

The role of fluid-phase immiscibility in quartz dissolution and precipitation in sub-seafloor hydrothermal systems: Implications for fluid inclusion studies of volcanogenic massive-sulphide deposits

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Volcanogenic massive sulphide (VMS) deposits represent the fossil analogues of the active hydrothermal vents in the seafloor found at oceanic spreading centres (Galley *et al.*, 2007). Fluid inclusion (FI) studies of both active (“black smokers”; *e.g.* Vanko *et al.*, 2004) and fossil (*e.g.*, Pisutha-Arnond & Ohmoto, 1983) VMS systems have provided much information on the physical and chemical conditions of formation of these deposits. The vast majority of all FI studied from VMS deposits are hosted in quartz. Primary inclusions in quartz are trapped as quartz precipitates from hydrothermal fluid – primary inclusions are unlikely to be trapped where and when quartz is being dissolved. Therefore conditions of quartz dissolution will be under-represented in the primary inclusion record (although those conditions may be preserved in secondary inclusions). The properties of quartz-hosted FI can provide information on the physical and chemical conditions of quartz precipitation. Ideally those conditions should be related to hydrothermal processes, but sub-seafloor processes cannot be directly observed, and must therefore be modelled and/or inferred.

The occurrence of phase separation in sub-seafloor hydrothermal systems is now widely recognized based on vent chemistry data (*e.g.*, Von Damm, 2004), although less is known about how sub-seafloor immiscibility affects fluid-rock interaction in VMS deposits. Fluid inclusion assemblages in quartz from active VMS-forming systems sometimes contain coexisting liquid-rich and vapour-rich inclusions, revealing that the host quartz was precipitated from immiscible fluids (de Ronde, 1995; Vanko *et al.*, 2004). Halite-bearing inclusions are commonly found in VMS quartz, and may represent brine generated during phase separation, although they are usually found without coexisting vapour-rich inclusions.

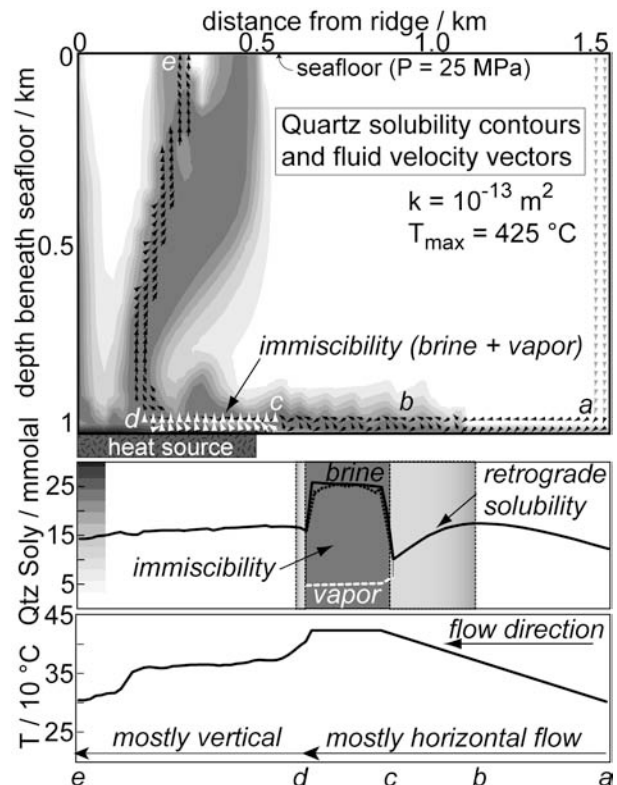


Fig. 1. Example flow path (small arrows are fluid flow vectors, top), and the quartz solubility (middle) and temperature (bottom) along the path. Points labelled a-e on the top diagram correspond to the x (distance) axis on the middle and bottom plots. Fluid flows from the lower right (a) towards the lower left (c-d, the heat source) and then flows upwards to vent at the seafloor (e). Quartz solubility first increases (a-b), then decreases (b-c), with increasing temperature. Immiscibility occurs at $T > 410$ °C (c-d), and quartz solubility is higher in the high-salinity brine and lower in the low-salinity vapour (white vectors). Bulk quartz solubility in the two-phase zone is dominated by the brine. There is an abrupt drop in quartz solubility going from two-phase to one-phase conditions (d).

In the present study, we couple numerical fluid flow modelling of sub-seafloor hydrothermal systems with a model for dissolution and precipitation of quartz, in order to predict where, when and by what mechanisms quartz is likely to be precipitated (and to trap FI in VMS deposits).

We model miscible and immiscible hydrothermal circulation using the code *FISHES* (Lewis and Lowell, 2009) which solves the conservation equations governing multi-component, multi-phase fluid flow in porous media. We calculate quartz solubility in H₂O-NaCl fluids (representing seawater and phase-separated seawater) using the model of Akinfiyev and Diamond (2009). Quartz dissolves when the quartz solubility in the fluid increases along its flow path, and quartz precipitates in the opposite scenario.

Effects of retrograde quartz solubility and fluid-phase immiscibility are observed in this model (Fig. 1). Retrograde quartz solubility causes quartz to be precipitated as fluid heats along part of the flow path (elsewhere, quartz dissolves as the fluid heats). Immiscibility creates a region in which quartz solubility is significantly higher in the higher-salinity brine phase and lower in the lower-salinity vapour; overall quartz solubility in that region is controlled mostly by the brine (Fig. 1). Across the interface from two-phase to one-phase conditions, there is a steep decline in quartz solubility over a relatively short distance (Fig. 1), resulting in large amounts of quartz precipitation.

As fluid flows along the solubility gradients shown in Figure 1, quartz is precipitated and dissolved, and the rate of dissolution/precipitation depends on the solubility gradient as well as the fluid mass flux. Two examples of distribution of dissolution and precipitation are shown in Figure 2. In both examples, quartz is precipitated where fluid flows upward as hot plumes and vents at the seafloor. There is a significant difference between the two examples: in the upper example (Fig. 2, top) the bottom temperature is set just high enough so that phase separation occurs, whereas in the lower example (Fig. 2, bottom) the temperature is kept just below that needed for phase separation. Where immiscibility occurs (Fig. 2, top), a zone develops in which quartz is precipitated at the highest rates, along the two-phase to one-phase

interface (where immiscible brine and vapour flow upwards and re-mix). This outcome suggests that FI in quartz from VMS deposits may preferentially record conditions *just above* the two-phase region.

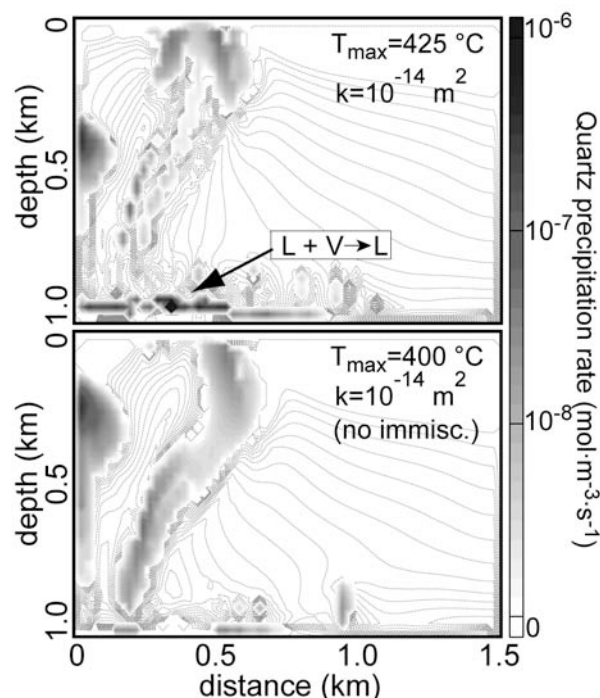


Fig. 2. Contoured rate of quartz precipitation throughout the system, comparing a simulation in which immiscibility occurs (top) with one in which immiscibility does not occur (bottom). Maximum rate of quartz precipitation is found in the simulation with immiscibility, at the interface between two-phase and one-phase conditions (L + V → L).

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