11ICI

Saskatoon, Saskatchewan
Canada | July 23-27, 2023

11th International Conference on Isotopes

Abstract Book

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Introduction

The 11th ICI Conference was held in Saskatoon, Saskatchewan, Canada from July 23 – 27th, 2023.

11ICI hosted by Sylvia Fedoruk Canadian Centre for Nuclear Innovation in partnership with the University of Saskatchewan and Discover Saskatoon under the umbrella of the World Council on Isotopes, provided a platform for discussions, multifaceted interdisciplinary exchange between developers and producers of isotopes, scientists, members of the industry and academia working in the areas of medicine, industry, national security and other fields.

Selected papers will be published in the Journal of Radioanalytical and Nuclear Chemistry (JRNC).

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Oral Presentations

INVESTIGATING THE CONCERNTRATION OF RADIONUCLIDES IN WELLS USED AS DRINKING WATER IN NORTHERN NIGERIA. A CASE STUDY OF JOS METROPOLIS.

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Introduction

The increasing health effects of nuclear radiation occasioned by the enhanced human activities in the environment necessitated the need for constant investigation and assessment of radiological impact on the general populace within a confined area. Based on this, Twenty two (22) (Hand dug and motorized) well water samples were collected from various locations distributed across Jos Metropolis, Jos North LGA, Plateau State, Nigeria and analyzed for the concentration activity of these radionuclides (^{40}K, ^{210}Pb, ^{224}Ra, ^{232}Th, ^{238}U) usingradiochemical analysis technique, a high resolution gamma ray Spectrometry and a radon emanometry technique.

Description of the Work or Project

The activity Concentrations of the dug and motorized well water samples were measured using an N-type coaxial HPGe gamma-ray detector at the laboratory of Nigeria Atomic Energy Commission, Abuja with ORTEC Multichannel Analyzer (MCA) and MAESTRO-32 evaluation software for spectrum acquisition and processing. The relative efficiency of the detector was 28.5 % with energy resolution of 1.8 keV at gamma ray energy of 1332 keV of 60Co. The specific activity concentrations (Asp) of ^{226}Ra, ^{232}Th, ^{238}U, ^{210}Pb and ^{40}K were determined in Bq *l*-1 for the drinking water samples.

Results

The mean concentration ranges from 1.36±0.51 Bq l_1 to 5.75±1.30 Bq l_1. The mean concentration of ^{40}K in well water samples ranges from 3.80±1.19 Bq l_1 to 2.05±0.30 Bq l_1. The mean concentration of dissolved ^{224}Ra in well water samples collected varies from 5.75±1.30 Bq l_1 to 1.95±0.58 Bq l_1. ^{210}Pb has an average concentration of 2.68 ± 0.80 Bq l_1 to 1.97±0.87 Bq l_1. ^{232}Th and ^{238}U had average concentrations of 3.09 ± 0.57 Bq l_1 to 1.89±0.24 Bq l_1 and 5.41 ± 1.37 Bq l_1 to 1.36±0.51 Bq l_1 respectively. ^{210}Pb and ^{224}Ra were slightly above the recommended limits of 0.10 Bq l_1 and 1.00 Bq l_1 respectively, this can be attributed to the geological formation of the sampled area. ^{232}Th and ^{238}U where within the accepted standard limits of 1.00 Bq l_1and 10.00 Bq l_1 recommended value by WHO (World Health Organization) and ICRP (International commission on radiological protection).

Conclusions

Activity concentrations of measured radionuclides are in general decreasing in this order: ^{238}U > ^{40}K > ^{224}Ra > ^{232}Th and > ^{210}Pb for well water samples. These mean activities exceeded the World Health Organization recommended guidance level in all the water samplesof this study for ^{210}Pb, ^{224}Ra and ^{232}Th. This is a sole reflection of radiological contaminated surface and underground water and further consumption could be detrimental to the well-being of Jos North residents.

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NATURAL RADIOACTIVITY LEVELS IN THE UNSATURATED SOIL PROFILE OF SOUTH-WEST PUNJAB, INDIA.

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Introduction

Radioactivity either natural or man-made is ubiquitous in the Earth's crust though in different amount. In agricultural environments, information about the regional distribution of natural radionuclides is very less. Elevated levels of uranium concentration have been observed in the groundwater of central part of South-west Punjab, India. Uranium concentration in the groundwater reaches up to 250 μgL^{-1}, higher than the permissible limit of WHO (30 μgL^{-1}). The geochemical processes and source responsible for the alarming levels of U is poorly understood. Thus studying the natural radioactivity level in the soil profile of region has become really important.

Description of the Work or Project

In this context soil samples were collected from the agricultural fields with the help of hand auger from 10 different locations in the central part of SW Punjab. Sampling covers depth of up to 35 feet. As per the lithology observed with depth, a few soil samples were selected for analysis from each location. The radionuclide concentrations of the primordial radionuclides ^{238}U, ^{232}Th and ^{40}K were measured by applying the technique of gamma ray spectrometry using High Purity Germanium (HPGe) detector at Saha Institute of Nuclear Physics, GOI in Kolkata, India.

Conclusions

The average activities recorded in the soil vary from 27.97 to 67.48 Bq/kg for ^{238}U, 44.05 to 85.45 for ^{232}Th. The data shows that there is no significant variation in the radioactivity concentration with depth. The concentration level obtained was compared to International values and previous studies in the area. Levels are found to be higher than worldwide average concentration of these radionuclide reported by UNSCEAR, 2000. Also ^{40}K activity is higher than the global average. It can also be concluded that such high level of U and Th values points out toward the geogenic origin of uranium present in the groundwater of region.

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Keywords: Radionuclides, Groundwater, Gamma Ray Spectrometry, South-west Punjab.

COORDINATED INDUSTRY EFFORTS FOR RELIABLE ISOTOPE SUPPLY

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Introduction

The international supply chain for Mo-99 and other isotopes has experienced periods of instability due to unplanned outages of research reactors, production facilities, and disruptions in international commercial flight operations due to the COVID-19 pandemic. This has led to occasional shortages of both Tc-99m generators and – in recent years - I-131 for therapy.

International stakeholders including Governments, international organizations, nuclear medicine companies, trade associations, medical societies and others have undertaken efforts to address the root causes and to implement new or strengthened measures to improve reliable supply of Mo-99 and other isotopes. Nuclear Medicine Europe (formerly AIPES) and CORAR (Council on Radionuclides and Radiopharmaceuticals) have played important roles.

Description

The presentation will describe efforts of Nuclear Medicine Europe and CORAR to improve international radioisotope supply. Nuclear Medicine Europe, including with the European Observatory on Medical Radioisotopes, has focused on enhancing coordinated industry activities including responses to unforeseen disruptions and improved research reactor scheduling. The presentation will discuss and highlight in particular the Nuclear Medicine Europe Security of Supply Working Group and Emergency Response Team (ERT) in improving international supply of Mo-99 and I-131, and to monitor isotope supply developments related to projected increases in demand of Lu-177. CORAR's efforts in advocating for U.S. Government support for domestic production of Mo-99 and other isotope supply efforts will also be reviewed.

Conclusions

Nuclear medicine industry and its trade associations have played a critical role among international stakeholders to strengthen the Mo-99 supply chain as well the supply of other isotopes. Such measures have been successful in improving the reliability and security of supply of Mo-99 and other isotopes.

Keywords: research reactors, Mo-99, I-131, Lu-177

CROSS SECTION MEASUREMENT AND THICK TARGET PRODUCTION OF TERBIUM RADIONUCLIDES BY ENRICHED GADOLINIUM TARGETS IN BIOMEDICAL CYCLOTRONS

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Introduction

Short-lived radioisotopes of the terbium (Tb) family show great prospects in theranostics: the ^{149}Tb can be used for alpha therapy, the ^{152}Tb, as a positron emitter, can be applied for the positron emission tomography (PET), the ^{155}Tb can be used for the single photon emission tomography (SPECT) and for Auger therapy, and finally, the ^{161}Tb can be an alternative to ^{177}Lu for β-therapy. Nevertheless, the applications of Terbium are limited at the moment due to its insufficient production and high cost: except for ^{161}Tb, the other radionuclides are produced by nuclear spallation reactions. The use of enriched Gadolinium (Gd) targets can help to increase their availability according to the following production reactions: ^{152}Gd(p,4n)^{149}Tb [1], ^{152}Gd(p,n)^{152}Tb [1], ^{155}Gd(p,n)^{155}Tb [2] and ^{155}Gd(d,2n)^{155}Tb [3]. In this work, the ^{155}Tb is taken as a case study, and Gd_2O_3 enriched in ^{155}Gd is used. Objectives of this work are on the one hand to measure the cross section of the ^{155}Gd(d,2n)^{155}Tb nuclear reaction induced by deuteron, and on the other hand to irradiate enriched Gd_2O_3 targets for thick target production with deuteron.

Description of the Work or Project

For the cross section measurement, thin targets (10-20 µm) are required while thicker targets are preferred for production. Therefore, two types of Gd targets with different thicknesses have been developed through two different techniques.

Thin targets were manufactured via the electrochemical co-deposition technique. Uniform Ni/Gd_2O_3 composite targets with a thickness of 10-20 µm containing about 2 mg of enriched Gd were obtained after 35 min of deposition. These targets were irradiated at GIP ARRONAX cyclotron with deuteron beams. Cross sections of ^{155}Tb and other Tb radionuclides (^{153}Tb, ^{154}Tb and ^{156}Tb) were measured from 8 MeV to 30 MeV. These measurements give the first experimental results for the reaction ^{155}Gd(d,x)Tb. From these results, the thick target yield and the purity of ^{155}Tb were estimated. The irradiation parameters for thick target production were also determined from the simulation.

Thicker targets were manufactured through the pelletizing technique. A uniform and compact target with a thickness of 390 µm was obtained using 0.6 g of enriched Gd_2O_3 powder. This target was irradiated by deuteron beams with an incident energy of 15.1 MeV and a beam intensity of 368 nA for 1 h. The production yield of ^{155}Tb was 10.2 MBq/µAh and the purity was 89% after 14 days of decay. These results are consistent with the estimation obtained by the measured cross sections.

Conclusions

This work shows the possibility of using enriched gadolinium targets to produce terbium radioisotopes via biomedical cyclotrons. Cross sections of deuteron-induced reactions on enriched Gd were measured and a test of thick target production was carried out.

As for large batch production, higher intensity and longer irradiation time will be necessary. To this end, specific encapsulation and cooling systems will also be designed and in addition, pure metal Gd targets with better thermal conductivity will be developed.

Keywords:
cross section measurement; radionuclide production; terbium; gadolinium; cyclotron; theranostics radionuclide

Acknowledgments:
The ARRONAX cyclotron is a project promoted by the Regional Council of Pays de la Loire and financed by local authorities, the French government, and the European Union. This work has been, in part, supported by a grant from the French National Agency for Research called “Investissements d’Avenir”, Labex No. ANR- 11-LABX-0018-01. It has been also supported partly through funding from the European Union’s Horizon 2020 research and innovation program under grant agreement No 101008571 (PRISMAP).

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Production of Pb-203 from target manufacturing to chemical separation Pb/Tl

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Introduction

Lead-212 ($t_{1/2}$ = 10.6 h) and lead-203 ($t_{1/2}$ = 51.9 h), can be used as a theranostic pair of isotopes for theranostic applications in nuclear medicine. ^{212}Pb can be obtained from ageing ^{232}Th and is used for alpha targeted therapy [1] whereas ^{203}Pb can be produced by proton or deuteron irradiation of a thallium target and allows for single photon emission computed tomography (SPECT) thanks to its 279.2 keV (80.9%) photons. Current production of ^{203}Pb uses ^{nat}Tl bombarded by a proton beam. In this work, we consider alternative production routes using enriched ^{205}Tl and a deuteron beam in order to limit the level of ^{201}Pb ($t_{1/2}$ = 9.33h) impurities and to avoid the production of ^{200}Pb ($t_{1/2}$ = 21.5h). Starting from cross section measurements, we have defined optimal production parameters and yields. In parallel, we have developed a manufacturing process of the target by electroplating as well as a separation chemical process using a Pb resin from Triskem. This scheme will allow us to produce ^{203}Pb in the near future.

The electrodeposition technique is used to prepare enriched Tl target for both cross section measurements and mass production. A pulse reverse potential is used to reduce Tl^+ to Tl on gold substrate. The temperature and pH of the solution are fixed respectively at 20°C and 8. The solution contains EDTA as complexing agent, hydrazine to prevent Tl^+ from oxidation to Tl^{3+} and Brig-35 as surfactant.

For the study of ^{203}Pb production cross sections and its impurities, thin deposits of ^{205}Tl (thicknesses ranging from 10 µm and 15 µm) with a circular shape (4 cm^2) were made. Experiments were done with a deuteron beam whose energy is ranging from 22 MeV and 34 MeV.

For the future routine production, a large deposit of enriched Tl is needed (14 cm^2 area) to reduce the heat deposition density. To this end, we are using our rabbit system that accommodate a 15° tilted angle target. A 40 µm thick was successfully obtained with a smooth deposit and good adhesion on gold backing. In parallel, we have studied the chemical separation of Pb/Tl using ^{203}Pb and ^{202}Tl ($t_{1/2}$ = 12.31 d) obtained by irradiation of thin deposits of ^{nat}Tl (20 µm) with an intensity of 50 nA deuteron beam during 1 h beam. Large Tl targets were dissolved in 1 mol/L of hot nitric acid (70°C) and tracers were added to be able to follow the different species. The solution was then poured in the column containing the Pb resin previously

conditioned with 1 mol/L of HNO_3. Tl was recovered by further eluting with 1 mol/L of nitric acid. For the elution of Pb, the solution of 1 mol/L of ammonium acetate at pH 7 is used. The recovery of Pb is 92% with less than 1% of the presence of the impurities of Tl in the solution containing Pb. In parallel, the recovery of Tl is also studied and the yield is more than 90%.

Some productions of ^{203}Pb with enriched thallium target will be produced and our results will be presented during my presentation.

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Orano Stable Isotopes Laboratory: A new occidental supplier of stable isotopes

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Introduction

Orano has been looking at leveraging on its unique competencies and technologies in chemistry and enrichment with a new activity: stable isotopes production.

Description of the Project

In 2018, Orano launched a new activity for producing stable isotopes based on its best in class separation and chemistry technologies and unique knowhow. The Stable Isotope Laboratory, an activity of Orano Chemistry & Enrichment, has all assets to be a stable isotopes separation benchmark player in the Western world:

- a state-of-the-art isotopic separation technology and among others for stable isotopes: ETC, co-owned by Orano;
- A thorough expertise in fluoride chemistry widely used for stable isotopes production;
- Proven technologies for downstream production including oxides and metals;
- An industrial platform located in Tricastin in the South of France, a recognized center of expertise with all the necessary infrastructure to host this new activity.

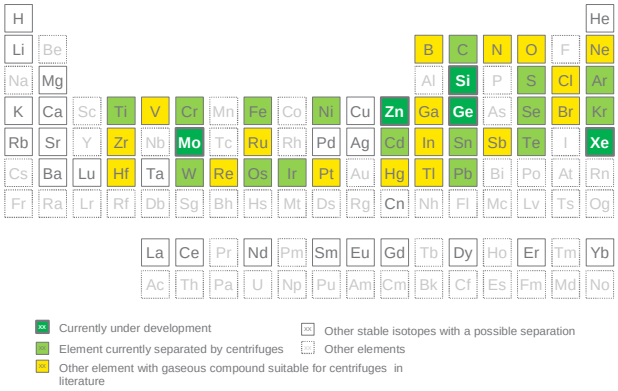

Figure 1: Stable Isotopes Catalogue

Around 20 elements can be enriched or depleted by centrifugation. First elements are being developed by Orano and will be produce as of 2023 with a high grade of purity for leading-edge applications such as medical imagery, oncology, advanced research… The first cascade has been designed to produce a wide range of volumes: from 10g to a couple of kilograms. Based on customized concept through the use of flexible cascades, the project will continuously develop new elements to serve its customers' needs.

The project is well advanced: civil works has been completed in October 2021, equipment will be erected through 2022 for a first commercial production in 2023.Thanks to its modular concept, production capacities will be increase according to market needs.

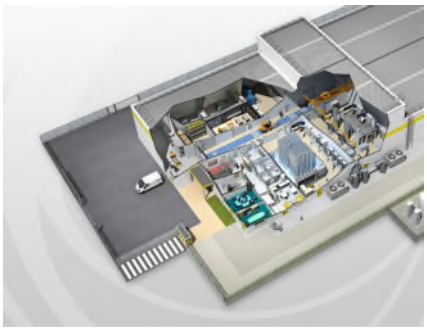

Figure 2 : Orano Stable Isotopes Laboratoy

Conclusions

The Orano Stable Isotopes project is based a proven technology and unique knowhows. Located in the heart of Europe, it is a welcomed alternative for stable isotopes users. The construction is on track and the production will start in 2023.

STRUCTURAL AND FUNCTIONAL CORRELATES OF ACUTE LUNG INFLAMMATION

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Introduction

The outcome of lung inflammation is important to host survival as lungs are necessary for oxygen exchange and fighting pathogens or any injurious stimuli. Thus, diagnosing and understanding the kinetics of lung inflammation is an emerging technological area of research and development. The premise of my thesis is to test the utility of longitudinal non-invasive imaging modalities, namely sequential [^{18}F]-fluoro-deoxy glucose (FDG) positron emission tomography-computed tomography (PET-CT) and synchrotron multiple image X-radiography (MIR), to assess the progression of acute murine low-dose ozone or bacterial lipopolysaccharide (LPS) induced lung inflammation over time.

Description of the Work or Project

Both ozone and LPS induced an increase in lung FDG standard uptake ratio (SUR) and a heterogenous lung distribution which was unlike the craniocaudal FDG gradient observed in lungs from mice before any exposure. The whole-body distribution revealed that lung FDG activity was higher and prolonged up to 28 h in LPS compared to ozone exposed mice. While FDG is a useful marker to highlight areas with high metabolic uptake of glucose in cells such as neutrophils and macrophages recruited during inflammation, the resolution of PET-CT precludes the evaluation of microscopic histopathologic changes especially in the alveoli. Using, binary masks of available stained lung cryosections, I assessed the ratio of lung and alveolar parenchyma to air spaces in mice left lungs exposed to 0.05 ppm ozone for 2 h. Our results from the X-ray CT lung tissue volume quantifications as well as the histologically derived percent-stained lung or alveolar area quantifications point towards significant damage that is observed as reduced percentage area as well as variability or standard deviation (S.D.) of binary lung images in mice immediately i.e. at 0 h and 6 h after exposure to 2 h of 0.05 ppm ozone exposure. Alveolar damage was also significant at 0 h as shown by reduction in percentage area and S.D. in the binary image region restricted to alveoli.

Conclusions

Thus, our study reveals that longitudinal FDG-PET imaging offers a tool for deep phenotyping of lung inflammation to understand the response to new targeted treatments in animal models and later in clinical trials. X-ray CT lung tissue volume quantifications as well as the histologically derived percent-stained lung or alveolar area quantifications point towards significant damage due to ozone.

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Keywords: *Acute lung inflammation, ozone, LPS, PET-CT, histology, image quantification*

^{99}Mo PRODUCTION VIA ^{99}Tc (μ^-, ν) ^{99}Mo REACTION ON RECYCLED ^{99}Tc.

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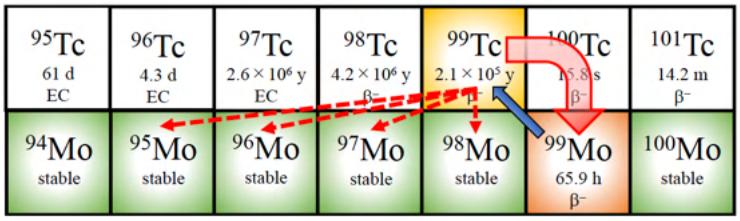
Muon nuclear-capture reaction produces a variety of useful radioisotopes for nuclear medicine [1, 2]. Muon is an elementary particle classified to electron group. The negative muon (μ^-) is captured by atomic orbits of nuclei, and cascades down to the lowest orbit to make muon nuclear capture. The muon is combined with a proton in the nucleus and converts to a neutron (n) and a neutrino (ν). Since a proton turns into a neutron, the atomic number of produced nucleus deceases by one. The muon nuclear-capture reactions on nucleus ($^{A}_{Z}N$) produce ($_{Z-1}{}^{A}N'$) isotopes by emitting x neutrons, where Z and A stand for atomic and mass numbers. The reactions are generally expressed $^{A}_{Z}N(\mu^-, x\text{n}\,\nu)^{A-x}_{Z-1}N'$.

It is well known that ^{99}Mo is an important parent-nuclei to generate ^{99m}Tc for imaging diagnostic in nuclear medicine. The ^{99m}Tc produced in ^{99}Mo-^{99m}Tc generator is eluted out by milking technique. We have proposed a new production method of ^{99}Mo by ^{99}Tc(μ^-,ν)^{99}Mo reaction, where muon is irradiated on recycled ^{99}Tc targets [1]. The ^{99}Tc(μ^-, xn ν)$^{99-x}$Mo reactions (x=0,1,2,3,4) produce ^{99}Mo (x=0) together with stable Mo isotopes (^{95}Mo,^{96}Mo, ^{97}Mo, ^{98}Mo), and the reaction diagram is shown below. The reaction routes from ^{99}Tc are shown by a large red arrow to ^{99}Mo and dotted red arrows to stable Mo isotopes.



The branching ratio of ^{99}Tc(μ^-,ν)^{99}Mo reaction has not been measured, but is expected to be about 10%. The mass distributions of produced Mo isotopes, specific activity of ^{99}Mo and the production rate are calculated. The expected specific activity is higher than the ones by the other ^{99}Mo production methods. By the reactions, ^{99}Mo is produced in ^{99}Tc target. The $^{99}MoO_4^{2-}$ ions are separated from $^{99}TcO_4^-$ ions by adsorbing in chromatography column, and this separation is a reverse process of milking for $^{99m}TcO_4^-$ in ^{99}Mo-^{99m}Tc generator.

The ^{99}Mo ceaselessly decays to ^{99}Tc once it is produced, as shown by a blue arrow in the figure. The ^{99}Tc is collected and safely stored at any ^{99}Mo processing stages. The ^{99}Tc left in used generators is collected at ^{99}Mo recharging stages. In addition, high-purity ^{99}Tc of about 1 Kg is contained in spent nuclear fuel of 1 ton [3], and can be extracted. These resources give an inexhaustible supply to the recycled ^{99}Tc target for the ^{99}Mo production method.

References

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^{225}Ac PRODUCTION VIA ^{226}Ra (μ^-, n ν) ^{225}Fr REACTION WITH ^{226}Ra TARGET.

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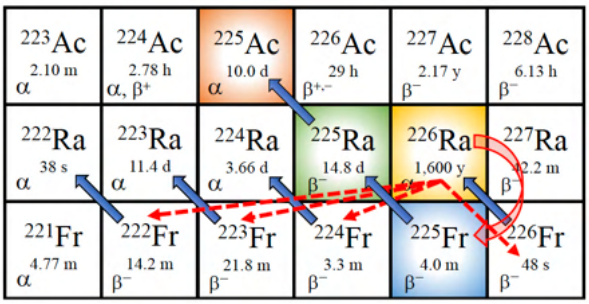
Actinium-225 (^{225}Ac) is known to be a promising radioisotope for Targeted Alpha Therapy and has been attracted global attention for the applications to cancer therapy. The demand has been increasing worldwide to develop clinical and pre-clinical researches.

A new production method of ^{225}Ac is proposed via ^{226}Ra (μ^-, xn ν) $^{226-x}$Fr muon nuclear-capture reactions on ^{226}Ra target [1]. The muon nuclear-capture reactions are generally expressed ${}^{A}_{Z}\mathrm{N}\ (\mu^-, x\mathrm{n}\ \nu){}^{A-x}_{Z-1}\mathrm{N}'$ and produce ${}^{A-x}_{Z-1}\mathrm{N}'$ isotopes from ${}^{A}_{Z}\mathrm{N}$ target, where A and Z stand for atomic and mass numbers. The chemical properties of produced ${}^{A-x}_{Z-1}\mathrm{N}'$ and target ${}^{A}_{Z}\mathrm{N}$ are different each other, and it makes the separation easier. By selecting the target nucleus, muon irradiation time and storage time, a variety of radioisotopes are produced [1].

The reaction rout to generate ^{225}Ac is shown below in the figure. Francium-225 (^{225}Fr) is firstly produced by ^{226}Ra (μ^-, n ν) ^{225}Fr reaction, and it decays with β^- emission to radium-225 (^{225}Ra). Subsequently, the ^{225}Ra decays with β^- emission to generate ^{225}Ac. The other Fr isotopes (^{222,223,224,226}Fr) are also produced by reactions with x=4,3,2,0, and decay with β^- emissions to ^{222,223,224,226}Ra, which are all α-emitters and do not decay to Ac isotopes. This is an advantage that ^{225}Ac is obtained free from the other Ac isotopes.



The branching ratio of ^{226}Ra(μ^-, n ν)^{225}Fr reaction has not been measured, but is expected to be about 45%. The mass distribution of Fr isotopes produced by muon irradiation and the generation yield of ^{225}Ra after a storage time are calculated. Consequently, the generation yield of ^{225}Ac obtained by milking from the ^{225}Ra is estimated by the calculation.

The batch and on-line production methods are designed. In the batch method, ^{226}Ra target is installed in a doubly sealed irradiation vessel and muon is irradiated from the outside to stop at the target. The ^{225}Ac generated in ^{226}Ra target after a storage time is chemically separated. In the on-line method, aqueous solution ^{226}Ra target is irradiated by muon for a short time. Employing chromatography column, the ^{225}Fr ions produced in the target are adsorbed and passed aqueous solution is recycled to the target. The ^{225}Ra is generated by β^- decay of ^{225}Fr in the column and eluted out to fabricate ^{225}Ra-^{225}Ac-^{213}Bi generator.

Reference

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CYCLOTRON-PRODUCTION OF INNOVATIVE RADIONUCLIDES: DIRECT ACTIVATION AND ISOL TECHNIQUE EXPERIENCE AT INFN-LNL

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Introduction

The cyclotron-based production of radionuclides for medicine is one of the research activities carried out in the framework of the SPES (Selective Production of Exotic Species) project at the Legnaro National Laboratories of the National Institute for Nuclear Physics (INFN-LNL). The heart of SPES is the 70 MeV proton-cyclotron with a dual-beam extraction, installed in 2015 in a new building equipped with ancillary laboratories currently under completion.

Description of the Work or Project

The SPES project aims at the construction of an advanced ISOL (Isotope Separation On-Line) facility for the production of re-accelerated exotic ion beams for nuclear physics studies. The double-beam extraction of the cyclotron also allows to perform multidisciplinary activities, such as radionuclides production for medical applications and neutron-based research. This work will mainly present the activities carried out in the unit "Radionuclides for medicine and applied physics", showing the major results obtained with the interdisciplinary projects LARAMED (LAboratory of RAdionuclides for MEDicine) [Esposito et al.] and ISOLPHARM [Andrighetto et al.]. LARAMED is based on the direct-activation method, and it includes the proton-based production of ^{99m}Tc, ^{67}Cu, $^{52/51}$Mn, ^{47}Sc and recently Tb-isotopes, from the nuclear cross section measurements to the preclinical studies. ISOLPHARM uses the ISOL technique for the development and the production of radioisotopes with high-specific activity, such as ^{111}Ag, going beyond the state-of-art in the field.

Conclusions

Thanks to a consolidated network of collaborations with national and international facilities, including the PRISMAP European consortium and several Italian universities and hospitals, the ongoing research activities on radionuclides production and their perspectives at the INFN-LNL will be presented at the 11ICI.

References

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SN-117M FOR METASTATIC BONE CANCER TREATMENT - MODERNIZING SN-117M-DTPA FOR REGULATORY APPROVAL

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Introduction

Sn-117m is a theranostic isotope used in for the treatment of metastatic bone cancer and bone pain with conversion electrons (CE) energy emission of ~0.14 MeV. The CE are mono-energetic with a maximum tissue range of 300µm. Early pre-clinical, and Phase 1/Phase 2 human trials utilized low specific activity (LSA) Sn-117m.[1] The specific activity (s.a.) of Sn-117m was 3 to 11 mCi/mg for use in the Sn-117m-DTPA therapeutic. The development of ≥1,000 mCi/mg, high specific activity (HSA) cyclotron produced Sn-117m has been used in animal trials in preparation for FDA regulatory qualification for human trials.[2] The Sn-117m-DTPA product can use LSA and HSA Sn-117m with FDA regulatory 'modernization' requiring additional animal species for biodistribution (BD) and dosimetry before qualifying HSA Sn-117m for human use.

Description of the Work

Additional background work for FDA regulatory qualification of new radioisotopes is required. Previously mouse studies using only LSA Sn-117m-DTPA were acceptable. Presently regulatory qualification of cyclotron produced HSA Sn-117m using both LSA and HSA Sn-117m-DTPA for BD and dosimetry and end-product validation in rat studies are required.

Results

	Srivastava mice	*ITG mice 9/10/2020*	*ITG rat 9/24/2020*	*ITG rat 10/14/2020*
		% Injected Dose		
Bone	36%	38%	60%	61%
Blood	0.02%	0.06%	0.03%	0.02%
Liver	0.47%	0.69%	0.52%	0.37%
Muscle	0.31%	0.60%	0.60%	0.58%
		Bone to soft tissue ratio		
Blood	1955	816	1945	2446
Liver	72	45	85	110
Muscle	860	472	614	652

Conclusions

Clinical trials utilizing reactor and cyclotron produced Sn-117m has been 'modernized' for FDA regulatory submission to include higher specific activity Sn-117m to match prior studies that are decades old. Additionally, regulatory agency requirements of pre-clinical studies have evolved to include changes in animal species and newer isotope validation studies.

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Modeling and assessment of Radioactive Iodine dispersion inside Egyptian radioisotope Production facility

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Abstract

Air quality is very important topic in radioisotope production facility. It is mandatory for some operators to be available behind hot cell to practice some activities concerning maintenance and operation. One of these tasks is redundant transferring Radioiodine from cell to QC lab and vice versa for measurements. Contamw is a simulation model from NIST (National Institute of Standards and Technology) is used to predict I^{131} concentration in air in hot cell and area of operator behind the cell in emergency case. Emergency is described by dropping small amount of I^{131} on cell floor. The model predicts the elapsed time for exhaust system to remove contaminants to dedicated filter and protect operator from inhalation. An emergency statue is also studied in case of opening I^{131} cell door hole (20 cm) by operators to pick the sample for quality control tests. Pressure interference occurs in this situation permitting some Iodine traces in the area under consideration. Ventilation system is responsible to evacuate and removes all radioactive species to settle inside dedicated filters to clean the area and keeps it in permissible safe limits.

Key words: contaminants, activity, simulation, air concentration, extraction air, Kinetic Reactions.

FIRST IN VIVO AND PHANTOM IMAGING OF CYCLOTRON-PRODUCED ^{133}LA AS A THERANOSTIC RADIONUCLIDE FOR ^{225}AC AND ^{135}LA

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Introduction:

Theranostic pairs in nuclear medicine involve labeling molecular targeting vectors first with a diagnostic radionuclide, followed by a therapeutic particle-emitting radionuclide. ^{225}Ac alpha-particle therapy has shown considerable efficacy in clinical trials for treating metastatic cancers, however existing positron emission tomography (PET) diagnostic radionuclides exhibit limitations such as short half-life (^{68}Ga), high positron emission energy (^{132}La), and excessive gamma ray dose (^{89}Zr). Owing to its chemical similarity with ^{225}Ac, low positron emission energy, and relatively long 3.9 h half-life, ^{133}La has strong potential to serve as a theranostic PET imaging partner with ^{225}Ac alpha-particle or ^{135}La Auger electron therapy.

Description of Work and Results:

^{133}La was produced on a 24-MeV TR-24 cyclotron using an aluminum–indium sealed target with 150–200 mg of isotopically enriched $^{135}BaCO_3$, $^{nat}BaCO_3$, or ^{nat}Ba metal. A NEPTIS automated synthesis unit performed barium/lanthanum separation. DOTA, PSMA-I&T, and macropa were radiolabeled with ^{133}La. Derenzo and National Electrical Manufacturers Association phantom imaging was performed with ^{133}La, ^{132}La, and ^{89}Zr and compared with ^{18}F, ^{68}Ga, ^{44}Sc, and ^{64}Cu. *In vivo* preclinical imaging was performed with ^{133}La-PSMA-I&T in LNCaP tumor–bearing mice. Proton irradiations for 100 μA·min at 23.3 MeV yielded 214 ± 7 MBq of ^{133}La and 28 ± 1 MBq of ^{135}La using $^{135}BaCO_3$, 59 ± 2 MBq of ^{133}La and 35 ± 1 MBq of ^{135}La using $^{nat}BaCO_3$, and 81 ± 3 MBq of ^{133}La and 48 ± 1 MBq of ^{135}La using ^{nat}Ba metal. At 11.9 MeV, ^{135}La yields were 81 ± 2 MBq, 6.8 ± 0.4 MBq, and 9.9 ± 0.5 MBq for $^{135}BaCO_3$, $^{nat}BaCO_3$, and ^{nat}Ba metal. $BaCO_3$ target material recovery was 95.4 ± 1.7%. National Electrical Manufacturers Association and Derenzo phantom imaging demonstrated that ^{133}La PET spatial resolution and scanner recovery coefficients were superior to those of ^{68}Ga and ^{132}La and comparable to those of ^{89}Zr. The apparent molar activity was 130 ± 15 GBq/μmol with DOTA, 73 ± 18 GBq/μmol with PSMA-I&T, and 206 ± 31 GBq/μmol with macropa. Preclinical PET imaging with ^{133}La-PSMA-I&T provided high-resolution tumor visualization with an SUV of 0.97 ± 0.17 at 60 min.

Conclusions:

With high-yield ^{133}La cyclotron production, recovery of $BaCO_3$ target material, and fundamental imaging characteristics superior to those of ^{68}Ga and ^{132}La, ^{133}La represents a promising radiometal candidate to provide high resolution PET imaging as a PET/α-therapy theranostic pair with ^{225}Ac or as a PET/Auger electron therapy theranostic pair with ^{135}La.

References:

Nelson B, Ferguson S, Wuest M, et al. (2021). First in vivo and phantom imaging of cyclotron produced ^{133}La as a theranostic radionuclide for ^{225}Ac and ^{135}La [published online ahead of print, 2021 Aug 12]. J Nucl Med. 2021;jnumed.121.262459. doi:10.2967/jnumed.121.262459

Keywords: PET; radiolanthanum; ^{133}La; ^{135}La; ^{225}Ac; theranostics; cyclotron

Efficiency Calibration of C-14 for LSC – It's Problems and Resolutions

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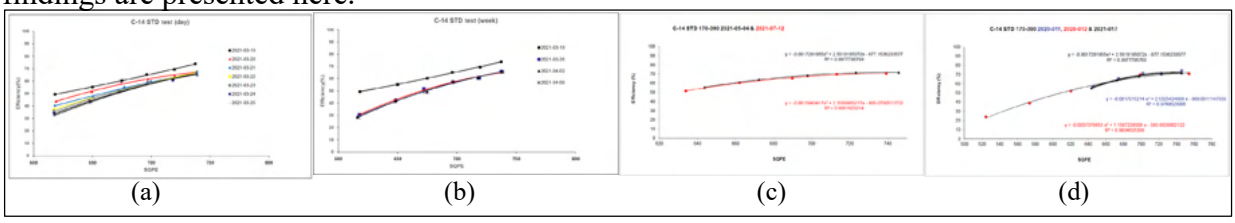
Introduction

^{14}C is one of fourteen radionuclides which must be identified from the radioactive wastes in order to get stored in a repository. Also it needs to be routinely monitored in air samples from NPPs. Prior to the routine measurements, the LSC should be properly calibrated for ^{14}C using a set of traceable calibration standard. It is common that the users use a set of commercially available calibration standard but those sets are not exactly same in geometry and chemical compositions etc. as those of actual samples prepared at laboratories for counting. If users want to use the set of commercially purchased standard for calibration, it is advised that such calibration details should be validated prior to the routine use.

Different between carbonate form of ^{14}C and toluene form of ^{14}C

The measured ^{14}C values of calibration sets were not stable over time which could lead to a significant failure of ^{14}C data. The prepared calibration standard sets were counted in a HIDEX 300SL liquid scintillation counter. The problem was investigated over a period of time and the findings are presented here.



(a) (b) (c) (d)

Figure 1. (a) Carbonate ^{14}C STD test(day), (b) Carbonate ^{14}C STD test(week), (c) Toluene ^{14}C STD test(2M) (d) Toluene ^{14}C STD test(year); .carbonate ^{14}C STD(set of 6), toluene ^{14}C(set of 10)

The radioactivity of ^{14}C in carbonate form is 173.79 Bq/g, Ref.Date: 2019-01-31, Supplier: Eckert & Ziegler and Diluted with: 0.1M NaOH. And toluene form is Activity : 338 Bq/g, Ref.Date : 2020-05-20, Supplier : Eckert & Ziegler and Diluted with : Toluene.

Conclusions

This study was conducted to evaluate the stability of ^{14}C carbonate and ^{14}C toluene when measured with a LSC. In the case of carbonate, it can be seen that the measurement efficiency decreases according to the SQPE when it is measured every day. However, it can be seen that the efficiency is the same after a week. In the case of toluene, it was confirmed that similar efficiency curves were drawn when measured at two-month intervals and one year later. When comparing the two, in the case of carbonate, the collection is not perfect, and it is judged that the efficiency decreases as it gradually evaporates.

Is ^{70}Zn(d,x)^{67}Cu the best way to produce ^{67}Cu for medical applications?

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Introduction

The theranostic approach combines diagnostic and therapy towards personalised treatment for patients. Among the different possibilities, associating two radionuclides of the same element is appealing, one for imaging purpose and the other for therapy. Copper offers such a pair of radionuclides with ^{64}Cu ($T_{1/2}$=12.7 h) allowing PET imaging and ^{67}Cu ($T_{1/2}$=61.8 h) for targeted beta therapy. If for ^{64}Cu the production route is well defined (a biomedical cyclotron delivering protons on a ^{64}Ni target), this is not the case for ^{67}Cu. Many production routes have been explored using neutrons, charged particles or photons. For charged particles, the main challenge relies on the limitation of ^{64}Cu co-production, even if its real impact on the patient and staff radioprotection needs to be studied and clarified. This work aims at identifying and studying an optimized production route for ^{67}Cu while limiting the impact of ^{64}Cu.

Description of the Work or Project

Within this frame, a preliminary research identified the production route ^{70}Zn(d,x). We have measured its production cross sections for ^{67}Cu up to 30 MeV. Measurements were done using the well-known stacked foils technique on 97.5 % enriched ^{70}Zn homemade electroplated targets. Beam intensity has been obtained using an instrumented Faraday cup. Cross sections were also measured for the monitor reactions ^{nat}Ti(d,x)^{48}V and ^{nat}Ni(d,x)^{61}Cu. They complement at higher incident energies the only set of data available in nuclear databases. The maximum of the cross section is reached for an incident energy of 23 MeV. Its value, 30 mb, is twice higher than the one obtained with a proton irradiation. With deuterons below 26 MeV, ^{64}Cu production is limited and directly connected to the enrichment of the target. Using this data, the production yield was calculated and production optimization parameters were proposed (energy incident of the deuteron, thickness crossed by the particles, irradiation time, waiting time).

Conclusions

This production route is of great interest as it limits strongly the production of ^{64}Cu that is directly linked to the level of ^{68}Zn impurity in the target. Using a 26 MeV deuteron beam and an enriched ^{70}Zn target, it is then possible to produce high purity ^{67}Cu. Activity of 16.4 GBq can be obtained for 26 MeV and 80 µA beam crossing 576 µm of ^{70}Zn thickness during 40 hours. By waiting 70 minutes after irradiation for decays, the activity of ^{67}Cu reaches 99.99 % of the total cooper activity and, at this time, the specific activity is 1.87 x 10^3 MBq/nmol or 2.79 x 10^4 GBq/mg. This specific activity value is very close to the theoretical maximum (2.80 x 10^4 GBq/mg). This production route can be of interest for future linear accelerators under development where mA deuteron beams can be available if adequate targetry is developed.

STUDIES ON SPECIES OF ARSENIC AND ITS COCONTAMINANTS IN WATER AND FISH BY NEUTRON ACTIVATION ANALYSIS

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Introduction

The toxicity of arsenic largely depends on its species. Arsenate, arsenite, dimethylarsinic acid (DMA) and monomethylarsonic acid (MMA) have been found in natural waters, and arsenobetaine (AsB) and arsenosugars in marine organisms. By far the most toxic as well as labile species of arsenic are As(III) and As(V). Antimony and selenium species can interact with arsenic species to either enhance or reduce its toxicity. Simultaneous multielement determination technique like neutron activation analysis (NAA) can be extremely helpful for such studies.

Description of the Work

We first developed a solvent extraction preconcentration and separation method involving ammonium pyrrolinedithiocarbamate (APDC) and 4-methyl-2-pentanone (MIBK) in conjunction with neutron activation analysis (NAA) for the simultaneous measurement of low levels of inorganic arsenic, antimony and selenium species in natural waters and fish samples. We studied several critical factors affecting the APDC/MIBK-NAA method in detail including the selection of chelating agent and solvent, aqueous pH for the extraction of six species as well as a few organoarsenic species, the stability of the complexes in organic phase, phase volume ratios for extraction and back-extraction steps, and the reduction of the species from higher to lower oxidation state.

We then combined solid phase extraction (SPE), coprecipitation, and NAA to develop a speciation analysis method based on green chemistry. In this method we separated and preconcentrated As(V), MMA, and DMA by strongly anion and cation exchange columns in tandem while As(III) remained in the effluent. We then selectively eluted these species and As(III) coprecipitated with bismuth sulphide. We applied this simple method to the analysis of water and fish reference materials with good results. The detection limits varied between 0.9 and 3.8 ng mL-1 for most arsenic species. We then extended the method to include the determination of Sb(III), Sb(V), Se(IV) and Se(VI).

Conclusions

The detection limits for arsenic, antimony and selenium were found to be as low as 0.026, 0.010 and 0.12 μg L^{-1}, respectively by the simple APDC/MIBK-NAA method. We used both this and SPE-NAA methods to study inter-element as well as intra-element species interactions. We found NAA to be superior to other conventional simultaneous multielement determination techniques. Details of the methods and results will be presented.

PRELIMINARY ASSESSMENT OF Tb-161 PRODUCTION IN MARIA RESEARCH REACTOR.

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Introduction

Terbium-161 (Tb-161) is of particular interest for targeted radionuclide therapy because of its similar properties to those of lutetium-177 (Lu-177), in addition it emits conversion and Auger electrons. Thus Tb-161 is considered to be more effective than Lu-177. Importantly, Tb-161 can be produced in nuclear reactors in the $^{160}Gd(n,\gamma)^{161}Tb$ nuclear reaction.

Description of the Work or Project

In order to assess the feasibility of Tb-161 production by neutron irradiation in MARIA research reactor in Poland the test irradiation of 0.87 mg of $[^{160}Gd]Gd_2O_3$ (enriched to 97.8%) was performed. The target material was irradiated for 184 hours in thermal neutron flux of 10^{14} n $\times cm^{-2} \times s^{-1}$. After two weeks cooling time, the target material was dissolved in 1 mL of 1 M HCl, evaporated to dryness and again dissolved in 1 mL of 0.1 M HCl. The aliquots of 0.05 mL were taken for measurement of radioactivity and radionuclide purity. Measurements were performed at 17, 28 and 44 days after the end of irradiation (EOB) by gamma spectrometry using HPGe detector, as presented in Table 1.

Table 1: Radionuclides and their measured radioactivities found in Tb-161 solution at EOB.

Measurement time	^{161}Tb [MBq]	^{160}Tb [kBq]	^{152}Eu [kBq]	^{154}Eu [kBq]
17 days	520 ± 10.7%	1.56 ± 37.9%	8.64 ± 64.0%	8.16 ± 38.9%
28 days	530 ± 10.7%	0.88 ± 99.4%	8.08 ± 40.7%	7.10 ± 39.7%
44 days	590 ± 10.7%	1,60 ± 12.3%.	9.03 ± 11.5%	6.94 ± 11.3%

The production yield of Tb-161 from highly enriched Gd-160 in the MARIA reactor was estimated at the level of 700 MBq/mg Gd. This value is similar to the results published by other authors [1]. The content of Tb-160 ($T_{1/2}$=72.3d) was $<1\times10^{-4}$%. Surprisingly, the two long lived europium radionuclides, ^{152}Eu ($T_{1/2}$=13.5y) and ^{154}Eu ($T_{1/2}$=8.6y), were found at the relatively high level up to 1×10^{-3}%. Their presence creates a concern for the reactor production route of Tb-161. Most probably they originated from the target material (according to the manufacturer's specification, the content of stable Eu in the target material is 1.1×10^{-3}%). Potentially, the radioisotopes of Eu can be removed during ^{161}Tb processing together with ^{160}Gd. However, in presence of long-lived Eu radionuclides the options for recycling of the Gd fraction for recovery of this costly target material are limited. Methods for purification of the target material (highly enriched ^{160}Gd) from Eu or for separation of the radionuclides of each of the three elements Eu, Gd and Tb need to be elaborated.

Conclusions

The feasibility of ^{161}Tb production in a medium neutron flux reactor such as Maria research reactor in Poland was demonstrated. The processing issues will be further addressed.

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PRODUCTION OF RADIOISOTOPES IN THE BR2 HIGH-FLUX REACTOR

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Introduction

The global production of radioisotopes for nuclear medicine, industry and research relies on a limited number of research reactors. The establishment of well-coordinated networks is required to ensure the security of supply. This is especially the case for the production of medical radioisotopes as Tc-99m used in 80% of the 35 million nuclear medicine procedures carried out worldwide annually for diagnostic. Additional irradiation capacity needs also to be developed in research reactors to face the explosive increasing demand for the production of therapeutic radioisotopes as Lu-177 for example.

Description of the Work or Project

The BR2 reactor operated by the Belgian Nuclear Research Centre is a 100 MW_{th} High-Flux Material Testing Reactor playing a key role in the production of medical and industrial radioisotopes. The availability of high thermal and fast neutron fluxes up to 1,2 x 10^{15} n/cm^2 s and 6,0 x 10^{14} n/cm^2 s respectively in dedicated irradiation devices allows the large production of various medical radioisotopes as Mo-99/Tc-99m, I-131, Xe-133, W-188/Re-188, Sn-117m, Ir-192, Lu-177, Sm-153, Y-90, Re-186, Er-169, Tb-161, Ho-166, P-32, Sr-89, Sc-47, ... The facility is also a major supplier of Ir-192 for non-destructive testing applications in the industry.

The successful refurbishment performed in the period 2015-2016 will ensure safe and reliable operation of the reactor for another period of 10 to 20 years. In addition, considerable efforts have been made to increase the irradiation capacity for the production of Mo-99 and Lu-177 in particular. Additional irradiation devices have been installed in an optimized reactor core configuration and the operating regime has been upgraded from 150 to 205 operating days per year since 2020 to secure the continuous supply of radioisotopes in a coordinated way.

The BR2 reactor has currently the largest world's installed irradiation capacity for Mo-99 production (7800 '6d' Ci/week) and is a major supplier of 'carrier-added' (c.a) Lu-177 and 'no-carrier-added' (n.c.a) Lu-177 from the irradiation of highly enriched Lu-176 and Yb-176 targets. The specific activities achieved by both production routes are respectively 35-40 Ci/mg and 80-95 Ci/mg at the end of a 7-day irradiation in a thermal neutron flux of up to 4,8 x 10^{14} n/cm^2s. Another achievement has been the successful validation work done to supply Y-90 and Ho-166 microspheres for liver cancer therapy.

Conclusion

The BR2 reactor will continue to play its key role in the global supply and the development of new radioisotope production routes in the next future.

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INTRA-ARTICULAR TREATMENT OF NATRAULLY OCCURRING CANINE ELBOW ARTHRITIS USING HOMOGENEOUS TIN-117M COLLOID

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Introduction

It has been suggested that as many as 20% of dogs over 1 year of age are afflicted with osteoarthritis (OA).[1] Radiosynoviorthesis (RSO) has been used successfully to treat human arthridities using other radioisotopes. This study determined whether homogeneous tin-117m colloid (HTC) a veterinary device could successfully treat naturally occurring canine elbow OA by RSO.

Description of the Project

There were three phases to this multi-centered observational safety and effectiveness study with each phase lasting 1 year in study dogs. Phase 1 was the intra-articular (IA) label dose treatment of a single elbow of companion dogs with mild or moderate elbow OA with HTC. Phase 2 was the IA label dose HTC treatment bilaterally or unilaterally as clinically indicated of companion dogs with severe elbow OA. Phase 3 was to reinject companion dogs a second time IA with HTC which previously had been treated in phase 1. In phase 3 both elbows were injected one for the second time and one for the first time. Comparisons were made with data collection times at 1, 3, 6, 9 and 12 months post treatments to baseline control values to determine successful improvement and safety.

Table 1: 3 phases of IA HTC canine elbow OA study

	Treated dogs	Treated elbows	Successful Improvement
Ph.1: unilateral IA treatment of mild-mod. OA	13	13	12
Ph.2: IA treatment of severe OA	14	25	11
Ph.3: reinjection of Ph.1 treated elbows	9	18	6

Successful improvement of each dog's condition with elbow OA was determined by composite scoring utilizing a validated client canine brief pain inventory survey, clinicians' lameness scoring and force plate evaluation when available.

Conclusion

Intra-articular injection of HTC, a veterinary device, to treat naturally occurring elbow OA in companion dogs was observed to be safe and effective and can be repeated a second time when indicated.

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Dissemination of Canada's National Standards for Radionuclide Metrology

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Introduction

Calibration of detector systems, traceable to the International System of Units (SI), is at the heart of any reliable measurement. For Nuclear Medicine applications, the knowledge of the amount of substance available in diagnostic or therapeutic radiopharmaceuticals can have a direct effect on patient outcomes. The National Research Council (NRC) of Canada is Canada's National Metrological Institute and is charged with developing, maintaining and disseminating Canada's standards for radioactivity. This work will describe the dissemination of three radionuclides standards – ^{99}Mo, ^{177}Lu and ^{90}Y – in 2020 and 2021.

Description of the Work or Project

Due to the short-lived nature of many radionuclides used in nuclear medicine, the standards for many γ-emitting isotopes are disseminated through the use of the NRC Secondary Standard of Ionizing Radiation Chamber System (SSIRCS). This is a collection of Ionization Chambers (IC) which have been calibrated using artifacts standardized by primary methods directly traceable to the SI.

The SSIRCS has been calibrated for many radionuclides in two primary calibration geometries; a 5ml serum vial and a 5ml flame sealed ampoule. If the calibration of the requested isotope has already been established, a standard can quickly be prepared by the end user or by the NRC and then exchanged. Measurements in the remote detection system (e.g.: IC or γ-spectrometer system) and the SSIRCS provide all the necessary information to the end user to calibrate their detectors. This was how a recent (2021) standard for ^{99}Mo was carried out.

The request for a traceable standard of ^{177}Lu required more work because the end-user geometry was not one which was a standard geometry maintained by NRC. Moreover, at the time of the request, the SSIRCS had not been calibrated for this isotope. In this case, the end user sent a dose of the ^{177}Lu radioisotope in their calibration geometry to NRC. The dissemination of the standard was performed by certifying the activity concentration of the master volume provided to the NRC.

Calibration geometries are central to the dissemination of the standards. Occasionally, a geometry can be rather unique, as in the case of a glass sphere ^{90}Y product that required standardization. The approach taken here was to prepare a mock artifact with cold glass spheres surrounded by a standardized ^{90}Y solution.

Conclusions

The standard uncertainty with which a standard of ^{99}Mo, ^{177}Lu and ^{90}Y is provided by NRC is 1%, 0.5% and 0.3% respectively. Since the method by which this standard is disseminated varies according to the requirements of the end user, NRC has developed a range of techniques to address common nuclear medicine isotopes.

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DEVELOPING PRODUCTION AND RADIOCHEMICAL SEPARATION FOR EMERGING RADIONUCLIDES FOR TARGETED MEITNER-AUGER THERAPY AT TRIUMF

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Introduction

Targeted Meitner-Auger Therapy (TMAT) provides great potential for selective therapy of cancer on the cellular level due to the short range[1]. Several research candidates with combined suitable half-lives, number, and energy of Meitner-Auger (MA) electrons and the absence of other accompanying emissions are currently under investigation. Namely, ^{119}Sb, $^{197m+g}$Hg, ^{103}Pd, ^{135}La, ^{155}Tb, ^{165}Er are currently produced at TRIUMF and actively investigated for their therapeutic potential. In this presentation, radiochemical aspects of the production of selected Meitner-Auger emitting radionuclides will be discussed.

Description of the Study

One of the advantages of MA emitters in comparison to the more commonly used β^- and α-emitters for Targeted Radionuclides Therapy is their accessibility as they can be produced with small to medium energy cyclotrons that are widely available in medical centers and hospitals around the world. At the Life Sciences Division at TRIUMF we are actively using our smallest TR13 (13 MeV) cyclotron for the production of emerging research candidates for TMAT. Further to the production, radiochemical purification is required to isolate the produced radionuclides from the target material and other possible stable contaminants [2] as for the final medical application through conjugation with selective delivery systems (e. g. peptides or antibodies) high radiochemical and radionuclidic purity is required.

Results

Production of research quantities of the above-listed radionuclides for testing radiochemistry, chelation and pre-clinical evaluation was achieved. Several efficient radiochemical purification strategies were tested based on ion exchange and solid-phase extraction chromatography providing high recovery yield and purity suitable for further application in the synthesis of radiopharmaceuticals.

Conclusions

Production and radiochemical separation of emerging radionuclides for TMAT was developed that enables access to more advanced candidates for TMAT to be tested *in vitro* and *in vivo* and advancing the field of Targeted Radionuclide Therapy.

Acknowledgments

We gratefully acknowledge funding from the NSERC Discovery program (VR, CR, CH, PS), Canadian Cancer Society Innovation Grant, and Canadian Institute for Health Research Project Grant. TRIUMF receives federal funding via a contribution agreement with the National Research Council of Canada. We thank Prof. Raymond Reilly and members of his group at Leslie Dan Faculty of Pharmacy, the University of Toronto for their collaboration and the TR13 Cyclotron Operations Group consisting of Toni Epp, Ryley Morgan, Spencer Staiger, and led by David Prevost for regular irradiations of solid targets. We also would like to acknowledge Dr. Steffen Happel for fruitful collaboration and providing access to solid-phase chromatography technologies.

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Commercial Production of Actinium-225

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Introduction

NorthStar Medical Radioisotopes, LLC is in process of establishing commercial scale production of actinium-225 via electron accelerator irradiation of radium-226 targets.

Description of the Work or Project

For a number of years, NorthStar has investigated the various options for commercial scale production of actinium-225. [Harvey] After careful consideration of the options, NorthStar has chosen to establish commercial production using IBA supplied electron accelerators. This production route involves the irradiation of radium-226 targets with bremsstrahlung to knock out one neutron to yield radium-225. The reaction is:

$$Ra226\ (\gamma,n)\ Ra225$$

The radium-225 produced is then setup in “cows” and periodically milked for actinium-225 over a multi-week period. The actinium-225 yield is a function of the amount of radium-225 initially produced and the number of times each “cow” is milked.

This presentation will provide updated project status and timelines to commercial production of actinium-225.

Conclusions

The electron accelerator route for production of actinium-225 is capable of yielding commercial scale quantities of the product with little to no contaminants of concern.

References

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Commercial Production of Copper-67

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Introduction

NorthStar Medical Radioisotopes, LLC is in process of establishing commercial scale production of copper-67 via electron accelerator irradiation of enriched zinc-68 targets.

Description of the Work or Project

NorthStar has learned of an unmet need for commercial scale production of copper-67 which is a potentially significant therapeutic radioisotope in the near future. Understanding that NorthStar is already establishing the production of molybdenum-99 via use of an electron accelerator, we have chosen to build on this established expertise and produce commercial quantities of copper-67. This production route involves the irradiation of enriched zinc-68 targets with bremsstrahlung to knock out one proton to yield copper-67. The reaction is:

$$Zn68\ (\gamma,p)\ Cu67$$

The copper-67 produced is purified from the target material to yield a product for use in various therapeutic applications. The enriched zinc-68 is recovered, re-purified and then available for reuse.

This presentation will provide updated project status and timelines to commercial production of copper-67.

Conclusions

The electron accelerator route for production of copper-67 is capable of yielding commercial scale quantities of the product with little to no contaminants of concern.

References

None

APPLYING PRINCIPLES OF RADIOIMMUNOTHERAPY TO NON-CANCER CONDITIONS

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Introduction

Radioimmunotherapy (RIT) uses monoclonal antibodies to deliver cytocidal radiations to the cells with the high precision. So far this approach has been used successfully in cancer arena. However, the principles of RIT could be applied to other, non-cancerous diseases where the diseased cells express the antigens, distinguishing them from the healthy cells. Our laboratory has been developing RIT of infectious diseases and, more recently, of multiple sclerosis (MS).

Description of the Work or Project

Medical progress has resulted in high numbers of immunosuppressed individuals who are affected by the opportunistic fungal infections as their immune system is not functioning. There are also infections with the multidrug resistant bacteria, such as MRSA. Some infectious diseases such as HIV are still incurable. When applying RIT to the fungal and bacterial infections, we are targeting antigens which are expressed on the surface of the microbial cells, such as polysaccharides, beta-glucans, or certain components of the bacterial or fungal cell walls. In HIV we are not targeting the virus directly, but rather viral antigens such as HIV gp41 which is expressed on the surface of the infected cells (viral reservoirs) where the virus hides and propagates. We have demonstrated the efficacy and safety of this approach in several murine models of infections, as well as showed that radiolabeled antibodies to beta-glucans are safe in healthy dogs. In MS, which is a devastating neurological conditions, the activated T cells which attack the nerves protective sheath express high levels of PD-1. Thus, we have chosen PD-1 as our target for RIT of MS with the purpose of slowing down the disease progression and/or ameliorating the MS symptoms. The homing of the antibodies on PD-1 expressing T cells was visualized by microSPECT/CT in experimental autoimmune encephalitis (EAE) in mice which recapitulates well the MS disease in humans. The increasing doses of 225Ac-labeled anti-PD-1 antibody proved to be safe to mice and the efficacy experiments in mice with EAE are currently on-going.

Conclusions

RIT can address the unmet needs of millions of patients with systemic infectious diseases and multiple sclerosis. RIT is technologically mature and can be easily translated from the cancer field into infectious diseases and other fields. Tertiary hospitals around the world are now routinely treating cancer patients with targeted radionuclide therapy and are fully equipped for deployment of infectious diseases and other types of RIT. There is a considerable enthusiasm towards RIT among physicians and patients.

Evaluation of ^{225}Ac production rate and its uncertainty in the experimental fast reactor Joyo

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Introduction

^{225}Ac is a promising nuclide for targeted alpha therapy. The current supply of ^{225}Ac is mainly extracted from ^{229}Th generators, and the global supply is limited to 63 GBq/y[1]. In this study, the authors investigated the ^{225}Ac production method using the experimental fast reactor Joyo owned by Japan Atomic Energy Agency (JAEA), and evaluated the production amount and its uncertainty caused by uncertainties of neutron flux and nuclear reaction cross sections.

Evaluation of ^{225}Ac production amount

The direct production of ^{225}Ac in the nuclear reactor contains a large amount of byproduct ^{227}Ac, while the indirect method can produce pure ^{225}Ac by the milking from ^{229}Th or ^{225}Ra as generators. The authors focused on the ^{226}Ra(n,2n)^{225}Ra reaction which utilizes fast neutrons (>0.1 MeV) as one of the indirect methods (Fig.1)[2]. The burnup calculation code ORIGEN2(Version2.2) was used for the evaluation of ^{225}Ra and ^{225}Ac production amounts. The one group cross sections library for ORIGEN code was generated by using the neutron spectrum at the center of the Joyo MK-IV core and JENDL-4 nuclear data library. Consequently, it was found that the production amount of ^{225}Ra after 60 days irradiation and 6 days cooling was about 14.5 GBq/g-^{226}Ra and the production rate of ^{225}Ac was 11.6 GBq/y by 17 times milking every 17.5 days (Fig.2).

Fig. 1. ^{226}Ra transmutation path

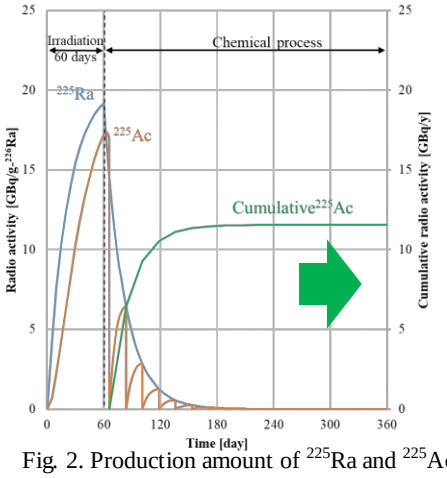

Fig. 2. Production amount of ^{225}Ra and ^{225}Ac

Evaluating the uncertainty of ^{225}Ra production amount

It is important to consider the uncertainty of ^{226}Ra (n,2n) reaction cross section and neutron flux in order to evaluate the ^{225}Ra production amount. The uncertainty of the ^{226}Ra (n,2n) reaction cross section was evaluated using covariance data prepared by the nuclear reaction calculation code CCONE, because the covariance data are not stored in JENDL-4. As a result, it was evaluated as about 18.4%. Assuming a neutron flux uncertainty of 10.0%, the Total Monte Carlo (TMC) calculation[3] was performed using these uncertainties. This method is to calculate nuclides production, depletion, and decay by the burn-up calculation code many times whereby at each run total neutron flux and one-group cross-sections are randomly sampled from a normal distribution with uncertainties width. As a result, the uncertainty of ^{225}Ra production was evaluated to be 21.0%.

Conclusions

The TMC calculation showed that the experimental fast reactor Joyo has potential to produce 14.5±3.1 GBq/g^{226}Ra of ^{225}Ra during 1 cycle (60days) operation.

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PROTON-INDUCED REACTIONS FOR ^{47}SC PRODUCTION: NEW NUCLEAR CROSS SECTION MEASUREMENTS ON ENRICHED TITANIUM TARGETS

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Introduction

The cyclotron-based production of medical radionuclides is one of the research activities carried out at the Legnaro National Laboratories of the National Institute for Nuclear Physics (INFN-LNL). The 70 MeV proton-cyclotron with a dual-beam extraction is installed in a new building equipped with ancillary laboratories, currently under completion. The interdisciplinary project LARAMED (LAboratory of RAdionuclides for MEDicine) [Esposito et al.] aims at the production of medical radionuclides for innovative radiopharmaceuticals, involving research activities from the nuclear cross section measurements to the preclinical studies. In this framework, the production of the theranostic ^{47}Sc (3.3492 d half-life) has been studied in the PASTA (Production with Accelerator of Sc-47 for Theranostic Applications) and REMIX (Research on Emerging Medical radionuclides from the X-sections) projects, funded by INFN for the years 2017/2018 and 2021/2023 respectively.

Description of the Work

This work mainly presents the proton-induced nuclear cross section measurements for ^{47}Sc production, considering also the co-production of contaminant radionuclides. Irradiation runs and γ-spectrometry measurements have been carried out at the GIP ARRONAX facility [Haddad et al.], by using the stacked-foils technique with a set of thin metallic targets. The enriched ^{48}Ti, ^{49}Ti and ^{50}Ti targets have been manufactured at the INFN-LNL, exploiting the HIVIPP technique (HIgh energy VIbrational Powder Plating) with the metallic powder deposited on a Al support. Targets have also been characterized with IBA (Ion Beam Analysis) methods to identify the ^{xx}Ti-amount deposited on the backing. Results on the ^{48}Ti(p,x)^{47}Sc cross sections and co-produced contaminants will be shown and compared with the scarce literature data and theoretical estimations; the preliminary results of the ^{49,50}Ti(p,x)^{47}Sc measurements will be also presented.

Conclusions

Thanks to a consolidated network of collaborations with national and international facilities, the ongoing research activities on proton-induced ^{47}Sc cross section measurements and their perspectives at the INFN-LNL will be outlined at the 11th ICI Session on Nuclear Data.

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TOTAL DOSE PERFORMANCE AT HIGH AND LOW DOSE RATE OF VOLTAGE REGULATORS SHOWING ENHANCED LOW DOSE RATE SENSITIVITY

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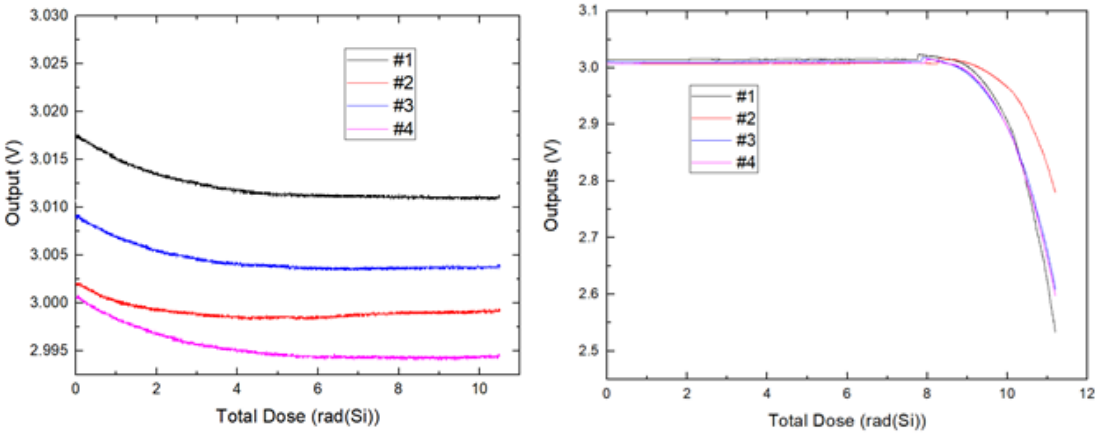
Introduction

The enhanced low dose rate sensitivity (ELDRS) of bipolar junction transistors and its failure mechanism have been extensively studied since it was first reported [1]. CMOS technologies are generally believed less susceptible to ELDRS due to the higher electric field in the oxide fields. However, recent studies showed that CMOS technologies and products could also be vulnerable to ELDERS. This damage could be caused by the much lower electric fields in the thick isolation oxide regions.

Description of the Work

This paper presents the total dose response of a CMOS low dropout (LDO) voltage regulator at 2.2 rad(Si)/s and 0.01 rad(Si)/s. Voltage regulators produce a constant voltage in theory independent of the loading, temperature changes, time and input voltage variation. The purpose of the lower dose rate irradiation was to determine the possibility of enhanced low dose rate sensitivity (ELDRS) of this CMOS microcircuit. The irradiations were performed at the University of Saskatchewan. The irradiation facility consists of two Gammacell 220 ^{60}Co irradiators. The total dose response of the microcircuits on each CCA at 2.2 rad(Si)/s and 0.01 rad(Si)/s are shown in the following left and right figures, respectively.



At the high dose rate the output voltage showed no significant degradation up to a total dose of 11.3 krad(Si) including post irradiation ambient and high temperature anneal. Drift at high temperature was well within performance provided on the manufacture datasheet. While, at low dose rate the voltage regulator showed significant degradation in output voltage at a total 11.3 krad(Si). The degradation recovered significantly during ambient annealing. Clearly, the device shows a significant ELDRS effects. It's likely due to voltage reference circuitry within the device.

Conclusions

The total dose performance of a CMOS LDO voltage regulator at high and low dose rates was presented, and the results suggest an ELDRS mechanism given the temperature sensitivity observed during experiments. It highlights the potential need to test analog and mixed signal CMOS commercial devices before application in the space environment.

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Measurement of activation cross sections for high energy neutron induced reactions of ^{59}Co, ^{nat}Ni, ^{nat}Zn and ^{nat}Cu

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At the MC-50 cyclotron facility, experiments are being performed to measure neutron induced reaction cross section of the ^{59}Co(n,x), ^{nat}Ni(n,x), ^{nat}Zn(n,x), and ^{nat}Cu(n,x) reactions. These measurements are done using off-line gamma spectrometric technique. Quasi mono energy neutrons are based on the ^{9}Be(p,n) reaction. The cross sections were measured at the Korean Institute of Radiological and Medical Sciences (KIRAMS) with quasi mono energetic neutrons in the 15.2-37.2 MeV energy regime. Theoretical calculations of neutron–induced reactions were performed using the nuclear model code TALYS-1.8. A few results are new, the others strengthen the database. Our experimental data were compared with results of nuclear model calculations and other available measurements. Adjusting TALYS parameters find best fit of model calculations well in both shape and numerical values with experimental values. The measured results for the formation of few radionuclides are new and the others measured data strengthen the database. The experimental and theoretical studies on the investigated reactions should lead to useful insight into mechanisms of ^{59}Co(n,x), ^{nat}Ni(n,x), ^{nat}Zn(n,x), and ^{nat}Cu(n,x) reactions.

Key Words: Neutron activation reaction cross section, Quasi-mono energetic neutron; Off-line γ-ray spectrometry; MCNPX 2.6.0 simulation; Talys-1.8., Theoretical calculations

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TERBIUM RADIONUCLIDES PRODUCTION BY DEUTERON BEAMS IRRADIATION FOR MEDICAL APPLICATIONS

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Introduction

The uniqueness in the terbium family consists in the fact that it has four radioisotopes of interest from a medical point of view: Tb-155 (half-life = 5.32 d) and Tb-152 (half-life = 17.5 h) can be used for SPECT and PET respectively; the properties of Tb-161 (half-life = 6.89 d) make it interesting from a therapeutic point of view because it allows a combined therapy with beta- and Auger electrons which has proved to be very effective in preclinical studies; finally, Tb-149 (half-life = 4.1 h) can be proposed for metabolic radiotherapy with alpha particles, therefore of interest for small metastases, with the associated possibility of diagnostics with PET due to the associated β^+ emission [1]. We present very preliminary results of the activation cross-sections of deuteron induced nuclear reactions for $^{nat}Dy(d,x)^{149,152,155,161}Tb$ production.

Description of the Work or Project

New measurements and data set for the $^{nat}Dy(d,x)^{149,152,155,161}Tb$ nuclear reaction are needed due to the lack of experimental data in literature. The stacked-foil activation technique irradiating at different energies thin natural Dy targets by deuteron beams was used to experimentally determine the excitation functions of the terbium isotopes and contaminants in the energy range between 23 MeV and 32 MeV. The irradiations were carried out at the ARRONAX Cyclotron Center, Nantes, France (E_d up to 35 MeV and 750 μA intensity) with a constant current of about 150 nA for a duration of 1 h. The irradiated targets were analyzed at LASA Laboratory in Milano. The thin-target yields have been plotted as a function of their average energy into the targets and were compared with the data present in literature [2] and with the predictions of computer codes like EMPIRE and TALYS.

Conclusions

The possibility to produce $^{149,152,155,161}Tb$ using deuterons beam induced nuclear reactions on ^{nat}Dy targets has been analysed. A comparison with the proton irradiation has been pointed out. A preliminary new set of excitation functions of the terbium radionuclides and the other co-produced contaminants for this nuclear reaction were obtained and compared with the other sets present in literature and the results of simulation with EMPIRE 3.2.2 and TALYS codes.

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RECYCLING USEFUL ISOTOPES FROM MOLTEN SALT NUCLEAR WASTE

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Introduction

The Molten Salt Nuclear Battery (MsNB) combines the benefits of nuclear power with the utility of a small-scale power generator. The MsNB is designed for onsite, continuous power generation for up to a decade. After completing operation, reusable isotopes, will be recycled. The remaining waste will undergo processing for safe disposal.

Description of the Work or Project

Before processing the waste, the reactor will rest for three years allowing for fission product decay and cooling of the reactor vessel. Once ready for processing, an addition between 19.8-27 mol% of zirconium hexafluoride (ZrF_4) must be added to the salt mixture to enable successful removal of uranium from the salt via fluorination. This is due to the ionizing nature of FLiNaK, the MsNB's working fluid [1].

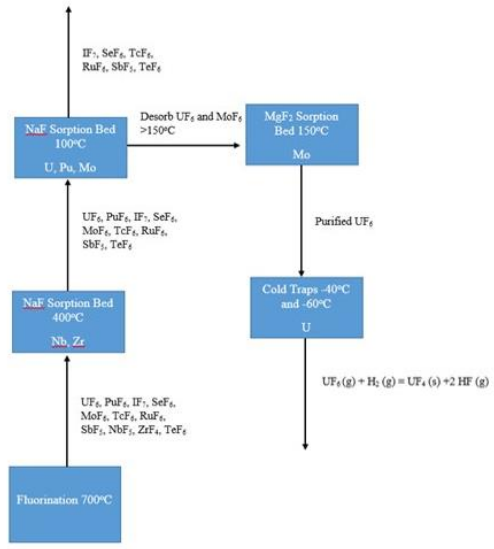

Figure 1: Flow chart of the uranium recycling process.

The salt mixture is initially heated to 700°C. Fluoride gas is then bubbled through the system creating volatile fluorides of uranium and fission products. The fluorides move into an off-gas system containing several sorption beds and cold traps [2]. The first is a NaF sorption bed heated to 400°C which absorbs niobium and zirconium from the gas stream. A second NaF sorption bed held at 100°C captures uranium, plutonium, and molybdenum. The remaining fluorides continue in the off-gas where they are separated by cold traps specific to each isotope. The uranium containing NaF bed is heated to above 150°C allowing the UF_6 and MoF_6 desorb from the bed. Molybdenum is sorbed into the MgF_2 bed held at 150°C leaving purified UF_6 in the gas stream to continue toward the first cold trap. The cold traps, kept at -40°C and -60°C, captures UF_6 which forms UF_4, the MsNB fuel form, when reacted with hydrogen gas [3].

Conclusions

This paper presents details of the fluorination process, which results in 95-99.5% recovery of uranium from MsNB spent fuel [2].

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IODINE RADIOLABELLED MESENCHYMAL STEM CELL (MSC)-EXOSOMES AND THEIR CD73 ENZYMATIC ACTIVITIES

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Introduction

MSC-derived exosomes have shown therapeutic potential in the areas of cardiovascular, orthopaedic, ophthalmologic, immune, dermatologic diseases and radiation sickness. Efficient radioisotope-labelling of exosomes remains as a challenging process. We demonstrate iodine-131 radiolabelled exosomes using both chloramine-T and Pierce Iodination methods, and characterized I-labelled exosomes via their CD73 enzymatic activities.

Experimental methodology and results

Two classic radio-iodination methods have been used to label exosomes due to several advantages: relatively long half-life of I-131 (half-life 8 days) and I-124 (half-life 4.2 days, 25.6% positron emission) could enable a desired tracking kinetics of exosomes in vitro and in vivo; radiolabelling of iodine to peptides and antibodies is a well-established chemistry; the unlabelled free iodine after radio-labelling can be easily removed to reach high radiochemical purity. By using chloramine-T, the radiolabelling yield of ^{131}I-labelled-exosomes achieved ~30-40% with a radiochemical purity > 90% after running through PD10 column purification. Using Pierce Iodination, the radiolabelling yield drops to ~15-20% (Table 1), radiochemical purity achieved >90% after the same purification process.

Table 1: Iodine-labelled of exosomes with radiolabelling yield, CD73 activity and particle size of post-labelling

	Radio-labelling yield (%)	CD73 activity	Particle size
Chloramine T	35-40	No	increased
Pierce	15-20	50%	same

The integrity of I-labelled-exosomes is important in the reproducibility and development of exosome clinical therapeutics. No radioactive iodine was labelled to exosome for characterization of their integrity. The results showed that chloramine T radiolabeling affected the structures of I-labelled-exosomes as the CD73 enzymatic activity of I-labelled exosomes was destroyed, the particle size became much larger and caused broader exosome size distribution. While with Pierce iodination the CD73 activity drops by 50 % when compared to that of the unlabelled exosomes, and the particles kept the same size (Table 1).

Conclusions

Using chloramine T method showed that the CD73 enzymatic activity of I-labelled exosomes was destroyed, suggesting the labelling process damaged the structure of exosomes. By comparison, using Pierce Iodination method preserved the CD73 enzymatic activity, indicating that exosomes can be radiolabelled using Pierce Iodination for in vitro and in vivo tracking and pharmacokinetic studies.

Development of decontamination technology for contaminated soil using non-radioactive isotopes and radioisotopes

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Introduction

In this study, a pilot-scale soil washing-based soil decontamination process was developed and applied by mixing surface fine-soil around nuclear power plants artificially contaminated with non-radioactive cesium with native soil. In addition, operational factors of critical processes for optimal physical distribution and cleaning were optimized and operating performance was carried out.

Evaluation of pilot-scale decontamination system performance

Artificially contaminated soil was manufactured by spiking non-radioactive and radioactive Cs into soil of less than 0.19 mm and mixed with surface soil for at least three to four weeks to obtain homogeneity of the specimen. Due to the environment in which radioactive soil cannot be obtained, the decontamination performance of the pilot system was evaluated using 100kg of experimental soil, which produced a ratio of 1:25 of non-radioactive cesium contaminated soil and raw soil.

Table 1. Cs concentration by particle size after soil washing

Sample	Before washing Weight (%)	After washing Weight (%)	1th Washing Cs Conc (mg/kg)	2th Washing Cs Conc (mg/kg)
Initial soil Conc		4.18±0.06		
25mm↑	1.2	1.1	N.D	N.D
5mm~25mm	11	8.1	N.D	N.D
2mm~5mm	15	15.8	N.D	N.D
0.2mm~2mm	67.5	37.4	0.72±0.08	0.07±0.05
0.05mm~0.2mm	5.2	25.5	0.64±0.09	0.34±0.09
0.05mm		12.2	1.23±0.02	0.44±0.09
0.05mm~2mm		0.007		0.005
0.05mm ↓		0.005	0.005	

Conclusions

Using the developed soil decontamination system, contaminated soil was classified into six levels according to particle size. Separation efficiency was best when the ratio of soil to water was 1:15 and about 63% of the contaminated soil could be decontaminated to the self-disposal level that could be classified as general waste. The concentration of trace-contaminated soil could be reduced to a self-disposal level by secondary cleaning. In the future, it will be planning to evaluate the decontamination performance of contaminated soil by spiking the standard radioactive materials for more reliable cleaning efficiency evaluation.

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MONTE CARLO SIMULATIONS OF THE NON-COLLINEAR GAMMA-RAY CASCADE EMISSIONS FOR MEDICAL IMAGING

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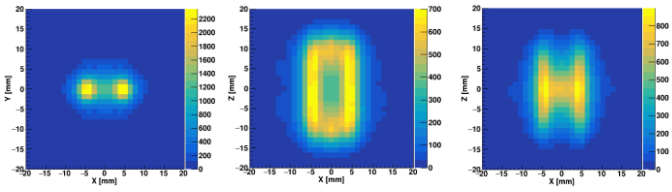
Introduction: Medical imaging by non-collinear gamma-ray cascade decay may be superior to the conventional PET and SPECT for at least two reasons: event-by-event analysis determines the decay vertices of stationary nuclei,[1] which enhances the image quality as back projections are not necessary (unlike SPECT), and the emissions are devoid of blurring from random positron motions (unlike PET).[2]

Description of the Work: A small animal PET composed of GSO detectors[3] can be adapted to the new modality by retrofitting with a SPECT-like collimators and software modifications. We developed a GATE-based collimated small animal PET model and a custom image reconstruction algorithm. We performed simulations of tungsten collimators with several point and line sources of ^{111}In isotope of various activities.

Results and Discussion: The reconstructed images were of the same shape as the geometry of the simulated source distributions (Figure 1). The image sensitivity of a single point source in air and in cubic PMMA phantom were 20.7 cps/MBq (2.1×10^{-5}) and 17.4 cps/MBq (1.7×10^{-5}), respectively. Their corresponding coincidence efficiencies are given in brackets. While the geometrical efficiency is decreased by many-folds compared to the uncollimated system; this loss is compensated by the image reconstruction algorithm, as each detected event constitutes the decay vertex of an image point. Typical spatial resolutions (FWHM) of 4.0 mm and 8.0 mm in transaxial and axial directions, respectively, were obtained.



(a) (b) (c)

Figure 1: Reconstructed images (a) transverse view of 2-point sources, (b) and (c) are coronal view of '0' and 'H' shaped line sources.

Conclusions: The results show that, within the visible region, image points reconstruction efficiency of the prototype 3D non-collinear cascade gamma-rays coincidence imager more than compensates for the loss of geometrical reconstruction efficiency of image points in standard PET and SPECT imaging systems.

Keywords: Small animal PET, ^{111}In, collimator, GATE, Medical imaging.

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Formulation and preliminary evaluation of a lyophilized kit of Arginine-Glycine-Aspertic acid (RGD) peptide derivative for convenient preparation of PET radiotracers

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Introduction

Radiolabeled arginine-glycine-aspartic acid (RGD) tripeptide based SPECT and PET tracers are already established as potent molecular imaging agents for the non-invasive imaging of various malignancies over-expressing integrin $\alpha_v\beta_3$ receptors like osteosarcomas, neuroblastomas, glioblastomas, lung carcinomas, ovarian and breast cancer [1,2]. In addition, these tracers can be useful tools in the monitoring of myocardial angiogenesis [3]. Rapid and convenient formulation of these radiotracers at hospital radiopharmacy is a prerequisite for their wider applicability. Against this backdrop, the present work is aimed towards the formulation and preliminary evaluation of a lyophilized kit for convenient and reproducible ^{68}Ga and ^{64}Cu-labeling of a dimeric cyclic RGD peptide derivative DOTA-E-[c(RGDfK)]$_2$ (E = glutamic acid, f = phenylalanine, K = lysine) for use as potent PET radiotracers for imaging angiogenesis.

Description of the work

A lyophilized kit containing 25 µg DOTA-E-[c(RGDfK)]$_2$ (Make: ABX Advanced Biochemical Compounds) and 21 mg ultrapure sodium acetate (Make: Sigma-Aldrich) was formulated under asceptic condition in a GMP certified clean room facility. Lyophilization was carried out for 24 h. Radiolabeling with ^{68}Ga was performed by adding [^{68}Ga]GaCl$_3$ solution in 0.05 M HCl (3 mL, 370 MBq) eluted from a 1.11 GBq ^{68}Ge/^{68}Ga generator (ITG GmbH) into the kit vial and heating the mixture for 15 min at 90° C. Subsequently, the reaction mixture was cooled to room temperature and subject to quality control tests. For the preparation of ^{64}Cu-radiotracer, ^{64}Cu was produced by ^{64}Zn(n,p)^{64}Cu route and radiochemically separated from irradiated ZnO target (99.2% enriched in ^{64}Zn) to obtain [^{64}Cu]CuCl$_2$ solution in 0.05 M HCl [4]. Copper-64 radiolabeling was performed by adding ^{64}CuCl$_2$ solution (2 mL, 370 MBq) eluted into the kit vial and heating the mixture for 20 min at 90° C. Both the radiolabeling yield and the radiochemical purity of [^{68}Ga]Ga- and [^{64}Cu]Cu-DOTA-E-[c(RGDfK)]$_2$ were found to be >97% (Table 1) as ascertained by radio-ITLC and reverse-phase HPLC following a gradient elution program.

Table 1. Radiochemical purities of RGD radiotracers

Radiotracer	Radioactivity (MBq)	Radiochemical purity (%)
[^{68}Ga]Ga-DOTA-E-[c(RGDfK)]$_2$	300 ± 25	99.0 ± 0.5
[^{64}Cu]Ga-DOTA-E-[c(RGDfK)]$_2$	370 ± 40	98.2 ± 0.7

Conclusion

Preliminary radiolabeling investigations carried out with the lyophilized kit demonstrated promising attributes of the developed kit formulation. However, it still warrants extensive pre-clinical and clinical investigations to establish its biological efficacy.

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Keywords

Lyophilized kit, RGD peptide, integrin $\alpha_v\beta_3$, non-invasive imaging, PET tracer

Neutron lifetime anomaly as a quantum correction to the classical time of flight

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Introduction

There is a long-standing tension between the beam and the bottle measurements of neutron's lifetime. The most recent precise bottle measurement gives an average lifetime of 878 seconds while the beam result is around 888 seconds. Using time operator formalism, we show that time measurements can be different if the details of preparation are different.

Description of the Work or Project

In principle, one can trace the beta decay products to the point and instant of interaction. That is, the neutron's wave function must be localized both in space and time for the decay to proceed. One way to realize the unitary localization of wave function is through the eigenfunctions of time-of-arrival operator. Thus, we can interpret the neutron's lifetime as an expectation value of time-of-arrival operator. For an initially prepared Gaussian wave packet with position uncertainty σ_0 and initial momentum of $\hbar k_0$, one can relate the quantum time expectation value with the classical result via the relation:

$\tau_{quant} = \tau_{class} \int_0^\infty dv \sin v \exp\left(-\frac{v^2}{8k_0\sigma_0}\right)$. Here, τ_{quant} is the quantum time and τ_{class} is the expected classical time. The important piece of the result is the quantum correction factor given in the integral above. Its behavior is shown in Figure 1.

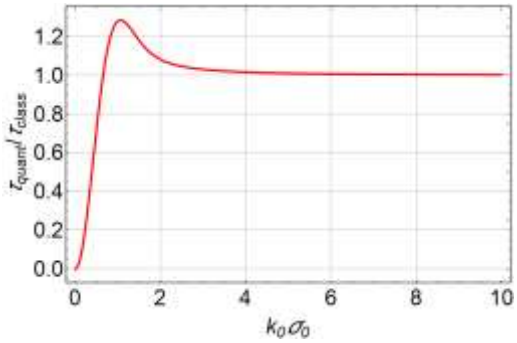

Figure 1: Quantum correction factor for an initial Gaussian wave packet.

The quantum correction effect is more prominent in the small $k_0\sigma_0$ region. This is the same region where the bottle and beam measurements belong since cold and ultracold neutrons have small initial momenta.

Conclusions

We provide an alternative explanation to the neutron lifetime puzzle. Our result suggests that the difference in the experimental results is due to quantum effects. The lifetime anomaly might be a useful avenue to explore the role of time in quantum mechanics.

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DISCRIMINATION OF GEOGRAPHICAL ORIGIN OF WHITE RICE IN THAILAND BASED ON STABLE ISOTOPES

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Introduction

Rice is a staple food for a large part of the world population. It is a main source of energy, vitamins, and essential elements for human especially in Asia, America, and some European countries. Thai rice is one of the main economic crops and export of Thailand. The geographical origin of rice product is an essential issue in order to prevent mislabeling and adulteration problem. Stable isotope ratio has been widely used to investigate the authenticity of food such as meat, dairy products, and rice. Therefore, the objective of the study was to discriminate the geographical origin of white rice cultivated from the northeastern, central, and southern Thailand using the stable isotopes combined with linear discriminant analysis (LDA).

Description of the Work or Project

Geographical origin of white rice samples was discriminated based on stable isotopes ($\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$). White rice samples cultivated from the northeastern, central, and southern Thailand were analysed using elemental analyzer isotope ratio mass spectrometer (EA-IRMS). In this study, the mean $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values found in white rice samples were in the ranges of -28.62‰ to -26.30‰, +1.87‰ to +8.67‰, and +23.41‰ to +28.58‰, respectively. There are significant differences in mean $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ found in white rice samples cultivated between 3 different regions of Thailand at 95 % confident interval with the p-values of <0.001. The stable isotopes combined with LDA could discriminate the geographical origin of white rice cultivated from 3 different regions of Thailand with 100% correct classification.

Conclusions

The stable isotopes ($\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$) in white rice samples cultivated from the northeastern, central, and southern Thailand were successfully determined by EA-IRMS. It is possible to discriminate the geographical origin of Thai white rice using stable isotopes in combination with LDA. The stable isotopic database and the established procedure in this study are useful in applications for other commodities and countries.

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GATE SIMULATIONS OF γ–γ CASCADE CORRELATIONS FOR AN ENTIRE-BODY SCANNER OF DIAGNOSTIC IMAGINGS

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Introduction: Recently, our international collaboration has made substantial progress in simulations and preparing for the prototype measurements of medical imaging from the spatial correlations of cascade gamma ray emissions of β^- emitting nuclei [1,2]. The project was undertaken to overcome two known deficiencies of Positron Emission Tomography (PET):

a) individual events do not locate the image points, necessitating the use of higher doses to the patient and involved statistical analyses for image reconstruction
b) blurring of the final image due to the positron's movement from the parent nucleus' decay site before annihilation

We simulate for an entire-body scanner, which is a full-body scanner in one setting to minimize the radiation dose to the patient and decrease the data acquisition time [3].

Description of the Project: We simulate an entire-body imaging system comprising of 40 rings each of 60 cm diameter for a total length of 190 cm. For non-collinear correlations, the detectors will be fitted with tungsten cylindrical collimators of 2 mm diameter holes of 4 mm wall thickness and 1 cm length to provide one-to-one correspondence of hit crystal and photon trajectories, just as in Single Photon Emission Computed Tomography (SPECT) arrangements. The detector assemblies are LSO crystals of 85% relative output.

The Monte Carlo simulations are carried out using GATE (GEANT4 Application for Emission Tomography), an open-source simulation toolkit. The isotopes employed are ^{111}In and ^{43}K, which have nearly 100% of their decays emitting pairs of photons, and they are widely used for medical purposes. The assembly performance will be assessed for image reconstruction efficiency, purity, and resolution of the system in comparison to the PET system of same geometry without collimators employing the workhorse FDG of ^{18}F isotope.

Conclusions: We have been working on novel medical imaging modality of exploiting well known directional correlations of cascade gamma rays in nuclear beta decays. The results will show the relative performance of this modality with respect to the conventional PET imaging system for its merits as a diagnostic tool.

Keywords: Medical Imaging, Entire Body Scanner, Cascade Gamma Ray Non-Collinear Correlation, Monte Carlo Simulations

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An Approach to Calculation of Individual Doses on Accident scenarios during Decommissioning of nuclear facilities

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Introduction

After the nuclear power plant is permanently shut downed, it is different from the accident type considered at the operation stage. One of the most difference in decommissioning is the occurrence of accidental criticality is not envisaged in shutdown nuclear reactors from which the fuel elements have been completely removed. However, given that there is radioactivity that can be emitted off-site if the existing confinement barriers are damaged by an accident or malfunction, it is necessary to evaluate the individual dose for accidents that might occur during decommissioning activities.

The accidents are conservatively approach that all radioactivity emitted to the off-site is released without considering any filters. The main exposure pathways are (1) external exposure by radioactive clouds from gamma and beta ray, and (2) internal exposure by suction of contaminated air. And the methodology for evaluating the impact of public due to radiological accident is the same as in operation phase. In addition, the atmospheric diffusion factor (X/Q) is applied at the Exclusion Area Boundary (EAB) and Low Population Zone (LPZ) for 2 hours, and conservatively, the emission type can be applied to the ground release. However, the methodology for defining released amount is different approach from the operation phase.

According to the NUREG-0586 document, the types of accidents that may occur during the decommissioning activities can be broadly classified into drop, fire, explosion, loss of confinement, liquid spill. In this paper, an explosion accident during the RV cutting activity was analyzed. Using the RV source term presented in IAEA SRS No.95, the amount of radioactivity released by cutting and the individual dose was calculated.

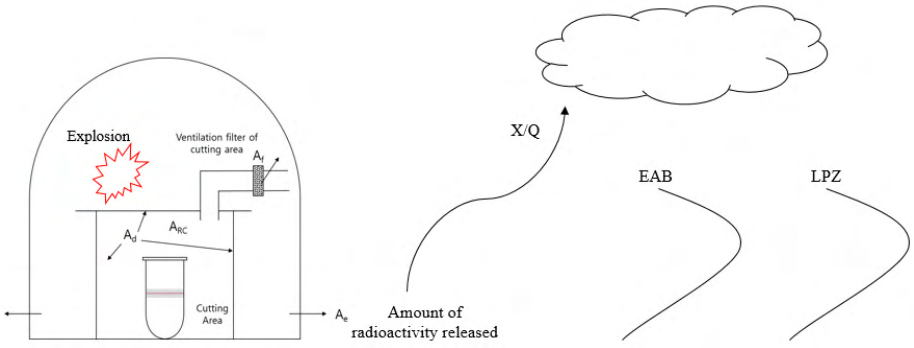

Fig. 1. An approach for individual dose assessment on explosion accident

References

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SOURCE PREPARATION TECHNIQUE OF ASTATINE-211 WITHOUT ELECTROPLATING FOR ALPHA SPECTROSCOPY†

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Introduction

^{211}At has attracted much attention because of its potential advantages in targeted alpha therapy. The high-resolution alpha spectrometry is one of the most important techniques in radiochemical analyses or precise activity measurements. The electroplating method is widely used as a conventional technique for the α-spectrometry. However, it might be difficult to adapt this method to ^{211}At due to its short half-life or volatility. To overcome these difficulties, another practical approach using a silver plate was employed to prepare sources for the α-spectrometry. In addition, a coprecipitation technique using $AgNO_3$ was also studied.

Methods and Results

In the present study, carrier-free ^{211}At produced via the ^{209}Bi(α, 2n)^{211}At reaction at the RIKEN AVF cyclotron was used[1)]. Around 1 MBq of dried elemental ^{211}At was dissolved with 10 mL of ion-exchanged water. Silver plates were used to fix astatine. Around 10 μL of the ^{211}At solution was directly dropped onto the silver plates. As another approach an ^{211}At source was also prepared with ascorbic acid to avoid ^{211}At loss due to volatilization. All sources were dried in atmosphere. To investigate time dependence of the deposition yield of ^{211}At adsorbed on the plate, the ^{211}At solution dropped on each plate without ascorbic acid was washed off with ion-exchanged water after 15, 30, 45, 60 and 75 min. As for the source preparation using the coprecipitation technique, 0.1 mg/g $AgNO_3$ solution was used. In this case 30 μL of the ^{211}At solution was directly deposited on the plastic plate and 10 μL of the $AgNO_3$ solution was added. These sources were dried in a desiccator with silica gel. All sources prepared were measured using a ZnS(Ag) scintillation detector with 2π geometry and/or an ion-implanted Si detector with the same source-detector geometry in vacuum. The deposition yield of ^{211}At adsorbed on the silver plates was determined as ratios of the activity on the plate to that determined from the activity concentration of the ^{211}At solution and a weight of its drop on the plate. As a result, the deposition yield was saturated to around 75% after 30 min. In case of the sample dried with ascorbic acid, the deposition yield was reached to 98%, while additional study using more samples should be required. In addition, no spectrum distortion due to self-absorption was found in the α-spectrum, resulting few differences could be found as compared with the source prepared without ascorbic acid. On the other hand, the deposition yield of the sample with silver nitrate was 70% and a significant spectrum distortion was found as compared with other two types of sources. Although this technique is one of the reliable methods to prepare radio-iodine sources with negligible volatility for standardization of their activities, it might be difficult to adapt the same procedure for ^{211}At as it is.

Conclusion

Among the three sources, the source prepared on a silver plate with ascorbic acid could have potential advantage to employ as a source preparation technique for activity measurement using the solid angle α counting technique with a small correction for loss of activity during drying process due to its volatility.

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PRISMAP, The European Medical Radionuclides Programme

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The field of nuclear medicine, the development of new radiopharmaceuticals and its related technologies has seen major advances, notably with the successful tests and marketing of companion drugs combining diagnostics and treatments, in the so-called field of theranostics. While the development of radiobioconjugates proceeds with the identification of a proper biological target, the synthesis and screening of different ligands, their tests in a staged approach, access to appropriate types of radionuclides associated with relevant information and the generation of new data is key for rapid and eventually successful new developments.
The supply chain for non-conventional radionuclides often proceeds through bilateral collaborations or through specific access at dedicated facilities. While some important programmes have already been launched, eg with the DOE-supported National Isotope Development Center, Europe was lacking until recently a coordinated consortium to supply researchers in biomedicine. *PRISMAP, the European Medical Radionuclides Programme*, was granted in the H2020 Infrastructures programme of the European Commission and kicked off last year, Fig.1. A first call for projects was recently launched for potential users and will select projects based on their scientific merit through an evaluation board [1].

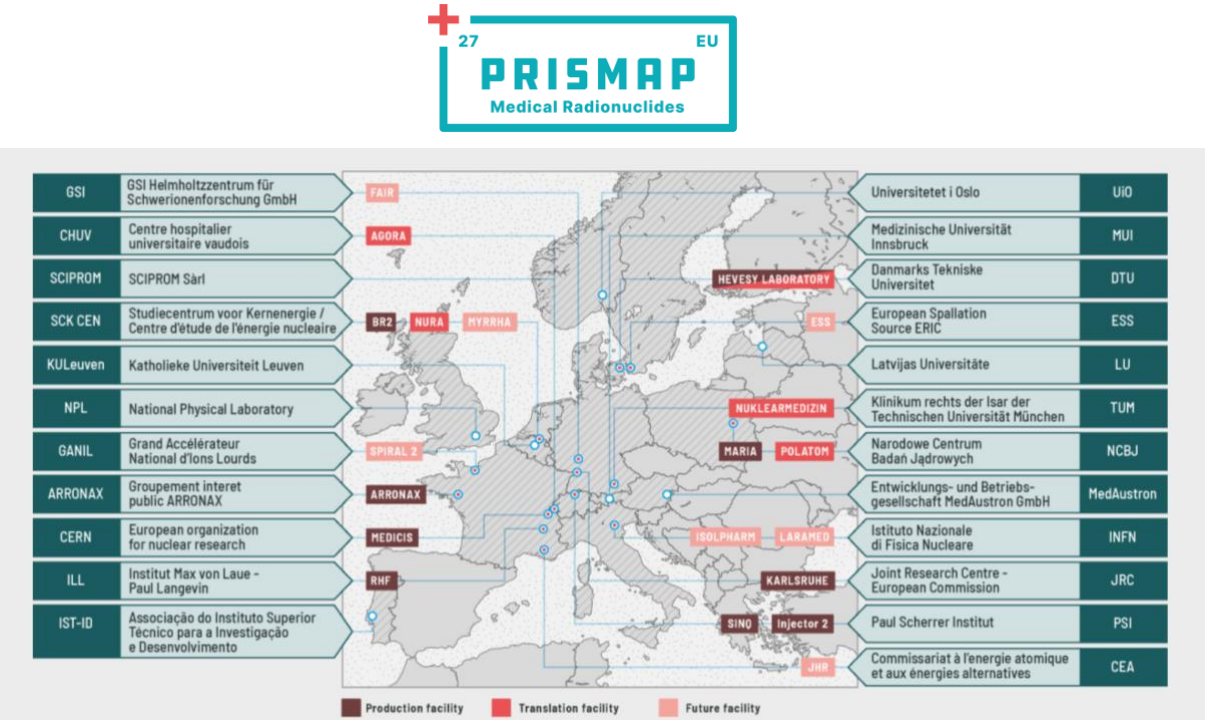

Figure 1: Overview of the PRISMAP consortium

Details on PRISMAP, its consortium and of Day-1 radionuclides will be reported here.

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This project has received fundin@g from the European Union's Horizon 2020 research and innovation programme under grant agreement No 101008571 (PRISMAP).

THE EVALUATION OF THE MICRO-GE DETECTOR FOR USE WITH HIGH ACTIVITY SAMPLES

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Introduction

Since their invention in the 1970s, high purity germanium (HPGe) detectors have been the gold standard for the detection of and spectroscopy with gamma rays. Developments in the fabrication, packaging, and operation of HPGe detectors, as well as in readout electronics and signal processing, have led to their application in fields as diverse as fundamental nuclear and particle physics, nuclear security and safety, and medicine. For example, position-sensitive HPGe detectors have been used to develop portable gamma-ray imaging systems, while extremely large HPGe detectors arrays are under development for the search for neutrinoless double beta decay. The supreme performance of HPGe is achieved through cooling to liquid - nitrogen temperatures and requires the detector to be mounted in a vacuum chamber so that the sensitive detector surfaces are protected from moisture and contaminants. Based on decades of experience in electrical cooling technology, in recent years Mirion Technologies has developed rugged germanium detectors that can be used in difficult conditions traditionally considered too challenging for HPGe detectors. An example of such a detector is our newest product: the MicroGe^{TM} detector. Due to it is small form factor and rapid deployment capability, the MicroGe^{TM} detector can be used in medical radioisotope production facilities where high resolution is critical to identify impurities in samples.

Description of the Work or Project

A small form-factor, rugged, easily deployable HPGe detector called the MicroGe^{TM} has been developed by Mirion Technologies and is commercially available. The ultracompact, lightweight design makes it very easy to deploy in the field, bringing high-performance energy resolution to industrial applications such as medical radionuclide production facilities, the measurement and identification of radionuclides in complex samples, and the measurement of highly radioactive materials. The palm-size MicroGe^{TM} detector can be brought very close to a sample in highly confined environments, and the rugged and compact design ensures the excellent energy resolution performance expected of HPGe performance, even in high-activity and high-throughput measurements. The MicroGe^{TM} detector is capable of going from storage to measurement in less than 30 minutes which is a revolutionary achievement for HPGes.
Portable systems and rapid deployment features of instrumentation are highly valued for many applications. Combining state-of-the-art technology with exceptional performance, Mirion is dedicated to providing unique solutions in radiation detection and instrumentation.

Conclusions

Detailed characteristics and performance of the new MicroGe^{TM} detector developed by Mirion Technology will be presented. This work will also describe how having the gold standard for spectroscopy (HPGe) in an ultracompact, rapidly deployable instrument can help quantify and identify gamma rays from different isotopes in challenging applications.

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https://www.mirion.com/products/search-measurement-analysis-instrumentation

HYPOXIA AND ANTIOXIDANT SIGNALING INDUCED IN HUMAN COLON CARCINOMA BY COPPER-64 AUGER ELECTRON EMISSIONS

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Introduction

^{64}Cu has been lately demonstrated to be a valuable theranostic in solid tumors, combining imaging and therapeutic properties. Nevertheless, there is an urgent need to describe the molecular background underlining the action mechanisms of ^{64}Cu and further identifying candidate molecular targets aimed to improve the therapeutic outcome, as well as the radio-toxicological profile of ^{64}Cu.

Description of the Work or Project

We assessed *in vitro* the effects of ^{64}Cu Auger electrons radiation on different cancerous cell lines and one reference normal cell line, testing for radio-induced oxidative stress by GSH and MDA content, activation of certain gene involved in cells oxidative stress response and protection. Cellular viability was tested using the MTS and LDH assays. Morphological staining methods were used to determine the cell death by apoptosis or necrosis. Experiments indicate that cell lines with higher proliferation rate are more susceptible to cell death by apoptosis, as a result to incubation with [^{64}Cu]$CuCl_2$.
To identify *in vitro* the response of normal and tumor cells to $^{64}CuCl_2$ using a transcriptomic approach, hypoxia signaling and redox responses were investigated. Human colon carcinoma HCT116 cells and human BJ fibroblasts (ATCC) were incubated with 20 MBq $^{64}CuCl_2$. Adherent cells were harvested in TRIzol at 24 h for gene expression studies performed by qRT-PCR using an array of 84 stress genes (Stress and toxicity pathway finder, Qiagen) and GAPDH / ACTB as housekeeping genes (RefFinder analysis). The treatment drastically reduced the number of viable cells in culture, both tumor and normal cells. The genotoxic stress inflicted by ^{64}Cu in vitro was highlighted by the up-regulation of CDKN1A and DDB2. Both adherent viable HCT116 and BJ cells had at 24 h a gene expression profile characterized by marked hypoxia signaling evidenced by up-regulation of the HMOX1, SERPINE1, SLC2A1 and VEGFA genes. The hypoxia-related genes ADM, EPO and MMP9 were found distinctively over-expressed only in tumor HCT116 cells. A molecular fingerprint of antioxidant response was detected in HCT116 tumor cells (up-regulated GSR, HMOX1 and PRDX1), with the HMOX1 gene at the crossroad of hypoxia and redox signaling.

Conclusions

^{64}Cu triggers genotoxic stress accompanied by hypoxia and oxidative stress in HCT116 colon carcinoma cells and normal BJ fibroblast, with distinctive patterns of gene expression in normal and tumor cells. Pharmacologic modulation of these signaling pathways may enhance the therapeutic effect of ^{64}Cu in tumors.

Theoretical Modeling of Tungsten-188 Production in the High Flux Isotope Reactor

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Introduction

Tungsten-188 (^{188}W) is produced through the irradiation of enriched ^{186}W in the High Flux Isotope Reactor (HFIR). This requires two neutron captures, one in the ^{186}W target material and one in the short-lived intermediary nuclide, ^{187}W ($t_{1/2}$ = 24.00 h). Accurately predicting the production yield of ^{188}W and osmium-191 (^{191}Os) from enriched ^{186}W metal rings irradiated in HFIR is not trivial. Two key factors that can lead to significant uncertainty or inaccuracy in yield predictions are the effect of neutron flux depression in the dense tungsten metal target and the inaccuracy of the currently accepted value for the ^{187}W thermal neutron absorption cross section.

Description of the Work or Project

Three different modeling and simulation methods/codes—HFIRCON, IsoChain, and MCNP6 combined with ORIGEN (MCNP6-ORIGEN)—were used to predict the theoretical ^{188}W and ^{191}Os yields for an irradiation sequence in HFIR. Theoretical values were then compared with experimental specific activity determined through high-purity germanium (HPGe) gamma ray spectroscopy and inductively coupled plasma mass spectrometry (ICP-MS). The theoretical and experimental specific activity of ^{188}W (in Ci/g) for one of these irradiations is shown in Figure 1.

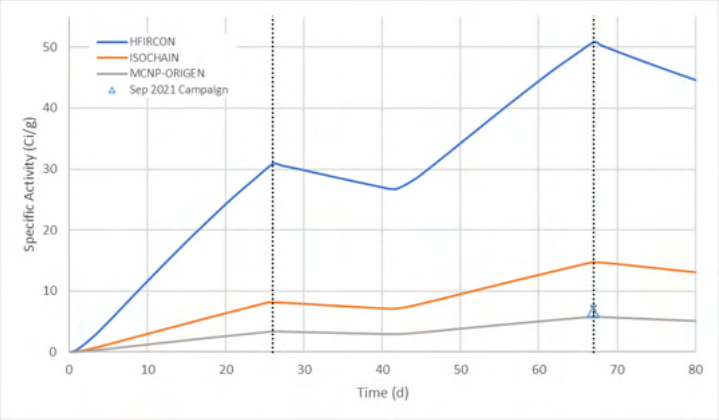

Figure 1. Theoretical specific activity of ^{188}W produced in HFIR using three different methods vs. experimental specific activity measured via HPGe gamma ray spectroscopy and ICP-MS.

Conclusions

Of the three modeling and simulation methods presented, the MCNP6-ORIGEN method resulted in a theoretical specific activity that was generally within 20% of the experimentally measured values. Possible modifications to HFIRCON and IsoChain that lead to more accurate predictions will be discussed.

Acknowledgment

This research is supported by the US Department of Energy Isotope Program, managed by the Office of Science for Isotope R&D and production.

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TALES OF AN ESSENTIAL WORKER DURING COVID, OPERATION AT THE UNIVERSITY OF WASHINGTON MEDICIAL CENTER CYCLOTRON THROUGHOUT THE PANDEMIC

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Introduction

The University of Washington Medical Cyclotron Facility (UWMCF) consists of a Scanditronix MC-50 cyclotron, four beam lines, three vaults, and two isotope production stations. Primary functions include isotope production, oncology patient radiotherapy, radiation effects/hardness testing, FLASH radiotherapy research, and neutron activation PET imaging.

Description of the Work or Project

Radionuclides are used across a wide range of industries and scientific disciplines including nuclear medicine, gas and oil exploration, nuclear forensics, etc. The Scanditronix MC-50 cyclotron at the University of Washington (UW) is recognized as a valuable resource for radionuclide production and research due to its high (50-MeV) beam energy and capability of accelerating multiple charged particles (i.e., H^+, H_2^+, D^+, $^3He^+$, $^3He^{2+}$ and $^4He^{2+}$).

The UWMCF is a member of the DOE University Isotope Network. We have been providing ^{211}At and high specific activity ^{117m}Sn to investigators through the National Isotope Development Center (NIDC) for preclinical and clinical research. In addition to routine production of ^{211}At and ^{117m}Sn, an active research and development effort has been focused on improving the targetry and purification processes of ^{211}At, ^{117m}Sn and ^{186}Re to achieve higher yields and purity.

The unique capability of the cyclotron has enabled several DOE Isotope Program funded research projects that are evaluating atypical production routes for radionuclides. |Collaboration with researchers at the University of Missouri, Brookhaven, Argonne, Los Alamos and Oak Ridge National Laboratories are ongoing for novel radionuclides of high interest to the community. We are currently developing new targets and methods to produce high specific activity ^{186}Re, ^{189}Re, ^{72}Se/^{72}As, ^{85}Sr, ^{155}Tb and ^{230}U/^{226}Th at UW through these collaborative research projects.

Recent upgrades to the facility have included a 7-port switching magnet, a larger quadrpole triplet, D-Pace Unibeam25 scanners, 4 sector split collimators, 3D FDM/FFF printing, and a compact isotope target station with auto load/retrieval.

Conclusions

This presentation will provide an overview of our cyclotron and radiochemistry facilities. We will highlight the ongoing collaborative research projects directed toward providing radionuclides in short supply.

OPTIMIZING THE ENCAPSULATION OF RADIOISOTOPES IN POLY(LACTIC-*CO*-GLYCOLIC ACID) NANOPARTICLES

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Introduction

Delivery strategies for α-emitting radioisotopes in targeted alpha therapy are based on self-targeting or the use of radioimmunoconjugates.[1] Effective targeting is achieved with these methods; however, improvements in retaining radioisotopes and decay daughters at the tumor site are required to maximize therapeutic outcomes and minimize side effects. Nanoparticles (NPs) have shown promising encapsulation and retention of α-emitting radioisotopes and their decay daughters.[2] Poly(lactic-*co*-glycolic acid) (PLGA) NPs have been widely used as delivery platforms for a variety of anticancer agents.[3] PLGA NPs offer a biodegradable and biocompatible delivery vehicle that can be engineered to improve administration and targeting and to minimize the relocation of radioisotopes from the tumor site. PLGA NPs also have the potential to selectively deliver ^{223}Ra, which does not form stable radioimmunoconjugates in vivo, and to serve as a platform for theranostic agents.

Description of the Work or Project

PLGA NPs were synthesized by nanoprecipitation and emulsion methods to enhance the encapsulation of unchelated and chelated metal ions. Synthesis conditions including chelator hydrophobicity, payload solution, and PLGA concentration were optimized for each metal ion. PLGA NPs with a mean hydrodynamic diameter of <200 nm show high colloidal stability in biologically relevant media over time. Chelation of ^{225}Ac to a lipophilic ligand or encapsulating unchelated ^{225}Ac in a methanolic solution increased its encapsulation efficiency within PLGA NPs. Release of ^{225}Ac from PLGA NPs in phosphate-buffered saline decreased significantly when using ^{225}Ac chelated to a lipophilic ligand. Although similar attempts were made to encapsulate ^{223}Ra within PLGA NPs, the encapsulation efficiency was <10%. Functionalization of PLGA NPs included a fluorophore and polyethylene glycol groups to assess their biological fate and minimize their recognition by the reticuloendothelial system, respectively. Radio-PLGA NPs demonstrate enhanced cytotoxic effects in cancer cells.

Conclusions

PLGA NPs have potential as delivery platforms for α-emitting radioisotopes ^{223}Ra and ^{225}Ac and as theranostic agents in cancer therapy. Synthesis parameters significantly influenced the encapsulation efficiency of radioisotopes which were further optimized for cancer cytotoxicity applications. Future work will focus on conjugation of targeting vectors to PLGA NPs and co-encapsulation of chemotherapeutic agents.

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EFFECTS OF ^{225}Ac ON OVARIAN CANCER CELLS

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Introduction

Ovarian cancer is the fifth leading cause of cancer deaths among women, with >70% of cancers progressing to metastatic disease and a 5-year survival rate of 20–30%.[1] Confounding treatment of this type of cancer is the rise in drug-resistant disease. Targeted alpha therapy has the potential to overcome issues of chemotherapy resistance by directly delivering α-emitting radionuclides to cancer cells. The high-energy deposition and short pathlength of the alpha particle emission induces double-strand DNA breaks that are more difficult for the cell to repair, resulting in cancer cell death. This study evaluates the effects on ovarian cancer cells of ^{225}Ac, an α-emitting radionuclide with a half-life of 9.92 days that has alpha-emitting daughters in its decay chain, including ^{221}Fr ($t_{1/2}$ = 4.8 min) and ^{213}Bi ($t_{1/2}$ = 45.6 min).[2]

Description of the Work or Project

Patient-derived ovarian cancer cell lines designated CK6869 and CK8270 were obtained from the National Cancer Institute. Initial experiments evaluated the cytotoxic effect of exposure to ^{225}Ac at different concentrations ranging from ~30 nCi to 10 μCi over a 9-day period. Cell viability was measured using an Alamar Blue assay and was also assessed using brightfield microscopy in a Cytation 1 cell imaging and multimode reader. RNA was collected from cells prior to treatment and from surviving CK6869 cells posttreatment for subsequent RNA sequencing and data analysis, to elucidate potential protein response or DNA damage and repair pathways that are activated in the presence of ^{225}Ac in surviving cells. In parallel, the ability of these cells to form spheroids was assessed and their different properties and responses evaluated.

Conclusions

The two patient-derived ovarian cancer cell lines exhibited significantly different responses to ^{225}Ac. CK6869 cells showed a greater sensitivity to radiation emitted during the decay of ^{225}Ac at 24 h, 48 h, and 72 h compared to CK8270 cells, with an approximate tenfold difference in response. Morphological examination of the cells confirmed this effect with a significant detachment of CK6869 cells after exposure to 1 μCi of ^{225}Ac, while CK8270 cells maintained a confluent monolayer. RNA sequencing results will be discussed, in the context of targeted delivery of ^{225}Ac to these cell lines.

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BUILDING PET RADIOCHEMISTRY SUITE IN A HOSPITAL: RADIOPHARMACEUTICAL SCIENTIST'S POINT OF VIEW AND LESSONS LEARNED

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Introduction

PET cyclotron radiochemistry facility is a prerequisite for making short-lived radiopharmaceuticals for research or clinical imaging. With increase in the number of clinical PET scanners and with ever-growing interest in innovative diagnostic radiopharmaceuticals, hospitals install their own cyclotrons, what was seen earlier as a prerogative of universities and research institutions. This paper will discuss how hospitals can approach the challenge.

Description of the Work and Results

Construction of a PET radiochemistry suite is a complex project and requires specialized knowledge. Each cyclotron facility is unique as it is built for a specific purpose. Therefore, the end user's requirements are very important and must be considered from the beginning of the project, equally with relevant guidelines and regulations. A number of soft guidelines exists, including those from the Canadian Nuclear Safety Commission and Health Canada. These guidelines, however, are not specific to the production of PET radiopharmaceuticals with small medical cyclotrons, making experience and expertise in the field invaluable for interpretation. There are several publications from the International Atomic Energy Agency that address the facilities for production of short-lived positron-emitters, but these guidelines are often overlooked by construction professionals as not locally enforced. Radiochemists / radiopharmaceutical scientists play a crucial role in analyzing these guidelines and can help to design a facility according to its intended use when involved very early. The following steps should be undertaken: 1) assembly of a team of experts with previous experience at PET cyclotron sites; 2) identification of the stakeholders and evaluation of their needs (strategic vision, choice of isotopes), which will become the basis for formal user requirements; 3) identification of potential equipment and collection of technical documentation; 4) preliminary conceptual design of the laboratory space by an experienced specialized company (cyclotron suppliers may offer such service), including preliminary but realistic cost estimation (sufficient space, properly sized ventilation, suitable finishes, equipment customization). The laboratories at CHU de Québec-Université Laval will include a cyclotron, a GMP area, research laboratories, and a dedicated laboratory for the preparation of investigational radiopharmaceuticals. Each laboratory type and equipment therein must satisfy a set of distinct specific requirements, often quite opposite: while research environment seeks to be as flexible as possible, GMP-type laboratories are preferentially rigid. The facility design was devised with an attempt to marry all the requirements and to create efficient workflows during operations.

Conclusions

Participation of radiopharmaceutical scientists in the facility design is indispensable. Effective communication is key in the process. The community could benefit from national guidelines specific to the design of facilities where short-lived PET radioisotopes are produced.

Orano Stable Isotopes Laboratory: A new occidental supplier of stable isotopes

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Introduction

Orano is leveraging on its unique competencies and technologies in chemistry and enrichment to produce stable isotopes for the research, medical and industrial communities.

Description of the Project

In 2018, Orano launched a new activity for producing stable isotopes based on its best-in-class separation and chemistry technologies and unique knowhow. The Stable Isotopes activity of Orano Chemistry & Enrichment has all assets to be a stable isotopes separation benchmark player in the Western world:

- a state-of-the-art isotopic separation technology and among others for stable isotopes: ETC, co-owned by Orano;
- A thorough expertise in fluoride chemistry widely used for stable isotopes production;
- Proven technologies for downstream production including oxides and metals;
- An industrial platform located in Tricastin in the South of France, a recognized center of expertise with all the necessary infrastructure to host this new activity.

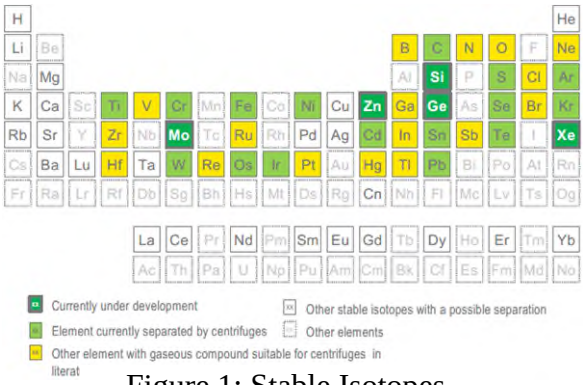

Figure 1: Stable Isotopes

Around 20 elements can be enriched or depleted by centrifugation. First elements are being developed by Orano and will be produced as of 2023 with a high grade of purity for leading-edge applications such as medical imagery, oncology, advanced research…. Based on customized design through the use of flexible cascades, the project will continuously develop new elements to serve its customers' needs.

This first phase of the project is well advanced: civil works have been completed in October 2021, the heart of the process and the key equipment are being installed for a first commercial production based on centrifugation technology in 2023, on time and budget. Thanks to its modular concept, production capacities will be increase according to market needs.

Orano is also studying the opportunity to launch the enrichment of other heavy stable isotopes such as lanthanides.

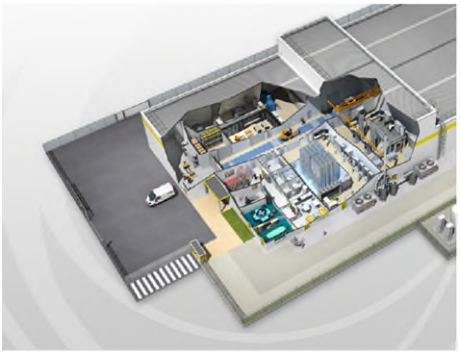

Figure 2 : Orano Stable Isotopes Laboratory

Conclusions

Orano Stable Isotopes on a proven technology and unique knowhows, is entering the production phase. This expansion of Western enrichment capabilities is a timely answer to the growing demand of OECD needs.

SAFEGUARDING CANADIAN LEADERSHIP: THE IMPORTANCE OF DIVERSITY AND REDUNDANCY IN MITIGATING MEDICAL ISOTOPE SHORTAGES IN CANADA'S ISOTOPE ECOSYSTEM

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Introduction

Canada's Isotope Ecosystem has been an important piece of the global supply chain for several medical isotopes for decades. Recently, Canadian companies are expanding their operations to produce emerging medical isotopes, offering new treatment options to patients at home. Yet, patients in Canada are still vulnerable to global supply shortages, which reveals the need to pursue redundancy and source diversity at the global level.

Description of the Work or Project

This paper will explore Canada's powerful Isotope Ecosystem by discussing the leading companies and organizations in the isotope community, as well as Canada's history as an isotope producer. Canada's Isotope Ecosystem boasts a network of producers, including a reactor fleet, cyclotron network, and research reactors, and the supply chain companies that provide specialized equipment to manipulate and transport medical isotopes. Canada's role in the supply for medical isotopes is therefore essential to modern medicine and the treatment of illnesses like cancer. However, when demand for an isotope surpasses the global production capacity, Canadian patients can be left vulnerable.

Conclusions

Canada's modern healthcare system relies on a diversified global supply chain network for isotopes and a ready supply of isotopes domestically. Global isotope shortages illustrate the importance of source diversity for patient supply and expose Canada's susceptibility without certain domestic sources. While Canada continues to play a leadership role in the global isotope community, we must pursue a collaborative, multi-stakeholder approach to achieve adequate redundancy and source diversity to mitigate future shortages.

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Keywords

Nuclear Imaging and Medicine, Production of Isotopes, Radiopharmacy, Radiotherapy, Theranostic

QUALITY CONTROL OF A NEW PORPHYRIN COMPLEX LABELED WITH ^{45}Ti

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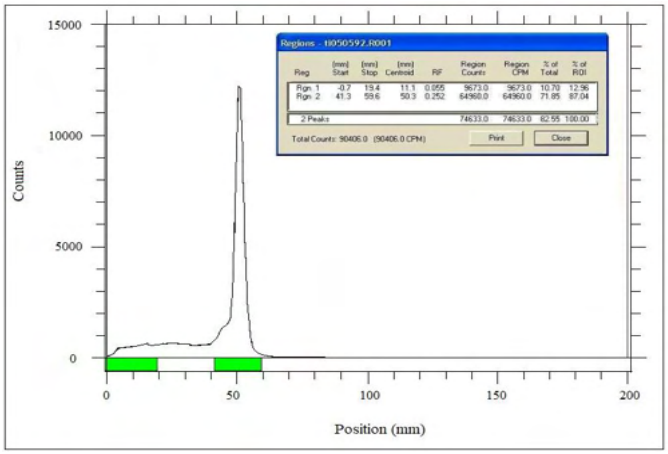
Introduction: The potential of accumulation in many types of cancer cells and noninvasive detection of cancer, mimicking other chemicals and combination with other treatments like photodynamic therapy and radiation therapy are interesting properties of porphyrins in cancer medicine and photodynamic therapy. ^{45}Ti is one of the most favorable radioisotopes for PET imaging. The purpose of this study is evaluation of new porphyrin complex labeled with ^{45}Ti. **Description of the work:** The cross-section of ^{45}Sc(p,n)^{45}Ti was investigated by TALYS-1.6 and the optimal target thickness and theoretical yield were calculated by SRIM code. ^{45}Ti was produced according to the work conducted by Fazaeli *et al.*[1]. The purified ^{45}Ti was labeled with the anticancer agent of tetrakis (pentafluorophenyl) porphyrin (TFPP). The radiochemical purity and the percentage of labeling were evaluated by radiation layer chromatography then the division coefficient of [^{45}Ti]-TFPP was calculated. The dual coincidence imaging system was used for imaging 1 and 2 hours after injection [^{45}Ti]-TFPP to rats. Immediately after imaging, the mean percent injected dose per gram and specific activity of different tissues including blood, heart, lungs, stomach, liver, bone, kidney, spleen, intestine, muscle, feces, and skin were measured. **Results:** The yield of 45Ti production was measured 468 MBq/µAh and the labeling rate was observed more than 98%. The highest activity was observed in the liver (%ID/g=2.27%, 1 h) and spleen (2.2%, 1 h), respectively, because of the high lipophilic of 45Ti-TFPP. SPECT images showed a significant uptake of radiopharmaceuticals in the abdomen. **Conclusions:** The labeling rate of 45Ti-TFPP was high and this compound has the potential for clinical application in different ways than PSMA, it can be joined with photodynamic therapy [2]. **Keywords**: Proton therapy, Proton Boron Fusion Therapy [PBFT], Boron, Brain, cancer, MCNPX, Bragg-peak

Figure 1: The paper chromatogram of 45Ti-TFPP by using a mixture of methanol and ammonium acetate 10% (1:1) on the Whatman no. 2 papers.



Reference

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AUGER ELECTRONS EMITTED FROM ^{119}Sb IN CANCER TREATMENT

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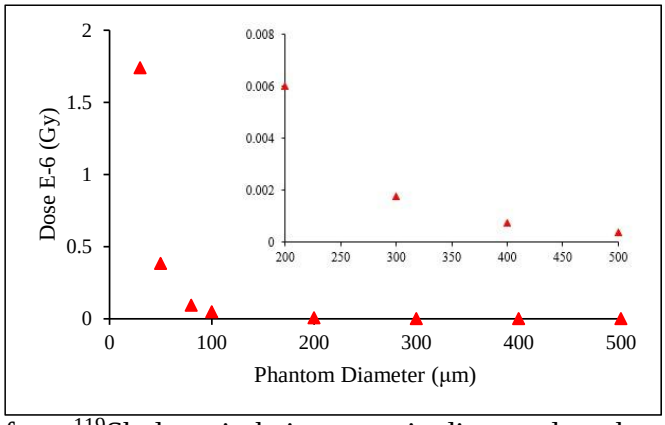
Introduction: High radiation dose delivery with high radiotoxicity at short distances, and minimizing toxicity for long distances are the opportunities to improve the radiation therapy by Auger emitters. The desirable reaction to ^{119}Sb production in low energy cyclotrons is ^{119}Sn(p,n)^{119}Sb with a half-life of $T_{1/2}$=38.19 h of ^{119}Sb. Most of the Auger electron energy emitted from ^{119}Sb deposits at distance less than 80 µm and the absorbed dose per Auger electron emitted from ^{119}Sb are significant.

Description of the work: The purpose of this study is dose evaluation induced by Auger electrons emitted from ^{119}Sb in a tumor and surrounding healthy tissues. Dose evaluation was done by using MCNP code in two nested cylinders as tumor and healthy tissue (thyroid follicles) with different diameters of tumor including 30, 50, 80, 100, 200, 300, 400, and 500 µm and a point source of electrons emitted from ^{119}Sb was assumed in the center of cylinders and the probability of electrons at different energies was inserted in the input of MCNP code. To validate the used method, the dose induced by emitted electrons from ^{131}I, ^{132}I, ^{133}I, and ^{134}I was calculated and compared with the work done by Campos *et al*. The results obtained from simulations of ^{131}I, ^{132}I, ^{133}I, and ^{134}I were matched with the similar work conducted by Campos *et al*. Auger electrons emitted from ^{119}Sb deposit their energy in distance less than 100 µm. The maximum absorbed dose per emitted electron was 1.74×10^{-6} with a tumor diameter of 30 µm and this value is significant in cell damages. By increasing the diameter of the phantom from 30 µm to 50 µm, the absorbed dose decreased by 78%.

Figure 1: The absorbed dose per particle induced by emitted electrons from 119Sb in phantoms with different diameters including 30, 50, 80, 100, 200, 300, 400, and 500 µm.



Conclusions: The dose induced by Auger electrons emitted from ^{119}Sb should not be ignored in dose calculations especially at short distances from the decay point. The effect of Auger electrons is significant especially at subcellular distances close to the decay region and it's an advantage for an accurate target therapy.

Keywords: Auger electron, ^{119}Sb, MCNP, Antimony-119, Radionuclide therapy

References

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STANDARDIZATION OF I-123 BY COINCIDENCE COUNTING

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Introduction

Iodine-123 is a short-lived radioisotope of the element iodine with a half-life of approx. 13.2 h. It decays predominantly to the first excited state of ^{123}Te by electron capture. The following deexcitation to the ground state gives rise to a gamma ray with energy of 159 keV. Iodine-123 is typically produced in medical cyclotrons and commonly used in nuclear medicine in combination with single photon emission computer tomography (SPECT). Depending on their chemistry, ^{123}I-labelled imaging agents can be used e.g., in thyroid cancer scans, to monitor the adrenergic status of the heart, or to study Parkinson's syndrome.

Description

In this work, a successful activity standardization of ^{123}I is reported, which was carried out as part of an international intercomparison campaign for medical isotopes involving the Transfer Instrument of the International Reference System (denoted as "SIRTI") of the Bureau International des Poids et Mesures (BIPM). The absolute activity determination was performed using the digital coincidence counting technique, realized in two different custom-built experimental configurations:
The first measurement setup consisted of a gas flow proportional counter (PC) for the detection of Auger electron, X-ray and conversion electron emissions following the decay of ^{123}I, while the gamma rays from the deexcitation of ^{123}Te were registered by two NaI inorganic scintillators. The second setup used a liquid scintillation (LS) counter (equipped with 3 photomultiplier tubes) in place of the proportional counter, which was complemented by an external $CeBr_3$ scintillator for the detection of gamma rays.
The measurements were performed using an aqueous solution of ^{123}I. For $4\pi\beta$(PC)-γ counting, the samples were produced by quantitative drop deposition on thin VYNS films to minimize self-absorption. Some of the samples were then covered by a varying number of absorber foils to change the detection efficiency in the proportional counter. In case of the $4\pi\beta$(LS)-γ setup, samples were prepared in standard high-density polyethylene (HDPE) vials. In this case, the efficiency was varied by chemical quenching of the LS samples and by the use of grey filters.
In both setups, a digitizer-based data acquisition was used, which recorded data from each detector independently, storing timing and energy information for each event in list-mode format. The coincidences were then evaluated during an offline analysis. The activity was determined by the efficiency extrapolation method.
The results from both setups were in excellent agreement with each other. The overall uncertainty for both activity determinations was less than 0.4%.

Conclusions

Primary standardization of medical isotopes is a challenging task due to their short half-lives and their requirement for intricate measurement methods. With the presented work here, the standardization of the versatile diagnostic agent ^{123}I was successfully carried out and an international traceability link was established.

PRECISION RADIONUCLIDE THERAPY FOR TREATING VETERINARY CANCEROUS TUMORS AND OSTEOARTHRITIS

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Introduction

Precision Radionuclide Therapy is an emerging class of treatment for arthritis and cancer. This presentation describes two specific examples that are currently being implemented in veterinary science: IsoPet® for tumor therapy using Y-90 and Synovetin OA® for osteoarthritis using Sn-117m.

Description of the Work or Project

Each therapy will be described in detail and example case studies presented.

IsoPet®

Injectable Y-90-IsoPet® comprises an insoluble Y-90-yttrium-phosphate radiation source mixed within an injectable, thermally reversible, temperature-sensitive polymer solution. After injection, the mixture gels within tumor extracellular spaces as it warms to body temperature to ensure that the Y-90 microparticles remain in tumors. Single-dose (300 to 400 Gy) treatment of tumors with high therapeutic ratio. Injection materials remain in the tumor and radioactivity is not excreted. IsoPet therapy has treated feline, canine and equine tumors. Three case studies will be reviewed.

Synovetin OA®

Synovetin OA® is a commercially available treatment in the USA for canine osteoarthritis. It is composed of insoluble Sn-117m hydroxide microparticles suspended in a saline solution. Intra-articular injection into the affected joint results in rapid assimilation into the inflamed tissue via phagocytosis by synovial macrophages. There is no leakage or systemic distribution away from the treated joints. Immediate release of dogs is achieved by owners conforming to straightforward handling restrictions based on the external radiation doses measured at release. Rat and canine trials [2] were undertaken in order to launch the veterinary medical device commercially. Sample case studies will be reviewed.

Conclusions

Precision Radionuclide therapy is a new promising therapy class for treating arthritis and cancerous tumors in veterinary science. Commercial products are now emerging for use in this field.

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ACTIVITY STANDARDIZATION OF CU-64 BY LIQUID SINCTILLATION AND COINCIDENCE COUNTING

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Introduction

Copper-64 is a rather unique short lived (12.7 h) radionuclide that comprises beta plus, beta minus and two electron capture decay branches. The radionuclide is of great interest for nuclear medicine due to its possible theragnostic applications. However, the special decay properties make precise activity determinations rather challenging. Here, we report on a recent activity standardization of ^{64}Cu using improved techniques as well as new approaches.

Description

In 2010, we reported on our first activity standardization of ^{64}Cu. At that time, we applied the liquid-scintillation-based CIEMAT/NIST efficiency tracing method and 4πβ(PC)-γ coincidence counting as primary methods. These methods were adapted in this work with various improvements: In the case of the CIEMAT/NIST method, we use updated decay data and a more accurate beta minus spectrum that takes the atomic exchange effect into account. Moreover, the conventional analog data acquisition of the 4πβ(PC)-γ coincidence counting system was replaced with a modern digitizer which records timestamps and energy data of each detector independently. The obtained list-mode data can then be used for offline analyses.

The above-mentioned methods were complemented by TDCR (triple-to-double coincidence ratio) liquid scintillation measurements. We show that this technique, which failed in the past due to outdated and inaccurate decay data, now yields good results. However, some efficiency ranges should be avoided, since the dependence of the counting efficiencies on the TDCR parameter or on the branching ratio can be very large. Thus, we propose a new hybrid method that overcomes such a high sensitivity. The hybrid method makes use of accurately measured data from a TDCR counter which are combined with quench indicating parameters obtained from a commercial liquid scintillation counter.

In addition, the 4πβ(LS)-γ coincidence counting was applied with a new setup that used a complete TDCR system rather than a proportional counter. Again, the data acquisition was carried out using a digitizer-based approach.

All of the above methods agree within their standard uncertainties and a combined activity concentration was obtained with a relative standard uncertainty of 0.5%.

The result was used to calibrate ionization chambers which are important tools for secondary activity standardization. Moreover, the activity standardization was used to participate in an international comparison using the Transfer Instrument of the International Reference System (denoted as “SIRTI”) of the Bureau International des Poids et Mesures (BIPM).

Conclusions

New developments in the activity determination of ^{64}Cu are presented and discussed.

References

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Sn-117m IN RADIOSYNOVIORTHESIS FOR CANINE OSTEOARTHRITIS: LEARNINGS FROM 2 YEARS OF COMMERCIALIZATION

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Introduction

After years of research and development, therapeutic use of radioisotopes for osteoarthritis in veterinary medicine has been implemented as a commercial enterprise. This presentation describes learnings from the first two years of commercialization, with emphasis on manufacturing, formulation, customer and pet owner education and communication.

Description of the Work or Project

In the US market, a Sn-117m colloidal suspension for intra-articular injection has now been available for over two years. This novel therapeutic device reduces joint inflammation by triggering the death of the inflammatory cells and their products which are largely responsible for the pain and tissue degeneration associated with osteoarthritis. Despite its promise, the challenges of commercialization and growth are many and varied.

The challenges of commercialization are generally related to scale. Ensuring adequate capability to manufacture product requires continuous process evaluation and improvement, as well as cultivation of multiple manufacturing and formulation sites. Expanding production volumes also forces consideration of radiation doses received by manufacturing personnel and appropriate mitigation measures. Field use of Sn-117m has provided a testing and proving ground for improvements in formulation, which have resulted in improvements in the patient and veterinary user experience.

Since the launch of the commercial product, the developers have been working to overcome the additional hurdle of convincing veterinarians that they should invest in licensing, training, and unfamiliar equipment and processes with the expectation that long-term patient benefits justify the investment. Most veterinarians had no prior experience or exposure to radionuclide therapy, making education and training a critical component of commercialization. In addition to educating veterinary professionals, they must be provided appropriate tools to educate pet owners on expectations and post-treatment contact limitations.

Conclusions

Sn-117m is a viable commercial addition to traditional therapies for canine osteoarthritis. Manufacturing and formulating require continuous evaluation and improvement to produce a quality product to scale. Successful adoption is largely dependent on thorough education of the veterinary professional and staff, as well as effective tools to educate pet owners.

References

[1] Lattimer J.C. *et al.,* Intraarticular injection of a Tin-117m radiosynoviorthesis agent in normal canine elbows causes no adverse effects, *Vet Radiol. Ultrasound.* 2019;60:567–574.

IMPROVING RADIOISOTOPE PRODUCTION AND PURIFICATION IN SMALL RESEARCH REACTORS: THE SZILARD-CHALMERS PROCESS

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Introduction

Szilard-Chalmers reactions using high surface area materials are being investigated at the Penn State Breazeale Reactor to develop higher specific activity isotopes for small research reactor applications. Emerging materials such as metal-organic frameworks (MOFs) and nanopowders with high surface areas offer a potential opportunity for increasing the specific activity in Szilard-Chalmers experiments, as the bulk of the captured recoil atoms are from the material surface. The effects of irradiation time (indicative of neutron and gamma ray influence), decay time, target quantity, capture matrix, and separation procedure on enrichment factor are being studied via experimental irradiations.

Description of the Work

Szilard-Chalmers target/capture matrix combinations are optimized based on solubility, filter performance, and gamma ray sensitivity tests. Novel Cu MOF and nanopowder targets are irradiated and separated via filtration and centrifugation methods. Measures for the Szilard-Chalmers reaction, such as yield and enrichment factors, are determined for each experiment to identify the parameters most influential to the enrichment factor.

Results

Initial results suggested the use of centrifugal filters for separation and a capture matrix of DI water (pH 7) or ethanol to optimize the Szilard-Chalmers capture of ^{64}Cu. Figure 1 reveals that smaller target quantities, neutral pH capture matrices, and shorter times in between irradiation and processing achieve higher yield and enrichment factors. The MOF and nanopowder materials achieved their highest enrichment factors of 26 and 2.2, respectively.

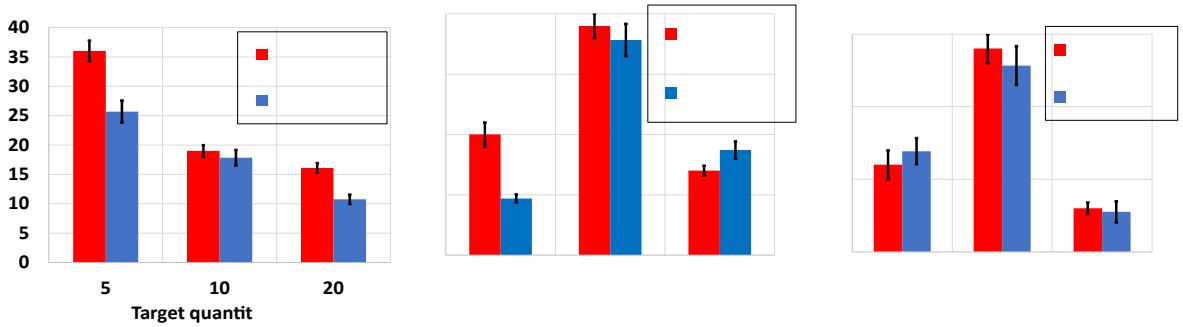

Figure 1. Effects of various testing parameters on MOF yield and enrichment.

Conclusions

Initial irradiations suggest that MOFs may serve as better Szilard-Chalmers targets than nanopowders for production of the ^{64}Cu isotope. Smaller target mass, shorter decay times, and a neutral water capture matrix have shown to be a few of the main drivers needed for increasing enrichment factors. Further optimization is currently being conducted to understand the effect of irradiation time, cooling time, and alternative capture matrices.

A NEW FAMILY OF HIGH-CURRENT CYCLOTRONS

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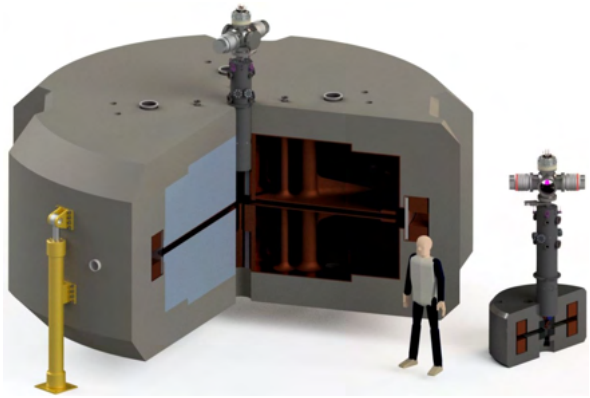
Introduction

We have developed a new family of compact cyclotrons capable of 10 mA of protons at energies up to 60 MeV. Designed for the IsoDAR neutrino experiment[1], the *x10* current increase could find important applications in isotope production, for products with long half-lives (^{68}Ge) or low cross sections (^{232}Th(p,X)^{225}Ac).

Achieving and Using High Currents

Beam current limits on today's isotope cyclotrons arise from extraction foil lifetime and central region erosion from inefficient injection. Our cyclotrons accelerate H_2^+ ions, reducing the effect of space charge, and bunch 90% of the beam into the RF acceptance window with an RFQ. A beam-dynamics effect discovered at PSI called "vortex motion" translates space charge forces in the cyclotron magnetic field into lateral motion that stabilizes the individual RF beam packets into compact bunches. Halo is generated but is collimated in the first turns. Stable packets are formed in about the first few MeV (10-15 turns), allowing for adequate turn separation to enable clean septum extraction of the H_2^+ ions without use of a stripper foil. Using these principles we can build cyclotrons for energies ranging from a few MeV up to 60 MeV. Turn separation above 60 MeV is lower, but use of structure resonances may enable extending the energy to higher values. Q/A of H_2^+ is 0.5, so cyclotrons are suitable for D^+, He^{++}, C^{6+} ions at the same energy/nucleon. At present, the H_2^+ ion source and RFQ are ready, the 1.5 MeV Demonstrator is being built, the IsoDAR experiment will be deployed in 5 years.



60 MeV IsoDAR and 1.5 MeV Demonstrator cyclotrons. They employ identical injectors.

x10 current means *x10* beam power. Developing high-power targets will enable the most efficient utilization of the available beam. In the interim, splitting the beam onto as many as 10 target stations is feasible, by employing either RF kicking of bunches into separate beam lines, and/or by insertion of stripper foils into the edges of the extracted H_2^+ beam to peel off adjustable amounts of protons using a dipole right after the stripper.

Conclusions

Our designs represent a paradigm shift in cyclotron performance, shattering the beam-current barrier, and opening the door for novel applications. In addition to isotope production, these machines are compact high-flux MeV-range neutron generators; planned uses extend from neutrino production to cost-effective IFMIF-style fusion reactor materials testing platforms.

References

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RADIATION DOSE ASSESSMENT OF THORIUM-CONTAINING GAS MANTLE IN CONSIDERATION OF USAGE STATUS IN REPUBLIC OF KOREA

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Introduction

The IAEA recommends various considerations for specific exemption of consumer products containing radioactive isotopes. One of which is that the results of the dose assessment for all predictable scenarios arising from the use of the products should meet the criteria of 10 μSv/y for general cases and 1 mSv/y for low-probability scenarios. It is necessary to regularly review justification of the radiation regulation exemption considering the usage of consumer products and the latest technology status. In this study, we performed the radiation dose assessment on the exposure scenarios according to the life cycle of the thorium-containing gas mantles in consideration of the usage status in Republic of Korea.

Description of the Work or Project

The gas mantles are used as a wick for gas lanterns, and the maximum mass of the product currently produced is 3.5 g. According to Nuclear Safety Act in Republic of Korea, the maximum radioactivity of thorium in the gas mantle exempted from regulation is 700 Bq/g. Therefore, it was assumed that the radioactivity per unit product of the gas mantle is 2,450 Bq. We evaluated the radiation doses for all predictable exposure scenarios when using the gas mantles (i.e., transport, distribution, use, accident, and disposal). The majority of the gas mantles exceeding the generic exemption criteria are imported from overseas, so in the case of the transport scenarios, we assumed that gas mantles are distributed from the Incheon Import Logistics Center to retail stores via the parcel distribution centers. In addition, classification at the parcel distribution centers and display at the retail stores were considered as distribution scenarios. Currently, the gas lanterns are mostly used for camping in Republic of Korea; hence, we developed use scenarios related to camping only. In the case of accident scenarios, not only traffic accidents and fires at warehouses and houses, but also cases in which children handle a used mantle at campground were considered. Finally, the gas mantles are discarded as general waste, and the disposal methods of general waste are classified as incineration and landfill. So, the incineration and landfill are considered as disposal scenarios.

Conclusions

Although we assumed the maximum activity of the gas mantle for conservative evaluation, the results of the dose assessment for all scenarios under considerations were far below the IAEA recommendations (i.e., 10 μSv/y and 1 mSv/y). Our next step is to conduct dose assessment for other consumer products containing radioactive isotopes.

Keywords: Consumer product, Radioactive isotope, Gas mantle, Exposure scenario, Dose assessment

PRODUCTION OF ISOTOPICALLY ULTRAPURE (99.999-99.9999%) PLUTONIUM244 FOR ISOTOPE DILUTION MASS SPECTROMETRY

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Introduction

The long half-life and extremely low abundance of ^{244}Pu even in irradiated nuclear materials makes this isotope an ideal mass spectrometry spike for quantitative Pu analysis. Historically residual contamination of enriched ^{244}Pu with the more abundant and analytically important Pu isotopes (238-242) hindered some measurements. Applying a Nier-type approach[1] using analytical mass spectrometry, ^{244}Pu with an isotopic purity of 99.999?% has been produced on a scale sufficient to meet the tracer needs for nuclear forensics applications.

Description of the Work or Project

The performance of analytical mass spectrometers has improved dramatically in the last decades to the point where they can be practically employed as a preparative tool for isotope purification. While it cannot match Calutrons for material throughput, enrichment factors after a single pass are far greater. In niche reference material applications where quantity requirements are low but high isotopic purity is required, such as for the rare actinide tracers ^{236}Np and ^{244}Pu then this approach becomes practical. For tracered Pu analysis, typical ^{244}Pu spiking levels are 10-100 pg/sample, therefore microgram scale production can meet analytical demand. In this presentation we will describe the use of a radiologically contained quadrupole Inductively Coupled Plasma Mass Spectrometer (ICP-MS) operating with nanoamp range ion currents to yield useful quantities of ^{244}Pu with previously unequalled isotopic purity. While operating at low ion energies limits available ion current due to space-charge defocusing, there are some subtle benefits especially for recovering the final purified product. As the source materials containing ^{244}Pu are exceedingly difficult to produce[2], recovery and recycling of material lost within the mass spectrometer are addressed, along with analytical measurements confirming isotopic purity.

Conclusions

A novel approach for small scale highly selective isotopic purification is outlined for ^{244}Pu. Analytical measurements indicate isotopic purity of 99.999?% of one 0.55 µg batch. This method could be well suited to the production of a new ^{244}Pu reference material for nuclear forensics applications that easily meets the most demanding analytical requirements.

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PNNL -SA-181033

DEEP DETRITIATION OF HEAVY WATER FOR CANDU END OF LIFE

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Introduction

Have you ever thought about the amount of heavy water in a CANDU nuclear reactor and where it goes at the plant's end of life? The Pickering Nuclear Generation Station may cease commercial operation in the next few years, posing several nuclear waste challenges for large-scale decommissioning of CANDU plants in Canada. One of these challenges is how to safely dispose of large quantities of tritiated heavy water.

Description of the Project

Tritium is a radioactive isotope of hydrogen that accumulates in heavy water (deuterium oxide or D2O) as part of normal operation in CANDU nuclear power plants. Though existing Tritium Removal Facilities are effective for normal operations, they are limited in their ability to fully remove tritium. Without "deep" tritium removal, the heavy water used in CANDU nuclear power plants is ultimately destined for disposal as radioactive waste.

Deuterium, in contrast, is a stable (non-radioactive) isotope of hydrogen with a demand that is expected to grow rapidly over the next 10 years. The demand for clean heavy water is driven by the recent advancements in industrial and commercial products including new deuterated drugs, robust OLED screens, semi-conductors, and Plastic Optic Fibers (POF) that support high data transmission for 5G networks and autonomous vehicles.

The current barrier to commercializing waste D2O is the water's tritium concentration, which is more than six (6) orders of magnitude higher than the acceptable limit for the non-nuclear market.

Conclusions

Laurentis Energy Partners (LEP), a wholly-owned commercial subsidiary of OPG, is developing a new and innovative deep detritiation project that will use combined electrolysis catalytic exchange (CECE) technology, to process tritiated D2O and produce virgin-grade equivalent heavy water. This first-of-a-kind remediation project will support sustainability of the nuclear energy sector.

Chemical Form and Adsorption Behavior of Radioiodine I-135 on the Graphite Surface in the Primary Circuit of HTGR

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Introduction

Due to the abrasion and corrosion of spherical fuel elements and some structure materials which are made of graphite, radioactive dust will be generated in the primary circuit of pebble-bed type high temperature gas-cooled reactors (HTGRs). Parallel to the helium, the dust can serve a new route for the transport of radioactive nuclides. Thus, radioactive dust is of great significance in the radiation safety study of the pebble-bed type HTGR, which is a first stage of the very high temperature reactor (VHTR), one of the six generation fourth advanced nuclear reactors. To interpret the interaction between the radionuclide and the graphite will be essential to grasp the characteristics of the radioactive dust with various absorbed radioactive nuclide, especially I-135 make non-negligible contributions on the source term in the primary circuit. (Moormann, 2018)

Description of the Work or Project

In order to explain the interaction between the radionuclide and the graphite, it will be a prerequisite to figure out the chemical form of radionuclide in a typical nuclear system. Then the adsorption behavior of the radionuclide on the graphite can be studied under a given chemical form. Since there are few researches about the chemical form and adsorption behavior of radionuclide on the graphite surface in the primary circuit of HTGR, we chose the I-135 as a representative which is a typical nuclide in the nuclear reactor. We proposed a research framework to study the chemical form and adsorption behavior of radioiodine I-135 on the graphite surface by coupling the microscopic quantum chemistry method and macroscopic thermodynamics theory with statistical mechanics. Specially, we adopted Factsage software to determine the chemical form of I-135 in the primary circuit in a typical HTGR circumstance. Then the Gaussion and Shermo were used to study the adsorption behavior of radioiodine I-135 on the graphite surface. Moreover, we considered the recoil energy during the decay process of the radionuclide unlike the stable isotopes, which may make an effect on the chemical form of I-135 and subsequently the adsorption behavior between I-135 and the graphite. As a new attempt, we will try to compare the unique behavior of radionuclide compared to its stable isotope, which may shed a light on the study of fission products behavior in the advanced nuclear energy systems.

Keywords: Graphite, Chemical Form, Radioiodine, Adsorption

Conclusions

Chemical form of I-135 in the primary circuit is CsI from the calculation of FactSage. Calculation from Gaussian and Shermo will give details about chemical potential and gibbs free energy which can judge chemical forms of radioiodine on the graphite surface. Comparing with stable nuclides, recoil energy during the decay process make an effect on debonding molecular with radioiodine, which is unique.

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PRODUCTION CROSS SECTIONS FOR THE PROTON INDUCED REACTIONS ON NATURAL FE WITH PROTON ENERGY OF 57 MEV

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Introduction

The production cross section for the proton induced reactions on Fe is useful for understanding nuclear reaction mechanism, improving nuclear reaction models and producing radioisotopes. Iron is a basic structural material used in reactor and accelerator, and the proton induced activation cross sections are important for the facility shielding. In addition, it is used as a target material for calibration sources and medical isotopes such as Co-55, Co-56, Co-57, Mn-52 and Mn-54. Therefore, we measured the production cross section in the ^{nat}Fe(p,x) reactions using a proton energy of 57 MeV at the KOMAC facility, Gyeong-ju, Korea. The measured data was compared with other experimental data in EXFOR and TENDL library.

Experimental methods

A high purity (99.9%) of natural Fe (102 μm thickness) was used for the cross section measurement. The Ti (108 μm) and Al (100 μm) were prepared for the determination of proton beam flux and the energy degrader. A total of 70 foils was stacked and irradiated with a proton beam for 30 minutes. The proton linac was operated with a repetition rate of 1 Hz and a beam current of 100 nA.

The proton energy incident on each sample was calculated from the SRIM code. The induced activity of activated sample was measured using the HPGe detector and analyzed from Gamma-vision program. The detection efficiency and energy calibration were determined with a number of standard sources. During the activity measurement, the dead time was kept below 1% by placing the sample at an appropriate distance from the HPGe detector.

The production cross sections of the Co-55, Co-56, Co-57, Mn-52 and Mn-54 in the ^{nat}Fe(p,x) reactions were determined by using the well-known activation formula. The total uncertainty was estimated by the statistical error on the γ-ray counting and the systematic error related to nuclear spectroscopic data, the detection efficiency, the beam flux and so on. The measured data was compared with the experimental data in EXFOR and the evaluated nuclear data from the TENDL library.

Conclusions

The production cross sections of radionuclides produced from the proton induced reactions on iron were measured by the stacked-foil activation method with a proton energy of 57 MeV at KOMAC facility. Our results were agreed with other experimental data, but showed significant difference in comparison with the data from TENDL library.

CROSS SECTIONS OF α-PARTICLE-INDUCED REACTIONS ON ^{nat}Ta

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Introduction

Tantalum is a frequently used material in accelerator technology. Irradiating it with an alpha particle beam several longer-lived radionuclides can be produced. In literature several experimental studies are available reporting cross section data on different reactions. However, the reported data have significant differences in peak position and in amplitude therefore, we decided to perform an experiment to investigate the ^{nat}Ta(α,x) reactions and to deduce more reliable cross sections data of these reactions.

Description of the Work

The experiment was performed at the AVF cyclotron of RIKEN RI Beam Factory. Stacked-foil target activation method was used followed by high-resolution γ-ray spectrometry measurements. The target consisted of pure metallic foils of ^{nat}Ta (primary target) and ^{nat}Ti (served as catcher foils and also as monitor foils). The average foil thickness was determined from the lateral size and weight of the original foils and found to be 14.37 and 2.24 mg/cm^2 for the Ta and Ti foils respectively. The original foils were cut into a size of 10×10 mm and stacked into a target holder served as a Faraday cup. Every Ta foil was followed by a Ti catcher foil. The other Ti foils were inserted into the stack at certain positions for monitoring purposes, to get most information on the beam parameters and the energy loss of the beam in the target stack using the ^{nat}Ti(α,x)^{51}Cr monitor reaction. The irradiation lasted for 1 h, the initial beam energy was 50.5±0.2 MeV measured by time-of-flight method. Energy degradation in the target was calculated using stopping power data derived from the SRIM code. The average beam intensity measured on the Faraday cup was 203 nA.
The γ-ray spectra of the Ta and its Ti catcher foil pairs, as well as of the Ti monitor foils were measured by a high-resolution HPGe detector (ORTEC GEM-25185-P) and analyzed using dedicated software. The spectra of each foil were measured several times to follow the decay of the reaction products. To keep dead time low the detector-foil distance was adjusted.

Conclusions

Activation cross sections were deduced for Re, W, Ta, Hf and Lu reaction products. The collected data were compared with the available literature data and the TENDL-2021 theoretical prediction. Based on the newly measured cross section data and the gathered information the quality of the excitation functions was improved for several reactions and the available deviations of some of the earlier datasets were able to be explained. The provided new datasets may also contribute to improve the prediction capability of the theoretical model codes.

NANOCONSTRUCTS FOR RADIONUCLIDE DELIVERY – AN ORNL PERSPECTIVE

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Introduction

Self-targeting and radioimmunoconjugates are effective delivery strategies for α-emitting radionuclides in targeted alpha therapy.[1] Transmetalation, transchelation, and the bond-breaking recoil energy of radionuclides will decrease therapeutic efficacy and increase side effects. Additionally, there is a need to selectively target ^{223}Ra, which does not form stable radioimmunoconjugates in vivo. The development of nanoparticles (NPs) as radionuclide delivery platforms is a promising alternative to overcome these limitations. Promising encapsulation and retention of α-emitting radionuclides and their decay daughters has been demonstrated in vitro.[2] Further development is required to engineer NPs for enhanced administration and targeting, while minimizing the relocation of radionuclides from the tumor site.

Description of the Project

Our efforts have involved the design, development, and evaluation of inorganic and organic NPs. Lanthanide-based inorganic NPs can encapsulate different α-emitting radionuclides and partially retained their decay daughters in vitro. Understanding the interaction of radionuclides with lanthanide-based NPs is critical to optimize the encapsulation and retention of radionuclides. Improving the colloidal stability, biocompatibility, and targeting of lanthanide-based NPs is a key step towards the development of radiopharmaceuticals. Poly(lactic-co-glycolic acid) (PLGA) organic NPs are a promising biodegradable and biocompatible delivery vehicle that must be engineered to improve the administration, targeting, and retention of radionuclides. Optimization of synthesis conditions and NP structure can enhance the encapsulation efficiency and retention of radionuclides within PLGA NPs.

Conclusions

Lanthanide-based and PLGA NPs have potential as delivery platforms for α-emitting radionuclides. These NPs could be developed as a novel radiopharmaceutical therapy to treat cancer. A comprehensive understanding of the interaction of radionuclides with NPs and their biological effects will aid in their design and development as radiopharmaceuticals. An endeavor of such magnitude requires a multidisciplinary team with expertise in advanced characterization and modeling techniques.

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Keywords: Nanoparticles; Radionuclides; Radiopharmaceutical

WATER EXTRACTION OF TECHNETIUM-99m FROM NEUTRON IRRADIATED POROUS MOLYBDENUM DIOXIDE PELLET

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Introduction

Technetium-99m(^{99m}Tc) is the most widely used radionuclide for nuclear medicine diagnostic imaging procedures, almost all ^{99m}Tc used for this purpose is obtained from the radioactive decay of molybdenum-99(^{99}Mo). Currently, the dominant way for ^{99}Mo production is by fission reaction of high-enriched uranium in research reactors. However, due to the decommissioning, unplanned shutdown of nuclear reactors, and the principle of nuclear non-proliferation, it is necessary to look for a new production method to meet the demand of ^{99}Mo. ^{98}Mo used for the target to produce ^{99}Mo by (n, γ) reaction was proposed as one of the solutions, where the most commonly used target is composed of MoO_3. We have been developing a ^{99}Mo/^{99m}Tc generator production method using a MoO_2 target, which is insoluble in water. In the present study, the porous MoO_2 was irradiated and the extraction of ^{99m}Tc from MoO_2 was by using the difference in their water solubilities.

Experiment

The porous MoO_2 pellet was manufactured as an irradiation target and the Kyoto university reactor (KUR) was irradiated. ^{99m}Tc was leached from irradiated MoO_2 by pure water. Since MoO_2 is extremely insoluble, a solution of ^{99m}Tc with a low Mo/Tc ratio is expected to obtain. We evaluated the ^{99}Mo/^{99m}Tc ratio by γ-ray spectrometer, and the concentration of all stable isotopes of molybdenum was measured by ICP-MS for evaluation of the Mo/Tc ratio and ^{99}Mo/^{all}Mo ratio. In addition, we carried out the milking experiments of ^{99m}Tc every 24 hours, the milking experiments were repeated for 4 days.

Conclusions

We developed the novel production method of ^{99}Mo/^{99m}Tc generator by using (n, γ) reaction. This method is based on water extraction of ^{99m}Tc from an irradiated porous MoO_2 target. As a result, we obtained the (n,γ) production method of a ^{99}Mo/^{99m}Tc generator with a low Mo/Tc ratio.

Keywords,
^{99}Mo/^{99m}Tc generator, Mo target, (n,γ) reaction, low Mo/Tc ratio, molybdenum dioxide

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DESORPTION OF MERCURY FROM AN EXTRACTION CHROMATOGRAPHIC RESIN – EFFORTS TOWARDS THE ISOLATION OF MERCURY-197M/G FOR THERANOSTIC APPLICATIONS

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Introduction

Mercury-197(m,g) decays mainly by electron capture and has therapeutic potential in cancer care because of its Meitner-Auger and conversion electron emissions (1,2). This radioisotope can be produced via the ^{197}Au(p,n)^{197}Hg nuclear reaction on a low-energy cyclotron (< 15 MeV). Our recent work has suggested that on those chromatographic resins where mercury can be readily retained, further increasing acidity can aid its desorption (3). However, eluting mercury in high acid concentrations requires further treatment to reduce acidity for subsequent radiolabeling studies. This study sheds light on the recent progress we have made on eluting mercury from an extraction chromatographic (EXC) resin containing a tertiary amine (WBEC Resin) in neutral pH buffer solutions.

Description of the Work or Project

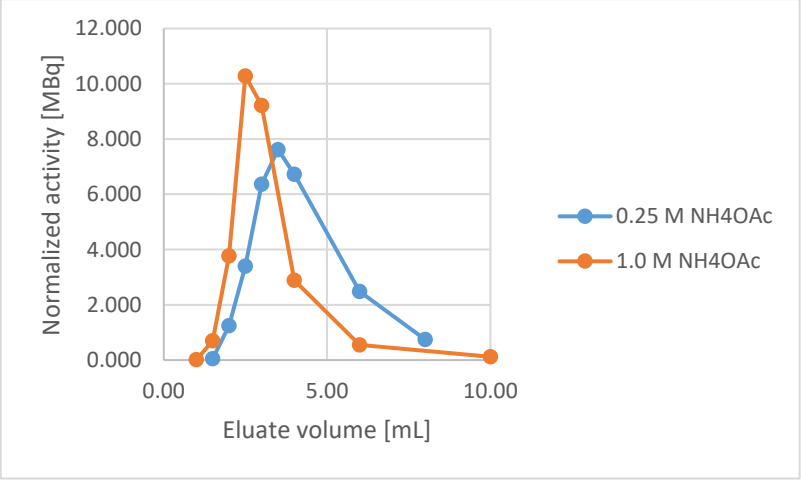

Figure 1: Elution curves of ^{197}Hg from WBEC column. Activity was normalized to per milliliter of eluate. Plot only shows volume of the eluate during the elution step.

$^{197m+g}$Hg was produced at the 13 MeV cyclotron (TR-13) at TRIUMF by proton irradiation of solid gold targets. The irradiated gold targets were dissolved in aqua regia and loaded onto a column packed with an EXC resin containing di(2-ethylhexyl)phosphoric acid (LN resin). Elution of mercury was achieved with 6 mol·L^{-1} HCl, while gold was retained on the column, judging by its bright yellow color (4). As mentioned in the introduction, such a high acid concentration cannot be applied during radiolabeling studies, thus the currently adopted separation procedure included three evaporation steps to reduce acidity. However, these steps are time-consuming (~ 8 h), and it is expected that by adopting

a second resin column, the overall amount of time spent on lowering acid contents is greatly shortened.

An aliquot of 0.5 mL of the eluate containing radiomercury in 6 M HCl was then loaded onto a column packed with WBEC resin. After loading the column was washed with 1.5 mL 0.1 M HCl. Elution of mercury was performed with 0.25 M and 1.0 M of ammonium acetate solutions. Eluted ^{197}Hg activity was measured by a dose calibrator. Obtained elution curves are depicted in figure 1. It should be mentioned that initial testing was performed with stable mercury samples at Eichrom, LLC.

During loading and washing steps, no noticeable activities were measured in the eluate. From the elution curves, it can be concluded that a higher concentration of ammonium acetate solution yielded a sharper elution curve. It should be noted that during both trials than 90 % of the activity introduced onto the column were recovered, despite the eluate volume being at least 16 times of the resin bed volume.

Conclusions

Our previous work suggests that decreasing acidity aids retention of mercury on certain extraction chromatographic resins. However, contrary to that trend, mercury elution from the WBEC resin was achieved with ammonium acetate solution, which is of neutral pH. Increasing the concentration of the eluting reagent not only yielded a sharper elution peak, but also contributed to a higher degree of mercury removal from the resin.

Acknowledgment

We would like to thank the TR-13 Cyclotron Operations Group consisting of Toni Epp, Ryley Morgan, Spencer Staiger, and led by David Prevost for regular irradiations of Au targets. We gratefully acknowledge funding from the NSERC Discovery program (VR, CH, CR), CCSRI, and CIHR. TRIUMF receives federal funding via a contribution agreement with the National Research Council of Canada. We also thank members of Reilly group at Leslie Dan Faculty of Pharmacy, University of Toronto for their collaboration.

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DEVELOPMENT STATUS OF RADIOISOTOPE POWER SYSTEM FOR SPACE MISSION IN KOREA

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Introduction

Radioisotope thermoelectric generator (RTG) is in the limelight as a power source for space mission owing to is long lifespan and high reliability. Korean government set the plan to launch a lunar lander in 2030, and establish a lunar base up to 2040. So, Korea Atomic Energy Research Institute (KAERI) has been developing RTG to supply heat and electricity to the lunar lander and rover during lunar night. In this study, 2 RTGs for satellite and lunar mission will be introduced.

Development of RTG

RTG converts the decay heat of radioisotope into electricity using thermoelectric materials. Components of RTG can be classified into radioisotope heater unit (RHU), thermoelectric converter[1], and assembly structure, and KAERI is developing all these technologies.
In the case of RHU, efforts are being made to secure a supply chain to secure Pu-238 and Am-241, which cannot be produced in Korea, through international cooperation. However, KAERI is developing a heat source protection technology using carbon composite to prevent leakage of radioisotope in spacecraft accidents and reentry accidents.
In order to generate maximum electric power under the specified heat source condition, KAERI also developed an optimal design program for thermoelectric module, and verified the program through prototypes.
Assembly structure should have an insulation structure to minimize heat loss of the radioisotope heat source. And it should be designed to protect the heat source and thermoelectric module from the vibration of the spacecraft and from the shock generated during fairing process. But, it should be designed to be lightweight.
KAERI started R&D on RTG in 2016 with the goal of supplying electric power to the lunar lander, and completed the development of an ETG, a simulated RTG, in 2021 and succeeded in space demonstration through Nuri (KSLV-II) satellite. The ETG satisfied all the environment test such as launch vibration, fairing shock, and thermal cycle at vacuum state. And, it has been operating normally since the first performance verification was succeeded in July 2022. From 2024, KAERI plans to start developing RTG for lunar lander using Pu-238, and technology to secure the safety of radioisotope heat source in spacecraft accident.

Conclusions

KAERI succeeded in demonstrating an ETG in low earth orbit. And the authors plan to develop an RTG to supply electricity at lunar lander in 2030. In order to ensure the safety of the RTG in spacecraft accidents, its safety and reliability test based on the UN safety framework will be conducted.

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A COMPARITIVE EVALUATION OF SELECT ALPHA-EMITTING RADIOISOTOPES AND THEIR FUTURE POTENTIAL IN RADIOPHARMACEUTICAL APPLICATIONS

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Introduction

With the recent radiopharmaceutical successes of Lutetium-177 (^{177}Lu) based radio-ligand therapy (RLT) demonstrated through the treatment of neuroendocrine tumors (NET) and metastatic castration-resistant prostate carcinoma (mCRPC), the importance of continuous high-quality radioisotope production and stable supply-chains is clearly identified for uninterrupted patient treatment.[1, 2] Alpha-emitters have shown significant promise towards the treatment of tumors refractory to beta-emitting isotopes used in RLT, but routine production methods to meet current and future global demands of alpha-emitting isotopes are still under development.[3]

Description of the Work or Project

This work provides an industry perspective from a radiopharmaceutical developer and radioisotope manufacturer to the current challenges of production of select alpha-emitting radioisotopes, namely Actinium-225 (^{225}Ac), Lead-212 (^{212}Pb) and Astatine-211 (^{211}At) with a focus on manufacturing methods capable of satisfying global demands and distribution requirements. An evaluation of the daughter nuclides of the respective alpha-emitters will also be critically analyzed with respect to short- and long-lasting radioisotopes and their potential dose-contribution including an assessment of the impact of the daughter nuclides of ^{225}Ac-labelled radiopharmaceuticals. Furthermore, a detailed comparison on ^{225}Ac production methods will be presented indicating the potential promises and pitfalls of each discussed method, highlighting the importance of dedicated ^{225}Ac production facilities and the significance of radioisotopic purity.

Conclusions

The global success of alpha-emitting radiopharmaceuticals for therapy will be dependent on the routine availability of alpha-emitting isotopes in large activities with insignificant levels of radioisotope impurities. ^{225}Ac offers promising potential due to its favorable decay characteristics and diverse production techniques.

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Factors Affecting Degradation of PLLA/Ho-166 Microspheres During Radioisotope Production

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Introduction

Microspheres of poly-L-lactic-acid (PLLA) coated holmium-166 (Ho-166) are used for selective internal radiation therapy of liver malignancies. The microspheres are fabricated using stable Ho-165, which generates the therapeutic Ho-166 via neutron capture in a nuclear research reactor. The microspheres are highly sensitive to the conditions within the nuclear reactor during Ho-166 production: minor changes to the environment can cause decomposition either during neutron bombardment or immediately after the microspheres are suspended in media. Multiple factors in-core are thought to degrade the PLLA coating;[1] however, the nature of these interactions is not well understood. The purpose of this project is to better explore the effects of temperature, reactive oxygen species, in-core shielding, and gamma radiation on the integrity of PLLA/Ho-166 microspheres.

Description of the Work or Project

Gamma radiation and temperature damage were assessed in conjunction. Non-radioactive microspheres were exposed to a range of gamma doses (0-800 kGy) using a cobalt-60 source. Samples were then divided and maintained at temperatures ranging from 20-100 °C for 4 hours. Unlike microspheres irradiated in-core, the microspheres in these tests did not show any notable degradation over time. Microsphere quality was most impacted by temperature, with a significant damage threshold observed at 65 °C, coinciding with the glass transition temperature (Tg) of PLLA. Absorbed gamma dose had minimal impact on microsphere quality apart from the 600 kGy samples. Further testing is underway to determine if the increased damage at 600 kGy is linked to the dose rate or an intersection of chain-scission/cross-linking dominance.

The effect of reactive oxygen species on microsphere integrity post-suspension was studied by adding radioactive Ho-166 as an aqueous solution of $HoCl_3$ to naïve microsphere samples. Initially testing has shown modest increases in microsphere damage over time.

Finally, an experimental irradiation tube was constructed to allow varying thicknesses of lead shielding to surround samples in-core to decrease the amount of gamma radiation impinging on the microspheres during neutron activation. It was seen that increasing the lead shielding corresponds to significant changes in temperature, neutron flux, and sample quality.

Conclusions

This research provides a new understanding of the factors affecting degradation of PLLA/Ho-166 microspheres, and provides insight into potential damage mitigation strategies and methods to increase microsphere specific activity, quality, and production. Our results will inform equipment design and site selection for nuclear reactors producing Ho-166/PLLA microspheres, thereby increasing patient access to this promising cancer therapy.

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Materials and Manufacturing for EmberCore: A Commercial Encapsulated Radioisotope

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Introduction

Ultra Safe Nuclear is developing a radioisotope known as "EmberCore". The concept focuses on radioisotopes encapsulated in a ceramic or other type of cladding, known as an "Ember". USNC-Tech has developed a patented (PCTUS2116982, PCTUS2116980) manufacturing method termed to reduce complexity and cost for radioisotope production as well as enable deployment in different environments for power applications. In this process, radioisotopes are manufactured using natural non-radioactive precursor material embedded within an encapsulation material. The precursor material is then activated or "charged" inside a radiation source such as a fission reactor and finally packaged in a hot cell.

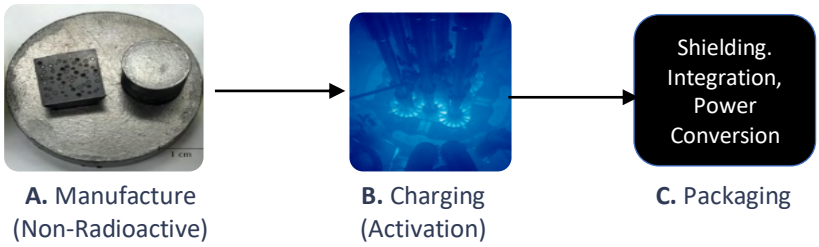

Figure 1. Ember Manufacturing Process

Embers can be manufactured in existing facilities and have a simple path toward a prototype using available. The main advantage of using radioisotope for energy applications is the energy density; $>10^6$ over electrochemical or combustion sources per unit mass. Radioisotopes also represent simple, robust, reliable heat sources suitable. Applications include thermal energy for deep space probes or lunar night operations. Current business development is focused on customer needs, power level, operation time. In the research and development space, development of the encapsulation "cladding" is the key feature. Two encapsulations have been explored and reported: alumina and silicon carbide (SiC).

Description of the Work or Project

EmberCore uses an integrated cladding using a ceramic encapsulation such as alumina and SiC. USNC has completed vertical integration of SiC over the last year, with nuclear grade, radiation stable forms of CVD SiC and NITE SiC within its materials portfolio.[1] These materials technologies leverage established bonding methodology from DoE fundamental research and development, which includes cladding seals and joining of ceramic-ceramic interfaces.[2] The irradiation is tailored to the activation cycle where Embers – both cladding and radioisotope – are exposed within similar temperature ranges at lower fluence/dose (i.e. within range of existing data).

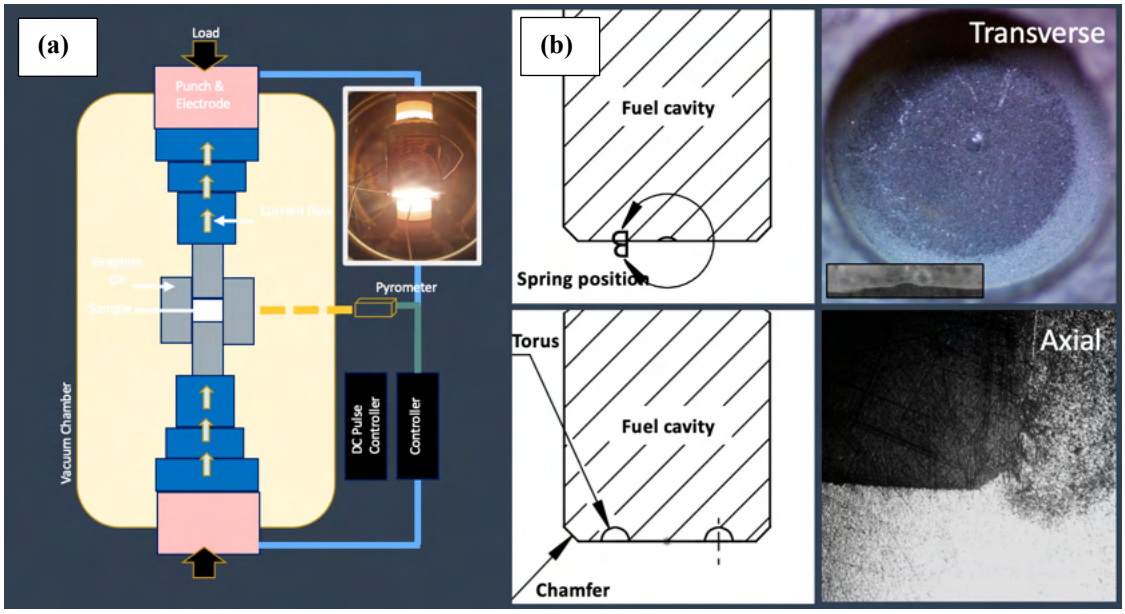

Figure 1. Cladding materials fabricated by (a) Pulsed Electric Current Sintering with schematic of key features of the fabrication instrumentation shown. Some of the ceramic cladding design features are shown in b) such as designed spring positions and chamfered vertexes during initial forming. The fuel cavity is not exclusive to containment of radioisotopes.

Cladding materials are fabricated by Pulsed Electric Current Sintering (PECS) shown in Figure 1(a). Applied pressure is often required to consolidate highly covalent ceramics such as SiC. Several key features are integrated into the encapsulation, some of which are shown in Figure 1(b) including a modeled swelling gap, interface release during fabrication, limited interphase formation (fuel-clad chemical interaction) and reduced internal stresses. Additive tooling methods eliminate the need for springs constraining the radioisotope during irradiation. Modeling has been coupled with fabrication design permit a prediction of the expected safety and performance as after neutron irradiation scheduled for late-2023 cycle at Oak Ridge National Laboratory.

Conclusions

EmberCore uses ceramic cladding to encapsulate a radioisotope. SiC encapsulation is currently being developed via Pulsed Electric Current Sintering. Fabrication and production of the encapsulation includes features that maximize robustness, reliability and production feasibility. The results highlight the mission of USNC to ensure robust barriers to radionuclide containment.

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Revival of Glovebox Alpha Instrumentation for Radiochemical Processing

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Introduction

Online monitoring of radiochemical processes can provide extremely valuable information to researchers and chemists that enable them to better perform chemical separations. Alpha and neutron detectors are used to monitor effluent activity for chemical processing column runs to determine elution composition and timing. These detectors can be used for the separation of any of the actinides that use column separations such as curium, berkelium, californium, etc. The alpha sensor provides the necessary measurements for the separation of neighboring actinides by monitoring elution drops from a column run such as a cation column–AHIB (α-hydroxyisobutyric acid) reagent. The corresponding measured alpha count rate can be used to relate the product coming from the column and alert the experimenter when it is time to switch to a new collection vial.

Description of the Work or Project

Alpha and neutron detectors are used to monitor effluent activity in Oak Ridge National Laboratory's (ORNL) Radiochemical Engineering Development Center (REDC) hot cells for separations. A similar effort successfully implemented using alpha detectors in REDC gloveboxes, but this work stopped in 2012 because the detectors were no longer providing useful information. Operation of these detectors within both hot cell and glovebox applications requires a packaged detector to prevent contamination and extend the detector's survival in the harsh environment. A project to revive these efforts for glovebox activities was undertaken. Initial investigations into the cause of the unsuccessful testing in 2012 revealed many possible causes of the failure, ranging from a failure of the electronic counting system, a change in material of the window to the detector, and degradation of the electronic feedthroughs for necessary connections through the glovebox. Each failure mechanism was addressed individually to ensure successful operation. A new count rate monitor was fabricated specifically for this purpose based on recent upgrades to the hot cell electronics.[1] A variety of Mylar windows were tested during packaging design to select an appropriate material that allows for alpha particle detection while also preventing ambient light response. An alpha detector was installed and tested for response inside a glovebox to test the feedthrough performance, which ultimately led to replacement of the feedthrough. Lastly, the alpha detector was used to aid with column separation of heavy actinides

Conclusions

Online alpha monitoring of radiochemical processing has been reestablished for glovebox operations at Oak Ridge National Laboratory. New detectors were purchased, packaged, and tested within a glovebox to ensure successful operation and aid in radiochemical separation processes. New electronics were designed for ease of operation and modernization.

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Development of a Selenium-75 Brachytherapy Source

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Introduction/Purpose: Selenium-75 (^{75}Se, $t_{\frac{1}{2}}$ = 118 days, $E_{\gamma,avg}$= 210 keV) is a radioisotope that is widely used in industrial gamma radiography. Its lower photon energy and longer half-life compared to Iridium-192 (^{192}Ir, $t_{\frac{1}{2}}$ = 74 days, $E_{\gamma,avg}$ = 380 keV) make it a viable candidate for use as a brachytherapy source. The goal of this study was to design a practical ^{75}Se source and investigate the feasibility of using the source for brachytherapy applications and its shielding properties combined with a novel rectal applicator developed for intensity modulated brachytherapy. Intensity modulated brachytherapy is a form of brachytherapy that uses static or dynamic shields to direct the dose to have a more conformal dose towards the tumour. This allows for dose escalation of the tumour and dose reduction towards healthy tissue.

Materials and Methods: A proposed ^{75}Se source (Figure 1a) was designed with its active core (0.65 mm diameter, 7 mm length, 3.7 g/cm3 packed density) encapsulated in a titanium (4.5 g/cm3, 0.90 mm outer diameter, 0.25 mm wall thickness) capsule. The length of the active core was chosen such that it can contain 23 Ci of ^{75}Se which gives a dose rate equivalent of 10 Ci ^{192}Ir. Since elemental selenium is highly volatile and chemically reactive at temperatures above its melting point (217°C), there are safety concerns when manufacturing a source, where temperatures exceeding the melting point will be achieved before and during irradiation. Prior to irradiation, elemental selenium must first be encapsulated by laser welding the capsule after packing, during which the air would be contaminated. To avoid this, the selenium source was chosen to be a compound (vanadium diselenide) where it will be irradiated in a thermalized neutron flux to activate the selenium, but due to small activation cross sections the titanium and vanadium will not be activated. The AAPM TG-43U1 brachytherapy dosimetry parameters were calculated for this source using RapidBrachyMCTPS, which is a Monte Carlo based treatment planning system. RapidBrachyMCTPS, was further used to calculate dose distributions in a 30 x 30 x 30 cm^3water phantom. Four different scenarios were simulated where a novel rectal applicator was combined with 3 types of rotating tungsten shields. These shields were developed for intensity modulated brachytherapy and include two rigid shields and a flexible (for patient comfort) shield. A no shield scenario was also simulated. The results were used to calculate the transmission factors (TF) for the different shield models. The results were compared with simulations performed with the conventional ^{192}Ir source. Simulations were performed so that the type A uncertainty at the 100% isodose line was less than 1%.

Results: The radial dose function and 2D anisotropy function were calculated and plotted in comparison with ^{192}Ir as presented in Figure 1b and 1c. The air kerma strength per unit activity and dose rate constant were calculated for ^{75}Se to be 4.75 +/- 0.01 x 10^{-8} U/Bq and 1.116 +/- 0.001 cm^{-2} respectively and for ^{192}Ir to be 9.79 +/- 0.01 x 10^{-8} U/Bq and 1.110 +/- 0.001 cm^{-2} respectively. Dose distributions in a water phantom were calculated and the dose colour maps for all scenarios can be seen in Figure 2a-e. For the rigid shields, ^{75}Se had TF values of 2.7 +/- 0.5 % and 2.3 +/- 0.7 %, and ^{192}Ir had TF values of 15.15 +/- 0.05 % and 13.2 +/- 0.2 %. For the flexible shield, ^{75}Se and ^{192}Ir had TF values of 16.0 +/- 0.5 % and 31.4 +/- 0.1 % respectively. This displayed that ^{75}Se had 4.23 and 4.03 times better attenuation than ^{192}Ir with the rigid shields and 2.08 times better with the flexible shield.

Conclusion: The designed ^{75}Se source was superior with regards to attenuation through tungsten shields due to its lower energy while still being able to produce an equivalent dose rate to ^{192}Ir. These results allow for justification of further analysis of this source for use in conventional

brachytherapy and intensity modulated brachytherapy as it is expected to deliver the same absorbed dose to the tumours as ^{192}Ir with similar treatment times while reducing the dose to surrounding organs at risk. This source will be manufactured and then irradiated at McMaster Nuclear Reactor for physical measurements in the coming months.

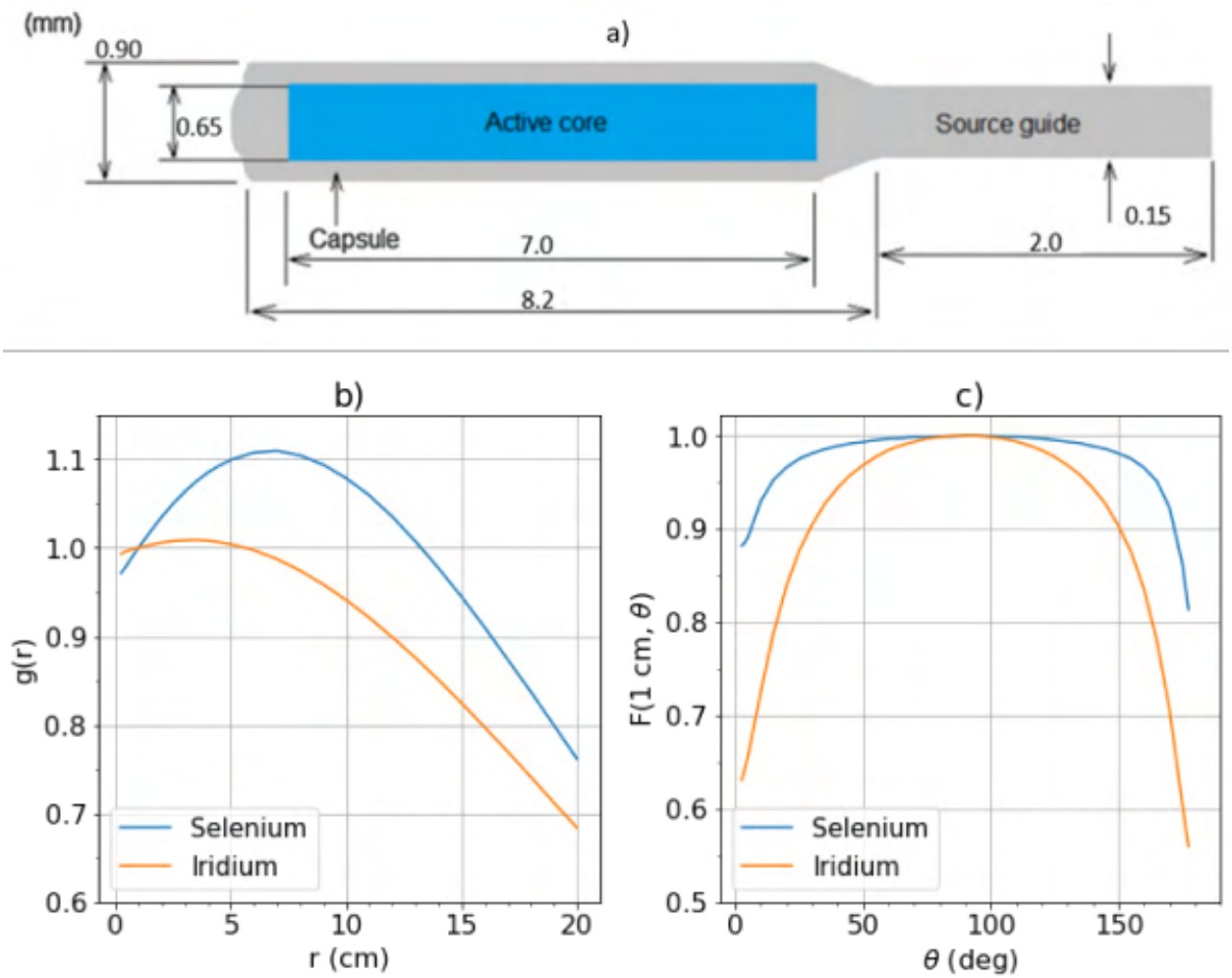

Figure 1: a) The selenium-75 source design with dimensions. b) The radial dose function of the selenium-75 source in comparison to the iridium-192 source. c) The same comparison of the 2D anisotropy function evaluated at r = 1 cm.

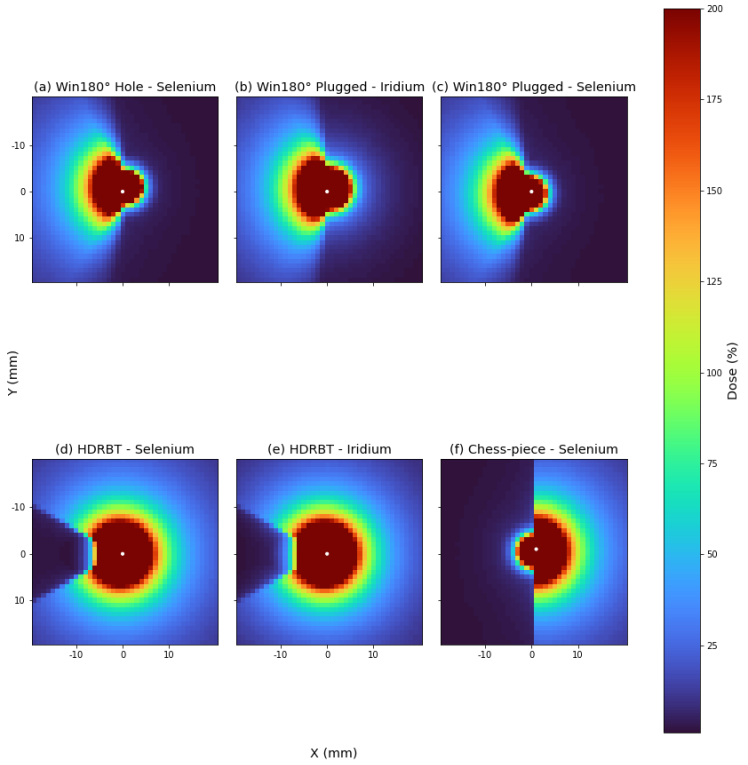

Figure 2: A comparison of dose distributions in water for selenium-75 and iridium-192 using the rectal IMBT shield (a-c) and conventional brachytherapy (d and e). The dose distribution in water for the flexible chess-piece applicator is also shown in f).

Latest development of α emitter imaging and quantification on a large Field Of View Targeted Alpha Therapy

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Introduction

Although very promising, the development of Targeted Alpha Therapy (TAT) requires the use of accurate techniques that can identify and quantify individual radionuclides in different matrices. However, currently, characterization of the nature and spatial distribution of radionuclides in a sample is time consuming and fastidious. Indeed, it necessarily requires the use of two distinct analytical techniques and detectors of different nature. Moreover, the resolutions of the two methods are often different from each other because they also depend on the nature of the detector used, which makes it difficult to integrate and interpret these two types of measurements together. To overcome these limitations, as well as to simplify and to accelerate the measurement process, it is now possible to use a digital autoradiograph capable of combining the measurement of the spatial distribution with the ability to separate and quantify each radionuclide.

Description of the Work or Project

For this purpose, a set of temporal and energy spectrometry techniques had to be specifically developed. On the one hand, the use of instruments capable of recording the location of each decay product allows to measure the evolution of the activity of the sample and thus, to deduce the contributions of several radionuclides. On the other hand, the development of an innovative method of autoradiography spectroscopy in particle energy also allows to separate them by measuring their initial energy. Even if the efficiency of energy spectrum reconstruction is low (<5%) compared to the efficiency of a simple autoradiograph (50%), this novel measurement approach offers the opportunity to select areas on an autoradiograph to perform an energy spectrum analysis within that area. Although if the samples usable on this type of instrument must generally be of solid nature, recent developments show that it is possible to use fluid samples (liquid or gas) with a cell system. Eventually, the proposed measurement system could allow dynamic imaging of radionuclides of interest.

Conclusions

From an application point of view, this opens up possibilities for theragnostic applications that typically use two radionuclides. Further upstream, it can optimize the production and distribution challenge of α radionuclides by allowing the identification and characterization of individual radionuclides in radionuclide chains such as 225Ac. Moreover, the measurement system and the associated method could strongly contribute to facilitate research on the biodistribution of α radionuclides.

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MO-99 PRODUCTION: THE COMMERCIAL REACTOR BREAKTHROUGH

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Introduction

Historically, the production of molybdenum-99 (Mo-99) via neutron capture has been limited, stemming from lower reaction efficiencies and extraction capacities based on low specific activity. A breakthrough in technetium generator technology, developed by BWX Technologies Ltd. (BWXT), based in the United States, has regained attention as a viable method for high-volume uranium-based Mo-99 production.

Description of the Work or Project

Through a partnership with Laurentis Energy Partners of Canada, a commercial CANDU (Canadian Atomic Natural Deuterium Uranium) nuclear reactor near Toronto, Canada, will generate Mo99 via neutron capture with subsequent processing at a local BWXT facility – both technologies being a first of a kind in the industry. Given the scale of power reactors, additional operating, and safety considerations beyond those incorporated in research reactors will be required for the production and handling of Mo99.

Conclusions

This paper discusses non-proprietary aspects of neutron capture-based Mo99 production through a commercial reactor in support of potential market expansion in other CANDU-type heavy water reactors or other future reactor designs.

RADIOISOTOPE PRODUCTION VIA MOLTEN SALT REACTORS

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Introduction

There is a world-wide push to develop the next generation of nuclear reactor technology. Molten salt reactors are one of the advanced reactor designs that are receiving a lot of attention. The United States has had two molten salt reactors that were developed in the 1950s and 1960s. The Aircraft Reactor Experiment (ARE) was developed and operated in 1954. The Molten Salt Research Experiment (MSRE) went critical in 1965. Today there is a renaissance in the exploration of molten salt reactors for power production. Variants on design include both a liquid core reactor and solid core reactors with molten salt as a coolant.

Description of the Work or Project

Molten salt reactors were assessed within the context of the needs of medical and industrial radionuclide needs. This work focused on liquid core reactors where process chemistry for purification is minimal in comparison to solid fueled reactors. Both fast spectrum and thermal spectrum designs were considered. Extraction of the liquid fuel salt and off-gas are two paths for radionuclide removal. Activity concentrations for radionuclides with medical and industrial interest are compared to current market needs. The potential for activation produced radionuclides is also explored.

Conclusions

Significant research is underway to develop, license and build advanced nuclear reactors. Molten salt reactors have the potential to significantly contribute to world radionuclide needs. Leveraging next generation nuclear reactor technology for radionuclide production will be necessary to address current isotope supply chain issues.

STATUS OF THE SHORT-LIVED RADIOISOTOPE SUPPLYING PLATFORM IN JAPAN

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Introduction

Radioisotopes (RIs) have been used in many fields of science taking advantage of their unique properties. Especially, their use in the medical field has recently become increasingly important. The Short-Lived Radioisotope Supplying Platform has been providing RIs under the auspices of the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) from FY2016 to FY2021. It continued with the support by Grant-in-Aid for Transformative Research Area since FY2022.

Detail of the platform

The platform consists of six accelerator facilities in Japan: Research Center for Nuclear Physics, Osaka University; RIKEN Nishina Center, RIKEN; Cyclotron Radioisotope Center, Tohoku University; Research Center for Electron Photon Science, Tohoku University; Institute for Quantum Medical Science, National Institute for Quantum Science and Technology (QST); and Takasaki Advanced Radiation Research Institute, QST. All these facilities operate their unique accelerators, beam irradiation apparatuses, and RI purification apparatuses. The complementary production and supply system for 93 nuclides in various physical and chemical forms is organized. More than 200 research programs in 7 years of our activity were carried out using the supplied RIs. The number of research programs per year is shown in Figure 1. It shows a yearly increase in the number of research programs. To meet the increasing demand for RIs, upgrading of the accelerator and the irradiation apparatuses has been carried out in each facility for increased supply capacity.

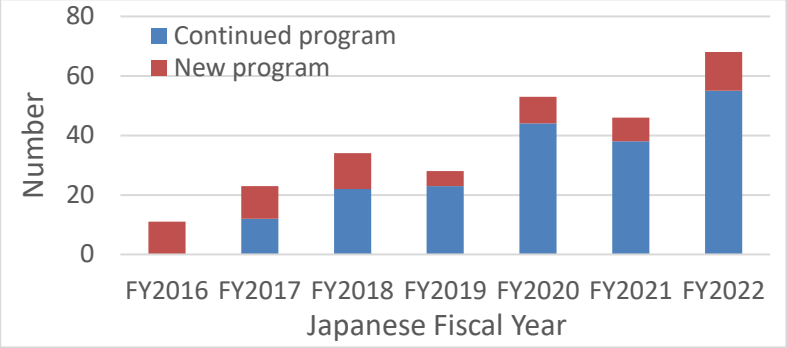

Figure 1: The number of research programs per Japanese fiscal year. Red bars show the numbers of new programs, and the blue bars show the numbers of continued programs.

Summary

The Short-Lived Radioisotope Supplying Platform in Japan has supplied various RIs for promoting the usage of RIs in broad field of science since 2016. The number of research programs is growing reflecting the increasing importance of the use of RIs. We continue supporting the research programs with the efforts of increasing the supply capacity.

This work is supported by Grant-in-Aid for Scientific Research on Innovative Area 16H06278 and Grants-in-Aid for Transformative Research Area 22H04924 by MEXT.

NEW NUCLEAR DATA AND COMPARISON OF PRODUCTION ROUTES OF THE NON-STANDARD POSITRON EMITTER Y-86

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Introduction

The positron-emitting radionuclide ^{86g}Y ($T_{½}$ = 14.7 h) is the most important imaging radionuclide for radiation dosimetry being used in combination with β^- emitting ^{90}Y-labelled therapeutics ($T_{½}$ = 2.7 d). Currently, the demand for this radionuclide is continuing. Several nuclear reactions have been investigated for the formation of ^{86}Y. The most feasible method recommended up to date is the proton induced reaction on highly enriched ^{86}Sr in the energy range of 14 to 7 MeV. In the scope of a large international collaboration that route was recently re-investigated together with the (d,2n)-reaction on enriched ^{86}Sr. In this work these two methods are faced with each other from the practical production point of view.

Cross Section Measurements and Yield Calculations

Cross sections of proton- and deuteron-induced reactions on highly enriched [^{86}Sr]$SrCO_3$ targets were investigated with three different cyclotrons at FZJ (Germany) and LBNL (USA) using protons up to 44.5 MeV and deuterons up to 40 MeV. Two other stacks were irradiated with 50 MeV deuterons using the CGR930 cyclone Cyclotron of the Université Catholique in Louvain-la-Neuve, Belgium. The excitation functions of the ^{86}Sr(d,xn)- and ^{86}Sr(p,xn)-reactions were measured by non-destructive γ-ray spectrometry of the activated samples. In addition to the main product ^{86g}Y the most important potential contaminants ^{85m,g}Y and ^{87m,g}Y were also investigated. The experimental results were compared with the results of nuclear model calculations using the codes TALYS and EMPIRE.[1] Based on the cross section measurements, many of which have been done for the first time in this work, the calculated yields of the two potential production routes of ^{86}Y using protons and deuterons were compared with each other, taking into account the level of co-produced isotopic impurities in each case.

Conclusion

Although from the yield point of view the production of ^{86}Y via both proton and deuteron induced reactions on enriched ^{86}Sr is feasible, in the energy ranges of E_p= 14 → 7 MeV and E_d= 22 → 14 MeV, respectively, the radionuclidic impurity considerations show that the ^{86}Sr(p,n)-reaction should be the most beneficial route.

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SECOND-PHASE FISSION Mo-99 PRODUCTION TECHNOLOGY DEVELOPMENT PLAN OF KOREA

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Introduction

Tc-99m has been the most commonly used medical radioisotope for past 60 years covering over 80% of total nuclear diagnostics procedures. Mo-99, mother of Tc-99m, productions have been mainly based on the fission of uranium in commercial-scale. During past decade, international supply of Mo-99 was unstable due to the aging of major isotope-producing research reactors.

In Korea, new research reactor project has been initiated after 2009-2010 Mo-99 crisis. The new reactor designed for dedicated medical radioisotope production. Especially, KAERI started first-stage development of fission Mo-99 production process development during 2012-2018. In this paper, we will present plan of the second-phase Korean Mo-99 development starting from 2023.

Description of the Work or Project

During the first-phase of the Mo-99 process development, KAERI performed hot test production of Mo-99 using depleted uranium target plates irradiated using HANARO. Own plate-type uranium target has been used for the project. The target had UAlx meat dispersed in aluminum matrix cladded with Al-6061 alloy.

For the second-phase development, quality of the Mo-99 solution will be controlled as commercial product. And, the production process will scaled-up to satisfy capacity of the new research reactor: 2,000 6-day Curie/week.

Conclusions

The new reactor construction started in May 2022, aiming 2027 as its first criticality. By the end of 2027, medical isotopes will be produced from the research reactor, and those will be supplied domestically and internationally since 2028. KAERI is aiming for the weekly production of 2,000 Ci (6-day calibrated) fission Mo-99 from the research reactor. The amount fulfills 100% of domestic, and 20% of international market.

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수출용신형연구로실증사업단/2023-01-25 18:28

A contribution to a stable supply of medical radioisotopes in Europe, TOURR and SECURE projects

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Abstract

The TOURR project "Towards Optimized Use of Research Reactors" is an Euratom-funded project among 9 partners across the European Union (EU), with 6 of them being research reactors (RR) operators.
Main objectives of the project are to assess the impact of the decreasing number of research reactors, identify future needs for the Eu RR fleet, draw a roadmap for the upgrade of the existing RR fleet, and develop a model for harmonized resource utilization. Another aim of the project is to evaluate the current and future need for neutron sources and medical radioisotopes in Europe [1].
While TOURR is at the end of its implementation after 3 years, the SECURE project has recently started and it aims to make a major contribution to the sustainability of medical isotope production and its safe application in Europe.
SECURE stands for "Strengthening the European Chain of sUpply for next generation medical RadionuclidEs" and will focus on promising developments in the design of irradiation targets, and production routes for existing and new isotopes in nuclear therapy and diagnostics. The ambition of the SECURE consortium is to identify and efficiently use the current resources for new radionuclides.[2]
Both TOURR and SECURE projects are part of the Strategic Agenda for Medical Ionising Radiation Applications (SAMIRA) which is the EU's first comprehensive plan for action to support a safe, high quality and reliable use of radiological and nuclear technology in healthcare. [3]
The TOURR project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945269. The SECURE Project is funded by the European Union. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

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STATUS AND APPLICABILITY OF NON-DESTRUCTIVE TECHNIQUES AT CNL FOR NUCLEAR FORENSICS

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Abstract

Nuclear forensics (NF) is the examination of radioactive or nuclear material, or evidence that is contaminated with radiological or nuclear materials in the context of legal proceedings under national or international law related to nuclear security [1]. Broader objectives also include nuclear non-proliferation, prevention of illicit trafficking of nuclear and radioactive materials, and emergency response in anticipated hazardous conditions following an incident [1] [2]. Collectively NDTs provide a wide range of information including mechanical, physical, isotopic, and chemical information, each of which render discerning information in the nuclear forensic investigative analysis. NDTs are important to nuclear forensics since samples/exhibits may be preserved for subsequent analysis/storage with little or no changes, and often have very quick turn-around times.

Canadian Nuclear Laboratories compiled a comprehensive list of non-destructive techniques available at its Chalk River campus that may find application in a nuclear forensic investigation. The list summarizes underlying working principles, figures of merit, and their relevance to potential samples that may be encountered in various scenarios. Examples include, radiological dispersive devices, materials falling out of regulatory control during civil unrest or wars, medical radioisotopes contaminating metal scrap and tackling suspicious objects including “hoax” items. The time required for analysis, ability to handle radioactive samples, and sample requirements (size, weight, volume, activity) have also been compiled. This information was used to sort the techniques into the often cited time frames issued by the IAEA [2] as part of an analytical plan.

This work not only facilitated the identification and applicability of NDTs to nuclear forensics, but also pathways for instrument development, multivariate analysis to further nuclear forensics science and support Canada’s contribution to the international community of practice.

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STATUS AND INSPECTION OF ISOTOPIC DECAY DATA FOR NORMALIZATION

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Introduction

Accurate and precise isotopic decay data are important for applied and basic sciences. There are more than 3,200 experimental decay data sets available in the Evaluated Nuclear Structure Data File (ENSDF). Of these, about 2,400 data sets represent β^- and β^++EC decay with approximately 1920 of the latter set are normalized, i.e. radiation intensities per 100 parent decays are known. While most of the isotopic decay data appear to be well known, inspection and improvement of the accuracy and precision have always been part of the experimental projects. Recently, we determined the positron emission intensity of ^{86g}Y by measuring the 511 keV annihilation γ-ray taking care of a number of correction factors. The electron capture (EC) intensity was also determined as an additional check by measuring the K_α and K_β X-rays[1]. The approach may further be used to inspect the accuracy of normalization of decay data set/s, when applicable - but not limited to, for cases where the normalization was done based on any assumptions.

Isotopes of interest for decay studies by 511 keV and X-ray measurements

In this work, isotopes related to medical, monitoring, dosimetry, etc. applications were inspected for which 511 keV and X-ray radiation measurements may provide confirmation of the normalization of the decay data, if done based on any assumptions. In general, these measurements need extra care, since both 511 keV and X-rays are not isotope specific. Of particular importance is therefore the high radionuclidic purity of the product under investigation.

Conclusions

Although the inspection/determination of isotopic decay data using the 511 keV and X-ray measurements appears to be challenging, yet the approach may be used for studying the decay data of special interest to confirm any assumptions used for normalization. In this presentation – some of the salient features of this method will be presented, with ^{61}Cu and ^{72}As as examples.

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DOMESTIC PRODUCTION OF Mo-99 AND Ac-225 USING COMMERCIAL PWR AND FAST EXPERIMENTAL REACTOR JOYO IN JAPAN

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Introduction

The production technology of medical radioisotopes (RI) using existing nuclear fission reactors has been studied to improve/achieve their domestic preparedness in Japan. The target nuclides currently considered in our project are Mo/Tc which is the most commonly used ones in medical diagnosis and Ac-225 which is recently known as effective alpha emitting nuclide for targeted alpha-particle therapy.

RI production in PWR and Joyo

This research project has launched in 2020 as a MEXT nuclear system R&D project by a team organized by Tokyo City Univ., Kanazawa Univ., Nippon Medical Isotope Co., Ltd. (NucMed), Mitsubishi Heavy Industries, Ltd. (MHI), and Japan Atomic Energy Agency (JAEA).

Fission reactors such as commercial light water reactors and experimental fast reactor are powerful tools to produce medical RI because they have 1) high neutron flux due to high power density, 2) high neutron fluence due to high capacity factor and long cycle length, 3) large irradiation area, 4) high heat removal capability and 5) excellent energy balance.

One of examined methods to produce Mo/Tc uses instrumentation tubes which penetrate the pressure vessel of PWR (Fig.1). The tubes are vacant and originally equipped for movable in-core sensors (fission chambers) to detect axial neutron distribution. Thin and short capsules containing Mo-98 enriched MoO_3 pellets are linked together by flexible connector and inserted into 4 tubes as 4 long strings of MoO_3 capsules for one week irradiation. The total weight of Mo in all MoO_3 capsules is about 500g and produce Mo-99 of about half of weekly demand (1000Ci/week) in Japan.

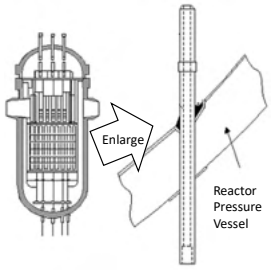

Fig.1 Guide thimble tubes and instrumentation tubes in PWR

There are several nuclear transmutation paths are available for Ac-225 production in thermal and fast neutron reactors. For instance, the fast experimental reactor Joyo (100Mt), which is currently only fast reactor in western countries, provides high energy and high flux neutrons required for threshold-type (n,2n) reactions to generate Ra-225 (parent of Ac-225) from Ra-226[1]. Joyo has potential to generate about 1Ci or more annually by irradiating 1g of Ra-226 for 60days/cycle × 3 to 5 cycles/year. The amount is more than half of the current world annual supply (2Ci/year).

Conclusions

Existing fission reactors, PWRs and Joyo, have potentials to work as excellent facilities for medical isotope production, as by-products of heat/electricity generation without consuming electricity and need for new plant construction.

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NUCLEAR FORENSICS CAPABILITIES AT CNL

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Introduction

Nuclear forensics (NF) uses a wide range of techniques to characterize materials in the context of a legal or national security investigation. Considerations are given to safety, preservation of evidence and ultimately may include a wide range of non-destructive and destructive analyses involving nuclear and/or radioactive material, such as nuclear fuel and radioactive sources used for medical and industrial purposes.

Canadian Nuclear Laboratories (CNL) has a long history of research and development in nuclear fuels as well as expertise in handling and characterization of radioactive materials. CNL has been leveraging its knowledge base and specialized facilities to support the enhancement of Canadian nuclear forensics capabilities. CNL's program of work on nuclear forensics focuses on several areas, including development of new methodologies, determination of signatures of nuclear materials and radioactive sources, and providing support to the national NF capability.

Description of the Project

Signatures of radioactive and nuclear materials can provide investigators with useful information about materials such as its origin, fabrication process, use, or history. Thus, one area of work for CNL is to investigate signatures of irradiated fuels, irradiated reactor alloys, and radioactive sources, using a range of techniques including isotopic analysis and radiochronometry. CNL has extensive information gained from post-irradiation examination (PIE) of nuclear fuels facilitating the investigation of isotopic signatures of nuclear fuels including power reactor, research reactor and other experimental fuels. Fuel types that have been investigated previously include uranium-based and mixed-oxide fuels, with the current focus on research reactor fuel. Future work is planned on thorium-based fuels, as well as advanced fuels such as those being considered for small modular reactors.

CNL has experience measuring nuclide ratios to estimate the fluence of reactor alloys for supporting ageing management and life extension of reactors. These techniques are being adapted for application to nuclear forensics on materials such as aluminum, stainless steel, and zirconium alloys. Samples of irradiated alloys with a known operating history are available for analysis and method development. The goal is to use measured isotopic ratios to "back-calculate" the irradiation history of the material which can then be compared to the operating history of the reactor and thereby validate the methodology.

Conclusions

CNL is continuing development work on new and innovative methodologies as well as leveraging its knowledge base and specialized facilities to support the enhancement of Canadian nuclear forensics capabilities. CNL's program of work on nuclear forensics focuses on several areas, including development of new methodologies as applied to radiochronometry and determination of signatures of nuclear materials and radioactive sources.

CANDU Role in Strengthening the Radioisotope Supply Chain

Author: Terry Campkin, Ontario Power Generation

Ontario Power Generation's (OPG's) 10 Commercial CANDU reactor units are responsible for generating about 60% of Ontario's electricity needs. In addition to providing low-cost, safe, reliable, and clean energy to 14.8 million Ontarians every day, OPG has also been reliably irradiating targets for the production of radioisotopes for over 50 years. Reactor reliability, high neutron flux, online fueling and capacity to produce in high quantities make power reactors an ideal source of neutrons for large scale radioisotope production. The predictable and reliable nature of commercial reactors enable dependable supply chains for some well established isotope markets (such as Cobalt-60), however there is opportunity to expand offerings to other isotopes markets providing greater stability to supply chains for decades to come.

In terms of existing and well-established isotope markets, OPG produces and sells 50% of the world's supply of Cobalt-60, a critical medical isotope used in medical radiotherapy to treat cancer, in medical device sterilization, and in food production. About 40% of the world's single-use medical devices, such as syringes, gloves, implants and surgical instruments, are irradiated and sterilized with Co-60.

Recent advancements in target delivery technology have made online irradiation and harvesting more effective and efficient, expanding the scope of radioisotopes that can be made available to the market through CANDU reactors. Laurentis Energy Partners (Laurentis), pending regulatory approval, will produce a stable supply of Molydenum-99, a critical life-saving isotope, from Darlington Nuclear Generating Station, for Canada and internationally. Laurentis leads this effort, in collaboration with OPG (as the irradiator) and BWXT. BWXT will utilize its newly designed proprietary generators, called NeuCap1, to process the Molybdenum-99 into Technetium-99m, the final product that will be used in diagnostic imaging. Darlington Nuclear will be the only source of Molybdenum-99 in North America, ensuring a stable domestic supply of this critical product.

In addition to having the ability to irradiate targets online and in the core, CANDU safety and recycling processes provide important isotopes, such as tritium, a nuclear by-product harvested through heavy water detritiation. Tritium is a radioactive form of hydrogen that occurs both naturally and as a by-product of the operation of nuclear reactors. Once thought of as waste, it is now seen as a strategic asset, valuable to other sectors including health care and high tech.

Advancements in nuclear by-product processing technologies are creating additional opportunities to reduce, reuse and recycle products that are needed for, or result from, the nuclear fission process.

The reliability of OPG's CANDU reactors and expanding the breadth of ways that isotopes can be generated and will be a key component to strengthening the radioisotope supply chain for the coming decades.

MCNP SIMULATION OF NEUTRON AND GAMMA-RAY IN SPENT FUEL AND VERIFICATION OF RELATIONSHIP WITH BURNUP HISTORY

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Introduction

At nuclear power plants, the spent nuclear fuel (SF), which generates energy, is removed from the core and kept in wet storage. Wet storage tanks for nuclear power stations are currently saturated worldwide, except in Finland and Sweden, where plans to construct permanent disposal sites for SF have been approved. To solve this problem, we have devised ways to store spent nuclear fuel further from an economic perspective, such as installing a dense rack while closely monitoring critical safety. The amount that can be stored is enhanced by precisely measuring the burn-up of used nuclear fuel, whereas previously it was presumed that new fuels were kept in a storage tank. In this regard, accurately assessing the degree of combustion is crucial. Since monitoring the neutrons and gamma rays released by spent nuclear fuel can be used to assess the burnup, it is important to collect data by monitoring for signals from the right detector. The development of non-destructive testing (NDT) based burnup measuring facilities has been done so far by different vendors.(Table .1)

Table 1: A comparison of combustion measurement equipment's features

Vendor	Characteristics	Detector Type	Signal Source
W/H(US)	-Simultaneous measurement of gamma rays and neutrons -Excellent radiation resistance	SiC(LiF coated)	Gamma-ray/Tritons-Neutron
EPRI/ Sandia (FORK, US)	Most validation cases in IAEA, US, Europe, etc. - Actual use for management in Sweden, Finland, etc.	CZT/Fission Chamber	Gamma-ray/Neutrons

Description of the Work or Project

Unless it is a real-scale experiment like the above measuring instrument, the situation in which the detector measures the dose is simulated through the MCNP code. To confirm that the detector effectively captures the radiation signal, the MCNP code will be used for the actual burnup history of CE16 x 16 operated in Korea. The detector's ability to produce the same burnup history when the correlation coefficient for SCALE's measurements of the burnup, cooling time, and enrichment are substituted will be tested.

Conclusions

In a conclusion, the correlation between burnup, cooling time, and enrichment obtained for the NPP's actual burnup history is consistent with the values identified in the MCNP's code, and the amount and impact of major nuclides that give the MCNP detected radiation.

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Nucleosynthesis Reactions with Active-Target Time Projection Chamber

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Introduction

The $^{12}C/^{16}O$ abundance ratio is of prime importance to understand stellar evolution and energy generation in the universe. A tiny change in the abundance ratio can change an entire scenario of the stellar nucleosynthesis from carbon burning to the Fe core in the last years of stellar life. Despite many experimental efforts in the last 50 years, none of the associated reactions like $^{12}C+\alpha \rightarrow ^{16}O+\gamma$ has yet been measured at the relevant stellar energies. Extrapolation from experimental data taken at high energies can only help theoretical modeling of stellar evolution and nucleosynthesis.

Description of the Work or Project

We propose the measurement of stellar nucleosynthesis reactions at low energies. We will develop a novel detector system consisting of a windowless active-target time projection chamber and a conduction-cooled superconducting magnet of the maximum magnetic field up to 3 T. The proposed research project (COREA, named after Carbon-Oxygen REaction with Active-target TPC) include the development of (1) a windowless active-target time projection chamber [1], (2) a conduction-cooled 3 T superconducting magnet [2] and (3) a $LaBr_3$/BGO array to perform direct measurements of (α,γ), (α,n) and (p,γ) reactions near the stellar energies using high-intensity heavy-ion beams at domestic accelerator facilities. High-intensity ^{12}C beam is available at the heavy-ion accelerator facility of Korea Basic Science Institute (KBSI) Busan Center, Korea. Highly-charged heavy-ion beams are produced by a superconducting 28-GHz ECR ion source at the facility and delivered to a 165-MHz radiofrequency quadrupole (RFQ). A high beam current for a fully-stripped ^{12}C beam is available. The beam can be accelerated up to 500 keV/u, which corresponds to E_{cm} = 1.5 MeV for the $^{12}C(\alpha,\gamma)^{16}O$ reaction.

Conclusions

In addition to the reaction $^{12}C(\alpha,\gamma)^{16}O$, the ^4He gas active target can be utilized to measure the reaction $^{16}O(\alpha,\gamma)$ for helium burning in a supernova and other radiative capture reactions of astrophysical interest, such as $^{14}N(\alpha,\gamma)$, $^{18}O(\alpha,\gamma)$, and $^{22}Ne(\alpha,\gamma)$ reactions in Asymptotic Giant Branch (AGB) stars, and $^{20}Ne(\alpha,\gamma)$, $^{24}Mg(\alpha,\gamma)$, and $^{28}Si(\alpha,\gamma)$ reactions in a supernova. We anticipate that the proposed research will extend the current knowledge of the stellar evolution and energy generation in the universe.

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Detecting Legacy sites that exist on the Korean peninsula

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Introduction

The recognition and management of legacy sites is a current issue worldwide. There is a high possibility that such sites exist even on the divided Korean Peninsula, and there is a need to develop systems and procedures to find and analyze these sites. In this study, we intend to develop a soil sampling plan to verify activities related to nuclear activity at legacy sites in the Korean Peninsula.

Description of the Work or Project

In the divided Korean Peninsula, it is not easy to determine the level of use and contamination of facilities such as uranium mines or nuclear facilities. Therefore, the experiment was conducted after assuming that the terrain of the legacy site on the Korean Peninsula was flat and no obstacles could cause disturbance within a certain radius. In Fukushima, an experiment was conducted by collecting soil of 5 cm from the surface to confirm contamination of the legacy site, and the study results were referred to for comparative analysis.

As a method for determining the sampling site, a Visual Sample Plan (VSP) program based on statistical analysis developed by PNNL (Pacific Northwest National Laboratory) was used. Through the VSP, the geometry of the Yeongbyeon area, where North Korea's activities related to nuclear activity were estimated, was reconstructed. Among them, the land that meets the experimental conditions was determined and the sampling spot was randomly selected.

Considering the physical environment between the selected spots, it was decided whether it was possible to apply and experiment with domestic legacy sites.

Conclusions

In this study, we developed a soil sample plan for legacy site detection in the Korean Peninsula. It is expected to be used as basic data for protocols for characterizing legacy site activity on the Korean Peninsula.

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Natural Radioactivity in the Upper Himalayas Region along the Manali-Leh Highway, India

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Abstract

Only a limited exploration of the upper Himalayas is carried out from the perspective of radionuclide mapping. However, this region is geologically very active and is the origin of many rivers that serve as drinking and irrigation water sources across the vast swathes of the Indian subcontinent. The present study aims to measure natural radioactivity in the upper Himalayas region along the Manali-Leh Highway. The water, soil, and rock samples were collected regularly along the highway between 4500 to 5200 meters in height. The level of Uranium in surface water samples collected from the region is determined using the LED Fluorimetry technique. The annual effective dose (AED), radiological and chemical toxicity risk and hazards quotient (HQ) due to the presence of uranium in water samples are also calculated. The concentration of heavy metals in water samples is also determined. The physicochemical parameters (pH, EC, TDS, and salinity) in water samples are measured, and an attempt is made to find the correlation between the concentrations of these parameters and uranium. The isotopic concentration of gamma-active radio-nuclei in soil and rock samples is estimated using HPGe detectors. The variation of radio-nuclei and their correlation with other parameters of importance has been estimated.

Keywords: Natural radioactivity, surface water, soil, uranium concentration, LED Fluorimetry, HPGe detector, annual effective dose

INTEGRIN $\alpha_v\beta_3$ RECEPTOR-BASED PEPTIDOMIMETIC ANTAGONIST (^{68}GA-DOTAGA-IAC) IN IMAGING TUMOR ANGIOGENESIS

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Introduction

Integrins are transmembrane glycoproteins comprising two noncovalently bound subunits (α and β). Out of all the integrins, the αvβ3 integrins are significantly overexpressed in various tumor cells undergoing angiogenesis. The absence of αvβ3 integrin on normal cells makes it suitable for the tumor angiogenic marker. In this study, a DOTAGA chelated peptidomimetic agent, Integrin Antagonist Carbamate derivative (IAC) is explored for its affinity towards αvβ3 integrin and application in angiogenesis imaging.

Description of the Work or Project

Radiolabeling of DOTAGA-IAC was optimized with $^{68}GaCl_3$ followed by various quality control assessments such as radionuclide purity, radiochemical purity, shelf-life, stability, sterility, and pyrogenicity. The in vitro saturation binding experiment was performed to determine the binding affinity (Kd) for recombinant human αvβ3 integrin receptors. The 96-well ELISA plate was coated with $\alpha_v\beta_3$ integrin receptor (2.5 µg/mL) and the wells were incubated with varying ^{68}Ga-IAC concentrations. The bound and total incubated radioactivity were measured and Kd along with the receptor density (Bmax) were derived. The pilot study was performed after institutional ethics committee clearance on histopathologically proven breast cancer patients (n=5; Mean age: 56.2 ± 14.3 y). Out of these, 2 were ER/PR positive and Her2 negative; 1 was ER/PR negative and Her2 positive; 1 was triple positive; 1 was triple negative. All these patients underwent whole-body PET/CT imaging post 45 minutes of ^{18}F-FDG (261.9 ± 36.6 MBq) and 35 minutes of ^{68}Ga-IAC (112.9 ± 9.25 MBq) intravenous administration on two consecutive days. The ^{68}Ga-IAC was prepared with more than 99.99 % radiochemical purity and passed all the quality control assessments. The K_d and B_{max} values were found to be 15.02 nM and 417 fmol, respectively. On WB ^{68}Ga-IAC PET/CT, the physiological biodistribution was seen in Thyroid (SUVmax 2.6), Lungs (SUVmax 1.3), Liver (SUVmax 3.62), and muscles (SUVmax 1.67). The high activity of ^{68}Ga-IAC in the Kidney (SUVmax 10.2) and bladder suggested renal clearance. In comparison to ^{18}F-FDG, ^{68}Ga- IAC also detected the primary lesions (SUV_{max} (IAC vs FDG): 3.85 ± 0.5 vs 16.01 ± 8.3) in all patients. Additionally, ^{68}Ga-IAC was also able to delineate the skull metastatic sites (SUV_{max} 3.7) which were not apparent in the ^{18}F-FDG scan.

Conclusions

The presented work demonstrates that IAC binds to αvβ3 integrin receptor with a high binding affinity. The clinical investigation showed that ^{68}Ga-DOTAGA-IAC could be a promising radiotracer in the evaluation of primary and metastatic breast cancer. A good tumor-to-background ratio also suggests the theranostic potential of DOTAGA-IAC for treating αvβ3 integrin expressing refractory and metastatic tumors when radiolabeled with ^{177}Lu or ^{225}Ac.

Key Words: αvβ3 integrin; Angiogenesis; DOATAG-IAC; FDG; PET/CT; ER; PR; Her2

DEVELOPMENT OF ANGIOSTATIN BASED RADIOPHARMACEUTICALS FOR PRE-CLINICAL EVALUATION OF LUNG INFLAMMATION IN MURINE

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Background

Angiostatin is the cleavage product of plasminogen to plasmin by phopshoglycerate kinase, serine proteinase-dependent release of kringle 1-$4_{1/2}$, and lastly matrix metalloproteinase dependent trimming of K 1-$4_{1/2}$ to K 1-$4_{1/2}$ or K 1-$3^{1,2}$. Angiostatin binds to various endothelial and neutrophil cell surface proteins, including ATP synthase F1F0, angiomotin, αVβ3 integrin, annexin II, and heat shock protein-$27^{3,4}$. Lung injury is a major component of lung damage, and it shows up in a variety of respiratory problems, including acute lung injury (ALI). The levels of angiostatin are elevated in bacterial endotoxin (LPS)-induced ALI and angiostatin alters the outcome of lung injury dependent on its bioavailability. We hypothesize that angiostatin (a native human form of glycosylated angiostatin containing 4 kringle domains) is a potential biomarker for detection of lung injury and thus aim to study the biodistribution of ^{89}Zr-tagged angiostatin (Zr-ANG) in a combined ozone and LPS induced murine lung injury.

Study Objectives

1. To test the purity, stability, and binding affinity of ^{89}Zr tagged angiostatin with endothelial tissue
2. To image ^{89}Zr tagged angiostatin by using PET/CT in ozone and LPS induced lung inflammation model of mice and test it against the prognostic significance of established inflammation markers such as ^{18}FDG and ^{68}Ga-citrate

Methodology

Angiostatin was conjugated with *p*-SCN-Bn-deferoxamine (DFO) for labeling with ^{89}Zr. For radiolabeling, Angiostatin-DFO was added to ^{89}Zr (1 MBq of ^{89}Zr: 0.25 µg of angiostatin-DFO) at a specific activity of 1 MBq/μg. The purity of the radiolabeled immunoconjugates was determined using SEC-HPLC and iTLC. Radioactivity was detected using a flow-through radio-HPLC-detector (Flow-RAM, Broomhill, UK). The chemical stability of ^{89}Zr labeled angiostatin in mice plasma were tested by using iTLC and SEC-HPLC for 5 days.

The treatment mice were first exposed to 0.05 PPM of ozone for 2 h and then intranasal inoculation (50µg) of LPS was performed. Tail vein injection of ^{89}Zr labeled angiostatin were performed in all the mice (both treatment and control) with approx. 10 MBq of activity. CT-SPECT images were acquired at 2, 24, and 72 h after injection using the Vector4CT scanner (MILabs, Utrecht). A 35 mins. full body SPECT scan was acquired in a list-mode data format with a high-energy ultra-high-resolution mouse pinhole collimator (HEUHR-1.0 mm) followed by 2 min full-body CT scan with a tube setting of 50 kV and 480 mA.

At the end of imaging, mice were sacrificed at 72 h post injection, harvested all the organs, followed by biodistribution studies. The activity was measured using a gamma counter, and the activity in organs was expressed as percentage injected dose per gram organ weight (% ID/g).

Results

^{89}Zr labeled angiostatin is chemically stable in plasma up to 120 hours. Standard uptake ratio (SUR) means the ratio of SUV (standard uptake value) of lungs and whole-body SUV. For the control group, the integral lines showing an upward trend of uptake ratio (sum of SUR for 2 h was 0.027, 24 h was 0.030 and 72 h was 0.032) while treatment group showing different pattern. For the treatment group sum of SUR for 2 h was 0.041 and it decreased at 24 h and 72 h showing the same SUR (sum) 0.022.

The uptake of ^{89}Zr labeled angiostatin was significantly higher only in case of lungs ($p< 0.0001$) compared to control group. The biodistribution results are statistically significant across control and treatment group ($p= 0.01$), organs ($p< 0.0001$) and their interactions ($p= 0.004$).

Conclusions

Acute lung injury causes significantly higher accumulation of ^{89}Zr labeled angiostatin in lungs compared to control group. It also increased in blood of the mice after ozone and LPS exposure.

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Funding source: Fedoruk Center.

NEUTRON ACTIVATION USING A MEDICAL CYCLOTRON

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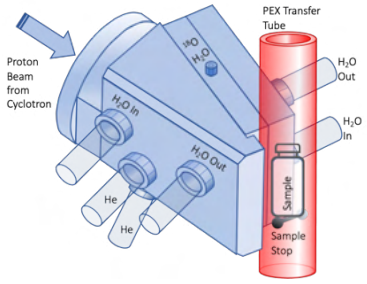
Introduction

Neutron activation is a well establish method of producing neutron rich radionuclides for non-destructive analysis, research, or medical applications. Traditionally this work has been carried out using the high neutron flux from a nuclear reactor. While reactors are extremely efficient at neutron activation, access to reactors isn't readily available, or can be cost prohibitive, for many researchers.

There are however an increasing number of cyclotron facilities located at hospitals, and research centres around the world. These facilities are often producing ^{18}F radiotracers one or more times daily. A byproduct of these production runs is a flux of "waste" neutrons resulting from the ^{18}O(p,n)^{18}F reaction. This work demonstrates the use of this neutron source for neutron activation.

Description of the Work

To allow for safe and efficient exposure of the samples to be irradiated, a simple pneumatic target transfer system was constructed using off-the-shelf parts available at most hardware or home improvement stores. The sample to be irradiated was loaded into a polypropylene vial and sent into the cyclotron vault using compressed air.



Application to Neutron Activation Analysis (NAA)

Initial studies looked at the activation of gold for analysis and quantification. Irradiation of showed a linear relationship between the activation and a product of beam current (μA), and beam time (h) on the [^{18}O]-water target and mass of gold in the sample (mg). Irradiated samples were counted using gamma spectroscopy and the area of the characteristic photon peaks used to quantify elemental composition. Application to the analysis of metals such as Au, Ag, W, in mineral samples as well as Hg and Cd in environmental samples will be demonstrated

Application to Radionuclide Production

Cyclotrons are well known to produce positron emitting radionuclides, through this work we will demonstrate the cyclotron production of short-lived beta emitting isotopes in quantities sufficient for research applications. While cyclotrons have a lower thermal neutron flux than reactors, they can produce high energy neutrons that open reaction possibilities beyond thermal capture. An example of this is the production of ^{31}Si via the ^{31}P(n,p)^{31}Si reaction. The production and purification of this isotope and others will be discussed. This route is valuable in ^{31}Si production as it gives the product in high specific activity and, the short half-life of ^{31}Si (2.6h) makes off site production impractical.

Conclusion

This work demonstrates the use of what would otherwise be a waste product from routine medical isotope production to preform analysis and the production of short-lived isotopes for research at no additional cost to the cyclotron facility.

The Kijang Research Reactor Project Status

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Introduction

The construction of Kijang Research Reactor (KJRR) has been started in May 2022, the first concrete will be poured in April and the initial criticality will be reached in April 2027. The domestic supply of Mo-99 will be possible from 2028 and the export of Mo-99 will be followed. The KJRR with the Radioisotope Research Center are located at the South-east Radiological and Medical Science Complex in Busan, Korea. Together with the Dongnam Institute of Radiological and Medical Science and Korea Particle Therapy Cancer Center, the KJRR will contribute to the enhancement of radioisotope research and industry as well as the global supply of important radioisotopes.

Description of the Work or Project

The KJRR is a dedicated isotope production reactor with the irradiation service for Silicon ingots. The KJRR facility includes reactor building, fission moly production building (FMPB), utility building, radioisotope production building (RIPB), radioactive waste treatment building (RWTB), and auxiliary buildings. There are two trains of 6 hot cells for Mo-99 production; transfer hot cell, receiving hot cell, dissolution hot cell, separation hot cell, purification hot cell, and packaging hot cell. The aluminum cake storage room and uranium filter cake storage room are located at the basement of FMPB. Three banks out of seven banks in RIPB are in GMP area. The cement solidification room and the fission moly intermediate level hold up tank rooms are located in the basement of RWTB.

There are six holes in the reactor core for uranium target irradiation by on-power loading to produce fission moly. The production capacity of fission moly is 2,000 6-day Ci/week. There are five holes in the beryllium reflector for neutron transmutation doping (NTD); two holes for 6 inch ingot and three for 8 inch ingot. The NTD production is more than 180 ton/yr when the resistivity is 500 Ω-cm. There are thermal pneumatic transport system (PTS) and epi-thermal PTS in the reflector region. The fast neutron irradiation (FNI) research will be performed using FNI hole in the outer reflector region.

Conclusions

The operating license will be requested and the commissioning plan is to be established in 2023. Early 2024, the radiation emergency plan will be submitted for regulatory review. The reactor performance test will follow the first criticality in 2027.

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KJRR Preliminary Safety Analysis Report, KAERI (2021)

Assessment of External Radiation Dose Rate after 18FDG-PET/CT Examination

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This study was conducted to assess the external radiation dose after 18FDGPET/CT Examination. 117 patients were enrolled in the study. Radiation exposure was measured using electronic survey meter, thermos scientific- red Eye PRD-ER personal radiation detector extended range the Calibrated Survey Meter at 0, 30, 100, 150, and 200cm distance; in the constant time interval. The time of measurement was; immediately post-injection, 30mint after the injection, 60mint later to injection, and at the time of releasing the patient. The proposed distances are mimicking variable distances of close relative or public in relation to injected patient. It is expected that the exposure from injected patient to close/ nearby person is not uniform as patient urinary bladder will have less radioactive compared to heart immediately at time of injection. Similarly, injected patient at a later time after injection of did not pass urine will have higher radiation exposure to nearby relative from urinary bladder.Thus, different distances Weber choose to mimic close contact (0 distance), close seat proximity (30 cm), passenger: driver distance (1 meter), front-back car seat (1.5 meter) and distance recommendation (6 feet).Times at different points reflect decay points with/ without urinary excretion.The result showed that the mean radiation equivalent dose rate at 0mint/0cm was 414μSv/h, at 30mint/30cm was 99.7μSv/h, and 60mint/100 cm was 18.3μSv/h. The radiation dose at different distances at the time of releasing the patient was 160.9μSv/h, 70.9μSv/h, 12.4μSv/h, 7μSv/h, and 3.7μSv/h respectively. The study concluded that the resulting exposure is below regulatory limits. The patient can be discharged safely with considered distance (at least 0.5 m) and time for more protection especially at first 60 minutes after the injection of radionuclide.

Keywords: 18FDG-PET/CT, Dose Rate, Radiation Exposure, Molecular Imaging, Patient Safety

Experiment and Monte Carlo simulations of gamma ray Back scatter off materials for security purpose Non-destructive Testing.

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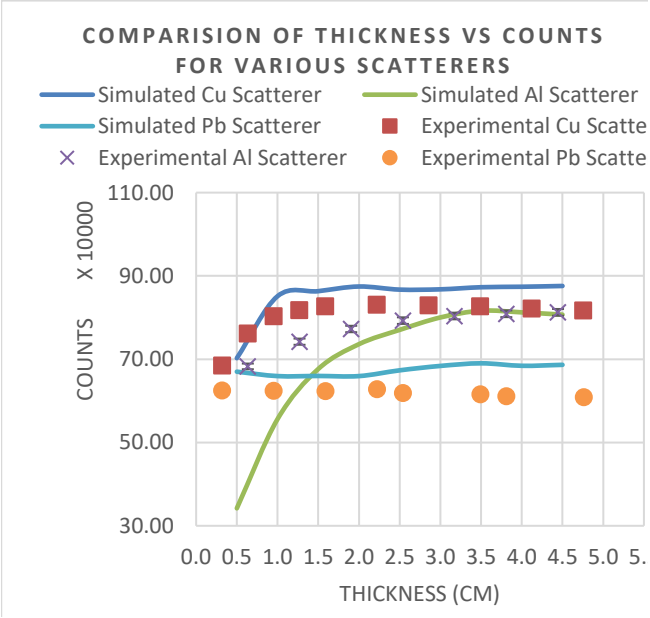
Introduction

The backscattering of gamma rays from the matter is of fundamental importance for radiation shielding designs and non- destructive testing of various samples from industry, medical and agricultural fields [1]. When gamma rays interact with thick target materials, they undergo multiple scattering within the target before they escape or get absorbed. Gamma ray backscatter yields are sensitive to the properties of the surfaces for their elemental composition. While photon transmission measurements are useful only for thin materials due to the absorption of photons in deeper depths, backscattering attain a saturation yield characteristic of the elements of sample.

Simulation and Experiments



A real time experiment and Monte Carlo simulation using GEANT 4 tool kit were carried out. In simulations, material foils of C, Al, Cu, Au and Pb of various thicknesses are exposed to a pencil beam of 661 keV photons. A 2" x 2" NaI (Tl) crystal assembly served as the back scatter photon detector. The figure shows initial results of Monte Carlo Simulation along with experimental data. The experiment was repeated with different absorbers of thicknesses ranging 0.5- 4.5 cm.

Simulations and experiment show that the backscatter yield reaches a maximum which is sensitive to the atomic number of the scatterer element. The normalized ratio of saturation yields vary as $Z^{1.5}$, The deviation from the expected exponent value of one from Klein-Nishina formula may [2], at least in part, be due to backscattering from the interior layers of the scatter medium.

Conclusion

From the simulations, as well as the real time experiment one, it is observed that thickness and Z of the material can be distinguished from backscattering phenomena, which has important implications in security industry for the detection of contraband and hazardous materials. The team is thankful to "Amity University-DTRA Technical assistance program for Amity University" Project Code: AUUP/2021-22/576.

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Modeling and assessment of Radioactive Iodine dispersion inside Egyptian Radioisotope Production Facility

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Abstract

Indoor Air Quality (IAQ) is very important topic in any radioisotope production facility. It is mandatory for some operators to be available behind hot cell that produce radioisotope to practice some tasks concerning maintenance, dosimetry and operation. One of these tasks is redundant transferring Radioiodine from cell to quality control lab and vice versa for measurements. Contam3.2 is a simulation model from NIST (National Institute of Standards and Technology) is used to study and predict I131 concentration in air in hot cell and area of operator behind the cell in emergency case. Emergency is described by dropping small amount of I131 on cell floor. The model predicts the elapsed time to remove contaminants by extraction ventilation system to deposit these contaminants in the dedicated filters and protect operators from inhalation. An emergency situation is also studied in case of opening I131 cell door hole (20 cm) by operators to pick the sample for quality control tests. Pressure interference occurs in this situation permitting some Iodine traces to be available in the areas under consideration. Ventilation system is responsible for removing all radioactive contaminants to settle it inside dedicated charcoal filters to clean the area and keeps it in permissible safe limits.

Key words: contaminants, activity, simulation, air concentration, extraction air, Kinetic Reactions.

A NEW ON-LINE MEASURING DEVICE FOR DISSOLUTION OF RADIOACTIVE DRUGS

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Introduction

Dissolution is one of the key indicators in the quality research of oral solid drugs. For radioactive solid drugs, such as therapeutic iodine [^{131}I] sodium capsules, how to detect their dissolution behavior in vitro in real time/online, is a difficult problem. At present, there is no online monitoring instrument for dissolution of radioactive drugs in global. Therefore, our company had developed an online measuring device for dissolution of radioactive drugs to realize online dissolution measurement of radioactive drug, provided the dissolution rate and amount of drugs in real time, and reduced the radiation dose of experimenter.

Description of the Project

According to the principle of drug dissolution measurement and considering the requirements of radiation protection, we designed a measuring device composed of a dissolution,circulating sampling device, measuring system and shielding device.

The dissolution adopted the commercially available equipment, on the basis of which was reformed to realize the automatic lifting of the dissolution basket rod and the automatic installation of the rotating basket, which can control the instrument remotely to ensure the safety of personnel.

The solution in the dissolving cup used three-way peristaltic pump and corresponding pump pipe to realize continuous online circulation sampling.

The measuring system of the device adopted a three-channel counter, including three sodium iodide detectors and corresponding electronic counter module and data processing software, which can directly give the dissolution results and dissolution curves and automatic comparison of multiple dissolution curves.

In order to eliminate the interference of environmental background and protect the radiation safety of the staff, we designed a shielding system for the device, including the probe shielding collimator, mobile shielding vehicle, waste liquid storage and transportation tools.

The design of the probe shield collimator not only ensures the accurate sample detection, but also shields the interference of the background to the measurement.

The design of a movable shielding vehicle and the waste liquid storage and transportation tool can effectively reduce the radiation dose of radioactive samples to the workers.

The device had been applied to the dissolution detection of therapeutic iodine [^{131}I] sodium capsules independently developed by our company, the results showed that the dissolution curves of three capsules measured at the same time were consistent.

Conclusions

A self-developed device for real-time/online dissolution detection of radioactive drugs for therapeutic iodine [^{131}I] sodium capsules in vitro dissolution behavior was studied, the realization of remote control/automatic measurement of capsule dissolution, and real-time dissolution curve, greatly reduce the radiation dose of workers, ensure the safety of workers and reduce environmental radioactive pollution. In addition, appropriate changes to the measuring conditions of the device detection system can be used for the automatic measurement of the generator leaching peak and relative activity.

Keywords: Radioactive drugs, dissolution, online measuring device, radiation safety

Disintegration of $^{158,166}Er^{*}$ isotopes formed via ^{16}O induced reactions across Coulomb barrier

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Introduction: To study the fusion-fission reactions at the low-energy regime, the heavy-ion induced reactions are used as useful tool to investigate the nuclear properties at various extremities. Such reaction mechanisms may lead towards the formation of compound nuclei of heavy/superheavy mass, which subsequently disintegrate into the binary fragments. In other words, it would be intriguing to understand the reaction dynamics of the compound systems belonging to Actinide, Pre-actinide and Lanthanide regions. In the present work, an attempt has been made to comprehend the decay dynamics of $^{158,166}Er^{*}$ isotopes, belonging to Lanthanide region, formed via $^{16}O+^{142,150}Nd$ reactions. To carry forward with the above idea, we have used the dynamical cluster-decay model (DCM) based on the collective clusterization approach of Quantum Mechanical Fragmentation Theory (QMFT).

Methodology: The Dynamical Cluster-Decay Model (DCM) [1] which is developed on the basis of well-known Quantum Mechanical Fragmentation Theory (QMFT). Based on QMFT, the DCM is worked out in terms of the collective coordinates of mass and charge asymmetries: $\eta = (A_1 - A_2) / (A_1 + A_2)$ and $\eta_Z = (Z_1 - Z_2) / (Z_1 + Z_2)$. Here in DCM, we define the decay cross-sections in terms of the partial wave analysis.

Description of Work: We have calculated the evaporation residue (ER) cross-sections for $^{158,166}Er^{*}$ compound nuclei, belonging to the Lanthanide series, formed via $^{16}O+^{142,150}Nd$ reactions over a wide range of incident energies $E_{c.m.}$ (MeV) spread across the Coulomb barrier. For the above said isotopes of Er^{*}, the experimental data of [2] has been addressed within the permissible limit of neck-length parameter (ΔR). Besides, the fragmentation and mass distribution analysis have been done for the spherical and deformed configurations of the fragments in the exit channel. The fission distribution which starts appearing at higher angular momentum values is symmetric for spherical choice. However for deformed choice some additional peaks are visible, and support emergence of intermediate mass fragments (IMFs) and heavy mass fragments (HMFs).

Conclusions: For $^{158,166}Er^{*}$ isotopes, the ER cross sections are duly addressed and relative emergence of competing channels such as intermediate mass fragments (IMF), heavy mass fragments mass fragments (HMF) and fission fragments (FF) is analysed in view of shape of decaying fragments.

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AN EFFECTIVE SIMPLE ESTIMATION METHOD OF PLUTONIUM MASS USING DIFFERENTIAL DIE-AWAY (DDA) ANALYSIS

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Introduction

Differential Die-Away (DDA) analysis is a non-destructive testing method for estimating the initial enrichment (IE) and burnup (BU) of the spent nuclear fuel using the measured die-away time. With the estimated IE and BU, a depletion calculation code can be used to estimate the mass of plutonium in the spent fuel. However, this method requires depletion calculation. In this study, we suggest a simple approach which can estimate the mass of plutonium in spent fuel using a simple formula based on DDA analysis results and a database, without depletion calculation.

Description of the Work or Project

Applying DDA analysis to spent nuclear fuel can calculate the die-away time (τ) and count rate (C). We showed that the mass of ^{235}U in a fixed spent fuel assembly and C/τ has linearity as shown in Figure 1: $m_{^{235}\mathrm{U}} \propto C/\tau$. The other relationships we found are the linearity between the reduced ^{235}U mass relative to the fresh fuel and C_r, and the linearity between the Pu (^{238,239,241}Pu) mass and τ_r with following definitions: $C_r = 0.6 \times C_0 - C$ and $\tau_r = 0.7 \times \tau_0 - \tau$. In these definitions, τ_0 and C_0 represents the die-away time and count rate for the fresh fuel assembly which has same IE, respectively.

In this work, the depletion calculations for 51 spent fuel assemblies with various initial enrichment and burnup were conducted using the ORIGAMI code and DDA analysis were simulated using MCNP6 for generating τ and C. 36 assemblies were used for linear regression to obtain the linearities addressed above. The plutonium mass estimation was calculated using the relational expression for 15 assemblies not used in linear regression. As a result of the calculation, the Pu mass could be estimated within 4% accuracy.

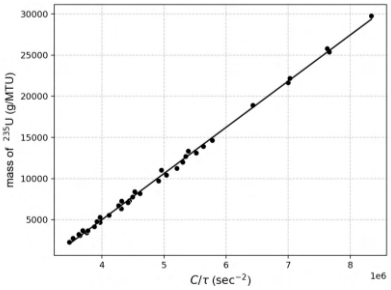

Figure 1: Linearity of ^{235}U mass and C/τ

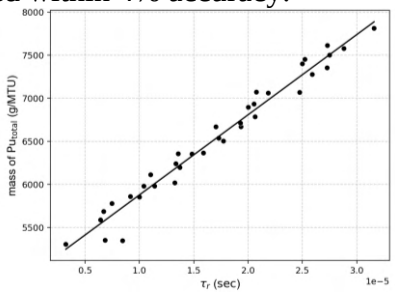

Figure 2: Linearity of m_{Pu} and τ_r

Conclusions

From this study, it can be concluded that the simple linearities based on DDA analysis results, and a database can estimate the plutonium mass in spent nuclear fuel assembly with 4% accuracy. This approach has the potential to simplify plutonium mass estimation and improve spent fuel management.

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PAIR ISOTOPES COPPER-64 AND COPPER-67 PRODUCTION IN RFT-30 CYCLOTRON FACILITY

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Introduction

^{64}Cu ($T_{1/2}$: 12.7 h) and ^{67}Cu ($T_{1/2}$: 61.83 h) are paired radionuclides with promising applications in the theranostic field for non-invasive diagnosis and therapy of various diseases. ^{64}Cu is an attractive radioisotope of significant interest for positron emission tomography. Its decay properties ensure high-resolution PET imaging, and it has a relatively suitable half-life corresponding to biological half-life compared to fluorine-18 ($T_{1/2}$: 110 min) and carbon-11 ($T_{1/2}$: 20.4 min). ^{67}Cu is a radioisotope with significant potential for therapeutic applications in nuclear medicine. Despite its potential, the use of ^{67}Cu for radionuclide therapy has been hindered for decades by its limited supply and low specific activity. In this study, we verified the irradiation, target preparation, chemical separation, and quality control of radioactive copper. We optimized the enriched target electrodeposition, proton beam irradiation, separation/purification, and quality control processes to enhance the routine capability of ^{64}Cu and ^{67}Cu production.

Description of the Work or Project

(1) Target preparation: Gold and silver were used as a cathode and a platinum rod was used as an anode. The backing materials were electrodeposited with ^{64}Ni and ^{70}Zn. After cleaning and drying, the plated ^{64}Ni and ^{70}Zn on the substrate were <150mg and <250 mg, respectively. **(2) Proton beam irradiation:** ^{64}Ni and ^{70}Zn targets were mounted using a self-produced cradle (6° tilted target system) and target transfer device, and then the proton beam was irradiated at 11 MeV and 17.7 MeV incident energy, respectively. **(3) Chemical separation:** ^{64}Cu and ^{67}Cu were purified using solid-phase resin with fraction volume of 500 μL at a flow rate of 0.5 mL/min using water (^{64}Cu and ^{67}Cu elution yield: >98% and >85%, respectively). **(4) Quality control:** The radionuclidic purity and metallic impurity contents were evaluated. It shows >99.9% high purity γ-ray spectra of purified ^{64}Cu and ^{67}Cu. The content of impure metals in the purified radioactive copper less than 1 ppm present, it will not affect the radio-labelling reaction.

Conclusions

The production procedure of pair-radioisotope ^{64}Cu and ^{67}Cu has been studies. This attempt is an imperative study that can guarantee the supply and quality of radioisotopes for diagnosis/therapy in the field of nuclear medicine. Each condition was established using prototype RFT-30 MeV cyclotron, equipment and chemical to optimize the entire process, including proton beam irradiation, targetry, chemical separation and quality control of ^{64}Cu and ^{67}Cu. Further studies on the Automation separating apparatus are underway for radioactive copper production.

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ADVANCING RADIOPHARMACEUTICAL TRANSLATION IN MALAYSIA: TACKLING REGULATORY AND MANUFACTURING ROADBLOCKS

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Radiopharmaceuticals are critical for diagnosing and treating various diseases, such as cancer, cardiovascular, and neurological disorders. However, the development and translation of radiopharmaceuticals from bench to bedside require extensive preclinical and clinical research, followed by regulatory approval and manufacturing processes. In Malaysia, the regulatory framework for radiopharmaceuticals is overseen by the National Pharmaceutical Regulatory Agency (NPRA) and the Department of Atomic Energy (Atom Malaysia). These agencies ensure that radiopharmaceuticals are safe, effective, and of high quality before they are administer to patients.

Due to the inherent risks associated with radioactive materials, the manufacturing of radiopharmaceuticals is subject to stricter regulations and controls than regular pharmaceuticals. Adherence to regulatory compliance becomes even more challenging as they can be opposing requirements between radiation safety and patient safety during their production.

Furthermore, the availability of specialized facilities, skilled personnel, and funding can also pose challenges to the manufacturing process. Addressing these challenges requires collaboration among regulatory bodies, academia, industry, and healthcare providers to establish a comprehensive and sustainable ecosystem for radiopharmaceutical development, translation, and access.

The use of PET and SEPCT radiopharmaceuticals in Malaysia is growing significantly owing to the demand for non-invasive diagnosis and effective cancer treatment. A surge in incidences of cancer, cardiovascular diseases, and other chronic diseases, a rise in the geriatric population, and increasing demand for targeted cancer treatment is propelling the need for nuclear imaging techniques and radiopharmaceuticals. As the demand for radiopharmaceuticals continue to rise, so will the number of facilities that can produce them. By recognizing the potential obstacles and regulatory hurdles that may arise, it can help streamline the process and minimize any difficulties.

Keywords: Good Manufacturing Practice, Good Radiation Practice, Radiopharmaceuticals, PET, SPECT

Production and evaluation of therapeutic radionuclides at SCK CEN

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Introduction

Targeted Radionuclide Therapy (TRNT) is currently one of the most promising developing treatments of cancer within nuclear medicine. Reliable supply of high quality (GMP grade) therapeutic radionuclides is key to ensure development and successful clinical introduction of the next generation therapeutic radiopharmaceuticals for the benefit of cancer patients. SCK CEN is currently establishing its own program on production and evaluation of the most potential therapeutic radionuclides for future nuclear medicine with several partners: Lu-177, Tb-161 and Ac-225.

Description of the Work or Project

The therapeutic radionuclide Lu-177 has already proven to be very successful in cancer treatments, and is now widely used routinely in clinical applications at hospitals. Also the interest for the β^- emitter Tb-161 in nuclear medicine is growing fast. Recent studies indicate a superior therapeutic for Tb-161 over Lu-177 in small volumes as a results of its high Auger/conversion electron emission probability. First clinical studies also show an added value of Tb-161 in TRNT.

With its high-flux BR2 reactor on-site, SCK CEN hosts one of the most important medical radionuclide production facilities in the world, for both diagnostic (Mo-99) and therapeutic (Lu-177) uses. So far, this production was limited to an irradiation service for third parties. In order to meet a worldwide exponential growing need of n.c.a. Lu-177 and n.c.a Tb-161 in the years to come, SCK CEN is developing a radiochemical processing and dispensing facility with external partners that will allow for a future large-scale production of GMP grade Lu-177 and Tb-161.

In support of the continuously expanding applications of Lu-177 and Tb-161 within the field of therapeutic radiopharmaceutical, SCK CEN is also deploying research infrastructure for radiopharmaceutical development. Full preclinical research will be covered, ranging from development of novel chelators and radiolabeling methods to in-vitro and in-vivo evaluation. In-house research lines on specific cancer types are established, for which full biological and radiopharmaceutical evaluation have been setup.

Conclusions

The presentation will give an overview of SCK CEN contributions to the future supply and support radiopharmaceutical evaluation of promising radionuclides for present and future applications of TRNT within nuclear medicine.

Design, Construction, Installation, and Refinement of a new Compact Radioisotope Production Station at the University of Washington Medical Cyclotron Facility

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Abstract. The University of Washington Medical Cyclotron Facility (UWMCF) has developed an auto-loading, auto-ejecting compact isotope target station to enhance isotope production capacity. The target station was originally designed to irradiate a novel bismuth target for ^{211}At production but has since been modified to accommodate targets of various thickness and target materials. Here we discuss the target station and ^{211}At target design criteria and methods, and our early experience producing isotopes with this station.

1. Introduction. The UWMCF began developing a compact isotope production target station and novel ^{211}At targetry in 2019 as part of its beamline expansion program intended to increase isotope production capacity. Early planning defined five major design constraints. 1) The ^{211}At target should withstand 100μA of 29MeV alpha beam, 2) The ^{211}At target should be size compatible with commercially available auto-transport systems (disc less than 25.4mm diameter and 6mm thick), 3) The target station should be able to auto load and eject targets to minimize personnel exposure, 4) The target station should be adaptable for other target materials, 5) The target station should be cost effective to produce and maintain. The development process began with a basic design of the ^{211}At target, which was carried forward and refined in parallel with the target station design to ensure compatibility. Finally, the target station was modified to accommodate thicker target material to support production other than ^{211}At.

2. Method & Materials. Target and cooling system designs were critical to handle 1.4kW of beam power. Minimum target material thickness was determined using published production cross-sections, SRIM, and MCNP. Energy deposition was modeled using MCNP. The target thermal model was produced in ANSYS Fluent using measured cooling water temperature, flow parameters and measured lateral beam distribution. The target mechanical model was developed in ANSYS SpaceClaim. Precision machining and metrology has been used to provide and confirm target material thickness. Bismuth target backing was constructed from 5N aluminum to reduce by-product activity without negatively effecting radiochemistry processing. The target station was designed using Autodesk Fusion360 and was mostly built with readily available off-the-shelf parts.

3. Results & Discussion. Verification of target loading, ejection, vacuum integrity, cooling integrity, and water flow rate was conducted prior to the first test irradiation. Since July 2021 we have conducted 20 test irradiations, gradually increasing the alpha beam current, on the bismuth target to produce a total of 250 mCi ^{211}At. Potential thermal hot spots were observed one time on

a target post irradiation, but we had lost beam profile scanning capability earlier on in the irradiation and most likely had an overly focused beam. We have also irradiated pressed powder and foil targets demonstrating that this system can be useful for a variety of target materials.

4. Conclusion. Multiple ^{211}At production runs have been completed on a novel compact target station using bismuth targets capable of retrieval by a commercially available target transport system. Determination of ^{211}At production rates with new targetry is ongoing with our radiochemistry group. Proton and deuteron irradiations have been conducted using the new station for production of high specific activity ^{186}Re, ^{189}Re, ^{85}Sr, ^{230}Pa. Various methods of applying target material to the target backing are being evaluated. Further reducing unwanted activity is paramount and we are layering other materials into the target backing for this purpose.

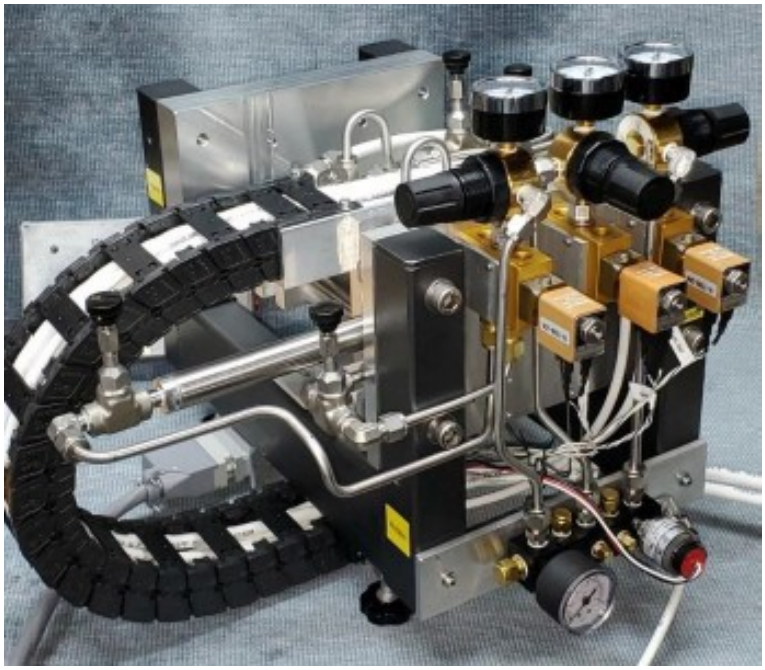

Figure 1.

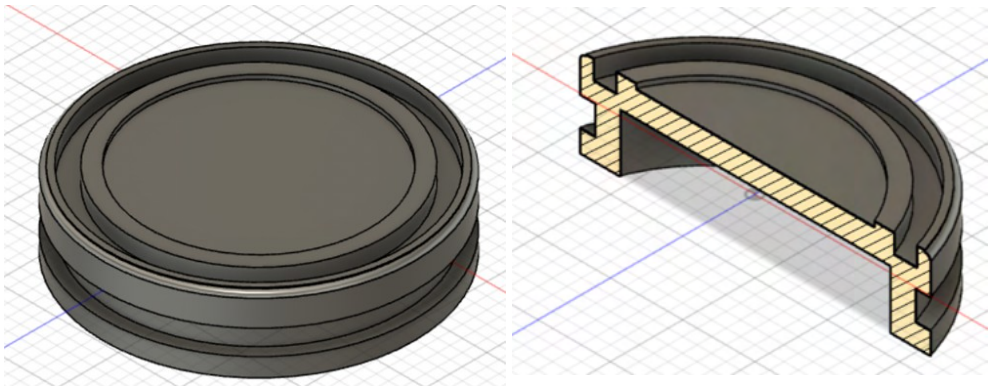

Figure 2.

Alpha production routes for medical radioisotopes

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Introduction

Over the past decade, interest has been growing in the use and application of radionuclides for medical diagnostics and therapeutics. Production of these radionuclides has historically been dominated by only a few nuclear pathways, those being either reactor production pathways utilizing neutron activation (n,γ) or cyclotron production pathways utilizing proton spallation (p,x). Other production pathways exist, for example photo-activation (γ,n) reactions using electron accelerators as well as spallation with heavier ions such as alpha spallation (α,n) and deuteron spallation (d,n). Only recently have these other production pathways seen modern investment and industrial development. Because of the lack of industrial investment, very few facilities exist which are capable of utilizing these alpha and deuteron production pathways. The focus of this work is to illustrate the unrealized benefits of using these pathways for radioisotope production. In this work we concentrate on several alpha production pathways to demonstrate the superiority of these methods for producing certain high-specific activity, non-carrier added radioisotopes for medical and industrial uses.

Description of the Work or Project

Using a purpose built Geant4 application the authors are able to compare the effectiveness of alpha vs. proton production pathways for several medically relevant radioisotopes, examples include Copper-67, Strontium-82, Cesium-131, Rhenium-186 and Astatine-211 among other proposed products. These production pathways are examined end-to-end, incorporating analysis of target materials, product yields, co-product and contaminate yields, competing reactions, post-bombardment decay, radio-chemical separation, radiopurity, waste, shielding and carrier added effects.

Conclusions

The results of this work illustrate that many factors impact the effectiveness and feasibility of a radionuclide production process. For a broad selection of specific radioisotopes, the alpha production pathway is superior for producing several high specific activity, high radio-purity non-carrier added products.

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KEYWORDS: Radioisotope, Copper-67, Strontium-82, Cesium-131, Astatine-211

NUSANO TARGETRY FOR THE PRODUCTION OF MEDICAL ISOTOPES

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Introduction

Nusano is constructing an accelerator facility in West Valley City, UT to produce radioisotopes. Our core technology is a patented and thoroughly tested source technology that enables us to produce alpha, deuteron, and lithium-ion beams in the multiple mA range. With these high current particle beams, Nusano can produce a wide range medical isotopes in yields unachievable at any other single facility. Some of these isotopes include ^{211}At, ^{117m}Sn, ^{67}Cu, ^{177}Lu, ^{186}Re, ^{82}Sr, ^{68}Ge, and ^{123}I. ^{211}At and ^{117m}Sn in particular are two radioisotopes that show great therapeutic potential but are only produced in high specific activity via an α-beam (or Li-ion beam in the case of ^{211}Rn/^{211}At). In Feng and Zalutsky (2021), it lists the α-accelerator facilities worldwide that have produced ^{211}At in the last 5 years, and the combined current of these facilities is ~350 μA. Nusano's single facility will have an order of magnitude more current than these combined facilities. One of the challenges with a high current alpha beam is the design of targets that can survive the heat loads.

Description of the Work or Project

Nusano's α-particle beam (50 MeV) has a relatively short range (a few hundred micrometers) in typical metal solid targets. This puts an incredibly high heat load into a relatively small volume. To take advantage of the full beam current, we plan to use a number of operational methods on our beamline and targets to ensure survival. These include fast-acting switching magnets that allow us to shift the beam pulse-to-pulse across multiple targets and octupole magnets to uniformly stretch the beam over an area up to 5 cm × 5 cm at the target. Nusano target designs consist of several techniques to improve predicted target survivability. A thin vacuum window will be employed to reduce sputtering and sublimation. A structural support grid will be clamped against the window for stability in long duration irradiations. Because Nusano's facility is under construction and no other facility can match Nusano's predicted α-beam current for testing, significant modeling and physical testing on lower beam current accelerators is needed. Detailed modeling of beam distributions (Elegant code), heat loads (MCNP6 code), and thermal and stress loads (ANSYS) are being performed to ensure target integrity at the desired average currents and irradiation lengths.

Results and Conclusions

Results of thermal and structural simulations performed indicate that targets can survive between 250 to 500 μA at 50 MeV. Currently, ranges of thermal contact resistances (i.e. between the window, target material, and housing) and radiation damage factors (thermal conductivity, swelling, and embrittlement) are being used in the models to estimate maximum currents targets can survive. Measurements are currently planned to reduce these uncertainties in the model. Modeling methods and results to assess target survivability will be presented.

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INVESTIGATING HIGH-ENERGY PROTON-INDUCED REACTIONS: IMPLICATIONS FOR LEVEL DENSITIES AND THE PREEQUILIBRIUM EXCITON MODEL

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Introduction

Multihundred MeV proton accelerators are promising sites for the large scale production of medical radionuclides due to the high production rates enabled by their high-intensity beam capabilities and the long range of high-energy protons. However, the ability to reliably conduct isotope production at these accelerators and model relevant (p,x) reactions in the 100–200 MeV range is hampered by a lack of measured data. The current suite of predictive reaction-modeling codes is only accurate to within approximately 20% for (p,x) and (n,x) reaction channels where a large body of experimental measurements currently exist. In cases where few data exist, these codes often exhibit discrepancies anywhere within a factor of 2–50.

Description of the Work or Project

In order to address this deficiency, stacked-target irradiations were performed at LBNL, LANL, and BNL, measuring proton-induced reactions on niobium, arsenic, lanthanum, antimony, and thallium targets from threshold to 200 MeV. Reaction modeling at these energies is typically unsatisfactory due to few prior published data and many interacting physics models. Therefore, a detailed assessment of the TALYS code was performed with simultaneous parameter adjustments applied according to a standardized procedure. Particular attention was paid to the formulation of the two-component exciton model in the transition between the compound and preequilibrium regions, with a linked investigation of level density models for nuclei off of stability and their impact on modeling predictive power, leading to global χ^2 improvements of up to 40x.

Conclusions

This assessment has revealed a systematic trend in how residual product excitation functions for high-energy proton-induced reactions on spherical nuclei are miscalculated in the current exciton model scheme. Additionally, adjustments made to the TALYS *ldmodel 4* (Goriely) and *ldmodel 6* (HFB+Gogny) level densities illustrate the reliance of reaction modeling upon well-characterized models of the nuclear level density at high excitation energy [1, 2]. The next stage of this effort is currently underway, using in-beam gamma spectroscopy and (γ,n) coincidence measurements on these targets to expand the measured channels to include short-lived and stable isotopes inaccessible via decay spectroscopy. This is expected to further improve the robustness of these model improvements via fitting to these additional independent reaction channels and neutron evaporation measurements.

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LOW GRADE URANIUM ORE ANALYSIS BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS USING A MEDICAL ISOTOPE CYCLOTRON

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Introduction

There has been a significant decline in number of research nuclear reactors world-wide over the past 25 years. Canada has not been immune to this trend, decreasing from a peak of ten operating research reactors in 1995 to three operating facilities in 2023. With this decline there has been a concomitant loss of neutron activation analysis and nuclear reactor radionuclide production capabilities, and of teaching, training and outreach opportunities. In contrast, the number of medical cyclotrons has increased significantly both world-wide and in Canada during the past 10-15 years. Canada currently has some 28 operating cyclotrons with plans for more. While the primary function of the majority of these cyclotrons is to produce radionuclides for medical purposes the cyclotrons simultaneously generate neutrons as a by-product *via* (p,*x*n) reactions, for example. In the case of the University of Alberta TR-24 cyclotron a goal of the author is to utilize these neutrons for the elemental analysis of samples (using instrumental neutron activation analysis, INAA) and for teaching purposes, while not interfering in the use of the cyclotron to produce medical radionuclides. In this work the functionality of the TR-24 cyclotron as a neutron source for INAA and teaching purposes is demonstrated through the analysis of low grade uranium ores (and other samples).

Description

Low grade uranium ore samples (*ca*. 0.1% to 2% U) and certified uranium reference standards, weighing between 1.000 and 1.500 g, were hermetically sealed in individual 1.5 mL polyethylene irradiation vials. The samples (previously analyzed for their U-content by delayed neutron counting, DNC) and standards were simultaneously irradiated in batches during extended (5-10 hr) TR-24 ^{64}Cu and ^{203}Pb cyclotron production runs, for example. A rotating multiple sample holder, located behind a paraffin wax moderator, was positioned adjacent to the cyclotron sample target assembly to maximize the irradiating neutron flux. Following irradiation, and a minimum decay time of 12 h, samples and standards were individually counted (typically 10 to 30 minutes each) using a 40% efficient ORTEC GEM-FX Series Profile HPGe detector. Uranium was quantified by comparator NAA utilizing the 278 and 228 keV γ-emissions of ^{239}Np (T½ = 2.357 d) produced *via* the ^{238}U(n,γ)^{239}U -- β$^-$--> ^{239}Np reaction. Other activation products (*e.g.*, ^{56}Mn, ^{76}As, ^{24}Na) and some trace fission products (*e.g.*, ^{97}Zr, ^{99m}Tc, ^{133}I) were clearly identifiable in the acquired sample γ-ray spectra.

Results

Good agreement was found between the INAA and previously determined DNC U analyses.

Conclusions

While the neutron flux generated by a typical medical cyclotron during radionuclide production is several orders of magnitude lower than that available with a small research reactor this work demonstrates that the accurate determination of select elements by NAA is achievable. With the increased availability of medical cyclotrons at university hospitals, *etc*. the use of cyclotron-generated neutrons can be utilized for INAA and incorporated into many university courses (*e.g*., archaeology, geochemistry, analytical chemistry, physics, *etc*.).

FATE AND TRANSPORT OF RADIOCESIUM IN TERRESTRIAL SYSTEMS FROM FUKUSHIMA DAIICHI NUCLEAR PLANT

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Introduction

The largest amount of radiocaesium released into the terrestrial environment since the Chernobyl nuclear accident occurred at the Fukushima Daiichi Nuclear Power Plant (FDNPP). The surrounding land received 2.7 PBq of radiocaesium in forests, agricultural areas, grasslands, and urban areas, from which the radionuclides migrated through soil and river systems. Compared with the activity concentrations of ^{137}Cs, the activity concentrations of ^{90}Sr were generally 3 to 4 orders of magnitude lower, and have a very small impact on the terrestrial environment of Fukushima.

Description of the Work or Project

This presentation discusses the deposition and distribution of radionuclides, especially radiocaesium, in the terrestrial environment resulting from the FDNPP accident. Anthropogenic activities such as rice and vegetable cultivation and residential activities in the upstream area have led to a rapid decline in the activity concertation of ^{137}Cs of suspended sediment (SS) transport in the river network. This decline directly controls the dissolved ^{137}Cs concentration in the river water. Environmental and anthropogenic factors, such as government-led decontamination efforts, are accelerating this decline, influencing the subsequent transport and impact of radionuclides through the environment. The environmental consequences of the Fukushima accident are compared with those of Chornobyl. The relatively rapid remediation of the Fukushima region compared to the Chornobyl region is explained.

The extensive data sets acquired in response to the FDNPP accident have the potential to advance the understanding of Earth and environmental science, both basic and applied, beyond the impact of this one event. For example, combining natural tracers such as ^{210}Pbex and ^{7}Be with stable isotopes and these data could lead to advances in tracking radioactive transport and fate.

Conclusions

The dense and continuous monitoring network established in the aftermath of the accident provides detailed information on the fate and transport of radiocesium and an opportunity to validate natural radionuclides as tracers of sediment sources and for mass budgeting. This allows us to evaluate the behavior of substances and contaminants from different sources and organisms at local scales and predict material cycle responses to global environmental change.

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RADIUM SEPARATION FROM THORIUM AND MANUFACTURING OF TARGETS BY PRECIPITATION METHOD

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Introduction

^{225}Ac is an α-emitting radioisotope that has received widespread attention due to the discovery of the therapeutic effect of high-energy alpha particles on cancer [1]. There are two main production methods for ^{225}Ac. One of the popular methods is the ^{229}Th/^{225}Ac generator. Since ^{229}Th is in the Np series, ^{229}Th must be generated by transmutation. One of the generation methods is extracted from ^{233}U. However, ^{233}U is not accessible to treat in Japan and many countries, because it is regulated as special fissionable material. Therefore, we proposed a new method to obtain ^{229}Th by generating ^{228}Ra(n,g)reaction, ^{228}Ra is separated from ^{232}Th in radioactive equilibrium. Since thorium exists in the form of oxide in nature, and the chemical property of ThO_2 is very stable and difficult to dissolve in acid and alkali, in order to separate ^{228}Ra from ^{232}Th, we have used the thermochemical conversion method to pretreat ThO_2 powder and successfully converted ThO_2 into soluble thorium halide compounds. We dissolved these thorium halides with 11.33M HCl and 13.14M HNO_3 respectively [2]. In the present study, the separation of ^{228}Ra from Th in the above-mentioned solutions and the manufacturing of the radium target for neutron irradiation by precipitation method. Our proposed target of radium is based on magnesium oxide, since the carrier is required because ^{228}Ra is radioactive, and I adopted magnesium because magnesium has the smallest neutron capture cross-section among alkaline earth metals. Our idea is the manufacturing precursor of the target, i.e., magnesium carbonate, and separation of Ra from Th at one time. Magnesium hydroxide can be easily converted to magnesium oxide.

Experiments and Results

First, we carried out the coprecipitation experiments by using stable alkaline earth metal ions (Ca^{2+}, Sr^{2+}, Ba^{2+}) for the prediction of Ra^{2+} behaviors. The solution with $CaCl_2$ or $SrCl_2$ or $BaCl_2$ of the same concentration as radium ion was added with Na_2CO_3 to neutralize the acid in the solution, and then we added 1M Na_2CO_3 and 1M $MgCl_2$ to this solution. After this treatment, white precipitates were immediately produced. The precipitates were filtered after standing for one hour. The ratio of the added 1M Na_2CO_3 to 1M $MgCl_2$ is 4:1.Next, we carried out the coprecipitation experiment using the solutions of ThO_2, which were obtained by the thermochemical conversion method. We confirmed that almost thorium remained in the filtrate by ICP-MS analysis, and the radium is distributed in the precipitation side by gamma-ray analysis of daughter nuclide, ^{228}Ac.

Conclusion

We confirmed that radium can be successfully separated from thorium by the coprecipitation method, and we can obtain the precipitate used as the target material.

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Keywords: ^{229}Th/^{235}Ac generator, coprecipitation, radium, alkaline earth metals

Alpha production routes for medical radioisotopes

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Introduction

Over the past decade, interest has been growing in the use and application of radionuclides for medical diagnostics and therapeutics. Production of these radionuclides has historically been dominated by only a few nuclear pathways, those being either reactor production pathways utilizing neutron activation (n,γ) or cyclotron production pathways utilizing proton spallation (p,x). Other production pathways exist, for example photo-activation (γ,n) reactions using electron accelerators as well as spallation with heavier ions such as alpha spallation (α,n) and deuteron spallation (d,n). Only recently have these other production pathways seen modern investment and industrial development. Because of the lack of industrial investment, very few facilities exist which are capable of utilizing these alpha and deuteron production pathways. The focus of this work is to illustrate the unrealized benefits of using these pathways for radioisotope production. In this work we concentrate on several alpha production pathways to demonstrate the superiority of these methods for producing certain high-specific activity, non-carrier added radioisotopes for medical and industrial uses.

Description of the Work or Project

Using a purpose built Geant4 application the authors are able to compare the effectiveness of alpha vs. proton production pathways for several medically relevant radioisotopes, examples include Copper-67, Strontium-82, Cesium-131, Rhenium-186 and Astatine-211 among other proposed products. These production pathways are examined end-to-end, incorporating analysis of target materials, product yields, co-product and contaminate yields, competing reactions, post-bombardment decay, radio-chemical separation, radiopurity, waste, shielding and carrier added effects.

Conclusions

The results of this work illustrate that many factors impact the effectiveness and feasibility of a radionuclide production process. For a broad selection of specific radioisotopes, the alpha production pathway is superior for producing several high specific activity, high radio-purity non-carrier added products.

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Balkin E., Cutler C. (2017) Scale-up of high specific activity 186gRe production using graphite-encased thick 186W targets and demonstration of an efficient target recycling process. Brookhaven National Laboratory BNL-114854-2017-JAAM

KEYWORDS: Radioisotope, Copper-67, Strontium-82, Cesium-131, Astatine-211

MICROFLUIDIC ION-EXCHANGE APPLICATIONS FOR LANTHANIDES

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Introduction

Microfluidic separation strategies are an emerging area of interest in radiological separations, with advantages including a reduction in radiological waste volume, decrease in dose to handlers, and potential for automation. Ideally, radiological separations procedures would be optimized at the microfluidic level and then scaled up to production levels. A promising case study for microfluidic separations are radiolanthanides, which are difficult to separate because of their similar chemistry. Separations of lanthanides have yet to be performed via chromatography on the microscale. We anticipate that comparing micro- and macroscale separations of radiolanthanides will facilitate production scale-up.

Description of the Work and Results

Polydimethylsiloxane microfluidic chips were fabricated using standard protocols. The chips were packed with AG 50W x-12 resin to study lanthanide separations. Separation strategies of naturally occurring lanthanides were tested initially with high-pressure ion chromatography. Different detection methods were used to gain insight on the separation performance and to fit the models. Optimized separation strategies were tested with radiolanthanides.

Ion exchange separation of Ce and Tm was achieved at the microscale. A step gradient of eluent alpha-hydroxyisobutyric acid was used to separate both radiolanthanides. Further optimization of the elution profile is needed to achieve adjacent lanthanide separation.

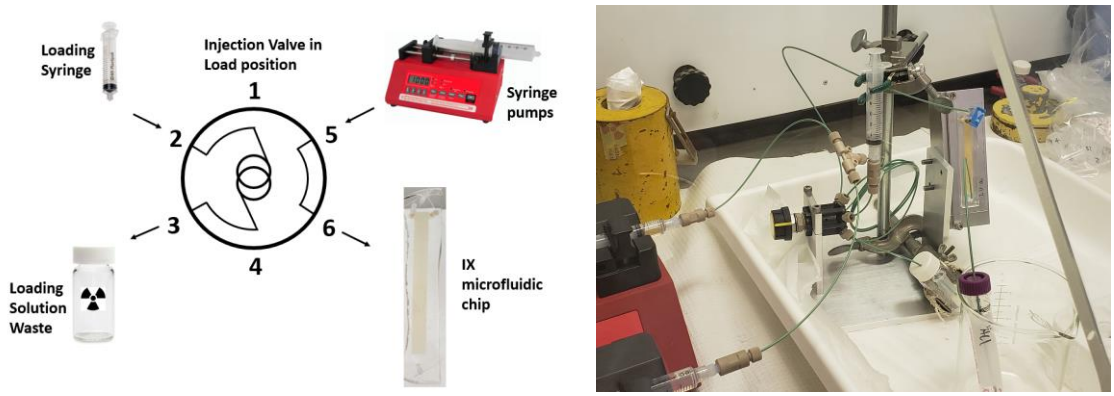

Figure 1. Microfluidic setup, including loading and injection stages that allow for isolation and automation of the radiological material.

Conclusions

Nonadjacent lanthanides can be separated in a microfluidic ion exchange chip. This microfluidic separation allowed for a reduction in radiological waste and dose experienced by handlers. Future work includes optimizing this separation strategy and developing a scale-up procedure.

EVALUATION OF AN INNOVATIVE ULTRA-COMPACT HPGe SPECTROMETER AS QUALITY CONTROL IN ^{177}Lu PRODUCTION PROCESS

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Introduction

Since their invention in the 1970s, high purity germanium (HPGe) detectors have been the gold standard for the detection and spectroscopy of gamma rays. Developments in the fabrication, packaging, and operation of HPGe detectors, as well as in readout electronics and signal processing, have led to their application in fields as diverse as fundamental nuclear and particle physics, nuclear security and safety, and medicine. This paper presents preliminary results of the evaluation of a novel ultra-compact electrically cooled germanium detector as a quality-control for radionuclide purity of ^{177}Lu samples.

Description of the Work or Project

In a relatively short period of time, ^{177}Lu has become a key radionuclide in a wide range of targeted radionuclide therapy (TRT) for treating various clinical conditions [1], primarily due to its relatively long decay time of 6.65 days, its chemical properties as well as the emission of moderate energy β- particles and low energy gamma rays, making it possible to handle relatively high activities with low dose during preparation while delivering localized treatment on the tissues and tumor cells near the surfaces of cavities [2].

In the direct production scheme of ^{177}Lu through irradiation of ^{176}Lu or ^{176}Yb in nuclear reactors, several Lu and Yb isotopes are produced having a variety of close gamma-ray lines. The use of HPGe detectors with their best separation power is key to identify and quantify the contamination lines from the lines of interest from ^{177}Lu (113 and 208 keV).

A small form-factor, rugged, easily deployable HPGe called the MicroGe [3] detector has been developed and evaluated for use is such quality-control applications for ^{177}Lu production process. The palm-size detector can be brought very close to a sample in highly confined environments, and the rugged and compact design ensures the excellent energy resolution performance expected of HPGe performance, even in high-activity and high-throughput measurements. The detector can go from storage to measurement in less than 30 minutes using miniature electrical cooling.

The detector system was used in different steps of ^{177}Lu radioisotope production process, for improving the purifications process. With respect to traditional HPGe detectors used in such applications, the new detector was optimized in terms of detection efficiency and overall size of the germanium crystal to ensure optimal efficiency at the relatively low energies of interest, while avoiding saturation of the detector due to high activity of the samples. A traditional germanium detector would have been relatively bulky and would have necessitated a significant collimator in these conditions.

Figure 2 shows a typical spectrum obtained with the MicroGe detector of the ^{177}Lu product on a Zn target. No clear gamma rays from ^{177}Lu are visible, mainly products form the activations of the Zn target. In Figure 3, the radioisotopes are chemically separated from the Zn target (black plot). In the following step, the purification of the solution is performed. At this stage, the activity of the ^{177}Lu is predominant, and the contaminants are difficult to detect. In order to obtain the percentage of contaminants, the purified solution is set to decay for 70 days. The resulting spectra is shown in red also in Figure 3.

Figure 1: MicroGe detector.

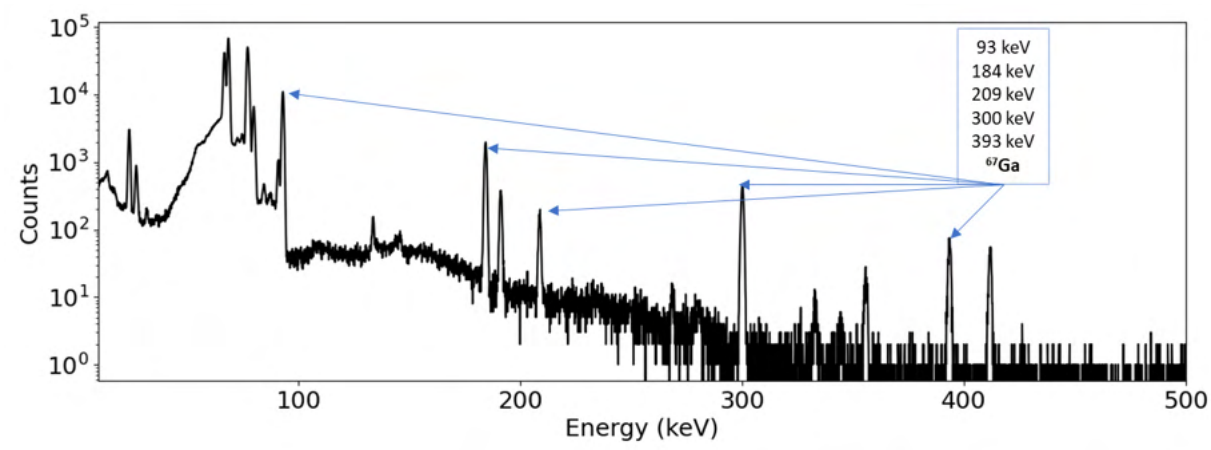

Figure 2: Typical spectrum obtained with MicroGe of the 177Lu product on a Zn target.

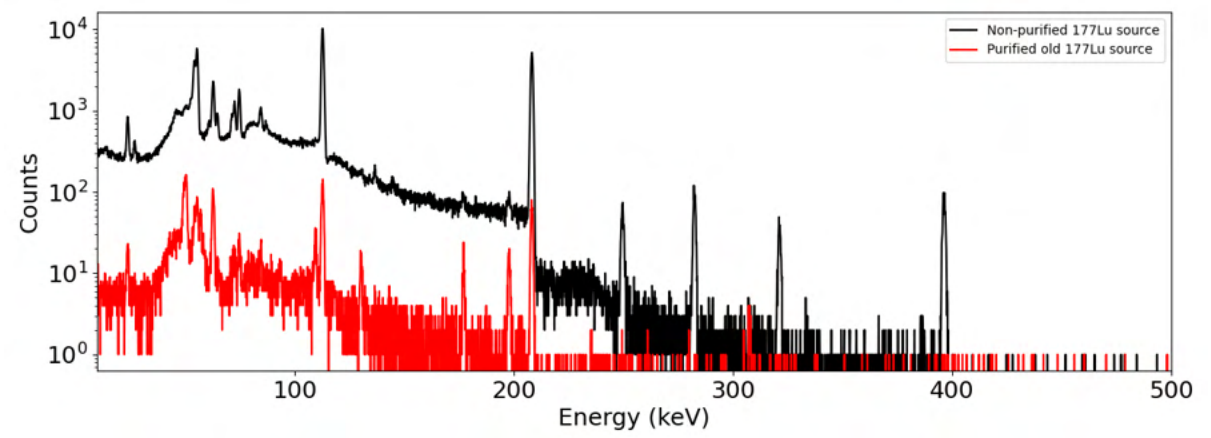

Figure 3: Typical spectrum obtained with MicroGe of the 177Lu solution (before and after purification and 70 days decay).

The energy density of the gamma-rays is particularly visible in the contaminant sample after the 70 days decay period, illustrating the need for the separating power of HPGe.

Conclusions

Detailed characteristics and performance of the new MicroGe detector will be presented. In particular, this work will show that the new detector is particularly suited for this application as it has an optimal efficiency for gamma ray detection in the range of 10-500 keV and allows the measurement of high activity sample at the beginning of the ^{177}Lu manufacturing process without the need of collimation. At the same time, bringing the MicroGe detector closer will allow to measure the decayed sample activity, thus avoiding any measurement bias that can exists if two different detectors were used, as well as reducing cost and complexity of the overall setup. Compatible with the LabSOC/ISOCS software, the MicroGe detector can be characterize and the ISOCS software can be used to compute efficiency calibration which are essential to determine the sample activities.

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PRECLINICAL EVALUATION OF LEAD-203 IMAGE-GUIDED LEAD-212 THERAPY FOR NEUROENDOCRINE TUMORS: THERAPY AND IMPLICATIONS FOR DOSIMETRY

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Introduction

Neuroendocrine tumors (NETs) are a rare, heterogeneous form of cancer. Current treatment using peptide-receptor radionuclide therapy with FDA approved ^{177}Lu-DOTATATE (LUTATHERA) improves progression free survival, but the objective tumor response observed in the Phase 3 trial was only 18%. Alpha-particle emitting radionuclides have emerged as a promising form of therapy for NETs. In our previous research, we optimized a peptide conjugate (PSC-PEG$_2$-TOC) for the Pb isotopes based on the structure of tyr^3-octreotide (TOC) to target somatostatin receptor subtype 2 (SSTR2), a G-protein coupled receptor that is highly expressed in NETs. This novel peptide conjugate demonstrated favorable *in vitro* binding affinity as well as *in vivo* pharmacokinetics and biodistribution characteristics in preclinical models. Here, we evaluated the therapeutic potential of the ^{212}Pb-labeled peptide conjugate and determine the estimated dose deposit in the tumor and normal organs of the mouse model using ^{203}Pb biodistribution results to evaluate the potential of dosimetry for treatment planning of ^{212}Pb targeted alpha therapy.

Description of the Work

A single (3.7 MBq) or thee fractions (3 × 1.2 MBq) of ^{212}Pb-PSC-PEG$_2$-TOC were administered to a AR42J tumor xenograft mouse model. Tumor progression, survival, and bodyweight changes of the treated mice and control mice were monitored over 120 days. The therapy study demonstrated the potential of ^{212}Pb-PSC-PEG$_2$-TOC for treatment of NETs. Three fractions of ^{212}Pb-PSC-PEG$_2$-TOC led to 70% complete response and an 80% survival rate at 120 days. The administrations of ^{212}Pb-PSC-PEG$_2$-TOC were well tolerated, presenting only early bodyweight reductions which were reversible in 10 days. Dosimetry analysis was conducted in the Medical Internal Radiation Dose (MIRD) schema. The biodistribution data of ^{203}Pb-PSC-PEG$_2$-TOC were used to obtain the time-activity curve of the ^{212}Pb-PSC-PEG$_2$-TOC in this model. The S-values in tumor and normal organs were estimated using the Particle and Heavy Ion Transport code System (PHITS) with the support of the DigiMouse voxel phantom model. The alpha doses per unit activity were estimated to be 8.65 Gy/MBq and 3.24 Gy/MBq in the tumor and the kidney respectively. Assuming no change in SSTR2 expression between the fractions, 3.6 MBq of ^{212}Pb-PSC-PEG$_2$-TOC resulted in 31 Gy in tumor and 11.7 Gy in the kidneys.

Conclusions

Our study demonstrates the significant potential of utilizing the theranostic pair, $^{203/212}$Pb, in conjunction with novel peptide-based radiopharmaceuticals for targeted alpha therapy in NETs. The dosimetry analysis conducted in this study revealed that 30 Gy of alpha particles could lead to a high probability of complete response, and the therapy was well tolerated. Further research is needed to investigate the long-term tolerability associated with this therapy and to better understand the alpha dose-toxicity relationship. The findings of this study have important implications for the development of targeted radionuclide therapy for NETs and could inform future clinical studies in this area.

Stable Isotope Target Fabrication at Oak Ridge National Laboratory

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Introduction

Oak Ridge National Laboratory (ORNL) is home to the National Stable Isotope Repository, a collection of more than 2,000 quality-controlled batches of 225 stable isotopes spanning 50 elements. The Stable Isotope Materials and Chemistry (SIMC) group maintains and dispenses the inventory forms of these materials and provides specialized technical services to convert materials to a customer's desired chemical form and fabricate a target of the desired physical form.

Description of the Work or Project

The SIMC group retains and utilizes a suite of techniques to prepare these customized target forms (Table 1). Due to the high monetary value of isotopically enriched material, these techniques are optimized for low-loss and material recovery. The SIMC group performs research and development to continually expand and improve upon these capabilities. This presentation will discuss the major processes and equipment used to manufacture a wide range of targets at ORNL and highlight recent method developments.

Table 1. Target fabrication techniques available at ORNL

Technique	Typical application
Pyrochemical and thermal conversions	Metal redox and contaminant removal
Inorganic synthesis	Convert metal and oxide samples to nitrate, carbonate, chloride, …
Powder pressing, melting, and sintering	Densified pellets (metals & complexes)
Hot and cold rolling	Thin metal foils
Wire casting, rolling, and swaging	Enriched wire fabrication
Arc melting, casting, alloying, and drop casting	Sputter targets, thick foils, ingots, pellets
Ion beam and plasma sputtering	Thin films (metals & complexes)
High vacuum evaporation	Thin films (metals & complexes)

Conclusions

Oak Ridge National Laboratory maintains an extensive collection of enriched, stable isotopes. Using a wide array of chemical and physical fabrication techniques, the SIMC group can provide these materials in the customer's desired form (e.g., powders, wires, foils, and discs) to help enable science across the world.

NEXT GENERATION RADIATION SPECTROSCOPY TOOLS

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Introduction

Spectroscopy tools designed in the 1960s relied on amplification, shaping and filtering techniques followed by DSPECs in the 1990s to bring about digital solutions, but continued to rely on the early NIM Bin style methods in one box. While offering improvements over prior systems, these spectroscopy tools still struggle with limited use cases, poor analytics and less than optimal resolution. Further, they offer little flexibility for analytics. Serva Energy has developed new radiation spectroscopy tools which allow for improved signal resolution and peak deconvolution for better isotope identification.

Description of the Work or Project

Serva Energy's DAQ system presents a flexible modular hardware architecture that includes separate preamplifier and ADC arrays. This design allows for compatibility with a variety of detector types (e.g. HPGe, SiLi, NaI, BGO, PIPSi, etc.). The system is capable of capturing time-synchronized data from multiple inputs and offers sampling rates of 250 MS/s. The accompanying software suite encompasses a Linux kernel driver, efficient low-level libraries for pulse detection, and a Python driver designed for high-level applications in data recording, visualization, analysis, and real-time radiation spectroscopy.

In contrast to Canberra/Ortec systems, these new systems preserve time information for each detector pulse, enabling the slicing of histograms along the time-axis and the application of a rolling window to observe the evolution of the histogram over time.

The ability to record and replay raw signals from the detector, coupled with seamless Python integration, enables the design of an effective Compton suppression system with just a few lines of code. Furthermore, this system offers the advantage of enhancing the results of previous measurements by applying updated tools and techniques to the existing data.

Previous Serva 25 MS/s spectroscopy hardware was capable of higher gamma peak resolution compared to commonly used DSPEC based systems[1]. The next generation Serva PCBs are equipped with 250 MS/s sample rates with substantially more flexibility through modular hardware architecture. This allows for new data analysis techniques of the raw signal in post processing to go beyond standard signal processing as seen below.

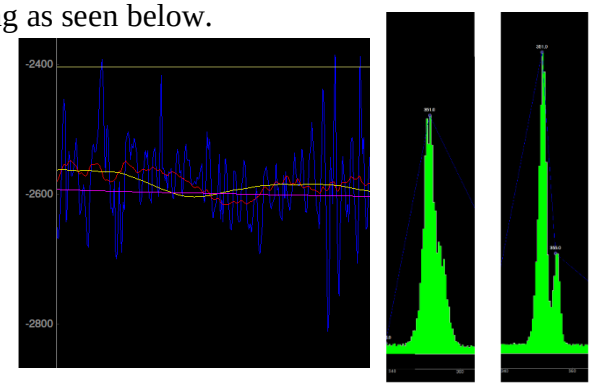

Figure 1 Detector signal - Blue is the raw signal noise; red, yellow and pink represent moving average stacking improved with genetic algorithms. Green peaks show unprocessed signal versus moving average stacking for peak deconvolution and improved resolution.

Peak convolution of differing isotopes with similar energy is often an issue; e.g. measurement of Ac225 in Ra226 and Ac227 where all Ac225 gamma emissions are separated by less than 2keV from those in the spectrum. Time stamps on recorded pulses allows for coupling to isotope tracking to automatically detect multiple peaks in even fully convoluted radiation energies due to known changes in activity over time.

Conclusions

Serva's modular ADC architecture coupled to raw data software analytics opens new avenues for radiation spectroscopy for identification of difficult to quantify radiation emission spectra across multiple detector types with higher precision than achievable with current systems.

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Novel Theranostic Platform for PET Imaging of Therapeutic Isotopes

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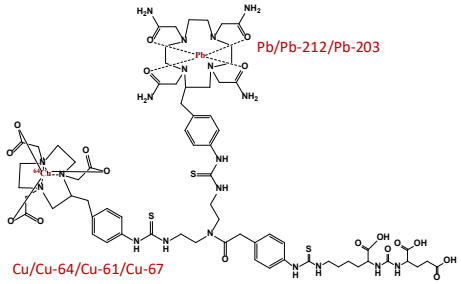
Introduction

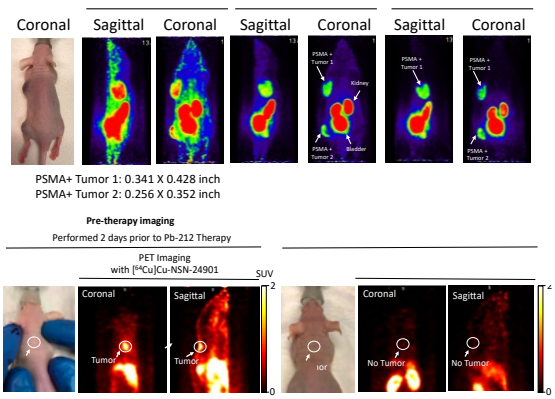
With a growing clinical interest in promising targeted alpha-radionuclide therapy, there is an unmet clinical need to accurately estimate the radiation dosimetry for the patients. There are only a few alpha-emitting radionuclides that could be potentially utilized for SPECT imaging, but others could not be. Alternatively, two chemically different molecules having different chelators and linkers can be employed, one for imaging and other for alpha radionuclide therapy with inherently different biodistribution and thereby suboptimal or inaccurate dosimetry. Herein, we propose a solution, which could solve the present problem and offers a robust platform approach to tailor the targeted alpha/beta-radionuclide therapy as a selective approach with a precise dosimetry estimation.

Description of the Work or Project

To solve this problem, we have created a novel molecular platform construct that allows accurate and precise dosimetry estimation by noninvasive PET imaging. In this construct, we have selectively added two chelators with a linker and a targeting vector in manner that creates two chemically identical/equivalent moieties one for PET imaging and other for alpha-radionuclide therapy with same biodistribution and thereby offering an accurate dosimetry estimation using PET. We have successfully created various versions of this construct and tested them in various in vivo pathological animal models with highly promising results both for PET imaging and alpha radionuclide therapy.



	Imaging pair	Therapy pair
NOTA	Cu-64	Cu-natural
TCMC	Pb-natural	Pb-212



Conclusions

Novel molecular construct have been successfully designed, developed, and evaluated in prostate tumor animal model to demonstrate its feasibility to accurately image with PET and treat tumor with alpha emitting radionuclide. Developed technology offers a simple solution to accurately measure the dosimetry for targeted alpha/beta radionuclide therapy.

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Poster Presentations

CERAD PROJECT AND 30 MEV CYCLOTRON FOR MEDICAL ISOTOPE PRODUCTION IN POLAND

Renata Mikolajczak

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In order to meet the high demand for radiometals with potential for medical applications, with a particular focus on their theranostic value, the new research facility is being built at NCBJ/POLATOM, with the name "Center of Design and Synthesis of Radiopharmaceuticals for Molecular Targeting, CERAD". It's main component is the 30 MeV cyclotron which will accelerate protons and alpha particles to 30 MeV and deuterons to 15 MeV. It will be a powerful tool for production of novel radioisotopes for medical use, which were not available in Poland up today. Among them the radioisotopes such as ^{18}F, $^{44/43}$Sc, ^{64}Cu, ^{67}Cu, Ge^{68}, ^{89}Zr, ^{123}I and ^{211}At will be produced. Installation of a new high-current cyclotron at NCBJ, with an equipment and infrastructure, combined with already existing scientific base, creates the unique and pro-development research capabilities.

The cyclotron of CERAD, Cyclone 30XP, has been already built by Belgian company Ion Beam Applications. Next to protons and deuterons it will also accelerate alpha particles. For that it is equipped with the alpha ion source. The cyclotron is installed in the new building which not only hosts the cyclotron but also a number of dedicated labs with hot-cells for radioisotope processing, with the QC and research laboratories. The entire facility offers a space of 2500 m^2. The upcoming infrastructure will be open to PRISMAP, SECURE and other user projects.

The infrastructure of CERAD can be used for both research and commercial activities, it creates the platform for comprehensive studies oriented at research and design of new medicinal products, in particular radiopharmaceuticals, and at implementing diagnostic and therapeutic procedures for diseases, which are currently treated ineffectively. The CERAD project has found its place on the Polish Roadmap of Large Research Infrastructure because it will not only offer new radioisotopes but also a possibility to design innovative radiopharmaceuticals. The research potential of NCBJ as consortium leader is supported by partner institutions: University of Warsaw, Warsaw Medical University, Institute of Nuclear Chemistry and Technology, Jagiellonian University Medical College and Medical University of Bialystok.

CERAD project is co-financed under Smart Growth Operational Programme 2014-2020, Priority IV: INCREASING THE RESEARCH POTENTIAL, Measure 4.2. Development of modern research infrastructure of the science sector.

Bulk-scale formulation and evaluation of 'ready-to-use' human clinical doses of [^{177}Lu]Lu-DOTMP using low specific activity ^{177}Lu

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Introduction

Lu-177 complex of cyclic polyaminophosphonic acid ligand DOTMP have been reported to demonstrate excellent features for palliative care of bone pain in preliminary clinical investigations in patients with skeletal metastases. Thanks to the macrocyclic ligand framework of DOTMP, [^{177}Lu]Lu-DOTMP complex could be formulated with excellent stability using significantly lower [L]/[M] ratio as compared to its acyclic counterpart, [^{177}Lu]Lu-EDTMP. Taking advantage of this, we have worked toward the utilization of low specific activity (LSA) ^{177}Lu produced by direct neutron activation of Lu_2O_3 target of natural isotopic composition in the formulation of [^{177}Lu]Lu-DOTMP for its human clinical use. Bulk-scale formulation of 'ready-to-use' doses of [^{177}Lu]Lu-DOTMP using LSA ^{177}Lu and its evaluation in pre-clinical as well as clinical settings is described.

The description of the work and results

Lutetium-177 produced by thermal neutron irradiation of Lu_2O_3 (natural isotopic composition, 2.6% in ^{176}Lu) at neutron flux of ~1.5×10^{14} n/cm^2.s for 14 d, which yielded specific activity of 19.6 ± 1.6 GBq/mg (530 ± 60 mCi/mg) at the end of irradiation (EOI). The radionuclide purity of ^{177}Lu produced was found to be 99.98±0.01% at EOI, with ^{177m}Lu being the radionuclidic impurity detected.

A one step procedure for formulation of ~ 37 GBq (1 Ci) of [^{177}Lu]Lu-DOTMP was optimized. In this, 100 mg DOTMP, 140 mg $NaHCO_3$ was dissolved in demineralized water and mixed with 37 GBq (~1 Ci) [^{177}Lu]$LuCl_3$. The mixture (pH ~ 7.5) was kept at room temperature for 30 min. Subsequently, the mixture was diluted with sterile physiological saline such that the radioactive concentration was adjusted to 740 MBq/mL (20 mCi/mL). ^{177}Lu-DOTMP solution is passed through 0.22 micron Millipore® filter to render the preparation sterile. The yields and radiochemical purities of the formulation were found to be 99.2±0.3% and 98.5±0.3% (n=8). The formulation was found to have a shelf-life of 14 d. Biodistribution studies in normal Wistar rats showed specific accumulation and retention of ~55% activity of injected formulation in skeleton along with clearance of remaining activity through urinary route. Post-therapy whole-body scan of a patient administered with ~2.6 GBq (~70 mCi) dose showed specific skeletal accumulation with preferential localization in the osteoblastic lesion (Fig. 1).

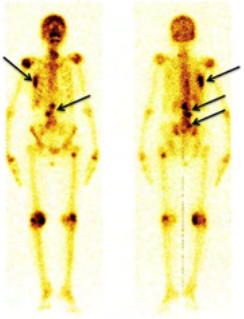

Fig.: ***Post therapy whole body scan of a 69 year old female patient with multiple skeletal metastases due to primary carcinoma in breast recorded 24 h post-administration of 2.6 GBq of ^{177}Lu-DOTMP prepared utilising ^{177}Lu produced from natural Lu_2O_3 target***

Conclusions

The studies reported herein, demonstrated excellent potential of ready-to-use formulation of [^{177}Lu]Lu-DOTMP prepared using LSA ^{177}Lu as a cost-effective radiopharmaceutical affordable to a large population of patients in the developing countries.

DEVELOPMENT OF C-14 ON-LINE MONITORING SYSTEM USING ROBOTICS TECHNOLOGY

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Introduction

Radioactive carbon is produced by a reaction of $^{17}O(n, \alpha)^{14}C$, $^{14}N(n, p)^{14}C$, $^{13}C(n, \gamma)^{14}C$ when operating a nuclear power plant, and is discharged as gaseous form of $^{14}CO_2$ and $^{14}C_nH_m$ through VCT. ^{14}C is a very important nuclide at the disposal site and NPPs. In this research a fully automated ^{14}C monitoring system that could perform, without an operator, all processes, such as collection of gaseous ^{14}C in NaOH solution, mixing with cocktail and measurement with LSC, etc. to continuously monitor gaseous ^{14}C discharged from nuclear facilities was developed using robotics technology.

Composition of ^{14}C Monitoring System

The ^{14}C monitoring system consists of a collection unit, a robot(mixing) unit, and a measurement unit. In the collection unit, gas samples including ^{14}C discharged in nuclear facilities are automatically collected in 1M NaOH solution at specified intervals. In the mixing part, the vial cap is opened and closed automatically using a robot arm and a gripper, and a collected solution and a fluorescent solution are quantitatively injected to vial using a peristaltic pump and a solenoid valve to produce a cocktail. When the robot arm opens the door of the LSC and places the vial in the specified position, and the robot arm closes the door, measurement begins. The above three parts are connected to each other and are configured to be able to operate without an operator through a PLC/HMI program. Figure 1 shows the 3D modeling and manufactured ^{14}C monitoring system developed in this study.

Figure 1. 3D modeling and manufactured fully automated ^{14}C monitoring system.

Conclusions

Using the developed ^{14}C monitoring system, $^{14}CO_2$ and $^{14}C_nH_m$ types of radioactive carbon can be automatically monitored, and if gas samples are collected every two weeks, ^{14}C can be monitored online for up to 16 weeks without an analyst.

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New study on the Preparation of Low Specific Activity $^{99}Mo/^{99m}Tc$ generator

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Introduction

^{99m}Tc is the most used diagnostic medical radioisotope for SPECT, comprising 80% of all diagnostic medicine.[1] ^{99m}Tc is obtained via the eluting of $^{99}Mo/^{99m}Tc$ generator system. Currently, ^{99}Mo is mainly generated from (n, f) reaction in reactor using highly enriched uranium-235 (^{235}U, HEU, Enrichment factor of ^{235}U > 90%) target all over the world.[2] But the use of uranium increases the probability of nuclear proliferation risk coming up. In addition, the purification of ^{99}Mo from irradiated HEU target is so complex and produce a lot of radioactive waste. Therefore, many new techniques for^{99}Mo production should be investiagted.

Reactor/accelerator-based production of ^{99}Mo using enriched Mo($^{98/100}Mo$) target such as the $^{98}Mo(n, \gamma)^{99}Mo$, $^{100}Mo(p, pn)^{99}Mo$ and $^{100}Mo(\gamma, n)^{99}Mo$, have been greatly explored. These are the least intricate ways to produce ^{99}Mo. However, the first problem faced that the ordinary alumina (2-20 mg Mo per g of alumina) which used for fission $^{99}Mo/^{99m}Tc$ generator is not suitable for preparation of low specific activity (LSA) $^{99}Mo/^{99m}Tc$ generator. So the development of column sorbent which has high sorption capacity for Mo ions is urgent. For another, the enriched Mo, which is so expensive, should be recycled and reused for irradiation. Thus, the LSA $^{99}Mo/^{99m}Tc$ generator is still worthy studying.

Description of the Work or Project

In this work, we proposed that hierarchically macro-/mesoporous γ-Al_2O_3 (HMMA) can be used for the preparation of LSA $^{99}Mo/^{99m}Tc$ generator. HMMA immobilizes the mesoporous (2-50 nm) into macropores (>50 nm) materials, can reduce the internal diffusion limitations and improve the high activity to absorb Mo ions. Firstly, HMMA was synthesized and characterized. Second, the absorption of Mo ions using HMMA as sorbent in static and dynamic conditions were studied. Fourthly, $^{99}Mo/^{99m}Tc$ generator used HMMA as column matrix was prepared. Lastly, the recovery of Mo absorbed on column was also investigated.

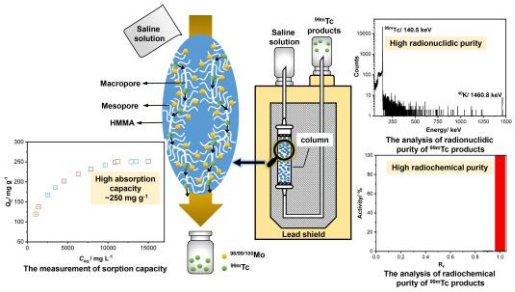

Figure The preparation of LSA $^{99}Mo/^{99m}Tc$ generator

Conclusions

HMMA exhibited high capacity to Mo ions with about 250 mg Mo per g of HMMA. A ^{99}Mo generator was prepared successfully. In the meantime, Mo were recovered in a high yield of 95%. HMMA has a potential application prospect for the preparation of LSA $^{99}Mo/^{99m}Tc$ generator.

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MEASUREMENTS OF NUCLEAR REACTION CROSS-SECTION ABOVE 70 MEV IN ENRICHED ZINC-70 TARGET WITH 100 MEV PROTONS

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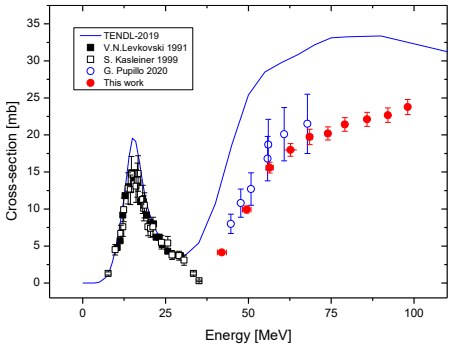
Introduction

Among various nuclear reactions for ^{67}Cu, the most famous reactions for ^{67}Cu production are ^{68}Zn(p,2p)^{67}Cu using a high energy proton beam, and ^{70}Zn(p,x)^{67}Cu using a low energy proton beam. Recently, Pupillo *et al*. reported [1] that the nuclear reaction cross-section of ^{70}Zn(p,x)^{67}Cu using enriched ^{70}Zn targets is 70 % higher than that of ^{68}Zn(p,2p)^{67}Cu in an energy range of 45-70 MeV. Despite their good results for the ^{70}Zn(p,x)^{67}Cu reaction, the data above 70 MeV have not been explored so far. In this study, for the first time, we obtained the ^{70}Zn(p,x)^{67}Cu reaction data in a range of 70-100 MeV using a 100 MeV proton accelerator.

Description of the Work or Project

In experiments, an enriched ^{70}Zn target with a concentration of 97.5 % was purchased from Neonest AB. A sample was prepared to the conventional stack-foils consisting of ^{70}Zn metal pellets, aluminum foils for beam monitor, and aluminum sheet for energy degrader. Proton beam irradiation was performed on a 100 MeV proton linear accelerator at Korea Multi-purpose Accelerator Complex (KOMAC), Korea. To calculate the nuclear reaction cross-section of ^{70}Zn(p,x)^{67}Cu and ^{70}Zn(p,x)^{67}Ga, the peak separation method developed by Park *et al.* was applied [2]. Figure 1 shows the excitation function of the ^{70}Zn(p,x)^{67}Cu nuclear reaction obtained from this work and other previous works.



Conclusions

We have successfully obtained the nuclear reaction from the enriched ^{70}Zn target using high energy protons above 70 MeV. IAEA announces ^{67}Cu for a new emerging beta emitter, “theranostic” radionuclide that requires to be as much supplied as possible. Therefore, we proposed using an enriched ^{70}Zn target in order to maximize ^{67}Cu production using a high-energy proton beam even if the price of enriched ^{70}Zn is more expensive than that of ^{68}Zn.

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EVALUATION OF CLINICAL OF ^{64}Cu-RMNP(RADIOACTIVE MAGNETIC NANOPARTICLES) FOR THE DEVELOPMENT OF ^{67}Cu UTILIZATION TECHNOLOGY

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Introduction

To date, for positron emission tomography (PET), chelator-based radio-labeling is the widely used technique for the synthesis of various kinds of radio-labeled nanoparticles (NPs). Nanoparticles with radioactive and magnetic properties can be used in PET-MRI contrast agents. Conventionally, PET-MRI contrast agents that label magnetic superparamagnetic iron oxide (SPIO) with radioactive isotopes on the surface of nanoparticles with a chelator such as DOTA or NOTA have been studied.[1] However, these contrast agents can partially dissociate radioisotope bound to the chelator in vivo.[2] As the result, the radioisotope and the magnetic property can separated, so it can be occured misdiagnosis. Therefore, the multimodality nanoparticle was synthesized to prevent such misdiagnosis. This contrast agent technology can play a significant role in developing nuclear medicine imaging technology.

Description of the Work or Project

This study synthesized nanoparticles with simultaneous radioactive and magnetic properties using ^{64}Cu by an optimized synthesis method identified as a stable isotope. Fe element was used for MRI images and ^{64}Cu was used for PET images. Nanoparticles were synthesized by a hydrothermal reaction at 200 ° C. using a direct stirrer at 200 rpm. The surface of the recovered nanoparticles was synthesized in core-shell form using tetraethylorthosilicate to reduce the toxicity of the metal. It was confirmed that the size of the recovered RMNP was optimized to about 5 nm, and the size of the core-shell nanoparticles was optimized to about 30 nm.

Conclusions

The synthesized product was confirmed that the size of the recovered RMNP was optimized to about 5 nm, and the size of the core-shell nanoparticles was optimized to about 30 nm. In addition, it was confirmed the magnetic properties by SQUID and MRI measurements, and the possibility of diagnostic was confirmed by PET images.

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DEVELOPMENT OF AN EFFICIENT TECHNIQUE TO SEPARATE CARRIER-FREE ^{47}SC ADEQUATE FOR RADIOTHERANOSTIC APPLICATIONS THROUGH ^{NAT}TI(N, P) REACTION

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Introduction

Cancer theranostics is a new emerging approach of employing one molecular vector to render diagnosis beside targeted radiotherapy, and subsequently facilitate personalized targeted radiotherapy. Recently, ^{47}Sc labeled with a definite ligand considered a promising radiopharmaceutical to be used in radiotheranostics. So, it appeared worthwhile to investigate the production and separation of ^{47}Sc, and this work focuses on the effective separation of no-carrier-added (NCA) ^{47}Sc from natural Ti target using newly synthesized resin material.

Description of the Work or Project

Natural Titanium target was irradiated in the Egyptian Second Research Reactor (ETRR-2) at a fast neutron flux of 1.63 x 10^{13} n cm^{-2} s^{-1} for 24 h. To allow short-lived radionuclides to decay, the irradiated target was left for three days, then a concentrated solution of Hydrofluoric acid was used to dissolve the target. ^{47}Sc produced from the fast nuclear reaction was effectively separated from bulk titanium target by a newly synthesized resin material which characterized by different techniques, such as FT-IR, SEM, TGA and DTA to confirm its structural morphology and mechanical properties. The separation process was carried out using chromatographic column method based on high separation factor between Sc and Ti on the synthesized resin material. Different parameters, such as flow rate, column internal diameter and amount of ion exchanger were studied for optimization the elution profile of ^{47}Sc. Finally, three quality control tests; radionuclidic, radiochemical and chemical purities was performed on the eluted ^{47}Sc to examine its validity for application in radiopharmaceutical preparations.

Conclusions

^{47}Sc was successfully produced at the Egyptian Second Research Reactor (ETRR-2) of 70 MBq after 24 h irradiation. Since the natural titanium target has different isotopes of different natural abundancies, many radioisotopes co-produced with ^{47}Sc, which need an efficient separation method to be separated. A low cost and effective resin material was synthesized by polymerization technique to increase the structural stability as well as the functional groups that responsible for the separation. After that, the batch method was applied to investigate the adsorption affinity of Sc and Ti ions towards the resin material under the influence of pH. Based on the Kd values obtained, ^{47}Sc was easily separated from bulk titanium and other radioimpurities by chromatographic column with elution efficiency of 96 ± 0.3 %. Furthermore, the eluted ^{47}Sc was of high chemical, radionuclidic and radiochemical purities and can be safely applied in radiotheranostics applications.

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Evaluation of Radiological Hazard parameter of building raw materials in South of Korea

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Introduction

Naturally Occurring Radioactive Materials (NORM) exist in all construction materials collected from nature, and as these are used as raw materials for apartments, humanity is being constantly exposed to natural radiation. Currently, apartments are over 80% of the total number of houses, some of which are 50 years old in Korea, and most construction materials are obtained from these natural materials. Building materials are managed by applying the radioactivity level index (EU RP112) in the S'Korea.

Building raw materials of Radiological Hazard Parameter

To measure natural radionuclides, uranium decay series ^{226}Ra, thorium decay series ^{232}Th, and ^{40}K were analyzed using a high-purity germanium gamma spectroscopy analyzer (HPGe).
The radioactivity level index, radium equivalent radiation, external exposure index, internal exposure index, absorbed dose, and annual effective dose can be calculated from the analyzed radioactivity concentration of the radionuclides (Radiological Hazard Parameter).
Gamma spectroscopy analysis was performed on the sand, aggregate, cement, concrete, brick, tile, and paint materials used as building raw materials, and radioactive hazard parameter values were calculated based on the measured radioactivity values.

Conclusions

Table 1. Building raw materials value

LIST	n	R_{aq}	I_r	H_{ex}	H_{in}	D	D_{eff}
[1)]Sand	24	230.59	0.87	0.65	0.73	108.79	0.53
[2)]Aggregate	124	201.23	0.75	0.57	0.65	94.79	0.47
Cement	39	150.38	0.54	0.44	0.57	69.44	0.34
Concrete	26	162.66	0.61	0.46	0.54	76.96	0.38
Brick	7	154.65	0.23	0.44	0.53	72.82	0.36
Paint raw material	7	116.38	0.07	0.35	0.45	54.68	0.27
[3D)]Europe member states, 23	10k	121.81	0.45	0.35	0.43	57.31	0.28
UNSCEAR 2008	-	370	≦1	≦1	≦1	84	0.48

1) not mineral sand (without Zirconium sand, Bauxite, Ilmenite, Rutile)
2) Crushed Aggregate (< 5 mm), Coarse Aggregate (10-25 mm)
3) ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in European brick, concrete, cement, natural gypsum: comparison with the European soil. Average values and ranges, UNSCEAR 2000.

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THE 1st ^{82}Sr PRODUCTION AND ^{82}Sr/^{82}Rb GENERATOR COLUMN CHARACTERIZATION IN KOREA

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Introduction

^{82}Sr/^{82}Rb generators are widely used for the diagnostics of cardiovascular decease using Positron Emission Tomography (PET). ^{82}Sr, a parent radionuclide of ^{82}Rb, is made by irradiating Rubidium Chloride (RbCl) targets with proton beams generated from high-energy linear accelerators or cyclotrons. While ^{82}Sr is produced in various countries around the world and two types of ^{82}Rb generators approved by the FDA are widely used, ^{82}Sr has not yet been produced or used in Korea. Since many years ago, studies have been conducted on the production of ^{82}Sr using KOMAC (Korea Multi-purpose Accelerator Complex), a 100 MeV linear proton accelerator. This paper reports the results of the whole process from the production of ^{82}Sr to the manufacture of ^{82}Rb generator columns and ^{82}Rb elution characteristics for the first time in Korea.

Description of the Work or Project

^{82}Sr was produced through a separate purification process using Chelex-100 resin after irradiating the proton beam on the RbCl target at the target irradiation facility installed at the end of the RI-dedicated beamline of the 100 MeV linear proton accelerator. The size of the target was 7.5 mm thick and 30 mm in diameter, and the average current of the proton beam was 1.2 μA for irradiation time of 150 minutes. The activity of ^{82}Sr produced was 43.4 μCi, and the separation purification yield was > 85 %. Hydrous tin oxide was selected as the adsorbent to be filled in the generator column, and the total length of the column was 4 cm and the volume of the adsorbent was 1.76 cm^3. The adsorption yield of ^{82}Sr into the generator adsorbent was > 99 %, and the total amount of ^{82}Sr adsorbed to the generator was 21.6 μCi as of the day of the ^{82}Rb elution experiment. In the ^{82}Rb elution experiment using saline, the ^{82}Rb elution yield varies depending on the flow rate. When the elution amount was 22 mL, the ^{82}Rb elution yield was 55.1 to 93.3%. When the flow rates were 5.0, 10.2, 14.5 and 22.0 mL/min, the yield was 55.1, 76.9, 86.2 and 93.3 %, respectively, and the elution yield increased as the flow rate increased.
After the eluted ^{82}Rb was filled in the correction phantom of the small PET for animals, a PET image was taken. The image capture time was set to 10 minutes, and the phantom PET image was successfully obtained. Impurity analysis was performed on eluted ^{82}Rb using ICP-MS, and Rb stable isotopes that compete in vivo of ^{82}Rb were identified as undetected levels and were determined to be No-Carrier-Added (NCA).

Conclusions

For the first time in Korea, ^{82}Sr production, ^{82}Rb generator column manufacturing, ^{82}Rb elution, PET phantom image acquisition using ^{82}Rb, and impurity analysis were successfully performed. In order to utilize the ^{82}Rb generator in Korea for research purposes, the activity of ^{82}Sr and the diagnosis of PET images of small animals using ^{82}Rb will be carried out continuously.

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ANALYSIS OF ISOTOPES PRESENT IN THE HAVAR® FOIL SAMPLE OF THE PETtrace 800 SERIES CYCLOTRON ACCELERATOR

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Introduction

Our medical cyclotron of 16 MeV protons is one among the many around the world which employ a Havar® foil as vacuum seal, exposed to prolonged irradiations of several months before they are replaced by a fresh foils. The radio activity levels of these foils are very high and they require very careful handling and storage. It is conceivable that more frequent changes of foils will mean easy to handle lower radio activities levels and the foils can be reused several times. We undertook a study to evaluate the possibility of such protocols.

Description of the Work

Havar® foil is an alloy containing Co (42%), Cr (19.5%), Ni (12.7%), W (2.7%), Mo (2.2%), Mn (1.6 %), C (0.2%), Fe (19.1%) [1], which upon irradiation by 16 – 24 MeV protons at cyclotrons result in myriad of radio isotopes of long and short half-lives with multiple gamma ray emissions. The shortest half-life is ^{60}Cu of $T_{1/2} \sim 0.4$ hours, while longest is ^{54}Mn of $T_{1/2} \sim$ 7500 hours. The major contributors for the radio activities are (p,a), (p,n), (p,2n) and (p,pn) reactions on Cr, Fe, Ni and Co isotopes for which the cross section data is available at the NNDC website [2] to be about a few micro barns to milli barns. We estimated the yields for a 25 – micron thick Havar® foil, 50 – micron thick Havar® foil and deduced the resulting radioactivities. It is found that the short – term radiation levels are dominated by those of ^{60}Cu, while the long – term activities are due to ^{56}Co and the rest of isotopes with varying levels of radiations.

Conclusions

We have made numerical estimates of the relative contributions of radio activities of isotopes produced in a Havar® foil by proton irradiation of medical accelerators in short – term and long – term. We are planning to quantify the numbers from experiments at our cyclotron. From these data, we will assess the protocols of irradiation to recycle the Havar® foils without accumulating radiations of hazardous levels. These results, which are of interest for cyclotron operators, will be presented at the conference.

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HEMISPHERICAL DOSE CALCULATION OF DISPERSED RADIOISOTPES

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Introduction

Releasing radioactive materials in the atmosphere is very important from viewpoint of radiological impact assessments. It is necessary to predict the behavior and absorbed dose of radioactive materials that are dispersed in the air in case of a hypothetical radioactive accident. We proposed different methods for modeling aerosol dispersed radioactive materials behavior in the air and an innovative way of calculating absorbed dose.

Materials and method

Gaussian plume model was used for calculating the amount of radioactive concentration in the air. Then, we assumed a hypothetical air hemisphere with the assigned radius for center dose calculation. After modeling, air concentration is assumed to be dispersed in the hemisphere and gamma flux reached to the center was calculated. Gamma attenuation in the air, with the probability of about 99.32%, is 5/μ, is the radius of the pre-assumed hemisphere.

Results and discussion

We compared and benchmarked the results of our work with PC-CREAM software for the same investigation conditions. For instance, for Cs-137 behavior with Table 1 conditions, activity concentration was calculated and comparative results are depicted in Fig. 1.

Table 1: Evaluation conditions of Cs-137 diffusion in the air	
Source height	100 m
Rate of release in the air	1 Bq/s
Ground type	Farm
Mixing layer height	1300 m
Wind speed	1 m/s
Stability class	A

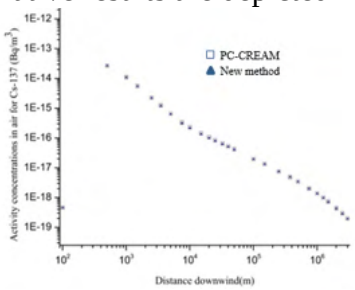

Figure 1: Comparison of results with PC-CREAM

Conclusions

Results show that our method has a good following of the PC-CREAM results and errors are negligible. We proposed a successful method of dose calculation of radioactive air dispersion.

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Keywords: Gaussian, Radioactive, Plume model, PC-CREAM, Absorbed dose

Isomeric yield ratios $^{59}Co(\gamma,n)^{58m,g}$ Co reaction photo-product at energy $E_{\gamma max}$ up to 80 MeV

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Introduction

Using the isomeric yield data, both the nuclear reaction mechanism and statistical properties of the excited state of atomic nuclei can be investigated. However, the information regarding the formation of isomeric-states is rather scanty and some discrepancies are still observed among the literature values which might be attributed to variations in experimental methods and/or the nuclear constants [1,2]. Moreover, the experimental database is also outdated because of the improvement of the sensitivity of detection system and the refinement of the spectroscopic features. Therefore, the update of the measurement of the isomeric yield ratios has been reissued recently.

Description of the Work or Project

The isomeric yield ratios for the ^{58m,g}Co were performed using the activation method at Pohang Accelerator Laboratory (PAL). We have measured the isomeric yield ratios for the $^{59}Co(\gamma,n)^{58m,g}$ Co reaction with bremsstrahlung end-point energies 60, 65, 70, 75, and 80 MeV by using the activation and the off-line γ-ray spectrometric technique in the electron linac Pohang Accelerator Laboratory, Korea. The aim of the present work is to measure the isomeric yield ratios of the ^{58m,g}Co with bremsstrahlung energy of 60- to 80-MeV with a step of $\Delta E = 5$ MeV from natural Cobalt. The induced activities in the irradiated foils were measured by the high-resolution γ-ray spectrometric system consisting of a high-purity germanium detector and a multi channel analyzer. The necessary corrections were made to improve the accuracy of the experimental results. The isomeric yield ratio of ^{58m,g}Co have been measured form reactions $^{59}Co(\gamma,n)^{58m,g}$Co. The IR values of ^{58m,g}Co from ^{59}Co target is reported to be 0.524±0.054, 0.503 ± 0.052, 0.507 ± 0.049, 0.515 ± 0.053 and 0.465 ± 0.049 for the bremsstrahlung photon energy 60, 65, 70, 75 and 80 MeV respectively. The present isomeric yield ratios for the ^{58m,g}Co induced by 60, 65, 70, 75, and 80 MeV bremsstrahlung are measured by using the activation method. The obtained isomeric yield ratios are compared with the calculated values based on the statistical model code TALYS.

Conclusions

The reaction $^{59}Co(\gamma,n)^{58m,g}$Co was studied for the first time which has no comparable literature data. The present experiment is based on (γ,n) reaction which is the first time measurement at intermediate energy bremsstrahlung 60, 65, 70, 75, and 80 MeV from ^{59}Co target. From the reaction studied in present measurement, it is observable that isomeric ratio is dependent on the spin of the target nuclei.

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Measurement of isomeric yield ratios of ^{174m,g}Lu produced in (γ,xn) photonuclear reaction of ^{nat}Lu

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Introduction

Studies of isomeric ratios are of considerable importance for both basic nuclear physics research and applications. The isomeric ratio depends on the spin distribution of the compound nuclei, the angular momentum carried away by the emitted particles, the character of the γ-cascade, and the spins of the isomeric states [1]. Since the interest of accelerator driven subcritical system (ADSs) is rapidly growing, a photo-nuclear data will be one of the useful data for the designing of such system [2].

In this work, the isomeric yield ratio of ^{174m,g}Lu produced in the ^{nat}Lu(γ,xn) reactions with end-point bremsstrahlung energies of 55-, 60-, and 65-MeV have been determined by using activation and off-line γ-ray spectrometric technique at Pohang Accelerator Laboratory (PAL), Pohang, Korea.

Description of the Work or Project

The photo-activation method was used to determine the isomeric yield ratios of the ^{nat}Lu(γ,xn)^{174m,g}Lu reaction. The produced nuclides in the irradiated foil together with reaction predecessors were identified based on the known spectroscopic data, such as energy and half-lives. The isomeric yield ratios were calculated from the measured activities of the high-spin state and the low-spin state of the produced radioisotope. The ^{174m,g}Lu isomeric pair were identified based on their characteristic γ-ray energies and half-lives. The isomeric-state ^{174m}Lu (high-spin state, 6-) with a half-life of 142 d decays directly to the unstable ground-state (low-spin state, 2-) by emitting the 76.47-keV γ-rays with a branching ratio 99.38 %. The measured values of IR of ^{174m,g}Lu are listed in Table 1

Table 1 : Isomeric yield ratios of ^{174m,g}Lu via photonuclear reactions with ^{nat}Lu

Nuclear reaction	Threshold energy (MeV)	Photon Energy (MeV)	IR ($Y_{high\text{-}spin}$ /$Y_{low\text{-}spin}$)
$^{nat}Li(\gamma,xn)^{174m,g}Lu$	14.04	55	1.035±0.08
		60	0.951±0.09
		65	0.930±0.08

Conclusions

The present experiment is based on (γ,xn) reactions which is the first time measurement at intermediate energy bremsstrahlung 55-, 60-, and 65-MeV from ^{nat}Lu target. The results provide complete data for this region and could contribute to the Nuclear Data.

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Y-90 SOL - GEL MICROSPHERES FOR RADIOEMBOLIZATION THERAPY AND DOSIMETRY METHOD FOR DETERMINATION OF ADMINISTERED DOSE

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Introduction

Radioembolization with radioactive microspheres has gained clinical acceptance as a therapeutic option for patients with liver malignancies. The presented work consist of two subjects, a first - a new technology for the production of radiopharmaceutical, ^{90}Y microspheres in the form of spherical yttrium oxide particles obtained by sol-gel method and the second-one - elaboration of a dosimetry method that allows the verification of the therapeutic doses evaluation in SIRT (Selective Internal Radiation Therapy) therapy.

Description of the Work

The authors present and discuss the results of investigations performed in the development of new production technology of yttrium microspheres and determination of their physic-chemical properties. The final product has the structure of spherical yttrium oxide grains with a diameter 25-100 μm, is stable and free from contaminants. Irradiation of 20 mg samples of grains with diameter of 20-50 μm in the thermal neutron flux of $1.7x10^{14}$ $cm^{-2}s^{-1}$ at the core of MARIA research nuclear reactor allowed to obtain microspheres labelled with the ^{90}Y isotope on the way of the nuclear reaction $^{89}Y(n, \gamma)^{90}Y$. Specific activity of irradiated microspheres has been determined by application of absolute triple to double coincidence ratio method (TDCR) and has been evaluated at 190 MBq/mg Y. [1]

Presented method consists in analysing the patient's PET-CT images after administering a radiopharmaceutical in the form of Y-90 microspheres. The subject of the presentation is the analysis of the distribution of activity and dose absorbed in a dedicated water phantom with vials containing $^{90}YCl_3$ solution with volumes and activities similar to the therapeutic ones. For the purposes of the work, a water phantom was designed and constructed, in which small-volume vials were placed to test the scanner's response to Y-90 activities. Dedicated software was developed for the analysis of image data. The development of the tool is consulted with the clinical community so that it can ultimately be used not only for scientific purposes, but also clinically.

Conclusion

^{90}Y microspheres prepared by the proposed technique can be regarded as a promising medical material for radioembolization of liver malignancies. The dosimetry method allows for verification of therapeutic dose administered onto liver.

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Research was supported by project OPUS-13 no 2017/25/B/ST7/01745 funded by National Science Centre of Poland.

VALIDATION OF A FULLY AUTOMATED PROCESS FOR ZIRCONIUM-89 PRODUCTION IN A VARIABLE ENERGY CYCLOTRON

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Introduction

Over the last several years, PET imaging with ^{89}Zr-based agents has been a dynamic research area; much further efforts are needed to accelerate the translation of promising ^{89}Zr-based tracers into clinical use. The new generation of immunoPET radiopharmaceuticals are expected to be used in early detection, screening, and follow up of human malignant diseases such as breast, prostate, ovarian and intestinal cancers.

Description of the Work or Project

The aim was to validate the production process of ^{89}Zr by irradiating Y-nat solid targets in a variable energy proton beam (14-19 MeV) cyclotron, using a commercially available automated solid target system. The irradiation and post-processing system, comprising modules for target preparation, pneumatic transfer, irradiation, dissolving and purification was employed for production of $^{89}ZrCl_4$, through the $^{89}Y(p,n)^{89}Zr$ nuclear reaction. Separation of ^{89}Zr and purification of $^{89}ZrCl_4$ using high molarity HCl and cation exchanger, were optimized, resulting in high purity and high molar activity/specific activity $^{89}ZrCl_4$ as radiochemical, to be further used for radiolabelling of antibodies. Quality control of the final radiochemical was carried out; irradiation yield, radiolabelling yield, pH, radiochemical purity and radionuclide purity were assessed.
The target used was a foil of Y-nat (99,9%, Alpha Aesar), 10 mm diameter and 250 μm depth. Nuclear reaction starts at 3.7 MeV, optimal proton beam energy at 13 MeV (according to Talys code symulations). The optimal irradiation parameters were set as follows: proton energy 14 MeV, degraded to 10.5 MeV with a pure Al foil, 40 μm; proton current intensity 8μA; irradiation time 4h (32 μAh). After irradiation, the target was dissolved in 5 mL HCl 6M and purified on a C18 column resulting 2 GBq [^{89}Zr]$ZrCl_4$. The radiochemical purity was assessed by radio-HPLC showing values higher than 99%, after purification.

Conclusions

The characteristics of the [^{89}Zr]$ZrCl_4$ meet the requirements for the biomolecules radiolabelling (antibodies) and comply to the translation to radiopharmaceutical production line, aiming to develop new generation of immunoPET agents for preclinal testing and clinical trials.

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IAEA RADIOISOTOPES AND RADIOPHARMACEUTICALS REPORTS No. 4, ALTERNATIVE RADIONUCLIDE PRODUCTION WITH A CYCLOTRON, INTERNATIONAL ATOMIC ENERGY AGENCY VIENNA, 2021

OPTIMISING THE PRODUCTION OF ^{67}CU/^{64}CU THERANOSTIC PAIR

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Introduction

Developing new theranostic approaches requires improved nuclear data to understand the best conditions to produce such isotopes. ^{67}Cu/^{64}Cu is one of the promising theranostic pairs that its use has been limited by lack of a sustainable supply of its therapeutic counterpart ^{67}Cu. Despite its increasing clinical relevance, the production route for ^{67}Cu and ^{64}Cu have not yet been evaluated [1]. Although there are several routes to produce the ^{67}Cu/^{64}Cu pair, a small cyclotron with low-energy protons (or deuterons) seems promising. Using a research beam line is of a great potential for the accurate cross section measurements, allowing the optimisation of its production route. The motivation behind this project is a search for alternative production routes of ^{67}Cu/^{64}Cu with increased yield and radioisotopic purity to achieve a secure supply of this pair. The approach is to produce the ^{67}Cu/^{64}Cu pair via a stacked target which involves proton- (or deuteron-) and secondary neutron induced reactions simultaneously.

Description of the Project

Efficiency of the production depends on a good target design. A dedicated stacked target comprising ^{70}Zn, ^{64}Ni and ^{67}Zn layers allows producing ^{67}Cu and ^{64}Cu by primary reactions ^{70}Zn(p,α) and ^{64}Ni(p,n), respectively. The added ^{67}Zn layer contributes to the production of ^{67}Cu through the secondary reaction ^{67}Zn(n,p)^{67}Cu. The measurements are carried out at The Birmingham MC40 Cyclotron, and the stack is optimised by Monte Carlo modeling.

Conclusions

This project identifies the optimal experimental conditions to maximize the ^{67}Cu and ^{64}Cu production yield in a small-sized cyclotron. Producing the ^{67}Cu/^{64}Cu theranostic pair being the overall objective of it, the project obtains the nuclear data relevant to the production of ^{64}Cu and ^{67}Cu radionuclides via proton and neutron induced reactions. The obtained experimental cross sections are compared with nuclear model calculations to develop evaluated excitation functions for each reaction.

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Newborn screening for the early diagnosis of Congenital Hypothyroidism in Bangladesh by Immunoradiometric Assay (IRMA)

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Abstract

Background: Newborn screening programs signify early and presymptomatic detection of treatable disorders which authorize commencement of prompt medication to alleviate notable fatality. Newborn screening (NBS) for congenital hypothyroidism (CH) has been a successful public health initiative in preventive medicine over the last few decades. Previous pilot (1999-2006) and phase 1 (2006-2011) NBS program in Bangladesh reported an increased incidence rate compared to global data which urged to initiate a second phase NBS program for the detection of CH.

Materials & Methods: Neonates' blood has been collected from the umbilical cord or heel and drawn on the Guthrie filter paper card. The thyroid-stimulating hormone (TSH) has been measured from this filter paper by Immunoradiometric assay (IRMA) or Dissociation Enhanced Lanthanide Fluorescence Immunoassay (DELFIA) method.

Results: 123 out of 2,60,150 neonates have been already diagnosed with congenital hypothyroidism from September 2018 to August, which represents an incidence of 1:2126.

Conclusion: Ongoing phase-2 project data is already showing some optimistic outcomes. Hence, a time-worthy sustainable policy should be implemented to mark this program as an effective one.

Keywords: Newborn Screening, Congenital hypothyroidism, TSH, IRMA, DELFIA

Cyclotron based PET radiopharmaceutical production: Present status and future prospectus in Bangladesh

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Bangladesh is the 8th most populated country in the world with a population of 166.3 million. Non-communicable diseases, such as cancer, have recently become a serious public health concern and it is the 6th leading risk factor for death in Bangladesh. Positron Emission Tomography - Computed Tomography (PET-CT) scan are extremely useful in determining the type of cancer and its stage. In Bangladesh there are seven (07) PET-CT facilities. The most successful and extensively used imaging radiotracer in PET-CT is 2-[^{18}-F]-fluro-2-deoxy-D-glucose (FDG). In the past, there was only one cyclotron (0.6 MeV, GE) in the private sector. An 18/9 MeV cyclotron (18MeV for proton and 9 MeV for deuteron, Model: Cyclone 18/9 IBA) was installed at the National Institute of Nuclear Medicine and Allied Sciences (NINMAS), Shahbag, Dhaka. The installation of this cyclotron will help to shorten the long lines of cancer patients and will be able to meet the demand for other radiotracer in PET-CT facilities. This cyclotron facility is being used to produce ^{18}F-FDG since October 2020 and 100 batches have been completed successfully. Appearance, Identity (Radionuclidic purity, Radiochemical purity, Half-Life), pH, Chemical purity, Residual solvents: acetonitrile and ethanol, Bacterial endotoxin, sterility test are the quality parameters that are used to maintain quality assurance. Synthesis of [^{18}F]NaF, [^{18}F]FLT, [^{13}N]NH_3, [^{11}C]Acetate, [^{11}C]Methionine and [^{11}C]choline are in the active consideration. With the development of solid target radiochemistry laboratory, we also have future plan to produce ^{124}I, ^{64}Cu, ^{68}Ga and ^{89}Zr radioisotopes.

PROTON BORON FUSION THERAPY FROM MONTE CARLO STANDPOINT

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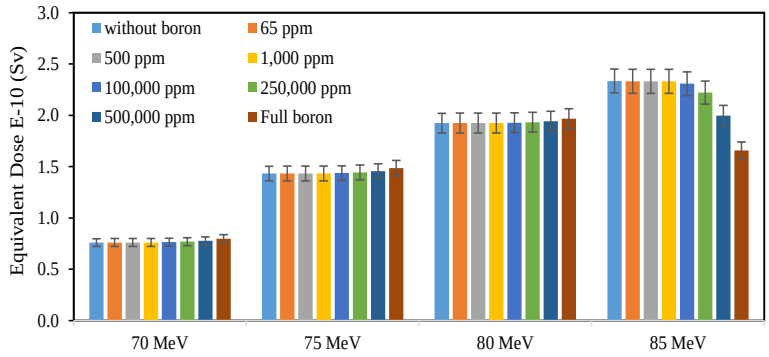
Introduction: Recently it has been suggested that the presence of boron-11 during proton therapy leads to a significant dose increasement in the boron uptake region [1, 2]. Three high-LET alpha particles with an average energy of 4 MeV are generated at the point of interaction between proton and boron-11. Nevertheless, the cross-section of $p + {}^{11}B \rightarrow 3\alpha$ interaction is negligible and dose increasement is unlikely. The purpose of this study is dose evaluation of the proton therapy with and without the boron-11. **Description of the work:** All simulations were performed using MCNPX 2.6.0 code at the Snyder head phantom. At the elderly stage, the range of Bragg-peaks was adapted to the tumor volume, with and without boron-11. Then, the different concentrations of boron-11 were assumed including 65, 500, 10^3, 10^5, 2.5×10^5, and 5×10^5 ppm in the tumor region. To investigate the maximum effectiveness of PBFT (proton boron fusion therapy), the entire tumor was assumed full of boron-11, and the dose components were calculated. **Results:** The dose amplification is negligible at all concentrations of boron-11 of 65, 500, 10^3, 10^5, 2.5×10^5, and 5×10^5 ppm as well as when the entire tumor was assumed full of boron-11. In the best case, the maximum dose amplification was less than 5%, in which the entire tumor was assumed full boron-11. **Conclusions:** The total number of alpha particles generated from $p + {}^{11}B \rightarrow 3\alpha$ interaction is negligible. As well as the presence of boron-11 during the proton therapy makes that the Bragg-peaks happen in greater depth. Hence, from the Monte Carlo standpoint, the effectiveness of the proton boron fusion therapy is not related to the alpha particles because the dose component of alpha particles is negligible.

Figure 1: The comparison of equivalent dose at 70, 75, 80, and 85 MeV at different concentrations of boron-11 in the tumor.



Keywords: Proton therapy, Proton Boron Fusion Therapy [PBFT], Boron, Brain, cancer, MCNPX, Bragg-peak

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DEVELOPMENT OF ^{64}CU-DOX/DOX-LOADED CHITOSAN-BSA MULTILAYERED HOLLOW MICROCAPSULES FOR SELECTIVE LUNG DRUG DELIVERY

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Introduction

The use of engineered particles as carriers for chemotherapy drugs has created a bright horizon for improving the performance of chemotherapy of cancers. When chemotherapy drugs such as doxorubicin (DOX), paclitaxel (PTX), cisplatin, etc. are intravenously injected in unloaded and conventional formulation, distribute freely throughout the body, resulting in undesirable side effects on healthy tissues and limiting achievement of adequate doses in the tumor site required to be effective on the cancerous cells. To overcome this problem, particles-based drug delivery platforms have emerged and so far various platforms like liposomes, dendrimers, polymeric nanoparticles (NPs), etc. have been studied.

Description of the Work or Project

To selective delivery of high dosage of doxorubicin (DOX) to the lung via I.V injection, it was loaded into chitosan-BSA multilayered hollow microcapsules fabricated based on the $CaCO_3$ template. The ^{64}Cu-DOX radiotracer was also loaded along with cold DOX for the evaluation of the biodistribution. Hollow microcapsules were non-toxic, compatible with red blood cells, and did not change the blood coagulation time.

Conclusions

The drug content of drug-loaded hollow microcapsules was %55–60 and their toxicity depends on the drug release rate. PET-CT images showed that, unlike unloaded DOX, the majority of the loaded drug (>%75) was delivered to the lung by the hollow microcapsules and then sustainably released from microcapsules into the lungs. The ratios of loaded drug accumulated in the lungs to the liver were 5.2, 2.52, 0.28, and 0.14, at 1, 4, 14, and 24 h post-injection, respectively. These ratios for injection of the unloaded drug were 0.04, 0.01,0.01, and 0.006 at the same time intervals. The results showed that the fabricated hollow microcapsules are capable to carry a high dose of the drug to the lungs.

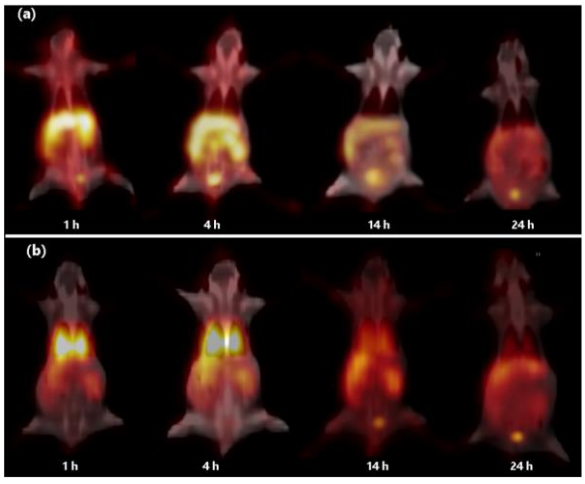

Figure 1: PET-CT images of the healthy mice after i.v. injection of (a) unloaded ^{64}Cu-DOX and (b) loaded ^{64}Cu-DOX.

Keywords: Hollow microcapsules Lung targeted drug delivery PET-CT

PRELIMINARY ASSESSMENT OF Tb-161 PRODUCTION IN MARIA RESEARCH REACTOR.

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Introduction

Terbium-161 (Tb-161) is of particular interest for targeted radionuclide therapy because of its similar properties to those of lutetium-177 (Lu-177), in addition it emits conversion and Auger electrons, thus Tb-161 is considered to be more effective than Lu-177. Importantly, Tb-161 can be produced in nuclear reactors in the $^{160}Gd(n,\gamma)^{161}Tb$ nuclear reaction.

Description of the Work or Project

In order to assess the feasibility of Tb-161 production by neutron irradiation in MARIA research reactor in Poland the test irradiation of 0.87 mg of $[^{160}Gd]Gd_2O_3$ (enriched to 97.8%) was performed. The target material was irradiated for 184 hours in thermal neutron flux of 10^{14} n $\times cm^{-2} \times s^{-1}$. After two weeks cooling time, the target material was dissolved in 1 mL of 1 M HCl, evaporated to dryness and again dissolved in 1 mL of 0.1 M HCl. The aliquots of 0.05 mL were taken for measurement of radioactivity and radionuclide purity. Measurements were performed at 17, 28 and 44 days after the end of irradiation (EOB) by gamma spectrometry using HPGe detector, as presented in Table 1.

Table 1: Radionuclides and their measured radioactivities found in Tb-161 solution at EOB.

Measurement time	^{161}Tb [MBq]	^{160}Tb [kBq]	^{152}Eu [kBq]	^{154}Eu [kBq]
17 days	520 ± 10.7%	1.56 ± 37.9%	8.64 ± 64.0%	8.16 ± 38.9%
28 days	530 ± 10.7%	0.88 ± 99.4%	8.08 ± 40.7%	7.10 ± 39.7%
44 days	590 ± 10.7%	1.60 ± 12.3%	9.03 ± 11.5%	6.94 ± 11.3%

The production yield of Tb-161 from highly enriched Gd-160 in the MARIA reactor was estimated at the level of 700 MBq/mg Gd. This value is similar to the results published by other authors [1]. The content of Tb-160 ($T_{1/2}$=72.3d) was $<1\times10^{-4}$%. Surprisingly, the two long lived europium radionuclides, ^{152}Eu ($T_{1/2}$=13.5y) and ^{154}Eu ($T_{1/2}$=8.6y), were found at the relatively high level up to 1×10^{-3}%. Their presence creates a concern for the reactor production route of Tb-161. Most probably they originated from the target material (according to the manufacturer's specification, the content of stable Eu in the target material is 1.1×10^{-3}%). Potentially, the radioisotopes of Eu can be removed during ^{161}Tb processing together with ^{160}Gd. However, in presence of long-lived Eu radionuclides the options for recycling of the Gd fraction for recovery of this costly target material are limited. Methods for purification of the target material (highly enriched ^{160}Gd) from Eu or for separation of the radionuclides of each of the three elements Eu, Gd and Tb need to be elaborated.

Conclusions

The feasibility of ^{161}Tb production in a medium neutron flux reactor such as Maria research reactor in Poland was demonstrated. The processing issues will be further addressed.

Acknowledgement

PRISMAP project has received funding from the European Union's Horizon 2020 research and innovation programme.

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ABSORBED ENERGY MEASUREMENT USING TUMOR-MODEL SCINTILLATION DETECTORS IN SKULL PHANTOMS

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Introduction

A Gamma Knife (GK, Eletka AB, Stockholm, Sweden) uses ^{60}Co sources to treat various intracranial lesions. Absorbed energy to a tumor in GK radiosurgery is an integration of dose to each point in the tumor and is related to DNA aberrations. Despite its importance, the treatment planning program's accuracy in calculating the absorbed energy has rarely been verified. In this study, absorbed energy to tumors calculated for GK radiosurgery was verified using 3D-printed scintillation detectors in skull phantoms.

Description of the Work or Project

The MR images of two vestibular schwannomas were exported to a 3D-slicing program in DICOM-RT format. The volume of the tumors was 0.722 cm^3 and 0.216 cm^3. Two hollow skull phantoms filled with water were built by 3D printing according to each patient's skull images. Tumor model scintillators (TMS) formed in the shape of the tumors were 3D-printed using self-developed scintillating plastic resin. The TMS outputs were measured in nine fields at the center of an Elekta solid water phantom (Elekta AB, Stockholm, Sweden) and calibrated to Monte Carlo simulation values. The mean adjusted R-square value of linear fitting of the calibration data was 0.9996 +/- 0.0020. TMS detectors were inserted into the tumor location inside the skull phantom, and CBCT images were obtained. Treatment plans were made to irradiate the TMS using a GK. 12.5Gy and 13Gy were prescribed to 50% isodose for each TMS. Irradiation was carried out in the same manner as the patient was treated. The energy absorbed by TMS was measured and compared with calculated values. The calculated energy was 14.24 +/- 2.44 mJ and 4.62 +/- 0.70 mJ, respectively. The corresponding measured energy was 14.29 +/- 0.11 mJ and 4.65 +/- 0.03 mJ.

Conclusions

The absorbed energy to a TMS calculated for GK radiosurgery agreed with measured values with differences less than 0.7%. We expect experiments with skull phantoms mimicking inhomogeneous intracranial materials to be necessary for more accurate verifications.

IMPROVING ACCURACY IN THE RADIOACTIVE ANALYSIS OF ASH PLANT SAMPLES

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Introduction

In this work, to study the effect of the Compton continuum of ^{40}K on the full energy peaks of ^{238}U and ^{232}Th and to improve the minimum detection activity of the analytical results of those in ash samples of plants, the isotope ^{40}K is separated from the samples by chemical method. The samples with and without ^{40}K are surveyed by the gamma spectrometer HPGe. The results of ^{238}U, ^{232}Th, and ^{40}K are evaluated and compared with each other.

Description of the Work or Project

The biota samples were studied on the campus of the VNUHCM – University of Science including Vera, Pine Spruce leaf, Vertiver grass, Red Amaranth, Water Spinach denoted by VERA, PINE, VERT, AMAR, SPIN, respectively. The influence of the ^{40}K on the energy peaks of other isotopes in ash plant samples was evaluated by analyzing the results of the spectrum of the ashes before and after the separation of ^{40}K. It is quantitatively estimated the influence of the Compton region of ^{40}K in the energy range of less than 100 keV. The peak-to-total ratio at these energy peaks (46.5 keV, 63.3 keV, and 92.6 keV of ^{210}Pb and ^{234}Th, respectively) on the spectrum before and after K-separation was calculated. The peak–to–the total ratio of the samples with removing K are greater than that of the samples without removing K.

Conclusions

It is demonstrated that the method of separating ^{40}K from the plant ash sample is obviously applied to increase the accuracy of the peak energy analysis results of the other radioisotopes in the sample, namely in the 100 keV region. The results figured out that the influence of the ^{40}K Compton background is proportional to the activity of ^{40}K in the sample. For plant samples with a ^{40}K activity dominant to other isotopes, and using low energy peaks, the separation of ^{40}K out of the sample will allow for rapid and accurate analysis.

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PURIFICATION OF At-211 USING WET EXTRACTION AND CHEMICAL APPLICATIONS

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Introduction

Alpha emitters have been studied extensively in targeted radiotherapy for last 10 years because of its strong cytotoxicity, which can induce non-repairable DNA double-strand breaks. ^{211}At production sites are still limited due to short half-life (7.2 h) and availability of alpha-beam irradiation at 28 MeV energy[1]. Korea Institute of Radiological and Medical Sciences (KIRAMS) is producing ^{211}At since 2021. Herein, production, quality control, and chemical application experiences of ^{211}At in KIRAMS will be described.

Methods

^{209}Bi metal was coated onto Al target plate and irradiated at 29 MeV of α-beam energy with 43-107 μA·h. Irradiated ^{209}Bi was dissolved in HNO_3 and evaporated. White residue redissolved in HCl was extracted by diisopropyl ether. ^{211}At in organic layer was taken to NaOH solution and neutralized by adding acid for radiolabeling. Gamma energy and metal contents were measured using high purity germanium radiation detector and inductively coupled plasma mass spectrometry. Radiolabeling of ^{211}At was performed using three precursors: *N*-succinimidyl 3-(trimethylstannyl)benzoate, *N*,*N*′-bis(tert-butyloxycarbonyl)-3-(trimethylstannyl)benzylguanidine, and 3-(trimethylstannyl)benzoyl-trastuzumab conjugates. Radiochemical yield and purity were confirmed by HPLC or radioTLC.

Results

260.5-910.2 MBq of ^{211}At in solution (pH 5-6) was produced using wet extraction in recent irradiation (n=6). 76.9, 79.3, 89.8, and 687.0 keV of characteristic gamma energy was observed and 2.5±1.4 ppm of Sn and 0.4±0.6 ppm of Al was measured from ^{211}At solution, respectively. Radiochemical yield of of *N*-succinimidyl 3-[^{211}At]astatobenzoate ([^{211}At]SAB) and *m*-[^{211}At]astatobenzylguanidine ([^{211}At]MABG) was 43.1±2.6% and 65.7±10.1% based on HPLC analysis. 3-[^{211}At]astatobenzoyl-antibody was radiolabeled in 68.6±12.6% yield after PD-10 purification. Radiochemical purity of compounds was higher than 95%.

Conclusions

^{211}At was produced successfully using wet extraction and chemical validation indicated that specific gamma energy was observed and ^{211}At solution contains significant low metal contents. Radioastatination to organotin precursors was carried out in high radiochemical yield. Further biological evaluation of ^{211}At-labeled compounds is planned.

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Keywords: Astatine-211, cyclotron, wet extraction, alpha-emitter, radioastatination

Development of Pm-147 betavoltaic batteries

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Introduction

Betavoltaic nuclear batteries are energy-conversion devices using low-energy beta particles emitting radioisotope. They have attracted much attention due to continuous power generation without affecting external environment, long-term lifetime and small-size fabrication. Various radioactive materials, such as H-3, Ni-63 and C-14, can be used in betavoltaic batteries, which decide their lifetime and power. KAERI have developed betavoltaic batteries and we recently succeed to develop 3uW Ni-63 betavoltaic battery and verify their long-term performance. In order to improve power, KAERI have developed betavoltaic batteries using Pm-147 radioisotopes, which have higher beta energy (average energy 62keV) than Ni-63 (average energy 18keV).

Development of Pm-147 betavoltaic batteries

For Pm-147 betavoltaic batteries produce high power, Pm-147 radioisotope with high purity and high specific activity should be required. We plan to produce Pm-147 through neutron irradiation of Nd-146 using HANARO (High-flux Advanced Neutron Application ReactOr) in KAERI. Also, we have developed high-purity separation process based on HPLC (High-pressure liquid chromatography). After irradiation and purification process, we can get Pm-147 as oxide form – Pm_2O_3. Based on this oxide form, we estimated the Pm-147 source thickness. Although the thicker source materials emits the more amount of beta particles, they interact with the source itself, called self-shielding effect, so there is optimum thickness for maximum emission of beta particles. In case of Ni-63 beta sources, they have 2-3um optimum thickness. We conducted MCNP (Monte Carlo N-Particle) analysis to find the Pm-147 source thickness that could emit the maximum amount of beta particles. As a result, Pm-147 showed lower self-shielding effect than Ni-63. Furthermore, we analyzed absorbed dose depends on the depth of 4H-SiC-based semiconductor. MCNP modeling was conducted with Pm_2O_3 beta source in planar surface configuration. We found optimal semiconductor depth that absorbed beta particles ~99.98%.

Conclusions

Betavoltaic batteries have much potentials on low-power electrical applications with long-term lifetime such as medical, space, military and IoT fields. KAERI have developed betavoltaic batteries for various applications and have research on core technology from production to fabrication. In order to generate target power for Pm-147 betavoltaic batteries, we have optimized the production/separation process of Pm-147 beta sources and the structure.

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An overview of the Radioisotope Electromagnetic Isotope Separation Capabilities at Idaho National Laboratory

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Introduction

Electromagnetic isotope separation (EMIS) has been an effective tool for the enrichment of isotopes for over 80 years. In the early 1990's the United States reduced or eliminated electromagnetic isotope separation capabilities, but in the last decade, increased demand for both stable and radioactive enriched isotopes have led to the reestablishment of small-scale EMIS capabilities in the United States National Laboratory Complex.

Description of the Work or Project

In the timeframe of approximately 2008 to 2013, Idaho National Laboratory (INL) refurbished and constructed two 90-degree sector isotope separators. The first one completed was originally designated for stable isotope production but is now used primarily as a non-radioactive component and method testing platform. The second separator is currently designated for radioisotope enrichment and is undergoing significant component upgrades under the stewardship of the U.S. Department of Energy Isotope Program (DOE-IP). The primary purpose of this effort is to produce high purity, highly enriched radioisotope spike solutions that can be used as reference materials for isotope dilution mass spectrometry (IDMS) analyses.

Conclusions

This work describes the status, capabilities, and future endeavors of the radioisotope EMIS at INL as part of the DOE Isotope Program.

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DEVELOPMENT NI-63 BETAVOLTAIC BATTERY FOR LOW-POWER SENSOR SYSTEM IN KOREA

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Introduction

A Betavoltaic battery that converts the decay energy of a radioisotope into electricity offers advantages for application requiring a long lifetime without recharging and minimum maintenance. In addition, it is used in many low-power applications because it can operate effectively under extreme environmental conditions such as in space and at deep sea.
In Korea, Korea Atomic Energy Research Institute (KAERI) has been developing a betavoltaic battery using radioisotope Ni-63 and silicon carbide (SiC) semiconductors. In this study, betavoltaic battery for low-power sensor system will be introduced.

Development of Ni-63 Betavoltaic battery

For a betavoltaic battery, beta rays emitted from a pure beta source such as Ni-63, Pm-147, and H-3 can cause the generation of an electron-hole pair (EHP) onto a semiconductor, and produce electricity. Ni-63, a pure beta-emitting source, is proper as the power source of a betavoltaic battery because of its energy spectrum (Eevg=17.4 KeV) and long half-life of 100.1 years.

Figure 1. Prototype of Ni-63 betavoltaic battery and application of sensor system to measure spatial radiation.

Ni-63 was fabricated in the form of a thin film using the electroplating method, and unit cells were fabricated by bonding Ni-63 plate and the SiC semiconductor. The battery pack was manufactured using a total of 64 unit cells. The output of the battery pack is 3.09 ㎼, the open circuit voltage is 4.14 V, and the short circuit current is 0.75 ㎂.
The battery pack is installed in KAERI's radioisotope production facility, and is used as an independent power source for radiation sensor that measure spatial radiation. It stores power in an all-solid-state secondary battery and transmits the radiation level in facility to the outside using wireless communication twice a day.

Conclusions

Betavoltaic battery has the advantage of being able to supply power stably for a long time without being affected by the external environment, so they can be used in various fields such as medical, national defense, aerospace and special industries. KAERI and ETRI (Korea Electronics and Telecommunications Research Institute) succeeded in demonstrating power generation using radioisotopes for the first time in Korea. In future research, it is required to develop a technology to improve the output and efficiency of betavoltaic battery.

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CONCEPTUAL DESIGN OF A LOW-ENERGY ION STORAGE RING AND A RECOIL SEPARATOR FOR STYDUING NEUTRON CAPTURE CROSS SECTIONS

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Introduction

The most elements heavier than iron are synthesized by neutron capture reactions occurring in the rapid (r) and the slow (s) processes. The s-process has been measured using the ToF or activation techniques. However, most of the capture products of r-process are too short-lived to be directly measured. An ion storage ring combined with a neutron generator and a new method to extract the products from the ring using a Wien filter, electromagnetic septa and a recoil separator has been proposed at TRIUMF based on the idea of ref [1]. The conceptual design of a storage ring and a recoil separator to measure neutron capture cross sections are presented.

Description of the Work

In order to determine the structure of the storage ring, ion optics calculations were carried out using COSY INFINITY code. $^{80}Zn^{15+}$ ions at 0.15 MeV/c are used as the primary beam for the calculations. The ring was designed as rectangular shape with a circumference of 46.5 m composed of four 90˚ dipoles, twenty quadrupoles, eight hexapoles and a Wien filter. The four straight sections would have an electron cooler, a neutron generator to make neutron reaction area, a Wien filter to separate the products from the beam by using the difference of their velocities and other devices, respectively. The results were verified using MADX code and it was found that the ring can store the beam over 10^6 turns inducing the reaction through the reaction area. G4beamline program was used to evaluate the trajectories of products and determine the specifications of septa. A pair of septa is located at the end of the third straight section to transfer the products to the recoil separator. The phase spaces at the end of septa were evaluated and it was applied as the beam input for the recoil separator. Ion optics calculations were performed to design the separator which separates the products by its charge states and masses. The separator has five quadrupoles, two hexapoles, an electric dipole and a magnetic dipole to disperse the beam by charge states and masses. It was evaluated that the products with selected charge state are dispersed by mass through the magnetic dipole and focused on a focal plane with mass-to-charge dispersion of 11.4 mm/% with transmission efficiency of ~99%.

Conclusions

An ion storage ring combined with a neutron generator and the extraction system to induce neutron capture reactions and to measure its cross sections directly were designed by using ion optics calculations and MC simulations. A recoil separator was also designed to separate the injected product beam by its charge states and masses with similar methods. The extraction system could transfer ions lighter than Z=82 in the energy range of 0.15 to 1.8 MeV/c to the separator. It can be improved if two or more Wien filters are used in the ring. It is planned to modify the ring structure to cover wider range of mass and energy of ions. It is expected that this facility can support to explore the origin of heavy elements and its abundance in the universe.

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FREE RELEASE OF HELIUM-3 FROM DARLINGTON NUCLEAR GENERATING STATION

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Introduction

Helium-3 isotope is a decay product of tritium, a byproduct from the ongoing operation of Canadian Deuterium Uranium (CANDU) Nuclear Reactors. Since the operation of Darlington Tritium Removal Facility in 1989 by Ontario Power Generation (OPG), the increasing inventory of tritium as a metal hydride has yielded a significant accumulation of helium-3 released inside the void space of Immobilized Tritium Containers (ITC's). This increased pressure from helium-3 has resulted in an operational burden to safely handle and transport ITC's, particularly as the vault allocated for ITC storage is nearing its inventory capacity.

In recognition of both the operational challenge within OPG and the growing global demand for a reliable supply of helium-3 isotope, Laurentis Energy Partners initiated a project to commercialize helium-3 by degassing ITC's through a series of zirconium-based alloy and removing trace quantity of tritium. The key features of the helium-3 extraction system and the results of the recent extraction campaign is discussed in this paper.

Description of the Work or Project

Simple design, minimal product loss and safety assurance to release helium-3 from the Darlington Nuclear Generation Station have been the key drivers for this project. Therefore, a decision was made to proceed with recovering helium-3 from the void space of ITC at room temperature, given the low equilibrium pressure of tritium-titanium. The compromise, however, was the overall efficiency in recovering helium-3 as significant volume is retained in the metal hydride matrix.

Once the ITC is degassed, the recovered helium-3 is recirculated in batches though zirconium-based getter beds, particularly chosen for being chemically inert in nitrogen, high reactivity with tritium and modest reactivation temperature [1]. A pair of 1 L in-line ionization chambers with maximum sensitivity of 1 $\mu Ci/m^3$ are utilized to monitor the tritium concentration.

Recent results of the helium-3 extraction campaign detected tritium as high as 35 $\mu Ci/m^3$ upon initial release of helium from ITC's but the series of zirconium getter beds were effective in reducing the tritium concentration down to <1 $\mu Ci/m^3$, far below the tritium Exemption Quantity.

Conclusions

Since the commissioning of the helium-3 extraction system in late 2021, Laurentis Energy Partners have become a reliable, strategic supplier of civilian produced helium-3 to the global market. The results of the recent extraction campaign demonstrate Laurentis Energy Partners' adherence to the principle of ALARA, reducing the tritium content in helium-3 below 0.01% of the Exemption Quantity.

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Title: Role of quantitative SPECT/ CT for thyroid uptake in patients with Graves' disease undergoing low dose I-131 therapy

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Introduction: Thyroid scintigraphy and uptake studies are usually performed prior to low-dose I-131 therapy in patients with Graves' disease. However, SPECT/CT plays an important role in quantification of uptake and absolute quantification has become possible due to the availability of commercially available software. SPECT/CT studies can be calibrated to convert the counts obtained into activity and further to SUV which may be preferable method for for volume quantification in patients for pre-therapy assessment and response evaluation in post-treated period.

Aim: The aim of study is to quantify and correlate the thyroid uptake and volume with the help of thyroid planar and SPECT/CT scans in patients with Graves' disease undergoing low-dose I-131 therapy.

Materials and Methods: In this prospective study, a total of 35 patients (25 women, mean age: 35±5; range 26-63 years) with the diagnosis of Graves' disease were included. Pre-therapy planar and quantitative SPECT/CT scans were acquired in all the patients at around 20 minutes of intravenous injection of technetium pertechnetate (3-5 mCi) as per department protocol. The acquired

data was reconstructed using the proprietary techniques and analysed using QMetrix software for thyroid uptake, SUV and volume.

Results: The correlation between planar uptake and SPECT volume for thyroid gland was statistically non-significant ($r(35)= 0.1044$, $p=0.550$).The correlation between SPECT uptake and SPECT volume was statistically significant at ($r(35)= .580$, $p<.005$). The SUV max value of 285.3 and SUV mean value of 98.1were positively correlated with elevated free T3 and free T4 levels based on ROC analysis with satisfactory sensitivity and specificity.

Conclusion: SPECT/CT can give absolute quantification of thyroid uptake with good reproducibility as compared to planar imaging which may be subjective. SPECT/CT can also reveal any incidental thyroid nodule with its functional status that might have been masked in planar imaging. SUV derived from thyroid SPECT/CT using QMetrix are useful for the pre-therapy evaluation in Graves' disease.

Light-Ion Irradiations of Platinum for Gold Radioisotope Production

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Introduction

Gold radioisotope production cross sections were investigated in support of nuclear data. The irradiations were performed at Lawrence Livermore National Laboratory's (LLNL) Center for Accelerator Mass Spectrometry (CAMS) facility. Light-ion irradiations of protons and deuterons were bombarded onto natural platinum targets.

Description of the Work or Project

The series of irradiations were done using the 10 MV FN tandem accelerator and the NEC Toroidal Volume Ion Source (TORVIS). The stacked target method was used to determine a suite of near-barrier cross sections. The experimental cross sections for $^{190\text{-}199}$Au radionuclides have been determined. All cross sections are compared to TENDL-2019[1]. Deuteron irradiations were performed for the sole motivation to extend the list of production radionuclides to include ^{199}Au.

Table 1: Novel Light-Ion Cross Sections (mb)

	E= 12.0±0.4 MeV	E=11.1±0.5 MeV	E=10.2±0.5 MeV
^{nat}Pt(p,x)^{190}Au	0.0717±0.0184	0.0483±0.0161	0.0446±0.0108
^{nat}Pt(p,x)^{191}Au	0.480±0.020	0.0357±0.0082	<0.0016
^{nat}Pt(p,x)^{198m}Au	0.0294±0.0078	0.0263±0.0084	0.0154±0.0030

Conclusions

This includes the first such proton-based cross sections of ^{190}Au and ^{198m}Au and lowest energy investigation of ^{191}Au. Literature comparisons to the remaining isotopes are also reported[2,3]. Additionally, isotope-specific reaction cross sections are derived using the natural platinum abundance values.

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Production and Incorporation of Br-82 into Organic Radiotracers

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Introduction

Radiotracers are used in chemical plants and the oil and gas sector to identify the presence and location of leaks within a system so as to prevent potential incidents with plant-wide or environmental consequences. The radionuclide bromine-82 (Br-82) is widely used in this application due to its high energy gamma emissions (E_γ = 777 keV, 554 keV) and 35 h half-life, which provides sufficient time for testing without incurring long-term contamination of the system.[1] Br-82 is a cost-effective choice because it can be produced via neutron bombardment of natural bromine with no need for purification. However, direct production of organobromine tracers is not possible because organic substrates decompose in-core. The objective of this work is to develop a safe, rapid, and effective method of incorporating Br-82 into organic molecules for industrial tracing applications.

Description

Standard synthetic approaches to make organobromides often rely on elemental bromine or hydrobromic acid as bromine sources[2]; however, neither of these compounds is sufficiently chemically or thermally stable for activation in a nuclear reactor. Instead, KBr was selected as the irradiation target as it has good thermal and radiolytic stability compared to alternatives such as NH_4Br and generates less incidental activation product than NaBr.

Non-active KBr was reacted with a series of organic substrates (1-dodecene, 1-dodecanol, 1-methoxynaphthalene, 9-methoxyanthracene) at room temperature. A solution of Oxone® ($2\ KHSO_5 \bullet KHSO_4 \bullet K_2SO_4$) was added slowly to the biphasic reaction mixture.[3] After 1-2 h, the organic layer was decanted; any residual inorganic material was removed using a silica gel cartridge. A mixture of 1,2-dibromododecane and 1,12-dibromododecane (80% combined yield) was obtained from 1-dodecene regardless of changes in reaction conditions; no reaction was observed from the 1-dodecanol.

The reactions were repeated using $K^{82}Br$ ($\leq$ 40 MBq) generated at the McMaster Nuclear Reactor. The process was found to be reproducible, with $\geq$ 80 % of the ^{82}Br converted into organic complexes and isolated from the reaction mixtures. There was no indication of volatile ^{82}Br escaping from the reaction vessel, which is essential for scale-up to industrial levels, as Br_2 gas is corrosive and could damage expensive (hot cell) infrastructure.

Conclusion

A rapid and efficient process has been developed for radio-brominating organic substances starting with inorganic bromide. Work is in progress to scale up the process to industrially relevant quantities (40-100 GBq), including adapting the process steps so they can be executed with tele-manipulators in a hot cell.

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Separation of Pm for betavoltaic battery

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Introduction

Beginning with the development of beta cells in 1954, research on beta cells has been conducted until recently. A betavoltaic battery is a device that generates electricity by absorbing beta rays emitted by radioisotopes at semiconductors. Nuclides such as H-3, Ni-63 and Pm-147, which emit 100% of beta rays, are mainly used. Since nuclides with long half-lives emit low energy, beta batteries can be made by selecting suitable nuclides based on the required power and duration.

Production technology of Pm-147

To make beta batteries using Pm-147($t_{1/2}$ = 2.62 years), which has a relatively short half-life, promethium must be produced. Although Pm-147 can be separated from the spent nuclear fuel, the author decided to develop a production process of Pm-147 through neutron irradiation technology for stable supply in Korea. Simulations were performed with Cinder 90 prior to the production of Pm-147 with HANARO (neutron fluxes 5.8 x 10^{13} $n/cm^2 \cdot s$). As a result, when Nd-146 absorbs neutrons, it generates Pm, Nd, Sm, Eu, Tb, Ir, and Co. After 8 cycles of irradiation, it was confirmed that the specific activity of Pm-147 was about 0.6 ~ 0.7 mCi/mg. In order to purify Pm-147 from the mixture, separation process should be developed. The process of increasing purity uses high-performance liquid chromatography (HPLC). Since lanthanides can be separated based on changes in pH, an optimum conditions by varying the type of eluent and the pH.

Conclusions

Simulation was carried out using the cinder 90 program to produce promethium for application in beta batteries. As a result, in addition to Pm-147, nuclides such as Nd-147, Sm-151, Eu-154, Tb-160, Ir-192, and Co-60 were also produced. The specific activity of Pm-147 was 0.6~0.7 mCi/mg. For use in betavoltaic battery high purity promethium is required. To purify Pm-147, the separation process of lanthanides will be developed using HPLC under the several conditions in KAERI.

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Research on Pressurizer Demolition Scenario Construction During Nuclear Decommissioning

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Introduction

Demolition during nuclear decommissioning is an operation performed close to the structure, with a risk of excessive radiation exposure for the workers. In the case of the pressurizer, one of the larger structures for demolition, there is a significant lack of research on the radiation exposure management for the workers. Through worker radiation exposure assessment and the establishment of a working scenario, this research aims to consider the applicable demolition technique for large structures during decommissioning and present meaningful preliminary data for establishing the method of demolition based on the as low as reasonably achievable (ALARA) principle.

Assessment method

This research used VISIPLAN and Fusion 360 in parallel for scenario formulation and dose assessment. The radiological information for the Bohunice A1 pressurizer and the data for the AP-1000 pressurizer were applied.

The demolition scenario was constructed based on the standard radioactive solid waste disposal drum. The 200 L scenario was established according to the 200 L disposal drum, and the 320 L scenario according to the 320 L disposal drum. Both scenarios were established with the condition that the demolished piece is sufficiently filled.

In addition, the radioactive decay was considered according to the initial pollution rate to derive the location of demolition, and the demolition speed was included to assess the effective dose. First, in the case of the location of demolition, a total of seven periods were established, from the time of the permanent suspension of operations to 30 years thereafter, at five-year intervals. The considered demolition speed was separated into 19 stages from 10 $cm \cdot min^{-1}$ to 100 $cm \cdot min^{-1}$ at 5 $cm \cdot min^{-1}$ intervals.

Assessment results

The assessment results showed that the ranges of the collective and individual effective doses for the workers from the permanent suspension of operations to 30 years thereafter, with respect to the effective dose and the 200 L scenario, were 1.00E-01–5.75E+01 man*mSv and 0.1–4.42 mSv, respectively. In contrast, for the 320 L scenario, the ranges of the collective and individual effective doses were derived as 5.9E-02–3.34E-01man*mSv and 0.06–3.72mSv, respectively.

Conclusions

The speed and location of demolition considered in this research can be accounted for during future actual decommissioning activities or may serve as prior research to be referenced while selecting the demolition technology optimized for the structural application. It is expected that this study can be used as base data for the delay, immediate decommissioning, and sequencing of demolition activities for each structure. In addition, during actual demolition, a comparative analysis of this research could lead to the suggestion of a direction for more reliable research.

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MEASUREMENT OF ^{224}Ra, ^{226}Ra AND ^{228}Ra IN NATURAL WATERS THROUGH GAMMA-RAY SPECTROMETRY

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Introduction

Radium in drinking water may expose the public to significant doses of radiation. In this study, a gamma-ray spectroscopic technique was established to determine radium isotopes, which were preconcentrated from natural waters as barium sulfate. Additionally, the concurrent determination of radium isotopes (^{224}Ra, ^{226}Ra, and ^{228}Ra) in hot spring waters and associated sludge was performed in the Beitou hot spring area in Taiwan.

Description of the Work

A field survey utilizing gamma-ray spectroscopic technique was conducted in the Beitou hot spring area, where radium activity in hot spring waters and sludge was found to be higher than elsewhere in Taiwan. According to the results, the activity of ^{224}Ra in spring waters was highly correlated with ^{228}Ra due to their identical chemical behavior and original decay chain series. Additionally, concentrations of these radium isotopes (^{226}Ra, ^{228}Ra) and some chemical analogues in sludge were linearly related to one another, revealing their similar chemical behavior and that they may transport and distribute together in the environment.

Table: Radioactivity in soil, water, and sludge globally, in East Asia, and in Taiwan.

Region	Radioactivity in soils (Bq kg^{-1})							
	^{40}K		^{224}Ra		^{226}Ra (^{238}U)		^{228}Ra (^{232}Th)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Global	400	140 – 850	–	–	35	16 – 110	30	11 – 64
East Asia	393	63 – 1011	–	–	56	14 – 219	60	16 – 180
Taiwan (elsewhere)	370	11 – 844	–	–	17	<2 – 37	27	<2 – 70
Taiwan (this study)	–	–	7.0	1.0 – 9.2	–	<0.02 – 0.035	1.2	0.2 – 1.3
Taiwan (this study)	416	179 – 722	–	–	98	24 – 271	1080	219 – 3671

Conclusions

Gamma-ray spectroscopic technique was sensitive and alternative way to determine radium isotopes in natural and drinking waters. Radioactivity of radium isotopes in Beitou hot spring waters was relatively high levels compared with elsewhere in Taiwan and not safe for use as drinking water. Concentration of ^{226}Ra/^{228}Ra was correlated with selected chemical analogues in sludge from moderate (Ba and Sr) to strong (Pb) correlation.

Keywords: Beitou; radium; gamma-ray; hot spring; natural waters

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F-18-FDG PET/CT IMAGING ON THERAPEUTIC EFFICACY OF NOVEL DRUG CANDIDATE 'CM1' ON DSS-INDUCED COLITIS

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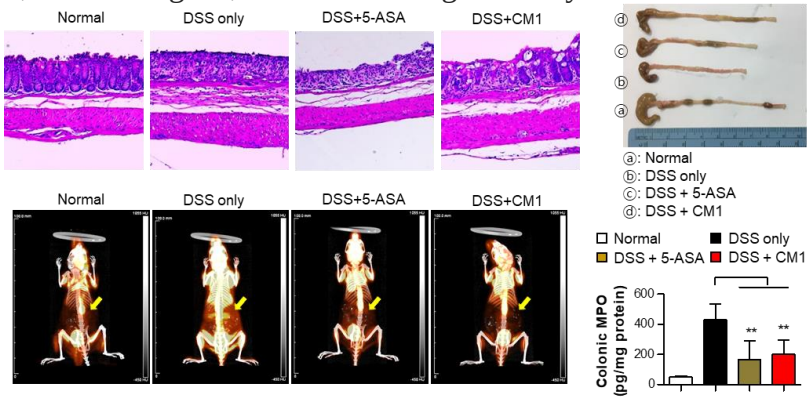
Introduction

Endoscopy and histological analysis are routinely used to evaluate the mucosal disease activity in individuals with inflammatory bowel disease (IBD) and in mouse IBD models. Due to the fact that mucosal healing is a primary therapeutic objective throughout IBD therapy, it is crucial to monitor how well the treatment is working. In previous study, we discovered a novel compound 'CM1', which produced from gamma ray-exposed chrysin, and CM1 has both improved water solubility and anti-inflammatory effects. Here, we examined, using histological, immunological, and microbiome analyses, the therapeutic efficacy of CM1 on dextran sodium sulfate (DSS)-induced colitis and compared it to the precision of F-18-FDG PET/CT imaging.

Description of the Work or Project

Our project has focused on the discovery of novel drug candidates from hit & lead compounds via radical reaction induced by ionizing radiation. We use SPECT/PET-CT imaging to monitor the therapeutic efficacy of novel drug candidates and also conduct radioisotope-ADME/PK *in vivo* studies.

In this study, to investigate whether F-18-FDG PET/CT could be a general monitoring methodology for IBD treatment, we compared colonic F-18-FDG uptake during the progression of disease. DSS-treated mice showed high FDG uptake in the medial colon and bone marrow, whereas both positive control (5-ASA) and CM1-treated mice showed significantly lower FDG uptake in the medial colon and bone marrow, which corresponded with histological, immunological, and microbiological analysis results.



Conclusions

The *in vivo* F-18-FDG PET/CT activity was correlated with histological damage of colon. F-18-FDG PET/CT is a non-invasive monitoring technique that can be used to track the progress of IBD therapy.

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Development of Qualification Standards and Test Procedures for Safeguards Equipment

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Introduction

When using safeguard equipments, in the Korean Peninsula, it is necessary to consider the situation of the divided country, unsmooth political cooperation, the environment, electric power, etc. It aims to develop qualification standards and test procedures by considering several factors, such as climate, risk factors, and power supply planning.

Description of the Work or Project

Safeguard equipment is used for gamma-ray spectrometry, neutron counting, spent fuel measurement, etc. The equipment consisted of InSpector 2000 MultiChannel Analyser, NaI, HPGe, xenon detector and so on. There are verification standards presented by International Atomic Energy Agency, European Union. "Qualification Test of IAEA Safeguards Equipment" explains Functionality Tests, Environmental Tests, Electromagnetic Compatibility Tests, Electrical Safety, and Irradiation Tests.

This study sought to identify the factors, climate, and terrain that may affect the equipment. But South and North Korea are divided nations, and accurate information about North Korea's nuclear activities is insufficient. We would like to propose a list of tests with suggested acceptance criteria and procedures that are compatible with the Korean Peninsula's conditions by synthesizing this. Tests include high temperature, low temperature, humidity, and shock test. The test procedure describes the sequence of steps along with the background conditions, and retention times. When taking the low-temperature test as an example, the qualification standard is -40°C for 16 hours. The procedure is described in the table.

Table 1: Low temperature test procedure

Test Procedure	1. Place the equipment in the chamber. 2. Start at 0°C and decrease by 10°C at intervals of at least 10 minutes. Check and record the normal operation of the equipment (resolution, channel peak, count rate change, etc.) at -10, -20, and -30°C. 3. Maintain the temperature at -40°C for 16 hours. 4. Final confirmation and recording of the normal operation of equipment.

It is also necessary to establish movement conditions and battery conditions. Factors that can affect equipment during transportation include shock and vibration. An appropriate counterplan should be considered for a power supply that is not smooth while the equipment is in use.

Conclusions

The qualification standards and test procedures for safety-related equipment were established. The materials constructed through research can be used as reference materials and minimize trial and error when verifying denuclearization in the future.

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Improvement of radiation utilization field classification criteria considering international classification standards and development of radiation protection record utilization model

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Introduction

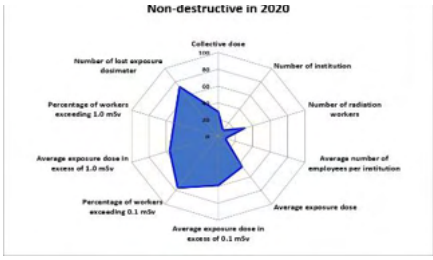
Along with the expansion of radiation use and development of technology, the number of radiation workers is continuously growing. Some international organizations are conducting continuous research on worker exposure, and demand for interpreting and evaluating exposure records is also increasing. In this study, we developed a model for the improvement of Korea's radiation use field classification criteria considering the international occupational exposure classification criteria and the utilization of exposure records.

Description of the Work or Project

To analyze the international occupational exposure classification criteria, the UNSCEAR, US, UK, France, and Canada classification systems were analyzed among countries for which data are open. Among them, UNSCEAR, which has 197 member countries and is conducting global level and impact assessments, was compared and analyzed as fundamental data for consistency and future data comparative analysis.

Korea's occupational exposure classification system is broadly classified into education, medical, research, military, public, and industrial. The public is classified into nuclear power plants and public institutions. The industries are classified into non-destructive tests, sales, production & sales, sales & use, and general. In total, it is classified into 11 categories. Through matching with UNSCEAR and international classification standards, we proposed improvement directions for seven major categories and subcategories, as well as detailed work codes for each industry.

Figure 1: radial polygon model



And then, we developed a tool using a radial polygonal model to utilize protection records further. We invented the model using Microsoft's VBA(Visual Basic for Applications) code. This model was developed by setting 10 evaluation factors, and it is possible to analyze the last 5 years by category and compare them with other categories.

Conclusions

In this study, We proposed improving Korea's classification criteria to enable search analysis with international occupational exposure. We developed a radial polygonal model tool for the utilization of protection records. It is expected to be used as basic data for increasing the efficiency of radiation exposure management.

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Development of a phoswich radiation sensor for simultaneous detection and energy spectrometry of beta and gamma radiation

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Introduction

Currently, measurements for radionuclide identification and quantification, which are time- and labor-consuming activities, are separately made. If any beta-gamma emitter is included in the sample, the radionuclide identification measurements are more complicated. Hence, in this study, a phoswich radiation sensor (PHORS) was developed to simultaneously detect beta and gamma radiation and identify radionuclides using radiospectrometry. To check its performance in terms of detection efficiency, the PHORS measurements were compared with those from a commercial Geiger-Müller (GM) counter. To check its capability in terms of energy spectrometry, energy spectra produced by the PHORS were compared with those from a commercial CdZnTe detector.

Description of the Work or Project

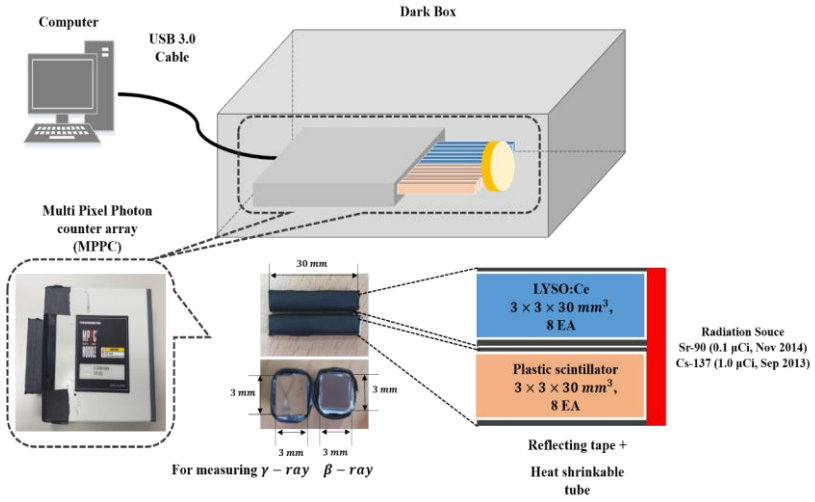

Fig. 1. Experimental setup

Fig. 1. shows experimental setup of PHORS. The PHORS was evaluated with respect to relative detection efficiency and reproducibility as a radiation sensor. To identify its relative detection efficiency, PHORS measurements were compared with those from a commercial Geiger-Müller counter. To test reproducibility, a $\chi 2$-test was applied to the PHORS measurements. Finally, the energy spectra measured by the PHORS were compared with those from a commercial CdZnTe detector to check if the PHORS could produce a reliable energy spectrum.

Conclusions

In this study, the PHORS was developed for simultaneously detecting beta and gamma radiation and identifying radionuclides using energy spectrometry. The test result of showed that the PHORS could reproduce statistically significant measurements. The energy spectra produced using the PHORS were compared with those from a commercial CdZnTe detector to check if the PHORS could produce a reliable energy spectrum. The comparison showed that, though the PHORS could produce energy spectrum of a radioisotope whose shape was similar to that from the CdZnTe detector. In summary, the evaluations showed that the PHORS has capabilities to simultaneously measure beta and gamma radiation and to produce energy spectra to differentiate radiation energy.

Keywords: Phoswich detector, Beta-Gamma radiation, Energy spectrum

Preparation and Characterization of Site-Specifically Radiolabeled ^{89}Zr-DFO-anti-PD-L1-mAb ImmunoPET Tracer

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Introduction

Site-specifically radiolabeled immunoPET tracers have been demonstrated to provide superior imaging ability in vivo than conventional radiolabeled one (random method). In this study, preparation and characterization of site-specifically radiolabeled ^{89}Zr-DFO-anti-PD-L1-mAb tracer will be investigated. The site-specific immunoPET tracer could detect the expression of immune checkpoint protein (ex. PD-L1/PD-1) on tumor for assessment of patient stratification before treatment and therapeutic efficacy after treatment.

Description of the Work

Materials & Methods: The technique of enzymatic glycan modification was utilized to prepare site-specifically radiolabeled ^{89}Zr-immunoPET tracer. In brief, GlyCLICK® and SiteClick® kits were used to prepare azide-activated anti-PD-L1-mAb with degree of labeling of 2 and 4, respectively. Then bifunctional chelator DBCO-DFO was attached to the azide-functionalized antibodies via SPAAC click reaction to form site-specific DFO-anti-PD-L1-mAb conjugates with different chelator-to-antibody ratio (CAR) of 2 and 4. The quality control of conjugates were conducted and then radiolabeled with ^{89}Zr in 1 M HEPES buffer, pH 7, and shaking with 300 rpm at RT for 40 min. In addition, in vitro stability of tracers was estimated in the PBS at RT after purification. **Results:** Both site-specific DFO-anti-PD-L1-mAb conjugates with CAR of 2 and 4 were performed as transparent, clear, without aggregation, chemical purity of 100%, pH of 7.0 – 7.5. Analysis of conjugates by LC-MS showed that CAR for GlyCLICK® and SiteClick® prepared DFO-anti-PD-L1-mAb conjugates was 2.04 and 3.62, respectively. Results from radio-TLC indicated that radiochemical purity of tracers with CAR of 2 and 4 reached 100% and 98.7%, respectively. In addition, the results from HPLC analysis revealed that radioimpurities in both tracers were less than 5%. For in vitro stability study, radiochemical purity of both tracers displayed no any decline until 7 d incubated in PBS at RT.

Conclusions

In this study, site-specifically radiolabeled ^{89}Zr-DFO-anti-PD-L1-mAb tracer with CAR of 2 and 4 have been prepared and characterized. The LC-MS results demonstrate that CAR for GlyCLICK® and SiteClick® prepared DFO-anti-PD-L1-mAb conjugates was 2.04 and 3.62, respectively. Radiochemical purity of tracers with CAR of 2 and 4 were large than 98% and both of them showed excellent stability in vitro.

Keywords: site-specifically radiolabeled; DFO; ^{89}Zr; immunoPET; PD-L1/PD-1

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NOVEL ADSORPTION BIO-MATERIAL FOR RADIOACTIVE COPPER SEPARATION

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Introduction

In this study, an absorbent for radioisotope separation was synthesized using Pectin and Chitosan, which are inexpensive, biodegradable, biocompatible, environmentally friendly, and available in large quantities. Also, since it is a natural product, it has the advantage that it is harmless even if it is introduced into the body in a small amount. Therefore, through the development of an inexpensive and abundant natural product-based radioisotope adsorbent, there is no need to worry about technology dependence and it is possible to produce and separate radioisotopes on their own.

Description of the Work or Project

The adsorption capacity of PC 2:8 and 4:6 was compared up to pH 2~6. As a result of comparison, in the case of PC 2:8, as shown in Fig. 2(a), it can be seen that the adsorption capacity for Zn(II) is high, but the adsorption capacity does not change constantly as the pH increases. However, in the case of PC 4:6, as shown in Fig. 2(b), at pH 2, it exhibits selective adsorption capacity for Cu(II), and at pH 3~6, it shows a similar pattern.

Conclusions

Resins commonly used for radioisotope purification generate many adjuncts and secondary wastes during the pretreatment process. Therefore, it is possible to solve the environmental pollution problem and to expect economic effects through the development of an adsorbent that can purify radioisotopes based on natural products that are easily available in nature. In this study, an adsorbent was synthesized using natural products Pectin and Chitosan, and it was confirmed that Cu(II) selectively adsorbed at a specific condition. Through this, we confirmed the possibility of application as an eco-friendly adsorbent that can be used for radioactive copper purification, which can be used as radiopharmaceuticals.

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Separation of ^{131}Cs from neutron irradiated Ba target for brachytherapy application

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Introduction

Brachytherapy for the treatment of prostate cancer can focus radiation dose on the target site by placing radioactive seeds near the tumor. Compared to other radioisotopes, ^{131}Cs is considered as an effective radioisotope for the brachytherapy due to its high radiation energy and short half-life time. The ^{131}Cs should be separated from ^{131}Ba parent radioisotope because high specific activity is a key parameter for *in vivo* therapeutic application [1, 2]. This paper demonstrates a research on the ^{131}Cs separation process from the neutron irradiated ^{131}Ba target. The separation process is based on the commercial resin packed column. We investigated the influence of parameters (e.g., eluent condition and injection flow rate) on the separation process. The ^{131}Cs radioisotope was obtained from the separation column by the selective elution under the optimized condition, and the radionuclidic purity was investigated using gamma-ray spectroscopy.

Description of the Work and Results

The Ba target was prepared by neutron irradiation of enriched ^{130}Ba carbonate target at the IP-15 hole (flux of thermal neutron: 1.6×10^{14} n·cm^{-2}·s^{-1}) of the HANARO research reactor. The Ba target was dissolved using the nitric acid, and the target solution was injected into the resin (Sr spec, Eichrom Technologies Inc.) packed column. The evaluation of distribution coefficient exhibited that 3 M HNO_3 was the optimum eluent condition for the separation process. The concentration behavior of eluted Ba ions from the separation column was analyzed at various injection flow rates: 0.5 mL/min flow rate was proper for the separation process. Under the optimized conditions, the eluent (i.e., 3 M HNO^3) was injected into the ^{131}Ba loaded column to elute the ^{131}Cs radioisotope selectively. The gamma-ray energy spectrum by the measurement of HPGe detector showed that the eluted solution had energy peaks corresponding to the ^{131}Cs (29.6 and 33.5 keV); the radionuclidic purity of ^{131}Cs was over 99.9%.

Conclusions

This paper demonstrated the separation process based on the chromatography method for production of ^{131}Cs radioisotope. No-carrier-added ^{131}Cs with high radionuclidic purity could be obtained using our process. We expect the separation process presented in this study will be a promising ^{131}Cs production method.

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