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

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1. Introduction

^{99m}Tc is the most important isotopes which covers more than 80% of overall nuclear diagnostics. Half-life of ^{99m}Tc is 6 hours. Preferentially, we should produce ^{99m}Tc's mother, ^{99}Mo. 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.

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

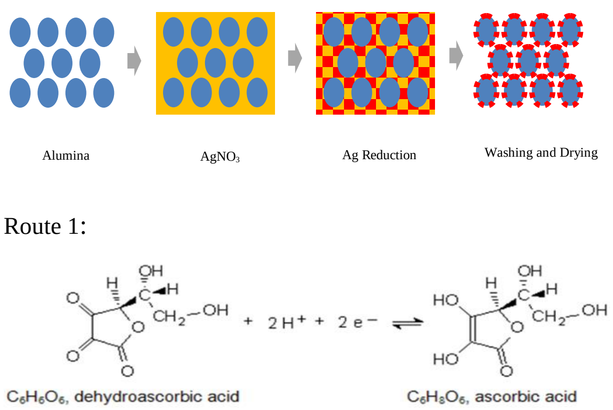
2. Methods and Results

In fission ^{99}Mo process, 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.

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. after washing with distilled water and drying in an oven. $AgNO_3$ solution is added dried alumina. Then compound is dried again. After heating ascorbic acid solution, solution is added to dried compound. Heat the mixture. 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 and then recovering by using the sieve.



$2Ag^+$ + ascorbic acid → 2Ag + dehydroascorbic acid

Route 2:

R-CHO + $2Ag(NH_3)_2OH$ → R-COOH + 2Ag + $4NH_3$ + H_2O

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

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, after washing with distilled water and drying in oven. It makes the Tollens' reagent solution in the following respective concentration. Alumina and Tollens' reagent solution is mixed and stirred. glucose solution and KOH are mixed and added to alumina and Tollens' reagent solution. After a few minutes, 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 and then recovering by using the sieve. The method of using a DAW-70 inert alumina is the same as the above method.

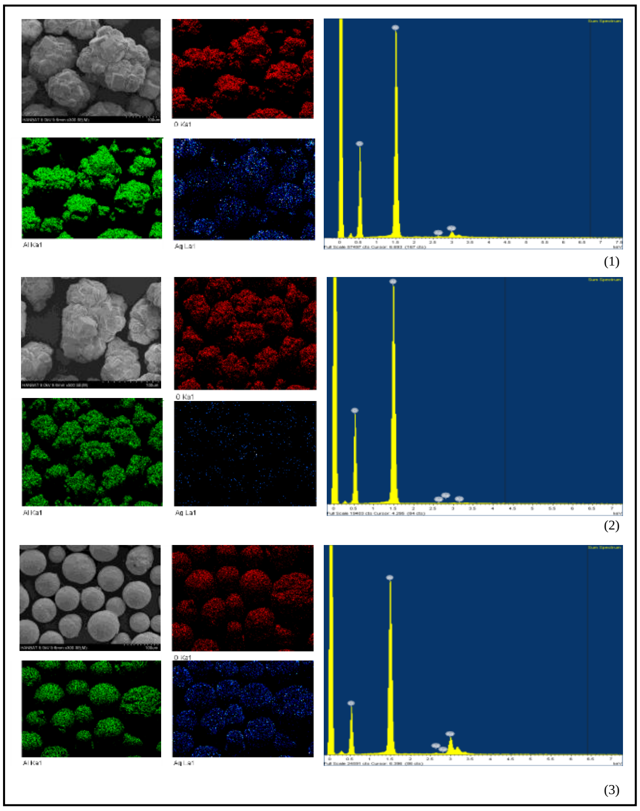


Fig 3. SEM/EDAX data of silver-doped alumina. (1) is data of route 1 method (Fig 2.). (2) and (3) are made by silver mirror reaction. (2) is used Aldrich alumina. (3) is used DAW-70 inert alumina.

2.2 Removal Efficiency of Silver-doped Alumina

Tracer experiments using I-131 have been performed for the proof of concept. Operation conditions: Adsorbent in column, loading iodine solution, Iodine recovered using Na_2S solution.

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 The supernatant analyzed

The supernatant of silver-coated alumina by using ascorbic acid is yellow color. But the supernatant of silver-coated alumina by using silver mirror reaction is transparent. Pure alumina of Aldirch, pure DAW-70 alumina, Ag-doped alumina(Aldrich) and Ag-doped alumina(DAW-70). Each of alumina put in test tube and added NaOH. After few minutes of stirring, supernatant is filtered and measured by UV-Vis.

Remelting of silver and aluminum in alkaline dissolution

Each alumina of Ardrich Company, DAW-70 alumina, silver mirror reaction Ag-coated alumina(Adrich) with $AgNO_3$ solution and silver mirror reaction Ag-coated alumina(DAW-70) $AgNO_3$, and NaOH solution are put in Test Tube. Stirring for few minutes. After filteration, samples are measured by ICP-MS.

3. Conclusions

In this study, silver-exchanged adsorbent is used to adsorb iodide from the solution. Adsorbed iodide can be recovered and recycled for radiopharmaceuticals. Silver-doped DAW-70 alumina by using silver mirror reaction is less impurities and simpler than method using ascorbic acid.

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