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THE APPLICATION OF EXPERIMENTAL PETROLOGY TO THE GENESIS AND CRYSTALLIZATION OF GRANITIC PEGMATITES

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ABSTRACT

Experiments in the hydrous granite system and related systems (natural and synthetic) have provided useful information on the liquidus phase relations of granitic and pegmatite-forming magmas at equilibrium. Because pegmatite-forming magmas usually intrude cooler host rocks, however, the actual conditions of emplacement and initial solidification do not necessarily correspond to those of the equilibrium liquidus field. Experimental studies aimed at establishing equilibrium phase relations also yield little information on the processes of crystallization that produce the coarse-grained and heterogeneous, anisotropic fabrics that distinguish pegmatites from granites. The results of a relatively few studies of silicate crystal nucleation and growth are applicable to pegmatite systems, which are best described as a product of disequilibrium fractional crystallization through liquidus undercooling. The degree of liquidus undercooling, and the concentrations of quartz-feldspar-incompatible components in the melt (particularly H_2O, B, P, and F), govern the textural development of granitic magmas by controlling the rate and number of stable crystal nuclei formed.

Keywords: experimental petrology, igneous, granite, pegmatite.

SOMMAIRE

Les expériences portant sur le système haplogranitique en présence de l'eau et sur les compositions semblables, soit naturelles ou synthétiques, ont fourni de l'information très utile à propos des relations sur le liquidus des magmas granitiques et pegmatitiques à l'équilibre. Comme les magmas qui cristallisent sous forme de massifs pegmatitiques sont mis en place généralement dans un encaissant plus froid, les conditions de mise en place et de solidification initiale ne correspondent pas nécessairement à celles du liquidus à l'équilibre. Aussi, les expériences visant l'équilibre parmi les phases ne sont-elles pas pertinentes aux processus de cristallisation donnant les textures grossières, hétérogènes et anisotropes qui distinguent les pegmatites des granites. Seuls les résultats d'une suite relativement restreinte d'expériences de nucléation et de croissance cristalline seraient applicables aux systèmes pegmatitiques, qui semblent le produit de cristallisation fractionnée sous conditions de déséquilibre par refroidissement sous le liquidus. Le degré de ce refroidissement et la concentration des composants incompatibles avec quartz et feldspath (surtout H_2O, B, P et F) dans le bain fondu déterminent le développement textural des produits de cristallisation par contrôle du taux de formation et du nombre de centres de cristallisation stables.

(Traduit par la Rédaction)

Mots-clés: pétrologie expérimentale, roches ignées, granite, pegmatite.

INTRODUCTION

Petrologists rely heavily on the results of experimental studies to interpret and understand natural igneous rocks and the phenomena that produce them. For most igneous petrologists, experimentation in liquid silicate systems has largely defined what magmas can and cannot do; thus the impact of experimental petrology on shaping current concepts of igneous processes cannot be overstated. In no subfield of igneous petrology has experimentation had a more important effect than in the study of granitic pegmatites. Early experimental investigations of pegmatites were logically part of the broader problem of the origin of granites, and the properties of silicic melts in general. Pegmatites were a likely target of experimental investigation, because the distinctive and complex features of pegmatites defy any simple or intuitive explanation. In reference to the pioneering experimental studies by R.H. Jahns and C.W. Burnham, Wyllie (1963) noted that "This programme, which soon achieved spectacular success, is a fine example of how natural rock samples may yield useful experimental data despite the fact that the system is too complex to be treated theoretically in terms of the phase rule."

Experimental studies of pegmatites are complicated by the large number of chemical components that are known to comprise some of these rocks. The vast majority of pegmatites, however, are compositionally simple granites (ranging from mildly alkaline to strongly peraluminous), and lack evidence for high concentrations of normally rare elements [*e.g.*, large-ion lithophile elements (LILE), high field-strength elements (HSFE), and fluxing components such as B, P, and F]. In addition, many of the intrinsic features of pegmatites are shared by the compositionally simplest to the most evolved types. This fact implies a common process of pegmatite formation that does not hinge on unusual components (*i.e.*, significant deviations from the haplogranitic system). Any experimentally based model for pegmatite formation, therefore, must account for the features of pegmatites essentially within the simple haplogranite system.

What are the characteristics of pegmatites that distinguish them from granites, and which must be explained by any (experimentally derived) model? The salient features include:

(1) *Extremely coarse grain-size*. Individual crystals greater than several centimeters are typical, but giant crystals over a meter in dimension are common in larger pegmatites.

(2) *Extremely variable grain-size*. The grain size of constituent major minerals (especially quartz and the feldspars) usually increases by several orders of magnitude from margin to center in zoned pegmatites. In addition, some pegmatite units display a porphyritic texture, in which comparatively fine-grained assemblages (including those with an aplitic texture) are interstitial to giant crystals.

(3) *Anisotropic fabrics*. Together with variations in grain size and mineral zonation, pegmatites commonly possess the following highly anisotropic fabrics: (A) a comb structure, in which the long axis of nonequidimensional megacrystic minerals (usually microcline, tourmaline, beryl, lithium aluminosilicates, and phosphates) are perpendicular to pegmatite contacts, and individual crystals flare (expand) toward the center of dikes; (B) a layered structure, usually fine-grained albitic rocks with flat or convolute layering subparallel to pegmatite contacts or internal zonal boundaries (with foliation and lineation of nonequidimensional minerals parallel to the layering), and (C) graphic, radial, and skeletal crystal habits. Graphic intergrowths of quartz in microcline, quartz in tourmaline, and phosphates in albitic units are common. Crystals of microcline, phosphates, oxide minerals, and tourmaline exhibit branching or skeletal crystal forms; branched skeletal crystals expand toward the center of pegmatites units (away from substrates). Micas and coarse-grained albite ("cleavelandite") commonly form spherically radial aggregates that appear to have nucleated on a substrate seed crystal at the center of the aggregate, or along the margins of megacrystic phases such as microcline, tourmaline, or lithium aluminosilicates.

(4) *Chemical heterogeneity at several different scales*. This includes zonation within individual crystals (*e.g.*, of feldspars, micas, beryl, tourmaline), and variations in the composition of the same phase in different textural–paragenetic assemblages within a pegmatite. The most striking manifestation of chemical heterogeneity is the characteristically sharp, megascopic zonation of mineral assemblages. Even in compositionally simple granitic pegmatites, the major minerals quartz, plagioclase, and microcline are commonly segregated in nearly monomineralic zones.

(5) *Chemical fractionation within individual pegmatites and among pegmatites of a group or field*. Many lithophile trace elements with significantly different chemical properties (large variations in ionic charge, ionic radius, electronic field-strength, and coordination number) are concentrated together by pegmatite-forming processes, such that these elements are enriched by a factor of 10^3 to 10^4 over their average abundances in typical granites. In the common class of peraluminous pegmatites, for example, there is a positive correlation of Li, Rb, Cs, Be, Ga, B, P, F, Mn, Nb, Ta, Zr, Hf, Sn, and U with increasing

differentiation of pegmatite (*e.g.*, Černý *et al.* 1985). In addition, some pairs of elements that exhibit chemical coherency in other geological environments (*e.g.*, Zr–Hf, Nb–Ta) may be completely fractionated by pegmatite-forming processes. The chemical fractionation is manifested on a regional scale, wherein the distal pegmatites at the margins of a cogenetic group are the most fractionated, and within individual pegmatites, in which chemical fractionation and trace-element enrichment can reach extreme values through internal differentiation.

The primary purpose of this paper is to summarize and evaluate how petrologists have used experimental petrology (in combination with other types of data) to understand pegmatite-forming processes. As a result, it will become evident that very few experimental studies have actually addressed the five distinguishing characteristics of pegmatites noted above.

Initially, I present a brief summary and discussion of the various experimental methods that are employed by petrologists in silicate crystal – melt – vapor systems. The way in which experiments are done has profound effects on the experimental results and the interpretations made from them.

A REVIEW OF EXPERIMENTAL PROCEDURES

Few mineralogists or petrologists appreciate the complexity and subjectivity that are inherent in experiments with silicate–volatile systems. This is partly because there are comparatively few experimental petrologists, and most have not adequately conveyed to their audiences what they have done and observed. Experimental results are usually reduced to depiction as phase diagrams or tables in final publications. Large uncertainties in the location of reaction boundaries, especially in isotherms and cotectics of liquidus diagrams, are commonly lost in the presentations. Most importantly, the textures and compositions of run products (especially with respect to the homogeneity of individual phases) have not been regularly or thoroughly described. The now-routine use of electron-beam microanalysis and back-scattered electron (BSE) scanning microscopy allows experimentalists to depict important compositional and textural details of experiments. Consider, however, that most of the experimental work on which granite–pegmatite petrology is based was conducted before the advent of electron-beam microanalysis or imaging, and some experimentalists still do not integrate visual (*e.g.*, BSE) images of their experimental products into final publications. The following review of general experimental procedures highlights some of the inherent difficulties and limitations of experimental design and interpretation that are especially pertinent to granite–pegmatite studies.

Approach to experimental run conditions

Any experiment in the field of silicate liquid + crystals ± vapor approaches this final condition from a metastable state (*i.e.*, with a metastable phase-assemblage). Figure 1 depicts the five commonly used approaches to liquid–crystal (-vapor) experiments. Each different approach imposes a specific mechanistic process on the resultant experiment, especially in terms of the kinetics of crystal growth or dissolution. Normally, changes in temperature from the start of an experiment to its final state and quench are accomplished isobarically. A decrease in pressure that accompanies quenching in some experimental devices, however, may promote vesiculation and a loss of volatile components from the resultant glass.

Paths D and E approach a final state of the experiment from below the solidus of the system, and are termed forward-direction experiments (they are prograde in temperature: Bowen 1913, 1914). For path D, starting materials may be crystalline or vitreous; path E is employed to devitrify starting glasses. The increase in temperature to the final state of the experiment is usually instantaneous with respect to run duration. Upon entering the stable field of crystals + melt, most starting materials produced by path E normally consist of very fine-grained (2–10 μm) crystalline phases; crystalline phases produced directly from glasses along path D tend to be slightly coarser grained. Forward-direction experiments, however, normally enter the final P–T field of an experiment with a very large number of existing crystal nuclei (path E), or a large number of favorable sites of nucleation inherited from starting glasses. Strictly speaking, forward-direction experiments with crystalline starting materials (*e.g.*, the famous Spruce Pine and Harding pegmatite rock composites) replicate the geological process of partial melting in solid rock. The single advantage of forward-direction experiments is that they may contain crystal nuclei upon attainment of run temperature. This eliminates the kinetic and energetic barriers to crystal nucleation that are prevalent in silicate systems (discussed further below). There are, however, a number of disadvantages. The lower reactivity of crystalline silicate phases (as opposed to glasses) may mean that reactions take longer to come to a final steady-state. Crystalline and especially devitrified materials may contain phases that are metastable at the final conditions of the run, but which persist metastably because of slow reaction-kinetics (or for crystalline solid-solutions,

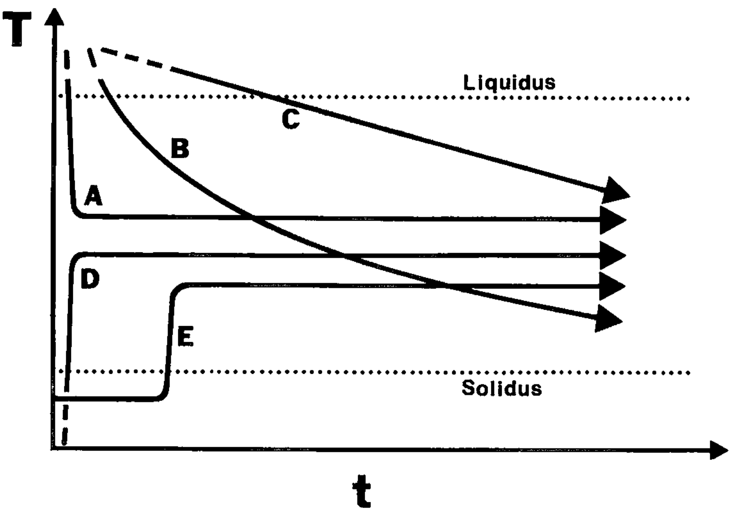


FIG. 1. Schematic illustration of various isobaric temperature (T) – time (t) approaches to run conditions, including: preconditioned at superliquidus T, followed by single-step cooling to run conditions (reverse-direction, path A); preconditioned at superliquidus T, followed by programmed cooling (reverse-direction, paths B and C); single-step approach to run T from cold start with glass or crystalline materials (forward-direction, path D); and subsolidus devitrification of glass, followed by single-step approach to run T (forward-direction, path E). From London *et al.* (1989).

initial compositions that fail to equilibrate with melt over the run duration). The final recognition and characterization of run products, and of potentially metastable phases, are complicated by two important factors. The crystalline products are normally very fine-grained (< 10 μm), and commonly too small for accurate optical identification and even electron-beam microanalysis (*e.g.*, at an accelerating voltage of 15 kV, electron-beam penetration of typical alkali aluminosilicates such as feldspars is greater than the thickness of the grains being analyzed). Because the reactions involve only melting, relict crystalline grains are typically rounded and embayed, rather than euhedral. Thus it is difficult to determine unequivocally if melting has gone to completion, and hence if all relict crystals and the proportion of crystals to quenched melt represent an equilibrium assemblage. Most importantly, forward experiments do not replicate the crystallization of silicate liquids.

In Figure 1, paths A–C involve the crystallization of silicate liquid from a crystal-free, superliquidus state. Starting materials (glasses or crystalline assemblages) are first taken above the liquidus of the system (termed preconditioning) for complete melting, followed by an isobaric decrease in temperature to final run conditions. Such experiments are reverse-direction (Bowen 1913), as they are retrograde in temperature. The primary disadvantage of reverse experiments is that like most liquids, silicate melts must be cooled to some temperature below the stable liquidus (termed undercooling) to initiate nucleation and growth of crystals. Apparent temperatures of the liquidus obtained from reverse experiments will always be lower than the values at equilibrium. In reverse experiments, the magnitude of the preconditioning step above the liquidus, the magnitude of the undercooling step below the liquidus, and the rate of undercooling all combine to dictate the final texture, morphology, grain size, numbers of crystals, and even crystalline assemblage of the final product. With increasingly large preconditioning steps above the liquidus, larger undercoolings and longer run-times are generally required for nucleation of crystals. Because of kinetic barriers to crystal nucleation at lower temperatures, large degrees of undercooling may entirely inhibit the formation of stable crystalline phases, or metastable phases may nucleate rather than stable ones.

It is apparent also that the experimentalist may control the rate of cooling of reverse experiments, with consequent effects on the nucleation and growth rates of crystalline phases. Single-step (A), nonlinear (B), and linear (C) cooling ramps (termed programmed cooling) may be used, and the choice is not arbitrary. Here another problem arises, because the experimental rates of cooling are normally far greater than those of natural systems.

One distinct advantage of reverse experiments is that the grain size of crystalline silicates is normally much larger (in granitic systems, commonly > 50–100 μm) than in forward experiments, which greatly facilitates identification and characterization of the phases. In addition, silicate and volatile components (*e.g.*, added water) may be more completely homogenized by preconditioning, thus eliminating inhomogeneity in starting materials and any stable or metastable crystalline phases that may be present or produced on run-up. Also, the recognition of crystalline phases that have actually grown from melt (in contrast with the partially resorbed phases in forward experiments with crystalline starting materials) is unambiguous.

Many experimentalists do not indicate whether forward or reverse experiments, or some combination of both, have been employed in their studies. From my own conversations with experimentalists studying granitic systems, most have used dominantly or exclusively forward-direction methods, because the objectives have been the accurate determination of equilibrium liquidus and solidus phase-boundaries.

From the earliest stages of experimental petrology, Bowen (1913, 1914), who established the nomenclature of forward- and reverse-direction experiments, recognized that silicate systems respond quite differently when brought to the liquidus surface by different means. Bowen (1914) noted that even melts as basic as orthopyroxene in composition do not yield equilibrium products when crystallized by reverse-direction methods. This observation formed the basis of Bowen's most famous concept, the process of fractional crystallization. Bowen (1914) elaborated on the fact that only forward-direction experiments, where starting materials were devitrified or recrystallized just below the solidus of the system, could be used to obtain diagrams showing the equilibrium liquidus in silicate systems. Thus, most experimental petrologists have shunned reverse-direction methods because of the nonequilibrium behavior in such experiments, especially with siliceous bulk compositions (*e.g.*, Tuttle & Bowen 1958). This is an important point to remember, as the pivotal experiments that shaped Jahns's concept of pegmatite formation [cited in Wyllie (1963) and Jahns (1982)] were performed by reverse-direction methods. Though liquidus diagrams are derived from partial melting experiments, geologists have applied these to cooling of silicate melt on the assumption that natural plutonic bodies cool much more slowly and achieve the equilibrium that short-term experiments do not. Whereas this may be true for most magmas in plutonic settings, pegmatite-forming magmas may be subjected to much more rapid cooling [*e.g.*, Chakoumakos & Lumpkin (1990), discussed further below], such that the cooling history (rate and magnitude of undercooling) of the natural magmas approaches that of the experiments. For this reason, reverse-direction experiments have applications to pegmatites that may not extend to other plutonic systems. Certainly, forward-direction experiments do not yield textural information pertinent to undercooled magmas. It is therefore impossible to understand pegmatites in terms of forward-direction experiments, because it is texture and fabric that distinguish pegmatites from granites (and rhyolites as well).

Experimental studies of nucleation and growth of silicate crystals from melt

Inasmuch as magmas solidify through changes in pressure, temperature, and composition, they represent dynamic rather than static systems. Crystal growth (or resorption) occurs only where and to the extent that systems deviate from equilibrium, *i.e.*, crystal growth or dissolution constitutes a response of a dynamic system toward equilibrium. The kinetics of crystal nucleation and growth, therefore, represent important time- and process-dependent factors that ultimately define the fabric of a body of crystalline igneous rock, including pegmatites. There are but few experimental investigations of rates of crystal growth in relevant hydrous haplogranite or alkali feldspar systems (notably Fenn 1977, 1986, Swanson 1977, Swanson & Fenn 1986, Petersen & Lofgren 1986; Muncill & Lasaga 1988). In addition, London *et al.* (1989) have presented some qualitative information on the kinetics of crystal growth in differentiated felsic melts.

Experiments such as these are conducted by reverse-direction single-step or programmed isobaric cooling to some temperature below the equilibrium liquidus for the crystalline phase(s) in question. After a characteristic time between undercooling and crystal nucleation (termed the nucleation delay or lag time), isothermal growth of crystals proceeds by fractional crystallization, such that the compositions of melt and crystals evolve toward the equilibrium values at the experimental temperature of undercooling. That is, the degree of undercooling of the system approaches zero with time, and consequently the rate of crystal growth decreases to zero when chemical equilibrium is attained between the rim of crystals and the residual melt. To this extent, experiments on kinetics of crystal growth generally fail to replicate the continuously changing state of natural silicate melts. An additional limitation of the experimental data is that the cooling rates are far too rapid to satisfactorily emulate natural plutonic systems. In most of the crystal-growth experiments cited above, nucleation densities are 10^2–10^4 higher than in typical granites, and in turn far greater than in pegmatites. There is an obvious problem in scaling densities of nucleation in experiments to those of rocks. Fractional crystallization and compositional changes in the melt during the course of an isothermal experiment also are inevitable consequences of crystallization in multicomponent systems containing crystalline solid-solutions (*e.g.*, feldspars) or non-eutectic compositions. Whereas these changes may be pertinent to natural systems, they introduce complexities in the interpretation of the factors that control crystal nucleation and growth.

Most data are reported in terms of nucleation density and growth rate as functions of the magnitude of the undercooling step below the liquidus of the bulk composition. The nucleation delay or lag time is not presented systematically in any of the published crystal-growth diagrams, and cannot be ascertained indirectly. This lag time between the initial step of undercooling and the first appearance of crystalline phases depends complexly on the bulk composition (*i.e.*, melt structure) of the system, the magnitude of the isobaric undercooling, and the duration of the experiment (*e.g.*, Dowty 1980, Kirkpatrick 1981, 1983). For alkali aluminosilicate systems, lag times for nucleation of minutes to thousands of hours have been reported. The lag time for nucleation contains potentially useful information on melt structure, cation diffusivity, and the probability of forming nuclei of critical size (*e.g.*, London *et al.* 1989). In most studies, growth-rate measurements apply only to crystalline phases that lie fully within melt (termed internal nucleation, which is not strictly equivalent to homogeneous nucleation: *e.g.*, Fenn 1977). Crystalline phases that nucleate (heterogeneously) on the surfaces at the melt-capsule interface are not considered, because the surface-energy contribution to the total free energy of substrate-catalyzed nuclei may be lower than that of crystals nucleated within silicate liquid (*e.g.*, Kirkpatrick 1981). The hosts to plutonic bodies of magma, however, do provide surfaces and potential nuclei that may facilitate incipient growth of crystals in melts. It will be argued below that heterogeneous (perhaps surface-catalyzed) nucleation is pervasive in the crystallization of pegmatites.

The meaning and experimental determination of vapor saturation

In seeking to explain what state or process makes pegmatites as opposed to texturally and compositionally homogeneous granites, Jahns & Burnham (1969) proposed that it is the presence of an aqueous vapor phase, together with silicate melt, that is essential to pegmatite formation. This concept has been widely accepted since its inception. Before reviewing the tenets of the Jahns-Burnham model, however, it is important to explain how experimentalists ascertain the presence of an aqueous vapor phase in systems containing silicate liquid, or more importantly, the point at which an experimental system is H_2O-saturated.

Experimental starting reagents (finely powdered rock or glass, and water) are loaded into precious-metal capsules in the presence of air; virtually all sealed experiments contain more air than can be dissolved into silicate melt at run conditions. Therefore, even anhydrous experiments are vapor-saturated, and the vapor phase is mostly nitrogen. This fact can invalidate one of the more commonly employed means of determining saturation in an aqueous vapor: the presence of vapor bubbles in quenched melt (glass) (Burnham & Jahns 1962). As the H_2O content of successive experiments is increased, the vapor phase (initially mostly nitrogen) contains an increasing mass fraction of H_2O, as controlled by the fugacity of H_2O in coexisting melt at the pressure and temperature of the experiment. In quenched experimental glasses, the filling density of vesicles (recognized as an increasing volumetric ratio of condensed water to vapor in vesicles observed at room temperature) will increase, as will the fraction of H_2O in the vapor space that exists outside of the melt bead. This fact negates another commonly used method of determining H_2O saturation of melt, wherein a capsule is punctured after the run, dried, and any subsequent weight loss after drying corresponds to the amount of free water in the capsule (presumed to be indicative of H_2O in excess of saturation in melt). It is apparent that weight loss will occur from capsules in which melt was not saturated in H_2O. If a silicate melt is oversaturated in H_2O, the composition and density of the vapor phase in capsules and vesicles trapped in glass do approach those of pure H_2O (disregarding the diminishing mass fraction of air and the fraction of solutes in the fluid phase) at the pressure and temperature of the run. Thus, the filling density of trapped vesicles increases up to H_2O saturation of melt, but remains essentially constant at any degree of oversaturation. Determination of the filling density of trapped inclusions may yield estimates of the point of H_2O saturation of melt within approximately ± 1 wt%

H_2O added (*e.g.*, London *et al.* 1988, 1989). Electron-beam microprobe analysis (EMPA) of quenched melt compositions also has been employed. With increasing H_2O up to saturation, EMPA totals (*e.g.*, the atomic or weight sum of all constituents) decrease without a change in the atomic proportions of analyzed elements; at and above H_2O saturation, analytical totals become nearly constant (assuming that the solubilities of melt components in vapor are negligible, or that melt dissolves congruently in vapor). Whereas this method may determine the point of H_2O saturation, the estimation of the H_2O content of melt by difference from EMPA totals can be inaccurate owing to the large uncertainties associated with the analysis of volatile components such as Na and loss of H_2O under vacuum and during analysis. For experimental systems containing elements whose vapor/melt partition coefficients deviate significantly from unity, changes in their abundances in quenched melts (based on relative increase or decrease depending on compatibility in melt *versus* vapor) above H_2O saturation have been used. London *et al.* (1988) employed all of these chemographic methods, including measurements of filling density and direct analysis of H by ion microprobe, to obtain accurate and consistent values for H_2O saturation in quenched rhyolitic melt. By these same methods, however, London *et al.* (1988) also determined that a significant mass fraction of H_2O that was dissolved in melt at run conditions exsolved on quench (approximately 17% of the total H_2O content of melt was lost). They and others have noted the difficulty of retaining H_2O in quenched experimental melts that are near or at H_2O saturation (*e.g.*, Stolper *et al.* 1983, Hamilton & Oxtoby 1986).

The experimental and analytical difficulties associated with determination of H_2O saturation of melt are important, as the experimental data form the basis of models for the dissolution of H_2O in melt. The problems are particularly germane to pegmatite studies, because the model of Jahns & Burnham (1969) attributes the transition from granitic to pegmatitic textures to the exsolution of an aqueous vapor phase from saturated melt.

EXPERIMENTS IN HYDROUS GRANITIC SYSTEMS

Experiments in haplogranitic and granodioritic systems form much of the basis of granite - pegmatite petrology. This extensive body of experimental work will not be reviewed fully here (*e.g.*, Tuttle & Bowen 1958, Luth *et al.* 1964, Burnham 1979, Whitney 1988, and references cited therein). Above all, experiments in simple granitic systems reveal the importance of H_2O in depressing liquidus and solidus temperatures (Fig. 2). Other significant effects include lowered viscosities of the melt (*e.g.*, Shaw 1965, 1972), changes in field boundaries on the liquidus and eutectic compositions (*e.g.*, Tuttle & Bowen 1958, Luth *et al.* 1964), and kinetics of crystallization (discussed below).

How H_2O promotes structural changes in polymerized, high-silica melts is still uncertain. A landmark study by Burnham & Davis (1971, 1974) led to a model wherein H_2O dissolves in melts as OH groups; the OH groups are formed partly at the expense of oxygen bridges in the TO_4 framework of the melt, which would account for the large decrease in melt viscosity with addition of H_2O (Burnham 1979). Subsequent spectroscopic investigations of quenched alkali aluminosilicate glasses have found little evidence for the Si–OH bonding predicted by the Burnham model (*e.g.*, Mysen & Virgo 1986a, b). Most of the H_2O in quenched, saturated glasses exists in molecular form (Stolper 1982); the portion of the H_2O dissolved as hydroxyl groups is associated with alkalis and Al (*e.g.*, Mysen & Virgo 1986a, b, McMillan & Holloway 1987). On the basis of a recent spectroscopic study, Kohn *et al.* (1989) proposed that H_2O does not interact with TO_4 framework components of melt, leaving unanswered the cause of reduction in melt viscosity that presumably involves breakage of oxygen bridges in the TO_4 framework. Perhaps the best observation to draw from this discussion is how much remains to be learned about one of the most important properties of silicate melts.

Through petrological and spectroscopic experiments, the solubility limits of H_2O in simple alkali aluminosilicate melts have been related to the variables of pressure, temperature, and bulk composition. With increasing H_2O pressures, minima in the haplogranite system migrate along a path toward the Ab–Or join (Luth 1976). The effects of an isobaric increase in the activity of H_2O in melt, however, are not yet resolved. Nekvasil & Burnham (1987) calculated that the isobaric minimum should migrate toward quartz as the activity of H_2O increases in melt. Experiments by Pichavant (1987) showed that an isobaric increase in the activity of H_2O in granitic melt induces a pronounced shift of melt compositions toward the Ab–Qtz join. An experimental study by Dingwell *et al.* (1984) demonstrated that the solubility of H_2O in alkali aluminosilicate melts increases with both peralkaline and peraluminous deviations from metaluminous haplogranitic compositions. This behavior was related to the decrease in the state of polymerization of the melt caused by excess alkalis or Al, and is consistent with the prevalent view that the solution of H_2O principally involves hydrolysis of alkali and aluminate species in the melt.

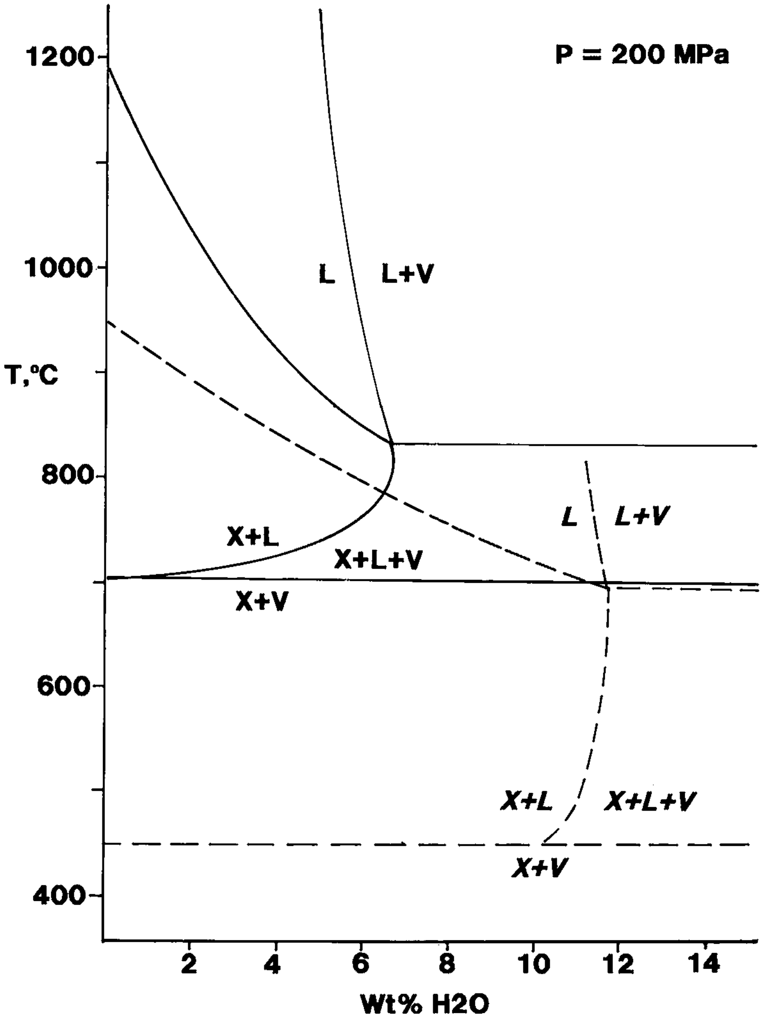


FIG. 2. Schematic depiction of experimental phase relations in two granitic systems at 200 MPa total pressure as functions of T *versus* wt% added H_2O (or H_2O content of melt). Solid lines and non-italicized labels are from Whitney (1975) for a simple synthetic granite composition (R1 of Whitney 1975); dashed lines and italicized labels are from London *et al.* (1989) for the Macusani rhyolite obsidian, which is a peraluminous natural glass with elevated Li, B, P, and F contents akin to peraluminous complex pegmatites. Note that the subliquidus vapor saturation curve for R1 represents the total water added to the system, whereas the comparable phase boundary for the Macusani glass portrays the water content of melt along the saturation surface. Phase labels are X (crystals), L (silicate melt), V (aqueous vapor).

Increasing solubility of H_2O in melts as caused by components outside of the haplogranite system is discussed below.

The role of H_2O in granitic melts is paramount in the model of pegmatite evolution proposed by Jahns & Burnham (1969; also Jahns 1982, Burnham & Nekvasil 1986). The depression in freezing point and decreased viscosities of the melt effected by H_2O helped explain the migration of pegmatite-forming melts into cooler rocks at distances of kilometers from their source plutons. Jahns & Burnham (1969; Jahns 1982) also equated reduced viscosities of the melt with depolymerization of pegmatite-forming melts, and postulated that such depolymerization would enhance rates of diffusion of melt components to the sites of crystallization. Increased rates of diffusion caused by H_2O also were believed to augment the formation of crystals of giant dimensions, and in large part to promote the mineralogical segregation that is characteristic of zoned pegmatites. The key concept of the Jahns-Burnham pegmatite model, however, lies not in the effects of H_2O in melt, but rather in the existence of an aqueous vapor phase that coexists with H_2O-saturated melt. This hypothesis of the Jahns-Burnham model has been so thoroughly ingrained in geological thought that most petrologists equate pegmatites with evidence of saturation in an aqueous phase in granitic systems. The validity of this hypothesis has been evaluated and tested experimentally by London (1986a, b, 1990a, London *et al.* 1988, 1989). The conclusions of this recent evaluation are summarized in following sections.

EFFECTS OF COMPONENTS OUTSIDE OF THE HAPLOGRANITE - H_2O SYSTEM

Much of the field and experimental study of pegmatites has focused on chemically evolved pegmatites whose compositions lie outside the haplogranite system. This is the case for the class of complex peraluminous pegmatites that tend to be enriched in Li, Be, B, P, and F. Some experimental investigations have utilized complex bulk-compositions, containing some combination of the components above (*e.g.*, Fig. 2), as analogs to the behavior of natural pegmatite-producing magmas (*e.g.*, Burnham & Nekvasil 1986, London *et al.* 1988, 1989). The individual effects of these five components on phase relations in granite systems, however, are summarized below. Lengthier discussions of the roles of B, P, and F are provided by London (1987).

Addition of lithium

Early reconnaissance experimental studies (Wyllie & Tuttle 1964) showed that the addition of up to 1 wt% Li_2O to H_2O-saturated melt (of the composition of Westerly granite) lowers the apparent eutectic temperature of this system by 90°C at 275 MPa. The addition of Li to the system Ab–Qtz at 200 MPa(H_2O) lowers the eutectic from 735°C on the Ab–Qtz binary to 640°C at the pseudoternary minimum of Ab–Qtz–Pet (petalite) (Stewart 1978). The studies by Stewart (1978) and Martin & Henderson (1984) demonstrate conclusively that the lithium aluminosilicates are stable with quartz and feldspars on the liquidus of pegmatite systems (*i.e.*, are magmatic in origin) at bulk Li_2O contents similar to those found in natural Li-enriched granitic pegmatites (Stewart 1978).

In experiments involving melt–vapor (London *et al.* 1988) and crystal–vapor equilibria (Sebastian & Lagache 1990, Lagache & Sebastian 1991), there is no measurable fractionation of Li from other alkali elements. In other words, the ratio of Li/[sum of alkalis] in vapor is equivalent to that of the melt or crystalline phases with which vapor coexists. This has important implications for the formation of zones consisting of lithium aluminosilicates + quartz (feldspar-absent), which Stewart (1978) and others have construed as precipitates from aqueous vapor. The recent experiments invalidate this hypothesis.

Addition of beryllium

There are no published studies of the effects of Be on liquidus relations in granitic systems, nor on the BeO content of granitic melts required to produce saturation in beryl at elevated melt or H_2O pressures. Experimental studies (*e.g.*, Cemic *et al.* 1986) and calculated equilibrium phase-relations (*e.g.*, Barton 1986) show a wide P–T field for the stability of beryl that overlaps the feasible conditions for crystallization of hydrous granites and pegmatites. Judging by the widespread occurrences of beryl in granites and pegmatites, reactions among beryllium and other aluminosilicates or their melt components (*e.g.*, Burt 1978, Barton 1986) probably buffer the Be content of the melt at low concentrations, such that Be should have a minor effect on liquidus relations.

Behavior and effects of boron

The common occurrence of accessory to abundant tourmaline in metaluminous to peraluminous granites and pegmatites attests to the elevated boron contents of these melts. As discussed below, however, the boron content of most granitic and pegmatite-forming melts is not conserved (as tourmaline or other borosilicates) within the crystalline bodies, and hence the actual magmatic concentration of boron is difficult to assess in these systems.

The ability of boron to lower melting temperatures and melt viscosities in silicate systems has long been utilized in glass technology. Chorlton & Martin (1978) provided the first experimental evidence that boron promotes significant freezing point depression of alkali aluminosilicate liquids at elevated H_2O pressures. From this initial study, subsequent experiments have elucidated many additional details of boron geochemistry (Benard *et al.* 1985, Pichavant 1987, London 1986a, 1987, London & Palmer 1990a, London *et al.* 1988, 1989, Morgan & London 1987a, b, 1989, Morgan *et al.* 1990, Palmer *et al.* 1992).

For the haplogranite system with boron,

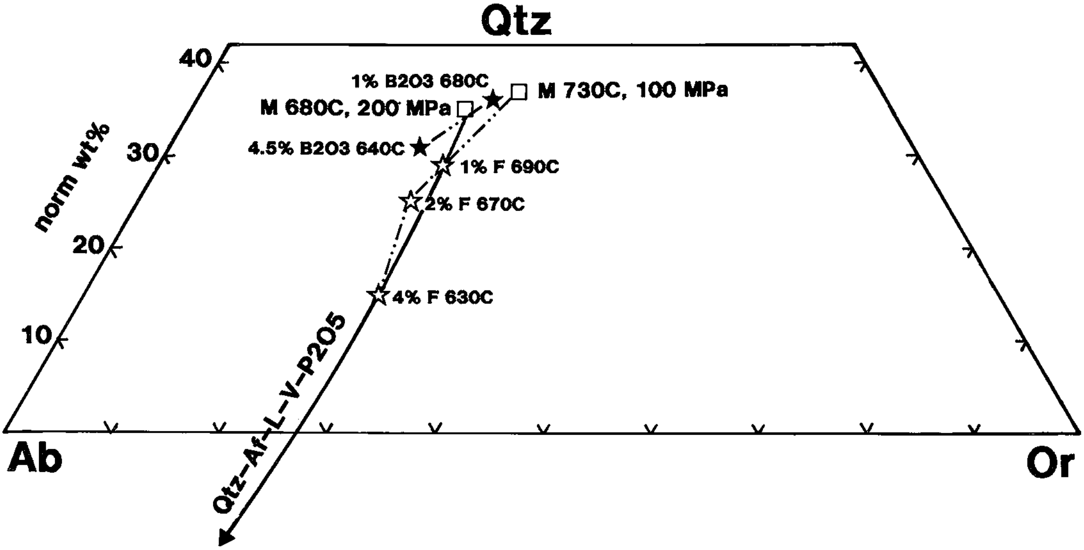


FIG. 3. Liquidus diagram of the H_2O-saturated haplogranite system (wt% components) showing the trace of reported "minimum melt compositions" and freezing point depression with the addition of fluorine (Manning 1981), boron (Pichavant 1987), and the trend of compositional change of melts in the haplogranite system with phosphorus (London *et al.* 1992). Granite minima at 100 and 200 MPa(H_2O) are from Tuttle & Bowen (1958).

Chorlton & Martin (1978) suggested a minimum melt temperature of 600°C at 100 MPa(H_2O). Their experiments achieved borate saturation only on quench of vapor, and hence their system was boron-undersaturated at P and T. The minimum melt temperature, therefore, lies below 600°C. Addition of boron to granitic melt causes an expansion of the liquidus field of quartz (Fig. 3) and evolution of residual melt toward Ab-enriched compositions (Pichavant 1987). Again, no boron-saturating phase was found in these experiments, so that the composition and temperature of the minimum at 100 MPa(H_2O) remain unknown. The effect on liquidus depression and expansion of the liquidus field of Qtz is somewhat greater in the system $Na_2O–Al_2O_3–SiO_2–B_2O_3–H_2O$ (Morgan & London 1987a). Perhaps more importantly, the addition of boron to hydrous sodium aluminosilicate melts ranging from peralkaline to peraluminous promotes an isobaric–isothermal increase in the solubility limit of H_2O in melt, by approximately 1.7 moles H_2O/mole B_2O_3 (Morgan 1988). Infrared absorption and NMR studies by Morgan *et al.* (1990) showed that most of the boron in hydrous alkali aluminosilicate melts exists in insular BO_3 clusters that are extensively hydrated and partially coordinated with alkalis and Al; there is no evidence for formation of B–O–Si bonds. In an experimental replication of natural liquid compositions from fluid inclusions with high boron content, London (1986a) reported complete liquid miscibility (*i.e.*, supercritical behavior) at 200 MPa in the system $Ab–Qtz–LiAlSiO_4–Li_2B_4O_7–H_2O$. Morgan & London (1987a) also encountered quenched experimental products indicative of complete H_2O-silicate liquid miscibility at run conditions (quenched, single-phase gels with plastic properties at room P and T). Thus, as the B content of granitic melt increases, H_2O and silicate melt become increasingly miscible to the point that a supercritical transition from melt to aqueous vapor may occur (*e.g.*, London 1986a, b).

Boron partitions slightly in favor of vapor over melt. Pichavant (1987) reported a *D*[B]vapor/melt of 3 for the haplogranite system, but this value is lower in peraluminous compositions and is dependent on temperature (London *et al.* 1988). Mass-balance calculations showed that boron significantly increases the dissolution of alkali aluminosilicate melt components into vapor (Pichavant 1987, London 1986a, Morgan & London 1987a, 1989), and particularly increases the solubility of Al in vapor relative to that of alkalis or Si (Morgan & London 1987a, b, 1989).

The stability of tourmaline is a key factor to accumulation of boron in granitic melts. Equilibria between tourmaline and other ferromagnesian silicates, particularly biotite, cordierite, and hornblende, buffer the boron content of melt at concentrations well below 1 wt% B_2O_3 (*e.g.*, Benard *et al.* 1985, Morgan & London 1989). This is true also of similar equilibria involving crystalline phases and aqueous vapor (Morgan & London 1989). The formation of tourmaline, however, depends largely on the Fe–Mg content of melt, which is normally quite low in evolved magmas. Once the buffer capacity of equilibria between tourmaline and biotite or hornblende is exhausted (*i.e.*, where tourmaline is the only ferromagnesian phase), then the boron content of melt may increase by essentially perfect fractionation. There are three types of evidence that pegmatite-producting felsic magmas commonly accumulate boron far above the expected concentrations of buffering equilibria involving tourmaline and other ferromagnesian phases. One is evidence from fluid inclusions, which may contain upward of 10 wt% B_2O_3 (London 1986a). A second is the common occurrence of very tourmaline-rich border zones within pegmatites, which are interpreted to record an influx of ferromagnesian components from host rocks into boron-rich pegmatite (London 1990a). Even more common is the formation of extensive metasomatic aureoles of tourmaline around pegmatites and similar granites that contain little or no tourmaline themselves (*e.g.*, Morgan & London 1989, London 1990a). In a detailed study of one such example, Morgan & London (1987a, b) combined experimental results with field evidence and mass-balance calculations to conclude that almost 80 mole% of the original boron content in the magma from which the Tanco pegmatite, Manitoba, crystallized was lost to the amphibolite host-rocks. All of these features indicate that the original boron content of any granitic pegmatite can be difficult, if not impossible, to evaluate, and may have been highest for those granites and pegmatites that contain little or no tourmaline and no other ferromagnesian silicates.

Behavior and effects of fluorine

Fluorine is a common constituent of hydrous silicates and phosphates in many granitic pegmatites, and therefore deserves consideration here. The abundance of fluorine in typical peraluminous pegmatites, however, has perhaps been overestimated. Mineral solid-solutions of $LiAlPO_4(OH,F)$, which historically have been called amblygonite (the fluorine end-member), have turned out to possess mostly montebrasite compositions (*e.g.*, Černá *et al.* 1972, London & Burt 1982a). Similarly, a large proportion of pink-lavender micas normally referred to as lepidolite are actually less fluorine-rich lithium-bearing muscovite (*e.g.*, Černý & Burt 1984). Fluorine-rich minerals, particularly topaz and fluorite, are normally absent or rare in these pegmatites. In one case study, Morgan & London (1987b) used internal and external mineralogical monitors to assess the fluorine content of the Tanco pegmatite, Manitoba. In terms of the fluorine index of Munoz (1984), the Tanco pegmatite lies closer to porphyry copper deposits than to the conspicuously F-rich porphyry molybdenum bodies.

The bulk effects of fluorine on liquidus phase relations in the haplogranite system have been determined experimentally by Wyllie & Tuttle (1961) and Manning (1981). Fluorine, when added as a component involving only the substitution F_2O_{-1}, lowers the minimum temperature of melting of the system to less than 550°C at 100 MPa (Manning 1981) and causes a marked expansion of the liquidus field of quartz. When projected onto the plane of the haplogranitic system, compositions of residual melt in equilibrium with quartz and alkali feldspar follow an isopleth of approximately Or_{30} toward the Ab–Or sideline (Fig. 3). The composition of the minimum melt in this system is not known, but is probably sodic and low in silica. Cryolite (*e.g.*, Manning 1981) and villiaumite are likely F-saturating phases at the minimum.

The F_2O_{-1} exchange in H_2O-saturated granitic or felsic melts apparently decreases the solubility limit of H_2O in melt (Dingwell 1985; *cf.* Sorapure & Hamilton 1984). One explanation of this behavior is that F and OH compete for similar coordination to cations in silicate melts. Spectroscopic studies of sodium aluminosilicate glasses show a preference of F for coordination with Na, Al and Si in the order Na > Al >> Si (Mysen & Virgo 1985).

Preliminary experiments by Manning (1981) show that for haplogranite compositions, the distribution coefficient for fluorine between vapor and melt is equal to 0.33 at 100 MPa. In a more extensive study, Webster (1990) observed values of *D*[F]vapor/melt from 0.2 to greater than 1, depending on melt or fluid composition, temperature, and pressure. For most compositions, however, *D*[F]vapor/melt was found to be less than 1. Small vapor/melt partition coefficients may partly explain why fluorine has little effect on the fractionation of other trace elements between vapor and melt (*e.g.*, Dingwell 1985). In the haplogranite system, however, fluorine does enhance the solubilities of all the major components of the melt, especially Al, in the aqueous vapor phase (Dingwell 1985).

Behavior and effects of phosphorus

Many classes of granitic pegmatites, particularly peraluminous ones, show a marked enrichment in phosphorus. Usually, this is revealed by a variety of common to abundant phosphate minerals, in addition to apatite, that include amblygonite–montebrasite, lithiophilite–triphylite and related phases, and monazite. Recent studies by London (1992, London *et al.* 1990c), however, reveal that a large fraction of the P content of peraluminous granites, pegmatites, and rhyolites resides in the feldspar phases. There are few reliable estimates of the whole-rock P_2O_5 content of phosphate-rich pegmatites. On the basis of modal data and mineral compositions, Morgan & London (1987b) estimated the P_2O_5 content of the Tanco pegmatite, Manitoba, at 1.2 wt%. Chemically evolved peraluminous granites and rhyolites possess elevated phosphorus contents as well (London 1992).

Other than a reconnaissance study by Wyllie & Tuttle (1964), and an investigation of the solubility of apatite in silicic melts (Watson & Capobianco 1981), there have been no pertinent experimental studies of phosphorus in hydrous haplogranitic systems. The results of a recent study of phosphorus in H_2O-saturated granite compositions ranging from metaluminous to peraluminous, however, will soon be available (London *et al.* 1990b, 1992). At 200 MPa(H_2O), the temperature of minimum melting in the haplogranite system with phosphorus lies below 550°C. Projections of melt compositions in equilibrium with quartz and alkali feldspar onto the "haplogranite" plane follow an Or_{28} isopleth past the Ab–Or join (Fig. 3). Melt compositions near 600°C are very alkaline, sodic, and low in silica. Qualitative electron-microprobe analyses indicate that glass (residual melt?) quenched from 550°C contains very little silica, and approaches the composition of the phosphorus-saturating phase in the system, a hydrous sodium phosphate. Small additions of phosphorus to the haplogranitic system also promote an increase in the solubility limit of H_2O in melt at 200 MPa. Spectroscopic studies of anhydrous alkali aluminosilicate glasses display evidence for coordination of P with Al and alkalis (Mysen *et al.* 1981, Gan & Hess 1989), which is consistent with expansion of the liquidus field of quartz and the compatibility of P in alkali feldspars (London *et al.* 1990b, 1990c, 1992, London 1992).

In the metaluminous haplogranite system, *D*[P]vapor/melt is less than 0.07 (London *et al.* 1990b, 1992), and is still less than 1 in complex peraluminous systems (London *et al.* 1988). Consequently, H_2O saturation promotes only limited redistribution of phosphorus between vapor and relatively unfractionated melts. As the melt evolves toward an increasingly alkaline, sodic composition, however, the solubility of P in vapor (as an Na phosphate component) increases substantially.

Comparative effects of boron, fluorine, and phosphorus

Figure 3 plots the reported minima for H_2O-saturated, nominally haplogranitic compositions with the addition of B (Pichavant 1987), F (Manning 1981), and a projected liquid line of descent for the granite system with P (London *et al.* 1992). Several points are evident:
(1) the compositions of crystalline phases and particularly residual melts cannot be represented within the haplogranite system. None of these systems can reach their respective minima within the "haplogranite" plane; (2) the minimum of each system cannot be attained until a B-, F-, or P-saturating phase becomes stable, and (3) at fixed pressure, each system possesses but one minimum melt composition and temperature. The minimum for each system probably lies and projects well out of the "haplogranite" plane at compositions that are sodic, alkaline, and low in silica.

All three components lower the temperatures of minimum melting substantially, and cause expansion of the liquidus field of quartz largely at the expense of albite. For the systems with P and F, the projections of residual melt compositions in equilibrium with quartz and alkali feldspar follow an Or_{28-30} isopleth toward the Ab–Or sideline. Changes of melt composition with the addition of boron do not appear to follow this trend. Unlike fluorine, both boron and phosphorus promote substantial increases in the solubility limit of H_2O in melt. Fractionation trends that increase the concentrations of B and P in melt, individually or together, will forestall H_2O saturation of derivative melts to later stages of crystal fractionation.

Spectroscopic studies, albeit mostly limited to anhydrous glass compositions, indicate a generally similar environment of coordination for B, F, and P (Mysen *et al.* 1981, Mysen & Virgo 1985, Gan & Hess 1989, Morgan *et al.* 1990). In metaluminous glasses, each component is associated principally with alkalis or Al, or both, but not Si. This speciation in glasses is similar to that of H_2O, as discussed above. The fractionation path of liquids with increasing B, P, or F parallels also that of increasing H_2O pressure.

Unlike oxides of the periodic Groups I and II, which are network-modifying components, F and the oxyanions of B and P (*e.g.*, BO_3, BO_4, and PO_4) do not contribute nonbridging oxygen to melt and hence cannot be construed as network-modifying components in the strict sense. Each of these components, however, reduces the viscosities of silicate melt (Dingwell *et al.* 1992, Knoche *et al.* 1992), and hence they have some network-interactive role. Addition of B, P, or F increases the degree of association (copolymerization) of SiO_4

tetrahedra in melt, but coordination of these components with Al or its charge-balancing cations removes them from network-forming roles.

EXPERIMENTS WITH PEGMATITE BULK COMPOSITIONS

Other than work in the hydrous haplogranite system, with or without added Li, B, P, or F, several experimental studies have utilized bulk compositions that were derived from or are considered equivalent to natural granitic pegmatites. These include early investigations using mineral composites believed to be representative of the simple pegmatites of Spruce Pine, North Carolina (Vaughan 1963, Fenn 1986), and the chemically evolved Harding pegmatite, New Mexico (Burnham & Jahns 1962, Vaughan 1963, Fenn 1986), and investigations by London *et al.* (1988, 1989) with the peraluminous rhyolite obsidian from Macusani, Peru. The major difference between the Spruce Pine experiments and those in the simple haplogranite system is the appearance of muscovite as a liquidus phase at pressures of 500 MPa, as the Spruce Pine bulk composition is peraluminous. Calculations by Burnham & Nekvasil (1986) predict that muscovite would be unstable on the Spruce Pine liquidus at 200 MPa, which places some minimum constraints on depth for these muscovite-rich pegmatites. As might be expected, the reported liquidus and solidus of H_2O-saturated Harding pegmatite are lower than those of the Spruce Pine sample, owing to the high content of Li and F in the Harding bulk composition (see Fig. 8 of Burnham & Nekvasil 1986). Lithium aluminosilicates (spodumene or petalite, depending on pressure) are stable liquidus phases (*e.g.*, Fenn 1986). The H_2O-saturated solidus of the Harding pegmatite (approximately 610°C at a P above 150 MPa: Burnham & Nekvasil 1986) was thought to represent the lowest temperatures at which pegmatite-forming magmas could exist, until studies of fluid inclusions by London (1986a, b, and several Soviet references therein) found evidence for magmatic compositions down to approximately 475°C. This lower limit of pegmatite-producing magmas was shown to be feasible in experiments with the Macusani glass (London *et al.* 1989), whose solidus lies at 450°C at 200 MPa (Fig. 2). This temperature represents the 200 MPa solidus for the bulk composition of Macusani glass. Solidus conditions for residual melts produced by H_2O-undersaturated fractional crystallization of Macusani glass were never obtained (London *et al.* 1989).

Many other observations were drawn from the experiments with Macusani glass (London *et al.* 1988, 1989). In the field of melt + aqueous vapor, London *et al.* (1988) noted surprisingly high reciprocal solubilities of melt and vapor (10–15 wt% dissolved solids in vapor, 11–12 wt% H_2O in melt). With respect to major and some minor components, the melt dissolves congruently in vapor, as reported in experiments by Kilinc (1969), which contradicts the frequently cited hypothesis that the generation of an aqueous phase promotes significant fractionation of major and minor components between vapor and melt, particularly with respect to alkalis. Indeed, London *et al.* (1988) examined the melt–vapor partitioning of 32 major, minor, and trace lithophilic elements, and found that only boron tends to partition into the vapor phase. All other elements, including Li, Rb, Cs, Be, Nb, Zr, *REE*, *etc.*, either showed no preference for vapor over melt or were strongly partitioned into the melt as opposed to vapor. Although traditional forward-direction experiments were used to locate liquidus and solidus boundaries (path E, Fig. 1), most experiments below liquidus temperatures of Macusani glass were made in the reverse direction. London *et al.* (1988, 1989) described some of the difficulties of working with the Macusani glass, including extreme metastable persistence of melt at H_2O-saturated and undersaturated conditions, and unusually long lag times for crystal nucleation in some experiments (discussed further below). The comparison of experimental results for H_2O-saturated *versus* H_2O-undersaturated compositions, for which the reader is referred to London *et al.* (1989) for details, clearly indicates that H_2O-undersaturated fractional crystallization of this composition replicates the general and many specific features of pegmatites, whereas H_2O-saturated experiments yield results that are distinctly unlike natural pegmatites.

PRESSURE – TEMPERATURE CONDITIONS OF PEGMATITE FORMATION

The pressures and temperatures at which granitic pegmatites are emplaced and crystallize have been estimated through melting experiments with granite–pegmatite compositions, by mineral geothermometry and geobarometry, combinations of fluid inclusion and stable isotopic analyses, the metamorphic grade of host rocks, and heterogeneous reactions among minerals in systems of low variance. As in other plutonic igneous systems, problems of exsolution and deviations from simple solid-solutions largely preclude accurate geothermometry based on the compositions of coexisting alkali feldspars or Fe–Ti oxides. Re-equilibration down to closure temperatures for exchange also hinders the utilization of stable isotopes for geothermometry. A multitude of problems inherent in the analysis of fluid inclusions (primary *versus* secondary origin, postentrapment modification, derivation of relevant isochores, estimations of pressure corrections to the temperatures of homogenization from the intrinsic properties of fluid inclusions) render this method nearly futile in the absence of other constraints. Because pegmatite emplacement commonly postdates regional dynamothermal metamorphism, it is difficult to accurately correlate the metamorphic thermal grade of host rocks with possible temperatures of emplacement of pegmatite-forming magmas; the use of metamorphic assemblages to delimit maximum possible pressures, however, is feasible. Experimental approaches currently provide the best pressure–temperature estimates.

The H_2O-saturated solidi and liquidi of haplogranite and pegmatite bulk-compositions have been widely used to delineate possible P–T conditions. Most petrologists utilize the H_2O-saturated liquidi to constrain emplacement temperatures of pegmatite-forming magmas, but this presumes *a priori* that the magmas are H_2O-saturated (*cf.* Fig. 2). If undersaturated, then the liquidus temperatures would be considerably higher (*e.g.*, Whitney 1988, Burnham & Nekvasil 1986, London *et al.* 1989, Webster *et al.* 1987).

The H_2O-saturated solidus in the haplogranite system does represent an applicable lower magmatic boundary for compositionally simple pegmatites if near-equilibrium crystallization prevails. Pegmatite-forming melts emplaced into cooler host-rocks may experience significant undercooling, however, before crystallization commences. The addition of components outside of the haplogranite system (*e.g.*, Li, B, P, F) also will lower liquidus and solidus temperatures. Notice in Figure 2, for example, that the liquidus of the Macusani glass lies below the solidus of the compositionally simple granite R1 (Whitney 1975) at 200 MPa(H_2O).

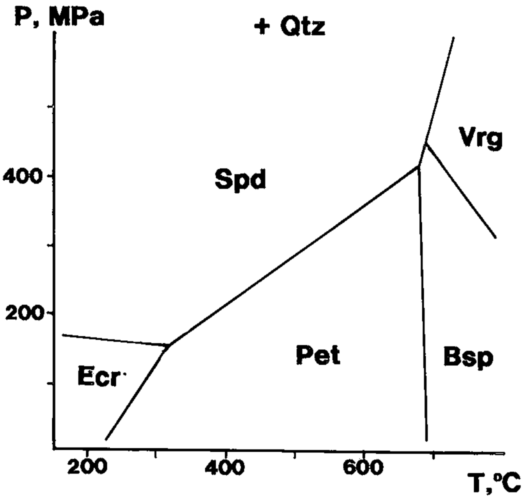


FIG. 4. Quartz-saturated subsolidus phase relations among the lithium aluminosilicates eucryptite (Ecr), spodumene (Spd), petalite (Pet), virgillite (Vrg), and β-spodumene (Bsp), from London (1984).

The quartz-saturated phase relations among lithium aluminosilicates (petalite, spodumene, and eucryptite in pegmatites) have been successfully utilized to ascertain the crystallization conditions of numerous lithium-rich pegmatites (Burt *et al.* 1977, see London 1984, 1990a for lengthier discussions). The value of lithium aluminosilicate – quartz phase relations as a frame of reference for fluid-inclusion studies has also been demonstrated (London, 1985a, 1986a). The primary pegmatitic phases spodumene and petalite are unstable with quartz above 700°C at low to moderate pressures (Fig. 4). Most massive and miarolitic pegmatites that contain lithium aluminosilicate assemblages, especially those with multiple (*i.e.*, successive) lithium aluminosilicate assemblages, crystallize at pressures in the range of 280–350 MPa (London 1984, 1986a, b, 1990a). In this pressure range, the primary crystallization of spodumene + quartz assemblages from melt (with or without an intervening vapor phase) must occur below 650°C. By combining phase relations of the lithium aluminosilicates with fluid-inclusion data, London (1986a, b) has defined solidi for the massive Tanco pegmatite and miarolitic rare-element pegmatites from Afghanistan and California near 475°C at pressures near 250 MPa for both types of pegmatite.

Rates of cooling

One critical parameter of pegmatite crystallization, the rate of heat loss from the magma (and consequently the rate and magnitude of undercooling of melt), is almost totally unstudied. Two variables needed to model cooling rates, the temperatures of host rocks at the point of magma emplacement and the mechanisms of heat transfer between magma and host rock, cannot be readily evaluated. If pegmatite-forming magmas are near thermal equilibrium with their hosts, then cooling will be constrained by the geothermal gradient of the tectonic regime. Cooling rates will be nearly linear, resembling path C in Figure 1. If pegmatite-forming magmas are hotter than their hosts, then the cooling rate will be nonlinear, and the rate of heat loss from magma to host will decrease from the point of emplacement until thermal equilibrium is attained (*e.g.*, path B in Fig. 1). Using a general model for magma cooling, Chakoumakos & Lumpkin (1990) have considered these two states of host-rock conditions to estimate the interval of magmatic crystallization for the Harding pegmatite, New Mexico. In their model, they constrained the temperature of the host rock to lie between 0° to 200°C below the inferred (H_2O-saturated) liquidus temperature of the pegmatite-forming magma, and assumed that heat was transferred through purely conductive processes. With these boundary conditions, they calculated that the interval of magmatic crystallization for the Harding pegmatite ranged from less than 100 to more than 1000 years. As the peak regional metamorphic temperature recorded by the rocks that host the Harding body is near 550°C (Chakoumakos & Lumpkin 1990), it is reasonable to infer that the host-rock temperature at the time of magma emplacement was at least 100°C lower than the pegmatite liquidus, and possibly much lower (contact metamorphic assemblages adjacent to the pegmatite are retrograde with respect to the regional metamorphic grade recorded by the unaltered host-rocks). Chakoumakos & Lumpkin modeled the thermal history of the center of the body of pegmatite-forming magma for the conditions of a liquidus at 650°C, the host rocks at 350°C, and other constraints as described in their Figure 7. For this realistic contrast in temperature, the magma at the center of the pegmatite body would have cooled below 550°C (*i.e.*, well below its solidus) in approximately 1 year. Such rapid rates of cooling approach those of experiments (*e.g.*, London *et al.* 1989) in which severe undercooling below the liquidus (*i.e.*, metastable persistence of melt and large lag times in crystal nucleation and growth) is evident.

The P–T path for the Harding body (Chakoumakos & Lumpkin 1990) is very similar to a well-constrained cooling–uplift curve for the Tanco pegmatite, Manitoba (London 1986a). Both bodies cooled almost isobarically to temperatures near 450°–500°C, from which point temperature and load pressures fell along paths close to reasonable geothermal gradients. The magmatic

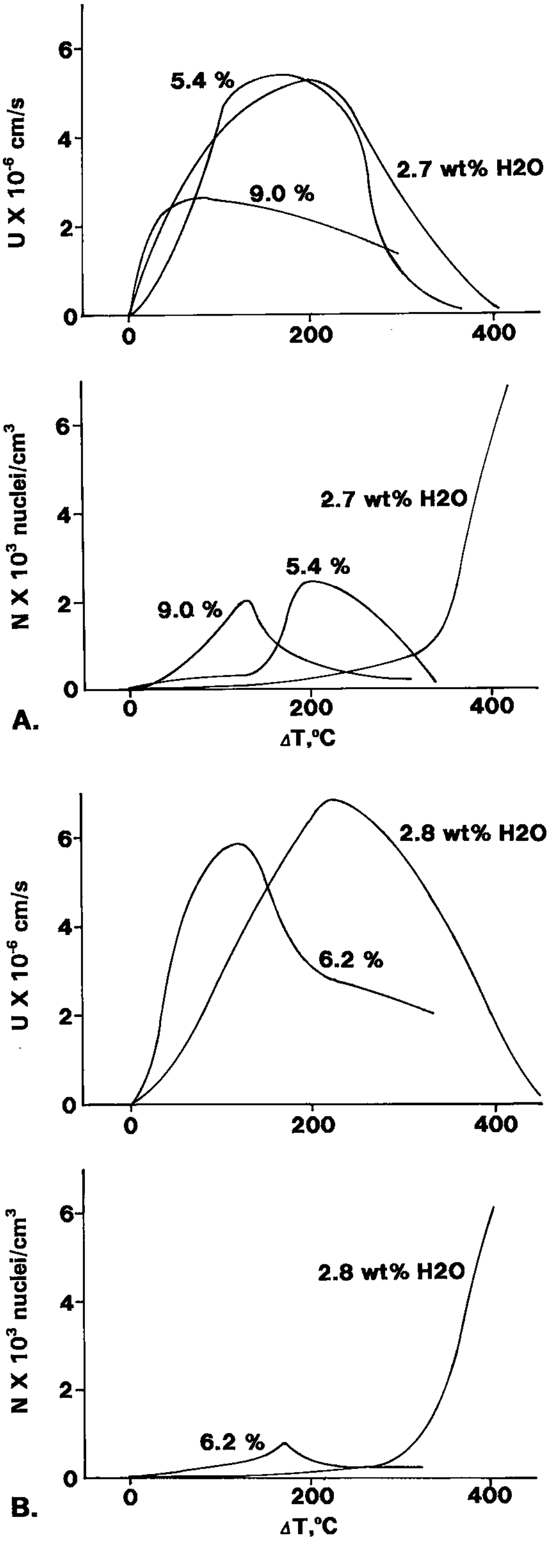


FIG. 5. Crystal growth (U X 10^{-6} cm/s) and nucleation densities (N X 10^3 nuclei/cm^3) for (A) sodic feldspar ($Na_{0.9}K_{0.1}AlSi_3O_8$) and (B) potassic ($Na_{0.5}K_{0.5}AlSi_3O_8$) feldspar as functions of liquidus undercooling (ΔT) and added H_2O content of melt (labeled on curves) for experiments at 500 MPa. Reprinted from Fenn (1977) by permission of the author.

interval corresponds to the nearly isobaric portion of the cooling curves. Such isobaric cooling through several hundred degrees Celsius is a qualitative indication of rapid cooling, as implied by the thermal analysis of Chakoumakos & Lumpkin (1990).

VARIATIONS OF GRAIN SIZE

Rates of crystal growth

The cooling model discussed above indicates that pegmatite-forming magmas, especially those responsible for chemically evolved pegmatites that are emplaced far from their sources, may experience rapid and substantial undercooling. The rate and degree to which melts are undercooled will dominate the kinetics of crystal nucleation and growth, and the resultant fabric of the rock produced. Figure 5 presents two representative sets of experimental data on feldspar nucleation and growth from Fenn (1977). Ranges of undercooling where most of the growth occurs give rates of growth in the range of 10^{-5} to 10^{-7} cm/s (Fenn 1977, 1986, Swanson 1977, Swanson & Fenn 1986, Muncill & Lasaga 1988). Taking these as average growth-rates for a system that experiences continuous cooling, crystals of quartz and the feldspars could grow to dimensions of 1 m in just a few years (*i.e.*, at a constant growth rate of 10^{-7} cm/s, a single crystal could grow to 31.5 cm in one year, or 31.5 X 10^6 s). Thus, the rapid rates of cooling deduced from the calculations of Chakoumakos & Lumpkin (1990) do not preclude the formation of giant crystals at normal rates of growth. What is necessary, however, is that the nucleation density must be much lower in pegmatite systems than what is considered typical of granites in the temperature range of undercooling in which most crystal growth occurs.

Effects of H_2O and other network-interactive components on nucleation and growth of crystals

In the course of any of the crystal-growth experiments cited above, the concentration of excluded components in melt, including H_2O, increases as crystal growth proceeds so long as the melt is not saturated in these components. Although the mechanism by which H_2O interacts with silicate melt is still a subject of debate, the spectroscopic data available from alkali aluminosilicate glasses indicate that the hydroxyl groups formed by dissolution of H_2O are associated primarily with alkalis and Al (*i.e.*, the feldspar-forming components) as opposed to Si (quartz-forming component). Spectroscopic studies cited previously report the same dominant speciation of B, F, and P with alkalis and Al in alkali aluminosilicate glasses. The observed liquidus relations in the granite system (expansion of the liquidus field of quartz at the expense of alkali feldspars with increasing B, F, or P in melt or increasing H_2O pressure) is perhaps the best evidence that the spectroscopic data for glasses have real significance for species in melts. For granitic systems, the speciation of H_2O, B, P, and F as inferred from liquidus and spectroscopic experiments suggests that the nucleation of the feldspars will be inhibited to a greater degree than that of quartz. Such behavior has been observed in experiments, and I believe it has important applications to pegmatites.

The experiments by Fenn (1977) revealed a substantial influence of H_2O on crystal nucleation in alkali feldspar compositions. For liquidus undercooling of up to 400°C, increasing H_2O content of melt (from low H_2O contents to saturation of melt) resulted in a nearly six-fold linear decrease in the maximum number of crystal nuclei per unit volume of sample. At very large undercoolings, increasing H_2O decreased the nucleation density by a factor of 30. Fenn (1977) recognized that this behavior was related to the interaction of H_2O with silicate liquid, and suggested that the reconstructive process required for crystal nucleation in melt changed in some way with the addition of H_2O. Molecular H_2O, or an association between protons or hydroxyl radicals and alkalis and Al in melt, could lower the nucleation density of the alkali feldspars by dilution of the crystal-forming components in melt adjacent to a critical nucleus of feldspar, thereby decreasing the probability of a nucleation event (*e.g.*, Kirkpatrick 1983). Effects of H_2O or its dissociation products on the activation energy for attachment of melt components to crystal surfaces remain uncertain until the speciation of H_2O in melt is better resolved (current estimates for the enthalpy of dissociation/association of H_2O in melt are 25 kJmol^{-1}: Silver & Stolper 1989, Dingwell & Webb 1990). A requirement that OH bonds be broken for the crystallization of anhydrous phases, however, would increase the activation energies for nucleation and growth of the alkali feldspars and perhaps quartz.

A second important feature of Fenn's (1977) experiments is that increasing H_2O was found to shift the maxima of nucleation density closer to the liquidus temperature and closer to the regions of undercooling for maximum rate of crystal growth (Fig. 5). Fenn (1977) observed, however, that the measured rates of crystal growth for the alkali feldspars actually decreased as melt attained saturation in H_2O, a result that he noted was at odds with the beliefs expressed by Jahns & Burnham (1969).

There are no published quantitative data on the effects of B, P, and F on kinetics of growth of silicates, but the data of Swanson & Fenn (1992) indicate that the addition of fluorine does not change the growth rates for quartz and feldspars. Though experiments by London and coworkers (*e.g.*, Morgan & London 1987a, London 1987, London *et al.* 1988, 1989, 1992) were not designed specifically to quantify the effects of B, P, and F on crystallization kinetics, we have observed that growth rates of alkali feldspars do not appear to be changed by the addition of B or P, but the nucleation densities decrease sharply at all degrees of melt undercooling. This is expected from the fact that boron and phosphorus oxyanions form largely covalent bonds with alkalis and Al (in glasses), and these bonds must be broken in order to nucleate feldspar from melt.

In experiments with Macusani glass (London 1987, London *et al.* 1989), which contains elevated concentrations of B, P, and F, the nominally anhydrous glass (0.5 wt% included and adsorbed H_2O) devitrifies instantaneously during forward-direction run-up to subliquidus temperatures at 200 MPa. This is in sharp contrast to the behavior of nominally anhydrous alkali feldspar and simple haplogranitic compositions, which show virtually no crystallization over extremely long run-times at comparable conditions. With the addition of H_2O to Macusani glass, reverse-direction experiments require longer lag times to initiate silicate crystallization (>300 hours with 3.5 wt% added H_2O), and no crystal nucleation occurs in melt with ≥ 4.0 wt% added H_2O (regardless of rate or magnitude of undercooling) for experimental durations up to 4000 hours. Similarly large lag times for nucleation (>5000 hours with no crystal nucleation) have been observed in H_2O-saturated experiments with F-rich ongonite (V. I. Kovalenko, pers. comm., 1988).

Correlations of these results with granite–pegmatite systems are limited by the differences between the experimental procedures and natural phenomena. Some potentially useful generalizations, however, can be drawn from the experimental studies:

(1) The observed rates of growth of the feldspars and quartz from silicate melt are adequate to produce crystals of pegmatitic dimensions in a geologically short period of time (<100 years, and perhaps <3–5 years). The addition of fluxing components (H_2O, B, P, and F) does not appear to substantially change the average rates of growth of silicates in the liquidus–solidus temperature interval, although there is evidence that growth rates decrease as magmas approach and achieve saturation in H_2O. The effects of B, P, and F, in addition to H_2O, are to promote sharply lower densities of nucleation in the temperature intervals of maximum growth (*e.g.*, the Macusani glass experiments cited above). As a result, crystals are larger and fewer in number. Based on the known or inferred mechanisms of speciation for these added components in granitic melts, the inhibition of crystal nucleation should be greater for alkali feldspars than for quartz.

(2) As average growth-rates for quartz and the feldspars change only slightly as a function of concentration of network-interactive components, the transition from granitic to pegmatitic grain-sizes must be promoted by a sharp decrease in the nucleation density in the temperature interval of maximum crystal-growth. In granitic systems and subsystems, this condition appears to be satisfied for haplogranitic compositions that are far below H_2O saturation, or for chemically evolved melts whose network structures are substantially disrupted by components such as B, P, and F in addition to H_2O.

(3) The lag time for crystal nucleation and growth, which is a qualitative reflection of melt structure and the probability of forming crystal nuclei, appears to be highest for feldspathic and granitic systems that are highly polymerized (*i.e.*, nominally anhydrous and with no other added fluxing components) and for compositions that are highly disrupted by the addition of quartz- and feldspar-incompatible components such as B, P, and F in addition to H_2O. There appear to be intermediate structural states of melt, related to the addition of small quantities of network-modifying (H_2O) or other melt-interactive components (*e.g.*, B, F, and P), that bring nucleation maxima and crystal growth rates into coincidence at the same degree of liquidus undercooling. In the absence of H_2O, moderate concentrations of the components B, P, and F (in the range of 1 wt% or less of each) appear to promote short lag times and high densities of nucleation in the interval of high rate of crystal growth (London 1987, London *et al.* 1989), which is comparable to the results of Fenn (1977) at modest H_2O content of melt. Thus, nominally dry but B-, P-, or F-rich silicic magmas should form granites as opposed to pegmatites, unless and until the H_2O content of melt increases substantially to promote a higher degree of melt depolymerization.

THE TRANSITION FROM GRANITE TO PEGMATITE

Granitic stocks commonly possess pegmatitic regions of two sorts: pegmatitic borders, and segregations of pegmatite formed *in situ* within granite. The development of such pegmatites normally occurs in the apices of these granites, and the granites themselves commonly produce aureoles

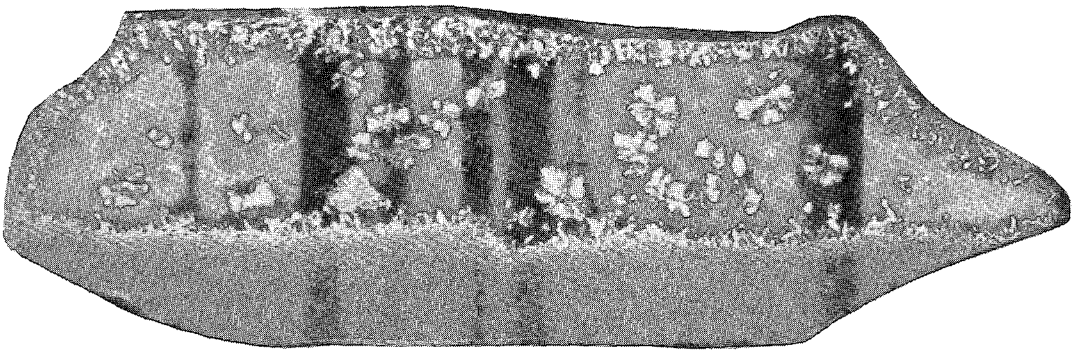


FIG. 6. Photomicrograph (in crossed nicols) reproduced from Figure 39 of Wyllie (1963) showing a reverse-direction experiment that was conducted by Jahns & Burnham as part of their experimental investigation of pegmatite-forming processes. The caption in Wyllie (1963) notes that the section measures 0.5″ across, and that a "fine-grained crystalline border encloses glass (isotropic) studded with radial groups of larger crystals...The development of a wide border facies at the bottom of the sample is due to a vertical temperature gradient." Reprinted from Wyllie (1963) by permission of the publisher.

of prograde thermal metamorphism in adjacent hosts. In other words, the granitic magmas have migrated into cooler rocks. Pegmatitic borders to larger masses of granite develop in regions that are likely to have experienced large undercooling below liquidus temperatures, as evidenced by the narrow, thermal metamorphic aureoles around these bodies (*e.g.*, Goad & Černý 1981, Černý & Meintzer 1988). In contrast, late-stage segregations of pegmatite within granite should not be subject to conditions of undercooling different from those of the adjacent granite. The transition from typical granitic to pegmatitic grain-size, however, is commonly abrupt (*e.g*, Fig. 6).

The traditional explanation of this textural change is that the development of a pegmatitic grain-size marks the point of aqueous vapor (over)saturation in magma, and that the actions of an aqueous vapor phase together with silicate melt make pegmatites as opposed to granites (*e.g.*, Jahns & Burnham 1969, Jahns 1982). Jahns (1982, p. 302) cited a critical experiment performed by himself and Burnham in 1958, in which the transition from granite to pegmatite was ascribed to the attainment of aqueous vapor saturation in initially H_2O-undersaturated melt [see Fig. 39 in Wyllie (1963), reproduced here as Fig. 6). The experiment was conducted by reverse-direction methods, and few details are given other than the general description by Jahns (1982) and additional remarks by Wyllie (1963). The experiment, involving a natural pegmatite bulk composition, was taken above its liquidus and then undercooled to an unspecified degree; the H_2O content of the experiment was such that the preconditioned (superliquidus) melt would be undersaturated (Jahns 1982). Wyllie (1963) commented in general and with specific reference to this experiment that the appearance of a "free vapor phase", which was equated with the formation of aqueous vapor by oversaturation of melt, could be "recognized by the preservation of bubble cavities in glass, the quenched liquid". As noted in the discussion of experimental methods, gas bubbles in glass are typical of all experiments with silicate liquids at pressure, including those that contain no H_2O (Burnham & Jahns 1962), and tend to be more prevalent as the H_2O content of melt is decreased. There are no details provided that would confirm if, let alone when, this particular experiment became H_2O-saturated. The experimental product (Fig. 6) is markedly asymmetrical, with the fine-grained granitic portion lying along one side of the run. Wyllie (1963) attributes this sharp boundary as "due to a vertical temperature gradient" through the capsule.

In experiments conducted by the same methods, London *et al.* (1989) have demonstrated that the observations of Jahns (1982) bear no causal relation to H_2O saturation of melt. In the experiments of London *et al.* (1989) and others (*e.g.*, Petersen & Lofgren 1986), similarly abrupt changes in grain size by factors of 10^2–10^3 over distances of 1 mm have been produced in melts far below bulk saturation in H_2O, and without local H_2O saturation in the boundary layer of melt in front of inwardly advancing crystals (ascertained from the low filling density of the gas bubbles in glass).

Experimental data such as those of Fenn (1977) provide insight into the probable explanations of sharp changes in grain size (granitic to pegmatitic).

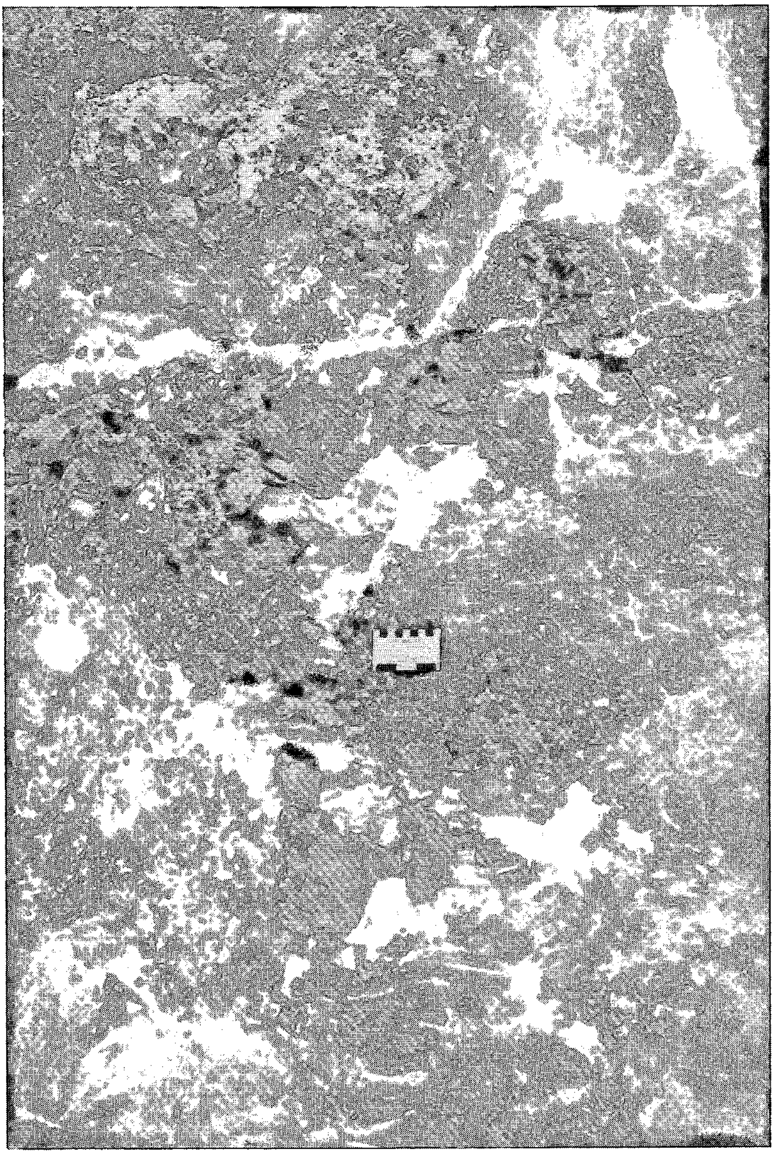
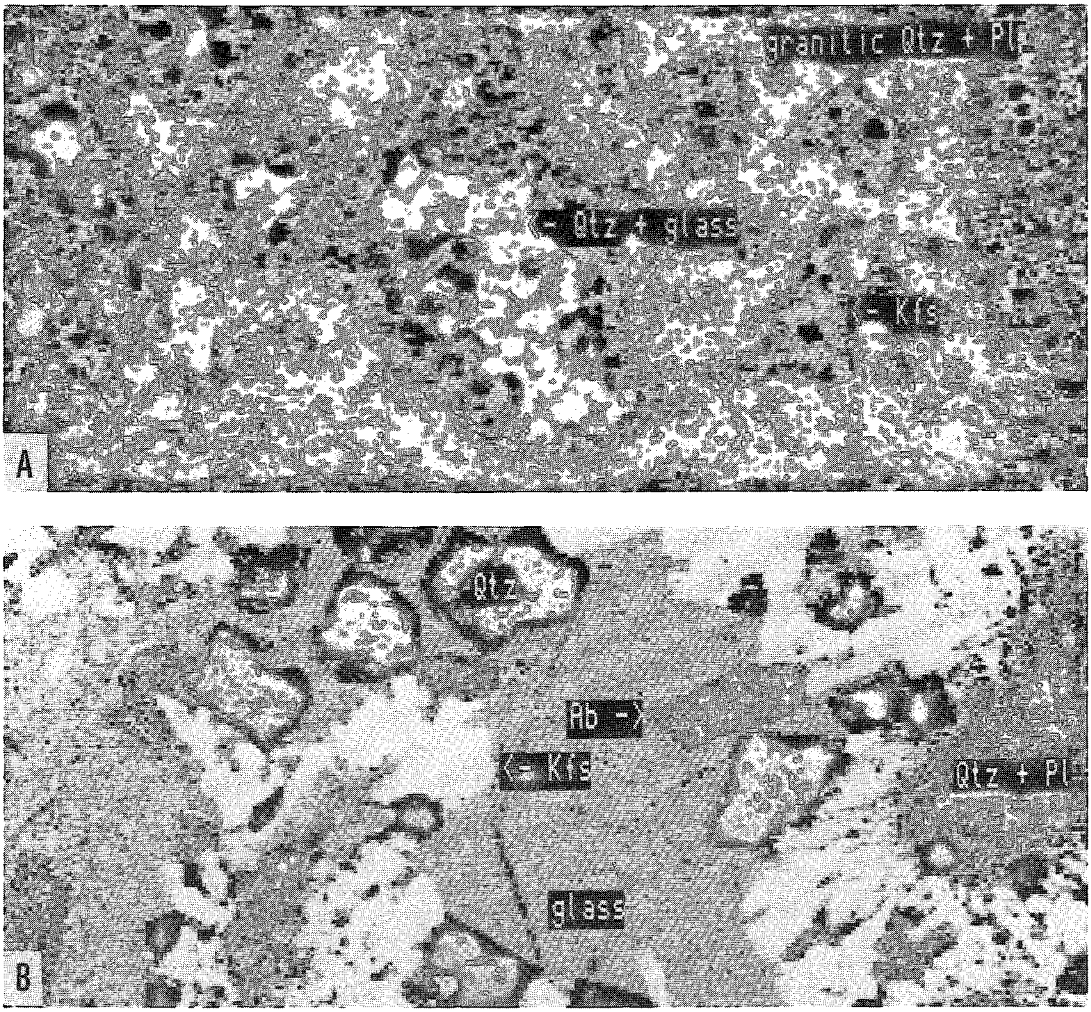


FIG. 7. Pegmatitic segregation within granite, White Rocks quarry, Middletown, Connecticut. Note the sharp transition from granitic to pegmatitic texture. The pegmatite consists of quartz (dark gray), microcline (white) and tourmaline (black). Top of scale marker is in cm.

Uniformly high rates of crystal growth prevail over a wide range of undercoolings below the liquidus, but nucleation densities in hydrous feldspathic melts possess sharp, narrow peaks within the undercooling interval of crystal growth. To demonstrate the critical relations of melt undercooling, H_2O content (or concentration of feldspar-incompatible but melt-interactive components), and grain size, the data from Figure 13 of Fenn (1977) at 4.3 wt% added H_2O can be recast as crystal dimensions, assuming a linear rate of growth at constant undercooling, over a duration of 10 years. Taking the total linear rate of growth over 10 years divided by the nucleation density (numbers of



individuals in the system of unit volume), individual crystals would attain maximum dimensions of 2.4 cm at undercooling of 150°C, 0.2 cm at undercooling of 200°C, and 1.6 cm at undercooling of 250°C. More realistic determinations of size of resultant crystals would entail consideration of nonlinear cooling rates (*e.g.*, Brandeis & Jaupart 1987), and variations in liquidus temperature and state of polymerization of the melt with fractionation. The example for this simple system, however, illustrates that relatively small changes in undercooling could promote large differences in resultant grain-size. The effect of increasing concentrations of fluxing components would be to drastically lower nucleation densities even further.

The general relationships of decreasing nucleation density with increasing concentrations of melt-interactive, crystal-incompatible components appear to lie at the heart of pegmatite formation. London *et al.* (1989) proposed a qualitative explanation for the decrease in nucleation density as a function of changing composition of the melt. Their model is based on the facts, known from experimental and spectroscopic studies of melts and glasses, respectively, (1) that components of H_2O, B, P, and F can dissolve into silicate melts in high molar quantities at elevated pressures, (2) that the dissolution of these components in melt involves some degree of reactive interaction with quartz–feldspar components of melt, and (3) that these components, except for P (London 1992, London *et al.* 1990b, 1992), are insoluble in quartz and in feldspar. London *et al.* (1989) suggested that local dilution of crystal-forming components and in-

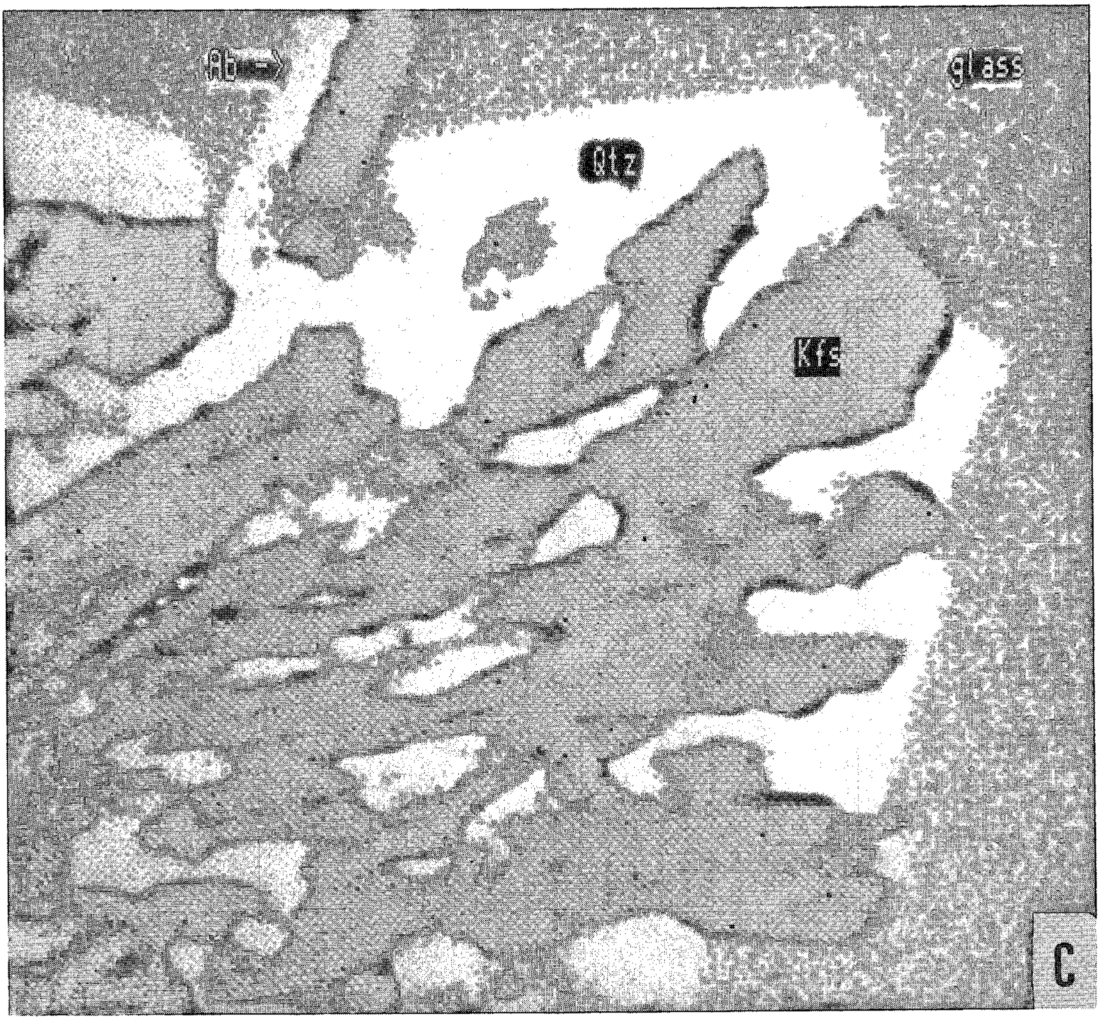
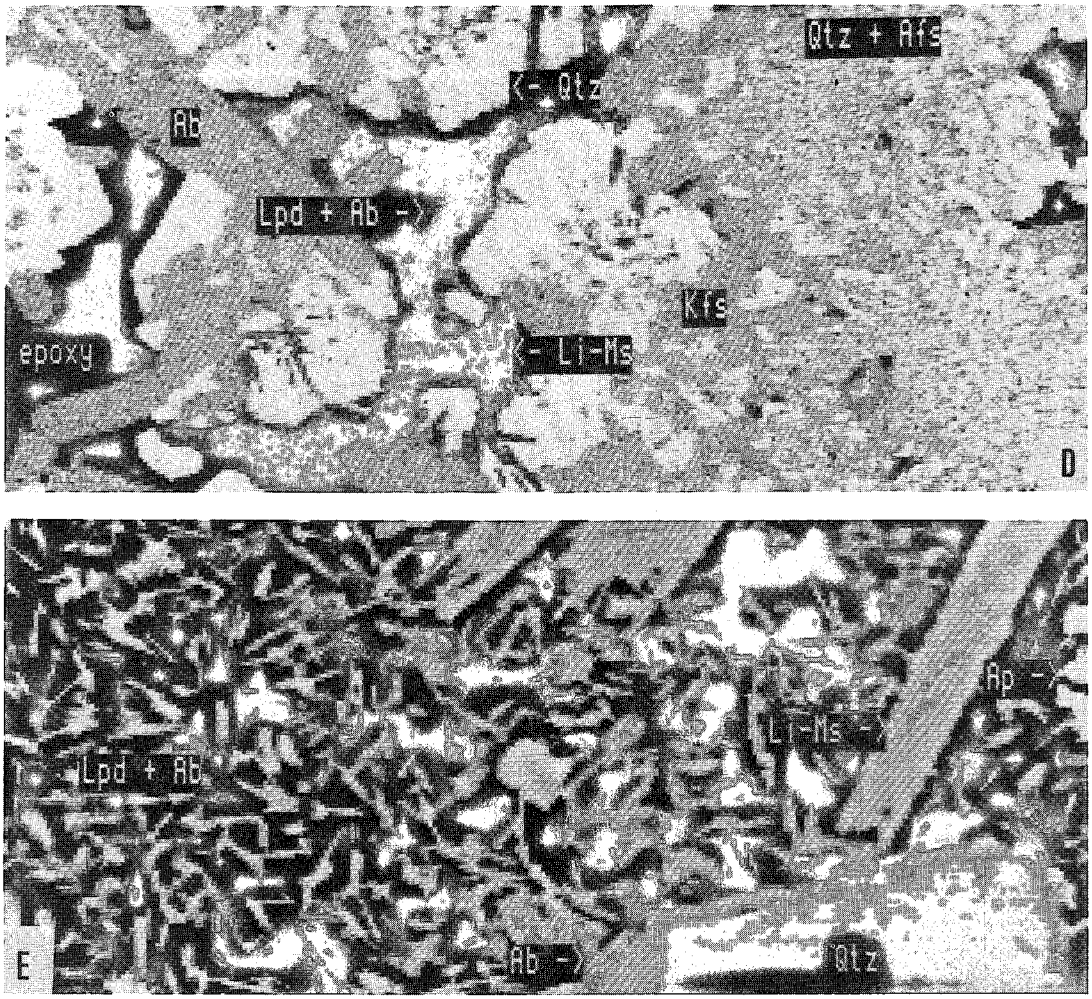


FIG. 8. Experimental fabrics produced by H_2O-undersaturated fractional crystallization of Macusani glass. All experiments are from London *et al.* (1989), and details of experimental conditions are described therein. (A) Potassium *Kα* X-ray map of experiment MAC 5 (no water added). Most of the sample consists of a fine-grained granitic intergrowth of quartz (Qtz) and sodic plagioclase (Pl). Dispersed within the granitoid texture are pegmatitic segregations, which are sharply defined by coarser-grained K-feldspar (Kfs) at their margins and aggregates of quartz in pools of residual melt. Photo covers an area of 512 × 256 μm. (B) Potassium *Kα* X-ray map of experiment MAC 149 (3 wt% water added, 575°C), showing the sequential development of pegmatitic texture and mineral zonation. Incipient crystallization, mostly along the margins of the capsule, produces graphic Qtz–Pl intergrowths. These are succeeded by a nearly monomineralic and coarse-grained fringe of K-feldspar (Kfs), and the accumulation of large crystals of quartz within pools of residual melt. Photo covers an area of 512 × 256 μm. (C) Potassium *Kα* X-ray map of experiment MAC 61 (3 wt% water added, 600°C). After the rapid growth of alkali feldspar (Kfs), monomineralic quartz (Qtz), as very large, composite crystals, is deposited along the feldspar growth-front. Photo covers an area of 89 × 89 μm. (D) Silicon *Kα* X-ray map of experiment MAC 102 (3 wt. water added, 500°C), showing many features that are present in MAC 61 and 149 (C and B above), but with the addition of a fine-grained intergrowth of lepidolite (Lpd) + albite (Ab); Li–Ms = lithium-bearing muscovite. Photo covers an area of 512 × 256 μm. (E) Digital BSE image of MAC 102, showing the texture of fine-grained lepidolite + albite in what were pools of residual melt. Note that apatite (Ap) is present only as a fine-grained phase in Lpd–Ab assemblages. Photo covers an area of 64 × 32 μm.



creased activation energies related to breaking bonds with crystal-incompatible components together served to severely inhibit crystal nucleation. London *et al.* (1989) reported experimental evidence that the nucleation of feldspars is suppressed to a greater degree than is the case for quartz. At large degrees of equivalent undercooling, quartz nucleated, but the feldspars did not. The increasing concentration of excluded components, which may be enhanced in the immediate vicinity of crystal nuclei, suppresses further crystal nucleation and growth.

For some specific conditions, a decrease in the degree of undercooling in a melt of constant composition will also promote a general increase in grain size, but this process is unlikely in the formation of the sharply bounded, late-stage pegmatite segregations that form *in situ* within larger masses of granite (Fig. 7). Changes in the degree of liquidus undercooling across this abrupt transition in texture must be small and insignificant. For this reason, and because small increases in the concentration of incompatible components can effect large decreases in the nucleation density, the process of chemical fractionation that increases the concentrations of fluxing components probably represents the dominant cause of the textural change from granite to pegmatite.

For components such as H_2O, B, P, and F to manifest changes in the nucleation density and consequent texture of rocks produced from granitic melts, the concentrations of these components cannot be fixed. Here it is important to consider the role of H_2O in compositionally simple granitic magmas (*i.e.*, those without demonstrably high concentrations of the other incompatible components). At aqueous vapor saturation, the activity of H_2O in melt is fixed, so that there will be no decrease in nucleation density and growth rate as a function of structural changes in the melt related to H_2O content. One way to interpret the transition from granitic (including the border zones that normally surround zoned pegmatite dikes) to pegmatitic textures in compositionally simple systems is to infer that the melts are not saturated in H_2O from the start, so that an increase in the H_2O content of melt may contribute to a decrease in nucleation densities and growth of fewer, larger crystals, as described above (*cf.* Figs. 6, 8A).

THE DEVELOPMENT OF ANISOTROPIC FABRICS IN PEGMATITES

A key distinction between most granites and pegmatites is that granites exhibit isotropic fabrics, whereas pegmatite fabric is characteristically anisotropic. The isotropy of granites is manifested by an essentially homogeneous distribution of mineral phases that possess random crystallographic orientations (neglecting flow foliation or imposed tectonism). Pegmatites are marked by two forms of anisotropic fabric: (1) highly oriented directions of crystal growth, mostly as comb-structure growth-induced orientations perpendicular to host rock or zonal contacts, and (2) a layered structure, with layering developed parallel to host rock or internal contacts.

Internal versus heterogeneous nucleation of crystals

In homogeneously nucleating systems, there is an equal probability that crystal nuclei will form spatially anywhere within the body. Purely random nucleation events are not likely in granitic melts, because these possess populations of different species (*e.g.*, Dowty 1980) and possibly relict or new crystals on which further growth and nucleation can commence; hence, the term *internal nucleation* is used. For internal nucleation, however, crystallographic orientations are random (as they would be for truly homogeneous nucleation). Heterogeneous nucleation occurs when the presence of a suitable substrate lowers the energy required to form stable nuclei below that needed to promote homogeneous nucleation. From solidification theory, there are a number of factors that may favor heterogeneous nucleation. These include pronounced effective undercooling of molten bodies (especially along their margins), a reduction in the critical size of stable nuclei caused by an increase in the radius of curvature of the crystal nucleus at its contact with the substrate, related effects caused by surface roughness, and epitactic inheritance of compatible crystal-structures from the substrate. In metallic and ceramic systems, the solidification of melts through heterogeneous nucleation usually produces a radially flaring fabric of crystals from the boundary of the melt toward its interior (Figs. 8B, C). When crystals nucleate on the substrate of the container, only crystals whose preferred or fast-growth axes point toward melt are able to sustain growth as the crystallization front advances. Crystal nuclei with unfavorable orientations may be starved before they grow to macroscopic size. Heterogeneous nucleation, therefore, produces anisotropic fabrics that exhibit comb structure and a high degree of preferred crystallographic orientation.

As magmatic systems are taken farther from equilibrium, for example by the imposition of large undercoolings or by structural changes in the melt that decrease the probability of crystal nucleation, the characteristic style of nucleation changes from random internal to comb-structured surface crystallization (*e.g.*, Lofgren & Donaldson 1975, London *et al.* 1989). Magmatic systems that manifest heterogeneous surface-nucleation do so because internal nucleation of stable phases is impeded by factors that inhibit the direct nucleation of phases from melt. Some form of catalysis is required to promote crystallization by lowering the activation or surface energy of crystal embryos below that of the bulk melt. In the crystal-growth experiments in compositionally simple systems (cited above), increasing the magnitude of initial undercooling leads to a progressive change from internal nucleation, in which crystalline phases do not possess a preferred crystallographic direction, to heterogeneous nucleation in which all crystallization starts from the substrate of the silicate melt (capsule walls) and proceeds inward in highly oriented and sharply bounded fronts of crystallization (Figs. 8B, C). There is a corresponding change in crystal morphology and habit, from euhedral, to skeletal or graphic individuals, to radial acicular spherulites as the degree of undercooling increases. As the lag times are large relative to the undercooling step, crystallization proceeds isothermally (neglecting changes in temperature caused by the latent heat of crystallization). The change from internal to heterogeneous nucleation, therefore, is not caused by thermal gradients in the experimental melts. The extent of disequilibrium and metastable supersaturation of melt are obviously rate-dependent; they are related to the rate of crystal nucleation and growth relative to the rate of cooling. They also are composition-dependent. That is, a chemically simple hydrous melt which, by virtue of its composition, might promote a high density of nuclei with short lag times at small degrees of undercooling, must be undercooled rapidly and by a large degree to initiate heterogeneous nucleation. Conversely, a melt whose composition decreases the probability of forming quartz or feldspar nuclei may require smaller degrees of undercooling to attain an equivalent degree of disequilibrium (London *et al.* 1989). Experimental granite–pegmatite systems with high concentrations of quartz- and feldspar-incompatible components also exhibit large lag times in crystal nucleation (some greater than six months), so that even with small initial undercooling and slow rates of cooling, the effective undercooling of melt may become great before crystal nucleation commences.

In the context of this discussion, the transition from isotropic granitic fabrics to anisotropic comb-structured pegmatite fabrics reflects a change from internal nucleation to strongly heterogeneous nucleation. This transition reflects an increasing degree of disequilibrium, the inhibition of nucleation and growth of crystals, caused by increasingly large and rapid initial undercooling of melt, by changes in melt composition (initially high or increasing concentrations of fluxing components) that inhibit nucleation and growth of crystals, or a combination of both factors. Rare-element pegmatites, which contain the highest concentrations of incompatible components, which on the basis of experiments will exhibit the longest lag times in crystal nucleation, and which may experience the largest initial undercooling by contact with cooler host-rocks, should and do possess the most pronounced comb-structure anisotropy.

Graphic intergrowths, skeletal crystals, and radial or plumose aggregates develop in conjunction with liquidus undercooling and metastable supersaturation of melt. Among the three major minerals of pegmatites, K-feldspar, which typically manifests a strong comb-structured growth orientation, commonly contains graphic or plumose intergrowths of quartz. The origin of graphic quartz – K-feldspar intergrowths has been explained through experiments in simple and complex granitic compositions (Fenn 1986, London *et al.* 1989) as the result of simultaneous deposition of quartz on growth fronts of K-feldspar. The saturation of quartz on a cellular (skeletal) growth surface of K-feldspar stems from the local increase in silica activity in a boundary layer of melt near the growth surface of the K-feldspar. A rapid rate of growth of the K-feldspar relative to the rate of diffusion of silica through melt is required. With slow growth of K-feldspar, or in melts with high diffusivity of elements (promoted mainly by the depolymerizing effect of H_2O or other fluxing components), the migration of silica down a chemical potential gradient into the bulk melt will preclude the formation of graphic intergrowths. Fenn (1986) noted that a graphic quartz – K-feldspar intergrowth could be produced experimentally only in melt compositions below bulk saturation in H_2O, but he proposed that melt in the boundary layer in front of advancing crystals might become oversaturated in H_2O. London *et al.* (1989) reported that graphic quartz–feldspar intergrowths were generated without local saturation of H_2O in the melt adjacent to fronts of crystal growth; vesicles trapped in melt at quartz – K-feldspar crystal surfaces were low-density air bubbles that had been pushed into the center of experiments by the advance of crystals. Indeed, the mechanism proposed by Fenn (1986) ceases to operate if a film of aqueous fluid separates the growth front of K-feldspar from melt.

Skeletal, branching, and radial habits of crystals are further indications of rapid disequilibrium-induced growth in silicate and other melts. The breakdown of faceted crystal surfaces into skeletal and dendritic forms with increasing degrees of undercooling is well documented (*e.g.*, Lofgren 1973, Lofgren & Donaldson 1975, Fenn 1977, Swanson 1977). Again, large-magnitude supercooling is but one means by which metastable supersaturation of crystalline components in melt may occur; a high proportion of incompatible network-interactive components contributes to a similar result at lesser degrees of undercooling. In pegmatites, branching habits may be developed in graphic quartz – K-feldspar intergrowths. Hollow, skeletal crystals of beryl and tourmaline are typical. Skeletal and branching habits are particularly common in phosphates and Nb–Ta oxides associated with coarse-grained albite–mica assemblages. In this same association, albite crystallizes as radial to parallel lamellar aggregates that nucleate on the surfaces of pre-existing megacrysts (usually K-feldspar, spodumene, or petalite) or small crystals (*e.g.*, minute crystals of zircon); micas develop plumose radial structures about similar centers of nucleation. Although these late-stage albitic rocks have in some cases been interpreted as metasomatic in origin, there is growing evidence that they represent crystallization from silicate melt (*e.g.*, London 1986a, b, 1987, London *et al.* 1989, Thomas & Spooner 1988). The characteristic skeletal and radial textures of this assemblage form at conditions where largely solidified pegmatite and host rocks are presumably near thermal equilibrium. At these conditions, metastable supersaturation of melt through rapid undercooling is not a viable process. Alternatively, inhibition of crystal nucleation in the late stages of pegmatite consolidation may be caused by increased concentrations of incompatible components, especially H_2O, B, P, and F, and by the low temperatures at which these late-stage assemblages crystallize.

Layered bodies

A distinctive anisotropic fabric of many pegmatites, both simple and complex in composition, is the presence of fine-grained saccharoidal to aplitic masses that possess a well-developed layering of minerals. They are mostly albitic in composition (far from the eutectic composition of granite), and contain lesser amounts of quartz, micas, and K-feldspar (although any one of these phases may be absent). They may contain a variety of accessory phases including individually or in combinations: garnet, tourmaline, and rarely gahnite, apatite, and Nb–Ta oxide minerals. Layering is manifested as nearly monomineralllic alternations. The thickness of layers is measurable in centimeters, and individual layers may be traceable for distances of up to a kilometer (*e.g.*, Foord *et al.* 1991). The layering varies from flat planar to highly convoluted. Where orientational anisotropy of grains exists, nonequidimensional minerals (tourmaline, micas, and albite) are foliated and may be lineated parallel to the layering. Perpendicular lineation (comb structure) is extremely rare. The layered albite-rich rocks are normally found near the footwall of shallowly dipping pegmatite dikes and sills. Exceptions do occur (*e.g.*, London 1985b, 1987). Note that texturally similar units exist without layering (in nature, and in the experiments of Jahns & Burnham shown as Fig. 6 here, and also London *et al.* 1989), that equivalent coarse-grained assemblages are common (in some cases referred to as "cleavelandite" complexes), and that coarse-grained and fine-grained, layered and non-layered variants of essentially the same assemblage can occur within a single pegmatite.

Four different mechanisms are commonly invoked to explain the development of layering within igneous bodies: (1) cumulus processes, (2) pressure fluctuations, (3) nonequilibrium sequential crystallization caused by significant undercooling below the liquidus, and (4) flow segregation. In rare instances (*e.g.*, the Tanco pegmatite, Manitoba), fragments of plastic, partially consolidated layered albite-rich rocks appear to have accumulated on footwalls of the pegmatite, and some are suspended in pure massive quartz (Fig. 9). An accumulation process for layering in pegmatite-associated aplites has been generally dismissed, however, on the grounds that the magmas are too viscous, and that the grain size and the density contrast between crystal and magma are too small to promote crystal settling (Jahns & Tuttle 1963). The other mechanisms warrant further consideration.

Jahns (Jahns & Tuttle 1963, Jahns & Burnham 1969) invoked pressure fluctuations caused by episodic loss of aqueous vapor due to overpressure to account for the three dominant characteristics of most pegmatitic aplites: their fine grain-size, sodic compositions, and layering. In this model, saturation in aqueous vapor and buoyant ascent of vapor bubbles at the inception of crystallization were considered to preferentially remove K from the lower portions of the haplogranitic melts, leaving them albitic in composition. Significant undercooling created by the shift in melt composition away from the granite minimum would drive rapid nucleation and growth of crystals, producing fine-grained rocks. Alternations between albite- and quartz-rich layers were explained by episodic release of aqueous fluid overpressure, which would cause isothermal oscillations in the compositional location of the albite–quartz cotectic.

The plausibility of this proposed mechanism has been diminished by subsequent experimental work and other mass-balance considerations. Experiments by Burnham and others (cited in Burnham & Nekvasil 1986, also see London *et al.* 1988) demonstrated that the K-number [100K/(K + Na)] of aqueous fluid is the same as that of the granitic melt with which it coexists at equilibrium, so that the initial proposition that the K-number of melt decreases with saturation in aqueous vapor is invalid unless the melt is saturated in muscovite (Burnham & Nekvasil 1986). Although some layered aplites contain muscovite, many do not. Where present, muscovite normally constitutes less than 5% modal (the K-number of vapor increases in proportion to the modal quantity of muscovite crystallizing from melt). By consideration of these facts, and other mass-balance calculations based on experimental data for alkali partitioning (Webster & Holloway 1988, Webster *et al.* 1989, London *et al.* 1988) and estimates of bulk chlorinities of pegmatite-forming magmas (London 1987), there is no apparent mechanism by which the K-number of a melt can be decreased sufficiently by saturation in aqueous vapor to produce such a K-depleted (and hence undercooled) sodic magma. If the K-number of melt could be shifted so far as to produce nearly monomineralic, fine-grained albite-enriched rocks in the footwall portions of pegmatites, then the corresponding addition of K to the hanging-wall portions would produce a complementary shift of the bulk composition (equal magnitude, opposite direction) away from the haplogranite eutectic. By the same arguments of Jahns & Tuttle (1963), this should produce a sharp rise in liquidus temperature

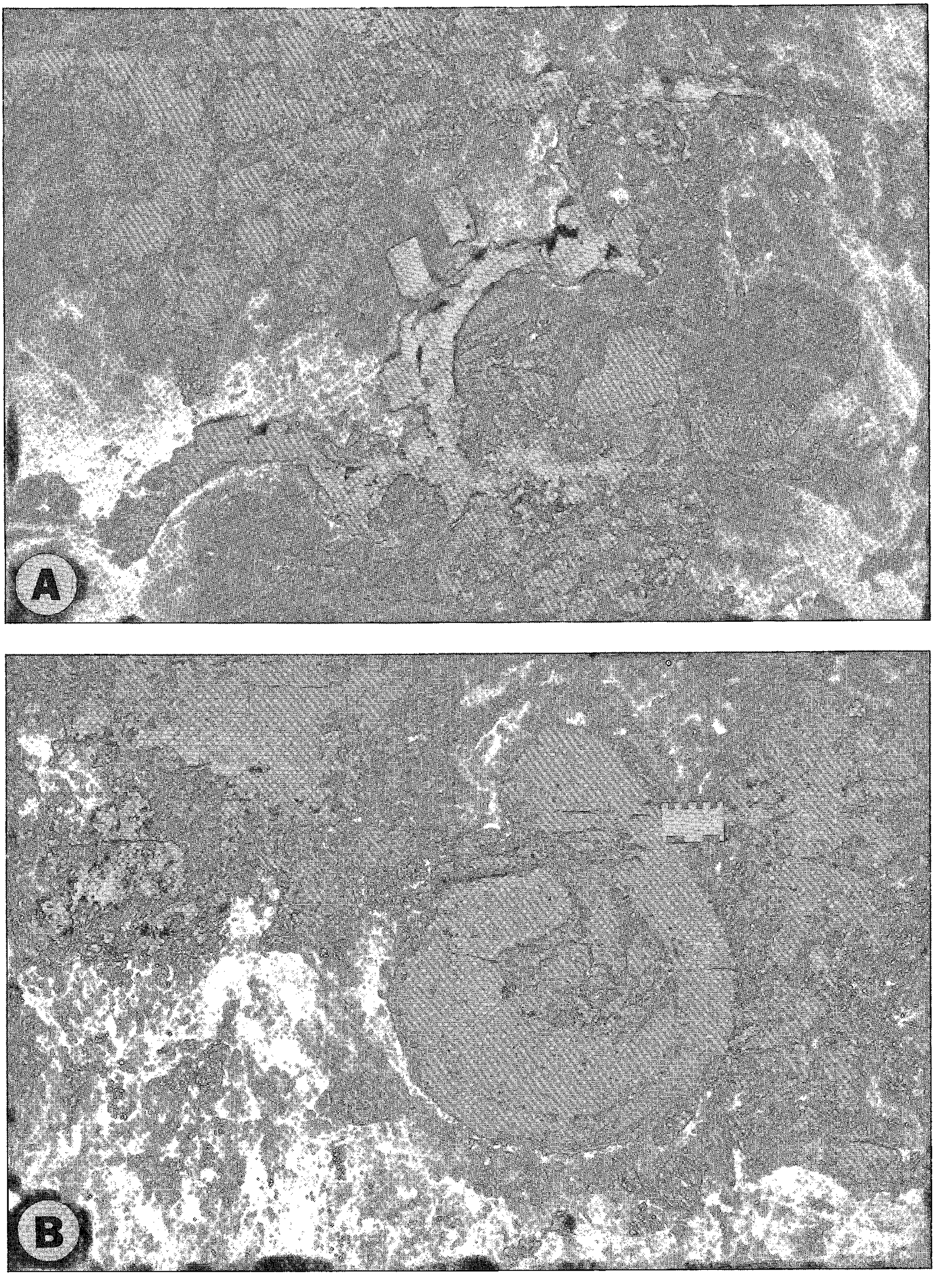

A
B

FIG. 9. Textural relations of saccharoidal layered albitic unit and massive quartz units in the Tanco pegmatite, Bernic Lake, Manitoba. (A) From lower right to upper left: faintly banded mass of saccharoidal albite, bounded by a thin band of Nb–Ta oxides (black), then white cesian beryl, and massive quartz (medium gray). The two large crystals of beryl in quartz are commonly 20 cm in length. Photo courtesy of P. Černý. (B) Curved, teardrop-shaped layer of banded saccharoidal albite, concave-upward, in massive quartz. Mining showed this mass of layered albitic rock to be completely surrounded by quartz (Peter J. Vanstone, Tantalum Mining Corporation of Canada, Ltd., pers. comm., 1985). Top of scale marker is in cm. (C) Rubble of bent layers of saccharoidal albite that have accumulated on the footwall of fine-grained albite unit, with massive quartz above and between layers. Standard geological hammer for scale. Photo courtesy of P. Černý.

(*i.e.*, effective undercooling below the liquidus) and result in similarly fine-grained, "quenched" grain sizes. This is clearly not the case, as the potassic hanging-wall zones, where developed, are among the coarsest-grained assemblages in any pegmatite. There are other important details of layered aplite bodies that are difficult to explain by the model of vapor exsolution that they proposed. These include the normally sharp marginal boundaries of layered aplites with other pegmatitic units (*i.e.*, how these could be developed in a vertical continuum of upwardly ascending aqueous vapor), and the fact that the alternations in layering are not always among quartz and feldspar (*i.e.*, cotectic in nature).

The model of sequential crystallization of nearly monomineralic layered rocks through liquidus undercooling has been developed by McBirney (1987) for application to layered mafic bodies. The essential components of this model, comparatively rapid cooling relative to rates of crystal growth and slow diffusion of excluded components through boundary layers in the melt, are even more likely in the context of small bodies of highly polymerized pegmatitic magma than in the case of large basic intrusions. Although this process has produced sodic albite – quartz border zones in experiments (*e.g.*, London *et al.* 1989), it has not been demonstrated that the process is viable on the scale of meters as in natural pegmatites. A potential obstacle to this model in natural pegmatites is the difficulty of diffusing K away from growth fronts in aplite so as to preclude local saturation in K-feldspar-rich layers. Many layered albite-rich aplites, however, do contain comb-oriented K-feldspar megacrysts, which are not normally included in the bulk compositions of the albite-rich units. These megacrysts indicate contemporaneity in albite - quartz - K-feldspar deposition, and possibly the effective removal of K from boundary layers in the melt as megacrysts rather than fine-grained crystalline layers. One additional problem with a model invoking rapid crystal-growth front is that flow deformation of layers, which has the appearance of gravitational slumping, is apparent in many cases. The implication is that interstitial melt is present at some point after the initial formation of layered sequences (*i.e.*, the growth front is not entirely solid).

Mineral segregation by viscous flow of partially consolidated aplitic bodies could account for at least some of the observed layering. Viscous flow is implied by the convolute shapes of some aplitic layers at their contacts with other pegmatitic units, and by the strong foliation and, in some cases, lineation of tourmaline and micas parallel to layering. There are, however, too many instances in which nonequidimensional minerals lack a preferred orientation to accept flow segregation as a primary process.

In summary, the textural development of layered aplites remains one of the major enigmas of pegmatitic rocks. Whereas any one of the three mechanisms cited above might be applicable in a particular circumstance, none of them provides a satisfactorily general or viable explanation, at least with the current development of the models.

INTERNAL ZONATION OF PEGMATITE BODIES

One of the most conspicuous features of granitic pegmatites is the common development of sharply bounded internal mineralogical zones. In its simplest form, this heterogeneous distribution of mineral assemblages, which represents another form of anisotropy that distinguishes pegmatites from typical granitic rocks, is manifested as concentric zoning in (1) granitic to markedly sodic outer margins, succeeded inwardly by (2) K-feldspar-rich units, and finally (3) quartz-rich interior bodies, often referred to as cores with the genetic connotation of being the last-crystallized, nonreplacive portions of pegmatite. A vertical asymmetry in concentrically zoned and especially layered pegmatites is apparent in the segregation of K-feldspar-rich units toward the upper portions and plagioclase-rich bodies near the bottom. Deviations from these simple generalizations, however, are numerous, and they tend to increase as the bulk compositions of the pegmatites become more evolved. Departures from the idealized zonation include chaotic and sporadic distributions of zones, or inversions and other irregularities in the typical zoning pattern (*e.g.*, footwalls rich in coarse microcline and layered sodic aplites on hanging walls: London 1987; bodies of monomineralic quartz at the margins of pegmatites: *e.g.*, Černý 1982b; lenses of monomineralic quartz that serve as the substrate for inwardly flaring spodumene–quartz–albite assemblages: *e.g.*, Jahns & Ewing 1977; and many others).

Unzoned pegmatites, including chemically evolved types, do certainly exist (*e.g.*, the Kings Mountain pegmatites in North Carolina: Kunasz 1982). Most of these, however, possess some features of anisotropic fabric, such as comb structure of microcline or spodumene, and small, dispersed lenses of mineralogically distinct (*i.e.*, zoned) assemblages, such as spodumene + quartz + muscovite at Kings Mountain.

Any discussion of mineral zonation in granite pegmatites is inseparable from the consideration of pegmatite fabric, which has been presented above. As a rule, the sodic border zones are comparatively fine-grained or granitoid in fabric (these are commonly difficult to distinguish, or are not distinguished from, the late-stage albite-rich rocks that represent the most fractionated of the pegmatite-forming liquids). The potassic zones that follow inward are sharply porphyritic in texture, usually with increasingly abundant, comb-structured megacrysts of K-feldspar surrounded by finer-grained quartz–plagioclase assemblages; or, fine-grained sodic borders terminate abruptly against uniformly megacrystic and monomineralic K-feldspar. The distinctly quartz-rich units are megacrystic. Noting this inward progression of textures associated with mineral zonation, Jahns (1953) affirmed the prevailing concept of pegmatite genesis of his time: that pegmatites crystallize from the margins inward by "fractional crystallization and incomplete reaction between successive crops of crystals and rest-liquid..., with or without end-stage deuteric or hydrothermal activity." In the ensuing decade, Jahns and others (principally with C.W. Burnham, O.F. Tuttle, and students) embarked on the first experimental studies of pegmatite genesis, aimed at defining the P–T–*X* variables attending pegmatite formation, and specifically to address the textural features of pegmatites as compared to granites. These experimental studies constitute the basis of the petrological understanding in the crystallization of siliceous melts at elevated H_2O pressures. Insofar as pegmatites fall within the range of the compositions studied, these experiments provided constraints on P and T for H_2O-saturated melts, as discussed above. They did not, however, resolve the questions of textural development and zonation. One reason is that experiments aimed at determination of equilibrium boundaries were performed in a forward direction, as in most subsequent studies in the granite system, and hence could not replicate the crystallization of silicate melt. An important outgrowth of this experimental period was a fundamental change in perception, that the crystallization of pegmatites can be described as an equilibrium process, which is wholly different from the disequilibrium fractional crystallization model that Jahns and others (*e.g.*, Cameron *et al.* 1949) had previously espoused. The requirements of equilibrium crystallization in the eutectic haplogranite system forced Jahns to abandon his views of sequential crystallization, and instead to propose that all of the three dominant zonal assemblages (sodic, potassic, and silicic) crystallized simultaneously. The mechanistic vehicle for zonal segregation centered on the role of a buoyant aqueous vapor phase in promoting mass transfer through the crystallizing pegmatite system, as discussed above for the generation of sodic aplites. Ignoring a few other suggestions that lack sufficient justification, therefore, the formation of mineral zones in pegmatites can be reduced to essentially two proposed mechanisms: (1) disequilibrium fractional crystallization of silicate melt by sequential, inward crystallization from the margins, or (2) eutectic equilibrium crystallization, in which the three dominant zonal assemblages develop simultaneously but separately because of incongruency in solubilities of alkalis, Al, and Si between silicate melt and aqueous vapor.

Evidence of saturation in an aqueous vapor

As the coexistence of an aqueous vapor phase with silicate melt is central to the model of Jahns & Burnham (1969; Jahns 1982, Burnham & Nekvasil 1986), it is necessary to review the various arguments for or against its presence early in the pegmatite-forming process. The list below contains some features of pegmatites that have been regarded as evidence of saturation in an aqueous vapor.

Miarolitic cavities. Miarolitic cavities, in which primary pegmatite minerals project into open space (locally filled with clay), constitute evidence of vapor saturation prior to the complete crystallization of pegmatite-forming melt. Such cavities are rare, however, and where present, they usually constitute a minute volume-fraction of space. Depending on the depth of emplacement, pegmatite-forming magmas that are vapor-saturated from the start should contain approximately 2.5 (500 MPa) to 8.5 vol.% (200 MPa) void space if crystallized as closed systems (Burnham & Nekvasil 1986). Pegmatites containing such large volume-fractions of void space are extremely uncommon. For example, over 24,000 pegmatite bodies have been recognized in the Black Hills district, South Dakota, and not one of them can be classified as miarolitic. Miarolitic cavities, which are found in the latest-crystallizing pegmatite units, cannot be taken as evidence that the pegmatite magmas were H_2O saturated at the onset of crystallization (see London 1986b, 1990a).

Presence of hydrous minerals. Although hydrous minerals are common constituents of pegmatites, they can be stable well below H_2O saturation of melt (*e.g.*, Maaløe & Wyllie 1975). The presence of fluorine, together with other substitutions, may extend the stability of hydrous phases in melt compositions that are well below saturation in an aqueous phase (*e.g.*, Webster *et al.* 1987, London *et al.* 1989). The presence of hydrous phyllosillicates, therefore, does not signify saturation in an aqueous phase.

Fluid inclusions. Dominantly aqueous fluid inclusions are common in pegmatite minerals, especially quartz. Where a suitable independent frame of reference (*e.g.*, the univariant reaction spodumene + 2 quartz = petalite: London 1984) can be used to ascertain origin of the inclusions, it can be demonstrated that the aqueous inclusions in quartz are almost always secondary, with entrapment temperatures well below any feasible magmatic solidus [London 1985a, 1986a, b, 1990a; *cf.* Thomas *et al.* (1988)] who suggested a magmatic solidus of 262°C for the Tanco pegmatite, Manitoba, based on an interpretation of inclusions in quartz as primary). Inclusions that can be reliably construed as primary are silicate-rich, and represent the entrapped products of hydrous silicate melts (*e.g.*, London 1986a, b, and references therein to Soviet studies). Hosts to these silicate-rich inclusions contain secondary planes of aqueous inclusions that are indistinguishable in properties from those of quartz. In the specific case of the Tanco pegmatite, Manitoba, London (1986a) used an experimental analog to the silicate-rich inclusions to show that this late-stage borosilicate melt could not have coexisted with an aqueous fluid phase. For the P–T–*X* conditions of the Tanco system, H_2O and borosilicate melt would have been completely miscible (*i.e.*, supercritical). If anything, the lack of evidence from fluid inclusions for coexistence of aqueous fluid and silicate melt in granitic pegmatites is striking. Although inclusions full of silicate daughter minerals can be found in tourmaline, topaz, beryl, feldspar, but almost never quartz, they are very common in spodumene. Perhaps this is because silicate melt tends to wet the surface of spodumene crystals and to displace aqueous fluid away from the surface (London 1986a). Post-entrapment modification of inclusion (*e.g.*, necking down) is prevalent and generally proceeds to low temperatures (<150°C). This process presents the greatest obstacle to meaningful studies of inclusions in pegmatites. In summary, aqueous inclusions in quartz are normally secondary, and their presence cannot be equated to the existence of primary inclusions that would or could confirm the coexistence of aqueous and melt fluid phases.

Wallrock alteration. Metasomatic alteration of host rocks by pegmatite-derived fluids is a common feature surrounding evolved types of granitic pegmatite. London (1986c) noted that the alteration assemblages around rare-element pegmatites are typical of greenschist-facies metamorphic conditions. Morgan & London (1987b) demonstrated that the temperatures associated with the pegmatite-induced wallrock alteration around the Tanco pegmatite were below 500°C. An important point, evident in the unique study of Morgan & London (1987b), is that the chemical nature of the wallrock metasomatism reflects the internal mineralogy of the spatially associated pegmatite. This feature implies that the chemical segregation of the pegmatite is already in place (*i.e.*, already crystallized, down to the latest, most differentiated units) prior to expulsion of fluid from the pegmatite.

Many pegmatites lack any evidence of fluid-induced wallrock alteration, even though the evolved mineralogy of some pegmatites suggests that there should be chemical disequilibrium and hence metasomatic reaction between pegmatite-derived fluids (if present) and surrounding hosts. For example, the Himalaya dike system in San Diego County, California, is remarkably rich in tourmaline (*e.g.*, Foord 1976). In large portions of the dike where tourmaline constitutes ten or more percent of the mode, the host norite is completely unaltered, except for a thin (< 1 mm) layer of biotite at the pegmatite margin, and sporadic argillic alteration of feldspars (possibly from weathering). Where rupture of miarolitic pockets has expelled pegmatite fluid into the norite, however, large aureoles of metasomatic tourmaline were developed. The implication is that the norite was reactive with the B-rich pegmatite fluid, as is generally the case for mafic hosts to boron-rich pegmatites (Morgan & London 1987b, 1989), but that these fluids were not present at the time when the pegmatite first began to crystallize, and had ample communication with the host norite. If anything, the pegmatite-forming magma was initially open to influx of components derived from the host rocks, as discussed below.

Other arguments. In connection with wallrock alteration or its absence, several other considerations are pertinent. Late-stage metasomatic alteration, where present, undeniably imposes the stable isotopic signature of the pegmatite-derived fluid on the host rocks (Taylor *et al.* 1979). Taylor *et al.* (1979) also noted that the border zones of pegmatites record influx of externally derived components (*i.e.*, the isotopic signature of the hosts). Influx of methane from host amphibolite is evident in fluid inclusions from the border zone of the Tanco pegmatite (Thomas & Spooner 1988). Thus, the exchange of components between pegmatite and host is not strictly bimetasomatic, but sequential, with initial influx of fluid components and solutes from hosts to pegmatite-forming magma, and subsequent outflow of pegmatite-derived fluids at the end-stages of pegmatite consolidation. London (1990a) utilized the occurrences of tourmaline in and around pegmatites to augment this argument. The formation of tourmaline-rich alteration aureoles is promoted by the introduction of a B-rich pegmatitic fluid into sufficiently ferromagnesian hosts. Tourmalinization, which proceeds rapidly, is localized at the site of mixing, *i.e.*, in the wallrocks. The inverse process appears to operate at the inception of pegmatite crystallization, wherein comb-structured concentrations of tourmaline (in some cases 80 vol.% of the border zone) radiate from the margins of the pegmatite toward its interior. At the early stages of crystallization, a B-rich pegmatite is open to infiltration of ferromagnesian components from the wallrocks, and again tourmaline deposition occurs at the site of mixing within the pegmatite-forming magma. Numerous occurrences of this sort could be cited, at which tourmalinization or any other alteration of mafic hosts is entirely absent next to tourmaline-rich marginal zones. It cannot easily be proven that H_2O infiltrates along with Fe–Mg from the hosts to pegmatite, but the isotopic signatures of border zones of pegmatite bodies are consistent with this hypothesis. The implication is that pegmatite-forming magmas are volatile-undersaturated, which creates a chemical potential gradient large enough to promote infiltration of host-rock-derived fluid components into the magma. Perhaps more importantly, if B-rich pegmatite-forming magmas are vapor-saturated upon emplacement, then it is inconceivable that such magmas could be open to infiltration from host rocks without leaking B-rich vapor to their surroundings. Yet as noted above, metasomatic tourmaline is mostly lacking along contacts between reactive host-rocks and B-rich pegmatites, except where demonstrably late-stage pegmatite fluids have been expelled into the host rocks.

Experiments by Morgan (1988) have shown that the isothermal–isobaric solubility of H_2O in granitic melt increases by approximately 1.7 moles of H_2O per mole of B_2O_3. Consequently, the crystallization of abundant tourmaline in a melt at H_2O saturation can liberate a large molar quantity of aqueous vapor (London 1986b). Someone asked me, several years ago why, if such large molar quantities of H_2O are given off by that reaction, is there no evidence of fluid infiltration into host rocks and only rarely are miarolitic cavities developed adjacent to tourmaline-rich border zones? In my opinion, the pegmatite-forming magmas are sufficiently H_2O-undersaturated that the H_2O released by tourmaline crystallization is still insufficient to bring the bulk magma to saturation.

There is ample evidence that most if not all pegmatite-forming magmas become H_2O-saturated near their solidi. Unless all magmatic H_2O is bound by hydrous phases, then the magmas must become H_2O-saturated at their minima or else pass continuously (supercritically) into the field or state of an aqueous fluid phase. If the arguments above are valid, however, then the evidence for early saturation of pegmatite-forming magmas in H_2O is hardly convincing.

Consequences of H_2O saturation

What are the effects of melt saturation in an aqueous fluid of low chlorinity (Burnham & Nekvasil 1986, London *et al.* 1988, 1989)? The potential effects on silicate crystallization kinetics have already been discussed. Once a magma is H_2O-saturated, there is no further depolymerization of melt nor change in nucleation or growth behavior of crystals as a function of changing activity of H_2O. When crystallization occurs by dissolution of melt through a vapor interface, as suggested by Jahns (1982), the resultant textures upon crystallization are uniform in size, shape, and distribution from the beginning to the end of crystallization (London *et al.* 1989).

The congruency of element partitioning between melt and vapor has been partly considered above in the context of the uniform K-numbers of fluid and coexisting melt. In the experiments of London *et al.* (1989) with an evolved peraluminous composition, all of the major-element components of melt dissolved congruently into vapor, including Al, such that vapor saturation promoted insignificant fractionation of major elements.

The exsolution of H_2O from melt is slightly exothermic (Burnham 1979), so that the buoyant ascent of aqueous vapor may create a thermal profile of increasing temperature from the lower to upper portions of the magma chambers (Burnham & Nekvasil 1986). In such a thermal gradient, the coexistence of albite and K-feldspar through an interconnected aqueous vapor phase (Jahns 1982) should promote redistribution of Na and K along the thermal gradient such that albite is enriched in the hotter, upper portions, and K-feldspar in the cooler, lower portions of the intrusive body (*e.g.*, Orville 1963). This, too, is contrary to field observations, and represents an important but overlooked contradiction to the aplite–pegmatite model of Jahns & Tuttle (1963).

Virtually all minor and trace lithophile elements display vapor/melt distribution coefficients of less than 1 (for many elements, near zero) in peraluminous systems of appropriately low chlorinity of fluid in the pressure range of 200–400 MPa (London *et al.* 1988, Webster *et al.* 1989). Thus, rather than "scouring" trace elements from melt (Jahns 1982), reasonable aqueous fluids in pegmatites may become depleted in trace elements relative to the coexisting magma. Among a wide variety of lithophile trace elements studied, London *et al.* (1988) observed that only boron consistently partitions in favor of the vapor phase. They noted that early separation of an aqueous vapor would fractionate boron from the wide range of melt-compatible trace elements (*e.g.*, Be, Ta) with which tourmaline is normally associated. If vapor/melt distribution coefficients for trace lithophile elements were much greater than 1, then vapor saturation at the inception of pegmatite crystallization would progressively deplete residual melt in incompatible components. The result would be an inversely zoned pegmatite, in which the early-formed units appeared to be most chemically evolved, and the latest unit were depleted in trace elements. Pegmatites, however, show normal trends of fractionation (increasing abundances of incompatible components), both within individual bodies and among cogenetic pegmatites.

As the equilibrium melt–vapor model of Jahns & Burnham (1969) does not account for the mass transfer necessary to explain the internal zonation of pegmatites, Burnham & Nekvasil (1986) suggested that "vigorous convection" of pegmatite melt plus vapor may aid the segregation of components by selective deposition. In consideration of the comparatively high viscosity of the melt and commonly narrow geometry of dikes, however, Jahns (1982) deemed convection unlikely. Even with the proposed convective transport of components, Burnham & Nekvasil (1986) concluded that the melt–vapor equilibrium model could not account for a magmatic origin for quartz-rich cores, and suggested that a large fraction (60%) of the silica in quartz-rich cores must be hydrothermal in origin. Although some late-stage redistribution of quartz is entirely possible, it is not apparent how such large volumes of silica could be transported within a closed system, nor what the driving chemical force would be for such remobilization of silica. Using data on quartz solubility from Fournier *et al.* (1982), applied to a low-chlorinity (<2 wt% Cl) aqueous fluid with a minor CO_2 component (London 1990a), a volume ratio of fluid to final rock (quartz) in excess of 10^3 would be required to transport and deposit a sufficient quantity of silica to form a typical quartz "core" body (representing 20–25 vol.% of the total pegmatite mass). Moreover, *all* of the silica transported by such a fluid would have to be deposited in the quartz body. The field and isotopic data (*e.g.*, Taylor *et al.* 1979, Taylor & Friedrichsen 1983) are not consistent with an open-system, hydrothermal influx of externally derived solutes, nor of fluid/rock volumes anywhere near this value.

Zoning by disequilibrium fractional crystallization

A point made earlier is that crystallization proceeds only when and to the extent that a system is perturbed from equilibrium. One can describe equilibrium crystallization, therefore, as a condition in which the perturbation is small relative to the response time of the system, such that at any given time in the cooling history of a magma, the assemblage of crystalline phases and melt are always close to their equilibrium proportions and compositions. For solid crystalline solutions, this requires that the compositions of the crystalline phases change (*e.g.*, by internal diffusion to promote homogenization) such that the activity of a given component anywhere in the crystalline phase is always the same and equal to the activity of that component in melt at a given set of state conditions. Another important point, known to students of petrology, is that equilibrium crystallization of H_2O-saturated eutectic melts produces no change in the assemblage of crystalline phases, compositions of individual phases (including that of melt), or proportions of crystalline phases. Whereas these facts of equilibrium crystallization of near-eutectic granitic melt plus aqueous vapor impose constraints that cannot be readily applied to the formation of pegmatites, there are no such restrictions on sequential fractional crystallization. Whereas the composition of eutectic melt does not change during equilibrium crystallization, sequential fractionation in multicomponent systems, even of a eutectic composition, can produce large variations in composition of residual melt. There occurs a corresponding variation in the compositions of individual crystalline solid-solutions, which is manifested as chemical zonation within an individual crystal, and as variations in the average composition of the phase at different stages of fractional crystallization. These facts are inherently appealing for an understanding of pegmatites, as these rocks record extreme fractionation of melt during their formation, and crystalline solid-solutions normally possess compositional zonation within individual crystals and among crystals of the same phase in different assemblages. It is obvious why the model of sequential fractional crystallization appealed to U.S. petrologists involved in the wartime study of pegmatites (*e.g.*, Cameron *et al.* 1949). Such a process could at once relate, albeit intuitively, both the textural and chemical development of zoned pegmatite bodies.

Jahns (1982) correctly noted that the crystallization of supercooled, H_2O-undersaturated granitic melts produces mineral zoning by sequential growth, and that the zoning patterns mimic those of pegmatites. This zonation has been shown again, and in more striking form, in experiments with a chemically complex pegmatite composition (Fig. 8). The sequential evolution of grain sizes, from relatively fine-grained granitic borders, porphyritic intermediate zones (megacrystic K-feldspar), and uniformly megacrystic interior units, also follows from the changes in melt composition that attend fractional crystallization (but not equilibrium crystallization of eutectic compositions). Concomitant with the variation in grain size is an associated change in fabric. The greater the degree of disequilibrium between melt and a (stable) crystalline assemblage, the greater is the tendency for oriented (comb-structured) surface nucleation over internal nucleation. Habits of individual crystals also evolve to more graphic and skeletal forms with increasing disequilibrium caused by undercooling. Thus, the development of comb-structured fabrics and associated graphic, branching, or radial habits of crystals is a measure of the degree of disequilibrium between melt and crystalline phases.

Boundary-layer effects and constitutional zone refining

A potential consequence of rapid growth of crystals, especially in viscous silicate liquids, is the development of a boundary layer of excluded components in the immediate vicinity of the front of crystal growth. In some instances, the rapid accumulation of excluded components may promote local saturation of a second phase at the margins of the growth front. Such is the explanation offered by Fenn (1986) for the origin of graphic intergrowths of quartz in K-feldspar. An alternative consequence is that an increase of excluded components that act as fluxes may cause crystals in the vicinity of the growth front to be resorbed into melt. In silicate crystal–liquid systems, this process of remelting by increased fluxing (and consequently lower temperatures of the liquidus) is equivalent to the metallurgical process of constitutional zone-refining (*e.g.*, McBirney 1987).

Constitutional zone refining is thought to contribute to the development of monomineralic layering in mafic intrusions (*e.g.*, McBirney 1987).

This hypothesis is built on models of the inferred cooling history and properties of silicate melts. Concrete natural evidence for this process might be obtained from careful analysis of volcanic glass away from the margins of spongy or skeletal phenocrysts, but I am not aware of any such studies. Experimental evidence in support of this process, however, has been observed in the relevant study of Macusani glass by London *et al.* (1989). They reported compositional gradients in glass extending to more than 150 μm away from K-feldspar megacrysts. Within this boundary layer, which is devoid of other crystals, concentrations of H_2O and B_2O_3 were found to be 15% higher than in the otherwise homogeneous glass beyond the boundary layer. Contrary to expectation, the K/Na ratio of glass increased toward the margin of the K-feldspar crystal.

As consequences of the model of rapid fractional crystallization described above, the combined effects of boundary-layer accumulation and constitutional zone-refining are particularly attractive to pegmatite formation. Within a geologically reasonable range of undercooling below the liquidus, Fenn (1977) demonstrated that nucleation maxima for sodic feldspar are higher than for potassic feldspar within the undercooling regions of maximum rate of crystal growth. The same is true for quartz, which exhibits high nucleation densities in experimental studies (*e.g.*, Swanson & Fenn 1986, London *et al.* 1989). In an undercooled pegmatite-forming melt, therefore, high nucleation-densities of albite and quartz would promote fine-grained products, and rhythmic alternations could result by the same process of boundary-layer saturation suggested by Fenn (1986) for graphic granite. Even though the concentrations of H_2O, B, P, and F might be low in bulk magma, they could be substantially concentrated as components excluded by rapid growth of crystals (*e.g.*, of feldspars and quartz). Their accumulation in advance of crystallization fronts would locally lower liquidus temperatures, causing resorption of crystals within the advancing boundary-layer, and inhibit further nucleation of stable phases by their interaction with crystal-forming components of melt. Rising concentrations of the fluxing components also may aid in the diffusion of K away from the advancing front of the crystal, so that boundary-layer saturation in K-feldspar was not attained. The result of this scenario would be that alternating fine-grained layers of albite and quartz would advance into the melt until the effects of constitutional zone-refining and disruption of melt structure by fluxing components would inhibit any further crystallization. At this point, the melt adjacent to the crystallizing front is grossly oversaturated in K-feldspar. The unexpected K/Na gradient in glass (increasing toward the K-feldspar) cited above from the work of London *et al.* (1989) may be a vestige of the accumulation of K in the boundary layer of the melt, generated by the initial advance of plagioclase + quartz. Crystallization at the growth front would recommence when sufficient quantities of fluxing components had diffused away from the boundary layer into the residual magma. Because of oversaturation, K-feldspar would be the dominant or exclusive phase to crystallize. Inherently low nucleation densities for K-feldspar, suppressed even further by the accumulation of fluxing components, would restrict this mineral to very low nucleation-density, and consequently to a very few, large crystals. Quite commonly, the megacrystic K-feldspar along the interface with albite-rich aplites in pegmatites is graphically intergrown with quartz, which suggests that the pegmatite system is still far from equilibrium at this stage and that crystal growth is rapid (Fenn 1986). The transition from layered albite–quartz aplites to graphic K-feldspar – quartz intergrowths, therefore, can be regarded merely as a change in feldspar mineralogy caused by the depletion in Na and increase in K in the boundary layer in the melt as the growth front proceeds inward. All other features of these texturally distinct pegmatitic units (layered albite-rich aplites *versus* comb-structured, megacrystic K-feldspar units) follow from the effects of rapid crystallization and the consequent imposition of boundary-layer effects. Because K-feldspar dominates the crystallizing assemblage in the K-enriched boundary-layer, silica is further concentrated in the diminishing volume of residual melt.

Quartz-rich zones and cores

For pegmatite systems, one of the more important consequences of recent experimental work pertains to the effects of B, P, and F on displacement of the quartz–feldspar field boundary toward albite [see the review by London (1987), and new data of London *et al.* (1990b, 1992)]. London (1987, 1989, 1990a, London *et al.* 1989) stressed the importance of the divergence in compositions of residual melt and crystalline phases with increasing concentrations of B, P, and F. The melt composition becomes increasingly alkaline, sodic, and silica-depleted as quartz-rich assemblages are deposited. In the H_2O-undersaturated experiments of London *et al.* (1989), residual pools at 85–90% crystallization were found to contain approximately 60% quartz, 5% mica, and 35% residual melt (*e.g.*, Fig. 8A). The crystalline silica produced in these experiments is actually a metastable solid-solution approaching virgilite, approximately $LiAlSi_5O_{12}$. Thus, the assemblage of the central portions of experiments was equivalent to the quartz - lithium aluminosilicate zones (petalite or spodumene as stable phases) that are common features of Li-rich pegmatites (*e.g.*, Stewart 1978). London *et al.* (1989) emphasized that these quartz-rich central zones of the experiments, including the formation of feldspar-absent quartz–mica–(LiAl silicate) assemblages, were generated by direct crystallization from melt in the absence of an aqueous vapor phase.

These experimental results help explain the formation of quartz-rich zones and cores as closed magmatic systems in pegmatites, without any tenuous or unreasonable requirements of vigorous convection, incongruent partitioning of elements and selective deposition from aqueous vapor, large aqueous vapor/melt ratios, or hydrothermal influx from external sources. Crystallization fronts of pure quartz develop over and sequentially after coarse-grained K-feldspar in hydrous but H_2O-undersaturated experiments that are significantly undercooled (*e.g.*, Fig. 8C). Despite the general success of the experiments by London *et al.* (1989) in producing quartz-rich central cores, and the intuitive appeal of the fractional crystallization model discussed above, it has not yet been demonstrated experimentally that similarly quartz-rich zones can be produced in melts of less-evolved composition. Attempts to replicate these features of natural pegmatites may be hindered by some of the scaling problems inherent in experiments.

The origin of bodies of massive quartz is made more problematic by excellent exposures in the Tanco pegmatite. There, bent and convoluted thin layers of albite-rich aplite that have been detached from the upper portions of the pegmatite are clearly suspended in massive quartz, and form a loosely associated rubble of bent layers where they came to rest on the footwall aplite (Fig. 9). Some unsupported layers curled into downward-pointing tear-drop shapes. All gaps between and created by the gentle disruption of aplite layers are filled by pure massive quartz. To those who have viewed these dramatic exposures, there is little doubt that the aplitic layers settled slowly and were suspended in a viscous fluid that is now pure quartz. These exposures make a hydrothermal origin for the massive quartz implausible, as such a low-density, low-viscosity fluid would be incapable of supporting the mostly crystalline aplitic layers. There is no indication that the aplite layers are draped around the shapes of pre-existing quartz crystals, either. These smooth, convolute contacts and suspended rafts of aplite in quartz are suggestive of contacts between two (immiscible) fluid phases. Although there is no problem generating the appropriately sodic melt at these low temperatures, the liquid analog to the massive quartz is difficult to explain. Although the experimental work of London *et al.* (1989) shows that quartz-rich bodies can crystallize from melt, there is no provision for a silica liquid of the type envisioned here. Furthermore, the suggested process of liquid–liquid immiscibility at Tanco hinges on the high concentration of boron and other incompatible components in these most fractionated melts (London 1986a). This would not be the case for compositionally simpler pegmatites that manifest many of the same textural features in and around masses of pure quartz.

Formation of rare-element orebodies

The accumulation of B, P, and F in melt promotes a sodic, alkaline liquid line of descent with increasing solubilities of H_2O and incompatible lithophile elements (London 1989, 1990a, London *et al.* 1989). In the experiments of London *et al.* (1989), residual melt at 85% crystallization, 200 MPa, were found to contain 15–20 wt% H_2O and still to be undersaturated. Trace-element abundances in this H_2O-undersaturated melt were generally an order of magnitude higher than in the H_2O-saturated system at the same degree of crystallization (Table 8 in London *et al.* 1989). The high concentrations of fluxing components and low temperatures precluded crystallization of the last 10–15% of melt (*i.e.*, the lag time for crystal nucleation increased to greater than the 4–6 month durations of these experiments) except in one experiment. In this unique experiment (Figs. 8D, E), the heterogeneous residual melt partially crystallized as extremely fine-grained lepidolite + albite (including apatite and presumably other accessory phases), and as such represents the latest-stage internal units of pegmatite that may be derived from sequential fractional crystallization, rare-element-enriched albite–mica aplites. There is therefore excellent agreement in the compositions of these latest-stage silicate liquids in experiments (London *et al.* 1989) and natural fluid inclusions (London 1986a), and in the correlation of these liquids with mineralized albite–mica bodies.

THE MAGMATIC–HYDROTHERMAL TRANSITION IN PEGMATITES

Eventually, all pegmatitic systems evolve an aqueous vapor phase. In views previously stated (London 1990a, London *et al.* 1989), this occurs very near the solidus of most pegmatites, or as they approach complete crystallization. For compositionally simple pegmatites, the miscibility gap between residual silicate melt and aqueous vapor must be quite large. In chemically evolved pegmatites, of which Tanco is but one representative, the miscibility between H_2O and fractionated silicate melt is extensive, so much so that the transition from magmatic to hydrothermal conditions may be continuous (supercritical) if it is not interrupted by the loss of components such as B, P, and F to late-crystallizing phases. Using mostly fluid-inclusion data, London (1986b) found support for a continuous transition from magma to aqueous vapor in miarolitic pegmatites of Afghanistan. In contrast, analyses of fluid inclusions show an abrupt change in the latest fluids present in the Tanco pegmatite, from a hydrous borosilicate fluid with complete H_2O miscibility to a low-density carbonic aqueous vapor (London 1986a).

The attainment of saturation in an aqueous vapor late in the history of pegmatite consolidation is indicated not only by the presence of primary miarolitic cavities, but also by subsolidus metasomatic replacement of pre-existing minerals. These are mostly pseudomorphic in nature; they are sporadic but most extensive in the most fractionated pegmatites, and the replacement assemblages are defined by the very local chemistry of the mineralogical zone in which they occur (*e.g.*, London & Burt 1982a, b, Burt & London 1982). The first stage of subsolidus metasomatic alteration is usually alkaline in chemical character (*e.g.*, London & Burt 1982a, b, Burt & London 1982). This chemistry of the first metasomatic fluids can now be explained as the excess alkaline components of melt that are not conserved by the crystallization of albite or albite–mica bodies (London *et al.* 1989, London 1990a). Alkaline metasomatic reactions are normally followed by a weakly acidic, potassic stage in which feldspars, lithium aluminosilicates, and montebrasite are replaced by sericitic micas. Sericitic alteration of feldspars liberates, among other components, phosphorus that may produce a diversity of secondary phosphates in fractures (London 1992). The conditions of subsolidus alteration, given by the local association of zeolites, sulfides, or clay minerals, are in the greenschist to zeolite facies of metamorphism.

WALLROCK ALTERATION

Alteration of host rocks to pegmatites is perhaps not so critical to pegmatite formation itself as it is to understanding how pegmatites form. Several points about wallrock alteration have already been noted. One is that pegmatite-induced alteration, whether in response to changing pressure, temperature, or fluid composition, is not prevalent around most pegmatite bodies. It is restricted mostly to evolved types of pegmatite, and is sporadic in its distribution. A second point is that the alteration assemblages normally indicate greenschist to zeolite metamorphic facies, as do minerals of metasomatic alteration within pegmatites. A third and perhaps most important point is that the chemistry of metasomatic assemblages in host rocks mirrors the composition of the innermost units of the adjacent pegmatite, indicating that fluids are expelled from pegmatites after they have fully crystallized, and that these fluids must have interacted with outer zones (*e.g.*, pegmatitic wall and border zones) as they passed through. This last observation makes it unlikely that any unit within a pegmatite fully preserves its original composition (elemental or isotopic).

There are many studies of the mineralogy of wallrock alteration around pegmatites. These have been pursued for scientific value, and as a guide to exploration for concealed pegmatite-related ores. A few important investigations pertain to stable and radiogenic isotope systematics (*e.g.*, Brookins *et al.* 1969, Taylor *et al.* 1979, Taylor & Friedrichsen 1983, Walker 1985) and the flux of fluids and solutes (*e.g.*, Shearer *et al.* 1986). Morgan & London (1987b) related the conditions of wallrock alteration to processes occurring within the Tanco pegmatite. The results of their study, summarized in Figure 10, revealed an excellent correspondence between the chemistry and conditions of wallrock alteration and internal processes within the pegmatite.

Experimental solutions to phenomena of pegmatite – wallrock alteration have been applied only by Morgan & London (1989). They considered the mechanism by which amphibolite, the host to the Tanco pegmatite, and gneissic compositions (biotite + plagioclase + quartz) can be converted to nearly pure tourmaline or tourmaline + quartz. This work showed that tourmaline stability is markedly dependent on fluid pH. Tourmaline was found to be unstable above pH values of the fluid near 5.5–6.5, regardless of the boron content of fluid. Hence, the alkaline chemistry of pegmatitic fluids and residual melts, in addition to low Fe–Mg content, precludes deposition of late-stage tourmaline within this or other similar pegmatites (*e.g.*, Harding, New Mexico: London 1986c). Through these and previous experiments (Morgan & London 1987a), coupled with mass-balance calculations (Morgan & London 1987b), it was shown that the conversion of amphibolite to tourmaline requires addition of Al (Fe and Mg are mostly conserved within the altered rock), and that Al is particularly soluble in aqueous borate fluids. At pH values of less than 5.5, amphibolites are very reactive with borate fluids, and the equilibrium of tourmaline + hornblende + plagioclase buffers fluid compositions at less than 1 wt% B_2O_3. Through this experimental study, Morgan & London (1989) were better able to constrain the mass balance of boron loss from the pegmatite. Almost 80% of the

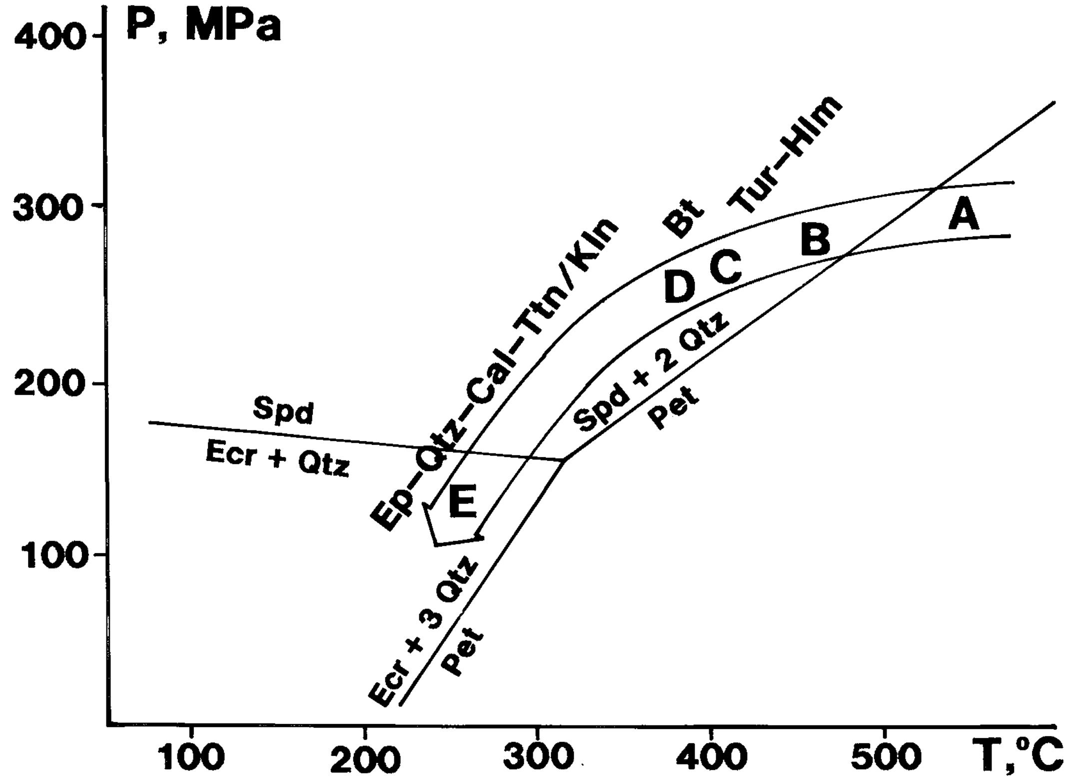


FIG. 10. Comparison of wallrock alteration of amphibolites to internal evolution of the Tanco pegmatite, Bernic Lake, Manitoba. The arrow denotes the P–T path of the pegmatite as deduced from analysis of fluid inclusions with reference to stability relations of lithium aluminosilicates (London 1984, 1986a). Phase abbreviations for Pet, Spd, Ecr, and Qtz are as in Figure 4; other phases are: tourmaline (Tur), holmquistite (Hlm), epidote (Ep), calcite (Cal), titanite (Ttn), and kaolinite (Kln). Late-stage sodic melt enriched in Li and B was present during the crystallization of petalite and spodumene (A → B). The magmatic products of this crystallization were aplitic rocks consisting mostly of albite and rare-alkali-enriched micas. This residual melt crystallized (*i.e.*, disappeared) in the narrow interval of temperature from B to C, with the exsolution of a low-chlorinity CO_2-bearing aqueous fluid. The excess Li and B that were not conserved during crystallization of the late-stage sodic melt were conveyed through the fluid to the wallrocks, where metasomatic reactions produced tourmaline plus holmquistite. Wallrocks record the influx of this fluid at about 425°C (garnet–biotite thermometry: Morgan & London 1987b). Between C and D, the carbonic aqueous fluid unmixed to separate H_2O- and CO_2-rich phases; sericitic alteration of microcline within the pegmatite provided the much of the K, Rb, and Cs that were incorporated as metasomatic biotite in the wallrocks. Between D and E, fluid inclusions within the pegmatite recorded an influx of wallrock-derived components. Concomitant alteration of wallrock produced a wide aureole of propylitic (Ep–Qtz–Cal–Ttn) and argillic alteration (represented by Kln, but the actual clay mineralogy is not known). With the buoyant ascent of the less dense CO_2 fluid phase through the pegmatite, calcite (Cal) became more prevalent in the altered amphibolite of the hanging wall as compared to the footwall. Figure from Morgan & London (1987b).

original boron content of the Tanco pegmatite was lost to the host rocks, and of this, probably not more than 60–70% was incorporated as tourmaline within the adjacent amphibolite.

CONCLUDING REMARKS

It is perhaps ironic that the experimentally based model presented here and in previous papers (London *et al.* 1988, 1989, London 1990a) is indistinguishable from the views held by Jahns (1953) before he began his experimental studies. Jahns's particular genius was his powerful insight derived from extensive field-based knowledge, and it is this aggregate of experience and understanding that formed the basis of his early model. Jahns (1953) concluded that the zonal features and progressive coarsening of pegmatite minerals could be "attributed to fractional crystallization and incomplete reaction between successive crops of crystals and rest-liquid...All known features of pegmatites seem reasonably explainable on the basis of crystallization from a silicate melt of low viscosity, with or without end-stage deuteric or hydrothermal activity. Many of these features also suggest that masses of pegmatite crystallized inward from the walls of an original chamber, under restricted-system conditions, rather than in some sort of channelway or thoroughfare under more open-system conditions...nearly all the giant crystals were formed during what has been designated by most investigators the primary, or magmatic, stage of pegmatite development, and hence by crystallization from liquid under conditions that permitted the remarkable growth of a relatively few individuals... The [giant] crystals are thought to have formed rapidly under restricted-system conditions involving rather delicate temperature and chemical balance. Temperatures were almost certainly below 600°C, and the confining pressures were sufficiently great to prevent major escape of volatile constituents during the period of giant-crystal development."

As he began experimental studies of pegmatite genesis, Jahns embraced the increasingly favored model of his time, equilibrium crystallization, in place of disequilibrium fractional crystallization. Jahns also recognized that the compositions of granitic pegmatites approach those of the haplogranitic eutectic at moderate pressures of H_2O. The constraints of a model for equilibrium crystallization of a eutectic composition forced Jahns into several quandaries, particularly the simultaneous (as opposed to sequential) development of segregated and texturally distinct zones of albite, K-feldspar, and quartz in pegmatites. Jahns (pers. comm., 1979) recognized that the textural and zonal features of pegmatites were inconsistent with equilibrium crystallization at the eutectic of haplogranitic melts, which required another agent, aqueous vapor, to accomplish what magmatic crystallization apparently could not do. So strong was this concept that I believe it led Jahns to misinterpret the causal relations of vapor saturation to textural development in the famous experiment cited by Jahns (1982) and Wyllie (1963). Recent studies (*e.g.*, London *et al.* 1988, 1989) have at least demonstrated that there is no unique causal relation between an aqueous phase and the textural transition from granitic to pegmatitic texture in experiments. Some experimental evidence (*e.g.*, Fenn 1977, London *et al.* 1988, 1989) also indicates that saturation in an aqueous phase may be counterproductive to forming pegmatites.

Whether an aqueous phase is actually present during the early stages of crystallization in most pegmatites is a separate and more contentious issue. There is good evidence from petrological and fluid-inclusion studies that pegmatites do not attain saturation in an aqueous phase until they approach their solidi. This hypothesis requires that the larger igneous masses (granites) from which pegmatites are derived are also H_2O-undersaturated at the time of withdrawal of the pegmatite-forming magma, which is implicit in the Jahns–Burnham model: granites themselves are the products of H_2O-undersaturated crystallization, and granites that lack pegmatites represent magmas that did not achieve vapor saturation in the presence of silicate melt.

The genetic link between granites and pegmatites is very poorly understood, despite the obvious relevance to the origin of granite, and to the evolution of silicic magma chambers in general. Granite–pegmatite relations have been studied by chemical trends (*e.g.*, Goad & Černý 1982, Černý *et al.* 1985, Černý 1991a, b) but not in terms of process. In total, the characteristics of most pegmatite groups suggest a single main pulse of magma injection from the cupolas of parental granites, followed by sporadic closure of dike conduits and subsequent internal (quasi-closed system) differentiation of restricted bodies of pegmatite (London 1990a). If pegmatite-forming magmas are produced by extensive fractionation of crystals from larger granitic sources, then their source granitic plutons must crystallize from the roots upward, and pegmatite-forming magmas, which appear to have been entirely liquid upon emplacement, must originate by extraordinarily efficient separation of silicate liquid from crystals. The field relations imply that much of the chemical differentiation within a pegmatite group is present at the time of magma emplacement, and hence is inherited in the liquid state from the source body of magma (Černý 1982a). This hypothesis suggests that granite–pegmatite relations should be viewed as part of the general study of vertical chemical zonation in silicic magma chambers, which is inverted on eruption of rhyolites (Hildreth 1981), but is preserved without inversion in granite–pegmatite systems. Not only is such chemical fractionation more common in pegmatite systems than in rhyolites, but it reaches extremes that are not found in most erupted magmas.

In summary, it may be surprising how little experimental petrology has contributed to unraveling the complex origins of pegmatites. Certainly progress has been made, and the experimentally and theoretically based model of disequilibrium fractional crystallization presented here, which reaffirm Jahns's original concepts (Jahns 1953), may answer many questions of texture and zonation that were not explained by the Jahns–Burnham model. However, numerous fundamental questions remain. It has not yet been demonstrated that absolutely pure quartz cores can be derived by closed-system crystallization of simple haplogranitic melts (with or without an aqueous vapor phase). In addition, chemically evolved pegmatites commonly possess a chaotic zonation, so much so that is it difficult to relate their crystallization to any existing model. The segregation of several thousand cubic meters of pollucite ($CsAlSi_2O_6$) in a few pure, pancake-shaped masses within the huge Tanco pegmatite (Černý & Simpson 1978) defies any explanation from known concepts of crystallization and mineral segregation. If there is hope for resolution of all features of the internal differentiation of individual pegmatite bodies, it will be through improved understanding of the dynamics of crystal nucleation and growth from silicate melts, the effects of bulk composition on changes in melt properties and crystallization, and of the conditions that pegmatite-forming magmas encounter upon emplacement.

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