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Extreme chemical heterogeneity of granite-derived hydrothermal fluids: An example from inclusions in a single crystal of miarolitic quartz

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ABSTRACT

Magma-derived fluids are important in geologic processes (e.g., metal sequestration and ore deposition) but are intrinsically transient. Samples of magmatic fluids represented by fluid inclusions in a single zoned quartz crystal from a miarolitic cavity within a porphyritic leucogranite hosting the Industrialnoe tin deposit, northeastern Russia, were studied by using modern in situ analytical methods (laser Raman spectroscopy, proton-induced X-ray emission). The fluid inclusions are either dominated by vapor or by complex multiphase brines. The inclusions within a given trapping plane have similar phase relationships; however, there are significant variations between inclusions in different healed fractures. Phase and chemical compositions of individual brine inclusions demonstrate significant compositional heterogeneity (in terms of absolute element concentrations and ratios) of high-temperature magmatic fluids accumulated in the miarolitic cavity. This finding suggests that fluids leaving a crystallizing magma may have variable initial compositions that are subsequently modified by reactions with the rocks while the fluid is in transit to a miarolitic cavity, as well as by processes in the cavity, such as mixing, crystallization, and boiling. The inferred chemical diversity and fractionation of granite-derived fluids at near-magmatic conditions imply that fluids entering a cooler hydrothermal system are extremely complex and their metallogenic signature may differ from that of related ore deposits.

Keywords: granite, fluid inclusions, ore deposit, PIXE, geochemistry.

INTRODUCTION

Fluids of different kinds are among the most important components in all geologic environments (e.g., igneous, sedimentary, metamorphic, metasomatic, hydrothermal) and events (e.g., melting, crystallization). However, the record of fluids and their properties is usually indirect and incomplete because of their transient nature. Fluids responsible for a variety of geologic process are prone to consumption and exhaustion in chemical reactions, and/or they leave Earth's interior for the atmosphere and hydrosphere by degassing.

Fluid inclusions trapped and sealed in crystals are by far the most representative of fluid compositions that existed during and after crystal growth (e.g., Roedder, 1979). In a way similar to melt inclusions trapped in early phenocrysts as instantaneous mantle-derived melt fractions (e.g., Sobolev, 1996), the compositions of individual fluid inclusions in igneous rocks are representative of magma-derived fluids, composing the ''primitive'' hydrothermal system, and are expected to differ significantly from each other and from the estimates of the ''bulk'' fluid composition. At present, reliable data on fluid compositions are hampered by many factors; i.e., the small size of fluid inclusions and their heterogeneous phase composition (unlike melt inclusions that can be homogenized and quenched into glass), the availability of quantitative analytical techniques, and the number of fluid inclusions that can be reasonably studied. Nevertheless, variations among individual inclusions have been documented through the estimates of fluid densities and salinities via cooling and heating experiments (see reviews in Roedder, 1984; Wilkinson, 2001), and variations in the element abundances have been detailed via in situ studies (laser ablation ICP-MS [inductively coupled plasma–mass spectrometry], e.g., Audétat et al., 1998, 2000a; Heinrich et al., 1999; Kamenetsky et al., 1999; Rankin et al., 1992; Ulrich et al., 1999; synchrotron X-ray fluorescence, e.g., Rankin et al., 1992; Vanko et al., 2001); and PIXE [proton-induced X-ray emission], e.g., Damman et al., 1996; Heinrich et al., 1992; Ryan et al., 1995, 2001a; Vanko et al., 2001; Williams et al., 2001).

Fluids involved in the transition from silicate magmas to hydrothermal fluids are of particular interest to many geologists because these fluids are thought to sequester metals from magmas and/or rocks and carry the metals to the sites of ore deposition. Although it is well known that the series of fluids derived from felsic magmas over a significant cooling interval (~200–300 °C) are predominantly aqueous chloride solutions, the metallogenic signatures and metal concentrations of a particular fluid fraction require further research. This paper provides an insight into the extreme compositional variability of individual magma-derived fluids represented by multiphase (aqueous solution + daughter minerals + vapor bubble) fluid inclusions trapped during growth of a single zoned crystal of quartz from a miarolitic cavity in a tin-bearing granite.

SAMPLES

The Industrialnoe tin deposit is located within porphyritic leucogranites of the Cretaceous (79 ± 4 Ma, according to K-Ar dating) Omsukchan massif, northeastern Russia (Naumov and Sokolov, 1981; Sokolov, 1980). Tin (cassiterite) mineralization is present in linear zones of disseminated metasomatic alteration of granites (quartz + sericite + chlorite ± tourmaline) and in related quartz and fluorite veinlets. The ore grades increase in the central, most iron-enriched parts of metasomatic columns. Orebodies have very simple phase compositions, i.e., dominant quartz, tourmaline, chlorite, and sericite, and minor cassiterite, fluorite, magnetite, and hematite. Other minerals (albite, calcite, arsenopyrite, scheelite, and adularia) occur sporadically, but iron sulfides are almost absent despite significant influx of iron during metasomatism (Sokolov, 1980). Miarolitic cavities filled with large (to 10 cm) euhedral quartz and fluorite-tourmaline interstitial ag-

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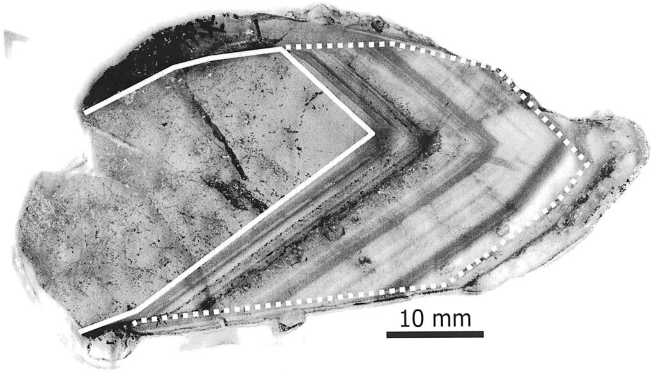

Figure 1. Section parallel to *c*-axis through central part of quartz crystal from miarolitic cavity. Solid line marks root zone that contains studied fluid inclusions. Dashed line contours middle zone with numerous growth planes.

gregates are common in the vicinity of orebodies, and are also recorded in less altered granites.

The studied crystal of quartz (Fig. 1) from a miarolitic cavity (0.5 m in diameter of the exposed part, Naumov and Sokolov, 1981) has a large unzoned core (root zone), surrounded by numerous growth planes (middle zone), and followed by several growth planes with tourmaline and fluorite on the prismatic faces (outer zone). Fluid inclusions and empty cavities are most abundant in the root zone, but are also present throughout the crystal.

RESULTS

Fluid Inclusions

Trails of inclusions are most commonly aligned with healed fractures crosscutting growth planes or randomly oriented in the root zone. The trails and inclusions in them are so abundant and so densely and randomly distributed that identification of truly primary inclusions (single or in small three-dimensional clusters; Roedder, 1979) was not practical. However, because almost all of the trails in the root zone terminate at the boundary with the middle zone and the trails in the middle zone are included entirely within growth planes, the inclusions in both zones are interpreted as primary or pseudosecondary.

The fluid inclusions vary significantly in size (to 300 μm) and phase composition. Two main types of inclusions present in different or the same trails are noted: (1) vapor rich with minor amounts of liquid, often containing a few euhedral crystals (Fig. 2A), and (2) brine inclusions with variable amount of liquid and solids (Fig. 2, B–G). The brine inclusions can therefore be subdivided into liquid rich (Figs. 2B and 3A) or crystal rich. In the latter, the number of crystals is typically >4 (Fig. 2, C–G, and Fig. 3, B–D); a few inclusions have as many as 14 crystals (Fig. 2D). The volume percentage of crystals does not vary much between brine inclusions in the same trail. The volume of the vapor bubble in brine inclusions is typically 10%–15%, and this ratio has been used to distinguish inclusions formed from a homogeneous fluid.

The crystals in brine inclusions are typically euhedral, although they may coexist with aggregates of tiny crystallites (Fig. 2, F and G). Cubic, pseudocubic, hexagonal, pyramidal, and prismatic forms are common, as are crystals with high relief and birefringence. However, there is no order or pattern to the alignment of the crystals and no clear relationships between the shape, color (except for hematite plates), birefringence, and relief of crystals. This diversity makes it difficult to recognize whether the same assemblage of crystals is present in inclusions belonging to the same trail. In addition, most daughter crystals are metastable, and thus seemingly different fluid inclusions do not necessarily represent chemically different fluid batches.

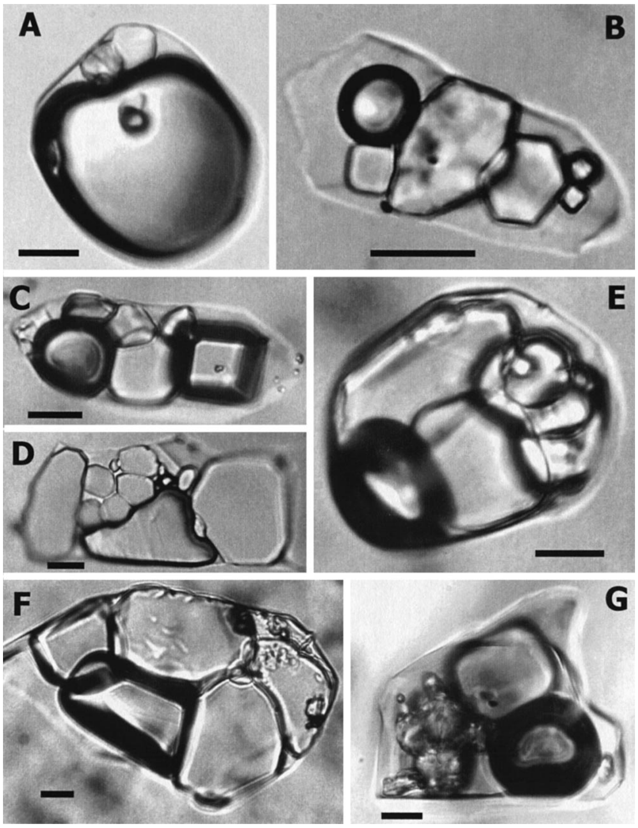

Figure 2. Representative fluid inclusions from root zone of quartz crystal. A: Vapor-rich inclusion containing small amount of liquid and several solid phases. B: Liquid-rich inclusion with vapor bubble, five clear euhedral crystals, and several small opaque minerals. C–G: Crystal-rich inclusions from different healed fractures. Note strong variability in number of crystals and in their size, morphology, and relief. Scale bars represent 10 μm.

Compositions of Brine Inclusions

The phase assemblage is variable from one inclusion to another, and at least 10 different solids have been distinguished by Raman spectroscopy (Dilor SuperLabram, Australian Geological Survey Organisation, Canberra). Usually one or two crystals in each inclusion did not produce a Raman spectrum, a result in accord with the predicted weak nature of the Raman spectra of diatomic ionic crystals. No carbonate, sulfate, phosphate, nitrate, or borate bands were observed in these spectra, but antarcticite ($CaCl_2 \cdot 6H_2O$) and $SnCl_2 \cdot 2H_2O$ have been tentatively identified. The weak bands between 600 and 750 cm^{-1} exhibited by some solids indicate the presence of silicates. Other crystals produced one or more relatively sharp bands below 400 cm^{-1}, and most of them also exhibited very intense OH-stretching bands between 3400 and 3450 cm^{-1}. Their Raman spectra do not correspond to those of the common minerals. Therefore, these crystals are thought to be complex hydrates that may only metastably exist in concentrated brines of fluid inclusions. Halite (isotropic cubes), hematite (reddish hexagonal plates), and magnetite (magnetic) are also present.

Nuclear microscopy (CSIRO-GEMOC Nuclear Microprobe; Ryan et al., 1995, 2001a, 2001b) by PIXE was used to image element distribution and quantify the compositions of 35 solitary (primary?) fluid inclusions within the root zone of the quartz crystal (Fig. 1). All these inclusions were trapped homogeneously (bubble size is 10–15 vol%), and most likely many of them belong to different trapping events.

The photomicrographs (before and after analysis) and PIXE element images of typical inclusions (Fig. 3) yielded the following observations. (1) The crystal phases were not stable during analysis, an

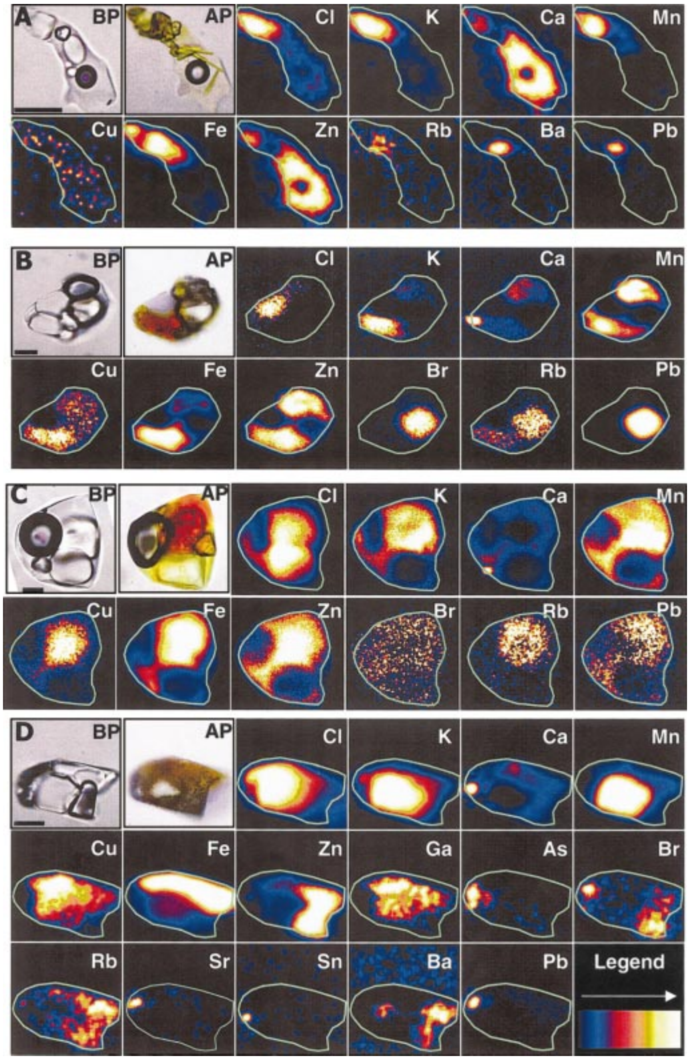

Figure 3. Optical images and proton-induced X-ray emission (PIXE) element maps of individual brine inclusions. A: Liquid-rich inclusion (7-1; see footnote 1). B–D: Crystal-rich inclusions (1-1, 3-3, and 5-3, respectively; see text footnote 1), analyzed by using a 0.3–0.7 nA beam of 3 MeV protons focused into ~2 μm beam spot with new CSIRO-GEMOC nuclear microprobe (Ryan et al., 2001a, 2001b). Legend shown in lower right image of D indicates increasing intensity (and thus concentration) as shown by arrow. Color scale in each element image is normalized to its own maximum. Optical images are shown for inclusions before (BP) and after (AP) PIXE analysis. Outlines on element maps mark boundaries of fluid inclusions. Scale bars on BP images represent 10 μm.

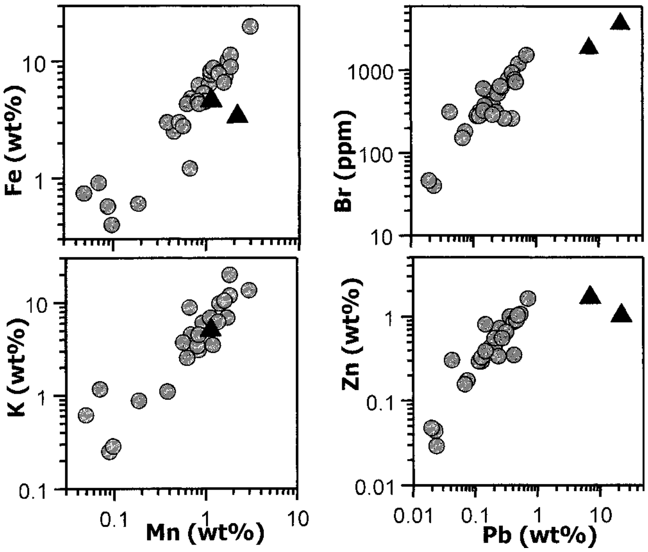

Figure 4. Representative covariations for compositions of fluid inclusions from root zone of quartz crystal. Separate symbol (triangle) is used to show two inclusions (1-1 and 1-2; see text footnote 1) anomalously enriched in Pb and Br (inclusion 1-1 is also presented in Fig. 3B). Error bars are smaller than symbol size.

unusual behavior for PIXE analysis. Some broke down or moved, some changed shape, size, or color, and new phases formed. (2) Vapor bubbles commonly moved and changed shape, and the liquid always acquired light yellow to reddish-brown coloration. (3) Individual solid phases were highly variable in terms of metal content. For example, we recognized crystals enriched in Pb and Br (Fig. 3, B and D); Pb and Ba (Fig. 3A); K and Mn (Fig. 3, A and D); Fe, Mn, and Zn (Fig. 3, B and C); Cu (Fig. 3C); Ca (Fig. 3, B and D); and Sn (Fig. 3D). (4) Cl is a major anion, present in crystals and aqueous solution (Fig. 3). (5) Composition of the liquid is undefined in most brine inclusions owing to strong fluorescence from crystals, although in one liquid-rich inclusion, the presence of Zn and Ca is unambiguous (Fig. 3A).

The PIXE-derived element concentrations (Fig. 4[1]) increase significantly (20–90 times) from low-salinity (3 wt% Cl) liquid-rich (e.g., Fig. 3A) to high-salinity (55 wt% Cl) crystal-rich (e.g., Fig. 3, B and D) inclusions. The abundances of major elements (Cl, K, Fe, Mn, Zn, Pb, Cu, Br) in all inclusions (except two Pb- and Br-rich inclusions; see footnote 1) are generally correlated (Fig. 4), although the element ratios deviate significantly (to 65%) from the average (Table 1).

DISCUSSION AND CONCLUSIONS

A miarolitic cavity forms in fluid-saturated conditions, and it is a semiopen system in which different magma-derived fluids accumulate, mix with each other, and fractionate owing to crystallization and boiling. The fluid inclusions trapped in crystals growing in such a cavity allow us to examine fluid compositions once present in the magmatic-hydrothermal system and a variety of processes (e.g., crystallization, mixing, reequilibration) that may have affected these fluids.

The coexistence of both vapor-rich and brine inclusions in the central part of the root zone (Fig. 1) indicates that boiling occurred in the initial stages of crystallization of the quartz crystal. These inclusions may represent the most pristine and hottest granite-derived and

TABLE 1. SELECTED ELEMENT RATIOS IN BRINE INCLUSIONS FROM A ROOT ZONE OF A MIAROLITIC QUARTZ CRYSTAL

	Fe/Mn	Fe/Zn	Fe/Cu	Fe/Pb	Fe/Rb	Mn/Zn	Zn/Pb	Pb/Br	K/Mn	K/Cl
Minimum	1.7	1.1	54	2.8	18	0.37	0.8	1.1	2.0	0.06
Maximum	14.6	32	617	103	227	3.1	7.1	17.5	15.7	0.61
Average	5.9	12	263	27	80	2.1	2.3	5.5	5.8	0.22
Standard Deviation	2.4	5.3	158	18	47	0.57	1.2	3.2	3.4	0.14

Note: Data for two Pb- and Br-rich inclusions (1-1 and 1-2) are excluded.

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[1] GSA Data Repository item 2002046, Appendix Table 1, Composition of fluid inclusions in miarolitic quartz from Industrialnoe tin deposit (northeast Russia), is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, editing@geosociety.org, or at www.geosociety.org/pubs/ft2002.htm.

granite-percolating fluids (in this case, to 840 °C, whereas the lower end of the range of magmatic temperatures of the host granite has been established by silicate melt inclusion study at 760 °C; Naumov and Sokolov, 1981). Inclusions within individual fractures have near-identical phase ratios and presumably similar compositions, indicating that they were trapped from a homogeneous fluid, but the compositions of inclusions representing different trapping events vary greatly (Table 1; Fig. 4; see footnote 1), suggesting vastly fluctuating fluid chemistry during the growth of this crystal.

The compositional heterogeneity among the inclusions (Table 1; Figs. 3 and 4; see footnote 1) supports the notion that ''a hydrothermal fluid is . . . an evolving or changing entity'' (Skinner, 1979, p. 12) and implies chemical variability among their parental fluids. As for fractions of primary mantle-derived melts represented by melt inclusions in magmatic phenocrysts (e.g., Sobolev, 1996), such fluids should show extreme compositional variability resulting from (1) the varying chemical parameters and physical conditions during fluid exsolution (see modeling of fluid compositions based on numerous experimental data in Audétat et al. [2000b] and Cline and Bodnar [1991]), (2) fluid-mineral exchange reactions and boiling (Heinrich, 1990), and (3) chromatographic effects during fluid percolation through the magma and solid rocks. Consequently, individual fluid fractions entering the miarolitic cavity could be largely responsible for the observed variability among inclusions (Table 1; Figs. 2–4; see footnote 1). Equally, chemical modifications may have occurred to a chemically homogeneous fluid during residence in the miarolitic cavity. The processes responsible for chemical fractionation of fluids are commonly assumed to be the precipitation of mineral phases, fluid mixing, and fluid effervescence or boiling (e.g., as represented by vapor-rich inclusions; Fig. 2A). Depending on the partitioning of certain elements between fluids and minerals and between liquids and vapors, these processes may have a dramatic effect on the absolute element concentrations and the element ratios in the residual fluid and subsequent metal distribution and transport. Dilution of magmatic fluids by meteoric waters may be another possibility, as has been shown in a similar study of individual inclusions from the tin- and tungsten-mineralized Mole Granite, Australia (Audétat et al., 1998, 2000a), but in our samples from the Industrialnoe tin deposit, the strongly variable element ratios (Table 1) do not support such progressive dilution.

This study provides ''snapshot'' information about varying chemical parameters during the growth of a single quartz crystal. It may be that some of the granite-derived fluids sampled by the fluid inclusions were genuinely primary (i.e., in equilibrium with parental granite magmas), and as such they demonstrate primary compositional variability governed by locally varying degrees of crystallization of the source felsic magma and different mineral assemblages present at the sites of fluid exsolution. We also envisage a scenario in which primary fluids follow a rock-dominated chemical path (see modeling in Heinrich, 1990). In this case individual fluid batches percolating through a magmatic conduit can be modified from primary compositions due to both reequilibration with residual melts and solid rocks (e.g., metasomatic alteration of granites) and partial crystallization (e.g., dispersed metal deposition in granites). Additional compositional heterogeneity can be introduced to fluids when they reside in the miarolitic cavity due to crystallization, boiling, and multistage mixing. Whatever the origin of the observed fluid diversity, it has occurred at magmatic and near-magmatic conditions. Thus the heterogeneity of near-solidus fluids can be a significant factor underlying the chemistry of later, hydrothermal fluids, and controlling the development of the metallogenic signature of related ore deposits. This magmatic diversity is likely to be eliminated if the fluids are thoroughly mixed in a fluid-dominated system (e.g., in a hydrothermal vein). However, primary fluid heterogeneity may still be important in studies of multiple hydrothermal events.

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