

Application Note

Alpha/Beta

ABA-004

Quantitation of Transuranium Elements in High Activity Waste (HAW) Using Alpha/Beta Liquid Scintillation Counting and Extractive Scintillators

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Abstract

Determining the content of transuranium elements, Americium (Am), Plutonium (Pu), Neptunium (Np), and Curium (Cm) in the nuclear fuel cycle and in nuclear radioactive waste is important for both economic and human toxicity reasons. The major concern is processing the sample rapidly and quantitating these transuranium alpha emitters in the presence of 10^5 fold excess of beta emitting contaminants. The combination of alpha/beta discrimination with a Packard Tri-Carb[®] Liquid Scintillation Analyzer employing the Pulse Decay Analysis (PDA) technique and extractive scintillators allows the quantitation of a small concentration of alpha radionuclides in the presence of a large excess of beta emitters without an excessive amount of sample preparation. Both the pulse decay analysis method and extractive scintillator performance are discussed.

Introduction

The quantitation of transuranium elements is important in radioactive waste management. The content of transuranium elements in high level radioactive waste (HAW) has a direct influence on the selection of the proper disposal method. HAW is characterized by its high beta and gamma radioactivity, complex chemical constituents (high salt concentration), and low concentration of transuranium elements. The total activity of this waste is approximately 3.7×10^{13} Bq/L with a concentration of 250 g/L. The concentration of the transuranium elements is only 0.07-0.35 g/L or a 3000 fold excess of beta to alpha particles.

Traditionally, alpha particles have been quantitated with ZnS(Ag) scintillation detectors and proportional counters. Alpha spectroscopy has been performed by surface-barrier detectors and grid ionization chambers. All of these methods measure solid samples on planchets. Sample preparation is a difficult problem because of the self-absorption of the alpha particle by the sample matrix or by the alpha particle itself. Several complicated and long procedures, including precipitation, filtration, chromatography, extraction, and stripping are required to separate the alpha emitting nuclides of interest from the sample matrix (HAW).¹ In addition, evaporation, electrodeposition, and vacuum sublimation are necessary to prepare the samples for accurate analysis by one of these methods.¹ These combined procedures are labor intensive and time consuming. They result in low counting efficiencies, low recoveries, and poor reproducibility. To reduce the amount of time and effort required to process and quantitate the alpha component in HAW, liquid scintillation counting employing Pulse Decay Analysis (PDA) and the use of extractive scintillators was investigated.

Experimental

The major radioactive components of HAW are shown in Table 1. All activities are relative to the activity of Neptunium. The total relative beta activity is 1.05×10^6 , and the transuranium activity level is from 1 to 400. To evaluate both the pulse decay analysis and extractive scintillator techniques, both synthetic and typical high activity waste (HAW) samples were examined and the radioactivity of the major components determined.



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Nuclide	Activity	Nuclide	Activity*
Np-237	1	Ru-106	2 x 10 ⁴
Pu-238	150-200	Rh-106	2 x 10 ⁴
Am-241	300-400	Ce-144	1 x 10 ⁴
Cm-244	5000	Pr-144	1 x 10 ⁴
Sr-90	2 x 10 ⁵	Pm-147	6 x 10 ⁴
Y-90	2 x 10 ⁵	Eu-155	1 x 10 ⁴
Cs-137	2.6 x 10 ⁵		
Ba-137m	2.6 x 10 ⁵	Sum F.P.	10.5 x 10 ⁵

*Relative Activity = activity of the nuclide/activity of ²³⁷Np.

Table 1.

Relative Radioactivities of Transuranium Elements and Some F.P.S. in HAW.

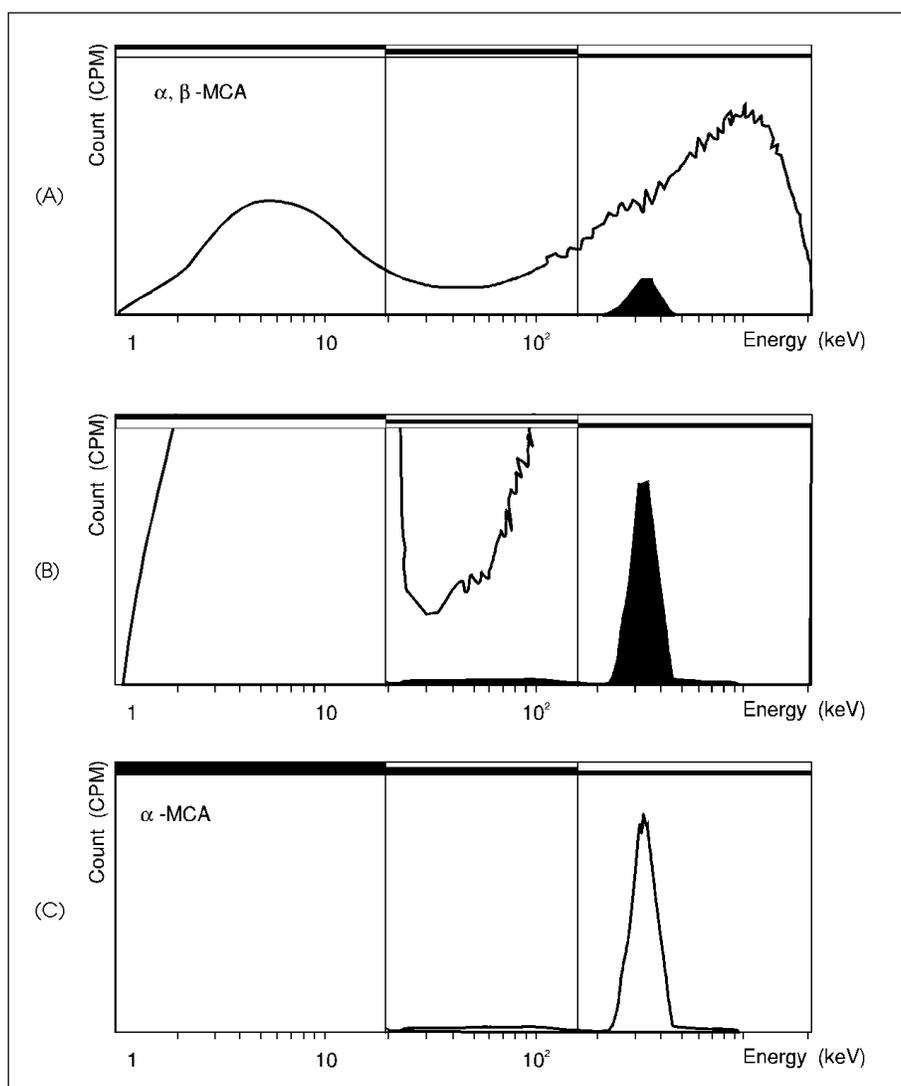


Figure 1.

Spectra of ¹⁰⁶Ru/¹⁰⁶Rh and ²⁴¹Am in a Mixed Sample
 10 ml 5 g/l PPO-0.5 g/l M₂POPOP-50 g/l Naphthalene-Toluene-Triton.

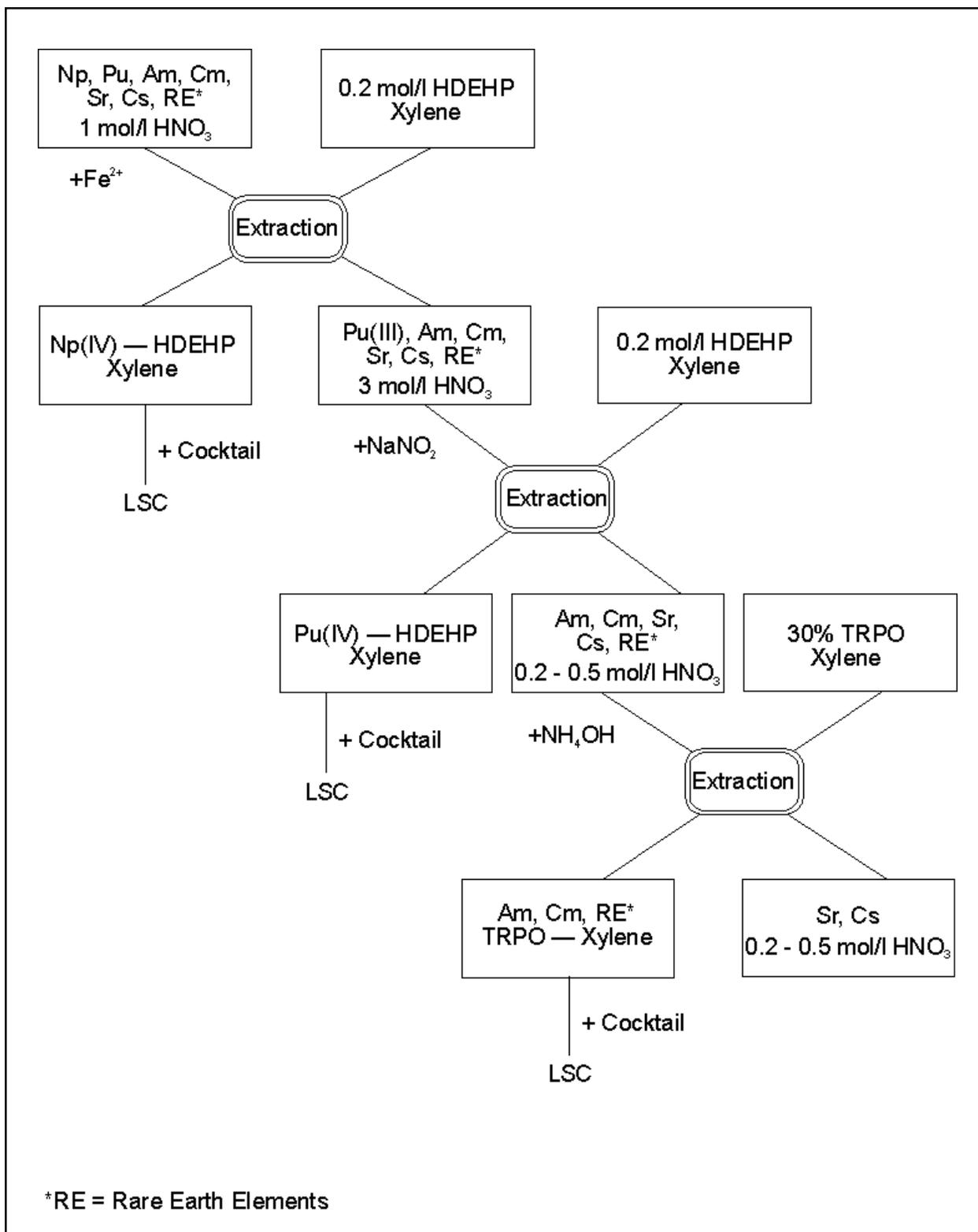


Figure 2.
Extraction Separation Procedure of Np, Pu, and Am from HAW

To quantitate the alpha emitters by liquid scintillation counting, a Packard Tri-Carb Alpha/Beta Liquid Scintillation Analyzer with special Pulse Decay Discrimination (PDD) circuitry is used. This circuit discriminates alpha particles from beta particles based on their pulse decay time and is the basis of the Pulse Decay Analysis (PDA) technique. Alpha particles produce pulses in a liquid scintillator which take several hundred nanoseconds to decay compared to beta pulses which take a few nanoseconds to decay. Once the PDD circuit is optimized for the cocktail, pulses which have decay times longer than the PDD setting are classified as alpha and placed in an alpha MCA and those that have shorter decay times are classified as beta and placed in a beta MCA.

Alpha/Beta Discrimination Without Extractive Scintillators

A mixed synthetic alpha/beta sample of HAW that contained a 150 fold excess of ^{106}Ru (beta emitter), and its daughter, ^{106}Rh (a mixed beta/gamma emitter), both commonly found in spent nuclear fuel, was mixed with ^{241}Am . The sample was counted directly in the liquid scintillation counter with pulse decay discrimination, without the use of extractants. The spectra in Figure 1A show the relative contributions to the alpha and beta MCAs. The y-axis is re-scaled in Figure 1B to better show the alpha peak in the alpha MCA relative to the beta activity in the beta MCA. Figure 1C is the alpha peak in the alpha MCA. It is obvious that with PDD, the contribution of beta activity to the alpha MCA is quite small even in spite of a large beta excess in the sample. In fact, the PDA method is effective for direct measurement of samples with a ratio of 250/1 beta to alpha.

As can be seen in Table 1, it is common for samples of HAW to have a beta/alpha ratio of up to $10^5/1$. To quantitate the alphas in these samples, a combination of PDA and extractive scintillators must be used. Extractive scintillators selectively isolate certain radionuclides from samples prepared at various acid concentrations. A scheme using extractive scintillators to isolate Neptunium (Np), Plutonium (Pu), and Americium (Am) is shown in Figure 2.¹

Alpha/Beta Discrimination With Extractive Scintillators

To investigate the use of both alpha/beta LSC and extractive scintillators, the activity of Pu in a HAW sample was determined. The total beta activity of the sample is 8×10^4 higher than the alpha activity. If the sample is analyzed directly with PDA, without any extractive processes, the spectral data shows that the beta MCA contains a large number of counts across the entire energy range (Figure 3). The alpha MCA shows only 0.3% of the total counts in this MCA, but no peak is apparent for the Plutonium. If the HAW sample is extracted with di-(2-ethylhexyl)-phosphate (HDEHP) in toluene cocktail (see Figure 2) and the organic phase is counted with Pulse Decay Analysis, the alpha component is easily isolated. Figure 4 shows the spectra of both the alpha and the beta components in their respective MCAs. Of the 180,000 CPM initially present in the HAW sample only 2,600 CPM are found in the extractant. Thus, extraction eliminates greater than 98% of the beta activity and the remaining 2% of beta can be discriminated from the alpha activity by PDA. The ^{239}Pu can easily be recognized as a peak in the alpha MCA (Figure 4) and quantitated using this combined technique.

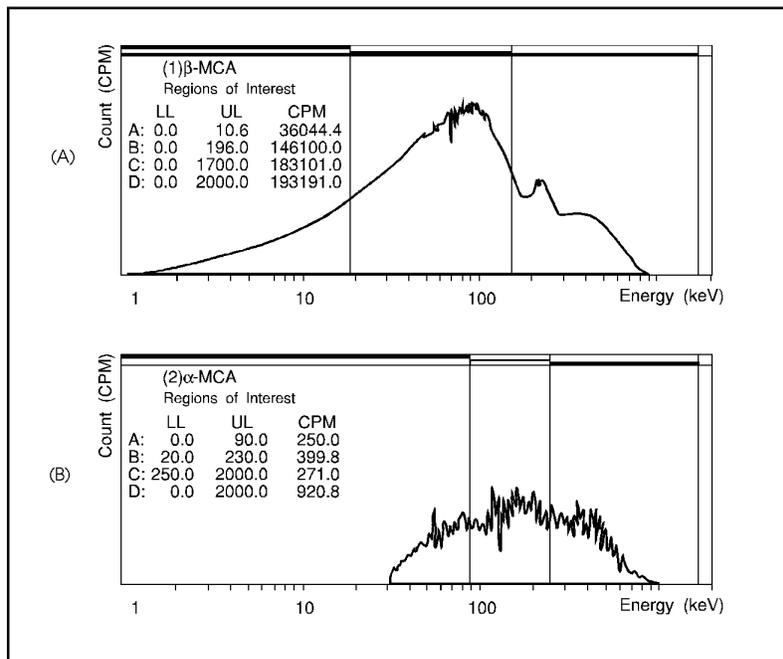


Figure 3.

Spectra of HAW without Separation; 10 ml 5 g/l PPO-0.5 g/l M_2POPOP -50 g/l Naphthalene-Toluene-Triton.

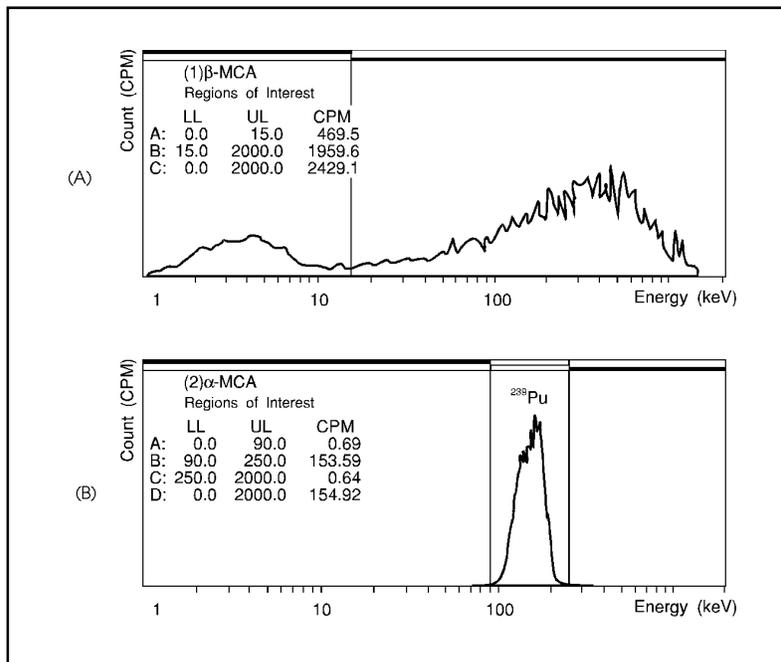


Figure 4.

Spectra of ^{239}Pu in HDEHP - Organic Solution After Extraction from HAW;
 10 ml 5 g/l PPO-0.5 g/l M_2POPOP -50 g/l Naphthalene-Toluene-Triton.

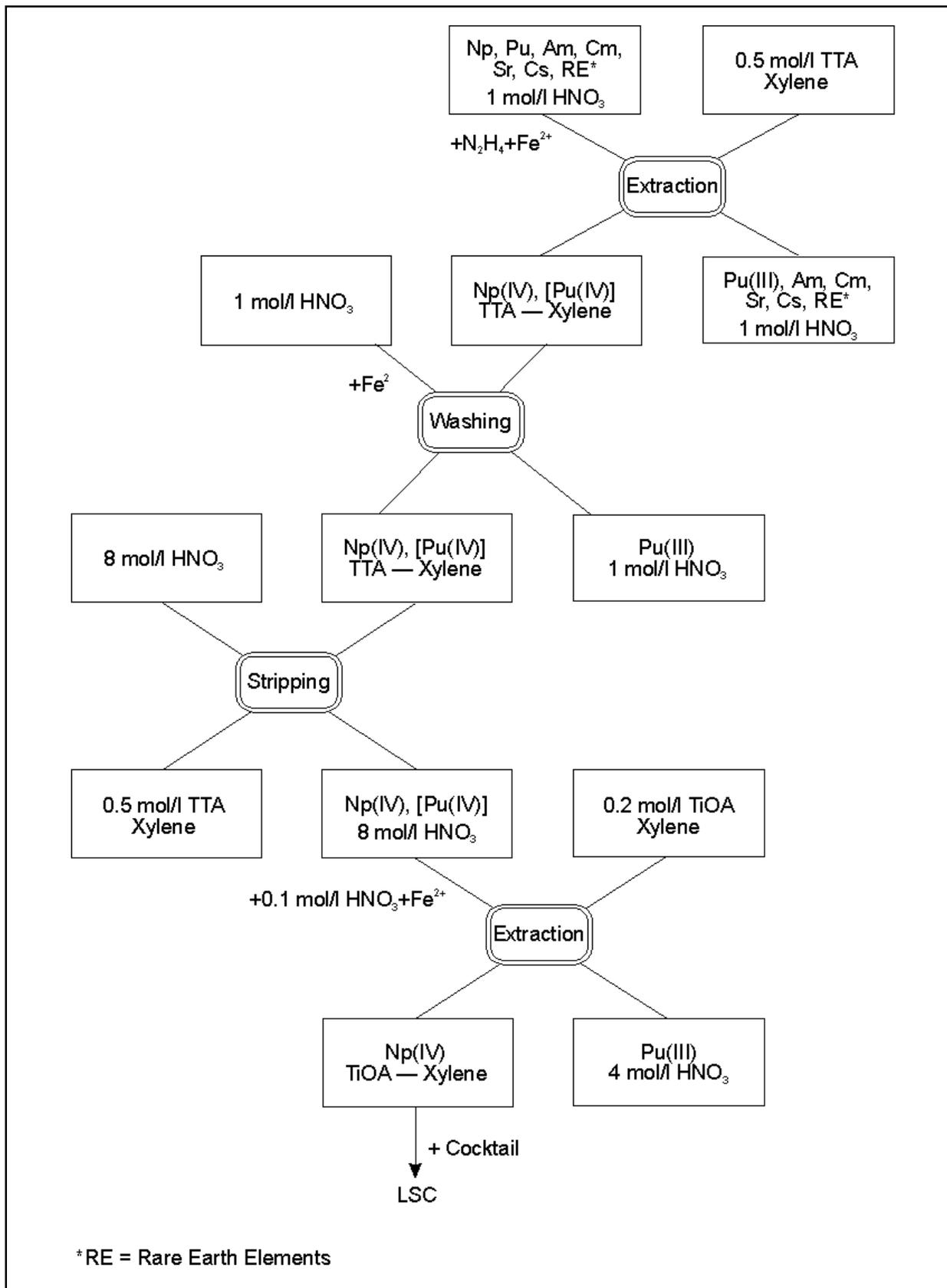


Figure 5.
Extraction Separation Procedure of Np from HAW.

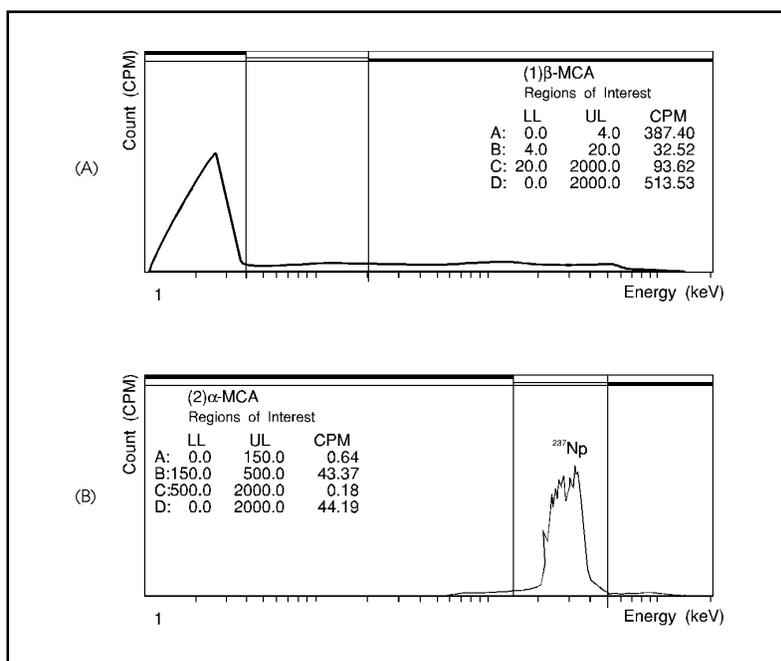


Figure 6.

Spectra of ^{237}Np in TiOA - Organic Solution After Extraction from HAW;
 10 ml 5 g/l PPO-0.5 g/l M_2POPOP -50 g/l Naphthalene-Toluene-Triton.

Conclusion

The quantitation of high level radioactivity waste from the nuclear fuel cycle or radioactive waste management is difficult, laborious, and time consuming because it is necessary to eliminate the interference of the large excess of beta and gamma radionuclides before measuring the alpha activity. To quantitate the alpha particles, several separations and sample preparation steps are required prior to analysis with alpha spectrometry. By using the liquid scintillation pulse decay analysis method, alphas can be accurately quantitated despite a 250 fold excess of betas without any extraction methods. Ratios in excess of 250/1 beta/alpha can be quantitated by combining the PDA technique with extractive scintillators. Together, these two techniques can be used effectively to quantitate alphas in the nuclear fuel cycle and nuclear waste samples in a simple, rapid, and accurate manner.

Reference

1. Yang, Dazhu, " Study on Determination of Np, Pu, and Am with Extraction-Liquid Scintillation Counting and Its Application to Assay of Transuranium Elements in High-Level Radioactive Waste, Ph.D. thesis, Tsinghua University, Beijing, P.R. China, 1990.