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Initial relocation behavior of control rod materials in boiling water reactors studied via time-resolved visualization

Shota Ueda[a,*], Byeongnam Jo[b], Masahiro Kondo[c], Nejdet Erkan[a], Takeshi Yajima[d], Koji Okamoto[a]

[a] Department of Nuclear Engineering and Management, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, Japan
[b] Department of Mechanical Engineering, Ajou University, Worldcup-ro 206, Suwon, Republic of Korea
[c] Nuclear Professional School, The University of Tokyo, 2-22 Shirakata, Tokai-mura, Ibaraki, Japan
[d] Institute for Solid State Physics, The University of Tokyo, 5-1-5 Kashiwa, Chiba, Japan

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ABSTRACT

For the Fukushima decommissioning, the distribution of boron species in the fuel debris must be determined to assess the risk of recriticality, the debris hardness and thus complicate its successful retrieval. As a result, the relocation behavior of boron carbide (B_4C) control rod materials has attracted significant attention. In this work, the influence of the thickness of its stainless steel (SS) clad on the initial relocation behavior of the control rod was investigated. In particular, the initial relocation behavior of the B_4C control rod materials was dynamically visualized using a technique previously developed by the authors. To simulate the control rod, a type-304 SS tube filled with B_4C powder containing particles with sizes of approximately 20–30 µm was heated to a temperature exceeding its eutectic point (1473 K). The three relocation modes of the obtained eutectic melt corresponded to film formation, droplet formation without collapse and droplet formation with collapse.

1. Introduction

It is considered that the early liquefaction and relocation of boron carbide (B_4C) control rods in boiling water reactors (BWRs) occur in severe accidents. Hence, it is important to investigate the relocation behavior of these rods as part of the establishment of a better severe accident management because of the effect produced on the subsequent accident progress. The obtained results can be also used for examining the boron distribution in fuel debris, which is of great importance to the Fukushima decommissioning because the presence of boron in the relocated core debris affects the possibility of re-criticality (Ikeuchi et al., 2017) and significantly increases its hardness (Takano et al., 2014; Nagase et al., 2017), thus making the process of debris retrieval very complicated. Therefore, a detailed elucidation of the relocation behavior of B_4C control rods must be performed.

A typical B_4C control rod is fabricated from a stainless steel (SS) clad and B_4C filling. During its relocation, various chemical reactions, such as the SS-B_4C eutectic melting (Hofmann et al., 1990) and oxidation of B_4C (Sepold et al., 2006; Repetto et al., 2010) occur. In particular, it is known that the B_4C-SS eutectic melting affects its relocation, further contributes to the degradation of the surrounding fuel rod claddings and modifies the composition and characteristics of the final melt that include fuel materials through integral effect tests; the DF-4 (Sandia National Laboratories) (Gauntt et al., 1989; Gauntt and Gasser, 1990) and Phebus FPT-3 (IRSN) (Clément et al., 2003; Girault et al., 2009; Haste et al., 2012) experiments. Thus, the B_4C-SS eutectic melting process must be examined in detail.

Fundamental studies on the progress of the eutectic reaction in a small specimen (so called a separate effect test) have been performed in the past (Hofmann et al., 1990; Nagase et al., 1997; Steinbrück, 2014; Shibata et al. 2015; Furuya and Morooka, 2015). Thus, Hofmann et al. (1990) found that the Arrhenius kinetics can be used in the mathematical description of the growth of the eutectic reaction layer, and Nagase et al. (1997) confirmed its applicability to a wider temperature range. In addition, the sudden increase in the reaction rate on the corresponding Arrhenius plots before and after the eutectic temperature was observed. Furuya and Morooka (2015) investigated the effect of oxidation during eutectic melting and found that B_4C oxidation affects the rheological properties of the melt, which affects convection of the melt, and composition profile of compounds. The ability to describe the growth of the eutectic reaction layer by constructing Arrhenius plots indicates that eutectic melting is a time-dependent phenomenon and that the behavior of the produced eutectic melt and its relocation properties depend on the thickness of the SS clad (the latter was confirmed experimentally in the previous studies conducted using a plate-shaped specimen (Ueda et al., 2016)). However, the influence of the SS

* Corresponding author.
E-mail addresses: ueda@vis.t.u-tokyo.ac.jp (S. Ueda), jo798@ajou.ac.kr (B. Jo), kondo@vis.t.u-tokyo.ac.jp (M. Kondo), erkan@n.t.u-tokyo.ac.jp (N. Erkan), yajima@issp.u-tokyo.ac.jp (T. Yajima), okamoto@n.t.u-tokyo.ac.jp (K. Okamoto).

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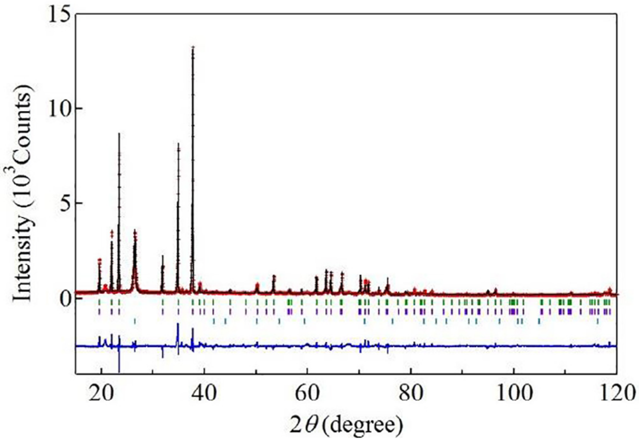

Fig. 1. XRD profiles of the B_4C powder used in this study.

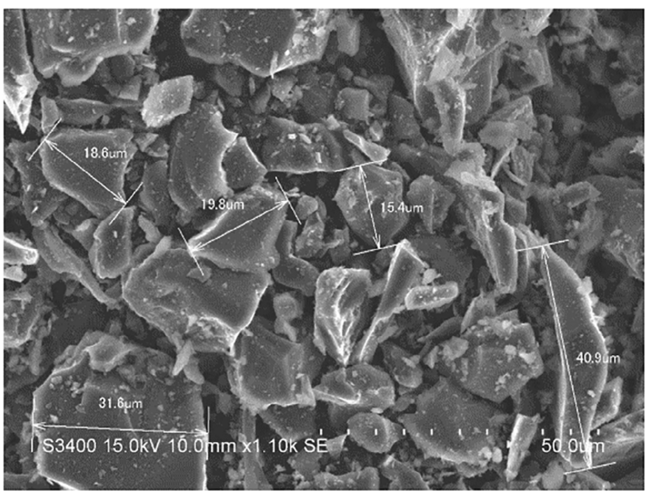

Fig. 2. An SEM image of the B_4C powder used in this study.

clad thickness on the relocation behavior of the control rod geometry has not been examined yet.

In the current study, to investigate the initiation of the control rod degradation due to eutectic melting, the dynamic visualization of the initial relocation behavior of the B_4C control rod materials has been performed by varying the thickness of the SS clad (the utilized approach has been developed in a previous study (Ueda et al. 2016)). Unlike other methods (such as a metallurgical approach), this technique allows systematic monitoring of the eutectic relocation reaction in a highly time-resolved manner. To simulate a control rod, a type-304 SS tube filled with B_4C powder (containing particles with sizes of approximately 20–30 µm) was used. The specimen was heated to a temperature exceeding 1473 K because Hofmann et al. (1990) reported that the eutectic reaction began at approximately 1473 K. The experiment was conducted in an argon atmosphere to eliminate the influence of B_2O_3 species (produced during the B_4C oxidation) on the relocation behavior of the studied rod. The specimen was analyzed via X-ray diffraction (XRD) after the experiment to confirm the absence of significant amounts of oxygen contaminants in the eutectic melt.

2. Experimental

2.1. Materials

In this study, type 304 SS (Nilaco Co.) and B_4C powder (Atom Shield Co., Ltd.) were used. The Rietveld analysis on XRD (Rigaku, SmartLab) data of the B_4C powder revealed the powder contains 5.8(4)% of graphite as an impurity. Fig. 1 is the refined XRD profile of the B_4C powder, showing observed (red cross), calculated (black line), and difference (blue line). The upper and middle ticks represent the positions of the Bragg reflections of the B_4C and the bottom ticks represent those of graphite. The SEM image of the utilized B_4C powder (see Fig. 2) suggests that its average particle size is equal to approximately 20–30 µm.

2.2. Experimental setup

The resulting melt flow was visualized in a time-resolved manner to characterize the initial relocation behavior of the control rod at super high temperatures using the techniques developed in our previous studies (Ueda et al., 2016, 2017). The schematic of the utilized experimental setup is shown in Fig. 3.

The specimen was placed between two tungsten heaters (connected to a power supply through copper electrodes) and then heated by thermal radiation. A traction system was attached to the bottom

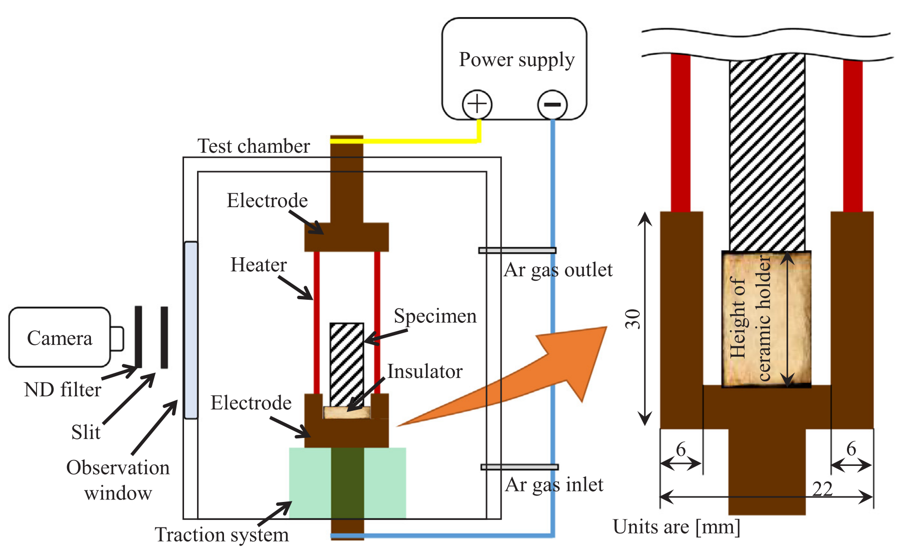

Fig. 3. A schematic of the experimental apparatus used in this study.