Recovery of Radioactive Cesium from Sewage Sludge Ashes —Paving the Way for Disposal of Sewage Sludge Ashes Containing Radioactive Cesium

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After the accident of the Fukushima Daiichi Nuclear Power Plant, the radioactive Cs was dispersed across large areas of eastern Japan. Some of the radioactive Cs has been discharged into sewage system. At sewage treatment plants, sewage is biologically treated and converted to sewage sludge. Sewage sludge is finally incinerated at 800–850 °C. The end product of sewage treatment is called sewage sludge ash (SSA). Since the Fukushima Daiichi accident, highly concentrated radioactive Cs has been detected in the SSAs produced at many sewage treatment plants in Japan. Most of the contaminated SSAs have been stored at those sewage treatment plants (as of October 2012, 120,000 t [1]).

Based on the experiences of decontaminating soils, some attempts were made to reduce radioactive Cs from SSA by dissolving SSA in organic acid or nitric acid. However, those attempts only evaluated the total amount of Cs dissolved under predetermined conditions without being accompanied by scientific grounds of the dissolution behavior of Cs.

Under these circumstances, we collected SSA samples contaminated with radioactive Cs fallout and investigated relations among elements such as radioactive Cs and iron dissolved from SSAs in acid solution to elucidate the chemical states of Cs, *i.e.*, the solid phase retaining Cs [2]. We found that radioactive Cs is mainly contained in iron oxides, and a part of the iron oxides is not easily dissolved in acid solution because it is present in dense sintered aggregates of numerous nanoparticles with silicate-framework. These aggregates are supposed to be formed during incineration of sewage sludge.

Accordingly, we pulverized SSA down to several-hundred nanometer (one nanometer: one billionth of one meter), and then dissolved in hydrochloric acid. Almost all of the iron oxides were dissolved by these treatments, and as a result, we succeeded in recovering more than 90% of radioactive Cs, while the maximum recovery had been only 70% without pulverization. By applying pulverization and subsequent dissolution in hydrochloric acid, the original radioactivity concentration of an SSA sample, 23k Bq/kg, was reduced to 4.8k Bq/kg, which is much lower than the standard value of radioactive Cs for the designated waste, 8k Bq/kg.

The dissolution residue is composed of three kinds of silicates: naturally-occurring quartz and feldspar, and unidentified silicate (not clay) probably formed by the incineration of sewage sludge. We confirmed that no radioactive Cs is dissolved into solution even when the dissolution residues were immersed in pure water or sea water at 60°C, indicating that Cs is tightly fixed in the silicate. On the other hand, several percentages of radioactive Cs is dissolved from the original SSA. Therefore, the dissolution residue has an advantage over the original SSA for landfill disposal.

The present results were obtained in the laboratory. There still remain many problems to be solved, such as the development of methods to reuse solution for dissolving more efficiently.

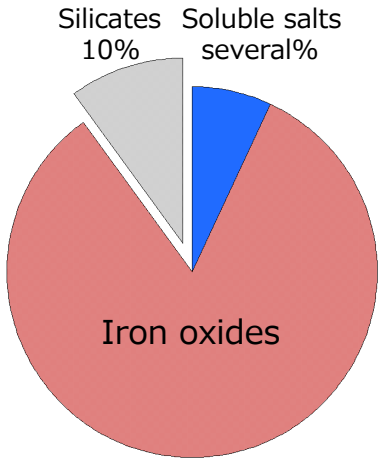

Fig. 1 Fractions of ^{137}Cs in sewage sludge ashes. The majority of ^{137}Cs is contained in iron oxides.

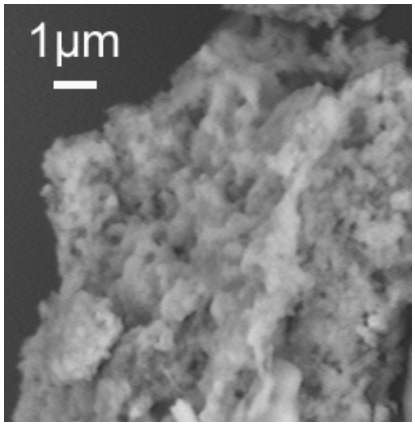

Fig. 2 SEM image of a typical particle of SSA. This particle looks sintered aggregate of numerous nanoparticles. In SSA, iron is contained in particles like this.

References

[1] 地方共同法人日本下水事業団資料 www.jswa.go.jp/gesuidouten/2013/pdf/h3.pdf
[2] N. Kozai *et al.*, Water. Res. **68**, 616 (2015).