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Phase transformation upon cooling path in Ca_2SiO_4 : Possible geological implication

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At the contact metamorphism zone two different Ca_2SiO_4 phases can be found; calcio-olivine (γ phase) and larnite (β phase). In-situ experiments illustrated the existence of five various polymorphs in Ca_2SiO_4 , i.e., α , α'_H , α'_L , β and γ . The path of phase transformation and the transformation temperatures are shown as follows.

$$\begin{array}{cccc} \gamma & \rightarrow & \alpha'_L(700^{\circ}\mathrm{C}) \rightarrow & \alpha'_H(1100^{\circ}\mathrm{C}) \rightarrow & \alpha~(1450^{\circ}\mathrm{C}) \\ \\ \alpha'_L \rightarrow & \beta~(680^{\circ}\mathrm{C}) \rightarrow & \gamma~(500^{\circ}\mathrm{C}) \end{array}$$

Experiments showed that the phase transitions at lower temperature is not reversible and seemed to be complicated; β phase is only stable from 500°C to 680°C upon cooling. To understand the possible mechanism of the β phase being metastable at room temperature, atmosphere condition, we were motivated to investigate the route of phase transition in Ca₂SiO₄ in different thermal process.

Powder samples were synthesized by the solid-state reaction. Pure reagent oxides $CaCO_3$ and SiO_2 were mixed in 2:1 stoichiometric mole. Two control factors were designated in the experiments; the sintering temperature of starting materials and the cooling path. The sintering temperature was set within the range of stable phase field of α'_L phase (\sim 900°C) and α'_H phase (1300°C). The cooling process was designed in three different routes: 1) the quenched procedure from sintering temperature with rate of 900°C/min and 1300°C/min, 2) the furnace cooling procedure, 3) set a slow cooling rate (0.265 °C/min). The products were examined for the crystal structure by X-ray powder diffraction. First-principle calculation was also applied to investigate the thermodynamic properties of α'_H , β and γ phases.

A major finding in this study showed that the γ phase presented in the final product when the sintering temperature was set at the stable field of α'_H phase; on the other hand, the β phase would present when the sintering temperature was set within the field of α'_L phase. It was noted that the existing phase in the product would be modified by the cooling procedures. Our calculation indicates the enthalpy of beta phase was slightly higher than that of the gamma phase at zero pressure, verifying the metastable β phase observed in the natural. In this meeting we present the detailed experimental results and discuss the potential implication for the thermal history of geological setting using the phase transition path upon cooling of Ca₂SiO₄.