The effect of high temperature on carbon solubility in cristobalite

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Silicates is one of the major and important components in the Earth's crust and mantle. Because an enormous amount of carbon is stored in the Earth's interior, it can be suggested that the silicates must be exposed to carbon under high-pressure and high temperature conditions (Sen et al. 2013). Recently, Santoro et al. (2014) demonstrated that carbon atoms can be substituted with silicon in cristobalite at P = 16-22 GPa and T > 4000K. Furthermore, the first-principles calculation showed the possibility that a continuous solid-solution can be formed between SiO₂ and CO₂ in a metastable cristobalite phase at ambient pressure (Aravindh et al. 2007). However, the effect of temperature on carbon solubility in silica has not been confirmed yet. Accurate knowledge of the carbon solubility in silica under high temperature can help us to understand the interaction between carbon and silicate minerals in the Earth's interior. In this study, we investigated the carbon solubility and structure change of cristobalite under high temperature condition. First, we prepared amorphous silica and graphite as starting materials. Powder amorphous silica and graphite were mixed sufficiently by an agate mortar and pestle. Second, the mixture was vacuum-enclosed in a quartz tube and heated at 1300°C for 3 days. Then, it was removed from the furnace and quenched at room temperature. The sample was carefully examined by powder XRD, EPMA, and IR spectroscopic analysis.

Regardless of the Si : C ratios of starting materials, all mixtures were crystalized into cristobalite. The result of the EPMA suggested that carbon was incorporated into cristobalite. The intensity of CK α was apparently increased with the carbon content. The amount of carbon in cristobalite was < 0.4 wt%. The powder XRD analysis revealed that all peaks corresponding to cristobalite were shifted to the lower angles, indicating the increases of unit cell and volume with the carbon content. Furthermore, cristobalite incorporating carbon in the structure was reheated in a quartz tube at 1300°C for 3days. The results of powder XRD showed that all of the peak positions came back to the original positions, which suggested that the carbon in crystobalite was resubstituted with oxygen atom. Therefore, these results support the following equation: SiO₂ + xC \leftrightarrow Si(C_xO_{2-x}) + (x/2)O₂ (0 ≤ x ≤ α)

Here, α is the solubility limit.

Consequently, it can be concluded that carbon can be substituted with oxygen in cristobalite at high temperature. In addition, we will report quantitative experimental results of temperature dependence on carbon solubility in cristobalite in detail.

Keywords: carbon solubility, silicate mineral, powder XRD, EPMA