Operational Experience of Chlorinated Compounds Waste Incineration System for Plutonium Contaminated Wastes - 10061

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ABSTRACT

The Japan Atomic Energy Agency (JAEA) has designed and manufactured a new type incineration system for chlorine containing wastes and combustible wastes generated from MOX fuel fabrication facilities.
The incineration system has been operating since June 2002. The plant has attained around 6000 hours of operation and treated about 46.4ton (290m^3) of plutonium contaminated solid wastes, as of today. Volatile chloride accumulated at the exhaust gas cooler, resulting in a flow obstruction. The incineration system has successfully and stably operated with periodic cleanup of the accumulated volatile chloride by using remote handling tools.

INTRODUCTION

Plutonium-contaminated solid wastes have been generated from MOX fuel fabrication process in the Japan Atomic Energy Agency (JAEA). The generated plutonium contaminated solid wastes can be classified into three categories, namely, combustibles, incombustibles and chlorinated wastes. Typical combustibles are cleaning fabrics, working clothes, polyethylene bottles, and vinyl acetate sheets. Incombustibles are dismantled glove boxes, equipments and filters. Chlorinated wastes are polyvinyl chloride (PVC) bags, chloroprene rubber and latex gloves. These wastes are packaged into 20ℓ carton boxes. Six carton boxes are put into 200ℓ drums. Chlorine containing solid wastes such as PVC bags and chloroprene rubber gloves have been steadily generated from of the MOX fuel facilities, because MOX fuel is fabricated in glove boxes. Therefore the percentage of chlorinated wastes in the MOX fuel facilities is higher than other nuclear cycle facilities. Due to the limited storage space, development of volume reduction technology for chlorinated wastes is necessary. JAEA had designed and manufactured a new type incineration system for the chlorinated and combustible wastes based on past experience of the Plutonium-contaminated Waste Treatment Facility [1]. This paper presents demonstrated operation results and issues revealed from the operation.

PROCESS DESCRIPTION

Figure 1 shows process flow of the incineration system. The system consists of waste feeder, water-cooling jacket type incinerator, primary ceramic filter, exhaust cooler, secondary ceramic filter, HEPA filter and other exhaust gas purifications units. All equipment is in an air-tight structure to prevent contamination in the work environment. Therefore, wastes feeder, ash extraction and equipment maintenance were carried out through glove boxes.
Corrosion resistance and cost performance were the primary criteria to be considered in the selection of materials for the incineration system. Main part of equipment is made from nickel alloy or ceramic coated stainless steel in order to prevent acid corrosion. Ceramic coated stainless steel was utilized as a component of the system wherever possible, if the highest operating temperature anticipated in the equipment is within the recommended operating range of ceramic coating. Nickel alloy was selected for the component where the equipment is operated above the ceramic coating design temperature. Furthermore, the equipment is maintained at the temperature above the acid dew point to avoid internal condensation.

Waste Feeder

The solid waste packaged drums are accepted from waste storage facilities at the receiving area of the PWTF. At the waste pretreatment process step, the waste drum is opened and the waste packages are taken out from the drum. The surface contamination of the package, weight, dose rate and absence of metal particles is checked. The waste packages are loaded into the glove box via an air lock. The waste packages are individually fed into the incinerator through two slide gates without any pretreatment. The two gates create a barrier between the glove box atmosphere and the incinerator.

Incinerator

The incinerator is bicylindrical structured, and cooling water circulates through the cylinders to prevent local temperature hot spots. Combustion air is supplied at high velocity from air nozzles arranged all over the inner wall of the incinerator to accelerate burning the chlorinated wastes. The waste is ignited by the primary burner. Throughput of the incinerator is 10kg/h.

Primary Ceramic Filter

Exhaust gas temperature is raised up over 550 ºC at the primary ceramic filter chamber in order to enhance the secondary combustion. Incinerator fly ash is trapped by ceramic filter elements. The collected ash is blown off from the surface of filter media by means of pulsed jets of compressed air.

Exhaust Gas Cooler

The gas cooler consists of spray cooling tower and air dilutor. The quench cools the flue gas exiting the first ceramic filter at gas exhaust temperature above 600ºC to below maximum operating temperature of HEPA filter (250 ºC) by mean of water atomization and air dilution.

Secondary Ceramic Filter

PVC sheets and chloroprene gloves contain not only high fraction of chlorine, but also significant amount of zinc and lead as additives. Zinc and lead react with chlorine to form volatile metallic chlorides ($PbCl_2$ and $ZnCl_2$) during incineration. The volatile metallic chlorides pass through the primary ceramic filter in a gaseous state because of its high vapor pressure at the temperature in the primary ceramic filter (as shown in Table I). The exhaust gas is cooled down to the temperature below maximum operating temperature of HEPA filter. Thus, the off-gas is cooled by mean of both water atomization and air dilution. Volatile chlorides which condense during cooling cause clogging of HEPA filter. Therefore secondary ceramic filter with reverse washing unit is placed before HEPA filter as a pre-filter to prevent clogging. Reverse washing unit can be performed in parallel manner with incinerator operation. Zinc chloride in particular is a very hygroscopic product which becomes sticky after absorbing moisture. To improve detachability of zinc chloride by reverse washing, the filter elements of secondary ceramic filter are coated with fine SiO_2 powder before operation.

Exhaust Gas Purification

After having passed the secondary ceramic filter, the gasses enter the HEPA filters, consisting of two inline units. The outlet temperature is 200 ºC. The alkali scrubber consists of a quench tower for the cooling down of gasses below 50 ºC, a counter current scrubbing tower with caustic liquid for removal of HCl and SO_2, condenser, and demister. The flue gases are heated above 30 ºC in order to decrease the relative humidity and to avoid condensation. Two extraction blowers in parallel ensure the evacuation of flue gasses into the atmosphere. One blower is stand-by. The dioxin is decomposed at 300ºC by a catalyser.

Important feature of this exhaust gas treatment system is that the radioactive liquid waste is not generated

because the alkali scrubber is installed after the HEPA filters.

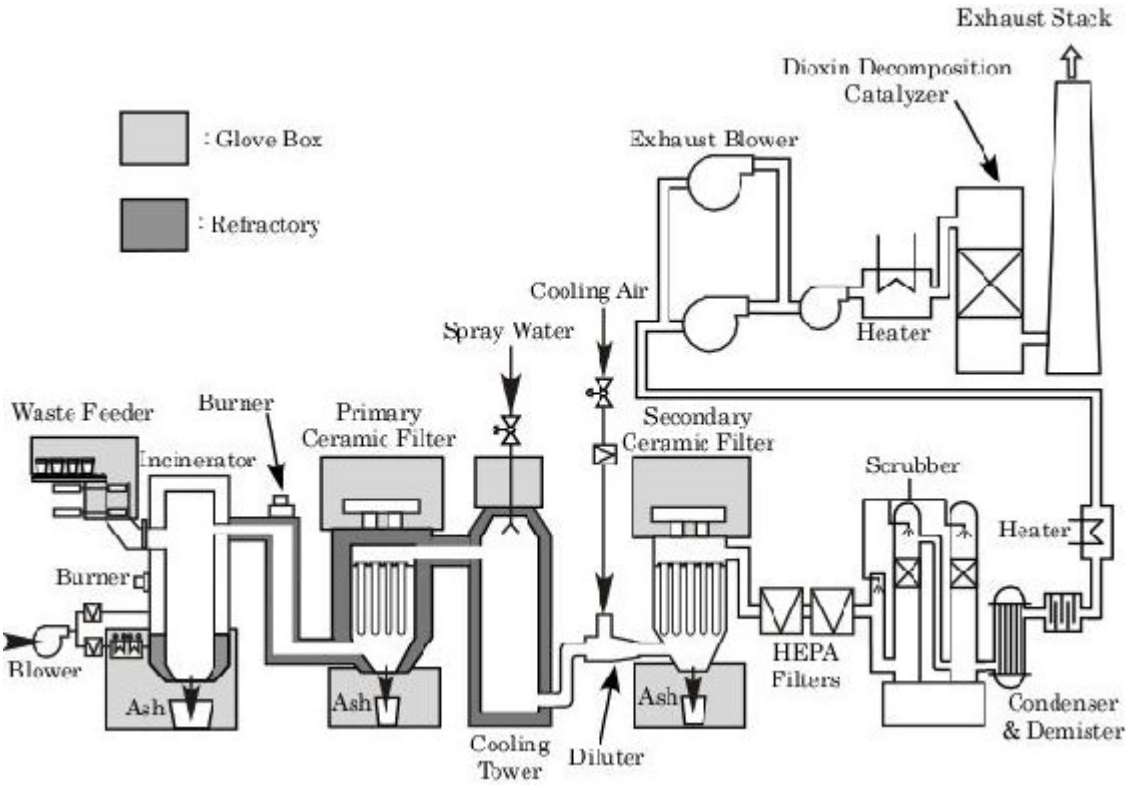

Fig. 1. Schematic flow diagram of incineration system

Table I. Vapor pressure of chlorides

Temperature / Material	Melting temperature /°C	Boiling temperature /°C	Vapor pressure* /Pa		
			600°C (Primary C.F.)	290°C (Cooling tower)	200°C (Diluter)
$ZnCl_2$	290	732	3.79×10^{3}	4.46×10^{-1}	1.19×10^{-3}
$PbCl_2$	501	950	1.37×10^{2}	1.35×10^{-3}	9.48×10^{-7}
$CaCl_2$	772	>1600	6.52×10^{-7}	3.09×10^{-16}	$6.71.\times10^{-22}$

*Vapor pressure was calculated by mean of using chemical equilibrium analysis code 'CHEEQ'[2].

VOLATILE CHLORIDE RETENTION

During the operation period from 2002 to 2004, incidents of pressure loss of exhaust gas cooler (between inlet of the cooling tower and outlet of the diluter) increased with increasing amount of treated waste (Figure 2). It is shown that there was obstruction at the exhaust gas cooler. Interior of exhaust gas cooler was examined using CCD camera to confirm the location and degree of flow obstruction. Locations of hold-up material in the flow path are shown in Figure 3. The pipe elbow and diluter showed the greatest hold-up obstruction, especially near the agitating blade. Samples of sediment were collected to analyze its property. Sediment gathered at the cooling tower and the pipe was hard and brittle while that at the diluter was soft and wet like icy slush. Qualitative analysis of sediment was conducted by using XMA (X-ray Micro Analysis) (Table II).

Although major elements of sediment were chlorine and lead, sediment in the diluter contained a large portion of zinc and the result suggest that the reason of sediment growth is considered to be the accumulation of deliquescent zinc chloride.

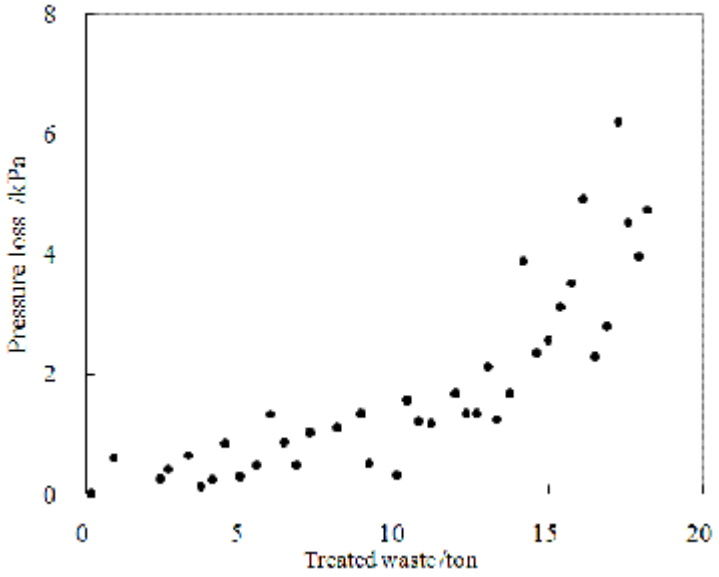

Fig. 2. Pressure loss of exhaust gas cooler

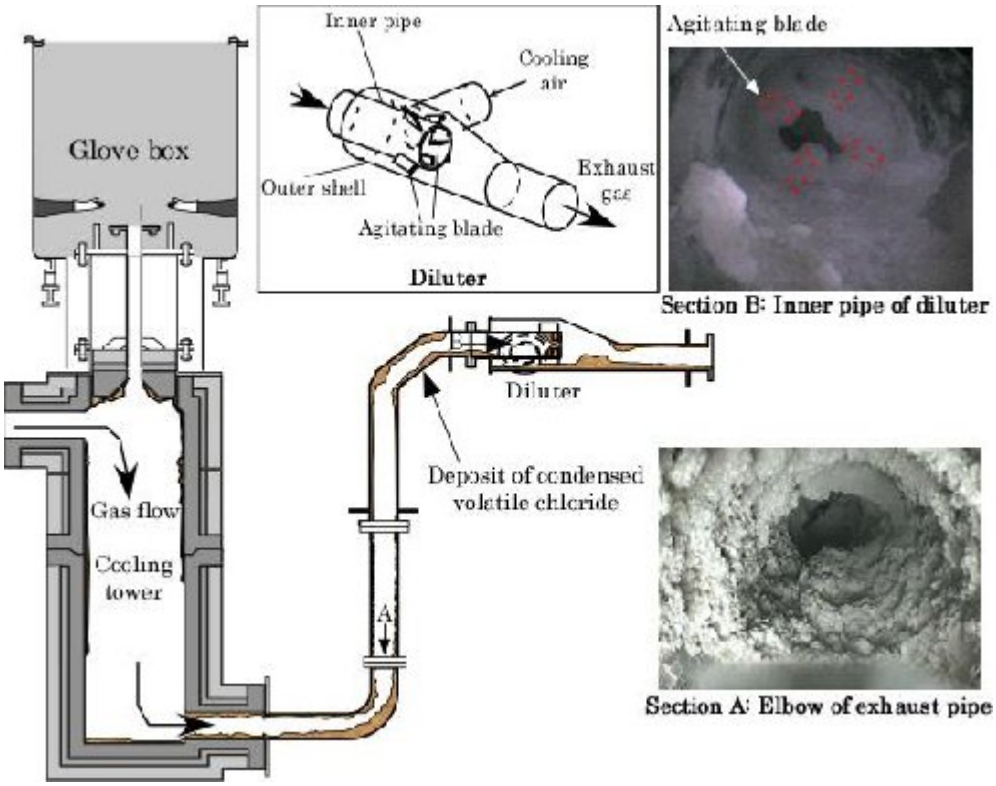

Fig. 3. Appearance of sediment in the exhaust gas cooler

Table II. Result of qualitative analysis by XMA

Element /wt.%		Cl	Zn	Pb	Ca	Si	Other (Na,Mg,P,S,K,Ti,Fe,Ni)
Sediment	Cooling tower	22.7	3.1	34.9	3.7	1.7	33.9
	Exhaust gas pipe	27.2	8.7	39.6	3.5	1.2	19.8
	Diluter	29.9	30.8	21.9	3.9	1.2	12.3
Reference	Incineration ash	24.5	0.0	0.0	38.2	10.6	26.7
	Fly ash (Primary C.F.)	1.3	3.3	23.4	27.3	12.7	32.0
	Fly ash (Secondary C.F.)	17.4	3.8	56.9	0.4	18.9	2.6

CLEANING UP THE SEDIMENT

At the end of the operational period in 2004, cleanup work was carried out with the following procedure.

1. Setting up a plastic enclosure tent
2. Cutting airtight cover of flange connector of exhaust gas pipe
3. Disassembly of the pipe from the cooling tower and diluter
4. Sediment removal from each part
5. Re-connection of the pipe and attachment of new airtight cover by welding
6. Decontamination and removal of enclosure tent

The amount of sediment recovered was 27kg. Material balance of the sediment production is shown in Table III. Weight of volatile chloride accounted for about 1.3% of the weight of treated waste. Most of volatile chloride was collected by secondary ceramic filter as fly ash, however 12% of it remained at the exhaust gas cooler as sediment.

Table III. Material balance of the incineration system

	Treated Waste*[1]	Incineration ash	Fly ash recovered from primary C.F.	Sediment	Fly ash recovered from secondary C.F.
Weight /kg	18460	1240	41	27	201*[2]
Ratio of weight to treated waste /%		6.7	0.22	0.15	1.1
Weight ratio of fly ash and sediment /%				12	88

*[1]: The breakdown is; PVC 74% and rubber 26% by weight.

*[2]: After deduction of pre-coating powder weight

It was expected to accumulate volatile chloride with incinerator operation. The cleanup work with pipe disassembled required a substantial amount of time for contamination control. Two type of remote handling tools were developed (Figure 4). A fracturing type tool was used for hard sediment retained at the cooling tower and the pipe. Sediment was crushed by run-out of a head part triggered by intermittent injection of compressed air. Crushed sediment was blown back to the bottom of the cooling tower by compressed air, and was recovered by vacuum cleaner. Owing to its simple and flexible mechanism, the remote handling tool could be applied to a more complex structure such as work that needs to be conducted using glove box method. The second, scoop type tool, was used for wet sediment retained at the diluter. Cleanup work was conducted once a year to prevent further flow obstruction. The period for cleanup work was shortened from three months to two weeks due to relive contamination control.

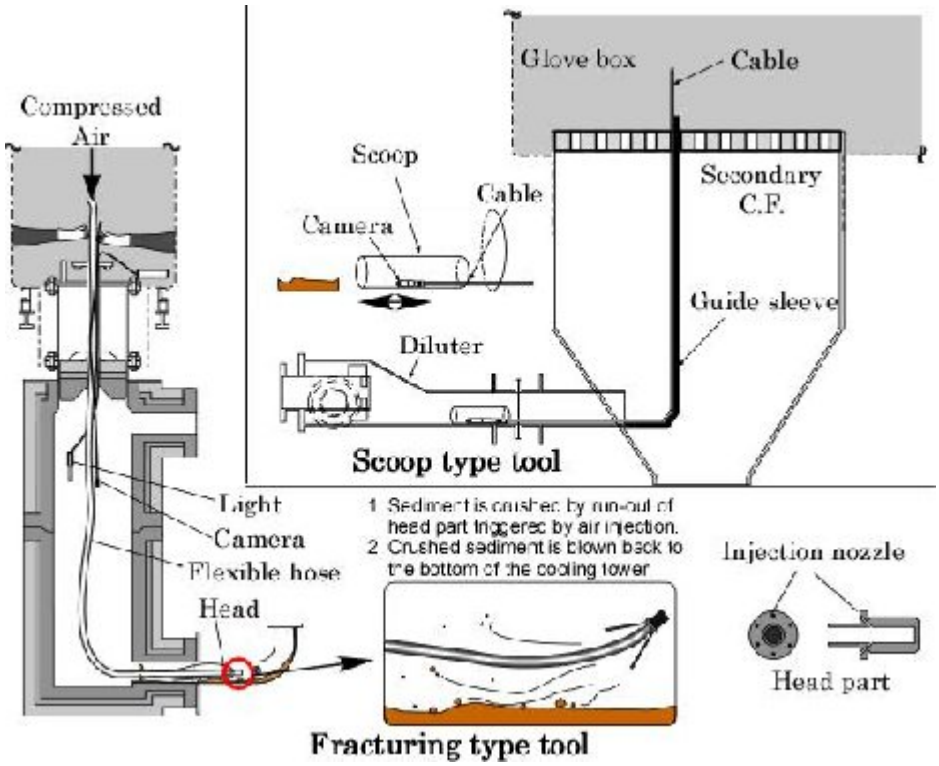

Fig. 4. Sediment removal tools for each retention part

OPERATION RESULT

Table IV shows the amounts of treated waste per fiscal year, the collected ashes, the weight reduction and volume reduction factors. The plant currently has about 6000 operational hours and has treated 46.4ton (290m^3) to date. The volume reduction factor has been approximately 47, and weight reduction factor approximately 12 (volume or weight reduction factor defined as the volume or weight of total waste fed into the incinerator divided by the volume or weight of total ash). A small quantity of sediment is observed at periodic maintenance checks. However as a result of periodic cleanup, pressure loss at the exhaust gas cooler has not increased significantly following long term operation.

Table IV. Operational results of the incineration system

Fiscal year	2002	2003	2004	2005	2006	2007*	2008	2009	Total
Chlorinate waste (ton)	3.6	9.3	5.6	2.2	2.8	-	5.9	4.9	34.3
Combustible waste (ton)	-	-	-	3.9	1.0	-	2.4	4.8	12.1
Total weight of waste (ton)	3.6	9.3	5.6	6.1	3.8	-	8.3	9.7	46.4
Total volume of waste (m^3)	24.3	55.7	33.2	41.9	23.8	-	51.1	61.7	291.7
Collected incinerator ash (ton)	0.24	0.64	0.36	0.32	0.2	-	0.57	0.63	2.96
Collected fly ash (ton)	0.07	0.19	0.11	0.08	0.04	-	0.09	0.09	0.67
Total collected ash weight (ton)	0.31	0.83	0.47	0.46	0.24	-	0.66	0.72	3.69
Total collected ash volume (m^3)	0.63	1.34	0.73	0.77	0.47	-	1.13	1.12	6.19
Weight reduction factor	11.6	11.2	11.9	13.3	15.8	-	12.5	13.4	12.5
Volume reduction factor	38.6	41.5	45.6	54.5	50.9	-	45.3	55.0	47.1

* It did not operate because of the government license renewal.

CONCLUSION

Operation of the chlorinated waste incineration system has been demonstrated for treatment of plutonium contaminated wastes generated from MOX fuel fabrication facilities. Plutonium contaminated solid wastes treated so far amount to 46.4ton ($290m^3$). The volume reduction factor was approximately 47 (weight reduction factor is approximately 12). Almost all volatile chloride in the exhaust gas was trapped by ceramic filter but some of it remained at the exhaust gas cooler. The remained volatile chloride could be removed by using remote handling tools. Pressure loss at exhaust gas cooler did not increase significantly following long term operation when periodic cleanup is performed.

REFERENCES

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