

Effect of Gamma Irradiation on Formation of Organic Iodide in Iodide and Ketone Mixed Solutions

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The aim of this study is to quantitatively measure the concentration of methyl iodide (CH₃I) and ethyl iodide (C₂H₅I) formed in methyl ketones under gamma irradiation in the range of 0 to 40 kGy. For each 1.0 mM of MIBK and MEK in the presence of NaI under 0 - 40 kGy irradiation, CH₃I was formed in a concentration range of 0 - 1.82 μM. The concentration of organic iodide generally increased, as the gamma dose and concentrations of MIBK and MEK increased.

I. INTRODUCTION

In the event of a severe accident in a nuclear power plant, the volatile fission products will be released into the containment from the damaged fuel bundles. It was reported that most of the iodine is accumulated in the sump water through both aerosol particles and a gaseous form.¹ Radioactive iodide can be oxidized into volatile iodine.^{2,3} Moreover, under gamma irradiation, organic iodides can be formed as a result of the interaction of radioactive iodine with organic compounds such as paint.⁴ Under this condition, the radiolytic decomposition of organic compounds can influence the formation of organic iodides.⁵ As one of the organic compounds, the decomposition of MEK has been successfully demonstrated using kinetic models.^{5,6} DRIVER et al.⁶ found that organic compounds will be decomposed into CO₂ and organic acids, while reducing the pH of the solution. Approximately 90% of 1 mM (mmol/l) MEK was decomposed over 4 kGy of irradiation, with a minimum pH being reached within 1 h.^{5,6} They also insisted that radiation rapidly decomposes organic solvents such as MIBK and MEK in the aqueous phase. Although many studies have been carried out on the formation of organic iodides.^{4,7,8} Additional studies will be helpful for a better understanding of the effect of gamma irradiation on the formation of organic iodide.

In this paper, we measured the concentration of organic iodide formed in iodide and under various concentrations of methyl ketone mixed solutions after gamma irradiation. We also investigated the major factors affecting the formation of organic iodides.

II. EXPERIMENTAL

The gamma irradiation experiment was carried out at a high gamma irradiation facility. The total dose was controlled to in the range of 0 - 40 kGy, and the temperature of irradiation was at 18° - 20°C. Four different NaI solutions of 0.1 to 5.0 mM with methyl ketone (MIBK and MEK) were prepared in 20 ml sealed vials. All vials were sealed with a Teflon cap to avoid evaporating the volatile organic iodides. After gamma irradiation, about 5 ml toluene was injected into the sealed vials using a gas-tight syringe. Liquid/liquid extraction was then performed for 1.5 hours to extract organic iodides into toluene phase. After extraction, we measured the concentrations of CH₃I and C₂H₅I dissolved in toluene using GC-MS (Perkin Elmer Clarus680/SQ8T, USA).

III. RESULTS

After 0 - 40 kGy of gamma irradiation of NaI (0.1 - 5 mM) and ketone (1.0 mM MIBK, 1.0 mM MEK) mixed solutions, we found that CH₃I was formed in a concentration range of 0.071- 1.82 μM. As total dose increased, the concentration of CH₃I formed generally increased. Unlike CH₃I, C₂H₅I was difficult to be observed. C₂H₅I was only detected in 5.0 mM MEK with 1.0 mM NaI after irradiation of 16 - 40 kGy. The concentration of organic iodides formed after irradiation increased, as the concentration of MIBK and MEK increased, respectively, in the presence of iodide. We found the concentration condition of MIBK under which CH₃I was formed to be the maximum concentration after irradiation. Consequently, we confirmed that both iodide and methyl ketone (CH₃-CO-) functional groups are the major ingredients for the formation of methyl iodide under gamma irradiation.

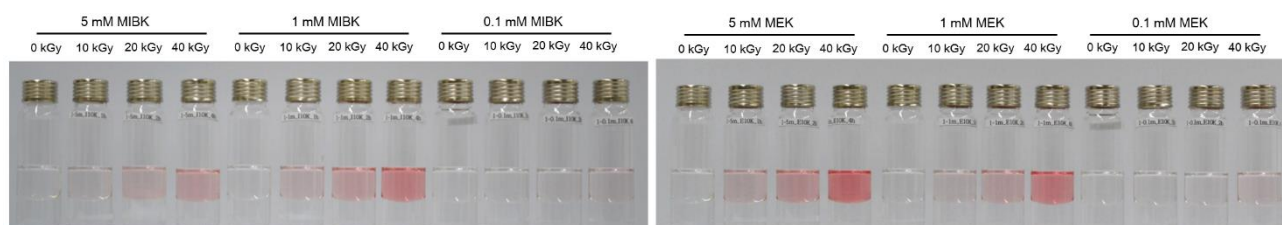


Fig.1 Photograph of vials after 0 - 40 kGy gamma irradiation of 1.0 mM NaI with three-different concentrations of ketone mixed solutions. The temperature in the irradiation laboratory was 18°C. The vials were replaced with new ones after gamma irradiation.

IV. CONCLUSIONS

We determined the concentration of organic iodides formed in iodide and ketone mixed solutions after gamma irradiation (0 to 40 kGy). The concentration of CH₃I increased as the gamma dose, and the concentrations of iodide and methyl ketone, increased. We found the experimental conditions under which the amounts of organic iodides were observed to be the maximum values.

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REFERENCES

1. J. C. WREN, J. M. BALL, and G. A. GLOWA, "Studies on the Effects of Organic-Painted Surfaces on pH and Organic Iodide Formation," OECD Workshop on Iodine Aspects of Severe Accident Management Workshop Proceed., Vantaa, **181**, 196 (1999)
2. C. C. LIN, "Chemical Effects of Gamma Radiation on Iodine in Aqueous Solutions," *J. Inorg. Nucl. Chem.*, **42**, 1101 (1980)
3. S.-H. JUNG et al. "The Oxidation Behavior of Iodide Ion under Gamma Irradiation Conditions." *Nuclear Science and Engineering*, **191**, 203 (2015)
4. F. TAGHIPOUR, and G. J. EVANS, "Radiolytic Organic Iodide Formation under Nuclear Reactor Accident Conditions," *Environmental Science & Technology*, **30**, 17 (2000)
5. J. C. WREN, J. M. BALL, and G. A. GLOWA, "The Interaction of Iodine with Organic Material in Containment," *Nuclear technology*, **337**, 362 (1999)
6. P. DRIVER, G.A. GLOWA, and J.C. WREN, "Steady-State γ -Radiolysis of Aqueous Methyl Ethyl Ketone (2-butanone) under Postulated Nuclear Reactor Accident Conditions," *Radiation Physics and Chemistry*, **37**, 51 (2000)
7. T. I. GORBOVITSKAYA et al. "Formation of Methyl Iodide in the Radiolysis of Aqueous Cesium Iodide and Acetic Acid Solutions," *High Energy Chemistry*, **161**, 165 (2001)
8. K. MORIYAMA et al. "Experiments on the Release of Gaseous Iodine from Gamma-Irradiated Aqueous CsI Solution and Influence of Oxygen and Methyl Isobutyl Ketone (MIBK)," *Journal of Nuclear Science and Technology*, **229**, 237 (2010)