Thermal Pretreatment For TRU Waste Sorting

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ABSTRACT

Japan Atomic Energy Agency conducted a study on thermal treatment of TRU waste to develop a removal technology for materials that are forbidden for disposal. The thermal pretreatment in which hot nitrogen and/or air is introduced to the waste is a process of removing combustibles, liquids, and low melting point metals from PVC wrapped TRU waste. In this study, thermal pretreatment of simulated waste was conducted using a desktop thermal treatment vessel and a laboratory scale thermal pretreatment system. Combustibles and low melting point metals are effectively separated from wastes by choosing appropriate temperature of flowing gases. Combustibles such as papers, PVC, oil, etc. were removed and low melting point metals such as zinc, lead, and aluminum were separated from the simulated waste by the thermal pretreatment.

INTRODUCTION

Japan Atomic Energy Agency (JAEA) has stored TRU waste generated from reprocessing and plutonium fuel fabrication R&D activities in Nuclear Fuel Cycle Engineering Laboratories. The total volume of the stored solid waste is approximately 22,000 m^3. JAEA has been developing the conceptual design for the TRU waste treatment facility to dispose of the stored TRU waste. The simplified process flow diagram of the TRU waste treatment is shown in Fig. 1. This treatment consists of four major steps, pretreatment, incineration, compaction, and conditioning. Conventional technologies used for low level radioactive waste treatment should be also used for the TRU waste treatment facility except pretreatment technology. Although the pretreatment (unpacking and sorting wastes) has been done by hand to remove harmful substances for disposal such as aluminum, zinc, lead, etc. at LLW treatment facilities in Japan, automation of the pretreatment process should be needed for the TRU waste treatment facility to reduce the risk of radiation exposure because of its relatively high dose rate. JAEA has tested and evaluated mechanical unpacking and sorting technologies for JAEA's TRU waste treatment such as wind sorting, magnetic sorting, etc. but no mechanical technology has been available for the treatment. Therefore, JAEA has been developing a new thermal pretreatment process since 2004 to automate the unpacking and sorting process for TRU waste. The thermal pretreatment is the method of removing combustibles, liquids, and hazardous metals from PVC wrapped wastes by introducing hot nitrogen and/or air to the waste. Combustible materials and hazardous metals are effectively separated from wastes by choosing appropriate temperature of flowing gases. Studies on zinc plating removal using a desktop thermal treatment vessel and on unpacking and sorting of simulated TRU waste using a laboratory scale thermal treatment system were conducted. The results obtained in this study indicate that the thermal pretreatment separates combustibles and low melting point hazardous metals from PVC wrapped TRU waste.

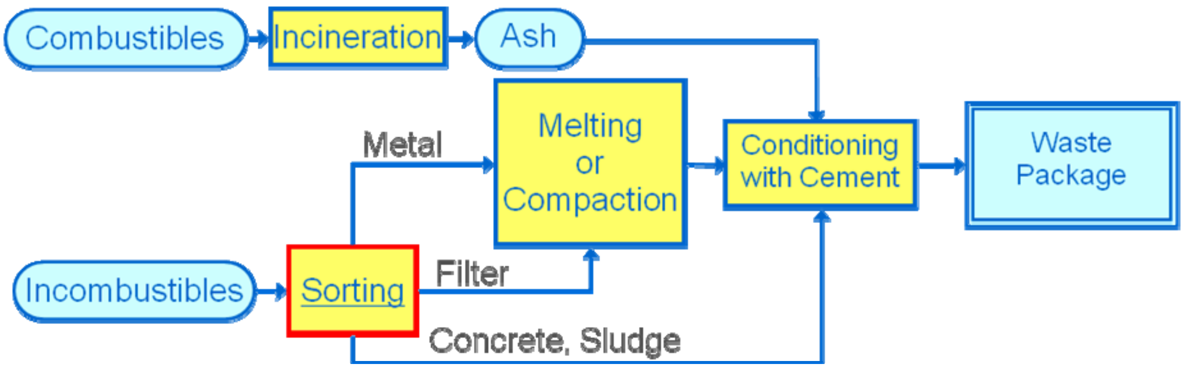

Fig. 1. Process flow diagram of the TRU waste treatment

EXPERIMENTAL

Zinc plating removal

A desktop thermal treatment vessel was used to study the removal of zinc plating from steel. The vessel was made of stainless steel, 108.3 mm in diameter, and 110 mm deep attached with a gas inlet and a gas outlet.
Test pieces of 1.2 mm thick, 40 mm wide, and 60 mm long zinc plated steel were set in the vessel and were heated up to 650 °C with continuous nitrogen flowing 100 ml/h. Residual zinc on the test pieces and in the vessel was dissolved by nitric acid and its concentration was measured using inductively coupled plasma atomic emission spectrometry.

Thermal pretreatment of simulated TRU waste

The waste sample used in this study simulated the waste generated from plutonium fuel fabrication facilities and a reprocessing plant in JAEA. The contents of the simulated TRU waste are shown in Table I. Each item was wrapped with papers and PVC sheets, followed by being packed in a PVC bag and sealed with packing tape.

Table I. Contents of the simulated waste

Materials	Shapes	Weight
Stainless steel	Plate	5.0 kg
Carbon steel	Plate, Pipe	3.5 kg
Aluminum	Plate	0.5 kg
Lead	Plate	0.5 kg
Brass	Bolt, Nut	0.5 kg
Silicone oil	-	0.05 kg
Grease	-	0.05 kg
Zinc plated steel	Plate	0.17 kg
Total		10.27 kg

Thermal pretreatment study on the simulated TRU waste was conducted using the laboratory scale thermal pretreatment system comprising a hot gas generator, a heating chamber, a burner, and a scrubber. The simulated waste sample set in a rotating cage (300 mm diameter x 500 mm length) in the heating chamber, shown in Fig. 2, were heated by hot nitrogen gas and/or air up to 700 °C. Organic substances such as papers and PVC sheets in the samples were pyrolyzed and gasified in the heating chamber and then oxidized in the burner. The off-gas from the burner was treated in the scrubber to remove acid gases and volatile metals. Melted metals such as lead and aluminum were collected at the bottom of the heating chamber.

In this study, simulated waste was heated at about 2 °C /min in nitrogen flow. It took five to six hours to reach treatment temperature. Then the waste was kept at the treatment temperature for one hour in nitrogen gas with 5% oxygen and cooled to room temperature.

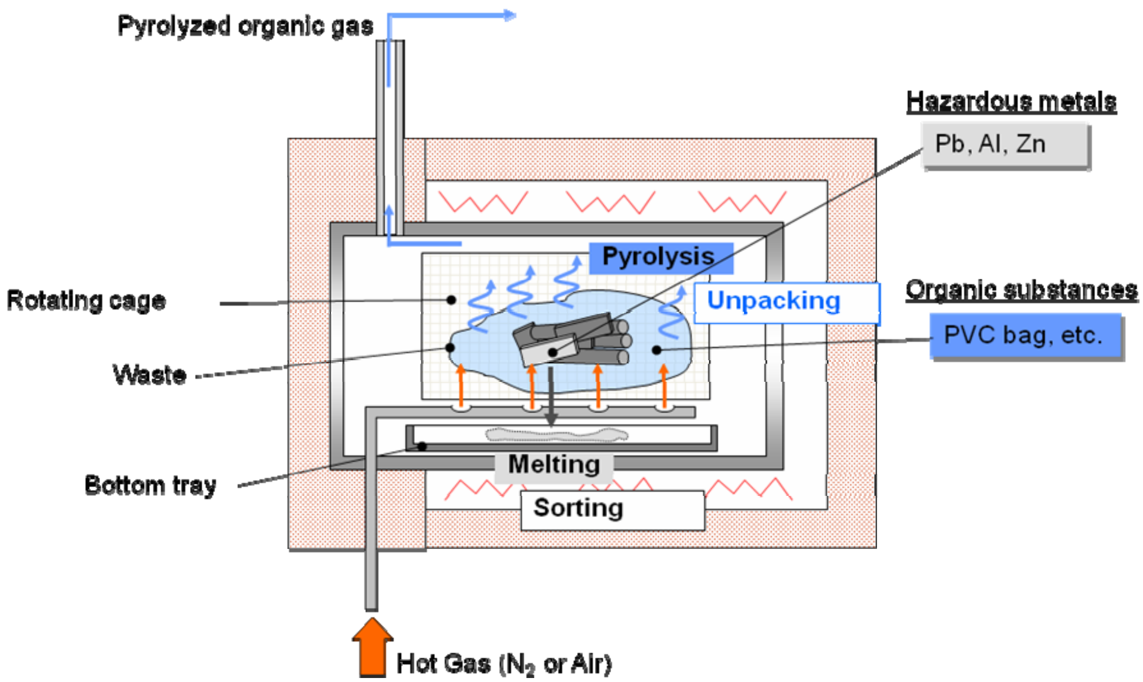

Fig. 2. Conceptual figure of the heating chamber for waste unpacking and sorting

RESULTS AND DISCUSSION

Zinc plating removal

The results of experiments on removing zinc using the desktop thermal treatment vessel are summarized in Table II. Almost all zinc remains on the test pieces at 450°C and 500°C even though the temperatures exceeds the melting point of zinc, 419.5°C. The removal amount of zinc from test pieces is increased with increase in temperature at 550°C and higher. At 650°C, over 90% of zinc is removed from steel surface and about half of it is entrained in off-gas. These results mean that zinc plating should be vaporized to be removed from steel plates and zinc in the off-gas should be trapped to prevent off-gas line blockage.

Table II. Distribution of zinc after thermal treatment of zinc plated steel

Temperature/ °C	Time/h	Distribution of zinc/%		
		Test pieces	Thermal treatment vessel	Off-gas
450	0.5	99.9	0.0	0.1
	1.0	97.2	2.3	0.5
	2.0	99.4	0.4	0.2
500	0.5	94.3	4.6	1.1
	1.0	89.7	10.6	0.0
	2.0	99.9	0.1	0.0
550	0.5	82.8	12.2	5.0
	1.0	78.8	15.5	5.7
	2.0	70.6	22.0	7.4
600	0.5	62.0	22.6	15.4
	1.0	49.0	25.3	25.7
	2.0	33.8	32.6	33.6
650	0.5	22.4	35.4	42.2
	1.0	6.7	39.1	54.2
	2.0	4.4	46.8	48.8

Thermal pretreatment of simulated TRU waste

The picture of the simulated waste which was treated at 650°C is shown in Fig. 3. Most PVC, packing tape, and paper was vaporized and a small portion was converted to char. Silicone oil and grease were removed. Unpacking and eliminating organic material processes were successfully conducted by the thermal pretreatment. X-ray fluorescence semi-quantitative analysis of the char collected in the rotating cage and the bottom tray indicated that the main elemental components of the char beside carbon are chlorine, calcium, and lead.

Fig. 3. Unmelted pipes and plates collected in the rotating cage

Table III shows the distribution of low melting point metals after the thermal treatment. Over 70% of lead is melted and accumulated in the bottom tray but about 10% of it remains in the rotating cage. The remaining lead in the cage has stuck to the char. Thus the char should be removed from the cage to separate all lead from the waste. Only 30% of the aluminum is collected at the bottom tray at 700°C even though the temperature is higher than the melting point of aluminum, 660°C. This low collecting rate could be due to the inhomogeneous temperature distribution in the cage. Gas circulation in the cage or higher temperature treatment (> 700°C) is required to increase the collection rate of aluminum. The collection rate of zinc in the scrubber at 500°C is higher than that conducted in the desktop thermal treatment vessel. It is probable that the reaction between zinc and chlorine produced by the pyrolysis of PVC increased the vaporization of zinc because the boiling point and melting point of the reaction product zinc chloride, 293°C and 732°C respectively, are lower than these of zinc.

Table III. Distribution of low melting point metals

Material	Temperature/°C	Distribution/%				
		Cage	Tray	Scrubber	HEPA Filter	Total
Pb	500	11.0 *1	73.1	2.7	0.0	86.8
	600	24.3 *1	76.0	3.3	0.0	103.6
	700	6.7 *1	93.2	5.0	0.0	104.9
Al	500	100.4	0.0	0.0	0.0	100.4
	600	75.6	38.8	0.0	0.0	114.5
	700	70.0	30.3	0.1	0.0	100.4
Zn	500	100.0 *2	0.0	1.8	0.0	101.8
	600	100.4 *2	0.0	2.2	0.0	102.6
	700	100.0 *2	0.0	2.0	0.0	102.0

*1: Sticking to carbon residue
*2: Brass

The picture of melted lead and aluminum collected in the bottom tray is shown in Fig. 4. Aluminum and lead were melted together and were difficult to separate. Lead must be separated from the melt and stabilized for disposal. A two step heating would be necessary to avoid this problem. The first step is keeping waste at 500°C to melt lead and the next step is heating waste to higher than 700°C to separate the aluminum.

Fig. 4. Melted lead and aluminum collected in the bottom tray

CONCLUSIONS

Studies on zinc plating removal using a desktop thermal treatment vessel and on unpacking and sorting of simulated TRU waste using a laboratory scale thermal treatment system were conducted to develop a removal technology for substances that are forbidden for disposal.

Study on zinc plating removal using a desktop thermal treatment vessel shows that zinc plating should be vaporized to be removed from steel plates at higher than 650°C.

In the thermal pretreatment of simulated TRU waste, over 70% of lead is melted and accumulated at the bottom tray but about 10% of it remained in the rotating cage. Only 30% of aluminum was collected at the bottom tray at 700°C due to the inhomogeneous temperature distribution in the cage.

Aluminum and lead collected in the bottom tray were melted together and they were difficult to separate. A two step heating process would be necessary to separate them.