Study on Formation of Volatile Organic Iodides under Severe Accident Conditions

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1. Introduction

The behavior studies of radioiodine are required to evaluate the source term of volatile radionuclides when a severe accident of a nuclear power plant occurs. In particular, the formation of radioactive organic iodides, such as CH_3I and C_2H_5I, causes considerable concern for the capture of radioactivity [1] because these species are hardly soluble in water. Under accident conditions, the radioactive organic iodides are generally formed by the reactions of radioactive iodine and organic compounds [2,3]. The radioactive iodine is released from the damaged fuels, and the organic compounds come from the organic materials such as epoxy paints and cable covers used in reactor building.

Various research works [1-11] have been carried out for evaluating the formation of radioactive organic iodides. Research activities include the amounts of radioactive iodine in the irradiated fuels, the ratio of iodine released from the damaged fuels, and the transport of iodine through the primary circuit of the reactor. In addition, the iodine chemistry, aerosol physics, and the iodine behavior based on the adsorption and desorption in the gaseous phase were also studied. Furthermore, the formation process and mechanism of organic iodides were studied under various irradiation conditions.

In the current work, we studied the formation of volatile organic iodides under the gamma irradiation conditions. The formation of CH_3I was influenced by various environmental factors such as the gamma dose and the solution pH. Our results will be helpful to understand and predict the formation of organic iodides under the accident condition of nuclear power plant.

2. Methods and Results

In this section some of the techniques used in this study and some experimental results are described.

2.1 Gamma Irradiation Facility and Devises

The gamma irradiation experiment was carried out at a high dose rate gamma irradiation facility. The gamma dose rates we used were maximum 10 kGy h^{-1}. The NaI with organic compound solutions were prepared in 20 ml vials. After gamma irradiation, toluene was injected into the vials using a gas-tight glass syringe, and the liquid/liquid extraction was performed to extract organic iodides into the toluene phase. After the liquid/liquid extraction, we sampled the 1 mL of the solution from the toluene phase by the syringe. After sampling, we measured the concentrations of CH_3I dissolved in toluene using a GC–MS (Perkin Elmer Clarus 680/SQ 8T, USA). The detection limit of the CH3I by GC–MS was determined to be 0.048 uM, and the extraction yield of CH_3I was ~85%.

2.2 The pH changes of alkaline solutions

Figure 1 shows the pH changes of NaOH solutions under gamma irradiation condition. It is shown that the pHs of the solutions decrease from pH 12 to pH 11.2 and from pH 11 to pH 6.4 respectively after 400 kGy gamma irradiation. It is known that the cause of the pH decrease is attributed by the air radiolysis products (NO_2, etc.) acting as acids. When the pH is lowered below 7, I_2 is chemically stable and organic iodides can be formed in this pH range.

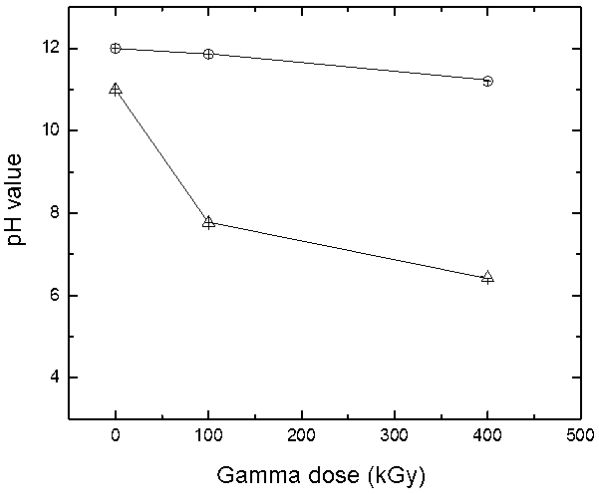

Fig. 1. The pH changes of NaOH solutions after high dose gamma irradiation. ○: 10 mM NaOH, △: 1 mM NaOH

2.3 Formation of CH_3I in presence of decomposition products of organic compounds

Chemical bonds of organic compounds are not easily damaged under gamma irradiation conditions. However, in the presence of water molecules, organic compounds are easily decomposed into small molecules by radiolysis products of water such as OH radicals. The decomposed small molecules such as acetic acid and formic acid are finally decomposed into single molecules such as CO_2 or H_2.

In our experiment, we investigated whether acetic acid or formic acid can act as a source of organic iodines. As a result of the experiment, it was confirmed that both organic acids can form CH_3I, the most basic organic iodide, under certain conditions. This result indicates that most organic material can act as a source forming organic iodides.

3. Conclusions

We confirmed that 0.01 M NaOH solution of the pH 12 decreases below pH 7 after 800 kGy gamma irradiation. It means that the iodide can be oxidized to I_2 by the continuous gamma irradiation, even though the initial pH values of the solutions are very high. We also confirmed that CH_3I was formed by the reaction of I_2 with organic compounds, even their molecular weight is very small, under gamma irradiation conditions. This indicates that iodines adsorbed on the surface of the organic materials could easily covert to the organic iodides in case of the severe accident.

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