Experimental determination of second critical endpoints in fluid/melt systems

R. Stalder
P. Ulmer
A. B. Thompson
D. Günther

Department of Earth Sciences, Sonneggstr.5, ETH Zentrum, 8092
Zürich, Switzerland

Fluid phases play an important role during many processes in the upper mantle and in the crust. They work as efficacious transport agents during mantle metasomatism and depress the solidus of silicates towards lower temperatures. Consequently, they have a prominent influence on the velocities of seismic wave propagation and on the rheological properties of rocks. However, even in modern literature the term 'fluid' is not adequately well defined and it is even used as a synonym for all non-solid phases. One reason for this loose definition is certainly the fact that fluids (especially water dominated ones) and silicate melts show converging similarites with increasing pressure. In many silicate-water systems, the water content of the melt and the silicate content of the fluid increase steadily with increasing pressures. In the silica-water system this behaviour is continued up to a second critical endpoint, where a fluid and a melt are no longer distinguishable and the term 'solidus' has to be reconsidered (Kennedy *et al.*, 1962). Investigations in other simple systems (e.g. diopside-water, Eggler and Rosenhauer, 1978) revealed a similar behaviour, but for more complex mafic and ultramafic compositions a second critical endpoint has - due to experimental limitations - not yet been determined.

The present experimental study started with an inspection of the system albite-water in order to evaluate the accuracy of a new technique to determine major element solubilities in fluid phases and water solubilities in melts. The system albite-water has previously been examined and a consensus with respect to phase relations, solidi and water solubilities in the melts has been achieved. Above 15 kbar the system approaches a second critical endpoint, where a distinction between fluid and melt can no longer be made (Boettcher and Wyllie, 1969; Paillat *et al.*, 1992) and therefore a water-saturated solidus does no longer exist. Therefore it seems to be a suitable system to test our technique, which previously has only been applied to fluid/mineral trace element partitioning (Stalder *et al.*, 1997).

In the next step this technique is being applied to the system $MgO-SiO_2-H_2O$ at pressures of 60 to 100 kbar and 900 to 1100°C, where the transition

$$\text{Fo} + \text{En} + \text{Fluid} \rightarrow \text{Fo} + \text{En} + \text{Melt}$$

is expected to occur (Irfune *et al.*, 1998).

Experimental runs in the system albite-water were carried out at near-solidius conditions in a 14 mm piston cylinder apparatus at 5–15 kbar and 645–775°C. Experimental charges contained low-albite, water and a layer of diamond crystals (grain size 50 μm). The pore space between the diamonds is preserved during the run, thus the fluid was able to circulate throughout the entire capsule. During quenching the material dissolved in the fluid precipitated between the diamond crystals and could be separated from the solid residue. The recovered capsules were embedded in epoxy and the diamond layers were analysed by laser ablation microprobe (LAM-ICP-MS). Ni, which was present in the diamond crystals at the level of 20 ppm, was used for internal standardisation.

Initial results indicate that both aqueous fluids (low silicate/water ratio) and hydrous albitic melts (high silicate/water ratio) can be properly analysed with the applied techniques.

Albite dissolves non-stoichiometrically at 5 and 10 kbar with silica being the most strongly dissolved component. The Al/Na ratio in the fluid increases with increasing pressure approaching stoichiometric values at 10 kbar. Although an incongruent dissolution was observed, phases other than albite were not detected. Probably the amount of dissolved material was low enough to allow the residual component to dissolve completely in the albite lattice.

Initial results for a synthetic serpentine composition reveal, that at 900°C and 60 kbar the silicate/water ratio of the non-solid phase is considerably lower than 1 and Fo + En + Fluid is the resulting phase assemblage.

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