Abstract
The liquefaction and relocation of control rods at temperatures over 1200 °C due to the eutectic reaction between boron carbide and stainless steel are important phenomena for the clarification of the Fukushima Daiichi Nuclear Power Plant accident. However, the mechanism and behavior of these phenomena have not yet been understood completely. The time-resolved visualization of the dynamic and unsteady characteristics of the eutectic reaction might provide important insights into the various phenomena occurring in a time sequence. Therefore, this study employed a time-resolved visualization technique under high-temperature conditions to analyze the eutectic reaction between boron carbide and stainless steel. Among the many heating methods available, radiative heating was chosen because it facilitates clear visualization. The main problems that made this visualization difficult were the large thermal expansion, reflected light from heaters, and change in the specimen geometry due to the reaction. These problems were solved by employing novel and simple techniques. In this study, experiments were performed at different temperatures using different specimens to confirm the effectiveness of the developed technique. The maximum temperature used in the experiments was 1250 °C. Clear images were recorded. It was observed that the change in the specimen geometry affected the melt behavior. The developed technique was effective in acquiring geometrical information on dynamic phenomena at high temperatures. The technique may provide a potential data set for the validation of numerical simulations.

Key words: Flow visualization, High-temperature measurement, Boiling water reactors, Control rod, Eutectic reaction, Severe accident

1. Introduction
Eutectic reactions at high temperatures are important phenomena for nuclear buildings that are built of several materials because the use of different materials substantially decreases the melting temperature of the original base material. High-temperature eutectic reactions have considerable influence on a structure, especially in cases of severe accidents in nuclear power plants (NPPs), where the temperature on the surface of fuel rods in the core exceeds 1200 °C. The latest severe accident experienced in an NPP was the Fukushima Daiichi accident in 2011. A boiling water reactor (BWR) was used in this NPP. In this accident, it is estimated that the early liquefaction of control rods due to the eutectic reaction between boron carbide (B₄C) and stainless steel (SS), which are widely used in many BWRs, occurred because their eutectic point is c.a. 1200 °C (Zheng et al., 2014). This melting temperature attributable to the eutectic reaction is relatively low compared to the melting temperatures of the other materials used in the core (Hering et al., 1994, Sepold et al., 2006). This control rod failure at the beginning of the accident can determine the subsequent nature of accident progression. Therefore, it is important to investigate the eutectic reaction between B₄C
and SS and their melt behavior through visualization.

The visualization of the melt behavior can be achieved mainly by two methods. One is a time-resolved visualization method in which time-resolved images are recorded using a video camera. The other is a static method for diagnosing samples. Previous studies have used static methods to investigate the eutectic reaction between B₄C and SS, with special focus on their chemical interaction, and the melt initiation accompanying the reaction. These studies visualized the molten material by observing samples after annealing tests (Hofmann et al., 1990, Nagase et al., 1997, Shibata et al., 2015, Sasaki et al., 2015). However, only few studies have performed experiments involving dynamic visualization even though the progress of melt growth and the melt behavior are unsteady and dynamic. The lack of research in this area is mainly due to two reasons. The very high eutectic temperature of the materials and the coexistence of both solids and liquids during the phenomena make dynamic visualization difficult. For example, the thermal expansion and deformation of a specimen and apparatus cause their mechanical failure, and the strong light emitted by heaters disturbs clear visualization.

For other applications, time-resolved visualization methods for the melt behavior have been developed, such as a radioscopic method (Koster et al., 1996), dynamic neutron computer tomography (Kureta et al., 2009), and ultrasonic visualization (Takeda, 1991, Tasaka et al., 2006). However, these methods have not been developed for the case where both solids and liquids coexist during the progress of the phenomena and the interaction between them is important. In addition, these methods do not consider high temperatures. Methods for the in-situ visualization of phase transformation at high temperatures have also been developed, such as high-temperature laser microscopy (Shibata et al., 1997, Komizo et al., 2006) and visualization using a high-speed video camera (Shinozaki et al., 2008). These studies utilize condensation heating or laser heating and a tiny specimen in the experiments. However, these methods cannot be used to observe the entire process from melt growth to melt flow in a time sequence. The specimen scale is also limited because of the features of the heating method (⌀4.3 mm × 2 mm). However, the visualization of the solid–liquid interaction at high temperatures is required. Thus, a novel and simple visualization technique for both the melt growth and melt behavior in a time sequence at high temperatures is required to realize the dynamic visualization of the eutectic reaction. Furthermore, a visualization experiment with well-defined boundary conditions can provide important insights into the mechanism and validation data for the numerical calculation of the phenomena.

In this study, a dynamic visualization technique is developed for the high-temperature eutectic reaction and the obtained results are presented in the form of a visualization experiment in a temperature range of up to 1250 °C; we measure the temperature using thermocouples. B₄C powder and SS are used as the reaction materials in the demonstration experiments. The advantages and disadvantages of our technique and its effectiveness are discussed with respect to the temperature measurement, boundary conditions for numerical simulations, and acquirable physical parameters.

2. Visualization technique and experiments
2.1 Heating method

Before developing the visualization technique for the high-temperature eutectic reaction, the requirements of the heating method are determined: the heating method should be able to heat a specimen to temperatures up to 1200 °C, non-contact heating that does not disturb the melt movement should be employed, no additional chemicals should be added (i.e., thermite reactions are not suitable), the method should be able to heat a sufficiently large specimen so that the melt flows, the input heat provided to a specimen should be independent of the change in the specimen geometry due to the reaction, and the heating method should be compatible with the visualization technique. If the input heat provided to a specimen is dependent on the specimen geometry (i.e., if direct heating is employed by allowing current to flow through a specimen), uncontrollable local heating may occur, wherein the physical parameters might be different from those in the case of real control rod failure. One of the most important aspects to be considered is how a test piece can be heated uniformly up to around 1200 °C while visualizing the phenomena. Among the many heating methods available, radiative heating using tungsten heaters is chosen. Radiative heating using heaters facilitates clear visualization because it can realize heating without the specimen being surrounded completely as is required in the case of induction heating and it provides a fixed boundary condition that is suitable for both the post-analysis of the experiments and numerical calculation of the phenomena because the heat flux from the heaters is constant throughout the experiment. Although this method is suitable for the objective of our research from many aspects, this method has a
disadvantage with respect to achieving clear visualization. Heaters usually emit very strong light at high temperatures. Consequently, the light might be reflected by the specimen and the inner structure of the test facility and might enter the visualization camera. To overcome this disadvantage, the specimen was designed carefully and some optical tools were employed to reduce the light.

2.2 Test facility and optical apparatus

The developed system is shown in Fig. 1. Tungsten heaters were installed beside the specimen because if the tungsten heaters were installed between the camera and specimen, it would have prevented clear visualization. In general, tungsten heaters expand considerably as the temperature increases. In our preliminary experiments, the thermal expansion of the heaters was not negligible because their estimated temperature was over 2500 °C whereas the desired temperature of a specimen was over 1200 °C. When the heaters were simply connected to the holder in the preliminary experiment, they expanded continuously, deformed exceedingly, and finally came in contact with the specimen, causing an electric short. To solve this problem, a traction device was used to release the stress and avoid deformation. This device used a weight and bearing to pull the heaters down. A heat-resistant material was used for this device because it was directly connected to the high-temperature heaters. In addition, thermal expansion will cause problems if two materials with different thermal expansion coefficients are joined together. For example, if the heater is joined to a specimen, mechanical failure will result between them because of the huge difference in their thermal expansion coefficients. A specimen should be physically isolated as much as possible. Thus, the specimen was hung from only one small point in midair to avoid mechanical failure. Copper electrodes were connected to the power supply using a cable. Two ceramic plates were installed between a copper electrode and specimen to ensure both electrical and heat insulation. The walls of the test facility were made of SS. A window was created in the front wall of the test facility for visualization using a high-resolution camera (FASTCAM SA-X, Photron co.). To prevent the reflection of direct light from the tungsten heaters, a slit was created on the observation window. A neutral density filter was set on the camera to reduce the surplus light uniformly. The gas inlet and outlet were located at the back of the test facility.

![Fig. 1 Schematic image of test facility](image)

2.3 Specimens and test conditions

Fig. 2 shows the two types of specimen designs used in the experiments. Both specimen types are used for the visualization of the interfacial reaction growth between the B₄C powder and SS part and the melt behavior. An SS plate (0.1 mm in thickness) was attached using cement to the back of both the specimen types. When cement was not used in the preliminary experiment, a reaction occurred at the middle of the specimen and the specimen separated from the plate at the molten portion. This reaction led to a sudden change in the specimen geometry, and the evaluation of the phenomena became difficult. B₄C powder was filled inside the SS plates. The void fraction for the B₄C powder was approximately 52% in each case, and the void was filled with air. To ensure clear visualization, the specimen shape is also important considering that radiative heating using tungsten heaters, which emit strong light, was used in the experiment. For example, a columnar specimen did not reflect light uniformly and a very-high-contrast image was obtained. Whiteouts and blackouts of major parts in the picture occurred, which prevented the accurate estimation of
the phenomena. From this viewpoint, a rectangular parallelepiped specimen is the best for the visualization experiment using radiative heating.

![Diagram](image)

(a) Front-uncovered specimen  (b) Front-covered specimen  (c) Front view of both specimen types

Fig. 2 Designs of two specimen types and positions of thermocouples (All units are mm)

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*: maximum temperature, **: constant temperature

Two experimental cases were considered to visualize the reaction growth and melt behavior and to confirm the effectiveness of the abovementioned visualization technique. Table 1 lists the experimental conditions. In the front-uncovered case, a 65-mm-high specimen was placed upside down. To confirm the visualization quality, the specimen was observed for a certain temperature gradient along the vertical direction. After the temperature reached 1220 °C, it was increased continuously at the rate of 0.08 °C/s. In the front-covered case, the temperature was set at 1220 °C. The thickness of the front cover was 0.1 mm. The temperatures were measured using thermocouples (K-type JIS standard thermocouples), which were inserted into the SS part of the specimen from the back in both the cases. In the front-covered case only, the temperatures on the front cover were also measured using the same two thermocouples. All the thermocouples were located 1.0 mm from each side of the specimen.

3 Results and discussion
3.1 Experimental results and discussion

Two types of experiments were performed. First, the front-uncovered case was considered, the results of which are shown in Fig. 3. Fig. 4 shows the temperature history during the experiment. Clear images of phenomena such as the interfacial reaction growth and melt behavior were obtained. For example, the small invasion of the formed melt into the B₄C powder region and the melt retention inside the SS part were clearly visualized (Fig. 3 (1) Small invasion). At around 1090 s, the B₄C powder in the middle part absorbed much of the melt and the melt dissolved the B₄C (Fig. 3 (2) Absorption of melt). Thus, B₄C powder was no longer present in the middle part. Because the test facility was filled with argon gas, only limited oxidation occurred at the bottom of the specimen though the oxidation potential of SS is high. As the temperature increased, the melt eventually liquefied completely. After the complete liquefaction and movement of the melt, it acquired a spherical shape because of strong surface tension. Thus, the melt reflected a
considerable amount of light, which resulted in images with relatively high contrast (690–1225 s, Fig. 3) Considerable light reflection). However, in spite of this relatively high contrast, the melt behavior can be understood from the images. At 890–1225 s, a downward melt movement was observed (Fig. 3 (4) Melt movement). The completely liquefied melt at the top and right side of the specimen stayed at its original position because of its high surface tension at 890 s. As the temperature increased, the surface tension of the melt reduced and it gradually rolled down along the specimen.

Fig. 3 Time-resolved images in front-uncovered case

Second, the front-uncovered case was considered, the results of which are shown in Fig. 5. The temperature was set at 1200 °C on average. Fig. 6 shows the temperature history during the experiment. The thermocouple installed on the right on the front cover (Surface/Right) got detached from the specimen and showed unreliable results. Fig. 5 shows the obtained images in a time sequence. Just after the temperature reached 1200 °C, eutectic material started to form at 600 s (Fig. 5 (1) Formation of eutectic material). In this case, no absorption of the melt by the B$_4$C powder was observed; rather, the melt was repelled by the powder. The repelled melt moved to the solid SS region because of the interfacial force between the melt and solids SS, and the inner B$_4$C region was exposure to argon gas (Fig. 5 (2) Melt movement and (3) Exposure of B$_4$C). As the melt moved downward, it became larger. The gathered melt formed a drop at the
bottom (Fig. 5 (4) Melt drop).

![Fig. 5 Time-resolved images in front-covered case (600–1100 s)](image1)

**Fig. 5** Time-resolved images in front-covered case (600–1100 s)

**Fig. 6** Temperature history of specimen in front-covered case

Fig. 7 shows the effect of different SS-plate thicknesses on melt behavior. The left and right panels show the cases with thick and thin SS plates, respectively. In the case with a thick SS plate, the melt tended to stay at the original position because all the surfaces of the melt were surrounded by solid SS, and hence, they resisted gravity. Eventually, the melt was absorbed by the B₄C powder because of capillary force. However, when the SS plate was relatively thin, the melt moved around easily just after its formation because the area of the melt that was in contact with solid SS was very small, which gave the melt much more free surface. Thus, in this case, the melt was rarely absorbed by the B₄C powder. In other words, this difference between the two cases can be attributed to the fact that the balance between the melt volume and the contacting area of the melt with the specimen plays an important role in this absorption phenomenon.
3.2. Discussion of effectiveness of method

In this section, the advantages and disadvantages of the abovementioned visualization method are discussed. As shown in Fig. 3 and Fig. 5, phenomena such as the melt formation, melt movement, and melt absorption by the B₄C powder were clearly visualized, and detailed and qualitative information was obtained in a time sequence. That is, the proposed dynamic visualization method provides this time-resolved information. This is one of the important advantages of this method. In this paper, the details of two experimental cases are presented: front-covered and front-uncovered cases. We obtain the boundary conditions by measuring the temperatures on the specimen using six thermocouples; hence, the data can be used for the validation of numerical calculations. Therefore, another advantage of this method is that it can create validation data with detailed time-resolved information of phenomena and boundary conditions such as the specimen temperature. The developed heating method and midair specimen holding using ceramic plates solve the problems caused by thermal expansion. These techniques do not interfere with the fluid flow of the melt. Therefore, these techniques are effective for the visualization of a system that consists of both solids and liquids and where the interaction between them is important. The physical parameters governing the melt behavior, such as the melt head velocity and timing of melt formation, can be measured. The combination of two-way radiative heating, appropriate design of the specimen and test facility, and midair specimen holding facilitates the visualization at high temperatures. However, the proposed method has a disadvantage, which is attributable to the heating method employed. The proposed method employs radiative heating using tungsten heaters, which emit strong light. In principle, an infrared camera is not suitable for temperature measurement by this method because the spectra of infrared light coming from the specimen (especially, the SS part) and tungsten heaters overlap each other. Although a parallelepiped specimen is designed, which reduces the amount of light from the tungsten heaters, even a small amount of light reflected by the specimen produces uncertainty in the temperature measured using an infrared camera. This implies that temperature distribution measurement along a specimen is difficult with this method.

Although the proposed method has its advantages and disadvantages, it can be said that this time-resolved visualization method for the eutectic reaction between B₄C and SS at high temperatures is an effective way to investigate the liquefaction and relocation of control rods at temperatures over 1200 °C.

4. Conclusion

In this study, a technique for the time-resolved visualization of the eutectic reaction between B₄C and SS at high temperatures was developed. The technique provided a clear view of the dynamic mechanisms in a time-resolved manner. Some problems were encountered during the visualization at high temperatures, such as thermal expansion, the reflection of light and compatibility between visualization and heating; these problems were solved, and clear image data were obtained. Two types of experiments were performed at different temperatures for different specimens to confirm the effectiveness of the developed technique at temperatures up to 1250 °C. The progress of the melt growth
and the melt behavior could be observed clearly, including melt absorption by the B$_4$C powder on the SS–B$_4$C interface and melt repulsion by the B$_4$C powder. It was confirmed that the developed method was effective in investigating dynamic and unsteady phenomena where both solids and liquids coexisted and the eutectic temperature was very high. The limitation of this method would be that temperature distribution measurement along a specimen using an infrared camera is difficult with this method. The proposed technique might provide a new perspective to the investigation of the mechanism of control rod failures in severe nuclear accidents.

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References