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Behavior of radioactive cesium during incineration of radioactively contaminated wastes from decontamination activities in Fukushima

Hiroshi Fujiwara [a, b], Hidetoshi Kuramochi [a, b, *], Kazutaka Nomura [a, 1], Tomoharu Maeseto [a], Masahiro Osako [a]

[a] Center for Material Cycles and Waste Management Research, National Institute for Environmental Studies, Onogawa 16-2, Tsukuba, Ibaraki 305-8506, Japan

[b] Department of Risk Management and Environmental Sciences, Yokohama National University Graduate School of Environment and Information Sciences, Tokiwadai 79-1, Hodogaya, Yokohama, Kanagawa 240-8501, Japan

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ABSTRACT

Large volumes of decontamination wastes (DW) generated by off-site decontamination activities in Fukushima Prefecture have been incinerated since 2015. The behavior of radioactive cesium during incineration of DW was investigated at a working incineration plant. The incineration discharged bottom ash (BA) and fly ash (FA) with similar levels of radiocesium, and the leachability of the radiocesium from both types of ash was very low (<1%). These results are significantly different from those obtained for the incineration of contaminated municipal solid waste (CMSW) reported in earlier studies. The source of radiocesium in DW-FA is chiefly small particles derived from DW and DW-BA blown into the flue gas, not the deposition of gaseous synthesized radiocesium compounds on the surfaces of ash particles in the flue gas as observed in CMSW incineration. This source difference causes the behavior of radiocesium during waste incineration to differ between DW and CMSW.

1. Introduction

After the accident at the Fukushima Daiichi nuclear power plant on 11 March 2011, a huge amount of radioactive cesium (r-Cs) was found to have contaminated the area around the plant. As a result, not only the environment but also ordinary municipal solid wastes became radioactively contaminated. Earlier studies on the incineration of contaminated municipal solid waste (CMSW) have revealed two important facts: 1) r-Cs in CMSW is mainly concentrated in fly ash (FA) (Kuramochi et al., 2016; Osako et al., 2012; Oshita et al., 2015), and 2) although the leaching rate of r-Cs from CMSW-FA is high, ranging from 64% to 97%, most r-Cs in the bottom ash (BA) from CMSW is not soluble in water (Iwahana et al., 2013; Kuramochi et al., 2016; Osako et al., 2012; Parajuli et al., 2013; Sakanakura et al., 2012). However, the leachability of r-Cs from FA differs among different types of contaminated wastes: its leachability from FA resulting from wood incineration is as high as that from CMSW-FA (Parajuli et al., 2013). In contrast, r-Cs in ash from the incineration of sewage sludge does not generally leach into water (Parajuli et al., 2013; Sakanakura et al., 2012; Tsushima et al., 2013). These results indicate that the chemical form of r-Cs and its leachability from FA depend on the type of waste feedstock that is being incinerated. Recently, it has been found that high leachability of r-Cs from CMSW-FA may trigger secondary contamination, and, thus, various techniques for immobilization or removal of r-Cs in CMSW-FA have been examined (Awual et al., 2014; Jing et al., 2014; Mallampati et al., 2014; Namiki et al., 2014; Yang et al., 2014a).

In Fukushima Prefecture's Special Decontamination Area, decontamination activities are being performed to remove large amounts of contaminated soil and vegetation and decrease the radiation dose level. Contaminated soil is transported to an interim storage facility in Fukushima, and flammable decontamination wastes (DW) are incinerated or melted to reduce their mass and volume. Depending on its r-Cs contamination level, the DW ash is stored at an interim storage facility or disposed of in a leachate-controlled type of landfill site. To be able to formulate suitable storage and disposal plans that take into account the impact on the

* Corresponding author. Center for Material Cycles and Waste Management Research, National Institute for Environmental Studies, Onogawa 16-2, Tsukuba, Ibaraki 305-8506, Japan.
E-mail address: kuramochi.hidetoshi@nies.go.jp (H. Kuramochi).
[1] Hitachi Power Solutions Co., Ltd., Horiguchi 832-2, Hitachinaka, Ibaraki 312-0034, Japan (present address).

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Abbreviations

DW	decontamination wastes
CMSW	contaminated municipal solid waste
BA	bottom ash
FA	fly ash
r-Cs	radioactive cesium
LHV	lower heating value
CV	coefficient of variation
SAP	steam air preheater ash
GCT	gas cooling tower ash

environment and human exposure to radiation, it is of key importance to understand the behavior of r-Cs, such as its distribution between FA and BA during the incineration of DW and the leachability of r-Cs from both FA and BA. These behaviors have not been reported in the literature. Because leachability depends on the type of feedstock, published information on CMSW incineration may not be relevant to DW incineration. Therefore, a working incineration plant that handles DW was investigated. The focus of this study was the behavior of r-Cs, and the safe management of incineration ash is also discussed. Furthermore, differences between DW and CMSW incineration, in particular with regard to the FA formation mechanism, were examined.

2. Materials and methods

2.1. Sampling and sample preparation

A working incineration plant equipped in series with a stoker furnace, steam air preheater, gas cooling tower, and two baghouses used exclusively for DW was investigated over two days in August 2015 (see Fig. 1). The temperature of the secondary combustor was 870–882 °C during the investigation period, similar to the temperature generally used for municipal solid waste incineration. The areas where DW and incineration residues were sampled were designated as radiation controlled areas. Therefore, all sampling was conducted by radiation workers under proper radiation controls in accordance with the Ordinance on Prevention of Ionizing Radiation Hazards (Ministry of Health, Labour and Welfare, 2015). For example, the air dose rate was monitored with a NaI scintillation survey meter, and suspended particles in air were collected on the filter of a high-volume air sampler during sampling at the work

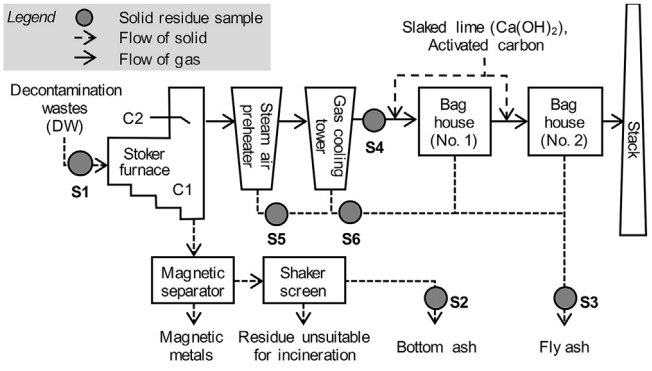

Fig. 1. Schematic diagram of the incineration plant. The stoker furnace has a primary combustion chamber (C1) and a secondary combustion chamber (C2). Sampling points: S1, decontamination wastes in the stock yard; S2, bottom ash; S3, fly ash; S4, fly ash before the addition of slaked lime and activated carbon; S5, steam air preheater ash; S6, gas cooling tower ash.

place. Then the concentration of radioactivity on the filter was measured with a Ge semiconductor detector. Collected DW and ash samples were transported to a non-radiation controlled area after surface contamination density had been surveyed with a Geiger-Müller survey meter. Five DW samples were collected at different points in the stock yard (sampling point S1; Fig. 1) and mixed together to prepare a composite sample for characterization. BA was collected at the plant eight times per day, and FA was collected four times per day. Both BA and FA (corresponding to sampling points S2 and S3 in Fig. 1) without any treatment were collected from the top of their respective storage silos by inserting a cylindrical tube with a semi-round aperture (aperture area ca. 10 cm × 10 cm). BA was collected before humidification, and FA was collected before chelate treatment. Particle size distributions of BA and FA are shown in Fig. A1. There was no significant difference in size distribution between MSW-FA and DW-FA, whereas DW-BA was smaller than MSW-BA (Chandler et al., 1997). Steam air preheater ash and gas cooling tower ash (corresponding to sampling points S5 and S6; Fig. 1) were collected three times per day with a vacuum sweeper from the conveyer belt that transported the ash to the FA storage silo. All ash samples collected at the same point during the same day were mixed to make a daily composite sample, except for the time series of r-Cs concentrations shown in Fig. A2. In the studied facility, slaked lime and activated carbon for neutralization of flue gas and adsorption of dioxins are mixed in the supply line and then added continuously before baghouse No. 1 and several times per day before baghouse No. 2. Hence, ash was collected from the flue gas at S4 (Fig. 1), before these reagents were blown in, for the elemental analysis of FA. A sample was collected at S4 once daily by a method for measuring dust concentration in flue gas (JIS Z 8808; JIS, 2013). For this sampling, a sampling probe was first inserted, pointing upstream, into the flue gas at S4, and then the gas was trapped by a filter paper thimble in the sampling probe. Because the measured r-Cs concentration at S4 was based on flow volume, the S4 values were not comparable to r-Cs concentration measurements of solid residues sampled at S2 and S3. DW analysis results were compared with those of a similar investigation conducted in December 2012 at an incineration plant for CMSW, which was equipped with a stoker-type furnace as well as an ash-melting furnace (hereafter referred to as facility A, as in an earlier study (Kawamoto et al., 2013)). In their investigation, CMSW-BA was collected after the removal of magnetic metals and residue unsuitable for melting and before it was passed to the ash-melting furnace. CMSW-FA was collected at the baghouse, after activated carbon had been blown into the flue gas but before slaked lime was added. In the present study, samples from facility A supplied by Dr. Kawamoto were analyzed along with DW samples.

2.2. Decontamination wastes

The characteristics of the DW composite samples on each sampling day (Days 1 and 2) are summarized in Table 1. The lower heating value (LHV) of DW was measured by a bomb calorimetric method (JIS M 8814; JIS, 2003) with a bomb calorimeter (1013-U, Yoshida Seisakusyo Co., Ltd., Tokyo, Japan). The appearance of DW (Fig. A3) is quite different from that of the municipal solid waste or wood chips that are ordinarily treated in incineration facilities. DW looks like composted vegetation because the collected vegetation decayed during storage. The weight fraction of combustibles in DW on both days was only about 17%, with the result that the LHV of DW on Day 1 and Day 2 was 2320 and 1530 kJ/kg, respectively. Waste can be burned without fuel only if its LHV exceeds 5000–7000 kJ/kg or the combustibles content of the waste is more than 25% (Lidia et al., 2015). Therefore, the incineration of DW requires the use of fuel or some other waste with a high caloric value.