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Research article

An assessment on the environmental contamination caused by the Fukushima accident

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ABSTRACT

The radiological releases from the damaged fuel to the atmosphere and into the cooling water in the Fukushima Daiich Nuclear Power Plant (FDNPP) accident are investigated. Atmospheric releases to the land and ocean mostly occurred during the first week after the accident whereas continuous release from the damaged fuel into the cooling water resulted in an accumulation of contaminated water in the plant during last six years. An evaluation of measurement data and analytical model for the release of radionuclides indicated that atmospheric releases were mainly governed by the volatility of the radionuclides. Using the measurement data on the contaminated water, the mechanism for the release of long-lived radionuclides into the cooling water was analyzed. It was found that the radioactivity concentrations of ^{90}Sr in the contaminated water in the Primary Containment Vessel (PCV) of unit 2 and unit 3 were consistently higher than that of ^{137}Cs and the radioactivity concentration of ^{90}Sr in the turbine building of unit 1 in year 2015 was higher than that in year 2011. It was also observed that the radioactivity concentration of long-lived radionuclides in the contaminated water in the FDNPP is still high even in year 2015. The activity ratio of ^{238}Pu/$^{239+240}$Pu for the contaminated water was in the range of 1.7–5.4, which was significantly different from the ratios from the soil samples representing the atmospheric releases of FDNPP. It is concluded that the release mechanisms into the atmosphere and cooling water are clearly different and there has been significant amount of long-lived radionuclides released into the contaminated water.

1. Introduction

The accident at Fukushima Daiich Nuclear Power Plant (FDNPP) is the third major nuclear accident following the Three Mile Island accident in the United States and the Chernobyl accident in the former Soviet Union. These accidents resulted in a significant melting of the nuclear fuel and subsequent release of radioactive materials. However, the Fukushima accident is unique in the aspect of core melt progression and radiological releases compared to the Three Mile Island and the Chernobyl accidents.

In the Three Mile Island accident, the reactor vessel maintained its integrity even though more than one third of the fuel in the core was molten. While a significant fraction of noble gases was released into the environment, the release of other nuclides such as iodine and cesium was negligible (Electric Power Research Institute, 1980). In the Chernobyl accident, radionuclides and part of the damaged fuel were released into the atmosphere due to an explosion in the reactor core and a graphite fire. The radiological materials were widely spread over the European continent by the wind (IAEA, 1986; IAEA, 2006). The remaining portion of damaged fuel was fixed by pouring concrete to form a sarcophagus around the shattered reactor to prevent further release. Due to sarcophagus aging, construction of a new safe confinement shelter is in progress (Peplow, 2011).

The FDNPP is located on the north-eastern Pacific Ocean coast of Honshu, about 200 km north-east of Tokyo. The atmospheric releases that contributed most to the observed deposition in Japan occurred during the first week after the accident. During this period, the radionuclides were released to both the land and the ocean (IAEA, 2015). By combining environmental data with atmospheric dispersion simulations it was possible to estimate the atmospheric release of ^{131}I and ^{137}Cs in the early phase of the FDNPP accident (Katata et al., 2012). An analysis of regional-scale atmospheric dispersion and deposition suggested that the distribution of a large amount of ^{137}Cs deposition in eastern Japan was produced primarily by four events that occurred on March 12, 15–16, 20, and 21–23 (Terada et al., 2012). Seawater samples in the North Pacific Ocean in April and May 2011 showed that the ^{137}Cs activity originated from FDNPP ranged from 1 to 1000 Bq m^{-3} with activity ratios of ^{134}Cs/^{137}Cs close to 1 (Aoyama et al., 2013). The total

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amount of radiological releases to the atmosphere in the FDNPP accident was estimated as about 520 PBq, which is about one tenth of the releases of 5300 PBq in the Chernobyl accident (Steinhauser et al., 2014).

As cooling water has been continuously injected into the reactor vessel and the Primary Containment Vessel (PCV) at a rate about 300–400 ton/day during the last six years, it resulted in an accumulation of contaminated water due to the dissolution of various nuclides into the water (IAEA, 2015). Controlled discharges and leakages of the contaminated water from the FDNPP (IAEA, 2015) led to a contamination of the ocean.

As the configuration of damaged fuel in the core and in the primary containment vessel is yet unknown (TEPCO, 2015), further scientific investigations are needed for better understanding of the radiological releases from the FDNPP accident. In addition, unidentified plant responses during the accident progression inevitably accompanied large uncertainties in the prediction of radiological releases from the FDNPP (IAEA, 2015; Kim et al., 2017).

In this paper, the unique nature of the radiological releases of the Fukushima accident, including the amount and species of radionuclides released to the atmosphere and the cooling water, are analyzed and discussed using available measurement data and information.

2. Methods

2.1. Identification of radiological release mechanisms

The earthquake and subsequent tsunami resulted in a complete loss of all power and loss of the main safety functions. Due to the inability to supply cooling water to the reactor vessel, the decay heat generated by the nuclear fuel resulted in a fuel failure. The failure of the fuel was accompanied by hydrogen generation from the oxidation of the cladding surrounding the fuel, and subsequent melting of the fuel. The melted fuel that has relocated to the lower head of the reactor vessel led to a failure of the reactor vessel. Then, part of the melted fuel was discharged from the reactor vessel and relocated to the pedestal area (TEPCO, 2015) of the Primary Containment Vessel (PCV). During the accident progression, radiological materials are released from the damaged fuel either in the reactor vessel or in the pedestal floor area. Fission gases in the fuel-cladding gap and other radionuclides retained in the fuel matrix are released to the atmosphere in the form of gases and aerosols during the accident progressions in the FDNPP unit 1, 2, and 3 which occurred mostly within a week. The released radionuclides from the damaged fuel to the atmosphere are transported to the environment mainly by two release paths as illustrated in Fig. 1.

The first path into the environment is through the stack. As the PCV pressure approached the failure pressure due to an accumulation of hydrogen, steam, and non-condensable gases, plant operators had to open the vent valve connected to the stack during the progression of the accident. A second path was formed along the breaches in the reactor vessel, leakages in the head flange of PCV. The radionuclides released from the damaged fuel migrated through the second path and were accumulated in the reactor building. As the reactor building was destroyed by a hydrogen explosion (IAEA, 2015), it resulted in a release of radionuclides to the environment. Through these two releases paths, the radionuclides in the form of aerosols and gases were released to the atmosphere and finally deposited on land and sea (IAEA, 2015; Tsumune et al., 2013).

There is another mechanism of release of radionuclides into the cooling water which has been injected into the reactor vessel and the PCV for the removal of decay heat from the damaged fuel, at a rate of about 300–400 ton/day during the last six years. A conceptual picture of the injection of cooling water and releases of radionuclides into the cooling water is illustrated in Fig. 2.

The cooling water has been in direct contact with the damaged fuel, whose physical form could be a mixture of solid ingots or particulate debris beds. Therefore, there has been continuous dissolution of radionuclides from the damaged fuel at low temperature to the coolant (Burns et al., 2012). The radionuclides dissolved from the fuel into the cooling water are accumulated in the PCV and the turbine building in the form of contaminated water.

As there were enormous quantities of ground water about 1000 ton/day passing through the floor of the FDNPP and there were leakage paths from the nuclear power plant site to the sea, an unintended release of contaminated water to the sea have occurred (IRSN, 2016; Tsumune et al., 2013). A frozen soil wall around the four reactor units was installed by TEPCO to block the flow of groundwater and prevent its mixing with contaminated water (TEPCO, 2017). However, until recently, TEPCO had difficulties in isolating the contaminated water.

2.2. Use of measurement and monitoring data for the estimation of radiological releases

Depending on the species of radionuclides, release characteristics from the damaged fuel during the core melt progression varies. From the VERCORS program (Ducros et al., 2013), the nature of the release of fission products (FP) and actinides during a core melt accident is characterized by four groups. The four categories consisted of 1) volatile FP including fission gases, iodine, cesium, antimony, tellurium, cadmium, rubidium and silver with very high releases at temperatures of around 2600 K, 2) semi-volatile FPs, a category composed of molybdenum, rhodium, barium, palladium and technetium with releases of 50–100%, which are very sensitive to oxygen potential, 3) FP displaying low volatility, such as ruthenium, cerium, strontium, yttrium, europium, niobium and lanthanum with releases of around 3–10% on average, and 4) non-volatile FP composed of zirconium, neodymium and praseodymium.

Comparisons of the atmospheric releases of volatile fission products, less volatile fission products and refractory materials for the FDNPP and the Chernobyl accidents are presented in Table 1. The data were taken from various sources (IAEA, 1986; IAEA, 2006; IAEA, 2015; Nishihara et al., 2012; Schwantes et al., 2012). The initial inventory in Table 1 indicates the amount of radionuclides in the fuel at the time of the reactor shutdown.

It is shown that noble gases such as Kr and Xe, and volatile radionuclides such as cesium and iodine, were major radionuclides released into the atmosphere in the FDNPP and Chernobyl accidents. It is noted that the initial inventory of radionuclides in the fuel of FDNPP is greater than that of the Chernobyl.

As the contaminated water stored in the PCV and the turbine building of FDNPP was released to the ocean by controlled the releases and leakages (IAEA, 2015; Tsumune et al., 2013), it was necessary to analyze the monitoring data for the radionuclides in the contaminated water and sea water near the FDNPP to estimate the amount and species of radionuclides released.

In this paper, continuous measurement data for concentration of ^{137}Cs in the contaminated water (TEPCO, 2016a) during last six years were used to estimate the amount of dissolution of ^{137}Cs from the damaged fuel into the cooling water. Also, measurement data for various radionuclides in the contaminated water of the PCV (JAEA, 2016) for units 2 and 3 and the turbine building of unit 1 (JAEA, 2017a) are analyzed to evaluate the characteristics of release into the water. The water samples were analyzed for ^{3}H, ^{60}Co, ^{90}Sr, ^{94}Nb, ^{106}Ru, ^{137}Cs, ^{144}Ce, ^{152}Eu, ^{154}Eu, ^{238}Pu, $^{239+240}$Pu, ^{241}Am, ^{242}Cm, and ^{244}Cm. Water samples from inside the PCV were collected at two depths: near the water surface (app. 0.1 m deep) and in the