# Determination of the Second Critical Endpoint in Silicate-$H_2O$ Systems

$H_2O$ plays an important role in magmatism and chemical evolution in Earth's interior. Two types of hydrous mobile phase, aqueous fluid and hydrous silicate melt, could exist in Earth's crust and mantle. In general, both the solubility of water in silicate melt and the solubility of silicate materials in aqueous fluid increase with increasing pressure. This could suggest that, above a certain critical pressure and temperature, silicate melt and aqueous fluid become indistinguishable from each other [1]. This critical condition is called the second critical endpoint.

Previous studies for direct observations of coexisting aqueous fluid and silicate melt, which have relatively low melting temperature [2], were conducted using a hydrothermal diamond anvil cell (HDAC). However, it is difficult to obtain the temperature optimal for observing fluid and melt with high melting temperature, such as basalt and mantle peridotite, using the HDAC. Our main purpose is to develop a new method for the determination of the second critical endpoint in basalt-$H_2O$ and peridotite-$H_2O$ systems at high pressure and temperature.

Experiments were conducted using an X-ray radiography technique [3] together with a Kawai-type multi-anvil high pressure apparatus (SPEED-1500) installed at beamline **BL04B1** [4]. The direct X-ray beam, which passes through the anvil gaps of SPEED-1500 and the sample under high pressure, is observed with an X-ray camera (Fig. 1). A hydrous strontium-plagioclase (mixtures of oxides and hydroxides) system is used as a starting material to obtain a better contrast in radiographic images. In addition to water in hydroxides, deionized water is added to the sample. The sample container should not react with hydrous samples, but should be X-ray transparent. We, therefore, developed a new sample container [5], which is composed of a metal (Pt or AuPd) tube and a pair of single crystal diamond lids placed on both ends of the metal tube (Fig. 1). The sample, which is placed inside the container, can directly be observed through the diamond lids and the aperture of the metal tube.

The experimental conditions are pressures from 3 to 4.3 GPa and temperatures up to approximately 1500°C. Pressure is applied first, and then temperature is increased. After the observation with the X-ray camera, the samples were quenched at the desired P-T conditions so that the X-ray radiographic images might be compared with the quenched textures.

At around 900 to 1000°C and 3 GPa, some light-gray spherical bubbles (100-500 microns diameter) moving upward in the dark-gray matrix were observed. The light-gray spherules that absorbed less X-ray are considered to be the aqueous fluid phase, whereas the dark-gray matrix is a silicate melt. With a further increase in temperature, small bubbles were coalescing to form large bubbles. Then, these large bubbles became hemispheres wetted on the upper wall of the metal tube (Fig. 2). In experiments up to 4 GPa, two phases (i.e., aqueous fluid and silicate melt) were observed, whereas at 4.3 GPa, no bubbles were observed during heating up to about 1500°C.

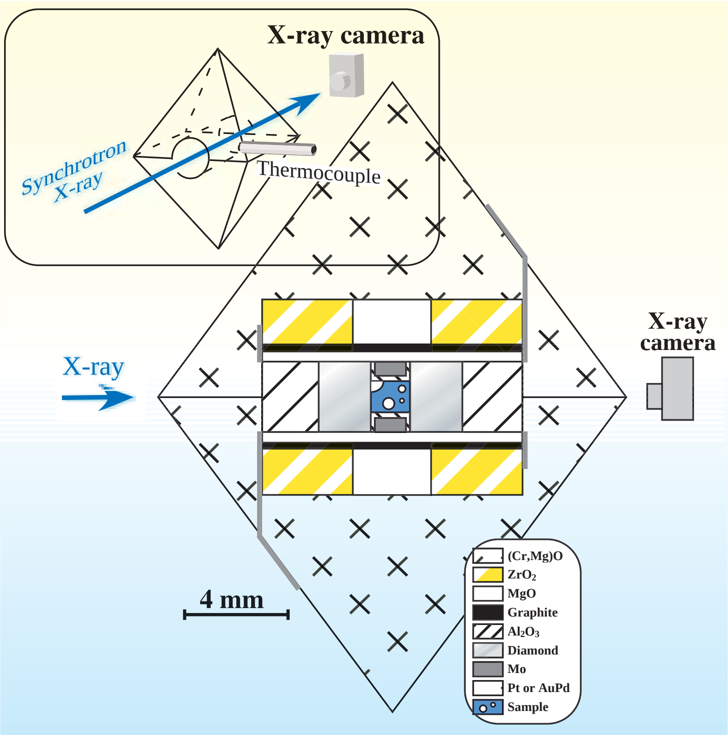

Fig. 1. Experimental configurations and the furnace assembly used in high-pressure and high-temperature experiments. Images are recorded with a digital video camera.

The image of the recovered sample quenched at 3 GPa and 1300°C is shown in Fig. 3. A large and hemispherical bubble, which is considered to be the aqueous fluid phase at high pressure and temperature, can be seen in the glass matrix, which on the other hand is considered to be the silicate melt phase during the run. This image (Fig. 3) is consistent with the X-ray radiographic image (Fig. 2) that showed two phases at this pressure and temperature. In the recovered sample quenched at 4.3 GPa and 1300°C, no bubbles were found. Instead, quenched crystals were homogeneously distributed in the entire sample capsule.

Both radiographic observations and inspection of quenched samples indicate that aqueous fluid and melt can coexist at pressures up to 4 GPa and there is no difference between these two phases above 4.3 GPa in a Sr-plagioclase-$H_2O$ system. Therefore, it is concluded that the second critical endpoint in the Sr-plagioclase-$H_2O$ system exists at pressure between 4 and 4.3 GPa. Our new technique could be applied to direct observations of various kinds of 2-fluid coexisting under deep mantle conditions that could not be achieved by the HDAC. Detailed studies for the determination of the second critical endpoint in the basalt-$H_2O$ and peridotite-$H_2O$ systems are now being undertaken for the better understanding of the properties of fluid and melt in Earth's deep interior.

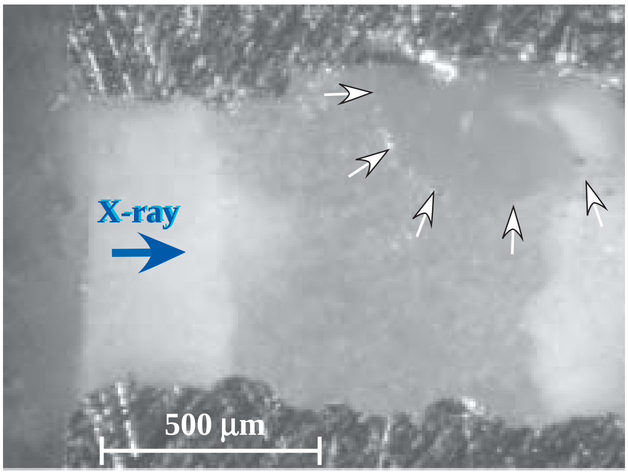

Fig. 3. Image of the recovered sample quenched at 3 GPa and 1300 °C.

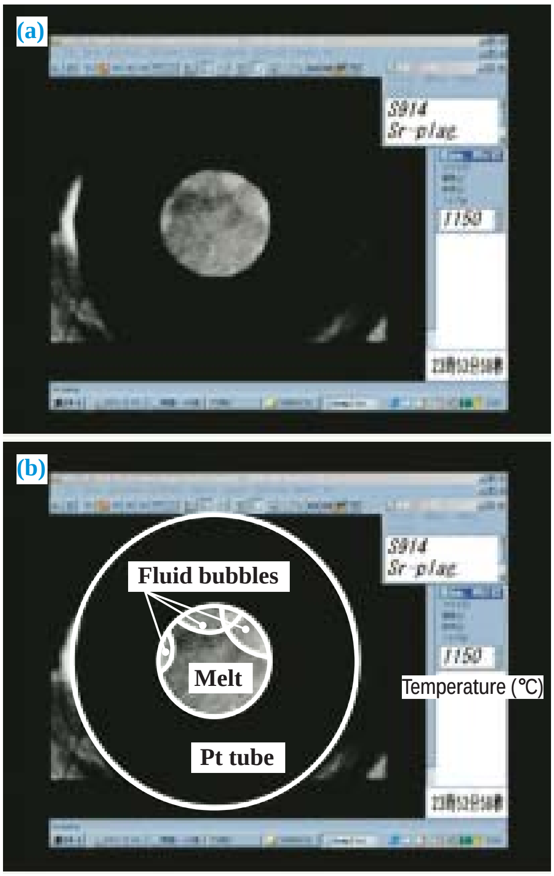

Fig. 2. X-ray radiographic images taken at 3 GPa and 1150 °C. **(a)** Image. **(b)** Explanatory notes along with **(a)**.

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