

Comparative study on the preparation of Uranium source using a Cerium fluoride Co-precipitation method and an Electrodeposition method

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1. Introduction

In nuclear safeguards application uranium particles in the air around a suspicious building can be analyzed for radioactive materials. This analysis can be used to identify the undeclared nuclear activities of North Korea. The international community has developed a method of collecting and analyzing uranium particles within a limited time in the field to determine whether or not undeclared nuclear activities have taken place [1]. A fast and convenient method of the uranium source is important in order to finish the inspection within a limited time. There are two different methods used to prepare a uranium source. One method is called the electrodeposition [2-4] and the other is the co-precipitation method [5-7]. The purpose of this study is to develop an optimal method of a uranium source in the field.

2. Methods and Materials

2.1 Filter and Air trapped preparation

Ashless material filters (Staplex TFA810) for trapping particles of uranium were prepared. The filter has an efficiency of 95%, 1 μm particles. A ^{232}U (NPL) tracer and SRM 3164 (Standard reference material : NIST) were used to obtain a chemical yield from the sample. Comparative values following the pre-treatment were obtained. After filters moistened by ^{232}U tracer and SRM 3164 were dried for one day in a fume hood, sampling started. Particles of uranium in the atmosphere were then collected.

Air trapping was carried out from the ground (1.5) m. Particles were trapped by the filter. The air sampler, which is used for sampling is represented in Fig. 1. The sampling flow rate used was about 500 SLM (Standard liter per minute) when the filter is clean. The sampling time was maintained at 12 hours. Each filter was placed in a clean square dish during transport and storage. A total of five samples were prepared.

2.2 Sample pre-treatment

Dust particles trapped by air filters were put into a glass beaker. Later, 40 ml of 3M HNO_3 was added. Figure 2 shows the Ultrasonic cleaner (MTH-3510, Branson).

Distilled water was poured into the cleaner water tank

and a glass was placed over the beaker as a cover. The operating time was one hour. The Ultrasound device frequency was 42 kHz. After finish the ultra-sonification, the filters in the beaker were removed. The experiments were carried out using the remaining solution. This solution was heated to 160 ~ 180 $^{\circ}\text{C}$, than stirred on a hot plate for several hours. After completing the drying process, the solution was cooled with 30 ml of 8M HNO_3 . This sample solution was then ready for the purification procedure with a column chromatography.



Fig. 1. Portable air sampler used for the collection of uranium particles



Fig. 2. Ultrasonic cleaner

Table I : Experimental condition

	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
Sampling time	12 hr	12 h	12 h	12 h	12 h
Flow rate	500 SLM	500 SLM	500 SLM	500 SLM	500 SLM
²³² U tracer	500 μ l	500 μ l	500 μ l	500 μ l	500 μ l
SRM 3164	X	X	X	250 μ l	250 μ l
Column work	O	O	O	O	O
Source preparation	Electro-deposition	Co-precipitation	Co-precipitation	Electro-deposition	Co-precipitation

2.3 Separation of uranium on UTEVA resin

The UTEVA resin (100-150 μ m, Eichrom Industries Inc.) was soaked in 1M HNO₃ for one day. Aliquots of the slurry were then poured into a disposable column up to a bed volume of about 0.6 ml [8]. The UTEVA column was preconditioned with 8M HNO₃ prior to its use. The flow rate was maintained at about 0.15 ml/min by a gravity elution. After conditioning the UTEVA column with 20 ml of 8M HNO₃, 30 ml of the sample solution was added into the column. After loading the sample solution, the column was washed with 20 ml of 8M HNO₃. Later, the column was washed with 5 ml of 9M HCl and 20 ml of 5M HCl - 0.05M oxalic acid. Finally, the uranium was eluted with 30 ml of 0.01M HCl [9-10].

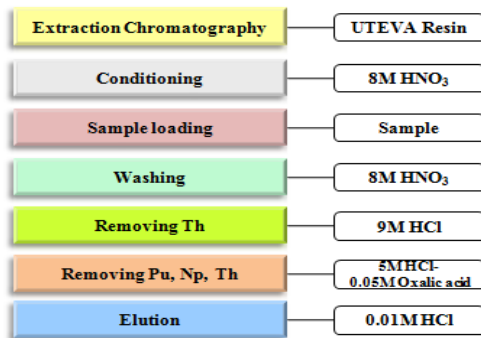


Fig. 3. Scheme of the column work using UTEVA resin

2.4 Sample preparation using electrodeposition and sample measurement

- 1) Eluted uranium solution was transferred into a 50 ml glass beaker and 2 ml of 1M HNO₃, 100 μ l of H₂O₂, 1 ml of HClO₄ were added.
- 2) The mixed solution was evaporated very carefully to dryness on hotplate.
- 3) The dry residue was re-dissolved in 500 μ l of concentrated HNO₃. If necessary, the solution was warmed up in order to dissolve the residue completely.
- 4) After finish the dryness, glass beaker was moved and cooled down.

- 5) 2 drops of Methyl red indicator added into the glass beaker.
- 6) The pH was adjusted to 1.8 ~ 2.0 with (1+9) H₂SO₄ and concentrated NH₄OH. This was done slowly, as the reaction is exothermic and the solution needs to cool to room temperature. At pH 1.8~2.0, the color of the solution changes to bright red.
- 7) The solution was transferred into the electro-deposition equipment with stainless steel disk.
- 8) Turn on a current, adjusted to 1 A, for 2 hours.
- 9) Before the end of electrodeposition, 1 ml of concentrated NH₄OH was added into the cell.
- 10) Rinse the stainless steel disk with the distilled water and hold it for 30 seconds in the flame of a torch.
- 11) Measure the alpha activity of the sample using alpha spectrometer manufactured CANBERRA Inc.

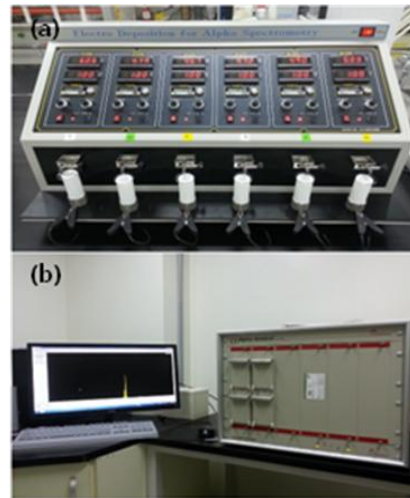


Fig. 4. (a) Electrodeposition system, (b) Alpha spectrometer

2.5 Sample preparation using CeF₃ coprecipitation and sample measurement

- 1) Eluted uranium solution was transferred into a capped 50 ml test tube and 100 μ l of cerium carrier solution, 500 μ l of TiCl₃, 1 ml of 48% HF were added.
- 2) The solution well mixed using mixer. Let stand at least 5 min.
- 3) Pour the solution filtering system with ResolveTM filter (0.1 μ m polypropylene).
- 4) Finally, the precipitate was deposited onto a filter using a vacuum filtration system.
- 5) The filter was cleaned with 5 ml of 80% ethanol and 1 ml of D.I water. Excess ethanol and distilled water eliminated under vacuum.
- 6) After filtering system disassembled, the filter with radioactive deposit was placed on a stainless steel disk.

- 7) It dried under an UV lamp for 5 minutes.
- 8) Measure the alpha activity of the sample using alpha spectrometer manufactured CANBERRA Inc.

CeF₃ coprecipitation method. The Electrodeposition method has many steps and is complicated. However, the CeF₃ coprecipitation procedure is so simple and robust that an unskilled person can easily perform the alpha source preparation.



Fig. 5. Vacuum filtering apparatus

Table II : Experimental Results

	Time (s)	Counts				Yield (%)
		²³⁸ U	²³⁵ U	²³⁴ U	²³² U	
Sample 1	100,000	X	X	X	580	96 %
Sample 2	100,000	X	X	X	541	93.5 %
Sample 3	100,000	X	X	X	503	92.6 %
Sample 4	100,000	95	9	195	456	86.2 %
Sample 5	100,000	85	3	109	479	89.8 %

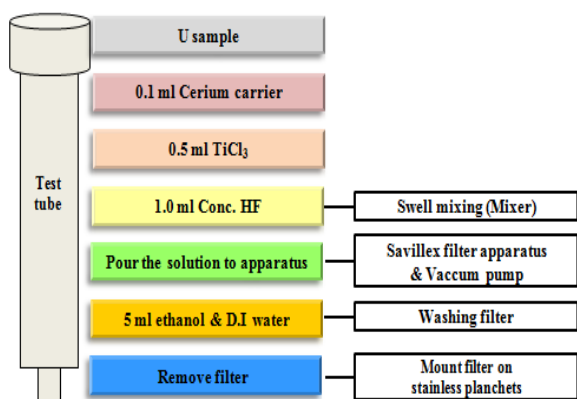


Fig. 6. Scheme of the CeF₃ coprecipitation for uranium sample

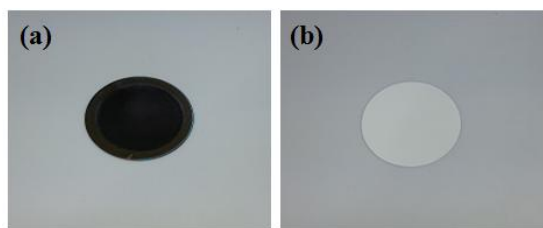
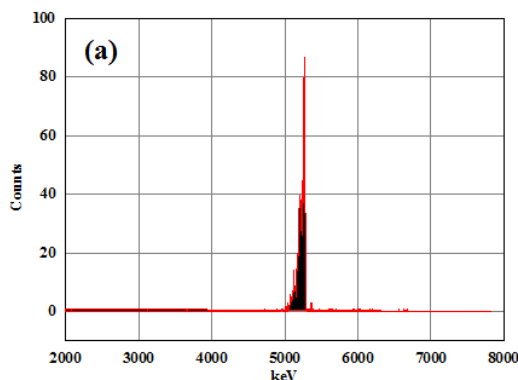
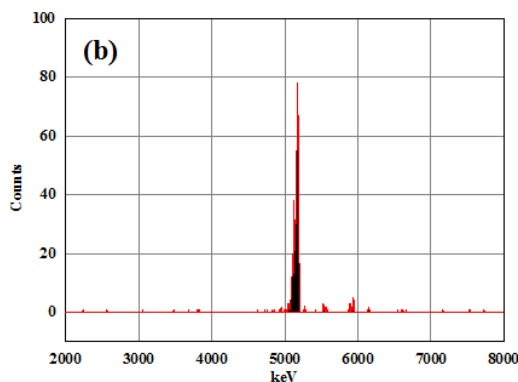


Fig. 7. (a) U source prepared by electrodeposition (b) U source prepared by CeF₃ coprecipitation.



3. Results and Discussion

The electrodeposition method and CeF₃ coprecipitation method were compared in this study.

As shown in Table II, the recovery of ²³²U using the CeF₃ co-precipitation method was similar to that using the electro-deposition method. Alpha spectra of uranium source prepared by electrodeposition and CeF₃ coprecipitation are shown Figure 8. An electrodeposition time required was more than two hours, while less than 30 minutes was needed for the

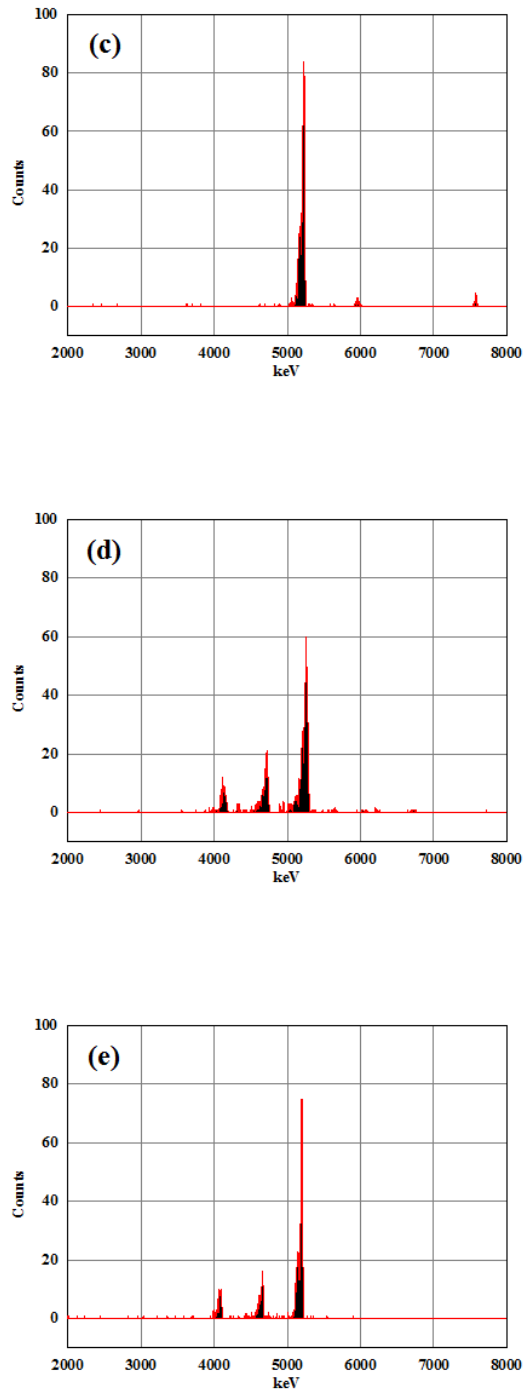


Fig. 8. (a) Sample 1, (b) Sample 2, (c) Sample 3, (d) Sample 4, (e) Sample 5 Alpha-particle spectra of uranium source isolated from air filter

4. Conclusions

This study shows the U source using CeF_3 coprecipitation has no significant difference. In addition, this CeF_3 coprecipitation method is simple and time saving. It is expected to be useful for rapid on-site detection of undeclared uranium enrichment activities.

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