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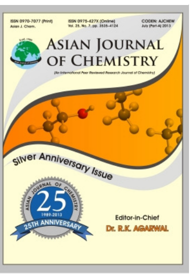

Deposition of B_4C Microparticles on Boiling Surface in Boric Acid Solution†

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The deposition of B_4C microparticles was carried out under boiling water conditions. We investigated the effect of heater power, the presence of Fe or Ca ions and the presence of pre-formed oxide layers on the deposition of B_4C microparticles. Starting from a power level inducing bulk boiling, the deposition amount of B_4C microparticles increased with an increase of heater power. However, a maximum deposition condition was observed between bulk boiling and film boiling states. The amount of deposited B_4C microparticles decreased in the presence of metal ions (Fe or Ca). The metal ions acted as flocculants to decrease the dispersive stability of B_4C microparticles rather than a source material for a porous oxide layer to increase the deposition of B_4C microparticles. It was observed that B_4C microparticles deposited much easier on bare stainless steel surfaces than on surfaces covered with an iron oxide layer under boiling water conditions.

Key Words: Deposition, B_4C microparticles, Metal ions, Oxide layer, Bulk boiling, Film boiling.

INTRODUCTION

In a nuclear power plant, the discharged fuels should be stored for a certain period in a water pool, which is used for its cooling function for the decay heat generated from high-heat-emitting radio nuclides. As many fissile materials remain in the used fuels, the fuels are carefully stacked with specific distances of separation in order to prevent the restart of nuclear fission reactions. In addition, neutron-absorbing materials are added to prevent the fuels from reaching self-propagating levels of neutrons for nuclear reaction. Boric acid is commonly used as a neutron-absorbing material because it is homogeneously dissolved in water[1-3].

However, in case of the loss of fuel cooling, the boric acid has some limitations as a neutron absorbing material. As boric acid is a weak acid with a solubility limit, it would be precipitated in the bottom of the storage pool by the evaporation of pool water. Consequently, it would easily lose its function as a neutron-absorbing material. In addition, it takes several hours for boric acid precipitate to re-dissolve in water and to reach several thousand ppm level of boron[4]. In order to complement the limitation of boric acid, it was considered to use B_4C microparticles as an additional neutron-absorbing material. Even though B_4C microparticles are a non-dissolving material, it can be dispersed in a colloidal form in aqueous solutions. It was reported[5,6] that the deposition of particles dispersed in aqueous solution could be promoted under boiling water conditions.

In this work, in order to evaluate B_4C microparticles as an additional neutron absorbing material, we investigated the effects of heater power, dissolved metal ions and pre-formed oxide layer on the deposition of B_4C microparticles. The electric power of the heater promoted water boiling and directly affected the deposition of B_4C microparticles. Metal ions and pre-formed oxide layers were observed to give negative effects on the deposition of B_4C microparticles.

EXPERIMENTAL

The B_4C powder was obtained from H.C. Starck, Germany. Most of the B_4C microparticles were less than 1 mm in diameter and were irregular in shape. The B_4C particle size distribution was a d_{50} of 0.65 mm and a d_{90} of 1.25 mm. The d_{50} and d_{90} represent the 50th percentile and the 90th percentile of the particle size distribution, respectively, as measured by volume. Distilled water and boric acid were used for the primary solution of the deposition experiments. Feric(III) nitrate and

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$Ca(NO_3)_2$ were respectively added to the primary solution in order to measure the effect of metallic ions on the deposition of B_4C microparticles. The pH of the solution was adjusted to be 9 by addition of 1 M NaOH solution.

Deposition experiments were carried out by a heating rod boiling system. The system was composed of rod-type heater, power supply, cooling unit and H-type glass tube. The schematic diagram of the system is shown in Fig. 1. Two heaters (116.2 Ω, 118.6 Ω at 25 ºC) were used for the experiment. The boiling surface area of each heater was 33 cm^2. After 2 h deposition experiments, the B_4C microparticles deposited on the boiling surface were removed by the use of ultrasonic cleaning. Filter paper (advantec, 0.1 mm pore size) was used for the separation of B_4C microparticles. The deposition amount of B_4C microparticles was determined from the net weight gain of filter paper after drying the filter paper in an oven.

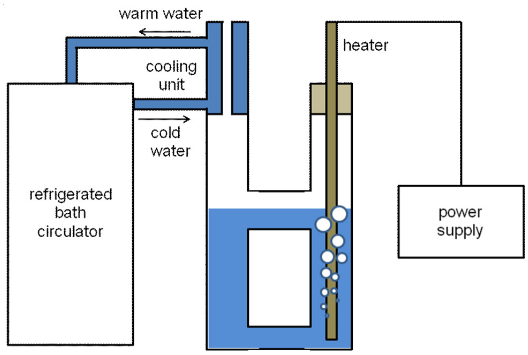

Fig. 1. Schematic diagram of experimental system for B_4C microparticle deposition

RESULTS AND DISCUSSION

Effect of heater power on the deposition of B_4C microparticles: The deposition experiments were carried out with varying heater powers in H_3BO_3 solutions. The concentration of B_4C and H_3BO_3 were 3,913 ppm B and 350 ppm B, respectively. Fig. 2 shows that the amount of deposited B_4C microparticles increased with an increase of the heater power. The amount of deposited B_4C microparticles started to increase at the onset of water boiling, which occurred at 300 W. The maximum deposition was observed at a heater power of 344 W. The deposition amount decreased slightly above the maximum deposition power of 344 W. Deposited B_4C microparticles were formed mainly around the water boiling spots.

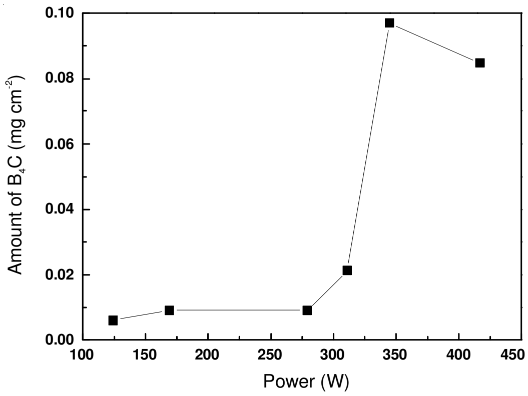

Fig. 2. Deposition amount of B_4C microparticles as a function of heater power

This result indicates that water boiling is the main factor to promote the deposition of B_4C microparticles. At low heater power (below 301.7 W), the size of boiling spots were very small and therefore only very small sizes of B_4C microparticles could be deposited on the boiling surface. Negligible weight changes of the filter paper occurred due to the limited B_4C deposition.

Above a heater power of 301.7 W, the local surface boiling turned into bulk boiling and the deposition amount of B_4C microparticles remarkably increased with an increase of heater power. The reason for a decrease in deposition above a power of 344.2 W was thought to be due to the occurrence of film boiling on the heater surface. As shown in Fig. 3, the film type boiling physically prohibits the contact of B_4C microparticles with the heater surface. In addition, a higher number of deposition spots were observed at 416.5 W compared to 344.2 W. This result also indicates that the deposition would be interrupted after the onset of film boiling.

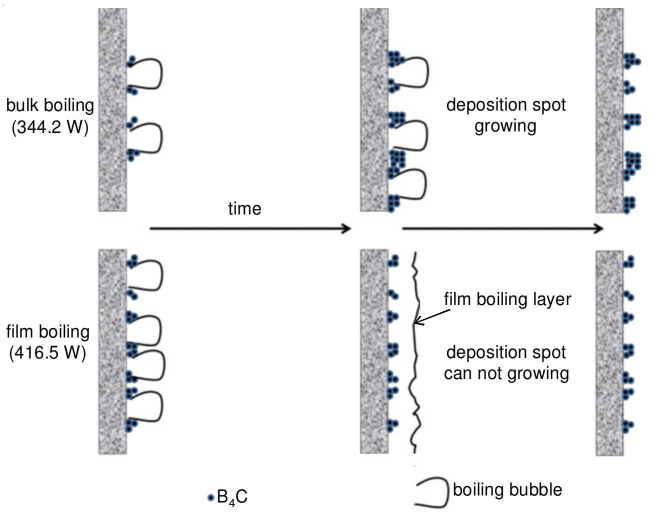

Fig. 3. Schematic diagram of B_4C deposition under bulk and film boiling conditions

Effect of metal ion on the deposition of B_4C microparticles: Table-1 shows the deposition amount of B_4C microparticles in presence of Fe and Ca ions. In all cases, Na ions were also present due to the use of NaOH to adjust the solution pH. It was observed that the deposition amount of B_4C microparticles decreased in presence of additional metal ions.

TABLE-1
DEPOSITION AMOUNT OF B_4C MICROPARTICLES IN THE PRESENCE OF METAL IONS

Metal ions	Deposition amount of B_4C microparticles ($mg\ cm^{-2}$)
Without metal ion	0.097
Fe ions	0.045
Ca ions	0.063

Metal ions have been reported to promote the deposition of particles on heat exchange surfaces through the formation of porous metal oxide layers[5]. However, following B_4C deposition experiments, we observed the agglutination of B_4C microparticles in the presence of Ca ions as shown in Fig. 4.

This result indicated that the decrease of B_4C deposition would be attributed to the weak dispersive power of microparticles in the presence of Fe and Ca ions.

Consequently, it was evaluated that metal ions such as Fe and Ca ions would act as flocculants to decrease the dispersive stability of B_4C microparticles rather than a source material of porous layers to increase the deposition of B_4C microparticles.

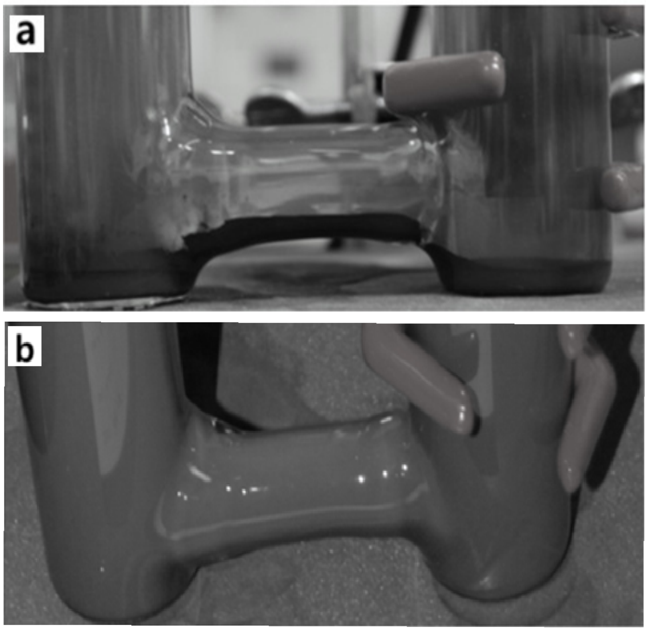

Fig. 4. Photos of B_4C microparticles dispersed in two different boric acid solutions. a: with 0.25 mM Ca^{2+}, b: without Ca^{2+}

Effect of iron oxide layer on the deposition of B_4C microparticles: In a reactor water pool system, various metal ions and colloidal particles are present from the release of structural materials such as stainless steel and Ni-based alloys. From the differing metal oxide solubilities and the presence of boiling water, various metal oxide layers would form on the surface of spent nuclear fuel. Therefore, we investigated the role of a pre-formed iron oxide layer on the deposition of B_4C microparticles. A yellowish iron oxide layer was formed on the stainless steel heater rods under boiling condition for 2 h prior to B_4C particle deposition.

In Table-2, the deposition amount of B_4C microparticles with and without the presence of an iron oxide layer is denoted. The deposition amount of B_4C microparticles with the presence of an iron oxide layer decreased by 65 % compared to that without an iron oxide layer. The result indicates that the B_4C microparticles could adsorb much easier on the stainless steel surface than on the iron oxide layer under water boiling conditions.

TABLE-2
DEPOSITION AMOUNT OF B_4C MICROPARTICLES ON PRE-FORMED IRON OXIDE LAYER

Surface	Deposition amount of B_4C microparticles (mg cm^{-2})
On stainless steel bare surface	0.042
On pre-formed iron oxide layer	0.015

Conclusion

The amount of deposited B_4C microparticles increased with an increase of heater power. From the onset of bulk boiling, the deposition amount remarkably increased. A maximum deposition heater power was observed between bulk boiling and film boiling states. The amount of deposited B_4C microparticles decreased in the presence of metal ions such as Fe and Ca. The metal ions acted as flocculants to decrease the dispersive stability of B_4C microparticles rather than a source material for porous oxide layers to increase the deposition of B_4C microparticles. B_4C microparticles deposited much easier on the stainless steel surface than on the surface of stainless steel covered with a pre-formed iron oxide layer.

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