Radiolysis of Boric Acid Solutions under Mixed Thermal and Fast Neutrons

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1. Introduction

Nuclear reactions of neutron absorbers such as ^{10}B or ^{6}Li in solid materials with thermal neutrons have been widely studied, and their applications are quite broad [1,2]. Nevertheless, as we know, the thermal neutron irradiation of water samples containing neutron absorbers has not been published except for a paper reporting the experimental data obtained at high temperatures [3]. However, irradiation [4] or simulations [5,6] of water and voluminous liquid samples with fast neutrons and gamma rays are frequently discussed in several published papers.

Several water samples containing ^{10}B-enriched boric acid, and natural and ^{10}B-enriched mixed boric acids in the range of 0 to 2000 μg/mL for the function of ^{10}B concentration, were irradiated to study the radiolysis of the cooling water containing boric acid. The concentration of natural boron in the primary coolant of pressurized water reactors (PWRs) is known to start at 1500 μg/mL, and boric acid is used for the purpose of nuclear reaction control.

2. Experimental

Different amounts of natural (H_3BO_3, 99.999%, Aldrich) and ^{10}B-enriched ($H_3{}^{10}BO_3$, 99%, Aldrich) boric acids were dissolved in deionized water at various sample concentrations. 1 or 2 mL of each prepared boric acid solution was poured in a one-edge sealed quartz tube and placed under a vacuum to 1×10^{-4} torr in liquid nitrogen. The other edge of the tube was sealed to a bullet-shaped ampoule (diameter: 8 mm, height: 55 mm, and thickness: 2 mm) and was put in an aluminum (Al) inner capsule and then a radio isotope (RI) target Al capsule. The samples were irradiated during 60, 120, and 240 min relatively in a IP04 hole (Thermal neutron flux : 2.81×10^{13} n cm^{-2} sec^{-1} ; fast neutron flux : 7.46×10^{9} n cm^{-2} sec^{-1}) of the high-flux advanced neutron application reactor (HANARO) at the Korea Atomic Energy Research Institute (KAERI) [7].

The radiolysis of a boric acid solution under mixed thermal and fast neutrons was examined quantitatively and qualitatively using a inductively coupled plasma mass (ICP-MS) spectrometer for measuring the ^{10}B and ^{11}B concentration ratio; a flameless atomic absorption (AA) spectrometer for the concentration of a product, ^{7}Li,; a gas mass (Gas-MS) spectrometer for the measurement of H_2, O_2, and H_2O concentrations; and titration methods for the H_2O_2 concentration.

3. Results and discussion

The experimental data of water decomposition to gases by the irradiation of boric acid solutions in the IP04 hole are shown in Figs. 1 and 2. Each of the produced total gas pressures in the examined ampoules shown in Fig. 1 was converted into the total gas volume shown in Fig. 2.

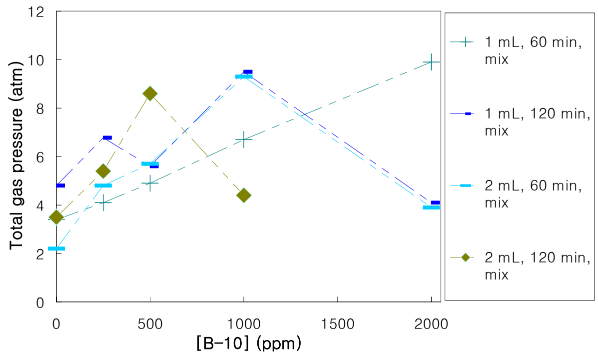

Fig. 1. Pressure changes of produced gases according to the concentration of ^{10}B after irradiation of boric acid (natural and ^{10}B-enriched mixed boric acid) solutions in a IP04 hole of the HANARO research reactor.

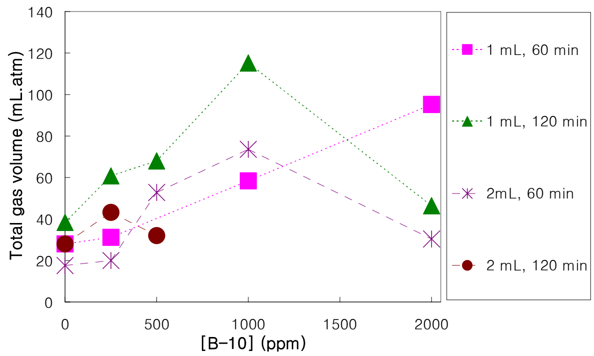

Fig. 2. Volume changes of the produced gases according to the concentration of ^{10}B after the irradiation of boric acid (^{10}B-enriched boric acid) solutions in a IP04 hole of the HANARO research reactor.

The boric acid concentration increased the extent of water decomposition compared to the absence of boric acid because of the nuclear effect from $^{10}B(n, \alpha)^{7}Li$, which is due to the radiation issued from a ^{10}B reaction with thermal neutrons. Some cases, a big pressure difference between the inside of the ampoule containing high concentration of ^{10}B and outside of it caused a flame spark when the ampoule was broken for a gas analysis, so the gas amount and ampoule pressure at the point were out of linearity and dropped suddenly. The flame spark consumed H_2 and O_2 gases and produced CO_2 gas.

To support the data shown in Figs and know the radiolysis degree of boric acid solutions under neuron irradiation, pH changes of the boric acid solutions, and the concentrations of produced ^{7}Li and diminished ^{10}B were also obtained.

4. Summary

To study the radiolysis of the cooling water and calculate the amount of gases that may have been produced in this manner, we irradiated several water samples containing ^{10}B-enriched boric acid, and natural and ^{10}B-enriched mixed boric acid. The amount of gases produced from water radiolysis in a fast neutron flux was insignificant compared to the gas amount obtained from the radiolysis of a boric acid solution in a thermal neutron flux.

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