MELT INCLUSION EVIDENCE FOR IMMISCIBILITY BETWEEN SILICATE AND SALT LIQUIDS IN A COMMON GRANITE MAGMA

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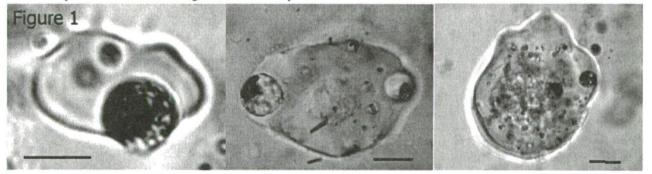
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Volatiles dissolved in aluminosilicate magmas (dominantly H₂O, CO₂, S and Cl) play a key role in the processes transitional from magmatic to hydrothermal systems. When the volatiles are exsolved from parental magmas, due to the processes of unmixing or immiscibility, and form a separate volatile-rich phase (fluid), a major change in the element partitioning occurs. It is believed that volatile phases exsolved during the crystallisation of magmas carry significant amounts of metallic elements. For many felsic magmas, the evolution of immiscible volatile phases takes place at temperatures near the magmatic solidus. This interval of magmatic cooling and crystallisation has proved extremely difficult to document and understand, because of the transient, reactive nature of immiscible volatile phases. The questions about physical state and chemical compositions of late magmatic fluid can be answered by employing melt and fluid inclusion research on samples representative of magmatic-hydrothermal transition. Despite the contention that "... interpretation of the inclusion record is commonly ambiguous and difficult, particularly that stage between silicate melt and the hydrous saline melts and aqueous solutions..." (Roedder, 1992), in recent years the inclusion approach has gained strength from the use of modern microbeam techniques (laser Raman spectroscopy, laser ablation ICPMS, PIXE - proton-induced X-ray emission). We aimed the study of inclusions in phenocrystic and miarolitic quartz from the tin-bearing Omsukchan granite (NE Russia) to describe the occurrence of immiscibility between the silicate melt and late magmatic non-silicate, volatile-rich phases, characterise their composition from microthermometric experiments and in situ analysis, and attempt to constrain the physical nature and chemical signature of magmatic derived fluids and their high temperature evolution.

Inclusions in quartz phenocrysts

Prior to heating we recognised two main types of coexisting primary magmatic inclusions: (1) birefringent microcrystalline aggregates of presumably silicate minerals with a misshapen vapour phases filling the spaces between solids, interpreted to be inclusions of crystallised silicate melt, and (2) magmatic volatile-rich phase consisting of variable proportions of aqueous vapour, liquid and euhedral crystals. The endmembers within this type are represented by essentially vapour (>90 vol%) and essentially crystalline (>70 vol%) inclusions. At least nine different mineral species (halite and hematite are tentatively identified) present among solids, and often within a single inclusion. In highly crystalline volatile-rich inclusions (hereafter, brine or salt melt) liquid exists interstitially, and a vapour bubble is deformed.

After bulk heating of inclusion-bearing quartz phenocrysts (1 atm furnace, 850-950°C, 20-50 hours) and consequent air quenching we found that the majority of silicate melt inclusions were converted into a clear glass containing spherical globules, composed of microcrystalline material with variable proportions of spherical to deformed vapour phase (Fig. 1; scale bars are 10 µm). The number (from 1-2 to 10's) and sizes (<1 - 15 µm) of globules vary significantly. Commonly in larger inclusions these crystal- and liquid-bearing globules form an emulsion (globules are suspended in the glass "matrix"). The globules are also present without silicate glass, in the host quartz.



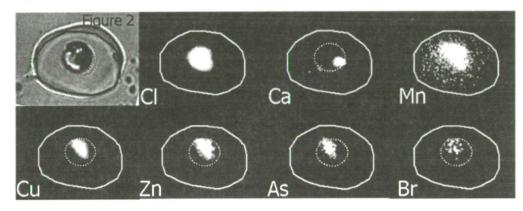
Heating stage experiments with the spherical globules inside silicate glass inclusions show the following (1) globules remain constant in size, do not change their position with respect to silicate melt, and do not mix with silicate melt even at temperatures exceeding liquidus (900-1100°C); (2) first recrystallisation and melting occur at 120-190°C; (3) vapour phase disappears at 420-510°C, and then renucleates at ~580°C. (4) melting of solid phases becomes noticeable at >600°C and is seen as appearance of liquid (melt) surrounding a cubic crystal; (5) above 630°C crystals decrease rapidly, and melt completely at 670-735°C; (3) vapour bubble shrinks and moves randomly and rapidly, until final disappearance at 790-935°C. The above phase transformations take place almost simultaneously in all entrapped globules within an individual silicate melt inclusion. The observed differences between globules in different inclusions can be tentatively attributed to the effects of internal

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pressure. Similar behaviour is observed for glass-free, spherical inclusion embedded in quartz, although the melting and homogenisation temperatures can vary by 100-150°C for several tens inclusions studied, even for those within same quartz phenocryst.

Composition of the dispersed phase in magmatic emulsion

Based on the phase composition of inclusions in quartz phenocrysts, the crystallising system of the Omsukchan granite consisted of at least three-phases - silicate melt, saline fluid (brine) and low-density vapour. Following the earlier studies (e.g., Reyf, Bazheyev, 1977; Frezzotti, 1992), we interpreted these phases as formed due to immiscibility of the precursor granitic magma that happened before or during quartz crystallisation. A clear case of immiscibility with at least two liquids simultaneously present during quartz growth, with a meniscus between them, is best demonstrated by the emulsion textures of many inclusions, in which multiple brine globules (i.e., the dispersed phase) are randomly distributed within silicate glass (Fig. 1). The distribution and abundance of elements heavier than Cl within brine globules were studied by PIXE using the CSIRO-GEMOC Nuclear Microprobe (Fig. 2; outlines on element maps mark boundaries of the inclusion and brine globule). Both types of globules (i.e. trapped with silicate melt and included in quartz) have the same geochemical signature. They are highly enriched in Cl (up to 50 wt%), moderately in K, Fe, Cu (up to 20 wt%), and to a lesser extent in Ca, Mn, Zn and Pb (0.2-2 wt%). Even elements with concentrations of several hundred ppm (Se, As, and Br) can be regarded as highly enriched relative to granitic magmas. Therefore, the globules are metal-rich chloride complexes, although the element abundances and ratios vary significantly, even for the globules, co-trapped within a single silicate melt inclusion. There may be numerous reasons for



this, e.g., exsolution of globules (now co-trapped) from different magma batches. But the most immediate is an extreme fractionation of most elements between immiscible silicate melt and non-silicate "fluids" and the disequilibrium character of exsolution.

Although it is expected that coalescence of immiscible volatile-rich

phases may reduce chemical variability, however, we anticipate that different batches of magmatic "fluids" have different histories in transit to miarolitic cavities (in effect fluid chambers) where they accumulate. If this is the case, discrete batches of fluids, following a rock-dominated chemical path, are prone to modifications caused by interaction with a granite (solid rocks and residual magma pockets). Moreover, implied unmixing of the fluids due to boiling or effervescence, cooling and related crystallisation, and mixing of fluid batches with each other and possibly external fluids can be responsible for even greater fluid variability, and the change of primary magmatic geochemical signature, as observed in inclusions trapped by quartz from miarolitic cavities in the Omsukchan granite (Kamenetsky et al., 2002). Although some element ratios remain relatively constant for brine inclusions in earlier phenocrystic quartz, and later miarolitic quartz, the concentration of Cu and proportion of Cu to other metals decrease dramatically in miarolitic brines (from wt% to hundreds of ppm). Such discontinuous evolution of the fluid and the absence of Cu mineralisation in the Omsukchan granite pose the question whether magmatic fluids in passage to miarolitic cavities experienced separation and loss of vapour-rich phase in which Cu strongly partitions.

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