Magmatic-hydrothermal transition in tourmaline-bearing miarolitic pegmatites: Hydrosaline Fluids or Silica Gels?

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ABSTRACT

Inclusions in quartz from tourmaline-bearing pegmatites show a marked evolution between early, massive pegmatites and late miarolitic druzes. Miarolitic fluid inclusions are associated to unusual hydrosilicate inclusions, containing silicate aggregates, composed mainly of daughter mica. These inclusions are believed to derive from silica gels, exsolved from magmatic aqueous fluids at magmatic-hydrothermal transition in pegmatites. These gels play an important role in element fractionation during the final stage of pegmatite crystallization.

Keywords: magmatic-hydrothermal transition, silica gels, fluid and melt inclusions.

For decades, the nature of fluids involved in the crystallization of granitic pegmatites has been a central question in magmatic petrology. Experiments on compositionally simple granitic systems, notably by W. Burnham and co-workers in the early sixties, have led many petrologists to believe that the transition between magma (melt) and fluid (hydrothermal) did occur at the onset of pegmatite formation, and that typical pegmatite fluids were low-salinity, dilute aqueous solutions (see e.g. Jahns and Burnham, 1969; Jahns, 1982). This conclusion however did not cope with the fact that in many cases fluid inclusions in minerals from pegmatites, especially from those enriched in F, B and rare alkali, are highly concentrated, complicated solutions, containing up to 85% dissolved silicate and salt components (Kosukhin et al., 1984; London, 1986, Kovalenko et al., 1996, Peretyazhko et al., 2004; Rickers et al., 2006). These inclusions have very unusual chemical and physical properties that make them considered, either as highly concentrated aqueous fluids or specific volatile-rich hydrosilicate or hydrosaline melts. In order to explain these extreme compositions, a number of hypotheses have been proposed, either related to the fluid itself (boiling), or to complexities of fluid/melt interaction during the final stages of pegmatite crystallization. Tourmaline-bearing miarolitic pegmatites are especially interesting in this respect, for a number of reasons:

- The relative abundance of «hyperfusible» elements (B, F, Li) drastically lowers melting temperatures, while extending silicate liquid-H₂O miscibility field.
- Many pegmatites show a well-defined zoning, from typical granitic early feldspar + quartz zones to late, idiomorphic crystallization in open space (druses). This zoning indicates that the pegmatite has evolved as a nearly closed system, with a limited influence of external sources.
- P-T conditions of mineral crystallization or subsolidus re-equilibration are relatively well known, from a number of experimental studies and some geo-thermobarometers.

A special problem relates however to the great size of many occurrences: not easy to have a complete picture of the distribution of small inclusions, typically few micrometers in size, in meter-size crystals or hectometer-size pegmatites. This may explain why the number of documented cases is small, the Volhyn pegmatite, Ukraina, (Kalyuzhni, 1966, Kovalenko et al., 1996) being a notable exception. Present study, done in the Institute of Geology and Mineralogy, and the Institute of Geochemistry of the Russian Academy of Science, Siberian branch, reports on a number of tourmaline-bearing miarolitic occurrences from Transbaikalia, Russia, and Pamirs, Tajikistan. Despite minor regional differences, common features are so striking that it can be safely assumed that the results of the present study apply to tourmaline-bearing miarolitic pegmatites in general.

From the edge towards the center of the open cavity (miarole), three distinct mineral assemblages are identified:

1) Early pegmatite (about 95% by mass) (Main: Qtz + Fsp, subordinate: schorl tourm. + garnet)

2) Near miarolitic assemblage (Main: Qtz + Ab or Ab + lepidolite, sub. color tourm., petalite, pollucite, Cs beryl, Ta-Nb minerals, etc..

3) Druze complex (Main: Qz, Ab, color tourm., lepidolite.

Melt inclusions accompanied by primary inclusions of aqueous magmatic fluid are present in all zones (except the final stages of the druze complex), with however significant differences:

- Early pegmatite contains tiny inclusions that represent heterogeneous system containing silicate melt and low salinity aqueous fluid, trapped at T > 600°C and P ~ 2kb (Fig. 1, next page).
- Quartz of the near miarolitic assemblages and root parts of miarolitic quartz contain numerous inclusions of silicate liquids (SiO₂=58-72 wt%) that have extremely unusual compositions (Fig. 2, «hydrosilicate inclusions», next page). They were trapped at T not higher than 550°C, precisely at the magmatic-hydrothermal transition. Some are strongly enriched in B, F, Rb, Cs, Ta and Nb, and they may have a very high water content (up to 12-18 wt %). They contain a large number of crystalline phases, from which quartz, micas, topaz, tourmaline, and scarcely jeremeevite were identified. However only quartz and Li- and Cs-rich micas are considered as daughter phases. Others are most probably co-trapped minerals. Typically

crystalline phases form cryptocrystalline aggregates, which resemble devitrified melt.

Although, the average composition corresponds to Qtz +Mica (against Qtz + Fsp in typical melt inclusions from granitic minerals), hydrosilicate inclusions show significant variations according to the preceding pegmatite mineralization: enriched in rare elements, e.g. Be, Ta, Nb, Cs, if the neighboring miarolitic assemblage is depleted in corresponding minerals (I type), strongly depleted in those elements when the near miarolitic assemblage contain great amounts of Be, Li, Cs and Ta-Nb minerals (II type). Hydrosilicate inclusions are associated with boron-alkali-rich (up to 12-24 wt% of H₃BO₃ and up to 4 wt% of total alkali content) aqueous inclusions (Fig.3).

Previously many experimental studies revealed that hydrous silicate liquids typically appear within P-T range of magmatic-hydrothermal transition due to reactions between aqueous fluids, rich in alkali and/or F and quartz or other silicates (e.g. Wilkinson et al., 1996, Peretyazhko et al. 2004b and references therein). Their compositions and some physical properties suggest that they are silica hydrogels rather than common silicate melts. Our experimental data (Smirnov et al., 2005) suggest that such pegmatitic hydrosilicate liquids could be exsolved from magmatic boron-alkali-rich aqueous fluids. Our experimental data have shown that hydrosilicate liquid formation in the system at Na₂O-B2O3-SiO2-H2O depends mainly from Na-content (Na₂O>3 wt%) and much less from concentration of B or other weak acids. H₃BO₃ (10-20 wt%) for example does not exert influence on the formation of silica gel. Similar hydrosilicate liquids were documented by Peretyazhko et al., (2005) in the system NaF-B2O3-SiO₂-H₂O and Kotelnikova and Kotelnikov (2004) in the system NaF-SiO₂-H₂O. Regarding the close similarity in appearance of common silicate melt inclusions and inclusions of the hydrosilicate liquids the term "melt-like gel" was proposed by Peretyazhko and co-authors (Peretyazhko et al. 2004b) for pegmatitic hydrosilicate liquids.

In conclusion, hydrosilicate gels may co-exist immiscibly with the latest portions of silicate melts and aqueous fluids at the end of pegmatite magma crystallization. We believe that cryptocrystalline aggregate (clay \pm mica \pm zeolite) that frequently fills the space between miarolitic crystals is a result of transformations of the latest portions of pegmatitic hydrosilicate gels. Our experimental data show that hydrosilicate liquids possess strong capability to accumulate B, F and some rare elements (e.g. Ta, Mo) (paper in prep.). Importantly they are also able to exchange some of them with low-temperature diluted hydrothermal fluids. Therefore, these gels act as an additional agent of element fractionation during pegmatite formation, and thus they play an important role in the nature and the sequence of crystallization of different pegmatite minerals.

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FIGURE 1. Recrystallized melt (a) and aqueous (b) inclusion in quartz from graphic pegmatite. Melt inclusions homogenize at 600°C, fluid inclusions contain aqueous solution and CO₂-bubble. Oktyabrskaya pegmatite, Transbaikalia.



FIGURE 2. Hydrosilicate inclusions (ex-silica gel) in miarolitic quartz from Oktyabrskaya pegmatite (a) and Savvateevo pegmatite (b), Transbaikalia. Inclusions contain aggregate of silicates (primarily mica), co-trapped topaz, fluid with daughter sassolite and low-density CO₂-bubble. They re-melt completely at 650°C and 3 kbar confining pressure.



FIGURE 3. Crystal rich fluid inclusions that accompany hydrosilicate inclusions in the miaroloitic quartz form Octyabrskaya pegmatite (a) and Savvateevo pegmatite (b), Transbaikalia. These inclusions typically show high ice melting temperature (>-8°C) and sassolite dissolution temperature up to 95°C that indicate up to \sim 24 wt% of H₃BO₃ in homogeneous fluid. Other crystalline phases are either sassolite or unidentified soluble phases that may persist up to 120°C.

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