The pH Change of Ketone and Iodide Solutions after Gamma irradiation

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1. Introduction

The pH values of iodide and ketone solutions under gamma irradiation are essentially required to evaluate volatility of radionuclides under accident conditions [1-7]. In particular, the iodine is generally released from damaged fuels as iodide aerosol including CsI and other hydroxide species [1-5, 8, 9], and then dissolved in coolant and oxidized into I_2. After that, I_2 can be changed to volatile organic iodide such as CH_3I by reacting with volatile organic materials [10-17]. During the formation of volatile organic iodides from iodide ions, the solution pH is one of the important factors to control the reactions related to the volatility of iodine species [1, 2, 4, 5, 11-14, 18, 19]. In this study, we investigated the effects of ketone and iodide on the solution pH under gamma irradiation.

2. Methods and Results

2.1 Experimental

The high gamma radiation was carried out by using Co-60 source. 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), MIBK (4-methyl-2-pentanone, 99.5%, Sigma-Aldrich), MEK (2-butanone, 99.7%, Sigma-Aldrich), and the deionized water (>18.2 MΩ·cm, EMD-Millipore). A pH meter (Thermo scientific Orion Star A211) calibrated with three different buffer solutions. The pH measurements of all the samples were carried out before and after gamma irradiation under same temperature.

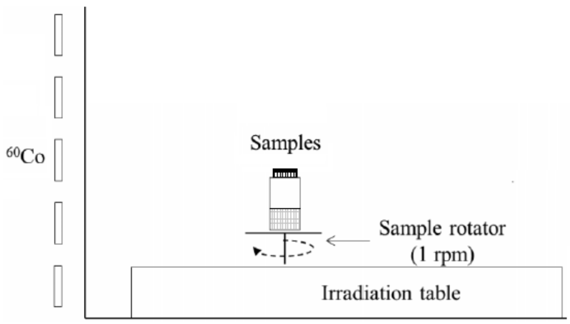

Fig. 1. Schematic diagram of Co-60 gamma irradiation facility.

2.2 Effect of ketones on the pH changes of solutions under gamma irradiation

Three different solutions of 1.0 mM MIBK, 1.0 mM MEK and deionized water were irradiated respectively and measured pH values after irradiation were showed in Fig. 2. Both types of ketone solutions had similar pH decrease by gamma irradiation. Within 10 kGy irradiation, the pH of two ketone solutions changed from about 7.5 to below 5. The pH decreased to about 4.5 after 20 kGy irradiation. In contrast, the pH of deionized water was almost unchanged after irradiation. The decreases in pH were evaluated that the ketones decomposes to form acidic chemical species.

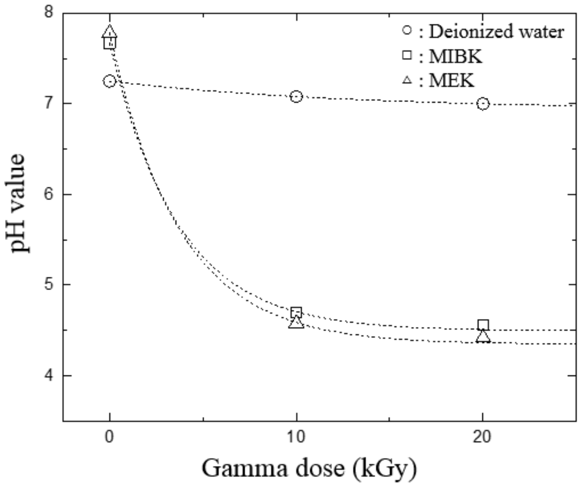

Fig. 2. The pH changes of three different MIBK, MEK, and deionized water solutions after gamma irradiation.

2.3 Effect of iodide on the pH changes of solutions under gamma irradiation

Figure 3 shows that the pH in the methyl alkyl ketone and NaI mixed solutions under 40 kGy. In the two ketones with NaI mixed solutions, the pH increased as the concentration of NaI increased after gamma irradiation. At 0.1 mM NaI with methyl alkyl ketone mixed solutions, the pH decreased from about 7.3 to 4. On the other hands, at 5.0 mM NaI with methyl alkyl ketone mixed solutions, the pH decreased a little from about 6.8 to 6.3. In this phenomenon, at 0.1 mM NaI and methyl alkyl ketone mixed solutions, the amount of oxidation of I^- into I_2 is relatively small compared to the over 1.0 mM concentration of NaI conditions. These results showed that the gamma oxidation of I^- into I_2 can increased pH in ketone and iodide mixed solutions.

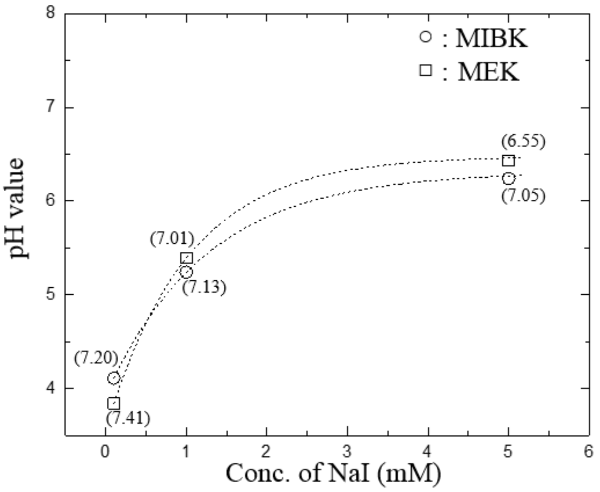

Fig. 3. The pH changes of three different concentrations of NaI in ketone solutions after gamma irradiation: Initial pH values in parentheses.

3. Conclusions

The methyl alkyl ketones reduced the solution pH after gamma irradiation. It was understood that they were decomposed by •OH into smaller molecules such as acetic and formic acids. There were few differences in pH decrease between MIBK and MEK solutions after gamma irradiation. In presence of iodide ion, the pH values were small changed with increase in the concentration of NaI in ketone and iodide mixed solutions.

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