Concentration measurement of volatile iodine species formed by thermal decomposition of iodate salt

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1. Introduction

In the event of a severe accident at a NPP, a large amount of radioactive aerosols could be released from the irradiated fuels [1-4]. Iodine oxides among the volatile radioactive aerosols have been reported to convert into I_2 under operating conditions of passive autocatalytic recombiner (PAR), and then the I_2 may adsorb on catalysts and reduce the performance of PAR [1-4]. Therefore, in order to suppress the performance degradation of PAR at high temperature and understand the behavior of iodine species at high temperature, it is necessary to understand the thermal decomposition behavior of iodate.

In this study, we investigated the thermal decomposition behavior of $NaIO_3$, one of the iodine salts (IO_x), under various conditions. And the effects of H_2 and H_2O on the decomposition behavior were analyzed by measuring the volatile iodine species. Our experimental results showed that the I_2 and NaI were easily formed by the decomposition of the iodates under our experimental conditions. The amounts of I_2 formed at 550℃ were about 1.3 times more compared to those formed at 650℃.

2. Experimental and Results

2.1 Experimental

All chemical reagents were analytical grade. $NaIO_3$ (≥ 99%, Sigma-Aldrich), The biphasic reagents and alkaline solution were made from toluene (≥ 99.8 %, Sigma-Aldrich), HNO_3 (70 %, Sigma-Aldrich), deionized water (>18.2 MΩ•cm, EMD-Millipore) and NaOH (≥ 97%, Sigma-Aldrich).

The thermal decomposition system used in the experiment is composed of 4 parts. First, gas supplies parts including MFC (mass flow controller) and humidifier. Second, furnace parts including quartz boats for loading sample. Third, iodine species capturing parts using biphasic gas scrubbers. Fourth, outlet line is then terminated by gas scrubbers filled with a diluted alkaline solution to trap other gaseous species. The schematic diagram of the decomposition system is shown in Fig. 1.

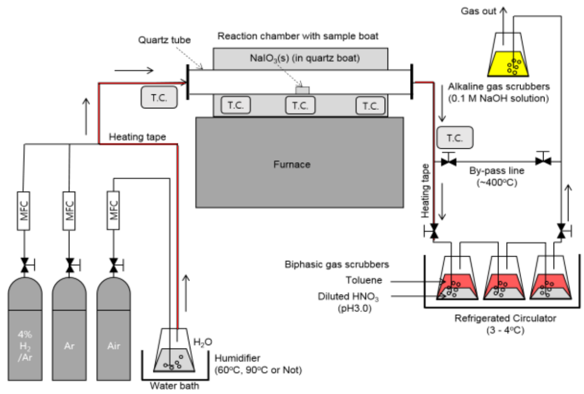

Fig. 1. Schematic diagram of thermal decomposition system

2.2 Concentration of I_2 formed from thermal decomposition $NaIO_3$

We investigated the effects of H_2 and H_2O on the formation of volatile I_2 at high temperature. At 400℃ under all gas conditions, no iodine species could be observed in biphasic gas scrubbers. Whereas the amounts of I_2 at 550℃ were up to 1.3 times more compared to those formed at 650℃. Exceptionally, the I_2 amount formed at 650℃ with H_2O condition a little increased more compared to that at 550℃ with H_2 condition (dry condition).

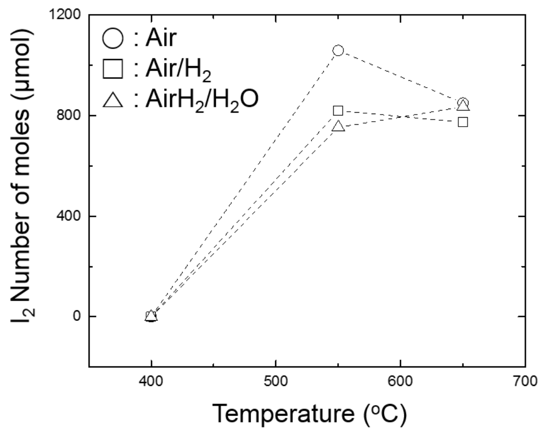

Fig. 2. Amounts of I_2 formed from decomposition of $NaIO_3$ at different temperature and gas conditions

2.3 Concentration of I^- and water-soluble iodine species formed from thermal decomposition $NaIO_3$

We investigated the effects of H_2 and H_2O on the formation of water-soluble iodine species (I^-, IO, IO_3^-) at high temperature. At 400℃ under all gas conditions, no iodine species were detected in biphasic gas scrubbers. Other iodine species were detected at the temperature above 550℃, and the major species of the other iodine species were evaluated to be I^- by using UV-VIS spectrophotometer and ICP-MS.

At 550℃, a very small amount of I^- was detected. On the other hand, at 650℃, a relatively large amount of I^- was formed. In particular, the amount of I^- formed under H_2 (reducing) condition was greater than that under air (oxidizing) condition. The amount of I^- formed at H_2O condition was relatively increased compared to those under other conditions.

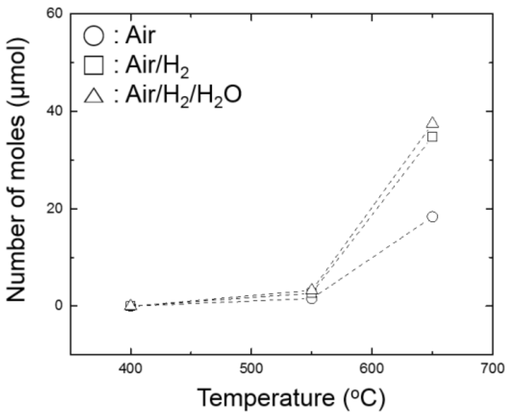

Fig. 3. Amounts of I^- formed from decomposition of $NaIO_3$ at different temperature and gas conditions

3. Conclusions

We investigated the thermal decomposition behavior of $NaIO_3$ under various conditions. The differences of I_2 amounts were explained by using different decomposition paths of $NaIO_3$ depending on the composition of gases and temperature. And the differences of I^- amounts were can also be explained in a similar way.

$$4NaIO_3 \rightarrow 2Na_2O + 2I_2 + 5O_2 \quad (> 510℃) \quad (1)$$
$$2NaIO_3 \rightarrow 2NaI + 3O_2 \quad (> 460℃) \quad (2)$$

On the presence of steam, thermal decomposition reaction of the first path (eq. 1) is more dominant. On the other hand, on the presence H_2, I_2 can be converted into I^-. The formation of iodine species from thermal decomposition were complexly affected by temperature and gas compositions.

Acknowledgments

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REFERENCES

[1] E. Hoinkis, Review of the Adsorption of Iodine on Metal and its Behavior in Loops (No. ORNL-TM-2916), Oak Ridge National Laboratory, Tennessee, 1970.
[2] F. Morfin, J.C. Sabroux, and A. Renouprez, Catalytic Combustion of Hydrogen for Mitigating Hydrogen Risk in Case of a Severe Accident in a Nuclear Power Plant: Study of Catalysts Poisoning in a Representative Atmosphere, Applied Catalysis B: Environmental, Vol.47, p. 47, 2004.
[3] L. Bosland, F. Funke, N. Girault, and G. Langrock, Paris Project: Radiolytic Oxidation of Molecular Iodine in Containment during a Nuclear Reactor Severe Accident. Part 1. Formation and Destruction of Air Radiolysis Products. Experimental Results and Modeling, Nucl Eng Des., Vol.238, p. 3542, 2008.
[4] J.C. Sabroux, F. Deschamps, Iodine Chemistry in Hydrogen Recombiners, InEurosafe Forum, Brussels, 2005.