

**PWR**

**Determination of Plutonium in PWR Spent Fuels  
by Controlled-Potential Coulometry**

150

가

(AG MP-1)

가

가

0.25 3.0 mg

99.36 %

가

250 450  $\mu\text{g}$

(

, RSD) 0.38 %

**Abstract**

Separation and coulometric titration method were applied for the determination of plutonium content in samples of PWR spent fuel. Plutonium was separated on an anion exchange(AG MP-1) column and determined by the controlled-potential coulometric titration. In this study, we discussed some experimental conditions related to the separation and determination of plutonium in PWR spent fuel samples. Average accuracy(recovery of plutonium) for the determination of 0.25 3.0 mg plutonium standard was 99.36 %. Average precision(relative standard deviation, RSD) for the determination of 250 450  $\mu\text{g}$  plutonium in PWR spent fuel samples was 0.38 %.

1.

[1], [2] [3-7]

[5-7].

[8].

As,

Cd, Cr, Co, Cu, Au, I, Fe, Pb, Mg, Mo, Ni, Pd, Ru, Ag, U Zn

PWR

[9].

glove box

8 M HNO<sub>3</sub>

Pu(IV)

Pd, In, Pt,

Np(IV), Au Th 가

8

M HNO<sub>3</sub>

가

H<sub>2</sub>O<sub>2</sub>

H<sub>2</sub>O<sub>2</sub>가 Pu(VI), Pu(V) Pu(IV) Pu(III)

가

[10].

가 PWR (Kori-1)

## 2.

### 2.1

PWR,  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ,  $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ , Sulfamic acid,  $\text{NH}_2\text{OH} \cdot \text{HCl}$ ,  $\text{H}_2\text{O}_2$ ,  $\text{HCl}$ ,  $\text{HNO}_3$  Aldrich Merck G.R A.C.S. BIO-RAD AG MP-1 (ORNL,  $\text{PuO}_2$ ,  $^{242}\text{Pu} = 99.93\%$  NBL CRM 122,  $\text{PuO}_2$  powder,  $\text{Pu} = 87.790\%$ ) 1.5 M  $\text{HNO}_3$  - 0.01 M HF 1 M  $\text{HCl}$  1 L  $\text{AlCl}_3$ , Fe (5 mg Fe/mL) 1.5 M sulfamic acid 8 mL, 1 mL 8 mL

가

### 2.2

Potentiostat/ Galvanostat(EG&G PARC 263A), Digital coulometer(Scribner 279A) Titration cell system(EG&G PARC 377A)

(EG&G PARC E0002)

(saturated calomel electrode, SCE)

glove box titration cell glove box

가

### 2.3

2.3.1 : PWR Hot cell

$\text{HNO}_3(1+1)$  가

2.3.2 : 8 M  $\text{HNO}_3$

, AG MP-1(200 400 mesh)

8 M  $\text{HNO}_3$  0.02 M  $\text{NH}_2\text{OH} \cdot \text{HCl}$  - 0.5 M  $\text{HNO}_3$

2.3.3 : Hot plate  
 0.5 1.0 mL가 25 mL  
 15 purging + 250 mV(vs. SCE) + 900  
 mV(vs. SCE) 30  $\mu$ A, +560 mV(vs. SCE) 20  $\mu$ A 가  
 3 blank

### 3.

#### 3.1

가 AG MP-1  
 , 8 M HNO<sub>3</sub> Pu(IV) 8 M HNO<sub>3</sub>  
 0.02 M NH<sub>2</sub>OH · HCl - 0.5 M HNO<sub>3</sub>  
 Fig. 1  
 100 mg 1.0 mg <sup>242</sup>Pu (ORNL ) 3 mL  
 8.0 M HNO<sub>3</sub> 0.1 mL 30 % 가 4 200 400  
 mesh AG MP-1 0.7 cm (i.d.) × 5.5 cm (h) , 8.0  
 M HNO<sub>3</sub> 0.02 M NH<sub>2</sub>OH · HCl - 0.5 M HNO<sub>3</sub>  
 1.0 mL 0.1 mL planchet  
 (gross  $\alpha$ -activity) . Table 1

1 6

가

가

#### 3.2

25 mL 6 9  
 mC  
 Pu(III) ⇌ Pu(IV) couple + 250

mV Pu(IV) Fe(III)가 Pu(III) Fe(II) , + 900 mV Pu(III) Fe(II)가  
 Pu(IV) Fe(III) , + 560 mV Pu(IV) Pu(III) .  
 가  
 Nd(III),  
 Zr(IV), Mo(VI), Ce(III), Ba(II), Pd(II), Sn(IV) Fe(III) . 25  
 mL 0.2 4.0 mg 가

, 가  
 가 가  
 , + 560 mV .  
 가 , 25 mL  
 (0.025 mg)

10, 50, 100 mg ,  
 0.02 M NH<sub>2</sub>O  
 H · HCl - 0.5 M HNO<sub>3</sub> NH<sub>2</sub>OH · HCl

7.2 × 10<sup>-3</sup> M NH<sub>2</sub>OH · HCl가 가 NH<sub>2</sub>OH  
 (ORNL )

, 120 450 μg

Table 2

(120 μg )가  
 가  
 = 1.0002, = 1.0378 μg, (r) = 0.9994 .

Table 2 , PWR (250 450  $\mu\text{g}$ )

(NBL )

Table 3

0.25 3.0 mg

2 4

0.25 3.0 mg

99.36

%

3.2

PWR

250 450  $\mu\text{g}$

PWR

Table 4

PWR

2 6

PWR

( , RSD) 6

0.18 0.53 %

mg

가

0.05 0.1%

250 450  $\mu\text{g}$  PWR

Table 4

가

가

가

가

4.

가

가

, 가

0.25 3.0 mg

99.36

% 가 250 450  $\mu\text{g}$   
( , RSD) 0.38 % .

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**Table 1.**

<b>Time, day</b>	<b>[Pu], <math>\mu\text{g/g}</math></b>
<b>1</b>	<b>84.8, 83.8</b>
<b>2</b>	<b>84.4, 83.2</b>
<b>4</b>	<b>84.6, 84.1</b>
<b>6</b>	<b>84.3, 83.5</b>
<b>uncontrolled</b>	<b>76.7, 75.2</b>
	<b>76.6, 75.8</b>

**Table 2. ORNL**

<b>Pu taken, mg</b>	<b>Pu found, mg</b>	<b>Recovery, %</b>	<b><math>\bar{X} \pm \text{sd}</math>, rsd</b>
<b>0.1179</b>	<b>0.1244</b>	<b>105.5</b>	<b><math>105.1 \pm 0.5</math>, 0.5 (%)</b>
<b>0.1189</b>	<b>0.1246</b>	<b>104.8</b>	
<b>0.2390</b>	<b>0.2347</b>	<b>98.2</b>	<b><math>98.3 \pm 0.3</math>, 0.4 (%)</b>
<b>0.2382</b>	<b>0.2335</b>	<b>98.0</b>	
<b>0.2362</b>	<b>0.2332</b>	<b>98.7</b>	
<b>0.4716</b>	<b>0.4772</b>	<b>101.0</b>	<b><math>100.4 \pm 0.9</math>, 0.9 (%)</b>
<b>0.4757</b>	<b>0.4745</b>	<b>99.7</b>	



Table 3. NBL CRM- 122

Pu taken, mg	Pu found, mg	Recovery, %	$\bar{X} \pm \text{sd}$ , rsd
0.2308	0.2294	99.41	99.32 ± 0.13, 0.13 (%)
0.2307	0.2294	99.43	
0.2291	0.2272	99.15	
0.2314	0.2298	99.30	
0.4795	0.4779	99.67	99.60 ± 0.06, 0.06 (%)
0.4828	0.4806	99.53	
0.4839	0.4819	99.59	
0.4809	0.4791	99.61	
0.9688	0.9622	99.32	99.29 ± 0.08, 0.08 (%)
0.9683	0.9611	99.25	
0.9701	0.9608	99.38	
0.9670	0.9592	99.20	
3.0221	2.9987	99.23	99.23 ± 0.01, 0.01 (%)
3.0235	2.9999	99.22	

Table 4. PWR

a)

Sample No.	Sample wt. mg	Results		Pu Content (%)
		[Pu], ug/g <sup>b)</sup> , (rsd, %)	Total Pu, mg	
1	512	48.9, (0.18)	2.615	0.511
2	507	85.5, (0.32)	3.717	0.733
3	551	99.4, (0.37)	4.230	0.768
5	737	84.3, (0.40)	5.756	0.781
8	511	106.2, (0.53)	4.413	0.864
11	324	75.5, (0.47)	2.797	0.863

a) PWR spent fuel samples from Kori-1. b) Number of titration : n = 2 - 6.