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Editorial

Molybdenum-99 (^{99}Mo): Past, Present, and Future

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Molybdenum-99 (^{99}Mo, half-life = 66 h) is a parent radionuclide of a diagnostic nuclear isotope. It decays in technetium-99 m (^{99m}Tc, half-life = 6 h), which is used in over 30 million procedures per year around the world. To meet worldwide demand for ^{99m}Tc-radiopharmaceuticals, fresh ^{99}Mo/^{99m}Tc must be delivered regularly to nuclear medical centers, week after week. The risks of global ^{99}Mo supply disruptions increased significantly since 1995 and have been experienced for different reasons. Since the last quarter of 2007, global Mo-99 supplies have been severely disrupted by recurring operational problems at a handful of aging research reactor (NRU Canada, HFR Netherlands, SAFARI-1 South Africa, BR2 Belgium and OSIRIS France) and processing facilities (MDS Nordion in Canada, Mallinckrodt in the Netherlands, IRE in Belgium, and NTP in South Africa). These few facilities are meeting the bulk of the worldwide demand. With the aging of the ^{99}Mo supply network, it is necessary for world Mo-99 market to consider new reliable supply sources. The present and future shortage of ^{99m}Tc or its parent is a worldwide issue. A research reactor is only one piece of the linear supply chain of ^{99}Mo/^{99m}Tc that exists today. In April 2010, the United States and 46 other countries signed an agreement to phase out HEU for civilian uses to reduce proliferation concerns. Now scientists and engineers involved in ^{99}Mo production are working to determine how to continue to make ^{99}Mo with (1) low-enriched uranium (LEU, <20% ^{235}U) or (2) other alternatives that do not require fissioning of ^{235}U. To overcome the shortage of various routes of its production by accelerators and reactors generating high and low specific activity ^{99}Mo or ^{99m}Tc directly is being researched.

Investigators were invited to contribute original research articles as well as review articles to stimulate the continuing efforts to understand the issues related to production of ^{99}Mo that is acceptable to end users and environmentalists. The article selected for this special issue represent the rich and many-faceted technical know-how that we have the pleasure of sharing with our readers. We would like to thank the authors for their excellent contributions and patience in assisting us. Finally, the fundamental work of all reviewers on these papers is also highly appreciated.

This special issue contains nine papers, where four papers deal with the production of ^{99}Mo using LEU targets in a reactor. Two papers are on ^{99m}Tc generators development and its utilization. One paper deals with the production of ^{99}Mo and ^{99m}Tc using a cyclotron. An article on immobilization of higher activity wastes from reactor production of ^{99}Mo is also included in this issue. One paper deals with research and development of chemical process, hot-cell infrastructure, and commercial production of ^{99}Mo.

In the paper entitled "*^{99m}Tc Generator Development: Up-to-date ^{99m}Tc recovery technologies for increasing the effectiveness of ^{99}Mo utilization*" by V. S. Le (Australia) presents a review on the ^{99}Mo sources available today and on the ^{99m}Tc generators developed up to now for increasing the effectiveness of ^{99}Mo utilization. The latest results of the endeavors in this field are also surveyed in regard to the technical solution for overcoming the shortage of ^{99}Mo supply. The technical topics are grouped and discussed to reflect the similarity in the technological process of each group. The following topics are included in this review.

(i) High specific activity ^{99}Mo (the issues for current production, efforts for more effectiveutilization, the ^{99m}Tc generator based on high-specific activity ^{99}Mo, and ^{99m}Tc concentration units)

(ii) Low specific activity ^{99}Mo (^{99}Mo production based on neutron capture and accelerators' direct production of ^{99m}Tc and the methods of increasing the specific activity of ^{99}Mo using the Szilard-Chalmersreaction and a high power isotope separator.)

The entitled articles "*Immobilisation of higher activity wastes from nuclear reactor production of ^{99}Mo*" by M. Stewart et al. (Australia) discusses progress in waste-form development and processing to treat ANSTO's intermediate-level waste (ILW) streams arising from ^{99}Mo production. The various waste forms and the reason for the process option chosen are reviewed. The options considered were cement-based, glass, glass-ceramic, and ceramic waste forms.

The entitled paper "*Evaluation of ^{99}Mo and ^{99m}Tc productions based on a high-performance cyclotron*" by J. Esposito et al. (Italy) describes possibilities to replace the current reactor-based method with the accelerator ones. A feasibility study was started in 2011 based on the new, high-beam-current, high-energy cyclotron scheduled to be available in the next coming years at Legnaro Laboratories (LNL). A molybdenum metallic target, enriched to 99.05% ^{100}Mo, has been assumed, as it is currently available on the isotopes market. A series of in-target quality parameters has thus been calculated for both radionuclides at the End of Bombardment (EOB), based on the maximum (i.e., 500 μA) proton-beam output current. TENDL 2012 theoretical excitation functions, extended up to (p,6n) (p,p5n) and (p,2p4n) levels, were used to get a detailed map of the radionuclides expected. Results point out that accelerator-^{99}Mo is of limited interest for a possible massive production, mainly because of in-target specific activities, which are a factor of 10^4 lower than reactor ^{99}Mo.

The article entitled "*History and actual state of non-HEU fission-based Mo-99 production with low-performance research reactors*" by S. Dittrich (Germany) discusses that 50 years ago, one of the worldwide first industrial production processes able to produce fission-Mo-99 for medical use had started at ZfK Rossendorf (now HZDR, Germany). On the occasion of this anniversary, it is worth mentioning that this original process (called LITEMOL now) together with its target concept used at that time can be applied still. LITEMOL can be adapted very easily to various research reactors and applied at each site, which maybe still of interest for very small-scale producers. Besides this original process, two further and actually proven processes are suitable as well and recommended for small-scale LEU fission Mo-99 production also.

The article entitled "*Influence of the generator in-growth time on the final radiochemical purity and stability of ^{99m}Tc radiopharmaceuticals*" by L. Uccelli et al. (Italy) describes both theoretical investigations and preliminary irradiations tests on ^{100}Mo-enriched samples. The authors argue that both the ^{99g}Tc/^{99m}Tc ratio and the ^{99m}Tc specific activity will be different in accelerator-produced Tc than that milked from generator, and this might affect radiopharmaceutical procedures. The aim of this work was the evaluation of possible impacts of different ^{99g}Tc/^{99m}Tc isomeric ratios on the preparation of different Tc-labeled pharmaceutical kits. A set of measurements with ^{99m}Tc, eluted from a standard ^{99}Mo/^{99m}Tc generator, was performed and results on both radiochemical purity and stability studies (following the standard Quality Control procedures) are reported for a set of widely used pharmaceuticals (i.e., ^{99m}Tc-Sestamibi, ^{99m}Tc-ECD, ^{99m}Tc-MAG$_3$, ^{99m}Tc-DTPA, ^{99m}Tc-MDP, ^{99m}Tc-HMDP, ^{99m}Tc-nanocolloids, and ^{99m}Tc-DMSA).

The entitled articles, the work of S. G. Govindarajan et al. (USA), "*Assembly and irradiation modeling of residual stresses in low-enriched uranium foil-based annular targets for molybdenum-99 production*" considers a composite cylindrical structure, with low-enriched uranium (LEU) foil enclosed between two aluminum 6061-T6 cylinders. A recess is cut all around the outer circumference of the inner tube to accommodate the LEU foil of open cross-section. To obtain perfect contact at the interfaces of the foil and the tubes, an internal pressure is applied to the inner tube, thereby plastically and elastically deforming it. The residual stresses resulting from the assembly process are used along with a thermal-stress model to predict the stress margins in the cladding during irradiation. The whole process was simulated as a steady-state 2-dimensional problem using the commercial finite element code Abaqus FEA. The irradiation behavior of the annular target has been presented and the effect of the assembly residual stresses has been discussed.

The article entitled "*The fission-based ^{99}Mo production process ROMOL-99 and its application to PINSTECH Islamabad*" by R. Muenze et al. (Germany and Pakistan) presents an innovative process for fission based ^{99}Mo production developed under Isotope Technologies Dresden (ITD) GmbH (former Hans Wälischmiller GmbH (HWM), Branch Office Dresden), and its functionality has been tested and proved at the Pakistan Institute of Nuclear Science and Technology (PINSTECH), Islamabad. Targets made from uranium-aluminum alloy clad with aluminum were irradiated in the core of Pakistan Research Reactor-1 (PARR-1). More than 50 batches of fission ^{99}Mo have been produced that meet the international purity/pharmacopoeia specifications using this ROMOL-99 process. The process is based on alkaline dissolution of the neutron-irradiated targets in the presence of $NaNO_3$, chemically extracting the ^{99}Mo from various fission products and purifying the product by column chromatography.

In the article entitled "*A solution-based approach for Mo-99 production: Considerations for nitrate versus sulfate media*" A. J. Youker et al. (USA) describe how Argonne National Laboratory is assisting two potential domestic suppliers of

Mo-99 by examining the effects of a uranyl-nitrate versus a uranyl-sulfate fuel or target solution compositions on Mo-99 production. Uranyl-nitrate solutions are easier to prepare and do not generate detectable amounts of peroxide upon irradiation, but the effect of a high radiation field (radiolysis of nitrate ion) can lead to a large increase in solution pH, which can lead to the precipitation of fission products and uranyl hydroxides. Uranyl-sulfate solutions are more difficult to prepare, and enough peroxide is generated during irradiation to cause precipitation of uranyl peroxide, but this can be prevented by adding a catalyst to the solution. A titania sorbent can be used to recover Mo-99 from a highly concentrated uranyl nitrate or sulfate solution; however, different approaches must be taken to prevent precipitation of uranium and other fission products during Mo-99 production.

In the article entitled "*Production cycle for large scale fission Mo-99 separation by the processing of irradiated LEU uranium silicide fuel element targets,*" A.-H. A. Sameh (Germany) describes experiments and related high-activity demonstrations that highlight the advantage of uranium silicide fuels as a target material for the production of fission Mo-99. Silicide targets combine features predestinating them as feed materials for a large-scale production of fission nuclides when starting from LEU.

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