Development of Silver-exchanged Adsorbents for the Removal of Fission Iodine from Alkaline Dissolution

Taewoon Kim*, Seung-Kon Lee, Suseung Lee, Jun Sig Lee
Division of Radioisotope Research, Korea Atomic Energy Research Institute, 989-111 Daedoek-daero, Yuseong-gu, Daejeon, 305-353 Republic of Korea
**taewoon@kaeri.re.kr*

1. Introduction

Molybdenum-99 (^{99}Mo), the mother of ^{99m}Tc, is the most important isotopes which covers more than 85% of overall nuclear diagnostics. Most of ^{99}Mo is produced by nuclear fission of ^{235}U due to their very high specific activity. For the production of the fission ^{99}Mo, irradiated uranium targets are dissolved in the sodium hydroxide solution, and divided into solid and liquid phase with some fission gas. Solid phase containing uranium and transuranic elements are separated from the liquid by filtration. Then, ^{99}Mo is extracted from the filtrate solution through column-based multistep separation and purification process. In the process, removal of radio-impurities from the solution is essential to acquire high-quality fission ^{99}Mo. Iodine is the main impurity having about 15% of total radioactivity among the whole fission products. Most of the iodine exists in the caustic dissolution as iodide form. KAERI is developing LEU-based fission ^{99}Mo production process which is connected to the new research reactor, which is being constructed in Kijang, Busan, Korea. In KAERI process, silver-exchanged adsorbent is used to adsorb iodide from the solution. Adsorbed iodide can be recovered and recycled for radiopharmaceuticals.

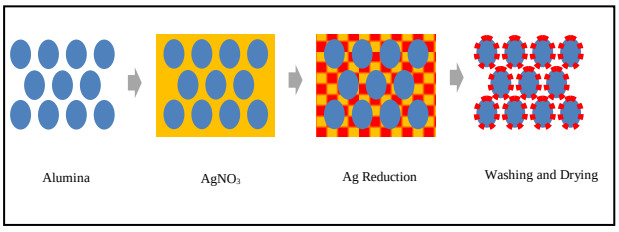
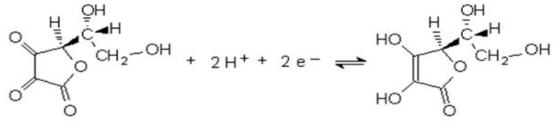


Fig 1. Synthesis scheme of silver-doped alumina

2. Methods and Results

In KAERI's fission ^{99}Mo process, irradiated plate-type LEU targets with UAlx meat and aluminum cladding are dissolved in the sodium hydroxide solution. Undissolved fission products including unreacted uranium and actinides are removed from the solution by filtration. After removal of radioactive iodine, ^{99}Mo can be extracted through the multi-step separation and purification process. Gaseous iodine is removed by copper oxide column installed in the off-gas treatment system. On the other hand, iodides remain in the alkaline dissolution is removed by silver-exchanged silica or silver-doped alumina.

Route 1:



$2Ag^{+}$ + *ascorbic acid* → *2Ag* + *dehydroascorbic acid*

Route 2:

$$R\text{-}CHO + 2Ag(NH_3)_2OH \rightarrow R\text{-}COOH + 2Ag + 4NH_3 + H_2O$$

Fig 2. Synthesis routes for silver-doped alumina. Route 1 is using the ascorbic acid as a reducing agent. Route 2 is silver mirror reaction.

2.1 Synthesis of Silver-doped Alumina

Synthesis of silver-doped alumina is conducted in two ways. One is using the ascorbic acid as a reducing agent. However, this method is impossible to control.
The method proceeds as in the following steps : A selection of alumina to 60~270 mesh, after washing with distilled water and drying in an oven to 80℃. 11ml of 1M $AgNO_3$ is added to 20g of dried alumina. Then compound is dried again. After heating 2M ascorbic acid solution to 50℃, 2ml is added to dried compound. Heat the mixture for 30 minutes. After removing supernatant, the mixture is washed with hot distilled water and then cool distilled water in the order named. Finally, the mixture is heated at 200℃ for 4 hours and then recovering a 60~270 mesh by using the sieve.

The other is silver mirror reaction by using glucose as reducing agent. This experiment was conducted using Aldrich alumina and DAW-70 inert alumina. The method is following steps : A selection of alumina to 60~270 mesh, after washing with distilled water and drying in oven to 80℃. It makes the Tollens' reagent solution in the following respective concentration. (0.5M $AgNO_3$ 5ml /25% NH_4OH 504 μl /H_2O 44.50ml) 10g of alumina and Tollens' reagent solution is mixed

and stirred. 0.5M glucose solution 1375 μl and 0.8M KOH 2740 μl are mixed and added to alumina and Tollens' reagent solution. After a few minutes, 200ml of H_2O is added to mixed solution to terminate reaction. After removing supernatant, the mixture is washed with distilled water many times. Finally, the mixture is heated at 200℃ for 4 hours and then recovering a 60~270 mesh by using the sieve. The method of using a DAW-70 inert alumina is the same as the above method. But DAW-70 inert alumina is used 20g. 0.5M glucose 2750 μl and 0.8M KOH 5480 μl are used. Because density of DAW-70 inert alumina is 2 times than the Aldrich alumina.

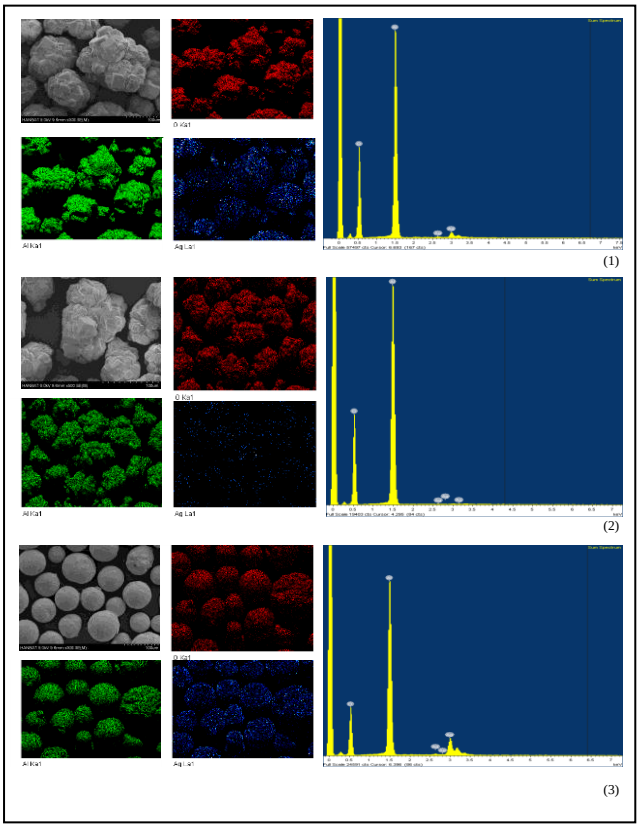


Fig 3. SEM/EDAX data of silver-doped alumina. (1) is data of route 1 method (Fig 2.). (2) and (3) are made by silver mirror reaction. (2) is used Aldrich alumina. (3) is used DAW-70 inert alumina.

2.2 Removal Efficiency of Silver-doped Alumina

Tracer experiments using I-131 have been performed for the proof of concept. Operation conditions: 1.23 g adsorbent in column (9.3 mm ID), 66 mL loading solution (1.6 mg/L iodine), 10 mL/min flow rate, Iodine recovered using 20 mL Na_2S solution. S^{2-} (k_{SP}=6×10^{-51})

Iodine Removal	99.55%	95.38%	99.65%
Iodine Recovery	76.37%	76.41%	70.14%
Iodine Adsorption per1 g Ag-alumina	0.0815 mg/g	0.0828 mg/g	0.0861 mg/g

Table 1. Removal efficiency of silver-doped alumina column

2.3 Loss of Molybdenum

Operation conditions : 1.23 g adsorbent in column (9.3 mm ID), 66 mL loading solution (Moly in 3M NaOH/4M $NaNO_3$ 2mg/L), 10 mL/min flow rate, washing with 1M NaOH. This experiment was measured by AAS. (AAS : Atomic absorption spectroscopy)

Sample Name	Conc(ppb)
Before Mo	28.0477
After 1	21.1383
After 2	3.4182
After 3	18.5156
Washing 1	4.2132
Washing 2	3.2133
Washing 3	2.3691

Table 2. Loss of molybdenum. Number 1 is column of route 1. (Fig 2.) Number 2 is column of silver mirror reaction by using Aldrich alumina. Number 3 is column of silver mirror reaction by using DAW-70 inert alumina.

3. Conclusions

In KAERI process, silver-exchanged adsorbent is used to adsorb iodide from the solution. Adsorbed iodide can be recovered and recycled for radiopharmaceuticals.

REFERENCES

[1] International Atomic Energy Agency, "Non-HEU Production Technologies for Molybdenum-99 and Technetium-99m", IAEA Nuclear Energy Series No. NF-T-5.4, (2013).
[2] International Atomic Energy Agency "Fission Molybdenum for Medical Use", IAEA-TECDOC-515, (1989).
[3] International Atomic Energy Agency , "Management of Radioactive Waste from ^{99}Mo Production", IAEA-TECDOC-1051, (1998).
[4] M. V. Wilkinson, A. V. Mondino, A. C. Manzini, *J. Radionanl. Nucl. Chem.*, **256**, 413-415 (2003).