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FORMATION OF GRANITE PEGMATITES IN THE LIGHT OF MELT AND FLUID INCLUSION STUDIES AND NEW AND OLD EXPERIMENTAL WORK

The origin of pegmatites, especially those which are complexly zoned, is very controversial. One group of researchers (e.g., Fersmann, 1931; Turner and Verhoogen, 1960) considers them to be of igneous origin, while another one believes that pegmatites are metamorphic or metasomatic (e.g., Barth, 1962; Ramberg, 1952; Gresens, 1967). In this paper we present evidence in favour of the igneous origin of pegmatites. This does not mean, however, that we completely rule out the second possibility. The model, presented in this paper, is based on experimental and inclusion studies, assumes that the vast majority of the granite pegmatites are primary of magmatic origin, but often affected by metasomatism.

According to Fersmann (1931) granite pegmatites can be defined as crystallization products of residual solutions of the granites, which are separated during the solidification of the granitic magma. They are characterized by an arranged sequence of mineral associations, by a remarkable size of the individual crystals, by a simultaneous crystallization of different mineral phases, by an enrichment of highly volatile constituent and disperse elements. Fersmann regards the pegmatite-forming process as part of a continuous physico-chemical process during cooling a granitic magma, starting at magmatic down to hydrothermal temperature and pressure conditions. This definition can mutatis mutandis be applied to pegmatites of other magmas.

This definition, however, suggests a simple, directional evolution from high to low temperatures, from magmatic, via pneumatolytic to hydrothermal stages. From this we, however, do not learn how the crystallization processes actually take place. Roedder (1984) wrote "During crystallization of various magmas rich in silica …, the most abundant solid phases that form initially are anhydrous quartz and alkali feldspars. Separation of these from melt enriches the residual homogeneous melt in all constituents that do not enter these particular crystal structures." Water is the most important constituent to become concentrated in the melt by crystal fractionation. Water has a tremendous effect on minerals and rocks lowering their liquidus and solidus temperatures. The presence of water decreases the viscosity of

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aluminosilicate melts. Water together with the other volatiles (F, B, Cl) enhances the transfer of ions through the melt to the growing crystal interfaces. The presence of large crystals in pegmatites is an excellent example of an unusual crystal growth.

Already Niggli (1920) has emphasized the outstanding significance of the water for the formation of the pegmatites. However, because the solubility of water in an aluminosilicate melt depends strongly from temperature and pressure and because an important number of granite pegmatites belong to the group of shallow pegmatites, formed at depths of about 3-5 km (see Kozlowski, 1978), the enrichment of water is limited. In the system Qz-Ab-Or-H_2O at 2 kbar and 680°C the water content at the eutectic point is about 6.4 wt%. The viscosity of such melt is than $10^{5.5}$ Pa.s (Holtz et al., 2001). At such high viscosities the formation of the typically coarse-grained or giant-textured pegmatites are heavily conceivable. London (1999) tried to solve the viscosity problem with the assumption of a boundary layer rich in excluded fluxing components, and Beus (1983) suggested a model for the giant growth of crystals embedded in a hard rock based on a metasomatic process. However, with the both last models it is not possible to explain all observations, for example connected with the formation of large chamber pegmatites and giant crystals.

Based on extensive field and laboratory studies Jahns and Burnham (1969) emphasized the role of water as the dominant constituent of a separate fluid phase that is in the supercritical state. They underlined the closed- or restricted-system crystallization in the presence successively of hydrous silicate melt, of melt and coexisting supercritical aqueous fluid, and finally of aqueous fluid alone. However, according to Bureau and Keppler (1999) a haplogranite forms a supercritical fluid at about 16.9 kbar and 825°C. Such P-T values have no significance for natural processes in the upper crust. In a study to the effect of fluorine, boron and excess sodium on the critical curve in the albite-H_2O system then Sowerby and Keppler (2002) could show, that in a complex pegmatitic system complete miscibility between melt and fluid may be important in the final stages of crystallisation even at lower temperatures and pressures. For their pegmatite system (Peg. 1) they have derived the following minimum conditions: 595°C and 5 kbar. The study demonstrated, that in principle complete miscibility between fluid and melt at moderate P-T conditions are possible in the crust. From melt inclusion studies, Thomas et al. (2000) have shown, that in the case of the Ehrenfriedersdorf granite-pegmatite complex, complete miscibility is possible even at considerably lower pressures. The cause for this peculiar behaviour can be seen in the complex interplay of the volatiles H_2O, F, and Cl, the semivolatiles B_2O_3 and P_2O_5, and fluxing components such as Li_2O, Rb_2O, and Cs_2O along with the SiO_2 and Al_2O_3 in the melt. The most intriguing peculiarity of this system is the existence of a solvus with two coexisting pegmatite-forming melts at pressures $\leq$ 100 MPa.

Complete miscibility was attained at 712°C and 21.5 wt% H_2O. Further studies have shown now, that this model represents a strong simplification, because in the reality three phases coexist at or near the critical point: two melts and a H_2O-rich fluid phase (Thomas et al., submitted). By pressure fluctuations result further complications: boiling and formation of a low-dense vapour phase, which then is in a "quasi"-equilibrium with the other phases. Such short-time fluctuations can pretend complicated, multistage mineral-forming processes (see for example Kamenetsky et al., 2002).

Recently the coexistence of aluminosilicate melt, with hydrosaline melt and lower-salinity aqueous fluid in a complex, multicomponent system over a broad range of P-T conditions was proved experimentally by Veksler et al. (2002) and Veksler and Thomas (2002). Earlier the importance of critical phenomena and immiscibility between two melts were shown by Valyashko and Kravchuk (1978) using the simple system SiO_2-Na_2O-H_2O. Now, the idea of three coexisting immiscible fluid phases is an important addition to the classical concepts of pegmatite development, and it provides a crucial confirmation of fluid and melt inclusion studies. The separation of three different phases is not only responsible for the element distribution but in particular also responsibly for the crystallization behaviour. Because the wetting and solvent behaviour of the different melts are surely different, immiscible melts are not necessarily trapped in the same proportions, as the free parent phases were present during crystal growth. Different melt phases can hamper or also accelerate the crystal growth. The experimentally produced graphic textures in feldspar is a very nice example. During crystallisation of potassium feldspar in the haplogranite-Li_2O-B_2O_3-H_2O system the originally homogeneous melt splits into two parts: a water-rich and B- and Li-poor

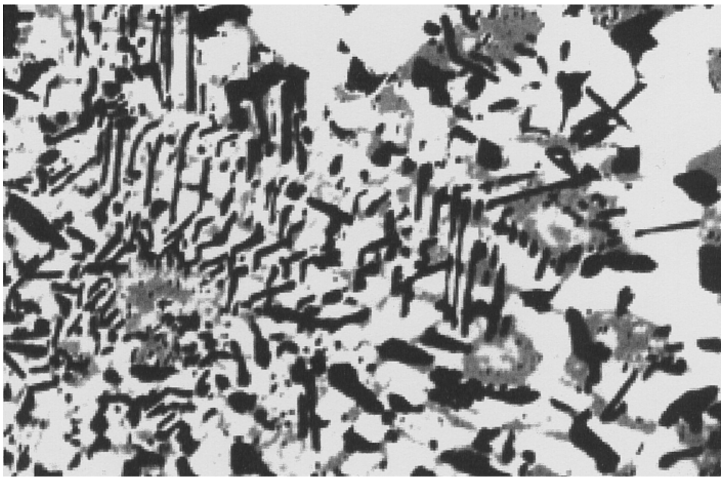

Fig. 1. The haplogranite-Li_2O-B_2O_3-H_2O system: formation of a graphic texture during crystallization at 500°C and 2 kbar (white: K-feldspar, grey: quartz, black: Li-tetraborate); image width: 250 µm.

aluminosilicate melt from which the feldspar obtains its components and droplets of a water-poor, B- and Li-rich melt, which being captured by the growing feldspar surface (Fig. 1).

According to our studies on natural melt and fluid inclusions and to our experimental work liquid immiscibility and the supercritical stage are of crucial meaning in the melt-dominated stage for the formation of pegmatites. Liquid immiscibility is not a singular event. These processes takes place at all times as long as there are melts. At the end of the melt-dominated stage maybe in small subsystem compartments separated by large crystals of quartz, feldspar and others.

In the following hydrothermal stage the primary melt signatures are very often blurred or wiped out completely. The lack of melt inclusions often led to misinterpretations of the pegmatite genesis (e.g. London, 1986). In large pegmatite bodies (Tanco, Mozambique) the relationship between different inclusion types often is not recognisable. This is also true for the large chamber pegmatites of Volyn, where Lemmlein et al. (1962) for the first time have studied silicate melt inclusions in topaz crystals. The often contradictory interpretations of the Volyn pegmatite genesis can be explained simple by the apparent incoherence of the different inclusion types. This connection is very seldom observable. In the relatively small pegmatite bodies of Ehrenfriedersdorf this relationship has been kept. From our observations on natural melt and fluid inclusions and our new experimental work (Veksler et al., 2000; Veksler and Thomas, in press; and ongoing work) the Jahns-Burnham model (1969) completed by the liquid-liquid immicible concept can explain more or less all steps of the genesis of granite pegmatites

Thus, studies of melt and fluid inclusions provide the critical evidences for liquid compositions and phase relations in nature. Our philosophy for further progress in this field is always to combine natural observations from inclusion research with experimental studies of key subsystems.

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