Transactions of the Korean Nuclear Society Autumn Meeting
Changwon, Korea, October 20-21, 2022

Understanding on Chemical Behavior of Iodine under Gamma Irradiation Conditions

Jei-Won Yeon, Minsik Kim
Nuclear Chemistry Research Team, Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon 34057, Republic of Korea
[*]Corresponding author: yeonysy@kaeri.re.kr

1. Introduction

After the accident of Fukushima nuclear power plant, the public's concern about the nuclear accident increased. At the same time the study on the nuclear accident began to be highlighted again. Our research group also started the study on the nuclear accident from the period. Radioactive iodine is a radionuclide of great interest in the early stages of a nuclear accident due to its relatively short half-life (~ 8 days) and volatile properties. And since iodine changes in volatility depending on its chemical oxidation state, it is one of the important parts to evaluate the behavior of iodine in the field of nuclear accidents.

At the beginning of our study, the oxidation behavior of iodine under the gamma-irradiation condition was studied[1-4]. Our gamma irradiation experiments were performed mainly by analyzing volatile iodine species present in the iodine solution after irradiation. However, we found that this kind of experiment had a limitation that it could not explain the phenomenon of volatilization of iodine at high pH even though it was a trace amount.

This study introduces a method to understand the volatilization behavior of iodine by separating the gamma oxidation of iodide ion[1,2] and reduction (or disproportionation) of molecular iodine[5] and evaporation behaviors of iodine[6]. Using this method, the change in the volatile behavior of radioactive iodine has been understood according to the solution pH, which was obtained by AECL-RTF test.

2. Methods and Results

2.1 Chemicals and Instruments

All chemicals used in this work were analytical reagent grade. The experimental solutions were made from NaI (≥ 99.5 wt.%), molecular iodine (≥ 99.8 wt.%), toluene (≥ 99.8 wt.%) and the deionized water (> 18.2 MΩ·cm, EMD-Millipore). The I_3^- and I_2 concentrations were respectively measured by the absorbance values at the characteristic wavelengths (I_3^-: 351 nm, I_2: 309 nm). We used a UV-VIS spectrophotometer (WPA II, Biochrom Ltd., Cambridge, UK) to obtain UV-VIS spectra and a pH meter (Orion Star A211, Thermo Fisher Scientific, Waltham, MA, USA) to control or measure the pH of the experimental solutions.

2.2 Gamma Irradiation

Gamma irradiation experiments were carried out at the same irradiation system (MDS Nordion, Canada) as previously used [1-3]. The gamma dose was controlled by adjusting the distance of the sample from the ^{60}Co source and the irradiation time, and the dose rate was confirmed before each irradiation experiment using a dosimeter (Alanine Dosimeter Reader, E-Scan, Bruker). The gamma dose rates were controlled in the range of 0-10 kGy h^{-1}. The irradiation experiments were conducted at a temperature of 25±2°C.

2.3 Prediction of Iodine Volatilization

To analyze the volatilization behavior of iodine, the gamma oxidation rate of iodine, the reduction rate of iodine, the disproportionation rate of iodine, and the evaporation rate of iodine were determined respectively. Currently, only a relative prediction of iodine volatility is possible with our data, so we adjusted the predicted volatility of iodine by inputting an arbitrary constant to match the absolute value of the trace amount of background volatility observed at high pH, which is the test result of AECL-RTF.

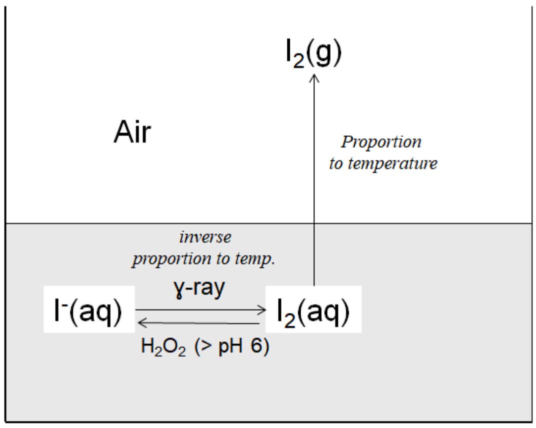

Fig. 1. Paths of iodine from I- ion dissolved in solution to I_2(g) in the air.

The increase in iodine volatility when the solution pH was decreased could be easily predicted only by the gamma oxidation behavior of iodine. However, it could be understood only through the separation evaluation method that the gradual decrease in volatility is due to the accumulated I_2 in the solution when the pH of the solution is suddenly increased. In addition, it was possible to calculate a very small amount of volatility at high pH through the method. In particular, it could be understood that the irregular change in iodine volatility with temperature was caused by the different effects of temperature on gamma oxidation and evaporation reactions.

3. Conclusions

The volatility analysis method that separated the gamma oxidation, reduction (disproportionation), and evaporation steps of iodine could well explain the volatilization behavior of iodine reported as a result of AECL-RTF. In particular, it was possible to understand the gradual decrease in iodine volatility when the pH of the iodine solution was suddenly increased, and the nonlinear change in iodine volatility according to the temperature increase.

Acknowledgements

This work was supported by the Korean government (MSIT: the Ministry of Science and ICT) (NRF Nos. 2017M2A8A4015281, 2021M2E3A3040092, and RS-2022-00144202) and KAERI Institute Program (Project No. 522330-22).

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