

The Sol-Gel Nature of Pegmatites

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In order to develop a viable model to describe the petrogenesis of pegmatites, several criteria must be satisfied. The foremost is that pegmatitic liquids must start with extremely low viscosities during emplacement, and then be able to support the growth of large and sometimes dense crystals in both viscosity and diffusivity. That is, in order to buoy crystals during growth, viscosities of the parent media must be functionally infinite ($\geq 10^{14}$ Pa s or 10^{15} poise) such that there are no preferred orientations or evidence of crystal settling. Moreover, diffusion rates and reservoir sizes must be sufficient to support growing crystals that have exponentially increasing surface areas and concomitant demands for components. Furthermore, crystal growth must at times be rapid, as some pegmatitic liquids must cool (days to 10^2 years; *e.g.*, Webber *et al.*, 1999) to equilibrate with their geotherms. Clearly, models based solely on direct crystallization from melts face the obstacle stated by Einstein (1905) that diffusivity decreases with increasing viscosity for these types of liquids, and such attenuated diffusion rates would preclude the formation of giant crystals in the given time frames. In the case of pegmatites with granitic compositions, specific criteria are required.

Massive quartz is the last primary “magmatic” phase to crystallize in both simple and complex granitic pegmatites, generally as cores or more centrally located zones. These zones of silica are composed of individual crystals or groups to >10 m and can occupy thousands of m^3 . Moreover, massive quartz is always anhedral with respect to all other primary phases, attesting to its crystallization during the very latest “magmatic” stage of pegmatite development. Indeed, studies of the structural state of massive quartz during crystallization indicate that silica crystallizes below the high-low inversion temperature of ~ 600 °C at 2 kbar, a temperature that depends on pressure and simple, coupled, and stuffed substitutions (*esp.* Al) into its structure that depress this temperature. However, quartz melts to a hydrous liquid in excess of 1000 °C in the silica-rich portion of the SiO_2 - H_2O system at 2 kbar, nearly twice its inversion temperature. Also, recent investigations indicate that massive quartz cores are homogeneous to within 1 to 2 ppm, *i.e.* no internal zonation whatsoever (R.B. Larsen, pers. comm., 2002; Götze *et al.*, 2005), unlike coexisting feldspars and other phases, and quartz of known magmatic origin. Hence, the formation of massive quartz-dominant zones from magmatic fractional crystallization is highly tenuous, especially in primitive pegmatites that lack an enrichment in fluxing components such as B, F and P that increase the solubility of H_2O in melts, lower their viscosities, and depress both liquidus and solidus temperatures. Just as perplexing, zones of massive quartz show none of the signs of precipitation from an exsolved silica-bearing aqueous solution under pneumatolytic (supercritical) to hydrothermal (subcritical) conditions, such as cryptocrystallinity and/or banding and zoning, nor extensive host rock alteration (hydration) due to the high water/rock ratio required by silica’s low solubility in aqueous fluid. Rather, segregations of silica generally occur as very coarsely grained monolithic zones within granitic pegmatites that possess only very highly attenuated to local or minor alteration haloes.

Euhedral crystals of feldspar, tourmaline, etc., grew into massive quartz zones similar to coral reefs into seawater, rather than competed with quartz for space. Also, replacement phenomena (*e.g.*, albitization, lepidolitization) are intimately associated with massive quartz and demonstrate that what eventually became quartz acted as the diffusive medium. Moreover, euhedral crystals commonly show mechanical deformation whereby they were bent or broken off of their matrix and displaced while enclosed within a single quartz crystal. Quartz deformation studies do not show evidence of plastic flow without dissolution-reprecipitation. But the complete dissolution and recrystallization of quartz on such a grand scale should also chemically affect feldspars, etc., which is not observed.

One model to describe the formation of pegmatitic textures is based on fluxed melt boundary layers that zone refine hydrous glass during its devitrification from sidewalls inward through liquidus undercooling (~ 75 - 150 °C), and experiments do produce granophyric and micrographic textures (London, 1999). However, undercooling is highly improbable for pegmatites hosted within their parents or pegmatitic leucosomes in migmatite terranes. Also, zone refining produces flat or increasing ferromagnesian trends from margin to core, while decreasing trends are the norm and suggest diffusive access to the whole “melt.” Furthermore, experimental devitrification temperatures (~ 450 - 550 °C) are much higher than pegmatite geothermometers are indicating (~ 425 - 250 °C; *e.g.*, Morgan & London, 1999; Sirbescu & Nabelek, 2003, Novák & Taylor, this volume), and the requisite fluxing components are not evident in most pegmatites. Finally, the experimental

microscopic textures have not been proven scalable to pegmatitic proportions. Therefore, this model is not valid to describe the internal evolution of pegmatites.

If the parent media is not highly viscous melt or aqueous fluid, then let us consider a 3-dimensional interpenetration of the two phases; in other words, a gel. Gels can form either from colloidal particles (~1-1000 nm) in solutions (sols) necking or polymerizing, or directly from viscous liquids by exsolving a fluid phase. Gels have functionally infinite viscosity but with finite shear strength; therefore, they can flow, fracture and reheel. And they are the perfect growth media for crystal nucleation and diffusion from within their entire volumes. Moreover, there is no Brownian motion or convection in gels, only the migration of components within their internal pore fluids and sponge-like frameworks directly to growing crystals. Pegmatitic textures (≥ 1 cm) of essentially every type can be routinely grown in seconds to months without undercooling in sol-gel systems; furthermore, increasing reservoir sizes can grow larger crystals. Oscillatory zoning in crystals and periodic precipitation (Liesegang phenomena) typically develop in gels, features that are commonly observed in pegmatites. Also in gels, the growth of late crystals sometimes pushes aside early ones rather than including them. Gels can transition back to sols through chemical or mechanical dispersion and be remobilized, or progress onward to solids, amorphous and crystalline. In essence, gels are self-organizing, disequilibrium systems.

A comprehensive view into the basics of pegmatology, the study of magmatically derived sols and gels (Taylor *et al.*, 2002, 2003, 2004), already exists by simply applying over a century of chemical engineering and materials research of sol-gel systems to solve the petrogenesis of pegmatites and other types of ore deposits. Alumina to silica sols, gels, and their products are extremely well characterized, including those with very high densities and adsorbed/polymerized metals. Many rare-metal deposits that do not contain anions or anionic complexes aiding metal solubility in melt or aqueous fluid, or any evidence of large-scale fluid-host rock interaction can now quite easily be described as a function of transport from high-density, metal-bearing sols and deposition as crystals from gels. Ultimately these gels order to crystalline solids, especially framework silicates. Taylor *et al.* (2002) proposed the term “tektogel” to describe such solids.

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