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Determination of plutonium isotopes in spent nuclear fuel using thermal ionization mass spectrometry (TI-MS) and alpha spectrometry

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Introduction

- The purpose of this paper is the separation of plutonium out of spent nuclear fuel solution and its qualitative and quantitative determination.
- Alpha emitters such as uranium, plutonium, can be measured only by destructive methods.
- Mass spectrometry is one of the techniques used for destructive characterization of spent nuclear fuel.
- Alpha spectrometry can also be used for destructive characterization of spent nuclear fuel, but it is difficult to determine the abundance of ^{239}Pu and ^{240}Pu isotopes in samples because the alpha emissions for the two isotopes are overlapped
- Several radiochemical procedures must be performed before spectrometry measurements.

Spent nuclear fuel dissolution and dilution of the concentrate solution

- The fragment of the spent nuclear fuel element was cut according to a sampling plan.
- Fuel dissolution is performed into a hot cell (Figure 1)
- The cut section of the fuel rod was dissolved in a stainless steel vessel with lid (Figure 2)
- During the process of chemical dissolution and dilution, we obtained a ***final solution*** where the uranium concentration is around 1mg/ml

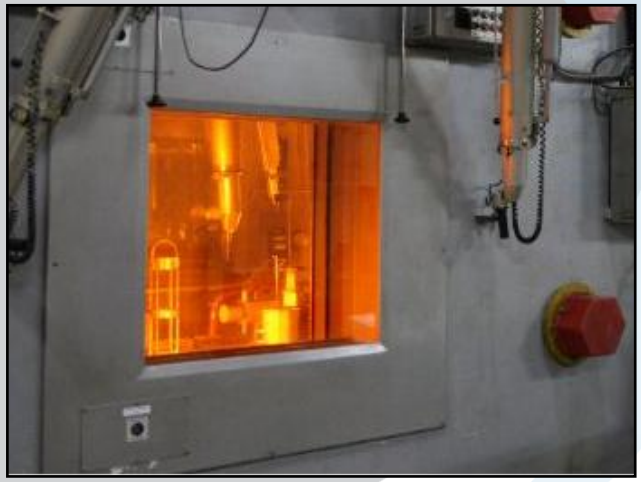

Fig. 1. Hot cell

Fig. 2. Dissolution device

Radiochemical processing of the samples

We processed two samples, first for alpha spectrometry and second for TI-MS measurements as follows:

• The sample for alpha spectrometry consisted of an aliqout sampled from the ***final solution*** and 5 ml of ^{242}Pu spike solution were added (^{242}Pu spike solution was prepared in 3M HNO_3 at around 22,6 mBq/ml). This sample was named "*Plutonium+^{242}Pu spike*"

• The sample for TI-MS consisted only from an aliqout sampled from the ***final solution*** and named just "*Plutonium*".

•The two samples were then subjected to repeated chemical purifications which are shown in figure 3.

Radiochemical processing of the samples

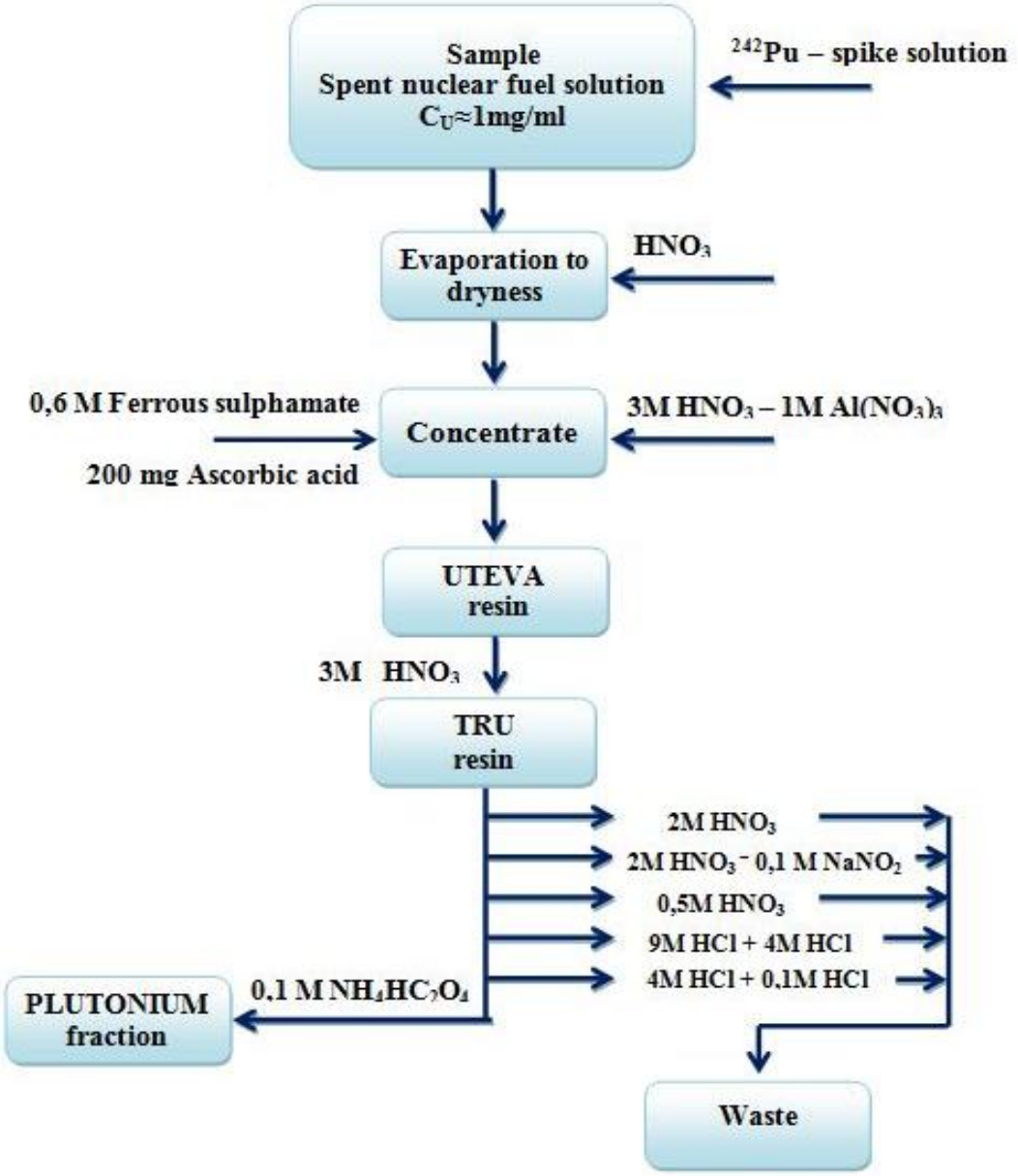

Fig. 3. Samples preparation procedure for plutonium analysis in spent nuclear fuel solution

Samples preparation for measurements

- The alpha source was prepared using the technique of co-precipitation using cerium nitrate.
- The precipitate was removed by filtration and washed using a Gelman system equipped with vacuum pump (Figure 4)
- The filter with the precipitate was then dried under an UV lamp (Figure 5) and fastened on a stainless steel disk (Figure 6)
- **For mass spectrometry measurements,** the plutonium fraction was dissolved in 50 µL 0,1M nitric acid and 1µL from it was deposited on the rhenium filament. (Figure 7)

Fig. 4. Gelman filtration system

Fig. 5. Filter's dry under the UV lamp

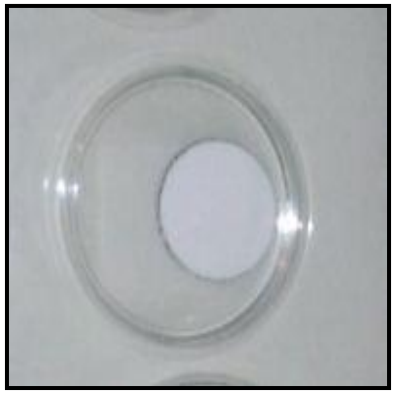

Fig. 6.The filter fastened on a stainless steel disk (Alpha source)

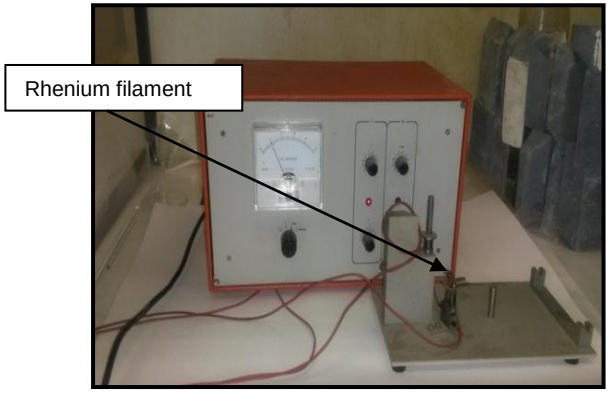

Fig. 7. Device for heating the filament

Alpha spectrometry results

The obtained alpha source is measured using the ORTEC 576 alpha spectrometry system, with two individual measurement chambers (Figure 8)

Fig. 8. ORTEC 576 alpha spectrometry system

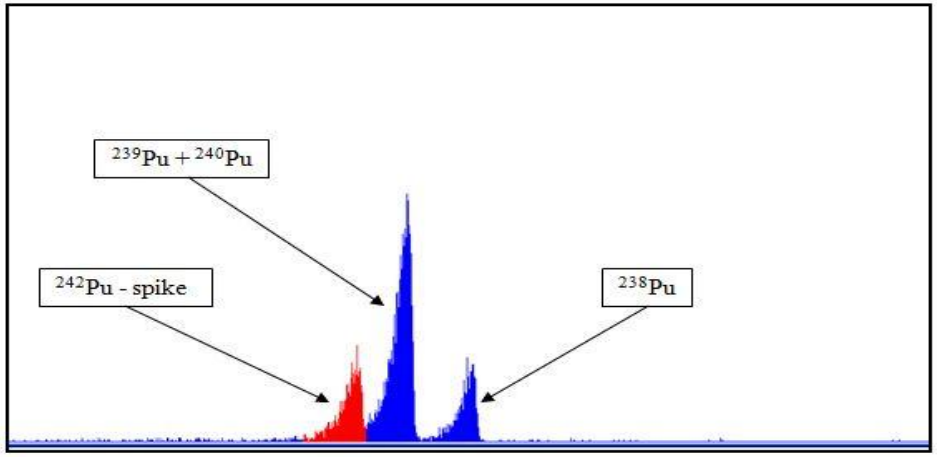

Fig.9. Alpha spectrum of the Plutonium+^{242}Pu spike source

Table 1. Activity of plutonium source measured using alpha spectrometry

Isotope	**Net aria (Counts)**	**Time (s)**	Measured source activity [mBq]	Activity of the added spike [mBq]	Yield [%]
^{242}Pu	1 804	80 000	89,8	113	**79,5**
^{239}Pu + ^{240}Pu	5 554		277,7		
^{238}Pu	1 504		75,3		

Thermal ionization mass spectrometry results

The rhenium filament which contains the evaporated plutonium sample is measured using the FINNIGAN MAT 261 mass spectrometer.

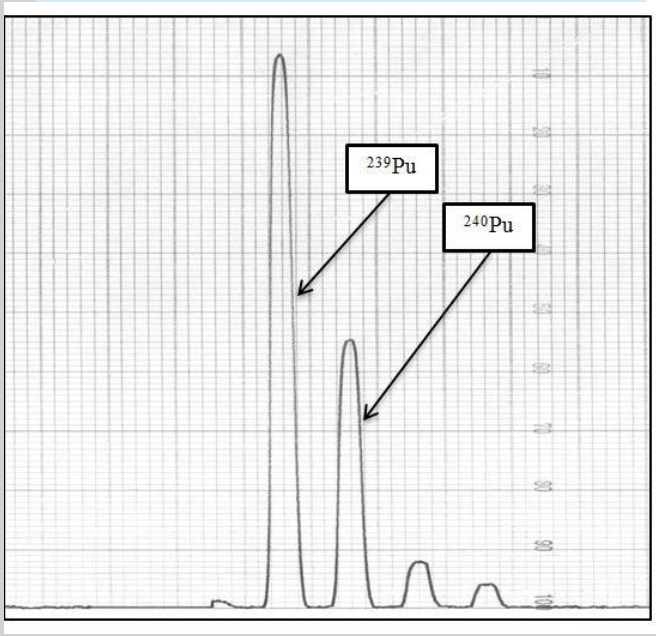

Fig.10. The spectrum of Plutonium sample

Table 2. The results using TI-MS measurements

Isotope ratio			
$R_{238/239}$	**$R_{240/239}$**	$R_{241/239}$	$R_{242/239}$
0.021518	**0.481854**	0.081469	0.038647

Concentrations – mass percent				
C-238	C-239	C-240	C-241	C-242
1.317347%	61.47853%	29.74773%	5.050569%	2.405827%

The ^{240} / ^{239}Pu isotopic ratio determined using TI-MS in spent nuclear fuel is **0.481854**. To calculate the ^{240}Pu and ^{239}Pu amounts we used the equation no. 1

$$N = \frac{m}{A} \times N_A \leftrightarrow m = \frac{N \times A}{N_A} \quad \text{(equation no. 1)}$$

Table 3. *$^{240/239}$Pu mass ratio*

Isotope	Amount (g)	$^{240/239}$Pu mass ratio – calculated	$^{240/239}$Pu mass ratio – by TI-MS measurements
^{239}Pu	$4{,}3432\times10^{-11}$	0,48386	0,48387
^{240}Pu	$2{,}1015\times10^{-11}$		

The dependence between $^{240/239}$Pu mass ratio and the fuel burn up using ORIGEN code

Using ORIGEN code we estimated the dependence between $^{240/239}$Pu mass ratio and the fuel burn up. As can be seen mass ratio determined by us corresponds to a fuel burn up around 8,000 MWd/tU (this being the burn up of the fuel studied in present paper).

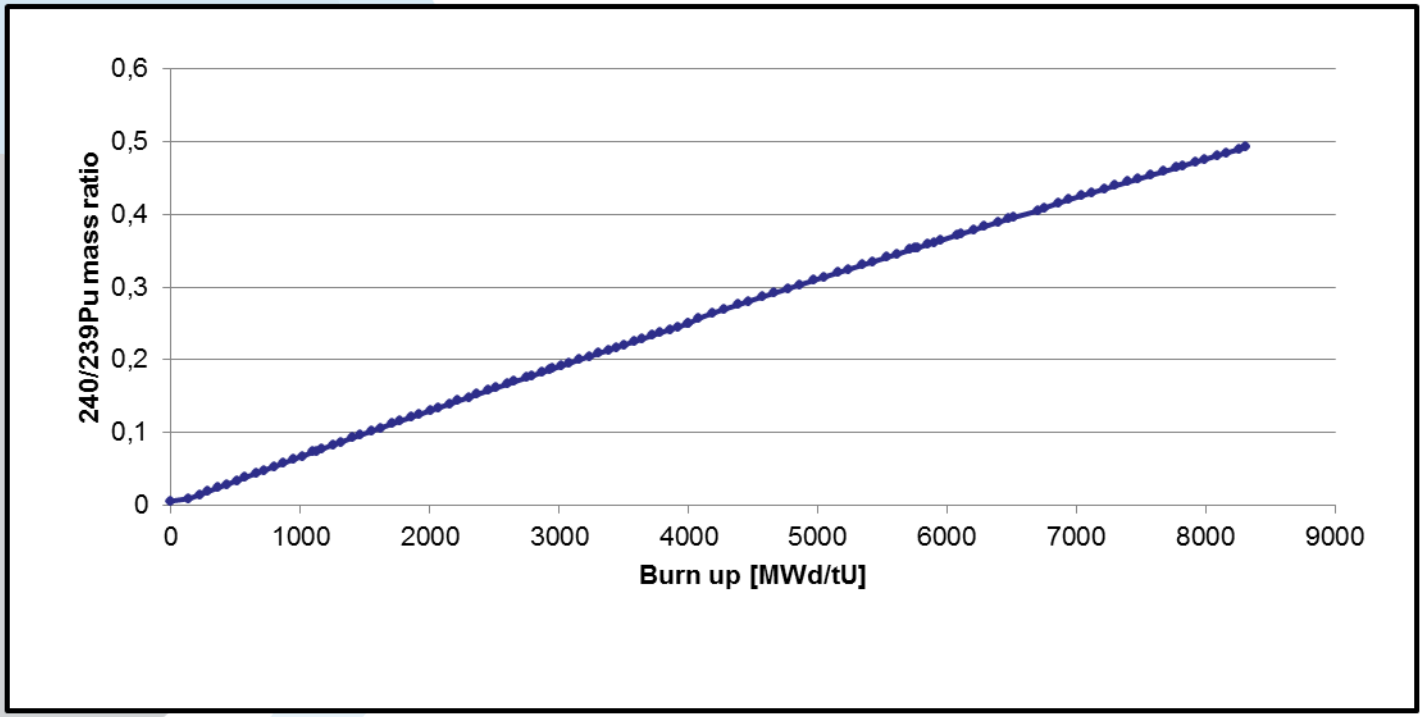

Fig.11. The dependence between $^{240/239}$Pu mass ratio and fuel burn up

Conclusions

- In this paper we estimated the $^{240/239}$Pu mass ratio in spent nuclear fuel using the techniques from our lab – alpha spectrometry and TI-MS
- The alpha source was prepared using the technique of co-precipitation as CeF_3.
- The chemical recovery for the ^{242}Pu spike was around 80%, but we cannot precise delimitate using alpha spectrometry ^{239}Pu isotope by ^{240}Pu isotope because alpha emissions for the two isotopes are overlapped
- Using TI-MS we could individually identify ^{239}Pu and ^{240}Pu and we estimated the isotope ratio and the mass ratio of plutonium isotopes.
- The $^{240/239}$Pu mass ratio estimated using TI-MS in spent nuclear fuel is **0,483870** - this value beeing the same with the calculated one (**0,483860**)
- Using ORIGEN code we estimated the dependence between $^{240/239}$Pu mass ratio and fuel burn. The $^{240/239}$Pu mass ratio determined by us corresponds to a fuel burn up around 8,000 MWd/tU (this being the burn up of the fuel studied in present paper).

Thank you for your attention!!!