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Removal of Radioactive Iodine Using Silver-Adsorbed Alumina from Sodium Hydroxide Solutions.

Taewoon Kim[a], Seung-Kon Lee[b,*], Suseung Lee[b], Jun Sig Lee[b], and Sang Wook Kim[a,*]
[a]*Analytical Chemistry Laboratory, Department of Advanced Materials Chemistry, Dongguk University, Gyeongju, Gyeongbuk 780-714, Republic of Korea*
[b]*Division of Radioisotope Research, Korea Atomic Energy Research Institute, 989-111 Daedoek-daero, Yuseong-gu, Daejeon 305-353, Republic of Korea*
**Corresponding author: seungkonlee@kaeri.re.kr, swkim@doungguk.ac.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. In this study, silver adsorbed alumina is used to adsorb iodide from the alkaline solution. Alumina is suitable under alkaline solution and is a suitable inert support that does not adsorb ^{99}Mo. The developed adsorbents with less impurities into the processed solution and the synthesis method is simple.

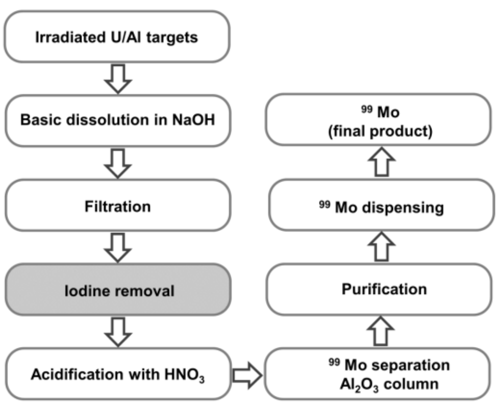

Fig. 1. Overall scheme of ^{99}Mo production. After dissolving the target with a strong base solution, filter the metal impurities. For purification of ^{99}Mo, the iodine present in the solution must be removed before the solution is acidified.

2. Methods and Results

2.1 Synthesis of Silver-Adsorbed 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.

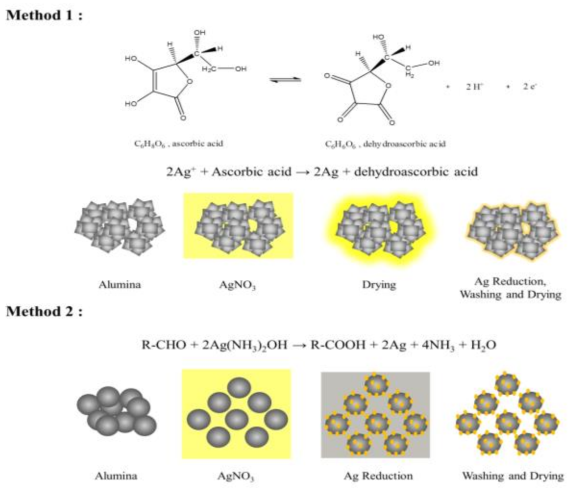

Fig. 2. Synthesis of silver-adsorbed alumina using ascorbic acid and the silver mirror reaction. In the case of Method 1, silver nitrate is mixed with acidic alumina, and the surface silver is reduced by using ascorbic acid in the presence of silver nitrate on the alumina surface. In the case of Method 2, silver is adsorbed on the surface of alumina while silver is reduced using reducing sugar while spherical alumina is stirred in a solution together with silver nitrate.

The other is silver mirror reaction by using glucose as reducing agent. This experiment was conducted using commercial alumina and Spherical 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 spherical alumina is the same as the above method.

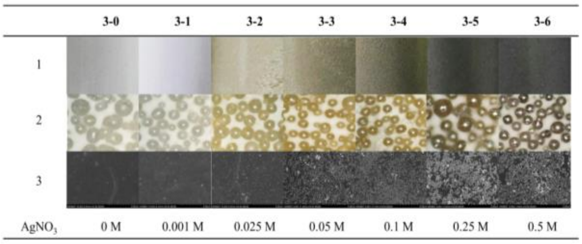

Fig. 3. (1), (2) Color of silver-adsorbed alumina (spherical) as a function of $AgNO_3$ concentration (silver mirror reaction). (3) SEM images of Ag NPs on the alumina surface. As the concentration of silver nitrate increases, the color of silver-adsorbed alumina (spherical) gradually increases (1), and it is confirmed by optical microscope (2). Silver nanoparticles adsorbed on the alumina surface increase in size and become larger (3).

Figure 3 shows that the color of silver-adsorbed spherical alumina and the size of Ag NPs thereon depended on the concentration of $AgNO_3$. The produced Ag NPs exhibited diameters of 75–340 nm, and the surface of silver-adsorbed alumina (spherical) had a silver content of 12.87%. Section 1 of the above figure shows the color of Ag-adsorbed spherical alumina, as seen by the naked eye, and section 2 represents optical images of Ag-adsorbed spherical alumina. Finally, section 3 shows SEM images of Ag NPs on the alumina surface. The increasing concentration of silver nitrate caused an overall darkening, increasing the size and density of Ag NPs adsorbed on the alumina surface.

2.2 Breakthrough test

For breakthrough experiment, silver adsorbed alumina was filled in a column with 3 cm height and 0.93 cm diameter. Prepared loading solution was loaded into the column at a rate of 0.9, 2.5 and 4 mL/min, respectively.

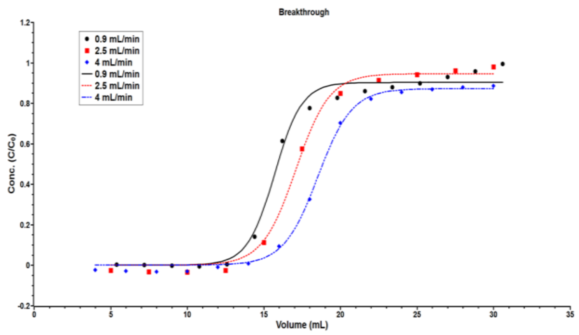

Fig. 4. Breakthrough curve. ● is measured every 2 minutes when the sample was passed through the column at 0.9 mL/min. ■ is measured every minute when the sample was passed through the column at 2.5 mL/min, and ◆ is measured every 30 seconds at 4 mL/min.

Figure 4 shows breakthrough curves according to different loading rates. 5% of saturation concentration was designated as breakthrough point. The amount of iodine removed from 1 g of adsorbent was 0.102, 0.102 and 0.106 mg, respectively. The average was 0.103 mg.

2.3 Degree of Ag and Al leaching

Ag/Al leaching was investigated to evaluate the stability of silver-coated alumina prepared using the silver mirror reaction in basic solution. The above evaluation was performed by treating 0.5-g of silver adsorbed alumina (commercial, Sample 2-2), silver adsorbed alumina (spherical, Sample 3-6) and commercial acidic/spherical alumina with NaOH solution and shaking at room temperature for 30 min. The mixtures were filtered, and the filtrates were analyzed using ICP-MS.

Table 1. Concentrations of Al and Ag leached from Ag-coated alumina samples prepared by the silver mirror reaction, as determined by ICP-MS.

	Concentration (ppb)			
Sample / Element	**Acidic alumina**	**Spherical alumina**	**Sample 2-2**	**Sample 3-6**
Al	213077.85	10209.98	151637.73	27498.11
Ag	1.34	0.32	11413.47	1752.21

The results of leaching tests performed for basic solutions of samples 2-2 and 3-6 using ICP-MS are shown in Table 1, revealing that acidic alumina showed a ~20 times higher concentration of leached Al than spherical alumina, confirming that the latter is more durable than the former in basic solution. Table 1 shows that the concentration of Ag leached from sample 3-6 was ~6.5 times lower than that leached from sample 2-2.

2.4 Removal of ^{131}I

Table 2. Removal efficiency of silver-adsorbed alumina column.

Ag-coated alumina	Sample 1-6	Sample 2-2	Sample 3-6
Total iodine removal	>99%	>99%	>99%
Iodine Adsorption per1 g Ag-alumina	0.0815 mg/g	0.0828 mg/g	0.0861 mg/g

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. Sample 1-6 is silver-adsorbed alumina by using ascorbic acid.

3. Conclusions

In this study, silver-exchanged adsorbent is used to adsorb iodide from the solution. Samples prepared using ascorbic acid or silver mirror methods achieved high ^{131}I removal efficiencies regardless of alumina type. Silver-adsorbed alumina (spherical) by using silver mirror reaction is less impurities and simpler than method using ascorbic acid.

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