

PLUTONIUM REMOVING TEST FROM UNPURIFIED URANIUM SOLUTION USING INORGANIC ADSORBENT IN JAERI'S REPROCESSING TEST FACILITY

Akinori Kawachi , Takeo Mimori , Kazutoshi Miyajima , Hironori Ishikawa
Japan Atomic Energy Research Institute. Ibaraki, Japan

ABSTRACT

JAERI's Reprocessing Test Facility (JRTF) was operated from 1968 to 1969 after which spent fuel (U/Al metal, burnup 600MWD/T) from a research reactor (JRR-3) was reprocessed. The reprocessing tests were carried out successfully, and about 200g of highly purified plutonium (Pu) was recovered. Because a uranium purification system was not installed in JRTF, about 1,700L of unpurified uranium solution containing U (350 mg/mL), Pu (1.0×10^{-3} mg/mL) and beta, gamma nuclides was generated from these tests. It was necessary to remove Pu from the unpurified uranium solution for interim storage in JAERI's Waste Treatment Facility (JWTF).

JAERI and Unitika Ltd. have jointly developed an inorganic adsorbent made from fibrous activated carbon (FAC), which has excellent characteristics to remove Pu from reprocessed liquid waste. Based on the results of laboratory-scale tests, JAERI constructed the engineering-scale apparatus using the inorganic adsorbent for the unpurified uranium solution in FY 1995. The treatment capacity of the apparatus was 22L/batch, which was 4 times the volume of the column loaded with the adsorbent. The treatment of 1,700L of liquid was completed in FY 1997.

The results of Pu removing tests by the adsorbent are summarized below;

The adsorbent showed high selectivity for Pu in the liquid waste containing a large amount of U. The removal capacity of Pu depended on space velocity (SV) and temperature. The enough condition was 0.64 h^{-1} for SV and 20 – 30 °C for temperature. Pu removal efficiency was up to 95% and the amount of adsorbed Pu was 0.02 – 0.03 mg-Pu/g-FAC.

It was confirmed that the inorganic adsorbent had an excellent ability to remove Pu from reprocessed liquid waste on an engineering-scale.

INTRODUCTION

It is probable that liquid waste containing TRU nuclides from reprocessing plants will increase in the future. It will be necessary to remove TRU nuclides from the liquid waste and to reduce the volume of the radioactive waste.

In JAERI, development of more effective treatment process for TRU waste has been studied using various kinds of radioactive liquid waste stored in JRTF.

JAERI and Unitika Ltd. have jointly developed an inorganic adsorbent made from fibrous activated carbon (FAC), which has excellent characteristics to adsorb Pu in the presence of nearly 1M nitric acid and reduce the volume by the incineration. We reported the results concerning the adsorption ability and the burning property of the adsorbent at Waste Management '96. (1)

The unpurified uranium solution is treated by the process using the inorganic adsorbent. This adsorbent is molded in a cartridge form by adding the inorganic binder to FAC which is easily applied in industrial settings. The adsorbent has the ability to remove Pu directly without using pre-treatment with a nitric acid solution, and large volume reduction of the waste is expected by incineration.

The treatment technology was developed by JAERI under a contract with the Science and

Technology Agency. In this paper, we described the outline of the treatment process for unpurified uranium solution and the results of a Pu removal test using the inorganic adsorbent.

THE OUTLINE OF TREATMENT PROCESS FOR UNPURIFIED URANIUM SOLUTION

Properties of the unpurified uranium solution

Because a uranium purification system was not installed in JRTE, 1,700L of unpurified uranium solution was generated from the reprocessing tests. The liquid waste contained alpha nuclides such as 350mg-U/mL and 1.0×10^{-3} mg-Pu/mL and beta, gamma nuclides. It consisted of 0.6M nitric acid solution and included a remarkable amount of dissolved metallic salts. The specific gravity of the liquid waste was approximately 1.6. Table 1 shows the properties of the liquid waste.

Table 1 Properties of unpurified uranium solution

Pu concentration	Bq/mL	2.3E+3
	mg/mL	1.0E-3
concentration	Bq/mL	8.7E+3
	mg/mL	3.5E+2
Gamma Cs-137 radioactivity	Bq/mL	1.3E+3
Gross beta radioactivity	Bq/mL	1.4E+4
Dissolved metal	mg/mL	Ca 1.5E-3 Cr 6.7E-3 Fe 2.6E-2 Mg 3.8E-4 Mn 7.4E-4 Na 3.8E-2 Ni 2.5E-3 etc
Acidity	M	0.6

Properties of the inorganic adsorbent

The adsorbent was molded in a cartridge form by adding the inorganic binder to FAC, with a specific surface area of approximately 2,000m²/g and high hydrophilicity. The cartridge consisted of 65wt% of FAC and 35wt% of inorganic binder (Type-A). The adsorption property of type-A was compared with that of another cartridge (Type-B) reducing the inorganic binder. Table 2 shows the properties of the inorganic adsorbents.

Table 2 Properties of inorganic adsorbents

Adsorbent	Molding Composition	Molding Density	Weight 1 cartridge (g/cm ³)	Physical properties			
	wt%			Specific Surface Area	Pore Volume	Equilibrium Moisture	
	FAC	Inorganic binder		g	(m ² /g)	(mL/g)	(%)
Type-	65	Glass fiber / Rock wool / Bentonite 35	0.28	260	1,700	0.9	10
Type-	80	Glass fiber / Lithium silicate 20	0.22	200	1,700	0.9	10

Procedure for treatment process for the unpurified uranium solution

Fig.1 shows a flow diagram of the treatment process using the inorganic adsorbent for unpurified uranium solution. The main parts of this apparatus were installed in the glove box (L3,000mm x W1,200mm x H1,750mm) and the treatment capacity was 22L/batch.

The unpurified uranium solution was pumped from the storage tank to the feed tank and passed through the column loaded with cartridges to remove Pu. A portion of treated solution was sampled and its radioactivity was measured. The liquid waste was treated again in cases where its radioactivity exceeded the level defined by the JWTF's regulation. The conditions of the treatment for the liquid waste are shown as follows.

Adsorbent	Type-A
diameter	122 mm
length	480 mm
volume	5.6 L
weight	1,560 g (FAC weight 1,000 g)
Flow rate	3.6 L/h
Space velocity (SV)	0.64 h ⁻¹
Level defined by JWTF's regulation	
Alpha radioactivity	< 370 Bq/mL

Alpha / beta (gamma) □ 0.1

After neutralization, the treated solution was transported to the JWTF and solidified with plastic. The used adsorbent was dehydrated and solidified with cement, canned, and stored in the JWTF.

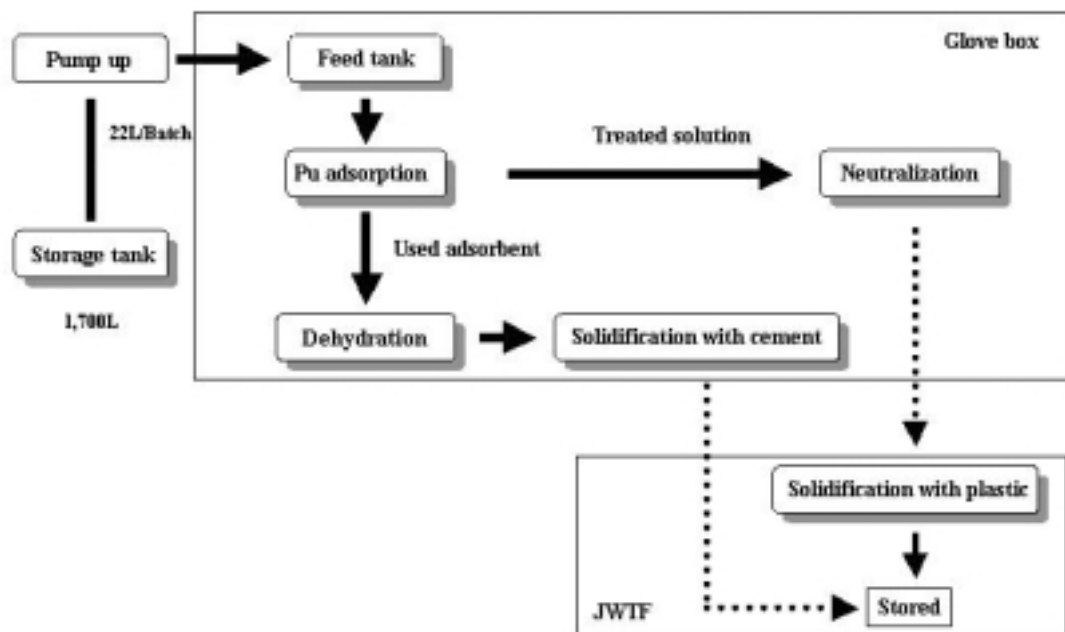


Fig. 1 Flow diagram of the treatment process using the inorganic adsorbent

Pu REMOVING TEST USING THE INORGANIC ADSORBENT

Experiment

In order to evaluate the process using the inorganic adsorbent, adsorption properties such as breakthrough properties, Pu removal efficiency and the amount of adsorbed Pu were examined under conditions of various space velocity (SV), temperature and adsorbent values.

A breakthrough property was derived from plotting the Pu concentration ratio (C/C_0) against the effluent volume (column volume: CV). The Pu removal efficiency, amount of adsorbed Pu and Pu concentration ratio (C/C_0) were calculated by the following equations.

$$\begin{aligned}
 \text{Pu removal efficiency (\%)} & : \{ (C_0 - C) / C_0 \} \times 100 \\
 \text{Amount of adsorbed Pu (Bq or mg)} & : (C_0 - C) \times V \\
 \text{Pu concentration ratio (} C/C_0 \text{)} & : C / C_0
 \end{aligned}$$

- C_0 : Pu concentration of the initial solution (Bq/mL or mg/mL)
 C : Pu concentration of the effluent or the treated solution (Bq/mL or mg/mL)
 V : Volume of the treated solution (mL)

Alpha radioactivity and gamma radioactivity were measured using alpha spectrometry with a Si (Li)-detector and a gamma spectrometer with Ge (Li)-detector, respectively. Gross beta radioactivity was measured by a 4 π gas flow counter.

Results and Discussion

Selectivity for Pu

Fig.2 shows a breakthrough curve of Pu. Approximately 0.8CV of effluent was held up by the adsorbent. Until 6.3CV of effluent, the Pu concentration ratio (C/C_0) was very low. After 10 % breakthrough, C/C_0 increased and the JWTF did not accept the treated solution. Accordingly, we regarded 10 % breakthrough as the limit of the adsorption ability. Pu removal efficiency was over 95% and the amount of adsorbed Pu was about 0.033mg-Pu/g-FAC. Table 3 shows properties of the initial solution and treated solution. As shown, the adsorbent has high selectivity for Pu even in the liquid waste containing a large amount of U and dissolved metallic salts.

Table 3 Results of Pu removing test using the inorganic adsorbent

		Initial solution	Treated solution
Pu concentration	Bq/mL	2.3E+3	8.4E+1
	mg/mL	1.0E-3	3.7E-5
concentration	Bq/mL	8.7E+3	8.6E+3
	mg/mL	3.5E+2	3.5E+2
Gamma Cs-137 radioactivity	Bq/mL	1.3E+3	1.2E+3
Gross beta radioactivity	Bq/mL	1.4E+4	1.3E+4

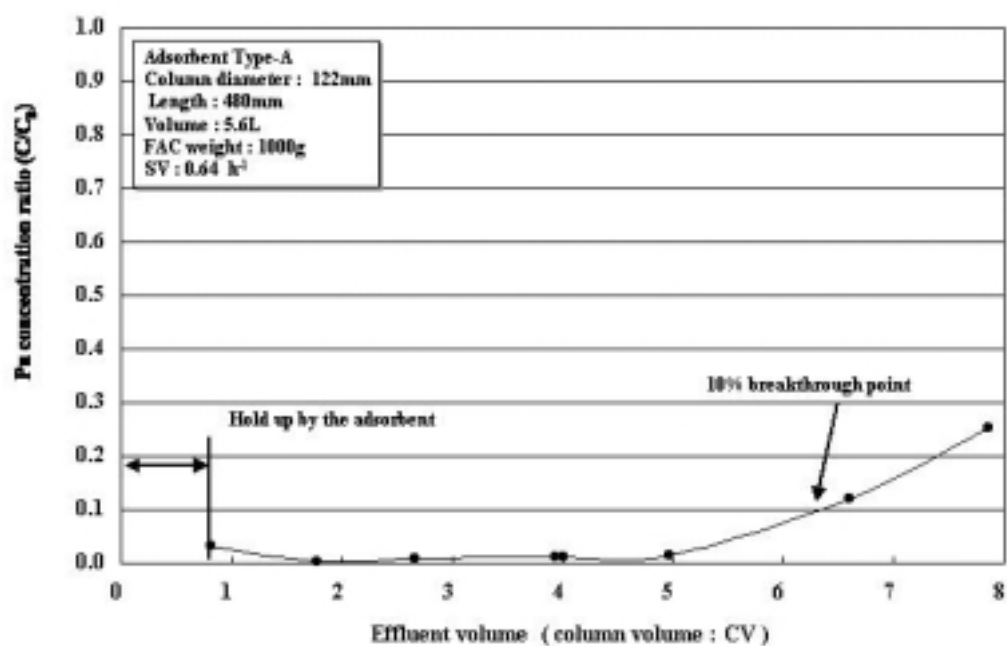


Fig. 2 Breakthrough curve of Pu (at 30°C)

Effect of SV

The breakthrough tests for Pu were examined at a range of SV 0.64 to 1.40 h⁻¹. As the SV increased, 10% breakthrough was shifted to a lower column volume as shown in Fig.3. The optimum condition for SV was 0.64 h⁻¹ in consideration of operation.

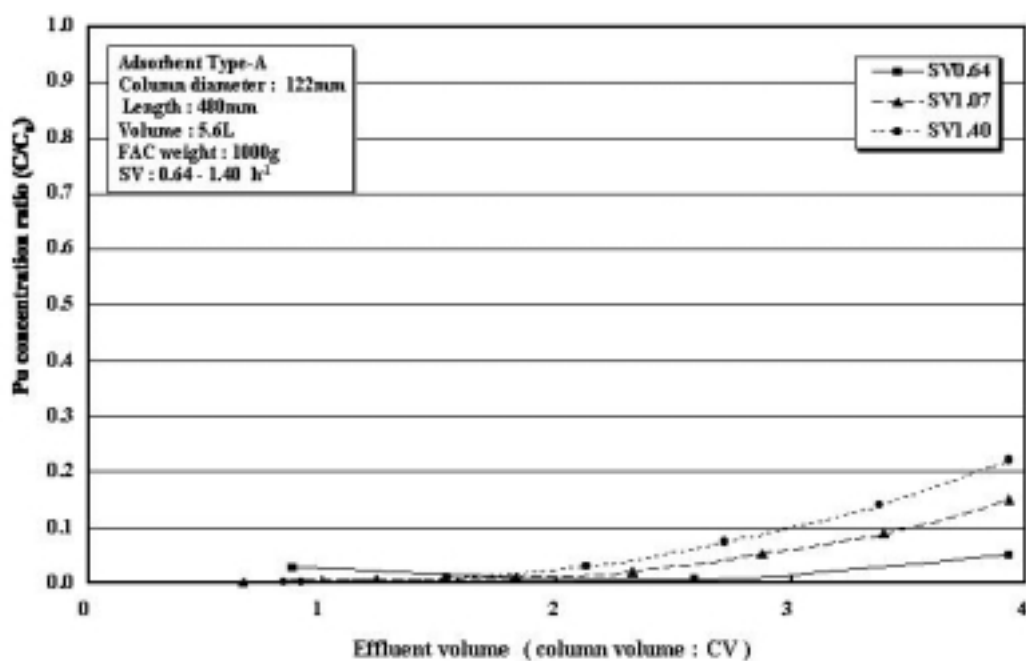


Fig. 3 Effect of Space velocity (at 20°C)

Effect of temperature

The breakthrough tests for Pu were examined at 20°C and 30°C. Fig.4 shows breakthrough curves of Pu. As the temperature increased from 20°C to 30°C, 10% breakthrough was shifted from 3.8CV to 6.3 CV. The amount of adsorbed Pu was 0.020mg-Pu/g-FAC at 20°C and 0.033mg-Pu/g-FAC at 30°C. It is believed that the temperature had an influence on a diffusion efficiency of Pu in a micro pore of FAC, which determined Pu adsorption ability.

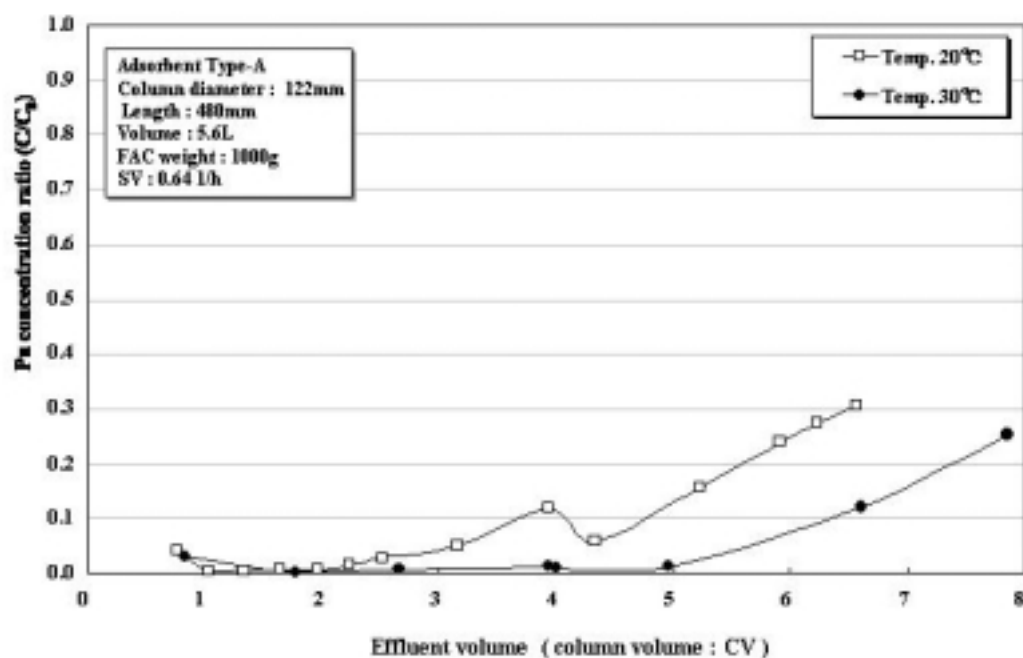


Fig. 4 Effect of temperature

Effect of adsorbent components

Fig.5 shows the comparison of the adsorption ability between type-A and type-B. The Pu adsorption ability of type-A decreased in proportion to increasing SV. But no decreasing Pu removal efficiency of type-B was observed at each SV. These results were attributed to the quantity of inorganic binder in the adsorbents. The surface of the adsorbent was partially covered with inorganic binder, and so contact with Pu in the liquid waste was inhibited.

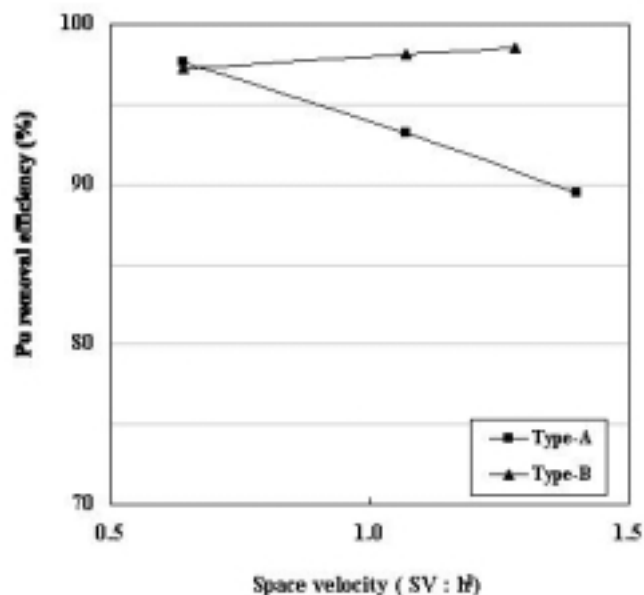


Fig. 5 Comparison between Type-A and -B

CONCLUSION

1,700L of unpurified uranium solution was treated from FY 1996 to 1997. The results of Pu removal tests by the adsorbent are summarized below.

The adsorbent showed high selectivity for Pu in the liquid waste containing a large amount of U. The removal capacity of Pu depended on space velocity (SV) and temperature. The enough condition was 0.64 h^{-1} for SV and $20 - 30^\circ\text{C}$ for temperature. Pu removal efficiency was up to 95% and the amount of adsorbed Pu was $0.02 - 0.03 \text{ mg-Pu/g-FAC}$.

These results suggest that the process using the inorganic adsorbent was quite effective in treatment of the reprocessed liquid waste on an engineering-scale.

As the inorganic adsorbent consists of more than 90% of carbon, it is easily converted to CO_2 and H_2O by incineration. It is possible to expect useful applications from the standpoint of volume reduction of the radioactive waste. In order to evaluate the reduction of the used adsorbent, the reducing process of the used adsorbent by incineration will start in FY 1999.

REFERENCE

1. T.MIMORI , K.MIJAJIMA , H.TAKAHASHI , M.OIWA , T.OKAMOTO , T.MORI , H.IWAYA ,
“ STUDIES ON PLUTONIUM ADSORPTION BY INORGANIC ADSORBENT “ , Waste
Management '96 Conference , February 25-29 , 1996 , Tucson , AZ