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Research article

Evaluation of physicochemical properties of radioactive cesium in municipal solid waste incineration fly ash by particle size classification and leaching tests

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ABSTRACT

After the Fukushima Daiichi-Nuclear Power Plant accident, environmental recovery was a major issue because a considerable amount of municipal solid waste incineration (MSWI) fly ash was highly contaminated with radioactive cesium. To the best of our knowledge, only a few studies have evaluated the detailed physicochemical properties of radioactive cesium in MSWI fly ash to propose an effective method for the solidification and reuse of MSWI fly ash. In this study, MSWI fly ash was sampled in Fukushima Prefecture. The physicochemical properties of radioactive cesium in MSWI fly ash were evaluated by particle size classification (less than 25, 25–45, 45–100, 100–300, 300–500, and greater than 500 μm) and the Japanese leaching test No. 13 called "JLT-13". These results obtained from the classification of fly ash indicated that the activity concentration of radioactive cesium and the content of the coexisting matter (i.e., chloride and potassium) temporarily change in response to the particle size of fly ash. X-ray diffraction results indicated that water-soluble radioactive cesium exists as CsCl because of the cooling process and that insoluble cesium is bound to the inner sphere of amorphous matter. These results indicated that the distribution of radioactive cesium depends on the characteristics of MSWI fly ash.

1. Introduction

Considerable amounts of radionuclides originating from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident were released into the environment. Both ^{134}Cs ($T_{1/2} = 2.06$ years) and ^{137}Cs ($T_{1/2} = 30.07$ years) are serious environmental pollutant because of their relatively long half-lives, respectively. The amount of released radioactive cesium in the environment has been estimated as 11.8–18 PBq (^{134}Cs) and 13–62.5 PBq (^{137}Cs), respectively (Steinhauser et al., 2014). The released radioactive cesium falls on the ground surface via dry and wet deposition. Such deposited radioactive cesium is stable because it binds to the inner sphere of clay minerals in the soil (Qin et al., 2012). As mentioned physicochemical properties such as the solubility and pH-dependence, it is necessary to monitor the long-term environmental behavior of radioactive cesium.

The contaminated fallen leaves, soils, and vegetation were collected as part of the decontamination works for environmental recovery. Collected samples were incinerated at an incineration plant to reduce their volume. The produced municipal solid waste incineration (MSWI) ash was classified into cinder called "bottom ash" and soot called "fly ash." Finally, both ashes were disposed at landfill. The Japan Ministry of the Environment (2011) has reported that the activity concentration of radioactive cesium in MSWI fly ash was higher than that in MSWI bottom ash. The activity concentration of radioactive cesium in fly ash was greater than the regulatory limit (8000 Bq kg^{-1}) at the plants in a high-dose area (Ministry of Environment, 2011).

After the FDNPP accident, studies reported on radioactive nuclides including radioactive cesium in MSWI fly ash were focused on its speciation (Saffarzadeh et al., 2014; Tojo et al., 2014; Iwahana

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et al., 2015; Shiota et al., 2015; Fujii et al., 2016). Shiota et al. (2015) used synchrotron radiation micro-X-ray analysis and revealed that stable Cs in the MSWI dust is similar to that of CsCl. Tojo et al. (2014) reported the presence of insoluble radioactive cesium in the amorphous matter in bottom ash. Generally, the leaching behavior of radioactive cesium in MSWI fly ash depends on its physicochemical properties such as solubility. However, to the best of our knowledge, only a few studies have reported the physicochemical properties of radioactive cesium in MSWI fly ash. Owing to the sequential extraction based on the method proposed by Tessier et al. (1979), approximately 60 percent of radioactive cesium in the fly ash became soluble form in water (Fujii et al., 2016; Ohbuchi et al., 2016).

Considering the demand for the reuse of a considerable amount of wastes, it is necessary to investigate the physicochemical properties for the purpose of evaluating the toxicity of not only radioactive cesium but also other heavy metals. Some studies focused on the physicochemical properties of heavy metals in fly ash (Okada and Matsuto, 2009; Chen et al., 2016; Raclavská et al., 2017; Li et al., 2017).

In this paper, the particle size distribution and physicochemical properties of radioactive cesium and other heavy metals in MSWI fly ash were investigated by a dry sieving method. After the size classification of that, the Japanese leaching test No. 13 (JLT-13) for industrial waste was carried out to evaluate the distribution of water-soluble radioactive cesium. The results obtained herein provide a good approach for understanding the adsorption behavior of radioactive cesium on the coexisting matter in MSWI fly ash.

2. Materials and methods

2.1. Sample preparation

The MSWI fly ash sample was collected in Fukushima Prefecture, Japan, in January 2013. The combustion capacity of plant is 300 ton day^{-1} with two stoker furnaces in the plant, where ordinary MSW is combusted. The fly ash was dried at 105 °C for 24 h before experiments. The activity ratio of $^{134}Cs/^{137}Cs$ in all samples was decay-corrected on March 11, 2011. An activity ratio of 1 has been reported previously in Fukushima-derived radionuclides $^{134}Cs/^{137}Cs$ (Komori et al., 2013). The behavior of ^{134}Cs is similar to that of ^{137}Cs (Saffarzadeh et al., 2014). Gamma-ray spectrometry results indicated that radioactive cesium in the fly ash originates from the FDNPP accident. Iwahana et al. (2015) reported that the activity concentration of ^{40}K in fly ash, corresponding to the natural radionuclides sampled before the FDNPP accident, ranges from 445 to 2600 Bq kg^{-1}. The activity concentration of ^{40}K in all fractions ranges from 634 to 2347 Bq kg^{-1}. The activity concentration of ^{40}K in the fly ash used herein was equivalent to that before the accident. Five hundred grams of fly ash sample was classified into 6 fractions (i.e., less than 25, 25–45, 45–100, 100–300, 300–500, and greater than 500 μm, respectively) by dry sieving. The stereoscopic microscope images of the classified samples are shown in Fig. 1. The cinder form was observed for large particles with a size of greater than 100 μm.

2.2. Gamma-ray spectrometry

The activity of radioactive cesium was determined by gamma-ray spectrometry using a p-type high-purity Ge/coaxial-type semiconductor detector (HPGe; IGC 10200, Princeton Gamma Tech Instruments, Inc., USA), which was surrounded by a 100-mm-thick lead shields with additional 50-mm-thick oxygen-free copper and 5-mm-thick acrylic plates. Each of the classified fly ash samples was compressed into the U8 container (height and diameter of 68 and 56 mm, respectively, Sekiya Rika Co. Ltd, Japan). The activity of radionuclides in samples was measured by gamma-ray spectrometry. The detection efficiency curve was drawn using a ^{152}Eu standard source (diameter and height of 25 and 6.0 mm, respectively, Japan Radioisotope Association, Japan). The calibration curve was corrected using the activity of 1461 keV gamma ray emitted from ^{40}K in KCl (Koike et al., 2017). The activities of ^{134}Cs and ^{137}Cs were determined by the emitted gamma rays, i.e., 606 and 662 keV, respectively.

2.3. X-ray spectrometry

X-ray diffraction (XRD) was employed to measure the crystalline phases of samples. The XRD system (SmartLab, Rigaku Co., Japan) was equipped with a 2 kW (Cu) X-ray tube (operated at 40 kV and 50 mA). A silicon drift detector was used as the detector. A Bragg–Brentano focusing optical system was used for data collection. The intensity of each peak was recorded from 5° to 90°/2θ in steps of 0.01° with a measurement speed of 0.5 s per min. Phase identification and quantitative analysis by Rietveld refinement were carried out using an integrated X-ray powder diffraction software (PDXL, Rigaku Co., Japan). The major and minor elements in fly ash were analyzed using an X-ray fluorescence spectrometer (XRF, ZSX Primus IV, Rigaku Co., Japan), which was equipped with an end window containing a 4 kW Rh X-ray tube. The analyzing crystals were LiF (200), Ge (111), PET (002), and RX25. A proportional counter flowed PR gas comprising Ar (90%) and CH_4 (10%) and scintillation counter were used as detectors. The PR gas flow rate was 50 cm^3 min^{-1}.

2.4. Japanese leaching test No.13 for MSWI material

The Japanese leaching test No. 13 (JLT-13) for industrial waste is one of the most rapid and easy method for determining the leachability of heavy metals from various types of wastes at a waste landfills. Sakai et al. (1995) reported the comparison of the leaching potential of heavy metals between JLT-13 and other availability test. JLT-13 has been employed to determine the leachability of radioactive cesium from various types of waste at a waste landfills (Pariatamby et al., 2006; Padmi et al., 2009; Harada et al., 2011; Oguchi et al., 2012). Therefore, it is effectively applicable for contaminated MSWI fly ash including radioactive materials. In the JLT-13, a mixture of pure water and classified fly ash (S/L = 0.1) was subjected to continuous shaking for 6 h at room temperature in a shaker. A mechanical shaker (Shaking Bath TBK 602DA, Advantec Inc., Japan) was used to prepare eluted solutions from fly ash. The velocity of the shaker was approximately 200 rpm (round per minutes). After shaking, the eluted solutions were separated from the classified fly ash using a filter paper (Whatman™ glass microfiber filters, 100 mmϕ, GE Healthcare Life Science, USA). The eluted solutions and residue powders were compressed into the U8 container and measured using HPGe. The distribution of the activities of radioactive cesium in the eluted solutions and residues were determined on the basis of the sum of that before extraction.

3. Results and discussion

3.1. Particle size distribution of MSWI fly ash including radioactive cesium

The distribution of the activity concentration of radioactive cesium and sample weight ratio in each fraction are shown in Fig. 2. The activity concentration of radioactive cesium in fly ash with a particle size of less than 500 μm (between 4000 and 5000 Bq kg^{-1})