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Fission-based Hot Test Production of Mo-99 Using HANARO

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1. Introduction

Molybdenum-99 (^{99}Mo) and its daughter ^{99m}Tc has been the most commonly used medical radioisotope which covers more than 80% of overall nuclear diagnostics. Majority of commercial-scale ^{99}Mo production is based on the fission of ^{235}U. The ^{99}Mo generated from the fission (fission ^{99}Mo) exhibits very high specific activity compared with ^{99}Mo generated from the other routes: neutron activation or accelerator-driven. [1] These days, international ^{99}Mo supply is unstable due to the frequent and unscheduled shutdowns of aged research reactors irradiating ^{99}Mo targets.

In the present, ^{99}Mo is imported from abroad, and stable supply of the isotope is important to provide essential diagnostic services. Besides, export of ^{99}Mo can be a strong driving force to promote radiation and radioisotope industry of Korea. For the purposes, KAERI is developing LEU-based fission ^{99}Mo production process from 2012 to be implemented to the new research reactor (KJRR), which is being constructed in Gijang, Busan, Korea.

Historically, the most ^{99}Mo producers have been used highly enriched uranium (HEU) targets so far. However, to reduce the use of HEU in private sector for non-proliferation, all producers are forced to convert their HEU-based process to use low enriched uranium (LEU) targets. Consequently, overall cost for the production of the fission ^{99}Mo increases significantly with the conversion of fission ^{99}Mo targets from HEU to LEU. It is not only because the yield of LEU is only 50% of HEU, but also because radioactive waste production increases 200%. Therefore, finding optimal treatment of radiowastes from fission ^{99}Mo production process become more important. [2, 3]

2.Development Progress of Fission ^{99}Mo Process

In 2012, development of fission Mo-99 production process has been initiated. Process development was tested via cold experiments with unirradiated depleted uranium (DU) or low enriched uranium (LEU) targets until 2017.

Today, all industrial-scale producers of ^{99}Mo use dedicated targets with a configuration similar to the reactor fuels. Since fuels of early times were generally uranium-aluminum alloy cladded with aluminum shell. KAERI developed plate-type LEU target composed of UAl_x meat dispersed in Al-6061 cladding.

After irradiation of the targets in the research reactor, the targets are dissolved in caustic solution to separate ^{99}Mo out of it. Other fission products including unreacted uranium and actinides are removed from the solution, and collected as a solid radiowastes. In the fission Mo-99 production process with caustic digestion, most iodine remains in the liquid phase as negatively charged iodide form. Isotopes of iodine and soluble elements are collected as a liquid radiowastes from the process stream. Radioisotopes of xenon (Xe) and krypton (Kr) are generated from the fission of Uranium. Major products from the production of fission-based radioisotopes are ^{131m}Xe, ^{133}Xe, ^{133m}Xe, ^{135}Xe, ^{135m}Xe, ^{85}Kr, ^{85m}Kr and ^{87}Kr. Emission of radioxenon from the medical radioisotope production is controlled via gaseous waste treatment system with multiple steps of mitigation and confinement. First, process equipment and production hot cells are made as closed-system with leak-tight parts to minimize effluence of Xe from the system. In spite of the leak-tight systems, it is impossible to completely confine Xe in the system. Therefore, proper combination of equipment to reduce the xenon emission is installed in the medical radioisotope production facility. Finally, medical-grade ^{99}Mo can be extracted after repeated separation and purification steps. [4, 5, 6]

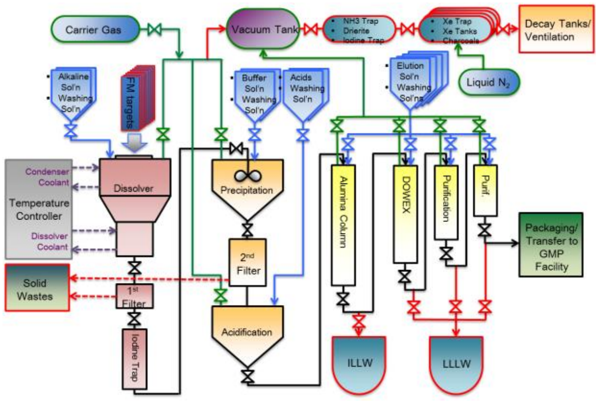

Fig. 1. Scheme for the KAERI's fission Mo-99 process

As a part of the project, KAERI developed technologies to treat radiowastes generated from the ^{99}Mo production. It covers generation, transfer, storage and disposal of liquid, gaseous and solid wastes containing various fission products and salts. KAERI developed new technology to facilitate waste treatment by converting sludge-type waste, which is difficult to handle, into independent solid and liquid wastes. Using this scheme, salt concentration in the ILW can be reduced significantly to make cementation much easier. KAERI developed compact xenon adsorption module using chilled carbon column. Compared with the normal carbon column, KAERI system requires only 1/3,700 carbon in weight or 1/980 in volume for equivalent performance.

3. Result and Discussion

In 2018, hot production test was performed in HANARO site. DU target plates were irradiated in HANARO. And then, the plates were transferred to the hot cells located in the Irradiated Material Examination Facility (IMEF) for the dissolution process. After removal of spent uranium, radioiodines and noble gas from the product, crude ^{99}Mo solution was transported to the hot cells located in the Radioisotope Production Facility (RIPF) for the separation. Inventory of Mo-99 in the irradiated target plates were estimated by calculation. Finally, total amount of produced Mo-99 at each production stage was characterized by gamma spectroscopy.

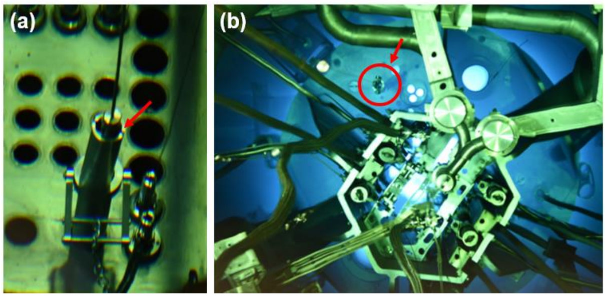

Fig. 2. Irradiation at HANARO. (a) Target capsule and irradiation rig assembly. (b) Loading target rig assembly in the IP5 hole of HANARO.

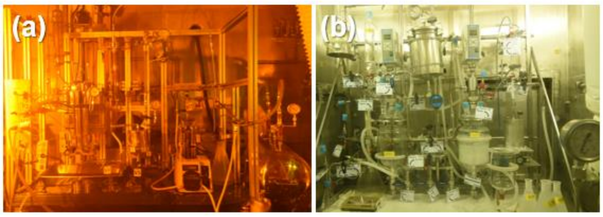

Fig. 3. Fission ^{99}Mo Hot test production systems. (a) System for target dissolution installed in the irradiated material examination facility (IMEF) of KAERI. (b) System for ^{99}Mo separation installed in the radioisotope production facility (RIPF) of KAERI.

5. Conclusions

From the hot test production in HANARO in 2018, over 2 Ci of fission ^{99}Mo has been successfully produced and separated. The result of the process development will be a first step toward commercial-scale ^{99}Mo production in the new research reactor of Korea. KAERI is aiming for the weekly production of 2000 Ci (6-day calibrated) fission ^{99}Mo from the KJRR. The amount corresponds to the 100% of domestic demand, and 20% of international market.

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