

Behavior of cesium in municipal solid waste incineration



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ABSTRACT

As a result of the Fukushima Daiichi Nuclear Power Plant accident on March 11, 2011 in Japan radioactive nuclides, primarily ^{134}Cs and ^{137}Cs were released, contaminating municipal solid waste and sewage sludge in the area. Although stabilizing the waste and reducing its volume is an important issue differing from Chernobyl nuclear power plant accident, secondary emission of radioactive nuclides as a result of any intermediate remediation process is of concern. Unfortunately, there is little research on the behavior of radioactive nuclides during waste treatment. This study focuses on waste incineration in an effort to clarify the behavior of radioactive nuclides, specifically, refuse-derived fuel (RDF) with added ^{133}Cs (stable nuclide) or ^{134}Cs (radioactive nuclide) was incinerated in laboratory- and pilot-scale experiments. Next, thermogravimetric (TG) and differential thermal analysis (DTA) of stable Cs compounds, as well as an X-ray absorption fine structure (XAFS) analysis of Cs concentrated in the ashes were performed to validate the behavior and chemical forms of Cs during the combustion.

Our results showed that at higher temperatures and at larger equivalence ratios, ^{133}Cs was distributed to the bottom ash at lower concentration, and the influence of the equivalence ratio was more significant at lower temperatures. ^{134}Cs behaved in a similar fashion as ^{133}Cs . We found through TG–DTA and XAFS analysis that a portion of Cs in RDF vaporizes and is transferred to fly ash where it exists as CsCl in the MSW incinerator.

We conclude that Cs-contaminated municipal solid wastes could be incinerated at high temperatures resulting in a small amount of fly ash with a high concentration of radioactive Cs, and a bottom ash with low concentrations.

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1. Introduction

Eastern Japan, especially the Pacific Ocean coast of the Tohoku region, was damaged by a huge earthquake and tsunami in March 2011. The tsunami washed away many buildings, and a large amount of disaster waste was generated. An estimated 22.8 million tons of waste accumulated in the most damaged prefectures (Iwate, Miyagi, and Fukushima), most of which had been remediated by the end of March, 2014 (Ministry of Environment, Government of Japan, 2014a). However, the Fukushima Daiichi Nuclear Power

Plant was also damaged by the tsunami, resulting in the release of radioactive nuclides, primarily ^{134}Cs and ^{137}Cs , to the atmosphere. The amount of ^{134}Cs and ^{137}Cs released was estimated to be approximately 1.8×10^{16} Bq, and 1.5×10^{16} Bq, respectively (IAEA, 2012). These radioactive nuclides contaminated the disaster waste, municipal solid waste (MSW), and sewage sludge from the surrounding area. According to the IAEA guidelines, incineration is the most commonly used thermal process for reducing the volume of these wastes (IAEA, 2006). However, enrichment of the radioactive nuclides in residue wastes is a problem in incineration. In Japan, wastes that contain ^{134}Cs and ^{137}Cs concentration exceeding 8000 Bq/kg activity is referred to as “specified radioactive waste”, which provides a safety limit of exposure to workers that does not exceed 1 mSv/y (Takeda and Kimura, 2012). Although, approximately 80% of these specified wastes consisted of incinerated ash

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and inorganic soils, the other 20% were combustible waste; i.e., sewage sludge, agricultural waste and MSW to be treated (Ministry of Environment, Government of Japan, 2014b).

Kawano et al. (2013) examined the incineration of refuse-derived fuel (RDF) containing only ^{133}Cs as Cs_2CO_3 and CsCl in laboratory and pilot-scale experiments. In the laboratory, using an electric furnace, RDF manufactured from MSW in other facilities was incinerated at 800 °C or 1000 °C. When the combustion temperature was increased to 1000 °C from 800 °C, the ratio of Cs in the waste decreased in both samples. This experiment examined only the effect of combustion temperature, not that of the equivalence ratio (ER). ER is one of most important factors for determining the efficiency of incineration and is defined as the ratio of the actual amount of air to the theoretical amount of air required for combustion (Andrés et al., 2011). Furthermore, radioactive nuclides in waste can concentrate in the resulting ash (fly ash and bottom ash) of incineration, and may be in different chemical forms in either ash. The chemical forms of Cs in ash are important for the determination of appropriate treatment and stabilization of ashes, but there are little information about it.

Some researchers used ^{133}Cs at high concentrations to predict the behavior of radioactive Cs in the incineration process (Amiro et al., 1996; Kawano et al., 2013; Saffarzadeh et al., 2014) and in the leaching from ash (Li et al., 2013). However some waste contains 8000 Bq/kg activity of ^{137}Cs equivalent to 2.5 ng/kg ^{137}Cs , making a direct analysis of Cs concentrations difficult.

Our study attempts to clarify the behavior of radioactive nuclides in waste incineration. Specifically, RDF with added ^{133}Cs (stable nuclide) was incinerated, and the changes in the distribution of ^{133}Cs to the bottom ash and fly ash in relation to the equivalence ratio (ER) and the combustion temperature were investigated. We compare results from a laboratory experiment using a tubular furnace and a pilot-scale experiment using a rotary kiln-stoker furnace. In addition, RDF with added ^{134}Cs (γ -ray radioactive nuclide) was incinerated at 800 °C and 900 °C in a laboratory experiment to clarify the behavior of radioactive nuclides and compare the tendencies of ^{134}Cs and ^{133}Cs . Finally, a thermogravimetric (TG) and differential thermal analysis (DTA) of stable Cs compounds and an X-ray absorption fine structure (XAFS) analysis of Cs concentrated in the ashes were performed to validate the behavior and chemical forms of Cs during the combustion.

2. Materials and methods

2.1. Materials

RDF pellets used in this study were manufactured from MSW from a plant located in west Japan. RDF was incinerated as pellets in the pilot-scale experiment, but in the laboratory experiment, RDF pellets were powder-milled and sieved under 2 mm for

Table 1
Elemental compositions of RDF.

Elements	This study ($n = 3$)		Reference values		
	Mean	S.D.	RDF ₁ ^a	RDF ₂ ^b	RDF ₃ ^b
C	41.2	0.346	41.7	46.6	41.7
H	5.79	0.153	6.00	6.80	5.00
N	1.35	0.176	0.90	1.28	0.75
O	40.8	0.240	36.3	34.5	36.3
S	0.21	0.006	0.17	0.13	0.17
Cl	0.801	0.030	1.20	1.08	1.00
Ca	4.52	0.110	0.25	2.67	2.7

S.D.: Standard deviation.

^a Kobayashi et al. (2005).

^b Piao et al. (2000).

Table 2
Experimental conditions.

	Temperature (°C)	ER	Nuclides	Concentration	
Laboratory experiment	900	2.2	Cs-133	5100 mg/kg	
		1.7			
		1.4			
	800	1.4	Cs-134	1.9×10^7 Bq/kg	
		2.2	Cs-133	5100 mg/kg	
		1.7			
800	1.4	Cs-134	1.9×10^7 Bq/kg		
		Cs-133	5100 mg/kg		
	700	2.2	Cs-133	5100 mg/kg	
		1.7			
1.4					
	Pilot-scale experiment	700	2.2	Cs-133	5100 mg/kg
			1.7		
1.4					

ER: Equivalence ratio.

homogenization of the sample. The composition of the RDF was analyzed with a carbon, hydrogen, and nitrogen elemental analyzer (Micro Corder JM10, J-Science Lab, Kyoto, Japan) and X-ray fluorescence spectrometry (XRF-1800, Shimadzu, Kyoto, Japan). The results are shown in Table 1. Dried RDF was measured three times with the use of each piece of instrumentation. RDF₁ (Kobayashi et al., 2005), RDF₂, and RDF₃ (Piao et al., 2000) are the elemental components of RDF manufactured from MSW in previous research. The four types of RDF were similar in composition, although the RDF used in this study had a higher Ca concentration than RDF used in previous studies.

An aqueous solution containing nuclides was added to the RDF, and this sample was adjusted to 30% moisture content. The nuclide concentration in the RDF was 5100 mg/kg for ^{133}Cs or 1.9×10^7 Bq/kg for ^{134}Cs . ^{133}Cs and ^{134}Cs were added as Cs_2CO_3 (Wako Pure Chemical Industries, Ltd, 1st grade), and CsCl (Japan Radioisotope Association, CZ010-0012), respectively (Table 2).

2.2. Laboratory experiment

A tubular furnace ($\phi = 45$ mm, $L = 1000$ mm) was used in the laboratory experiment (Fig. 1). Highly purified air (O_2 21%, N_2 79%; Air-Zero-A, Sumitomo Seika Chemicals, Osaka, Japan) flowed into the furnace at 200 mL/min. A sample was placed on a porcelain crucible and set in the center of the tubular furnace. Nuclides in the emission gas were collected in glass wool at the end of the quartz tube and in two bottles of collection liquid (5 mol/L: HNO_3 water solution) behind the quartz tube (Nakayama et al., 2006).

In the experiments using ^{133}Cs , the combustion temperatures were 700, 800, and 900 °C. The ER was modified to 2.2, 1.7, and 1.4 for each temperature by varying the amount of sample RDF. In the experiments using ^{134}Cs , the combustion temperatures were 800 and 900 °C, and the ER was 1.4 fixed. In these experiments, the incineration time was 50 min determined by preliminary examination to achieve complete combustion. In the ^{133}Cs experiments, each test was conducted twice, and ^{133}Cs analysis was conducted

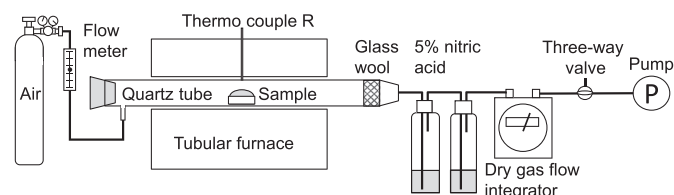


Fig. 1. Schematic of the tubular furnace used in the laboratory experiment.

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