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Quantity of Non-Condensing Gases (Such as Hydrogen and Oxygen) Produced from the Irradiation of Boric Acid Solutions in a Research Reactor

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1. Introduction

Several pipeline rupture accidents as a result of the detonation of a hydrogen-oxygen mixture accumulated due to radiolysis have been reported [1, 2]. However, non-condensing gases (such as oxygen and hydrogen molecules) are usually not detected in cooling systems of pressurized water reactors (PWRs) under high temperature, or research reactors even under room-temperature irradiation conditions. In the meantime, it was observed that the presence of certain amounts of boric acid, which is known as a water soluble thermal neutron absorber, produces an evolution of gases in significant quantity in the reactor at room temperature.

Nuclear reactions of neutron absorbers such as B-10 or Li-6 in solid materials with thermal neutrons have been widely studied, and the applications are quite broad [3]. Nevertheless, the irradiation of water samples containing neutron absorbers has not been published except for theoretical calculations using computer codes or a paper reporting experimental data obtained at high temperatures, written by B. Pastina et al [4].

To study the radiolysis of the cooling water and calculate the amount of gases that could have been produced in this manner, we irradiated several water samples containing natural, ^{10}B-enriched, and mixed boric acid in the ranges of 0 to 2000 μg/mL for the function of ^{10}B concentration.

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 for various concentrations of sample preparation. 1 or 2 mL of each boric acid solution was poured in a bullet-shaped quartz ampoule (diameter: 8 mm, height: 55 mm, and thickness: 2 mm) and placed under a vacuum to 1 × 10^{-4} torr in liquid nitrogen. The ampoules were sealed, put in Al inner capsules, and then RI target Al capsules. Several Co and Zr foils were placed together with the sealed ampoules for accurate neutron counting. The samples were irradiated during 60, 120, and 240 min relatively in a IP04 hole (Thermal neutron flux : 2.81 $\times 10^{13}$ n cm^{-2} sec^{-1} ; fast neutron flux : 7.46 $\times 10^{9}$ n cm^{-2} sec^{-1}) of the HANARO research reactor at the Korea Atomic Energy Research Institute (KAERI).

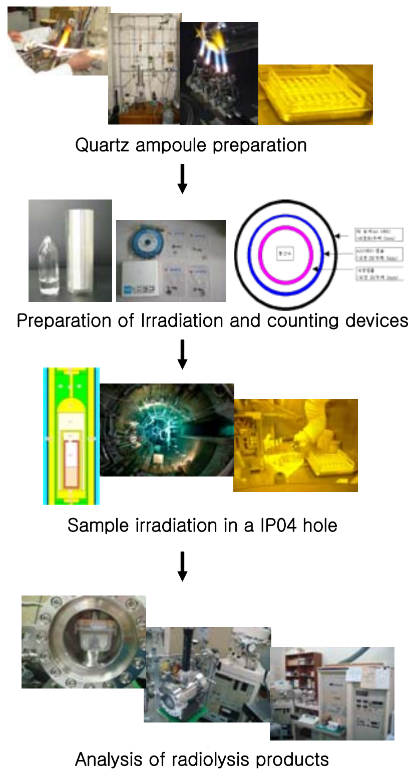

Figure 1. General flow for the whole experiments.

The radiolysis of a boric acid solution under mixed thermal and fast neutrons and gamma radiation 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. The total amounts of produced gases are discussed with a comparison of the theoretical calculation values.

3. Results and discussion

Heat generated from a boric acid solution, a container filling material (Al foil), and containers including quartz ampoule, an Al inner capsule, and an RI target Al capsule during irradiation in the IP04 hole, was calculated using a Monte Carlo N-particle Transport Code, Version 5 [5]. The estimated maximum total neutron heat and gamma heat are 16.049 and 39.609 watts (W) relatively. The temperatures of the samples and other materials during the irradiation were also computed by applying the heat values to a heat transfer code of GENGTC. The computed temperatures were all below 130 ^{o}C.

The total amount of gases produced from water radiolysis in a fast neutron flux (; the fast neutron flux is small as about 0.0265% of the total neutron flux.) of the IP04 hole was calculated based on the information that 270 mL/h of gases is generated when 1 mL of water is irradiated in a 10^3 n cm^{-2} s^{-1} neutron flux [6]. The amount of gas generated from water radiolysis in gamma radiation was also estimated based on known water decomposition G-values [7], which are radiolytic products yields in moles J^{-1}, and a gamma generating output in the IP04 hole in watts (W). These values were insignificant compared to the gas amount obtained experimentally from the radiolysis of a boric acid solution in a thermal neutron flux of an IP04 hole.

The experimental data of water decomposition to gases by irradiation of boric acid solutions in the IP04 hole are shown in Table I. 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.

Table I. The analysis results of gases produced by irradiation of boric acid solutions in a IP04 hole of HANARO research reactor*

^{10}B,(ug/mL)	Gas amount (cc.atm)			Ampoule pressure (atm)
	H_2	O_2	CO_2	
250	3.886	2.881	0.002	6.78
500	5.045	3.439	0.006	8.50
1000	9.020	5.332	0.003	14.36
2000	3.248	0.432	1.453	5.78

* Thermal neutron flux : 2.81 $\times 10^{13}$ n cm^{-2} sec^{-1} ; fast neutron flux : 7.46 $\times 10^{9}$ n cm^{-2} sec^{-1} ; irradiation time : 120 min ; solution volume : 1 mL .

However, in the case of a 2000 µg/mL ^{10}B-enriched boric acid solution, the gas amount and ampoule pressure were out of the linearity and dropped suddenly. A big pressure difference between the inside of the ampoule containing 2000 µg/mL of ^{10}B and outside of it caused a flame spark when the ampoule was broken for a gas analysis. The flame spark consumed H_2 and O_2 gases and produced CO_2 gas.

It is well known that the products of water radiolysis are molecular (H_2, O_2, H_2O_2) and radical (H, OH, e^-_{aq}, HO_2) species, and that high linear energy transfer (LET) radiation of $^{10}B(n, \alpha)^{7}Li$ produces more molecular species than radical ones. The high LET radiation of alpha rays, produced from the addition of boric acid in water, inhibits the recombination mechanisms of radicals and stops the chain reaction. Irradiation at a high temperature partially accelerates the recombination reaction of gases to return to water.

To support the data shown in Table I and know the radiolysis degree of boric acid solutions under thermal neuron irradiation, the concentrations of produced H_2O_2 and ^{7}Li and those of diminished ^{10}B will be discussed further.

4. Summary

Radiolysis gas molecular products from the decomposition of pure deionized water are not detected by thermal neutron irradiation. However, boric acid, dissolved in the water, produced detectable non-condensing molecular gases such as H_2 and O_2 under thermal neutrons.

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