

# Search for the second critical endpoint in basalt-H<sub>2</sub>O and peridotite-H<sub>2</sub>O systems

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H<sub>2</sub>O plays an important role in the physical and chemical evolution in the Earth's interior. Two types of hydrous mobile phase, aqueous fluid and hydrous silicate melt, can exist in the Earth's crust and mantle. In general, both the solubility of water in silicate melts and the solubility of silicate materials in aqueous fluid increase with increasing pressure. This could suggest that, above a certain critical pressure and temperature (the second critical endpoint), silicate melts and aqueous fluid become indistinguishable with each other.

Using previous experimental techniques, such as hydrothermal diamond anvil cell, it is difficult to obtain enough temperature to observe fluid and melts with high melting temperature composition. Therefore, we developed a new method for the determination of the second critical endpoint in basalt-H<sub>2</sub>O and peridotite-H<sub>2</sub>O systems at high pressure and temperature.

Experiments were conducted using X-ray radiography technique together with Kawai-type double-stage multi-anvil high pressure apparatus (SPEED-1500) installed at SPring-8, Japan. Direct X-ray beam, which passes through the anvil gaps of SPEED-1500 and sample under high pressure, is observed with an X-ray camera. The systems basalt-H<sub>2</sub>O and peridotite-H<sub>2</sub>O (mixtures of oxides and hydroxides) were used as the starting materials. In addition to the 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, which is composed of a metal (AuPd) tube and a pair of single crystal diamond lids put on both ends of metal tube. The sample in the metal container can directly be observed through the diamond lids with X-ray radiography.

The experimental conditions are at pressures from 1.7 to 4.0 GPa and at temperatures up to about 1400 deg. C. Pressure is applied first, and then temperature is increased. After the observation with X-ray camera, the samples were quenched at the desired P-T conditions so that the X-ray radiographic images could be compared with the quenched textures.

At around 1000 deg. C and 1.7 GPa in the system peridotite-H<sub>2</sub>O, a dark gray sphere appeared in the light gray matrix. The light gray matrix that absorbed less X-ray is considered to be aqueous fluid phase, whereas the dark gray sphere is silicate melt. With further increasing temperature, the drastic overturn was observed. In the experiments up to 3.6 GPa, two phases (aqueous fluid and silicate melt) were observed. At 4.0 GPa, we could not distinguish two phases in the radiographic images.

A large void space, which is considered to be an aqueous fluid phase at high pressure and temperature condition, can be seen in the recovered sample up to 3.6 GPa in the system peridotite-H<sub>2</sub>O. On the other hand, in the quenched sample at 4.0 GPa, no void space was found. Instead, quench crystals were homogeneously distributed in the whole sample capsule.

Both radiographic observations and inspection of quenched samples indicate that aqueous fluid and silicate melt can coexist up to 3.6 GPa and there is no difference between these two phases above 4.0 GPa in the system peridotite-H<sub>2</sub>O. It could be concluded that the second critical endpoint in the system peridotite-H<sub>2</sub>O occurs at pressure between 3.6 and 4.0 GPa. In the same way, our experiments in the system basalt-H<sub>2</sub>O revealed that the second critical endpoint could occur at pressure between 3.0 and 3.6 GPa.

At pressures below the second critical endpoint, chemical differentiation could be occurred by the fluid-liquid immiscibility. On the other hand, no such differentiation could happen above the second critical endpoint because the only supercritical fluid is stable. Our experimental results suggest important implications for the magmatism under hydrous conditions, such as the subduction zone magmatism.