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Study on the Accumulation Mechanism of Cesium in the Amorphous Phase around Mineral Particles in Incineration Bottom Ash, and its Application

Yasumasa TOJO
Graduate school of Engineering, Hokkaido University
Kita 13, Nishi 8, Kita-ku, Sapporo-City, Hokkaido 060-8628 JAPAN
E-mail: tojo@eng.hokudai.ac.jp

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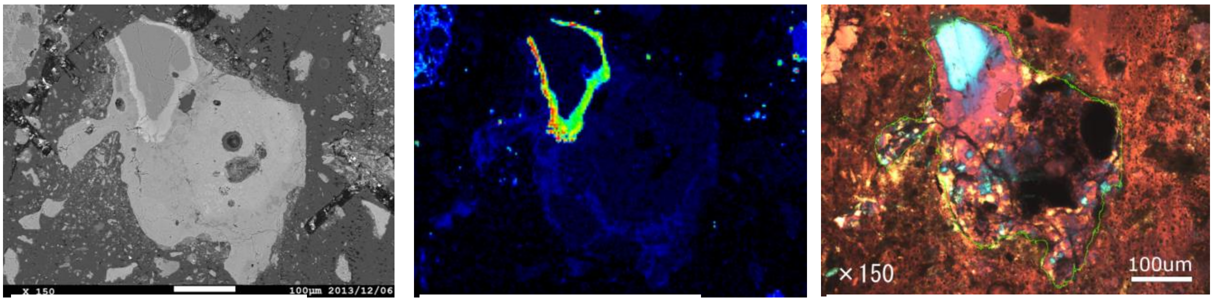
The Fukushima nuclear accident caused by a huge earthquake in 2011, resulted in the generation of solid wastes contaminated by radioactive cesium (Cs) in eastern Japan. Combustible wastes were subsequently thermally treated as usual, producing incineration residues containing high Cs levels. In addition, huge amounts of wastes and soils have since then been generated by decontamination activities. To reduce the final amount needing disposal, it has been decided that they should be thermally treated, and a part of the treatment has already commenced. Since the residue generated from such treatment has high radioactivity derived mainly from radioactive Cs, it must be strictly managed (stored) for a long time thereafter. What is important in this long-term management is that Cs never escapes from those storage sites. For this purpose, it would be better for the Cs to have less-soluble or insoluble properties, especially if it risks coming into contact with rainwater or other liquids.

From previous studies, it has been reported that Cs in incineration bottom ash is nearly insoluble. We investigated the reasons for that and found that Cs is captured in a glassy amorphous phase forming on the surface of a specific mineral in the ash particles as indicated in Fig. 1. This mineral was composed mainly of Si, Al and O.



Backscatter electron image   Elemental mapping of Cs   Image of polarized light microscopy

Fig. 1 Bottom ash particle containing nearly insoluble Cs.

It remained unclear, however, what this mineral was. Since Cs tends to concentrate predominantly in the mineral, it would be possible to make Cs insoluble by thermally treating these wastes/incineration residue/soils together with the mineral. For this purpose, identification of the mineral was indispensable.

This study set three objectives. The first (identification) was to identify the mineral capturing Cs by using various microscopic techniques such as μXRD, FT-IR and Raman microscopy. The second (confirmation of origin) was to elucidate the origin of the mineral, i.e., to clarify whether it originated from waste or was synthesized during incineration. The third (process development) was to confirm the possibility of Cs insolubilization by thermal treatment in which wastes/ashes/soils containing Cs were co-heated with the mineral.

The main results obtained were as follows.

By analyzing many bottom ash particles on which Cs specifically concentrated, these particles were identified as being composed of Al, Si, O and K or Na. By analyzing the mineral existing at the center of the particle by Raman microscopy, we found that they were alkali feldspar (microcline ($KAlSi_3O_8$) and albite ($NaAlSi_3O_8$)) as indicated in Fig. 2. When Cs (in the form of $C_{s2}CO_3$) was added to alkali feldspar (microcline) and then the mixture was heated at 900°C for two hours, capture of Cs similar to that confirmed in the ash was reproduced (Fig. 3) and almost 100% of the added Cs became insoluble (Fig. 4). Cs was also captured in particles that were created when heating Cs with feldspar-synthesizing reagent chemicals (Al_2O_3, SiO_2, K_2CO_3). In this case, Cs was trapped inside the particles, not on the surface. Due to the difference in location of the Cs, it became clear that the feldspar that had captured the Cs in the bottom ash had originally been present in the waste incinerated.

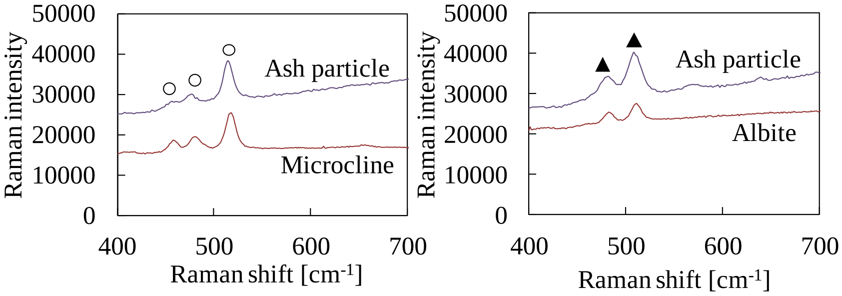
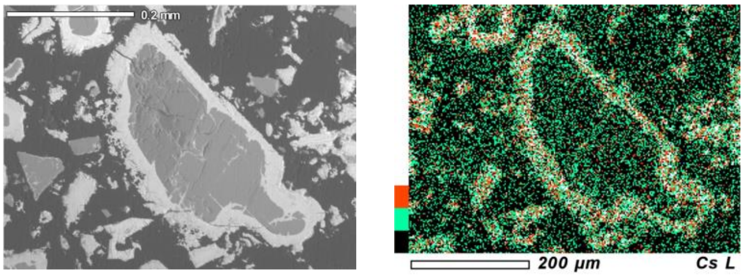


Fig. 2 Raman spectra of the two minerals existing at the center of a bottom ash particle that captured Cs and comparison with the spectra of microcline and albite.



(a) Backscatter electron image (b) Elemental mapping of Cs

Fig. 3 Reproducing Cs capture using microcline.

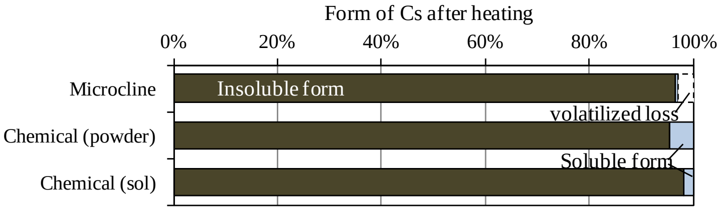


Fig. 4 Distribution of Cs after heating with microcline and feldspar-synthesizing reagent chemicals.

A co-combustion test of these feldspars with combustible waste containing Cs was conducted to confirm their inhibitory effects on volatilization of Cs and promotion of Cs insolubilization. Combustible waste was artificially created to simulate waste generated from the decomtamination activities. Co-combustion tests were conducted without feldspar, with feldspar, with powdered chemical reagent and with chemical reagent sol. As indicated in Fig. 5, the tests revealed that volatilization of Cs could be suppressed and the ratio of the insoluble form of Cs increased.

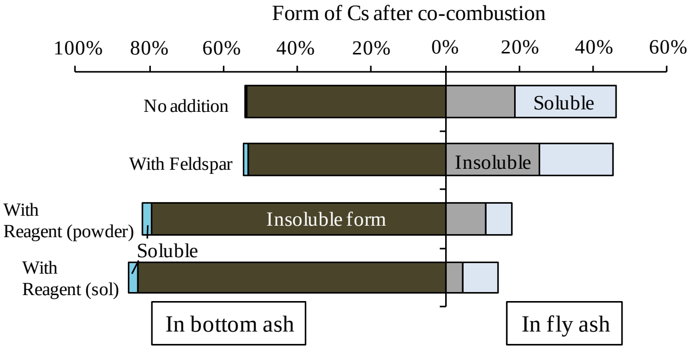


Fig. 5 Distribution of Cs after co-combustion of simulated decontamination waste with feldspar/feldspar synthesizing

In thermal treatment, Cs tends to concentrate in fly ash and become soluble. Therefore, we additionally investigated application of the insolubilization mechanism found in this study to Cs in fly ash. Since Cs in fly ash is CsCl, the Cs source was changed to CsCl and co-heating with feldspar was conducted. The capture and insolubilization mechanism worked even when the Cs was chloride. In case of CsCl, the optimum temperature for capturing Cs was 700°C. The efficiency of Cs capture and insolubilization could be increased even if inexpensive feldspar from the ceramic industry were used if its structure could be made amorphous.