Study on formation of CH_3I in NaI and $CH_3COC_2H_5$ (MEK) solution under gamma irradiation

Minsik Kim[a, b], TaeJun Kim[a, b], Jei-Won Yeon[a, b*]

[a]*Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, Daedeok-daero 989-111, Yuseong-gu, Daejeon 34057, Republic of Korea*
[b]*Radiochemistry and Nuclear Nonproliferation, University of Science and Technology, Korea, Gajung-ro 217, Yuseong-gu, Daejeon 34113, Republic of Korea*
[*]*Corresponding author: yeonysy@kaeri.re.kr*

1. Introduction

The chemical reaction and behavior data of iodine under gamma irradiation are essentially required to evaluate the source term of volatile radionuclides when a severe accident of a nuclear power plant occurs [1-7]. In particular, the formation of radioactive organic iodides, such as volatile CH_3I and C_2H_5I, causes considerable concern for the capture of radioactivity [8, 9] because both solubility are very low in water.

Many research scientists have been carried out pertaining to the radioactive organic iodide [6, 10]. The activities include the inventory of radioactive iodine species in irradiated nuclear fuels, the amounts of iodine released from the fuels under accident conditions, and the transport of iodine through the primary coolant circuit [11, 12]. Additionally, the iodine chemistry, aerosol physics, and the iodine behavior based on the adsorption and desorption in the gaseous phase were studied [12]. The formation rate and mechanism of organic iodide are still on-going research topics [10, 13-16].

In this study, we interpreted experimental results obtained from the gamma irradiation of NaI and MEK solutions by using CH_3I formation process, which was established in previous work [17].

2. Methods and Results

In this section the experimental method used in this work and results are described.

2.1 Experimental

The source of the high gamma radiation dose in the experiment was composed of ^{60}Co pieces with a source activity of about 280 kCi. The gamma dose rate was controlled to be within a range of 0.4 to 10 kGy h^{-1}. The schematic diagram is shown in Fig. 1. The experimental solutions were made from NaI (99.5 wt%, Sigma-Aldrich), MEK (2-butanone, 99.7%, Sigma-Aldrich), toluene (99.9%, Sigma-Aldrich), and CH_3I (99%, Sigma-Aldrich). The pH values of the solutions were maintained in the pH range of 6.7-6.9 before gamma irradiation. The concentrations of CH_3I dissolved in toluene were measured using a GC–MS (Perkin Elmer Clarus 680/SQ 8T, USA).

2.2 Formation process of CH_3I from NaI and methyl alkyl ketones

In previous work [17], a formation process of CH_3I was established as shown in Fig. 2.

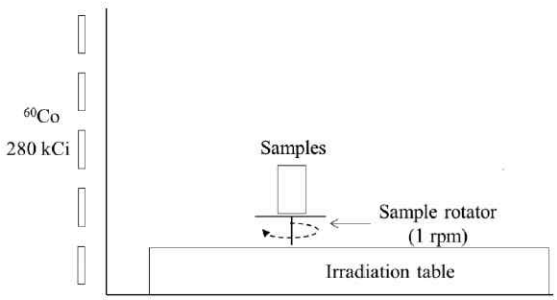

Fig. 1. Schematic diagram of ^{60}Co irradiation facility

At a pH below 6, I_2 is stable and reacts again with CH_3 radical, which is a decomposition product of methyl alkyl ketone, to form CH_3I. And then, CH_3I is finally formed in this condition. The first stage of CH_3I formation is the pH decrease by the decomposition of methyl alkyl ketone, since the gamma oxidation of I^- into I_2 can progress substantially in the pH range below 6.

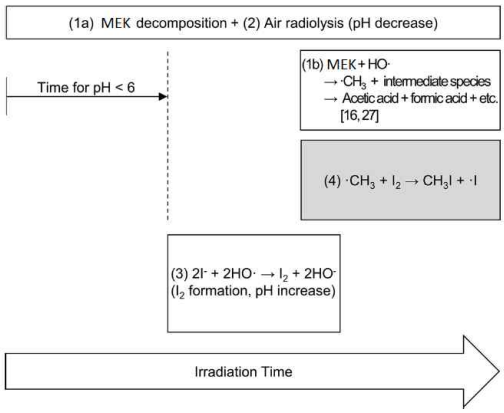

Fig. 2. Diagram of the CH_3I formation processes in NaI and

MEK mixed solutions under gamma irradiation

2.3 Formation of CH_3I in MEK and NaI solutions under gamma irradiation

Figure 3 shows that the concentration of CH_3I formed in the NaI and MEK mixed solutions under 10 kGy h^{-1}. We did not observe a significant amount of CH_3I in 0.1 mM MIBK mixed solutions. Based on the formation process, it indicates that a 0.1 mM concentration of MEK is too low to reduce the solution pH below 6. At 1.0 mM NaI and 1.0 mM MEK, the CH_3I formation was detected and the amount of CH_3I increased with increase in the gamma dose. In the 1.0 mM NaI and 5.0 mM MIBK solution, the amount of CH_3I also increased with increase in the gamma dose. And a considerable concentration of 1.6 μM CH_3I was observed at the irradiation of 40 kGy.

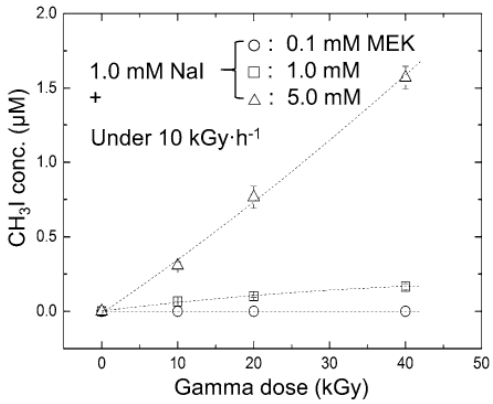

Fig. 3. The concentrations of CH_3I formed in NaI and MEK solutions under 10 kGy h^{-1} gamma irradiation condition.

3. Conclusions

We interpreted experimental results obtained from the gamma irradiation of NaI and MEK solutions by using CH_3I formation process, which was established in previous work [17]. First, it was confirmed that CH_3I was formed in NaI and MEK mixed solutions under gamma irradiation. And we confirmed that the formation amount of CH_3I was well accorded with the CH_3I formation process.

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