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Influence of the type of furnace on behavior of radioactive cesium in municipal solid waste thermal treatment

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ABSTRACT

Municipal solid waste (MSW) contaminated by radioactive cesium (r-Cs) has been incinerated since the Fukushima Daiichi Nuclear Power Plant accident. Eight thermal treatment plants with four different types of furnaces were comprehensively investigated to provide fundamental data to improve our understanding of the behavior of r-Cs in various types of MSW thermal treatment facilities. R-Cs tended to distribute to the fly ash (FA) more than to the residue from the bottom of the furnace (bottom ash, incombustibles or slag). The r-Cs concentrations in the FA depended on the type of furnace and followed the order; fluidized-bed incinerator < stoker type incinerator < gasification melting furnaces. Shaft-type gasification melting furnace separated r-Cs selectively into FA and simultaneously discharged decontaminated slag. The leaching rate of r-Cs from FA was high, 30–100%, and independent of the type of furnace, whereas r-Cs in the residue from the bottom of the furnace scarcely dissolved in water. Heat recovery ash e.g. gas cooler ash was characterized by intermediate r-Cs concentrations and leachabilities compared with bottom residue and FA in stoker type and fluidized-bed incinerator. In the case of shaft-type gasification melting furnace, however, heat recovery ash showed similar property to FA due to a cyclone followed by heat recovery process. We evaluated whether baghouses (air- pollution control equipment) successfully removed r-Cs from flue gas. In all cases, r-Cs in flue gas was below the limit of detection after baghouse. We concluded that different types of furnaces affected r-Cs distributions, but flue gases from baghouse systems of all types of furnaces were safe.

1. Introduction

Huge amounts of radioactive substances were released as a result of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident on 11 March 2011. The wide-scale contamination of eastern Japan that occurred because of the fallout of radioactive cesium (r-Cs) resulted in the generation of radioactively contaminated waste. In Japan, a densely populated country, thermal treatment of municipal solid waste (MSW) such as incineration and ash melting have played a significant role in sanitary waste treatment. Such treatment greatly reduces the mass and volume of the waste and can extend the lifetime of landfills. More than 80% of MSW in Japan is incinerated (MOE, 2017). Stoker-type incinerators equipped with fire grates account for 76% of thermal treatment plants in Japan, and several other types of furnaces exist (Fig. A.1) (MOE, 2017).

After the FDNPP accident, r-Cs was found to be highly concentrated in MSW thermal treatment residue because of the reduction in mass and volume effected by thermal treatment. Thermal treatment residues contaminated by r-Cs were found at many MSW incineration facilities in eastern Japan (MOE, 2011a) and were even identified in Kyusyu, 1200 km southwest of the FDNPP (Iwahana

Abbreviations: r-Cs, radioactive cesium; MSW, municipal solid waste; FA, fly ash; BA, bottom ash; Stoker, stoker-type incinerator; FBI, fluidized-bed incinerator; FB-MF, fluidized-bed gasification with melting furnace; S-MF, shaft-type gasification melting furnace; BH, baghouse.

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et al., 2013). Understanding the behavior of r-Cs during thermal treatment is necessary to prevent the secondary widespread r-Cs contamination due to thermal treatment and to reduce radiation exposure. The primary concern associated with thermal treatment of radioactive MSW is the possibility that wide diffusion of r-Cs discharged from stacks may result in re-contamination of surroundings by the r-Cs. In Japan, radiation dose limits of r-Cs for the air in public places have been set to satisfy the requirement that the additional dose be less than 1 mSv/year, even if the same person were to breathe the air from age 0 to 70 years. Based on this additional radiation dose limit, the concentration of r-Cs in flue gas from stacks must satisfy the criterion shown in Eq. (1) since the FDNPP accident. This criterion has always been used as the radiation dose limit of r-Cs for air in public places. Because flue gas is generally diluted with air by several orders of magnitude, this strict regulation assures the safety of the public.

$$\frac{\text{concentration of Cs-134 (Bq/m}^3\text{)}}{20(\text{Bq/m}^3)} + \frac{\text{concentration of Cs-137 (Bq/m}^3\text{)}}{30(\text{Bq/m}^3)} \leqq 1 \quad (1)$$

Thus, safety has been assured through the monitoring of r-Cs concentrations at the stacks (MOE, 2012). However, nobody knows if there is any difference in removal efficiency of r-Cs from flue gas among various furnaces because of a relatively high detection limit such as 2 Bq/m^3 other than investigation of stable Cs (Cs-133) (Yoneda et al., 2014). Osako et al. (2012) evaluated the r-Cs removal performance of flue gas treatment equipment at several stoker incineration facilities soon after the FDNPP accident. They concluded that use of air pollution control equipment such as a baghouse (BH) for contaminated MSW satisfactorily removed r-Cs and prevented the spread of r-Cs via flue gas into the atmosphere. Unfortunately, few studies have been conducted to evaluate the r-Cs removal performance of MSW incinerators other than stoker type incinerators. It is therefore necessary to evaluate the performance of flue gas treatment at other types of MSW incinerators. Especially, this information is very important for predicting safety of thermal treatment dealing with highly contaminated waste.

Laboratory and pilot-scale incineration tests involving the addition of high concentrations of non-radioactive cesium compounds such as CsOH or Cs_2CO_3 have also been conducted and have provided valuable information. Oshita et al. (2015) found that Cs tends to move to the fly ash (FA) as the combustion temperature and air equivalence ratio increase. Saffarzadeh et al. (2014) have investigated the chemical form of Cs in bottom ash (BA) obtained from pilot-scale stoker-type incineration plant and Cs were found in the matrix glass phase. As far as chemical form of Cs in FA, both experimental results (Shiota et al., 2015) and thermodynamic equilibrium calculations (Kuramochi et al., 2016) have indicated that r-Cs is present as cesium chloride (CsCl) in FA. CsCl is thought to be formed via $CsCaCl_3$ under high-temperature conditions (Jiao et al., 2016). These studies have provided useful understanding of the basic behavior of r-Cs, but investigations at real facilities are needed to confirm these results because real facilities are affected by additional factors such as the type of furnace, equipment layout, and operating conditions.

R-Cs concentrations in thermal treatment residue have been monitored and are publicly available on the website of each municipality. The existence of several types of thermal treatment plants means that several kinds of residues are being discharged. The effects of the types of furnaces on r-Cs concentrations in incineration residues have been compared by analyzing publicly available data (NIES, 2015). That study revealed that r-Cs concentrations in FA differed between different types of furnaces. The differences could be explained on the basis of the different amounts of FA discharged from the furnaces and different proportion of the r-Cs to be distributed to the FA. Some field surveys of the behavior of r-Cs at actual thermal treatment facilities have also been performed, and Kuramochi et al. (2016) have reviewed the results in terms of r-Cs distributions in FA and bottom residues as well as leaching rates from some types of thermal treatment plant residues. However, we consider that more detailed analyses are needed with respect to the following. (1) Comparisons of the behavior of r-Cs in plants with different types of furnaces, especially plants with shaft-type gasification melting furnaces, which have not been studied before. (2) Detailed analyses of all thermal treatment residues, including minor products such as metals and heat recovery ash are insufficient. The latter may be suspended or precipitated during heat recovery processes in each system component such as the boiler, economizer, and gas cooler. We therefore carried out a comprehensive investigation by collecting samples simultaneously at a number of thermal treatment facilities. We investigated the behavior of r-Cs in different types of furnaces, the relationship between r-Cs concentrations and amounts of thermal treatment residues, and r-Cs leaching properties. We also tried to evaluate the removal efficiencies of r-Cs by BHs.

2. Material and methods

2.1. Sample collection

We investigated eight commercial thermal treatment plants that processed contaminated MSW. Table 1 provides an overview of the investigation and the characteristics of each facility. Some published data were available for facility F (Harada et al., 2014). Investigations were performed over multiple days. We chose MSW thermal treatment plants with four different types of furnaces. Fig. A.2 is a schematic diagram of the sampling points at each facility. Every facility was equipped with a BH flue gas treatment system. The characteristics of the thermal treatment residues discharged from the plants depended on the type of furnace. The stoker-type incinerators (Stoker) (facilities A–D) discharged mainly two kinds of ash—incineration FA, which is dust or small ash particles in the flue gas collected mainly at BH; and bottom ash (BA), which is discharged from the bottom of the furnace along with the moving fire grate. The fluidized-bed incinerators (FBIs) (facilities E and F) generated incineration FA, and incombustibles such as metals and rubble were discharged from the bottom of the furnace instead of the BA. The fluidized-bed gasification with melting furnace (FB-MF; facility G) and shaft-type gasification melting furnace (S-MF; facility H) discharged slag instead of BA and produced a kind of FA called melting furnace FA. These thermal treatment residues (incineration FA, melting furnace FA, BA, incombustibles, and slag) were collected and are henceforth referred to as major thermal treatment residues. We also tried to collect minor thermal treatment residues such as metals (e.g., iron, aluminum) and heat recovery ash (e.g., gas cooler ash and boiler dust). In addition, fluidized-bed sand, which usually circulates in the furnace at FBIs, was also collected at facility E. Basically, all the samples were collected multiple times on the same day, and then individual samples were stored separately in airtight sealed bags to prevent exposure to the atmosphere and absorption of moisture. Composite samples were made by mixing samples of the same kind prior to analysis. As shown in Fig. A.2, minor residues were blended with major residues and then discharged together as a mixture of ash during actual operations at the facilities. Heat recovery ash was mixed with FA in our investigation, although heat recovery ash can be blended with either bottom residue or FA at thermal treatment facilities. Thermal treatment