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Towards Cleaner Methods for the Production of Mo-99 Using Refractory Ceramics and its Relevance to Actinide Partitioning and Transmutation

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Abstract

Mo-99 is the most utilized isotope in nuclear medicine accounting for over 30 million medical diagnostic procedures annually worldwide. The process for the production of Mo-99 through fission of U-235 normally involves the irradiation of UAl_x dispersion plate fuel in a research reactor, the subsequent dissolution of the fuel plate, the selective separation of the Mo-99 from all of the other fission products and possibly also the recovery of U-235 for future reuse. Compared to the amount of product recovered, copious radioactive waste is generated during the Mo-99 production process. Gaseous wastes are produced at the head-end during the plate dissolution and several liquid wastes are produced during the recovering of Mo-99 using solid extractants, typically polymeric ion exchange resins, which themselves constitute an additional waste stream. It would be extremely advantageous to devise a new process that generates little or no waste. We have been working on a new strategy for the production of fission Mo-99 that involves replacing the dispersion plate targets that are used in the traditional process with inert or active matrix fuel particles that do not need to be dissolved. In one embodiment of the strategy the preparation of new highly porous ZrC_x and graphite-ZrC_x composite target kernels are used that are prepared through polymer templating. The surface properties of these porous materials have been studied and are such that they can be easily loaded with uranium, or for that matter, with any other actinide. In our work we are exploring the possibility of selectively extracting the Mo-99 from the irradiated target kernels by either solution or gas-phase methods and then easily recover the uranium. The fission product-containing kernels can be oxidized in air to generate ZrO₂ that can act as a stable host material either alone or as part of a multiphase ceramic matrix or possibly even as an actinide transmutation host. At the conceptual level, this Mo-99 production strategy could be applicable to the recycling of actinides within the context of advanced fuel cycles and it could indeed act as a useful test-bed.

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1. Introduction

In order to assure the sustainability of nuclear fission energy the need for closed fuel cycles has been highlighted especially by the French researchers [1]. If the use of fast reactors and/or Accelerator Drive Systems (ADS) and closed fuel cycles are to be adopted at some point in the future it will be necessary to partition uranium and plutonium in order to fabricate MOX fuel and then the minor actinides (MAs) americium, neptunium and curium for burning by some other means. MA partitioning from the spent fuel, individually or as a group contemplates either hydrometallurgical (solvent extraction, SX) or pyroelectrometallurgy processing. Both are complex operations with inherent weaknesses, non the least of which is the radiolytic decomposition of extractants and solvents [2]. At the time of this writing hydrometallurgical processing appears to be the more industrially mature technology and reference processes have already been defined. Although the hydrometallurgical approach lends itself well to processing of aqueous PUREX raffinate, the organic solvents and extractants to be used in partitioning by solvent extraction will need to handle extremely high radiation fields that are well known to degrade both solvents and extractants. This weakness has started to receive close attention only relatively recently [3-6]. Even the PUREX process, which is carried out now on an industrial scale, requires a substantial infrastructure to handle waste arisings and improvements are continually being sought [7,8]. Managing these degraded solvents and extractants results in undesirable secondary radioactive wastes that need to be dealt with. Therefore, alternatives to SX should also be considered. What's more, in a P&T strategy some consideration needs to be given to reprocessing irradiated MA-loaded target materials, an area in which little has so far been done.

Molybdenum-99 is the most used radioisotope in nuclear medicine and globally accounts for more than 30 million procedures annually [9,10]. The most common route by far for the production of Mo-99 is through the fission of U-235 [11,12]. The process for producing Mo-99 shares many common elements with spent fuel reprocessing and MA recycling such that it can be used as a test bed. The main steps in the production of Mo-99 are as follows:

1. Fabrication of uranium targets
2. Irradiation of targets in nuclear reactor
3. Dissolution of target
4. Recovery and recycle of U-235 by PUREX or UREX process
5. Recovery of Mo-99 by solid phase extraction
6. Waste treatment and conditioning

The first three steps are similar to what occurs in the conventional nuclear fuel cycle for the production fission nuclear energy. Steps 4 and 5 involve efficient recovery of high value Mo-99 isotope which has a relatively short half-life of only 66 h meaning that during processing time is of the essence. The order of steps 4 and 5 can in fact be reversed depending on whether the head-end step of the process involves HNO₃ (acid-side) or NaOH (base-side) dissolution of the targets. The base-side process results in the precipitation uranium oxide that is filtered off and the Mo-99 extracted from the filtrate by solid-phase extraction. The U-235 can optionally be recovered from the filter cake by the PUREX or UREX liquid-liquid extraction processes, or some other conventional process. In the predominant acid-side process, the acid solution resulting from target dissolution is processed using Al₂O₃-containing columns and the U-235 that is deported to the raffinate could in principle be recovered as described. Both acid- and base-side processes suffer from the production of a range of secondary waste streams including spent resins, ILLW and LLW. In the case of the base-side process the ILLW is a solution of 4 M NaOH containing 0.4 mol/L Al that is difficult to process chemically. This ILLW is also hazardous since it contains an assortment of residual radioactive and non-radioactive contaminants. The major source of radioactivity comes from fission products such as Cs-137 and Sr-90. In the base-side process most of the Sr-90 is deported to the filter cake along with radiolanthanides. A shortfall of supply and waste generation are the primary drivers for the intense research on alternative cleaner and more efficient methods of production that has occurred over the past decade [13,14].

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