

## A Study of Simple $\alpha$ Source Preparation Using a Micro-coprecipitation Method

Myung Ho Lee,\* Tae-Hong Park, Byung Chul Song, Jong Ho Park, Kyuseok Song

Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, Daejeon 305-353, Korea

\*E-mail: mhlee@kaeri.re.kr

Received July 29, 2012, Accepted August 21, 2012

This study presents a rapid and simple  $\alpha$  source preparation method for a radioactive waste sample. The recovery of  $^{239}\text{Pu}$ ,  $^{232}\text{U}$  and  $^{243}\text{Am}$  using a micro-coprecipitation method was over 95%. The  $\alpha$ -peak resolution of Pu and Am isotopes through the micro-coprecipitation method is enough to discriminate the Pu and Am isotopes from other Pu and Am isotopes. The determination of the Pu and Am isotopes using the micro-coprecipitation method was applied to the radioactive waste sample, so that the activity concentrations of the Pu and Am isotopes using the micro-coprecipitation method in the radioactive waste sample were similar to those using the electrodeposition method.

**Key Words :** Pu isotopes, Am isotopes, Electrodeposition, Micro-coprecipitation

### Introduction

For the determination of actinides such as Pu, U, and Am isotopes, an  $\alpha$  spectrometric method is generally used after radiochemical separation and  $\alpha$  source preparation. An  $\alpha$  source with a remarkable energy resolution is required to accurately analyze such  $\alpha$  emitters of the actinide nuclides. The sources for  $\alpha$  spectrometric measurements have usually been prepared using vacuum sublimation,<sup>1</sup> direct evaporation,<sup>2</sup> electrodeposition,<sup>3-5</sup> and micro-coprecipitation.<sup>6,7</sup>

The direct drop deposition method, although this method is simple and rapid, has disadvantages in that an actinide cannot be deposited evenly on the surface of a stainless steel disk, and its energy resolution is inferior to that of sources prepared through other methods because of the self-absorption of the  $\alpha$  particles. In addition, vacuum sublimation is rarely used since equipment for  $\alpha$  source preparation is very expensive. Among the  $\alpha$  source preparation methods for  $\alpha$  spectrometry, electrodeposition is a commonly used method since it provides a good energy resolution of the  $\alpha$  peak. However, in the electrodeposition method, it is quite difficult precisely to control the pH in an electrodeposition solution. In addition, Fe ions and organic substances in the electroplating solution cause low electrodeposition recovery.<sup>10</sup>

Recently, in view of the difficulties associated with electrodeposition, alternative techniques making use of micro-coprecipitation with rare earths have been developed to yield sources for  $\alpha$ -spectrometry.<sup>8,9</sup> Although the micro-coprecipitation method is simple and more rapid than electrodeposition, the energy resolution (Full-Width at Half-Maximum, FWHM) in the  $\alpha$  spectrum with micro-coprecipitation is worse than that with electrodeposition. However, to improve the energy resolution, Resolve<sup>TM</sup> filters (0.1  $\mu\text{m}$  polypropylene) provided by the Eichrom Technologies have recently been used for  $\alpha$  source preparation, instead of conventional membrane filters.

In this study, using radionuclide tracers, the recovery and

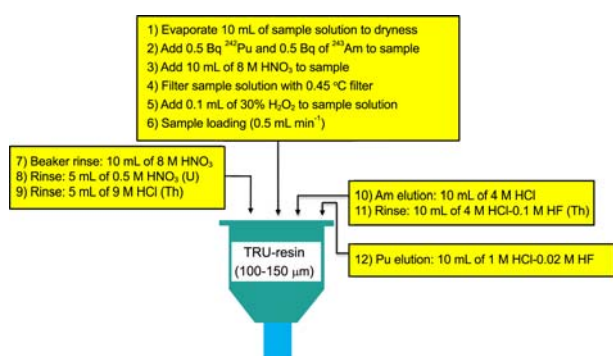
energy resolution for Pu, U, and Am isotopes through micro-coprecipitation methods were compared with those of electrodeposition methods. Also, the micro-coprecipitation method was applied to determine the activity concentration of Pu and Am in a radioactive waste sample.

### Experimental

**Comparison of an Electrodeposition and a Micro-Coprecipitation.** To investigate the electrodeposition and micro-coprecipitation yields,  $^{239}\text{Pu}$ ,  $^{233}\text{U}$ , and  $^{243}\text{Am}$  tracers (about 0.5 Bq) were added into the Teflon beaker. The solution was evaporated to dryness. The residue was dissolved in 1 mL of concentrated  $\text{HNO}_3$  and evaporated to a dryness. The samples were prepared using an electrodeposition method<sup>10</sup> and micro-coprecipitation method.<sup>9</sup>

**Pretreatment of the Samples.** A liquid sample (10 mL) leached from a radioactive waste sample with concentrated  $\text{HNO}_3$  was evaporated to dryness on a hot plate. To compensate for the chemical recovery,  $^{242}\text{Pu}$  (about 0.5 Bq) and  $^{243}\text{Am}$  (about 0.5 Bq), as yield tracers, were added into the sample. The residue was dissolved with 10 mL of 8 M  $\text{HNO}_3$ . The sample solution was filtered with a 0.45  $\mu\text{m}$  membrane filter. The oxidation state of Pu isotopes was adjusted to a tetravalent state with 0.1 mL of 30%  $\text{H}_2\text{O}_2$ .

**Radiochemical Separation of Pu and Am in the Radioactive Waste Sample.** The sample solution with an 8 M  $\text{HNO}_3$  medium was passed through a TRU Resin (Eichrom, 100-150  $\mu\text{m}$ ) column (inner diameter: 5 mm, resin bead length: 50 mm) pre-conditioned with 8 M  $\text{HNO}_3$  at a rate of 0.5 mL/minute. The column was then washed with 10 mL of 8 M  $\text{HNO}_3$  and 5 mL of 0.5 M  $\text{HNO}_3$  to remove the U isotopes and other hindrance elements. To remove Th, the column was washed with 5 mL of 9 M  $\text{HCl}$ . The Am was eluted with 10 mL of 4 M  $\text{HCl}$ . The column was rinsed with 10 mL of 4 M  $\text{HCl}$ /0.1 M  $\text{HF}$  to remove the residual Th. Finally, Pu was eluted with 10 mL of 1 M  $\text{HCl}$ /0.02 M  $\text{HF}$ .



**Figure 1.** Separation scheme of the Pu and Am isotopes in radioactive waste samples.

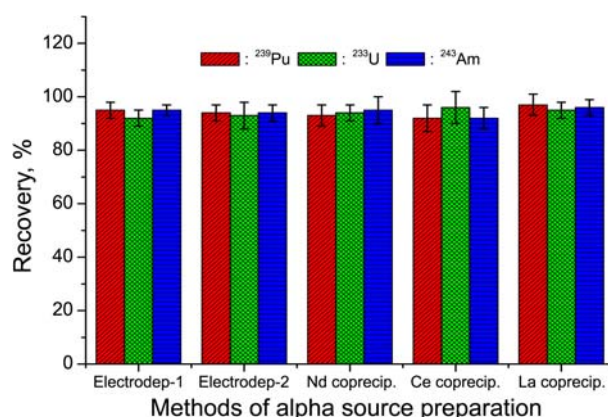
The sequential procedure for the Pu and Am isotopes using the TRU Resin (Eichrom) is shown in Figure 1.

**Source Preparation of Pu and Am Nuclides.** The purified Pu and Am fractions were evaporated to dryness on a hot plate. The residue was dissolved in 1 mL of concentrated HNO<sub>3</sub> and evaporated to dryness to destroy any organic substances. The purified isotopes were electrodeposited<sup>10</sup> or micro-coprecipitated with neodymium fluoride.<sup>9</sup>

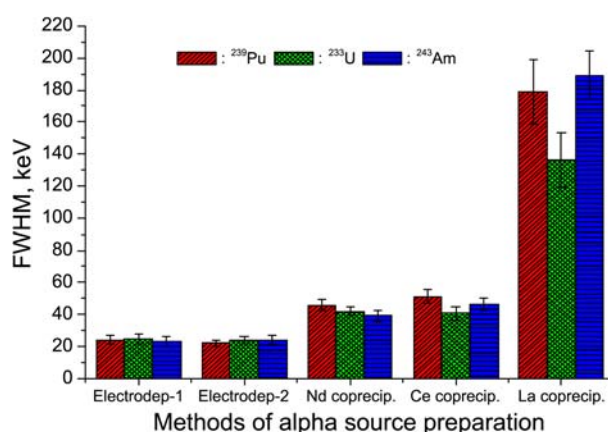
**Measurement of the Pu and Am/Cm Isotopes with an  $\alpha$  Spectrometer.** The activity concentrations of <sup>239,240</sup>Pu, <sup>238</sup>Pu, and <sup>241</sup>Am/<sup>244</sup>Cm were measured by an  $\alpha$  spectrometer. The measured activities for <sup>239,240</sup>Pu and <sup>238</sup>Pu were corrected for their chemical yields using the observed activities of <sup>242</sup>Pu. In addition, the chemical yields for <sup>241</sup>Am and <sup>244</sup>Cm were obtained by measuring the  $\alpha$  activity of <sup>243</sup>Am, as the chemical property of the Cm isotopes is similar to that of the Am isotopes.

## Results and Discussion

**Comparison of Electrodeposition and Micro-coprecipitation.** Among  $\alpha$  source preparation methods such as direct evaporation, electrodeposition, and micro-coprecipitation, the electrodeposition method and micro-coprecipitation method were compared in this study. As shown in Figure 2, the recoveries of <sup>239</sup>Pu, <sup>233</sup>U, and <sup>243</sup>Am isotopes using the electrodeposition methods were more than 95%. The recoveries of the actinides using the Electrodep-1 method as reported by Talvitie<sup>5</sup> were similar to those using the Electrodep-2 method reported by Lee and Lee.<sup>10</sup> However, for the preparation of the  $\alpha$  source, the Electrodep-2 method is more useful than the Electrodep-1 method as the Electrodep-1 method requires an accurate control of the pH and the electroplating solution contained the actinides should be free of Fe element and organic substances. In addition, the recoveries of <sup>239</sup>Pu, <sup>233</sup>U, and <sup>243</sup>Am isotopes using the micro-coprecipitation methods were similar to those using the electrodeposition method, as shown in Figure 2. However, the recovery of <sup>233</sup>U using the Nd-coprecipitation method was a little lower than that using the Ce-coprecipitation method. Therefore, the Ce-coprecipitation method is more reliable in the  $\alpha$  source preparation for uranium isotopes than the Nd-coprecipitation method.



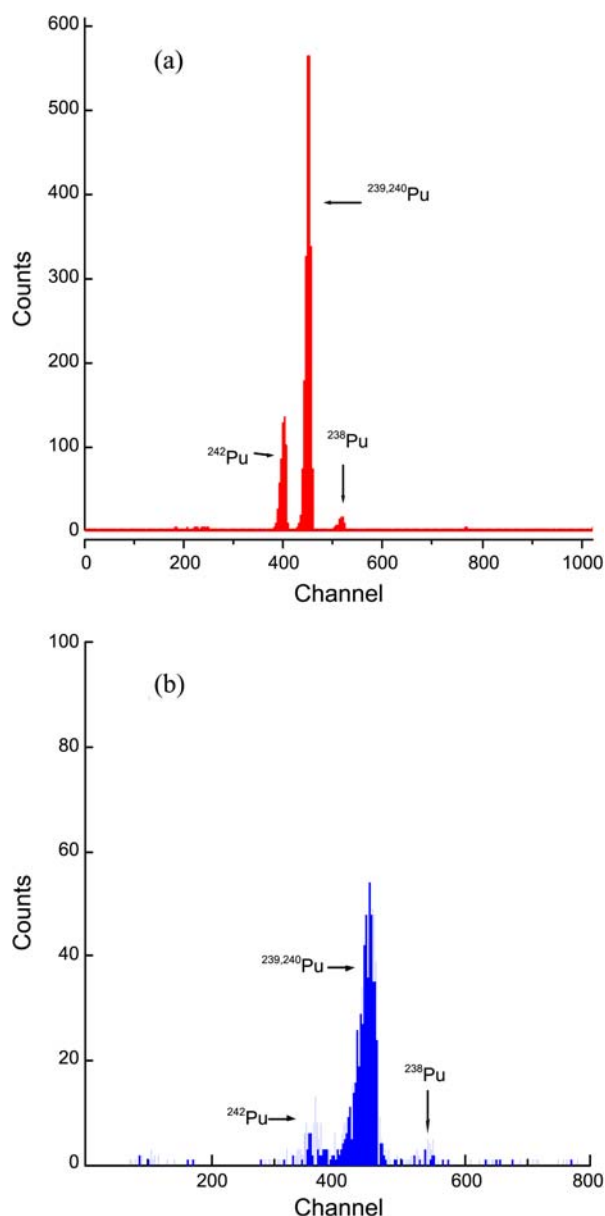
**Figure 2.** Recoveries for Pu, U, and Am isotopes with the electrodeposition and micro-coprecipitation methods.



**Figure 3.** Energy resolutions for Pu, U, and Am isotopes with the electrodeposition and micro-coprecipitation methods.

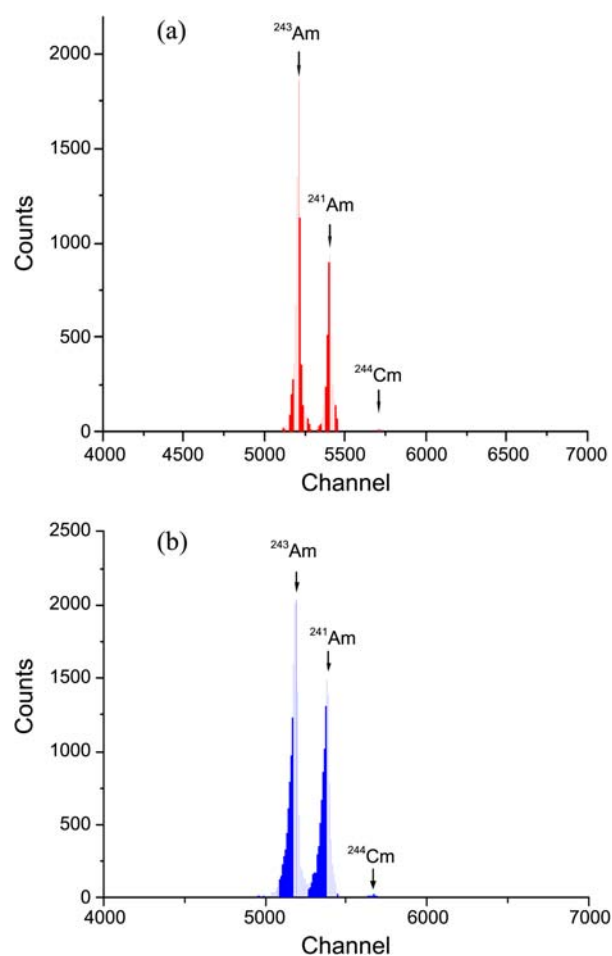
As shown in Figure 3, the energy resolution (FWHM) for <sup>239</sup>Pu, <sup>233</sup>U, and <sup>243</sup>Am isotopes using the electrodeposition methods was found to be below 30 keV, while the energy resolution for the <sup>239</sup>Pu, <sup>233</sup>U, and <sup>243</sup>Am isotopes using Nd coprecipitation or Ce coprecipitation was measured within the range of 40 keV to 50 keV. Although the energy resolution for <sup>239</sup>Pu, <sup>233</sup>U, and <sup>243</sup>Am isotopes with the micro-coprecipitation methods was worse than that with the electrodeposition, the peak of <sup>242</sup>Pu was distinguished from <sup>239,240</sup>Pu and <sup>238</sup>Pu with the Nd coprecipitation method, as shown in Figure 4(b). In addition, the peak of <sup>243</sup>Am was separated from <sup>241</sup>Am and <sup>244</sup>Cm in the  $\alpha$  spectrum using the Nd coprecipitation method, as shown in Figure 5(b).

An electrodeposition time is required more than 2 h, while less than 1 h is needed for the micro-coprecipitation method. Also, in the electrodeposition method, a lot of parameters such as the plating time, current, electrolyte concentration, pH, and volume of the electrolyte have influenced the electrodeposition of the actinides, so that it is necessary to optimize the electrodeposition conditions before electroplating the actinides. However, the procedure of the micro-coprecipitation with the Nd and Ce elements is so simple and robust that an unskilled person can easily perform the  $\alpha$  source preparation.



**Figure 4.** Typical  $\alpha$  spectra of Pu prepared by the electrodeposition method (a) and Nd coprecipitation method (b) in the radioactive waste samples (K1).

**Application to Radioactive Waste Samples.** The activity concentrations of  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  prepared through the

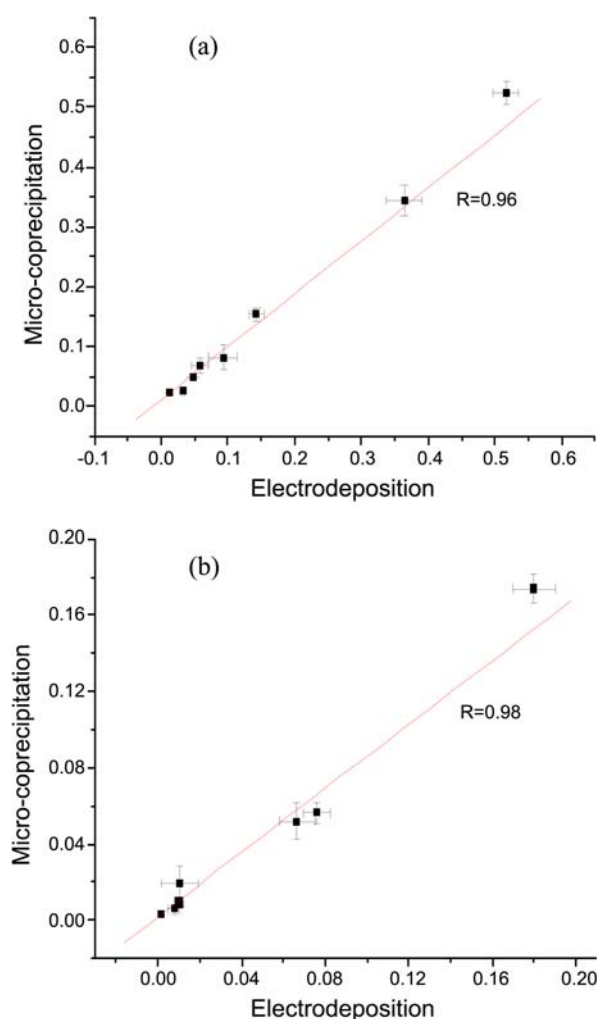


**Figure 5.** Typical  $\alpha$  spectra of Am prepared by the electrodeposition method (a) and Nd coprecipitation method (b) in the radioactive waste samples (K1).

electrodeposition and micro-coprecipitation methods for the radioactive waste samples are presented in Table 1. The activity concentrations of Pu and Am isotopes in the radioactive waste samples were variable, depending on the origin of the radioactive waste samples. Typical  $\alpha$  spectra of Pu and Am isotopes prepared by electrodeposition and micro-coprecipitation in the radioactive waste samples (K1) are shown in Figures 4 and 5, respectively. The peak of  $^{239,240}\text{Pu}$  in the radioactive waste samples was clearly separated from the peaks of other Pu isotopes, and thus the activity con-

**Table 1.** The activity concentrations of the Pu and Am isotopes in the radioactive waste samples

Sample	$^{239,240}\text{Pu}$ , $\text{Bq}\cdot\text{g}^{-1}$		$^{241}\text{Am}$ , $\text{Bq}\cdot\text{g}^{-1}$	
	Electrodeposition	Micro-coprecipitation	Electrodeposition	Micro-coprecipitation
K-1	$4.76\text{E-}02 \pm 5.18\text{E-}03$	$4.86\text{E-}02 \pm 5.20\text{E-}03$	$1.80\text{E-}01 \pm 4.80\text{E-}03$	$1.72\text{E-}01 \pm 4.72\text{E-}03$
K-2	$5.17\text{E-}01 \pm 1.92\text{E-}02$	$5.04\text{E-}01 \pm 1.93\text{E-}02$	$6.65\text{E-}01 \pm 1.21\text{E-}02$	$6.50\text{E-}01 \pm 1.17\text{E-}02$
Y-1	$1.43\text{E-}01 \pm 1.12\text{E-}02$	$1.54\text{E-}01 \pm 1.16\text{E-}02$	$1.07\text{E-}02 \pm 9.90\text{E-}04$	$0.87\text{E-}02 \pm 7.84\text{E-}04$
Y-2	$3.64\text{E-}01 \pm 2.59\text{E-}02$	$3.74\text{E-}01 \pm 2.63\text{E-}02$	$7.61\text{E-}02 \pm 6.15\text{E-}03$	$6.61\text{E-}02 \pm 5.64\text{E-}03$
W-1	$5.77\text{E-}02 \pm 1.22\text{E-}02$	$5.79\text{E-}02 \pm 1.24\text{E-}02$	$7.78\text{E-}03 \pm 2.98\text{E-}03$	$5.92\text{E-}03 \pm 2.76\text{E-}03$
W-2	$1.19\text{E-}02 \pm 7.26\text{E-}04$	$1.23\text{E-}02 \pm 7.29\text{E-}04$	$9.84\text{E-}03 \pm 5.05\text{E-}04$	$9.98\text{E-}03 \pm 5.21\text{E-}04$
U-1	$9.32\text{E-}02 \pm 2.08\text{E-}02$	$9.26\text{E-}02 \pm 2.00\text{E-}02$	$1.32\text{E-}03 \pm 1.99\text{E-}04$	$1.17\text{E-}03 \pm 1.82\text{E-}04$
U-2	$3.24\text{E-}02 \pm 1.81\text{E-}03$	$3.29\text{E-}02 \pm 1.83\text{E-}03$	$1.02\text{E-}02 \pm 9.08\text{E-}03$	$1.24\text{E-}02 \pm 9.24\text{E-}03$



**Figure 6.** The correlation of the  $^{239,240}\text{Pu}$  activity concentrations between the electrodeposition method and the micro-coprecipitation method (a) and  $^{241}\text{Am}$  activity concentrations between the electrodeposition method and the micro-coprecipitation method (b).

centration of  $^{239,240}\text{Pu}$  with the  $\alpha$  source preparation methods was easily determined in the radioactive waste sample. The peak of  $^{241}\text{Am}$  in the radioactive waste samples was clearly separated from the other peaks of the Am and Cm isotopes. In addition, as shown in Figure 5, the tracer level of  $^{244}\text{Cm}$  isotopes was measured with the Am isotopes as the chemical properties of Cm isotopes are similar to those of Am isotopes.

It is interesting to compare the correlation of activity concentrations for Pu and Am radionuclides prepared through the electrodeposition method and those prepared using the micro-coprecipitation method. As shown in Figure 6(a), the correlation of the  $^{239,240}\text{Pu}$  activity concentrations prepared through the electrodeposition method with those prepared using the micro-coprecipitation method is highly significant ( $R = 0.96$ ). Also, the correlation of the  $^{241}\text{Am}$  activity concentrations prepared using the electrodeposition method with those prepared through the micro-coprecipitation is highly significant ( $R = 0.98$ ), as shown in Figure 6(b).

## Conclusion

Micro-coprecipitation yields of  $^{239}\text{Pu}$ ,  $^{232}\text{U}$  and  $^{243}\text{Am}$  were over 95%. The recoveries of  $^{239}\text{Pu}$ ,  $^{232}\text{U}$  and  $^{243}\text{Am}$  using the electrodeposition methods were similar to those using the Nd, Ce, and La coprecipitation methods. Although the  $\alpha$ -peak resolution for  $^{239}\text{Pu}$ ,  $^{232}\text{U}$ , and  $^{243}\text{Am}$  using micro-coprecipitation methods was worse than that using the electrodeposition methods, the peaks of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  in the radioactive waste sample were easily discriminated from Pu and Am isotopes, respectively.

The activity concentrations of the Pu and Am isotopes in the radioactive waste samples were variable, depending on the origin of the radioactive waste samples. The correlation of the  $^{239,240}\text{Pu}$  activity concentrations prepared using the electrodeposition method with those prepared through the micro-coprecipitation method is highly significant. In addition, the correlation of the  $^{241}\text{Am}$  activity concentrations prepared using the electrodeposition method with those prepared through the micro-coprecipitation is highly significant.

## References

1. Maxmann, S. H. *Nucl. Instrum. Meth. Phys. Res.* **1967**, *50*, 53.
2. Aggarwal, S. K.; Chourasiya, G.; Duggal, R. K.; Singh, C. P.; Rawat, A. S.; Jain, H. C. *Nucl. Instrum. Meth. Phys. Res.* **1984**, *223*, 266.
3. Mitchell, R. F. *Anal. Chem.* **1960**, *32*, 326.
4. Handley, T. H.; Cooper, J. H. *Anal. Chem.* **1969**, *41*, 381.
5. Talvitie, N. A. *Anal. Chem.* **1972**, *44*, 280.
6. Puphal, K. W.; Olsen, D. R. *Anal. Chem.* **1972**, *44*, 284.
7. Dutton, J. W. R.; Lovett, M. B. *Nature* **1977**, *267*, 37.
8. Joshi, S. R. *J. Radioanal. Nucl. Chem. Article* **1985**, *90*, 409.
9. Hindman, F. D. *Anal. Chem.* **1986**, *58*, 1238.
10. Lee, M. H.; Lee, C. W. *Instrum. Meth. Phys. Res.* **2000**, *447*, 593.