K133-P014 **Room: Poster Session Hall** **Time: May 16**

Crystallization experiments in the system Ab-Qtz-H2O: Role of supercritical fluid and flux elements

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Supercritical fluid (fluid, hereafter) plays a central role in crystallization of magmas including formation of pegmatite (Jahns and Burnham, 1969). The flux elements such as B, F, P and Cl concentrate in the granitic melts and related fluids. Recent studies revealed that these elements significantly affect the characters of silicic melts by increasing H_2O solubility, decreasing solidus temperature, and inducing liquid immiscibilities.

The importance of free fluid phase for the formation of pegmatite texture in the hydrothermal experiments has been controversial. London et al. (1989) pointed out that a zoning which are characteristic of pegmatites, were reproduced in the fluid-undersaturated experiments. On the contrary, Baker and Freda, (2001) carried out fluid-saturated experiments and observed a formartion of spherulitic quartz-K-feldspar intergrowth and larger individual crystals of quartz or K-feldspar within the intergrowth. These textural changes were attributed to a change in the medium of crystal growth from a silicate melt to an aqueous fluid.

The purpose of this study is to understand the role of free fluid phase in the eutectic crystallization processes in the system Ab-Qtz-H_2O. The experiments were performed by using a piston-cylinder apparatus at 0.8GPa. First, we melted the starting materials at a temperature of 682 deg. C; 20 degrees higher than the eutectic temperature (662 deg. C;Boettcher and Wyllie, 1969), and held at 3000 minutes for the purpose of identifying the quench crystals (melting experiments). Since the solubility of H_2O at 0.8GPa in the system is 8.8 wt. % (Holtz et al., 1992), we added 8.5 and 15 wt. % water for the fluid undersaturated and saturated experiments, respectively. The $B(OH)_3$ and NaCl were also added fluid-saturated runs. Then, the crystallization experiments were performed from 682 to 512 deg. C at a cooling rate of; 0.05 deg./min, and the texture differences were examined to see the effect of differences in the fluid volume and flux elements.

In the quenched run products of the vapor-undersaturated melting experiments, several spherulites 800 micrometers in diameter were formed. The spherulites consist of submicron eutectic intergrowth of albite and quartz, partly showing the graphic texture (ca. 20 vol% of the melts). In the fluid saturation experiments, the melt was mostly quenched as homogeneous glass with a few small skeletal quartz quench crystals. In a $B(OH)_3$-added experiment, the H_2O solubility was increased, the Cl enhanced the formation of spherulitic intergrowth (up to ca. 40 vol% of the melts).

In the crystallization experiments, there was a great difference in the degree of crystallization between the fluid saturated and undersaturated runs. Coarse-grained euhedral albite and quartz crystals were formed in the cavities, the percentage of which was ca. 3 and 20 vol.% of the capsules in the fluid undersaturated and saturated experiments, respectively. The maximum size of the quartz crystals was 50 micrometers in the saturated experiments. The crystallinity and size of the crystals in the saturated runs are much larger than those expected from the slow diffusivity in the super-cooled melts and a short run duration (57hr). These results suggest that the crystallization in the fluid-saturated runs occurred through dissolution of silicate components into the fluid and precipitation from it, both of which are rapid process compared to the diffusion in the melt. Assuming that natural silicic melts follow this crystallization pathway, we may explain the observation that pegmatite crystallization is faster than crystals in the other plutonic rocks (Webber et al., 1999).

The zoning from the spherulitic albite-quartz intergrowth to the larger euhedral crystals of quartz and albite in the fluid-saturated runs resembles the characteristics zonation of pegmatitic veins.