Chemical state analysis of heavy metals and radioactive cesium in municipal solid waste incineration fly ash contaminated with radioactive cesium released by the FDNPP accident

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Abstract

The chemical states of heavy metals and radioactive Cs were estimated in fly ash sampled at Fukushima prefecture, Japan. Estimating the speciation of incinerator fly ash is important to ensure appropriate and efficient management of fly ash generated from disaster-related waste. In this study, fly ash collected at a waste incineration facility in Fukushima prefecture was treated using a sequential extraction test. The test results indicated that the solubility behavior of radioactive Cs was similar to that of NaCl and KCl, and approximately 60% of radioactive Cs was included as water-soluble chloride compounds in the fly ash sample. Most heavy metals eluted in three fractions, in the extraction steps for carbonate-bound, free oxide, and bound to organic matter species. The chemical state of elements in the three non-water-soluble fractions and residue showed minimal elution into the environment. Therefore, most heavy metals in fly ash exhibited minimal elution into the environment.

Keywords: municipal solid waste incineration fly ash, chemical states, sequential extraction test, heavy metals, radioactive Cs
Introduction

Considerable amounts of radioactive Cs ($^{134}\text{Cs}$: $T_{1/2} = 2.06$ y and $^{137}\text{Cs}$: $T_{1/2} = 30.07$ y) were released into the environment by the Fukushima Daiichi-Nuclear Power Plant (FDNPP) accident. Radioactive Cs has been intensively monitored because it has a relatively long half-life and high biological availability. During the ten years since the FDNPP accident, radioactive Cs have been analyzed for various environmental samples (e.g., soil, river water, seawater, sediment, agricultural and livestock products, marine products, and municipal solid waste (MSW)). Previous studies have also detected radioactive Cs in MSW incineration (MSWI) fly ash.\textsuperscript{1-4} Chemical state analysis is particularly useful information for estimating the behavior of radioactive Cs in MSWI fly ash contaminated with radioactive materials released by the FDNPP accident. Ministry of Environment in Japan\textsuperscript{5} has reported that the activity concentration of radioactive Cs in fly ash as soot was higher than that of bottom ash as incineration residue. In Japan, wastes containing radionuclides exceeding the regulatory limit (8000 Bq kg$^{-1}$) are classified as “specified radioactive waste”, and the exposure value of the radiation dosage for workers is limited to 1 mSV in a year.\textsuperscript{6} Although, approximately 80% of these specified wastes consisted of incinerated ash and inorganic soils, the other 20% was a combustible waste; i.e., sewage sludge, agricultural waste, and MSW to be treated.\textsuperscript{7} The activity concentration of radioactive Cs in fly ash was greater than the regulatory limit at the plants in a high-dose area\textsuperscript{5} owing to the concentration of radioactive Cs from MSW in fly ash. As discussed previously, fly ash that is within the regulatory limit is deposited at landfill sites. It is reported that radioactive Cs in fly ash deposited at landfill sites was eluted into the environment.\textsuperscript{8} To evaluate the environmental risk, it is necessary to investigate the leaching behavior and chemical
state of radioactive Cs in fly ash. Considerable research regarding the speciation of radionuclides, including radioactive Cs, in MSWI fly ash was reported.\(^9\)\(^{15}\) The leaching behavior of heavy metals in addition to radioactive Cs in MSWI fly ash depends on their chemical state and physicochemical properties such as solubility. The particle size distribution, determined via a dry sieving method, and physicochemical properties of radioactive Cs and heavy metals in MSWI fly ash were investigated.\(^14\) After particle size classification, the Japanese leaching test No. 13 (JLT-13) was carried out to evaluate the distribution of water-soluble forms.\(^14\) Ohbuchi et al.\(^15\) reported that radioactive Cs in MSWI fly ash was separated into soluble and insoluble forms.

Incineration processes are commonly employed for the treatment of MSW due to its advantages of mass and volume reduction, energy recovery, and sterilization of hazardous components.\(^16\) Finally, bottom ash and fly ash were disposed at landfill sites. Typically, the concentration of heavy metals, such as Cr, Ni, Cu, Zn, and Pb, in fly ash are higher than in the corresponding bottom ash. Therefore, fly ash is categorized as a “specially controlled general and industrial waste” under Japanese regulations. Both Japan and other countries require that fly ash must be disposed of carefully, because some of the hazardous materials in fly ash are leached at the disposal site. Therefore, the concentrations of hazardous materials are strictly controlled. It is also important to evaluate the leaching behavior of heavy metals in fly ash and suppress heavy metal leaching. Safety management of MSWI fly ash contaminated with radioactive materials released by the FDNPP accident and prevention of environmental pollution due to elution of harmful substances are required. Therefore, it is important to investigate the chemical behavior of heavy metals and radioactive Cs in fly ash containing radioactive Cs collected at an actual waste incineration facility. A few researchers have reported these properties, including the chemical state of the heavy metals and radioactive Cs in
fly ash. For reuse and recycling and an understanding of the leaching properties of fly ash, including considerable amounts of radioactive Cs, it is necessary to conduct a detailed investigation of the chemical state of radioactive Cs in addition to that of heavy metals. In this study, the chemical state of heavy metals and radioactive Cs were evaluated in detail using the sequential extraction method proposed by Tessier et al.\textsuperscript{17} In addition, a simple extraction method to evaluate the chemical state of each heavy metal was studied because the Tessier extraction method requires four stages of chemical extraction. These results provide a good approach for understanding the characteristics of radioactive Cs and heavy metals in MSWI fly ash.

The solidification and water repellent treatments are widely studied as a detoxification treatment method for landfilled fly ash. Elemental and chemical state compositions of MSWI fly ash are depended on compositions of municipal solid waste and incineration process.\textsuperscript{18} Therefore, it is significant that chemical state analysis of MSWI fly ash containing $^{137}$Cs collected in Fukushima Prefecture is necessary to comprehend characteristics of MSWI fly ash. This study would be useful for the stabilization with water repellent treatment and solidification treatment by using actual samples based on the management of landfill sites.

**Experimental**

**Fly ash sample preparation**

The MSWI fly ash sample was collected in Fukushima Prefecture, Japan, in January 2013. The combustion capacity of the plant is 300 t day\textsuperscript{-1} with two stoker furnaces in the plant, where MSW is combusted. The analysis sample was the same as the standard fly ash prepared by Fujii et al.\textsuperscript{19}
MSWI fly ash was heated using a muffle furnace (KDF S-70, Denken-Highdental Co., Ltd., Japan) at 900 °C for 20 min. The heated sample was added to a mixed solution of 10 mL (1:1) hydrogen fluoride (HF), 8 mL aqua regia, and 1 mL 1% NaCl aqueous solution, and acid-decomposed at 150 °C for 4 h. The residue after evaporation to dryness was decomposed at 220 °C for 24 h in 2 mL of HNO₃ and 7 mL of HClO₄ solution. The sample was then treated with 10 mL of aqua regia at 150 °C for 4 h. Finally, the acid-decomposed sample was dissolved in 5 mL of HCl at 100 °C and filtered through No. 5C filter paper. Elemental analysis was performed on the sample solution diluted to a constant volume after acid decomposition. The HNO₃ concentration used for dilution of the sample solution was adjusted to 0.1 mol L⁻¹.

The fly ash was dried at 105 °C for 24 h before conducting the experiments. The fly ash was compressed into the U8 container (height and diameter of 68 and 56 mm, respectively, Sekiya Rika Co. Ltd, Japan) at height of 5 cm and measured using gamma-ray spectrometry.

Apparatus

Inductively coupled plasma atomic emission spectrometry (ICP-AES; Thermo Fisher Scientific, USA) was employed for elemental analysis of the samples. The instrumental conditions of the ICP-AES were as follows: Radio frequency (RF) power was 1150 W, coolant Ar flow was 12 L min⁻¹, auxiliary Ar flow was 0.5 L min⁻¹, nebulizer Ar flow was 0.5 L min⁻¹, substituted flow quantity was 100 rpm, and analysis flow quantity was 50 rpm. A blank solution and a series of Na, K, Cr, Mn, Fe, Ni, Cu, Zn, Sr, Cd, and Pb multielement standards (0.01, 0.05, 0.1, 0.5 and 1.0 mg L⁻¹) was prepared using a serial dilution of 1000 mg L⁻¹ elemental standards (Wako, Japan) for the calibration of the ICP-AES. The concentrations of blank and standard solutions were
prepared to 0.1 mol L\(^{-1}\) using by HNO\(_3\). ICP-AES emission wavelengths of 589.5 (Na), 769.8 (K), 283.563 (Cr), 257.610 (Mn), 259.940 (Fe), 221.647 (Ni), 324.754 (Cu), 213.856 (Zn), 407.771 (Sr), 228.802 (Cd), and 220.353 nm (Pb) were used for determination of these elements. The solutions of each fraction by the sequential extraction method were prepared to 0.1 mol L\(^{-1}\) concentration using by HNO\(_3\) for the ICP-AES analysis.

The activity of radioactive Cs 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 100-mm-thick Pb shields with additional 50-mm-thick O-free Cu and 5-mm-thick acrylic plates. The resolution (full width half maximum) and the relative efficiency at 1332 keV, \(^{60}\)Co were 1.71 keV and 12.31\%, respectively. 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). To determine the radionuclides in environmental samples in a non-controlled area in Japan, Koike et al.\(^{20}\) developed a gamma-ray spectrometry calibration method for radionuclides using three natural radionuclides (\textit{i.e.}, \(^{40}\)K, \(^{138}\)La, and \(^{176}\)Lu), included in commercially available chemical regents, KCl, LaF\(_3\), and Lu\(_2\)O\(_3\), respectively. We used this method for the determination of radioactive Cs. The activities of \(^{40}\)K, \(^{134}\)Cs, and \(^{137}\)Cs were determined by the emitted gamma rays, \textit{i.e.}, 1461, 606, and 662 keV, respectively. Each radioactivity concentration was corrected by the background on the gamma-ray spectrum measured for 605427 seconds (\textit{live time}). The activity ratio of \(^{134}\)Cs/\(^{137}\)Cs in the fly ash was decay-corrected on March 11, 2011.
Sequential extraction method

The sequential extraction method was applied to estimate the chemical state of heavy metals and radioactive Cs in MSWI fly ash. This method was developed for the speciation of heavy metals in soil and sediment samples by classification of the five fractions using appropriate extraction reagents. This method has been employed for speciation of both heavy metals and radionuclides. Bunzl et al. reported the speciation of $^{238}$U, $^{226}$Ra, $^{210}$Pb, $^{228}$Ra, and stable Pb in the soil from a uranium mine. The Tessier method was modified for this study to incorporate the following steps. The procedure of sequential extraction method is shown Fig. 1. Solution of each fraction was analyzed by ICP-AES for heavy metals and gamma-ray spectrometry for radioactive Cs.

1. Water-soluble (WS) step

   A volume of 50 mL of pure water was added to 5 g of fly ash, and the mixture was agitated for 6 h at room temperature (20–25 °C). This suspension was then separated from the residue and extracted by filtration. The remaining residue was used for the next extraction.

2. Carbonate-bound (CB) step

   A buffer solution of 40 mL of 1 mol L$^{-1}$ sodium acetate and acetic acid (pH = 5.0) was added to the residue from step 1. The resulting solution was agitated for 6 h at room temperature.

3. Free oxide (OX) step

   The OX phase was extracted with 100 mL of 0.04 mol L$^{-1}$ NH$_2$OH • HCl solution containing 25% (v/v) acetic acid at 96 °C for 6 h.

4. Bound to organic matter, sulfide state (OB)

   The solid phase residue after step 3 was agitated in 40 mL of 30% H$_2$O$_2$ and 15
mL of 0.02 mol L\(^{-1}\) HNO\(_3\) at 85 °C for 3 h. The mixture was added to 25 mL of 3.2 mol L\(^{-1}\) NH\(_4\)OAc solution (HNO\(_3\) 25 vol%) and 20 mL pure water, and agitated for 3 h.

5. Residue (RES)

The residual solid material after step 4 was dried at 85 °C for 24 h.

**Results and Discussion**

*Concentration of heavy metals and radioactive Cs*

The total amount of heavy metals in fly ash was analyzed by ICP-AES in combination with acid decomposition. The concentrations of heavy metal in the fly ash are shown Fig. 2. The ranges of total concentration of heavy metals in MSWI fly ash from waste incineration in different countries were reported in IAWG\(^{22}\) and Hiraoka and Sakai\(^{23}\): 140–1100 mg kg\(^{-1}\) for Cr, 510–26000 mg kg\(^{-1}\) for Pb, 1620–70000 mg kg\(^{-1}\) for Zn, and 15.6–187 mg kg\(^{-1}\) for Cd. The concentrations of heavy metals in MSWI fly ash showed values falling within these ranges. Therefore, MSWI fly ash used in this study showed general compositions of heavy metals similar to other MSWI fly ash.\(^{22,23}\)

However, MSWI fly ash contained considerable amounts of radionuclides; these were analyzed by gamma-ray spectrometry. \(^{40}\)K, \(^{134}\)Cs, and \(^{137}\)Cs were identified on the gamma-ray spectrum of fly ash measured for 5492 seconds (live time). Activity concentration of \(^{40}\)K, \(^{134}\)Cs, and \(^{137}\)Cs were 2540±130 Bq kg\(^{-1}\), 882±17 Bq kg\(^{-1}\), and 4470±43 Bq kg\(^{-1}\), respectively. Iwahana *et al.*\(^{11}\) reported that the activity concentration of \(^{40}\)K in fly ash corresponded to the natural radionuclides before the FDNPP accident, ranging from 445 Bq kg\(^{-1}\) to 2600 Bq kg\(^{-1}\). The activity concentration of \(^{40}\)K in fly ash was found to be equivalent to the value before the FDNPP accident. Moreover, the activity concentrations of \(^{134}\)Cs and \(^{137}\)Cs showed higher values than before the FDNPP
accident. The activity ratio of $^{134}$Cs/$^{137}$Cs in Fukushima-derived radionuclides was reported as 1, and the behavior of $^{134}$Cs was similar to that of $^{137}$Cs. The activity ratio of $^{134}$Cs/$^{137}$Cs (4741±92 Bq kg$^{-1}$/5015±48 Bq kg$^{-1}$) which was decay-corrected on March 11, 2011 was determined as 1 in this study. Therefore, it is considered that the radioactive Cs in the fly ash originated from the FDNPP accident.

Leaching behavior of heavy metals and radioactive Cs

The Tessier sequential extraction method was applied to fly ash, and each effluent and the final residue after acid decomposition was analyzed by ICP-AES. The distributions of heavy metals and radioactive Cs in fly ash are shown in Fig. 3. In Japan, fly ash, including WS heavy metals at higher levels than the regulatory limit, was only deposited at a final disposal site after stabilization treatment. The concentrations of WS heavy metals in fly ash were higher than the regulatory limit (Table 1). Therefore, fly ash was deposited at a final disposal site and was not used for recycling.

Almost all the heavy metals, except Sr and Pb, were eluted by the three fractions of CB, OX, and OB. In general, the chemical state of elements in the four non-water-soluble fractions (CB, OX, OB, and RES) elutes very little into the environment. Therefore, most of the heavy metals in the fly ash hardly eluted into the environment. However, Sr and Pb were eluted by the WS fraction. Approximately 10% of Pb was eluted by the WS fraction; therefore, it is considered that Pb was present as a WS chloride. Almost all the Pb was eluted by the CB or OX fractions. The results indicated that stable Pb was present in at least three forms and it was more easily eluted than other elements. In contrast to transition metals, more than 50% of the alkali metals (Na, K, and Cs) were eluted in WS fraction. It is suggested that almost all alkali metals were contained in the MSWI fly ash formed chemical states such as chlorides.
Chloride-containing compounds were likely derived from kitchen wastes including vegetables, seasonings, and plastics.\textsuperscript{15} The alkali metal species in WS fraction had high solubility and showed a high elution rate.

After the FDNPP accident, researchers focused on the leaching behavior of Sr from fly ash because the majority of \(^{90}\)Sr was released to the environment. Owing to a long half-life, \(^{90}\)Sr has a high affinity to bone and can be a very dangerous radioactive isotope in the human body. Approximately 20\% of Sr was eluted by the WS fraction, which could potentially affect the ecological environment. This result indicates the possibility of migration behavior of \(^{90}\)Sr in MSWI fly ash to environmental water. Assuming of chemical state in the WS fraction\textsuperscript{8,25} of Sr was SrCl\(_2\), 45\% of Sr in MSWI fly ash was eluted in the CB and OX fractions. Yamamoto \textit{et al.}\textsuperscript{26} reported that the \(^{90}\)Sr concentration in incinerated fly ash was 2.3-7.75 Bq kg\(^{-1}\) although chemical state of \(^{90}\)Sr was unidentified. The comprehending of chemical state of Sr is an important to establish stabilize treatment of MSWI fly ash for elution suppression of \(^{90}\)Sr. It is difficult to estimate chemical state of \(^{90}\)Sr in solution of each fraction, and its analysis is for a future work. The other heavy metals—Zn, Cu, and Cd—were eluted by the OX fraction. Takaoka \textit{et al.}\textsuperscript{27} investigated the speciation of Zn, Pb, Cu, and Cd in fly ash using the sequential extraction procedure, and reported that almost of all these heavy metals existed in oxide form. The results in Fig. 3 showed similar results as in Takaoka \textit{et al.}\textsuperscript{27}. Funatsuki \textit{et al.}\textsuperscript{28} suggested that Cd was eluted in the WS fraction as CdCl\(_2\). The MSWI fly ash used in this experiment was collected by a bag filter as a reaction product with slaked lime. In Fig. 3, almost no Cd was eluted by the WS fraction. By spraying slaked lime, it is considered that Cd in this fly ash was contained as CdO, Cd(OH)\(_2\), and CdCO\(_3\) rather than CdCl\(_2\).\textsuperscript{27}

Chemical state analysis of radionuclides including radioactive Cs was
investigated. However, most of these studies used a simulation sample. The results of the sequential extraction method of MSWI fly ash indicated that elution rates of $^{134}$Cs and $^{137}$Cs in the WS fraction were 58.4% and 61.7%, respectively. The leaching behavior of radioactive Cs showed a similar behavior to that of alkaline metals, such as Na and K. The relationship between the elution rate of radioactive Cs and alkali metals (Na and K) in the fraction obtained by sequential extraction is shown in Fig. 4. Similar elution behaviors were observed for radioactive Cs and alkali metals during each extraction process. As per the Tessier method, Fujii et al. and Ohbuchi et al. indicated that WS radioactive Cs exists as CsCl. Therefore, it was estimated that radioactive Cs formed CsCl in fly ash, with the same chemical state as alkaline metals.

Radioactive Cs was detected in the gamma-ray spectrum of the residue after the extraction. Activity concentration of $^{134}$Cs and $^{137}$Cs in the residue were 53.8±1.3 Bq kg$^{-1}$ and 295±4 Bq kg$^{-1}$, respectively. Albite (NaAlSi$_3$O$_8$), gehlenite (Ca$_2$Al$_2$SiO$_7$), and amorphous phase were identified in the XRD pattern of the residue after the extraction. These crystalline species may occur as the result of oxidation during the combustion process of MSW. It is considered that non-soluble radioactive Cs in fly ash was bound to aluminosilicate or captured in the amorphous phase.

After undergoing intermediate treatment specified by the Minister of Health, Labor, and Welfare, incinerated fly ash is disposed as a specially controlled general waste at a final disposal site. It is considered that the long-term risk of radioactive Cs release after landfilled fly ash into the environmental water can be evaluated by comprehending the chemical state of radioactive Cs in MSWI fly ash. Chemical state analysis of radioactive Cs in incinerated fly ash has been conducted by simulation and simulated samples prepared by adding stable Cs. Currently, we have been studying the elution suppression method using of heavy metals and radioactive Cs in MSWI fly
ash samples as an actual sample collected from the incineration facility.\textsuperscript{31,32} The results of chemical state analysis will be useful information for elucidating the water repellent and cement solidification mechanism.

\textit{Leaching behavior of radioactive Cs with acid treatment}

The Tessier extraction method is applied to soil materials. Takaoka \textit{et al.}\textsuperscript{27} applied the modified sequential extraction method for fly ash to estimate the speciation of Zn, Pb, Cu, and Cd. However, several procedures are needed to obtain each chemical state. Therefore, a simple extraction method using HNO\textsubscript{3} was applied for evaluation of the chemical states of radioactive Cs.

Insoluble radioactive Cs was trapped into silicate as an amorphous phase by chemical treatment with HF.\textsuperscript{16} Therefore, acid treatment with HNO\textsubscript{3}, with less risk than HF, was applied to investigate the pH-dependent characteristics of the leaching behavior of radioactive Cs. The Japanese leaching test No. 13 (JLT-13) for industrial waste is one of the most rapid and easy methods for determining the leachability of heavy metals from various types of wastes in a waste landfill.\textsuperscript{33} The acid leaching test with HNO\textsubscript{3} aqueous solution or ammonia water was based on the JLT-13. In the acid leaching test, a mixture of HNO\textsubscript{3} aqueous solution adjusted to pH 1, 5, 7, and 13 and MSWI fly ash (S/L = 0.1: HNO\textsubscript{3} 50 mL of aqueous solution or ammonia water 50 mL, 5 g of fly ash sample) was subjected to continuous shaking for 6 h at room temperature (20–25 °C) in a mechanical shaker (Shaking Bath TBK 602DA, Advantec Inc., Japan). The velocity of the shaker was approximately 200 rpm. 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). According to the results of JLT-13, approximately 65\% of radioactive Cs in fly ash from municipal waste incineration
elutes into water at a pH of 6.7. Figure 5 shows the relationship between the pH of the aqueous HNO₃ solution and the elution rate of radioactive Cs. The leaching behavior of radioactive Cs in incinerated fly ash showed the pH dependency of the contact solution. Approximately 90% of radioactive Cs was eluted with the pH 1 eluate. This result corresponded to WS, CB, OX. In the pH 5 leaching test, a part of CB was eluted in addition to WS. In the case of MSWI fly ash, heavy metals present in the CB fraction are considered to be eluted into the environment by acid rain (pH 4.50–5.18). By adjusting the pH of the eluate, the chemical form can be easily estimated.

**Conclusions**

The chemical state of heavy metals was reported using the sequential extraction method. Few studies on chemical state analysis of radioactive Cs using samples collected from incinerators have been reported. In this study, the chemical state of both heavy metals and radioactive Cs were estimated in fly ash sampled at Fukushima prefecture, Japan. It is necessary to manage such wastes to reduce the risks of dose exposure to workers. Incinerator ash including radioactive Cs at concentrations less than the abovementioned regulatory limit can be placed in landfills as general waste. In recent years, landfill space has become limited owing to a large amount of waste. Hence, effective reuse of waste is proposed to address these concerns. For example, fly ash has been reused as eco-cement considering its cement-like characteristics. The potential risks of heavy metal and radioactive Cs elution from fly ash decrease as a result of solubilization treatment. The chemical state of radioactive Cs in fly ash can be utilized for water repellent treatment of MSWI fly ash.
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| Element | Concentrations of water-soluble heavy metals / ppm | Regulatory limit$^a$ / mg L$^{-1}$ |
|---------|-----------------------------------------------|---------------------------------|
| Na      | 1657                                          | –                               |
| K       | 1778                                          | –                               |
| Cr      | 0.869                                         | 0.05$^b$                        |
| Mn      | 0.0561                                        | –                               |
| Fe      | 2.93                                          | –                               |
| Ni      | 0.0120                                        | –                               |
| Cu      | 0.149                                         | –                               |
| Zn      | 1.11                                          | –                               |
| Sr      | 27.8                                          | –                               |
| Cd      | 0.0451                                        | 0.01                            |
| Pb      | 6.91                                          | 0.01                            |

$^a$ Environmental quality standards for soil pollution by the Ministry of Environment, Government of Japan

$^b$ Standard for Cr (VI)
Figure Captions

Fig. 1  Procedure of sequential extraction method for MSWI fly ash sample.

Fig. 2  Concentration of heavy metals in the fly ash sampled at municipal waste incinerators in Fukushima prefecture in Jan. 2013.

Fig. 3  Distributions of heavy metals and radioactive Cs in municipal solid waste incineration fly ash sampled at Fukushima prefecture.

Fig. 4  Relationship between elution ratio of radioactive Cs (\(^{134}\text{Cs}: \text{solid} \) and \(^{137}\text{Cs}: \text{open}\)) and alkali metals (Na: circle and K: triangle) from the MSWI fly ash.

Fig. 5  Relationship between the pH of the aqueous HNO\(_3\) solution and the elution rate of radioactive Cs.
Sample 5 g

- 50 mL of pure water.

The residue of WS

- 40 mL of 0.04 M NaOAc-HOAc buffer (pH 5.0) soln.

The residue of CB

- 100 mL of 0.04 M NH₂OH•HCl soln.

The residue of OX

1) 40 mL of 30 % H₂O₂
2) 15 mL of 0.02 M HNO₃
3) 25 mL of 3.2 M NH₄OAc soln. (25 vol% HNO₃)
4) 20 mL of pure water

The residue of OB

- 6 h agitate

Water Soluble
- WS

Non-water soluble
- CB
- OX
- OB
- RES

Fig. 1
Fig. 2
Fig. 3
Elution ratio of $^{134}\text{Cs}$, %

Elution ratio of $^{137}\text{Cs}$, %

Elution ratio of Na, %

Elution ratio of K, %

$r = 0.948$

$r = 0.962$

$r = 0.947$

$r = 0.969$

Fig. 4
Fig. 5
Graphical Index

Chemical state analysis of heavy metals and radioactive Cs in MSWI fly ash

Elements and nuclides

Distribution, %

Na, K, Cr, Mn, Fe, Ni, Cu, Zn, Sr, Cd, Pb, 134Cs, 137Cs

WS, CB, OX, OB, RES