

Study for Development of Magnetic Particles for Remove Radioactive Cesium and Iodine

Taewoon Kim^a, Seung-Kon Lee^{b,*}, Suseng Lee^b, Jun Sig Lee^b and Sang Wook Kim^{a,*}

^aAnalytical Chemistry Laboratory, Department of Advanced Materials Chemistry, Dongguk University, Gyeongju, Gyeongbuk 780-714, Republic of Korea

^bDivision of Radioisotope Research, Korea Atomic Energy Research Institute, 989-111 Daedok-daero, Yseonggu, Daejeon 305-353, Republic of Korea

*Corresponding author: swkim@doungguk.ac.kr and seungkonlee@kaeri.re.kr

1. Introduction

Radioactive cesium (Cs) is difficult to treat or remove, because Cs has a very long half-life. Among the nuclides that flowed out due to the Fukushima Daiichi nuclear disaster, Cs occupies a very large amount. Research is underway to remove Cs after an accident. Especially, Prussian blue (PB) is used to remove Cs [1,2]. Radioactive iodine (I) has many nuclides that are used in many fields such as industry, research and medicine. Also fission iodine is most often produced with molybdenum-99 from fission uranium. It is superior in sublimation and exists in liquid or gaseous state. Therefore, much research has been done to remove iodine. The purpose of this study is to develop magnetic particles that simultaneously remove radioactive Cs/I. Iron oxide, magnetic nanoparticles (MNPs), was used as a support and Prussian blue was used for cesium removal. Finally, silver was used to remove iodine.

2. Methods and Results

The iron oxide synthesis methods are various [3]. Among them, the low-temperature synthesis method was selected [4]. After that, Prussian blue is functionalized. Finally, the silver nanoparticles are coated using a silver mirror reaction with Tollens' reagent [5]. The removal of cesium and iodine with the completed Ag-PB-MNPs was tested respectively. All reaction use mechanical stirring without magnetic stirring.

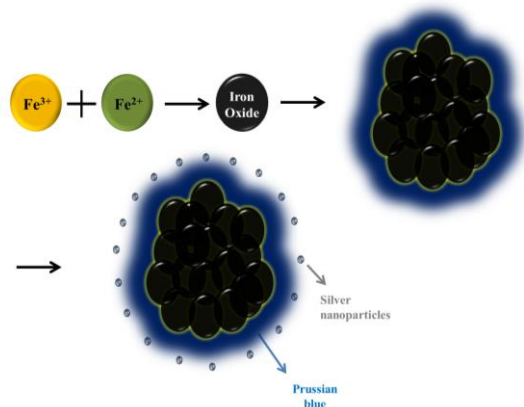


Figure 1. Synthesis of silver-prussian blue coated magnetic particles.

2.1 Synthesis of magnetic nanoparticles (Iron oxide)

There are various methods for the synthesis of iron oxide, such as coprecipitation, sol-gel, thermal decomposition and hydrothermal reaction. This experiment is a method of synthesizing iron oxide whose surface is hydrophilic at a relatively low temperature. The reaction proceeded under anaerobic and citric acid was used as a surfactant. $FeCl_3 \cdot 6H_2O$ and $FeCl_2 \cdot 4H_2O$ dissolved in distilled water. The solution is purged with nitrogen while heating to 70 °C. After adding ammonia solution and citric acid solution, raise the temperature to 90 °C and wait for 1 hour. Finally, centrifuge or magnetic decantation with distilled water wash. These synthesized particles become magnetized and react to the magnets.

2.2 Synthesis of PB-MNPs

The dried magnetic nanoparticles are completely dispersed in distilled water and then adjusted to a specific pH. Then add potassium hexacyanoferrate and stirring for 1 hour. Magnetic decantation is performed while washing with distilled water.

2.3 Synthesis of Ag-PB-MNPs

Silver has been widely used to remove iodine. In this experiment, silver was coated using a silver halide reaction with glucose as a reducing agent. When the solution is more basic than when it is acidic, the reduction of silver occurs more easily. Therefore, when glucose is added in this reaction, sodium hydroxide is added together. First, a Tollens' reagent containing a specific concentration of silver nitrate is prepared. At this time, when silver is added in excess, silver is aggregated and it is difficult to separate from synthesized Ag-PB-MNPs. In addition, the magnetism of Ag-PB-MNPs weakens and the magnet does not react. Conversely, if silver is added in too small amounts, it is difficult for Ag-PB-MNPs to remove iodine. Therefore, a suitable concentration of silver nitrate should be found. Add PB-MNPs to the prepared Tollens' reagent solution and stirring vigorously. Drop wise glucose and KOH solution to the mixed solution. When the color of the solution turns to light brown, the reaction is terminated

by adding distilled water. Magnetic decantation, washing with distilled water and drying in oven.

2.4 Removal of cesium

Ag-PB-MNPs were added to the cesium solution, and the mixture was stirred for 30 minutes. The supernatant was separated and measured by AAS.

Table 1. Concentration of cesium

	Cs standard	Sample 1
Abs	0.4026	0.1658
Conc. (ppb)	24.3848	10.6860

Table 1 shows that the concentration of sample 1 was lower than that of conventional cesium but lowered the cesium removal rate by the coated silver.

2.5 Removal of iodine

KI-I₂ solution is prepared and diluted. Add Ag-PB-MNPs to the solution and stirring for 30 minutes. Separate the supernatant and measure with UV-Vis.

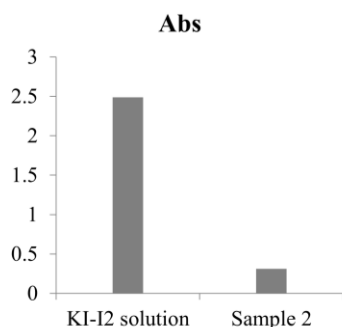


Figure 2. Removal of iodine measured by UV-Vis.

The figure 2 shows that the absorbance of sample 2 is significantly lowered. The iodine is almost completely removed by silver.

3. Conclusions

In this study, we have fabricated magnetic particles that can remove cesium and iodine present in water. The removal of cesium and iodine was confirmed and the magnetic properties of the particles were confirmed. However, there is a limit in that the dispersibility of the particles is lowered by coated silver and the removal rate of cesium is lowered. If the removal rate of cesium is increased, it can be commercialized.

REFERENCES

[1] S. C. Jang, S. B. Hong, H. M. Yang, K. W. Lee, J. K. Moon, B. K. Seo, Y. S. Huh, and C. H. Roh, Removal of

Radioactive Cesium Using Prussian Blue Magnetic Nanoparticles, *Nanomaterials*, Vol. 4, p.894-901, 2014.

[2] H. M. Yang, S. C. Jang, S. B. Hong, K. W. Lee, C. H. Roh, Y. S. Huh, and B. K. Seo, Prussian blue-functionalized magnetic nanoclusters for the removal of radioactive cesium from water, *Journal of Alloys and Compounds*, Vol.657, p.387-393, 2016

[3] S. Laurent, D. Forge, M. Port, A. Roch, C. Robic, L. V. Elst, and R. N. Muller, Magnetic Iron Oxide Nanoparticles: Synthesis, Stabilization, Vectorization, Physicochemical Characterizations, and Biological Applications, *Chemical Reviews*, Vol.108, No.6, p.2064-2110, 2008.

[4] S. Nigam, K. C. Barick, D. Bahadur, Development of citrate-stabilized Fe₃O₄ nanoparticles: Conjugation and release of doxorubicin for therapeutic applications, *Journal of Magnetism and Magnetic Materials*, Vol.323, p.237-243, 2011.

[5] H. R. Hwang, S. H. Kim, and S. M. Yang, Microfluidic fabrication of SERS-active microspheres for molecular detection, *Lab on a Chip*, Vol.11, p.87-92, 2011.