

# Chapter 15

## Properties of Radioactive Cs-Bearing Particles Released by the Fukushima Daiichi Nuclear Power Plant Accident and Trace Element Analysis



Kazuhiko Ninomiya

**Abstract** A large amount of radionuclides were released into the environment by the Fukushima Daiichi Nuclear Power Plant (FNPP) accident. By this accident, radioactive cesium (Cs)-bearing particles were also released. Since these particles stably exist in the environment, the influence of long-term exposure to radiation is concerned if the particle is ingested in the human body. In this review, in order to evaluate the influence of radioactive Cs-bearing particles on the environment, studies on radioactive Cs-bearing particles are summarized. Radioactive Cs-bearing particles can be classified into several kinds depending on their elemental composition and radioactivity. Although the production and distribution of radioactive Cs-bearing particles have still been unclear, details of the situation inside the nuclear reactor at the accident can be investigated from the classification and investigation of their production.

**Keywords** Fukushima Daiichi Nuclear Power Plant · Radioactive Cs-bearing particles · Radioactivity analysis · Cs contamination inventory

### 15.1 Introduction

On March 11, 2011, a large magnitude 9.0 earthquake occurred in the Pacific Coast of the northeast region of Japan and an accident occurred at the Fukushima Daiichi Nuclear Power Plant (FNPP) due to the large tsunami generated by the earthquake. This accident was classified in the most serious category of nuclear accident by the International Atomic Energy Agency (level 7), and a large amount of radionuclides were released into the atmosphere and ocean [1]. In addition to measuring the air

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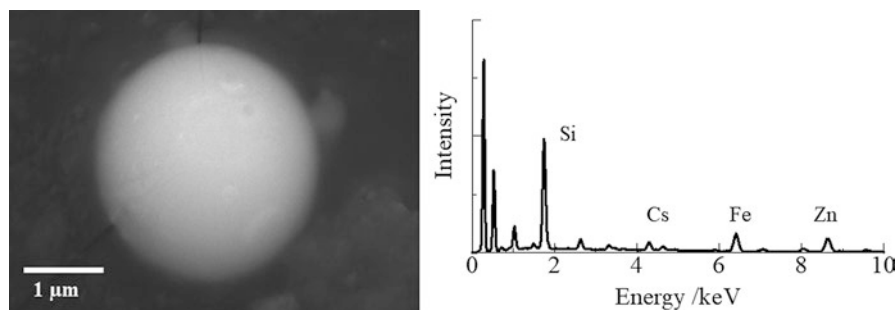
K. Ninomiya (✉)  
Department of Chemistry, Graduate School of Science, Osaka University,  
Toyonaka, Osaka, Japan  
e-mail: [ninokazu@chem.sci.osaka-u.ac.jp](mailto:ninokazu@chem.sci.osaka-u.ac.jp)

dose rate, the distribution of nuclides, such as iodine-131 ( $^{131}\text{I}$ ), tellurium-129m ( $^{129\text{m}}\text{Te}$ ), silver-110m ( $^{110\text{m}}\text{Ag}$ ) and radioactive cesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) deposited in soil was measured, and the initial contamination status was determined [2].

In a nuclear accident, volatilized radionuclides leak from the damaged fuel body and are released into the environment and captured by aerosols in the atmosphere. The aerosol behavior changes according to wind direction, speed and other factors at the time of the release event from the nuclear reactor, greatly affecting the transportation process [3]. Sulfuric acid is the most abundant aerosol in the atmosphere resulting in the radionuclide being transported as a sulfuric acid aerosol form. For the FNPP accident, by analyzing the contents of ionic compounds and radioactivities in the atmospheric aerosol for various particle sizes, it was determined that the amount of radioactive Cs strongly correlates with the amount of sulfuric acid aerosol [4].

On the other hand, heterogeneous spot-like contamination was reported via autoradiography analysis using imaging plates of environmental samples such as plant, soil and atmospheric filters just after the accident [5, 6]. Heterogeneous spot-like contamination was never washed out by water and was not the result of soluble sulfuric acid aerosols but arose from particles with concentrated radioactivity. Radioactive Cs is strongly adsorbed by minerals such as mica and some heterogeneous contamination was identified as soil particles [7–9]. However, Adachi et al. [10] reported in 2013 the detection of insoluble radioactive particles with spherical shapes of several micrometers in diameter from air samples collected in 2011. SEM image of the insoluble particle and its elemental composition is shown in Fig. 15.1. The particles are called variously such as radioactive Cs concentrated particles, Cs particles, insoluble particles, radiocesium-bearing microparticles (CsPs) and radioactive Cs-rich microparticles (CsMPs). The author calls the particles “radioactive Cs-bearing particles” hereafter in this article. Radioactive Cs-bearing particles are quite different from the “hot particles” found in the Chernobyl NPP accident [11] and various radioactive Cs-bearing particles have been discovered in Fukushima Prefecture and neighboring areas to date.

In this review, the author summarizes the recently reported papers on radioactive Cs-bearing particles that have been commonly detected after the FNPP accident. In addition to the properties of radioactive Cs-bearing particles, we examine their trace element analysis, distribution in the environment, formation processes and prospects for future research.

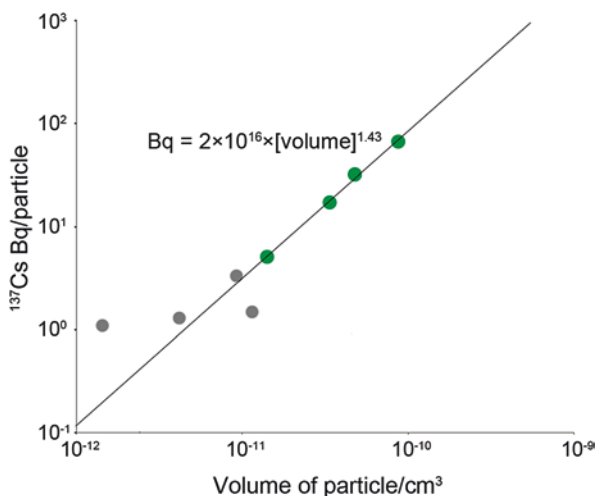


**Fig. 15.1** SEM image of a radioactive Cs-bearing particle (left) and its EDS spectrum (right)

## 15.2 Properties of Radioactive Cs-Bearing Particles

Radioactive Cs-bearing particles were isolated and analyzed for the first time from atmospheric samples (filters) collected in Tsukuba, 170 km southwest from FNPP [10]. From the scanning electron microscope (SEM) analysis, it is confirmed that radioactive Cs-bearing particles are spherical with a diameter of 2  $\mu\text{m}$  and contain Cs, iron (Fe), zinc (Zn), manganese (Mn) and chlorine (Cl). Those radioactive Cs-bearing particles also contain several Bq of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  with the particle radioactivity and size, indicating that a particle contains approximately 5 wt% of radioactive Cs. The particle is primarily composed of  $\text{SiO}_2$  though it is not clear because there are silicon (Si) background signals in the SEM analysis. They also reported radioactive Cs-bearing particles retain their shape even after washing with water. Abe et al. [12] conducted a more detailed analysis of radioactive Cs-bearing particles by synchrotron radiation X-ray (SR- $\mu$ -X-ray) analysis showing that radioactive Cs-bearing particles contain molybdenum (Mo), barium (Ba), tin (Sn) and a trace amount of uranium (U). Furthermore, the chemical state of the metals contained in radioactive Cs-bearing particles was determined by SR- $\mu$ -XANES (synchrotron radiation X-ray absorption near edge structure) analysis, and Fe, Zn, Sn and Mo are present in their oxidized states as  $\text{Fe}^{3+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Sn}^{4+}$  and  $\text{Mo}^{6+}$ , respectively. Radioactive Cs-bearing particles can also be isolated from leaves, soil and dust in the high dose area of Fukushima Prefecture [13–15]. Sato et al. [16] performed detailed analysis of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  radioactivity for radioactive Cs-bearing particles found in soil collected 20 km northwest of FNPP and reported the  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio just after the accident to be  $1.03 \pm 0.01$ . The authors also reported the clear linear relationship between particle size and radioactivity, that is, the specific activity is the same, indicating that radioactive Cs-bearing particles originate from the same source (Fig. 15.2).

**Fig. 15.2** Relation between the size of a radioactive Cs-bearing particle and its activity obtained by Sato et al. [16]



**Table 15.1** Characteristics of radioactive Cs-bearing particles [15]

Characteristic	Particle type	
	A	B
Size distribution	1–10 $\mu\text{m}$	40–400 $\mu\text{m}$
$^{134}\text{Cs}/^{137}\text{Cs}$	1.04	0.93
Other radionuclides	N/A	Antimony-125 ( $^{125}\text{Sb}$ )
Distribution	Wide	Limited (North)
Suspected emission date	March 15, 2011	March 12, 2011
Source of reactor(s)	Unit 2 or 3	Unit 1
Specific radioactivity	High	Low
Elements in common	Si, O, Zn	
Elements contained	Fe, Sn, Cl	Na, Mg, Ca, Ba

Sato et al. [15] classified 19 radioactive Cs-bearing particles collected from soil and dust samples in Fukushima Prefecture and found two types of radioactive Cs-bearing particles depending on the discovery location (Table 15.1). Both radioactive Cs-bearing particles are primarily composed of  $\text{SiO}_2$  as a base material [17], but their detailed properties are different. The “type A” particles are similar to the particles found in Tsukuba [10, 12]. These particles are characterized by several Bq of radioactivity per particle, but the specific activity is very large due to the small particle size, and typical radioactive Cs content is approximately several wt%. The  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio was approximately 1, so the source of the particle is likely Unit 2 or Unit 3, as estimated by Nishihara et al. [18]. The particles classified as “type B” have large activities, some exceeding 20 kBq per particle. On the other hand, the specific activity is much smaller than that of type A particle because the size of the type B particle is typically several tens of  $\mu\text{m}$  or more. The  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio is approximately 0.9, and the source of the type B particles is likely Unit 1 [18]. In addition, some type B particles exhibit Antimony-125 ( $^{125}\text{Sb}$ ) activity in addition to radioactive Cs.

### 15.3 Detailed Structure of Radioactive Cs-Bearing Particles by Destructive Analysis Method

Yamaguchi et al. [13] and Kogure et al. [19] sliced radioactive Cs-bearing particles into a thin film using a focused ion beam (FIB) instrument and analyzed the film by transmission electron microscopy (TEM), X-ray microanalysis with scanning TEM (STEM) and energy-dispersive X-ray spectroscopy (EDS). These analyses clearly showed that radioactive Cs-bearing particles are primarily silicate glass composed of Si and O. From the semiquantitative analysis by TEM of the entire particle, the content of  $\text{SiO}_2$  was estimated to be 70%. It is also reported that the constituent elements are distributed almost uniformly in the radioactive Cs-bearing particle. However, the concentration of radioactive Cs outside the particles is approximately

twice as high as the inside concentration, and potassium (K) and rubidium (Rb) are in the opposite distributions. The authors also reported that sulfide nanoparticles are implanted in the particles [13].

Furuki et al. [14] found that radioactive Cs-bearing particles contain fine pores 10–200 nm in size as determined by TEM analysis. In addition, they reported that crystalline nanostructures such as fine CsOH and Fe-Zn-oxide are formed inside the particles.

Imoto et al. [20] analyzed the isotope ratios of U, Cs, Ba, Rb, K and calcium (Ca) by secondary ion mass spectrometry (SIMS). They reported the  $^{235}\text{U}/^{238}\text{U}$  isotope ratio to be 0.29, which is significantly higher than the natural abundance of U (0.00729). In SIMS analysis, it is difficult to distinguish between Cs and Ba isobars, but the isotopic ratio of Ba indicates that most Ba contained in radioactive Cs-bearing particles originated from the decay of radioactive Cs. The  $^{87}\text{Rb}/^{85}\text{Rb}$  isotope ratio also indicated that Rb derived from fission products. On the other hand, ratios of K and Ca were consistent with their respective natural isotopic ratios. The authors also attempted to measure the amount of  $^{90}\text{Sr}$ , but the concentration fell below the limit of detection. Ochiai et al. [21] reported that U in radioactive Cs-bearing particles is present in iron oxide nanoparticles detected by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM).

## 15.4 Separating Radioactive Particles

Radioactive Cs-bearing particles are very small and contain a  $\text{SiO}_2$  matrix. Many environmental particles have similar elemental compositions, so identification and extraction of radioactive Cs-bearing particles can be difficult. Although bulk extraction has not yet been developed, other separation methods have been developed to date, as discussed below.

Adachi et al. [10] succeeded in isolating radioactive Cs-bearing particles by fragmenting the sample. From autoradiography measurements using an imaging plate (IP), the position of radioactive Cs-bearing particles in the environmental sample was identified. They transferred the environmental sample including radioactive Cs-bearing particles to a carbon tape and separated it into small pieces. By repeating the visualization by IP and fragmentation of the carbon tape, the particle could be completely separated from other components in the environmental sample. Furuki et al. [14] developed a similar method for isolating the radioactive Cs-bearing particles from soil samples. The authors disperse the soil sample on grid paper and identify the position of radioactive Cs-bearing particles by autoradiography and then drop pure water on that position. The droplet is spreaded thinly and the position of the radioactive Cs-bearing particles is determined using IP again. By repeating this procedure, radioactive Cs-bearing particles are separated from the bulk soil sample.

Radiation measurement using a semiconductor or scintillation detector instead of autoradiography by IP is also used to separate radioactive Cs-bearing particles.

Kurihara et al. [22] placed the environmental samples including radioactive Cs-bearing particles in a vial and divided each sample into two parts after mixing. After radiation measurement, water was added to the higher activity sample and subsequently divided into two samples. By repeating this procedure approximately 30 times, the radioactive Cs-bearing particles can be completely separated from the bulk soil.

## 15.5 Environmental Fate of Radioactive Cs-Bearing Particles

Ikehara et al. [23] conducted an autoradiography experiment with IP to determine the total amount of radioactive particles in soil samples. The soil was divided with a 114-mesh filter and the number of radioactive particles was determined by counting the spots of radioactivity on IP. The authors identified radioactive particles ( $> 0.06$  Bq) in surface soil collected from Okuma Town and Iitate Village and found 48–318 radioactive particles per gram of soil, and the ratio of total radioactivity of the particles to the entire soil sample was 8.53–31.8%.

Igarashi et al. [24] reported the amount of radioactive particles in environmental samples by separating radioactive Cs-bearing particles from soil samples collected in Okuma Town, Fukushima Prefecture. Initially, the number of radioactive Cs-bearing particles (with radioactivity of  $> 1$  Bq) was identified by autoradiography and was separated using Kurihara's method [22]. The radioactivity ratio was approximately 0.2%, differing from the results of Ikehara et al. [23]. This indicates that the proportion of radioactive Cs-bearing particles differs greatly depending on the sampling location.

Miura et al. [25] successfully isolated radioactive particles from suspended particles in rivers. From 2011 to 2016, suspended particles were collected by passing river water collected from the Kuchibuto River in Fukushima Prefecture through a filter. The authors isolated radioactive Cs-bearing particles found on the filter with a radioactivity more than 0.4 Bq, and the activity ratio of radioactive Cs-bearing particles to the filter was estimated to be 0–46%. The particles with weaker radioactivity (0.1–0.4 Bq) were identified via the IP analysis and total radioactivity on the filter was estimated to be 1.3–67%.

Yamaguchi et al. [26] discussed the deterioration of radioactive Cs-bearing particles in the environment and determined the weathering of the particles and heterogeneity of the constituent Cs and Rb [13, 19]. The authors concluded that these phenomena are caused by several environmental factors including dissolution by cycling of wet and dry states as well as shape change caused by weathering kinetics of silicate glass. The lifetime of the particles is predicted to be shorter than several decades.

Okumura et al. [27] reported the stability of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  by heating radioactive Cs-bearing particles. The authors conducted a heating experiment on six radioactive particles and showed that the radioactivity decreased at 600 °C and was completely lost when heated to 1,000 °C. The size and characteristics of the heated

particles were unchanged after heating. Upon heating, elements other than Cs, such as Cl and K, were also lost as they were likely released from the particles in a similar manner. The authors also reported the heating experiment of radioactive Cs-bearing particles with soil samples and showed that the radioactivity in radioactive Cs-bearing particles moved to soil, causing significant dilution. They concluded that radioactivity in radioactive Cs-bearing particles can be decomposed by the treatment in incinerators with a burning temperature of approximately 900 °C.

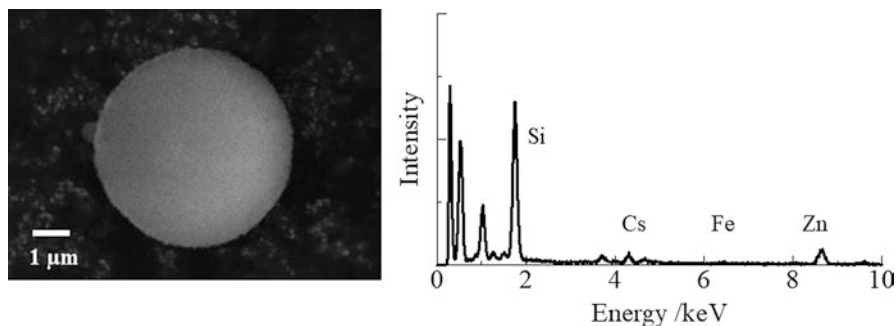
## 15.6 Production of Radioactive Cs-Bearing Particles

Radioactive Cs-bearing particles are not produced in the environment due to their high specific radioactivity and stability and are only produced in nuclear reactors and can be released by accidents. Determination of their production process is important to estimate the total amount of radioactivity released to the environment and to investigate the conditions in the nuclear reactor during the accident. Although the process of producing radioactive particles is unclear, several hypotheses have been proposed through detailed analysis of the produced radioactive particles.

The source of Si and O, the main components of the particle, also remains unclear. Kogure et al. [19] showed that  $\text{SiO}_2$  was derived from concrete and fine particles of  $\text{SiO}_2$  which were formed by the molten core concrete interaction (MCCI) after the meltdown. Satou et al. [16] indicated that  $\text{SiO}_2$  is also contained in the heat insulating material with a lower melting point than that of concrete and is located near the nuclear reactor containment vessel. Kobata et al. [28] characterized the production of  $\text{CsFeSiO}_4$  under high temperature conditions by CsOH chemisorption to stainless steel containing Si.

Kogure et al. [19] measured the diffusion coefficients of alkali metals and investigated that the diffusion coefficient of Cs is two orders of magnitude smaller than K or Rb. The different distribution of alkali metals is derived from the various rates of diffusion of volatilized alkali metals from the surface of the silicate particles. The authors also fabricated glass with the same composition in an attempt to reproduce the results of radioactive Cs-bearing particles. Imoto et al. [20] detected Zn-Fe-oxide nanoparticles in radioactive Cs-bearing particles, noting that iron nanoparticles are formed under high temperature conditions by MCCI and the evaporated  $\text{SiO}_2$  and CsOH can aggregate on nanoparticles. Ochiai et al. [21] pointed out that U is simultaneously taken into iron nanoparticles because U is found only in iron nanoparticles.

Fujita et al. [29] proposed a particle production process including the alkali melting reaction of CsOH. It is well-known that radioactive Cs evaporated from the fuel body in the form of CsOH, which is a strong base. The authors succeeded in producing particles by dissolving insulation materials or concrete mainly composed of  $\text{SiO}_2$  into droplets of CsOH melted at 300 °C as shown in Fig. 15.3.



**Fig. 15.3** SEM image of a reproducing radioactive Cs-bearing particle produced by the method of Fujita et al. [29] (left) and EDS spectrum of the reproducing particle (right)

## 15.7 Future Study Prospects

Herein, the author discussed the properties of concentrated radioactive particles found in the environment after the FNPP accident. These particles are insoluble in water and contain  $\text{SiO}_2$  as the main component. Although the production and distribution of radioactive Cs-bearing particles remain unclear, their detailed elemental composition is reported and classified into several distinct types of particles present in the environment. Studies related to particle classification and investigation of their production can clarify the release process of radionuclides at the time of the accident.

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