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# Treatment of Radiowastes from Fission Mo-99 Production

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## 1. Introduction

Molybdenum-99 ($^{99}$Mo) and its daughter isotope $^{99m}$Tc has been the most commonly used medical radioisotope which covers 85% of overall nuclear diagnostics. 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 (~$10^4$Ci/g) compared with $^{99}$Mo generated from the other routes: neutron activation or accelerator-driven. [1] These days, international $^{99}$ Mo supply is very unstable due to the aging of the main $^{99}$Mo production reactors causing frequent and unscheduled shutdowns. Situation in Korea is even worse, because 100% $^{99}$Mo is imported from abroad.

Under these circumstances, 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. Fission $^{99}$Mo Production Process

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. The targets are irradiated in the KJRR reactor core for about 7 days. Then, irradiated targets transferred from the reactor to the fission $^{99}$ Mo production facility for processing. The targets are dissolved in sodium hydroxide solution to extract $^{99}$Mo from the the solution. Other fission products including unreacted uranium and actinides are removed from the solution. Medical-grade $^{99}$Mo can be extracted after series of separation and purification process. [4, 5, 6]

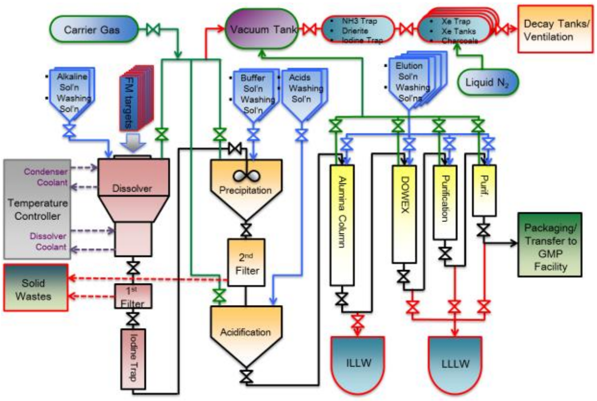

Fig. 2. Scheme for the KAERI's fission Mo-99 process

## 3. Radiowastes from Fission $^{99}$Mo Production

***Liquid Radiowastes:*** From the conversion of target material from HEU to LEU, radioactive wastes increase by decreased production yield. It is because of the increase in number of targets used to have same productivity. Consequently, aluminum and digestion solution in the waste stream increase, too. Especially, reduction of the intermediate level liquid wastes (ILW), directly resulting from the dissolution of target, is very important not only because of their high activity but also difficulties in the solidification treatment for final disposal. Despite of the high salt content in the uranium digestion, alkaline dissolution process was the most efficient for HEU-based targets. However, needs for the development of advanced fission $^{99}$Mo process have been presented due to the dramatic increase of ILW caused by the conversion to LEU. To be solidified for final disposal, the ILW with high salt concentration should be significantly diluted and conditioned with water or other conditioning solutions. Finally, total volume of the wastes increases by 25 folds when solidified with conventional cementation method. Every year, about 15,000 L of ILW is expected for the production 10,000 6-day Ci/week fission $^{99}$Mo from LEU targets, to cover global needs. After cementation, total volume will become even larger. In few years, for the production of fission $^{99}$Mo from LEU-based targets, advanced fission $^{99}$Mo production process with less wastes generation will be required.

In the fission Mo-99 production process with caustic digestion, most iodine remains in the liquid phase as negatively charged iodide form.

***Gaseous Radiowastes:*** 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. However, the conventional system for Xe treatment is huge and costly. This makes difficult to introduce xenon reduction system to the medical radioisotope production facilities. Therefore, development of a compact Xe adsorption module is requested.

## 4. Result and Discussion

***Liquid Radiowastes:*** 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.

***Gaseous Radiowastes:*** Xenon is inert gas with low adsorption characteristic. Therefore, typical xenon treatment system has bulky size with low efficiency. This requires huge space and cost for the establishment of the proper xenon treatment system with desired performance. This is a main obstacle for the radiopharmaceutical producers to equip the xenon treatment system. KAERI developed compact xenon adsorption module with chilled carbon column. At the operation temperature of -20 °C, the system exhibited 149.6 hr delay with 689 g carbon. Compared with the operation at room temperature (20 °C ), only 1/3,700 carbon quantity (in weight) is required (1/980 in volume). It stands for 32 tons of carbon required to achieve 5 GBq/day of CTBTO recommended xenon emission guideline can be reduced down to only 8.5 kg.

## 5. Conclusions

For the weekly productivity of 2000 Ci fission $^{99}$Mo from the KJRR, KAERI is developing the fission $^{99}$Mo production process and is designing the facility. It will cover 100% domestic demand, as well as 18% of international market. 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.

## REFERENCES

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