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A DYNAMIC MODEL FOR GRAPHIC QUARTZ–FELDSPAR INTERGROWTHS IN GRANITIC PEGMATITES IN THE SOUTHWESTERN GRENVILLE PROVINCE

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ABSTRACT

During the formation of U-, Th-, Mo-, *REE*-, and Nb-bearing granitic pegmatites in the Grenville Province, conditions of crystallization were such that widespread graphic quartz–feldspar intergrowths were formed. The alkali feldspar - quartz intergrowth is characterized by angular rods of quartz that have an irregular inner interface and a planar outer interface with the host alkali feldspar. The quartz is interpreted to have nucleated epitactically on rough edges and corners of alkali feldspar crystals. Rapid growth, at or near volatile-saturated conditions, resulted in quartz saturation along the irregular melt–feldspar interface. Slow diffusion of Si and Al species (network formers) in the boundary-layer melt was likely the rate-controlling step for quartz saturation, which occurred along corners and edges, where the feldspar grew most rapidly. Diffusion-limited growth resulted in SiO_2 buildup at the interface, producing oscillations from quartz-oversaturated to quartz-undersaturated conditions and thus the rhythmic quartz–feldspar intergrowths. The transition from planar, to edge, to cellular growth, and changes in the lobate inner feldspar–quartz boundary occurred in response to changes caused by crystallization that affect rates of Si–Al diffusion. Evidence of saturation in a volatile phase in these pegmatites indicates that water was a catalyst for feldspar growth and that lower activities of H_2O in the melt decrease Si diffusivity at the crystal interface.

Keywords: graphic texture, granophyric, growth kinetics, diffusion, granitic pegmatite, quartz, feldspar, Grenville Province, Ontario, Quebec.

SOMMAIRE

Au cours de la formation de pegmatites granitiques enrichies en U, Th, Mo, Nb et les terres rares dans la province du Grenville dans le sud-est de l'Ontario, les conditions de cristallisation ont favorisé une intercroissance graphique de quartz et feldspath alcalin. L'intercroissance est faite de bâtonnets angulaires de quartz possédant des interfaces interne irrégulière et externe planaire par rapport au feldspath alcalin hôte. Une nucléation épitactique du quartz aurait favorisé les arêtes rugueuses et les coins des cristaux de feldspath alcalin. Une croissance rapide, sous conditions de saturation, ou de quasi-saturation, en phase aqueuse, aurait causé une saturation en quartz le long de l'interface irrégulière entre bain fondu et feldspath. Une diffusion lente des espèces porteuses de Si et Al, composants de la trame, à travers la couche de liquide près de l'interface, aurait régi le taux de saturation en quartz. Cette saturation a eu lieu aux coins et le long d'arêtes, où le feldspath a pu croître le plus rapidement. La croissance limitée par le taux de diffusion a mené à un enrichissement en SiO_2 à l'interface, causant des oscillations entre conditions de saturation et de sous-saturation en quartz, et donc une intercroissance rythmique de quartz et de feldspath alcalin. La transition d'une croissance le long de plans, à une croissance le long d'arêtes, à une croissance cellulaire, et les changements dans la morphologie de l'interface interne entre feldspath et quartz, ont eu lieu suite à des changements qui ont influencé les taux de diffusion de Al et Si. Des signes de saturation en phase volatile dans ces pegmatites indiquent que l'eau a agi comme catalyste pour la croissance du feldspath, et qu'une faible activité de l'eau dans le magma mène à une diminution de la diffusivité du Si à l'interface magma–cristal.

(Traduit par la Rédaction)

Mots-clés: texture graphique, granophyrique, cinétique de croissance, diffusion, pegmatite granitique, quartz, feldspath alcalin, province du Grenville, Ontario, Québec.

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Introduction

The graphic texture refers to the regular cuneiform intergrowth of two mineral phases, typically quartz and alkali feldspar. The granophyric texture is the irregular finer-grained counterpart of the graphic texture. Detailed textural, petrographic, and chemical examination of twenty-five samples of graphic granite pegmatites from ten localities in the southwestern Grenville Province has provided insight into their formation.

Numerous hypotheses have been proposed for the formation of the graphic texture, but simultaneous growth of quartz and feldspar near the thermal minimum, originally proposed by Vogt (1931), is considered to be the controlling mechanism (*e.g.*, Barker 1970, Černý 1971, Hughes 1971). Replacement of feldspar by quartz-saturated solutions (Wahlstrom 1939, Drescher-Kaden 1948, Augustithis 1962, Seclaman & Constantinescu 1972) and vapor-phase crystallization (Simpson 1962) also have been proposed. Schloemer (1964) produced the texture by hydrothermal coprecipitation of quartz and feldspar. Lofgren (1971) also observed the texture by devitrification of rhyolitic glass in experimental charges. Maaløe (1974), Smith (1974), Walker (1976), Carstens (1983), Fenn (1986), and London *et al.* (1989) have proposed that the textures are formed near the cotectic or eutectic for the system but are not necessarily an equilibrium phenomenon. Fenn (1986) and London *et al.* (1989) experimentally produced the texture from granitic bulk compositions within the primary field of K-feldspar and concluded that the texture is related to the interplay of diffusion and growth kinetics.

Graphic and granophyric textures have been interpreted to result from rapid crystallization at volatile-saturated conditions (Mehnert 1968, Barker 1970, Smith 1974, Hibbard 1980, Martin 1982, Shannon *et al.* 1982, Černý & Meintzer 1988, Kirkham & Sinclair 1988). Both these textures occur within marginal facies of pegmatite, pegmatite pods, miarolitic cavities, aplites, and are also associated with hydrothermal mineralization.

This paper identifies morphological and compositional features of the intergrowths that support a diffusion-controlled model of growth. However, the growth dynamics differ from those proposed by Carstens (1983) and Fenn (1986) for granophyre and the graphic texture, respectively. Their mechanism involves short-range continuous diffusion of quartz- and feldspar-forming components to a crystallizing surface containing both minerals. Observation and interpretation of the lobate growth-boundaries between feldspar and quartz suggest that quartz nucleated on feldspar growth-surfaces. The effects of a dissolved and free volatile phase are examined because there is an empirical relationship between the development of the graphic texture and evidence of volatile saturation in these pegmatites.

Geological Setting

The U-, Th-, Mo-, *REE*- and Nb-bearing granitic pegmatites in the southwestern Grenville Province contain abundant examples of a well-developed graphic texture. In general, granitic pegmatites form both concordant and discordant dykes in the gneisses, marbles, and amphibolites. They can attain widths of tens of meters to hundreds of meters and have lengths up to several kilometers. Some of the larger pegmatites are associated with mineral deposits, for example the Faraday uraniferous granitic pegmatite in Bancroft, Ontario. Figure 1 illustrates the distribution of sample localities within the Central Metasedimentary Belt of the Grenville Province. Several of the samples of graphic granite pegmatites studied are hosted by mixed marbles, calc–silicate rocks, and paragneisses (samples 26, 43, 49, 52, 108 and 122), as this study forms part of a larger investigation of pegmatite-related skarns (Lentz 1992). The remaining samples (57, 105, 109 and 118) are from discordant pegmatites that cut biotite-bearing quartzofeldspathic gneisses. The local geology and setting of eight of these occurrences may be found in Lentz (1991b), and a brief description of the mineralogy and geology of the Blue Sea Lake pegmatite (122) and the Perth Quarry (118) may be found in Sabina (1987).

The granitic pegmatites under consideration are late tectonic (1020 to 1060 Ma, U–Pb zircon; Rimsaite 1982, Easton 1986) with respect to the Grenville Orogeny (*ca.* 1090 Ma; Easton 1986) and were emplaced into the Grenville rocks at pressures and temperatures less than peak metamorphic conditions (Lentz 1991b), which is compatible with their generally undeformed nature (Fowler & Doig 1983). $^{40}Ar/^{39}Ar$ cooling curves for the Bancroft terrane of the Central Metasedimentary Belt (CMB) suggest that the pegmatites were emplaced while temperatures remained between 500° and 550°C (Berger & York 1981), which is consistent with the development of mineral assemblages in the exogranitic skarns (Lentz 1991b, 1992). Pressures have been estimated to be between 300 and 500 MPa based on fluid-inclusion studies on the MacDonald zoned pegmatite (Peach 1951), and are consistent with the pressure – temperature – time paths presented by Martignole (1986). These conditions differ from those inferred for the peak of metamorphism (upper amphibolite facies; 650°C and 600 to 700 MPa) for the host rocks (Davidson 1986).

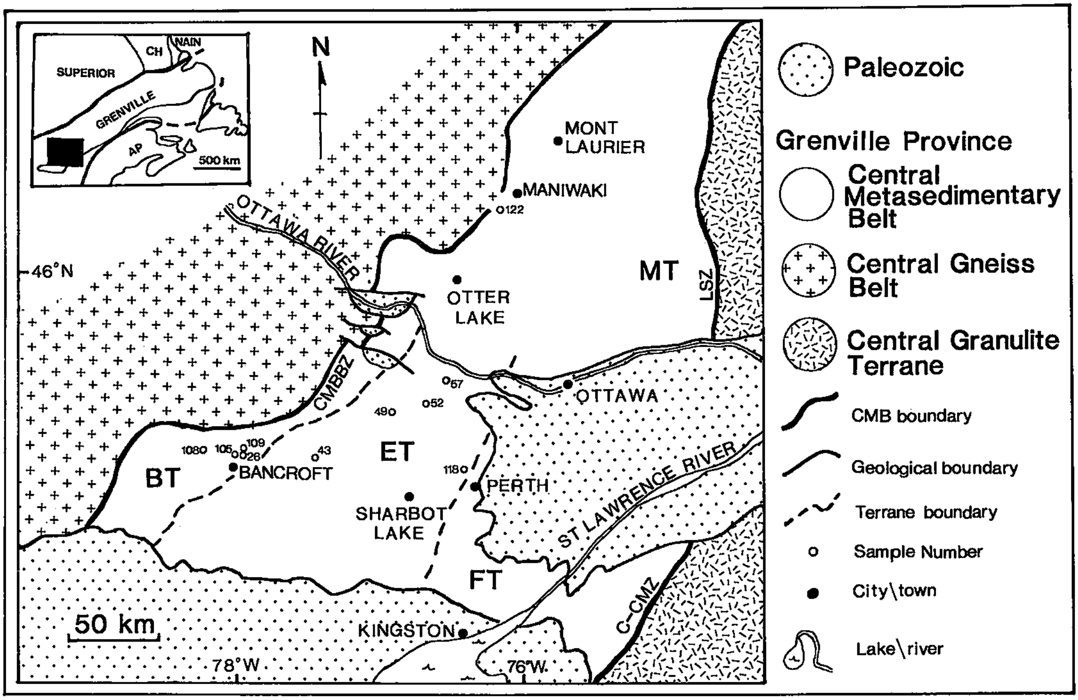

FIG. 1. Sample localities of granitic pegmatite displaying the graphic texture within the Central Metasedimentary Belt (CMB) of the southwestern Grenville Province (after Davidson 1986). BT Bancroft terrane, ET Elzevir terrane, FT Frontenac terrane, MT Mont Laurier terrane, CMBBZ Central Metasedimentary Belt Boundary Zone, C–CMZ Colton-Carthage Mylonite Zone, LSZ Labelle Shear Zone. Sample numbers and names: 26 MacDonald U-*REE*-Mo pegmatite, 43 Liedke Mo pegmatite, 49 Legris U-*REE* pegmatite, 52 Belanger's Corner granite, 57 Goshen "B" U-*REE* pegmatite, 105 McCormack U pegmatite, 108 Armac Mo pegmatite, 109 Genesse U pegmatite, 118 Perth U pegmatite, and 122 Blue Sea Lake amazonite pegmatite. Within the inset, the black square outlines the map; CH Churchill Province, AP Appalachians.

GRANITIC PEGMATITES

Granitic pegmatites are composed essentially of quartz, microcline and plagioclase, with lesser amounts of biotite, amphibole, calcic pyroxene, titanite, magnetite, zircon, allanite, calcite and fluorite. Field evidence suggests that some of the ferromagnesian phases, locally concentrated along the contact zone, originated by a complex process of hybridization or skarnification (Satterly 1956, Shaw 1958, Masson & Gordon 1981, Haynes 1986, Lentz 1991a, b). Partial melting of rocks in the Central Gneiss Belt during post-tectonic uplift produced H_2O-undersaturated, anatectic melts deep within the crust (Lentz 1991b); an origin in the lower crust or upper mantle for these melts has also been proposed on the basis of radiogenic isotope evidence (Fowler & Doig 1983). Fractional crystallization of anhydrous silicates (quartz and feldspar) during the ascent of these melts within the crust caused their enrichment in rare metal and volatile components, to the point of saturation (Lentz 1991b). In addition to H_2O, HF (< 1 wt.%), HCl, and alkali chlorides also were constituents of the melt and vapor phase, based on the presence of micas, fluorite and scapolite in and around the pegmatites (Lentz 1991b, 1992). Goad (1990) found that the feldspars in many of these pegmatites have trace-element abundances characteristic of rare-metal-bearing pegmatites. These feldspars crystallized at temperatures above 600° to 650°C, based on two-feldspar geothermometry (Sempels 1971, Pride 1974) and trace-element (Rb) partitioning between biotite and K-feldspar (Lentz & Kretz 1989). This temperature is consistent with that of the thermal minima in the haplogranite system; the phase relations are quite comparable to those for the Spruce Pine pegmatite at a pressure of 500 MPa (Fenn 1986, Strong 1988), although biotite is more prevalent than muscovite in the samples studied.

Pegmatite textures

Numerous textural variations occur within Grenville pegmatites, indicative of the variable conditions of crystallization. The pegmatites occur as both zoned and unzoned types (Hewitt 1967). Most bodies are unzoned and contain fine-grained aplitic (<1 mm) to coarse-grained pegmatitic rocks (tens of cm); however, the average grain-size is 1 to 3 cm. The crystals are dominantly subhedral and equigranular. In the zoned bodies of granite pegmatites, grain size varies from medium grained to very coarse grained (3 cm to >1 m). Ferromagnesian silicates are euhedral, whereas quartz and the feldspar are subhedral to anhedral, except in the core zone. Perthitic microcline is ubiquitous. Feldspar exsolution postdated the formation of the graphic texture, as the traces of exsolution lamellae intersect the intergrown quartz.

Description of graphic granite pegmatites

Storey & Vos (1981) have described the occurrence of the graphic texture in 40 of the 50 pegmatite deposits examined in the Bancroft–Renfrew area of Ontario. De Schmid (1916) and Spence (1932) noted that the mining of feldspar and quartz from many zoned pegmatites ceased after coarse- to fine-grained graphic granite pegmatite was encountered at depth. The graphic texture is extremely variable even on outcrop scale; grain sizes range from 2 mm to greater than 10 cm (Spence 1932, Hewitt 1967, Storey & Vos 1981). The reported grain-size of the graphic texture, the average distance between adjacent quartz domains, varies from fine (0.25 to 1 mm) to coarse (greater than 10 mm).

The quartz forms discontinuous plates, triangular rods, and angular corners (arrows), which impart the characteristic cuneiform texture in two dimensions (Figs. 2, 3). The graphic quartz has an anhedral (irregular) inner boundary, whereas the outer boundary is euhedral (planar). This observation indicates that quartz nucleated on the anhedral boundaries of the host feldspar and grew to a more euhedral form (Fig. 3). The cuspate protuberances are particularly obvious in the coarser-grained varieties of the graphic texture (Fig. 3b). In the finer-grained samples, quartz has a saw-tooth-shaped inner surface (Fig. 3e).

Surprisingly, the irregular nature of the inner surface of the quartz does not seem to have been documented. This may be in part due to the lack of preservation of the irregular interface (*e.g.*, Smith 1974). This relationship indicates that quartz nucleated onto an irregular feldspar interface and grew to a more regular (prismatic) outer surface. The formation of feldspar protuberances into the

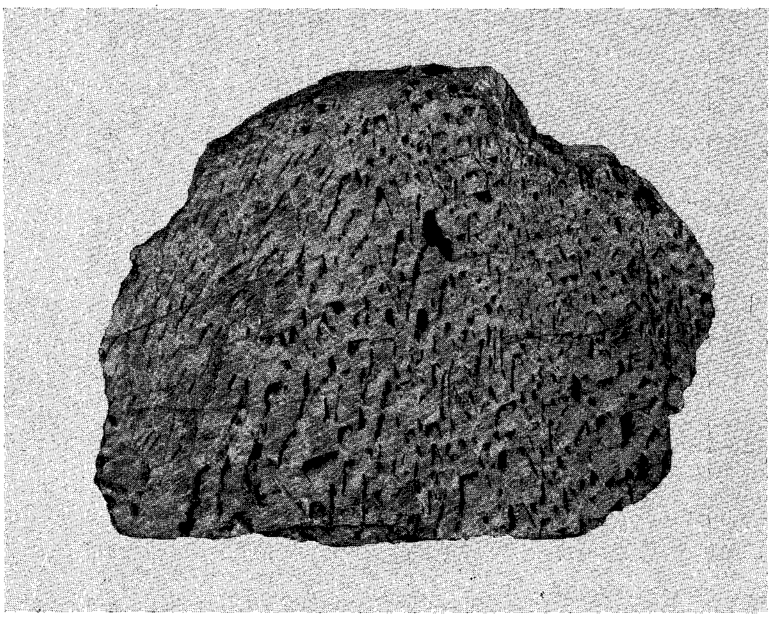

FIG. 2. Coarse-grained graphic granite pegmatite (unknown Grenville locality; 35 cm wide; GSC photo #204994).

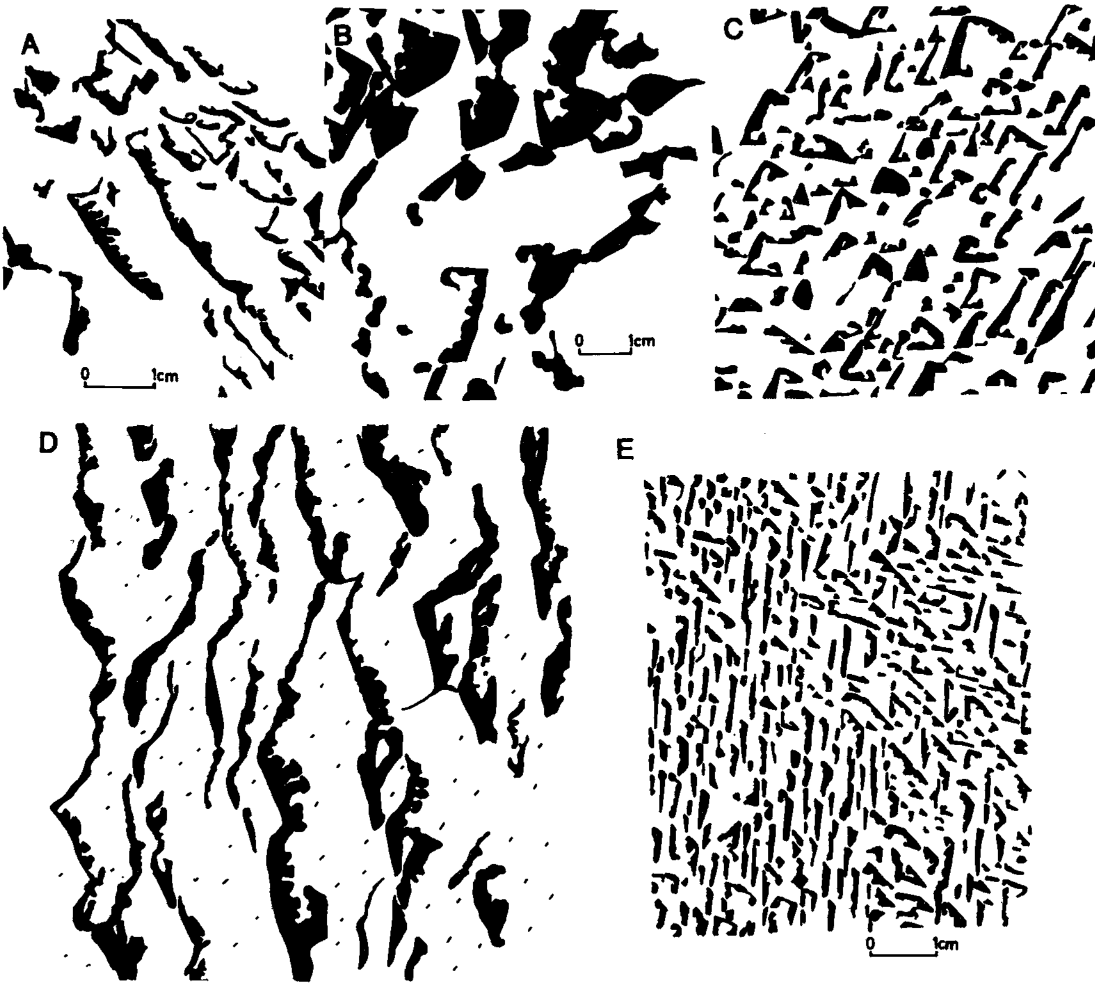

FIG. 3. A. Tracings of coarse-grained graphic texture with irregular lobate quartz–feldspar interface (parallel to edge) (sample #105–1). B. Sketch of coarse-grained graphic texture with faceted quartz–feldspar outer interface and irregular interface (perpendicular to edge or at corner (sample #57F). C. Sketch of coarse-grained graphic granite pegmatite from Hybla pegmatite field near Bancroft, Ontario (perpendicular to edge; Figure 10.81, Hurlbut & Klein 1977). D. Sketch of coarse-grained lobate-textured graphic granite pegmatite from Topsham, Maine (no scale) (Appalachian) (after de Schmid 1916). E. Sketch of medium-grained graphic granite pegmatite with sawtooth and barbed shaped inner quartz–feldspar margin (after de Schmid 1916).

melt is a result of a change from planar (equilibrium) to cellular growth (nonequilibrium; *e.g.*, Fenn 1986). The hook-shaped protuberances at the feldspar–melt interface (irregular inner Qtz–Kfs surface) have the appearance of screw-dislocation growth in side view. Quartz saturation did not occur until the outermost portion of the feldspar protuberances crystallized. The bulbous tips of the protuberances suggest that either 1) quartz nucleated in the embayments and overgrew the protuberances, or 2) the feldspar protuberances extended beyond the silica-enriched zone but were overgrown upon quartz saturation. *Note added in proof:* Stel (1992) documented the existence of rugose inner feldspar–quartz boundaries and euhedral outer boundaries as evidence that the graphic texture is a primary magmatic feature.

Composition of graphic granitic pegmatites

A comprehensive compilation of the chemical compositions of Grenville pegmatites and aplites

TABLE 1. COMPOSITION OF GRAPHIC GRANITE PEGMATITES IN THE SOUTHWESTERN GRENVILLE PROVINCE

Sample	52-1	52-2	26C	43E	49-1	49-2	49-3	49-4	49-5	49-6	49-7	57F	105-1
Texture	mg	mg	mg	fg	cg	fg	mg	cg	mg	mg	mg	cg	mg
SiO_2 wt.%	73.3	72.6	76.1	74.3	74.2	75.2	75.5	73.7	75.7	74.5	74.4	75.5	71.5
TiO_2	0.01	0.01	0.00	0.01	0.01	0.00	0.00	0.01	0.01	0.01	0.00	0.00	0.00
Al_2O_3	14.28	14.38	14.46	13.75	14.00	13.48	13.59	14.46	13.38	13.33	13.67	12.78	14.10
Fe_2O_3	0.07	0.10	0.17	0.12	0.18	0.20	0.14	0.15	0.14	0.19	0.21	0.23	0.40
MgO	0.24	0.21	0.1	0.14	0.00	0.01	0.12	0.07	0.06	0.08	0.21	0.14	0.28
CaO	0.17	0.09	1.64	0.19	0.11	0.10	0.07	0.09	0.09	0.09	0.07	0.66	0.11
Na_2O	2.24	1.79	6.23	2.86	2.72	2.73	3.01	3.24	2.89	2.81	2.94	2.71	2.50
K_2O	9.60	10.28	0.85	8.11	7.88	7.61	7.71	8.31	7.61	7.57	7.92	7.11	8.82
MnO	0.02	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.01	0.00	0.01	0.01	0.01
P_2O_5	0.00	0.01	0.00	0.01	0.02	0.02	0.01	0.00	0.01	0.03	0.00	0.00	0.01
Total	100.0	99.5	99.7	99.5	99.1	99.3	100.1	100.0	99.9	98.7	99.6	99.1	98.0
Ba ppm	70	80	90	480	490	430	440	460	460	440	330	1380	1050
Sr	17	61	234	177	383	319	344	383	351	373	350	848	491
Rb	567	501	33	296	614	508	517	573	497	509	538	361	612
q wt.%	22.83	22.30	33.54	26.24	28.28	30.19	28.04	22.74	29.51	28.94	26.52	31.21	22.58
or	56.73	60.75	5.08	48.16	47.04	45.27	45.56	49.11	44.97	44.74	46.81	42.02	52.12
ab	18.95	15.15	52.21	24.29	23.27	23.27	25.47	27.42	24.45	23.78	24.88	22.93	21.15
an	0.56	0.45	8.12	0.76	0.42	0.37	0.28	0.37	0.38	0.25	0.35	1.71	0.48

Sample	105-2	108-1	108-2	108-3	108-4	109-1	109-2	118-1	118-2	122-1	122-2	122-3
Texture	fg	mg	cg	cg	mg	cg	cg	mg	cg	cg	cg	mg
SiO_2 wt.%	74.5	75.1	75.8	74.8	73.9	77.0	74.7	72.8	72.3	75.7	72.8	73.3
TiO_2	0.00	0.01	0.01	0.00	0.01	0.00	0.03	0.01	0.00	0.00	0.00	0.00
Al_2O_3	13.70	13.26	13.16	13.74	14.00	12.48	14.13	13.90	14.51	12.80	14.33	14.10
Fe_2O_3	0.19	0.17	0.20	0.15	0.24	0.16	0.32	0.18	0.12	0.11	0.12	0.08
MgO	0.00	0.16	0.18	0.13	0.12	0.05	0.21	0.06	0.11	0.00	0.00	0.00
CaO	0.21	0.42	0.12	0.11	0.19	0.08	1.17	0.15	0.26	0.09	0.06	0.18
Na_2O	2.25	3.60	3.76	4.13	3.93	1.84	5.74	1.89	2.00	1.94	1.97	2.08
K_2O	8.36	6.27	6.21	6.49	6.65	8.31	2.09	9.83	10.30	8.30	9.55	9.39
MnO	0.01	0.01	0.00	0.00	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01
P_2O_5	0.02	0.02	0.01	0.01	0.01	0.00	0.00	0.01	0.01	0.00	0.00	0.02
Total	99.2	99.1	99.5	99.5	99.1	99.9	98.4	98.8	99.6	98.9	98.8	99.0
Ba ppm	490	710	710	760	690	370	180	590	580	140	120	110
Sr	298	506	480	495	396	177	159	136	145	30	37	32
Rb	955	221	221	219	237	405	144	306	318	1247	1533	1618
q wt.%	29.67	29.51	29.86	25.85	25.28	34.26	31.02	24.12	20.44	33.05	24.50	25.19
or	49.41	37.41	36.88	38.47	39.65	49.17	12.53	58.80	61.34	49.58	57.80	56.02
ab	19.04	30.72	31.99	34.69	33.59	15.57	49.33	16.16	17.31	16.25	16.84	17.77
an	0.91	1.55	0.53	0.00	0.88	0.40	5.90	0.43	0.00	0.45	0.30	0.76

*** Notes: fg: fine-grained, mg: medium-grained, cg: coarse-grained graphic texture. The X-ray fluorescence (Philips PW 1410) analyses for major- and trace elements were performed on fused disks at the University of Ottawa. GA and SY-2 were used as internal standards to determine accuracy, and several duplicates were used to determine precision. Estimated precision for major elements: less than 2%, except for Na_2O (3%); for trace elements, precision is less than 5%.**

indicates an average composition near the volatile-saturated minimum or eutectic in the haplogranite system at approximately 400 MPa (Lentz & Kretz 1989). Results of twenty-five chemical analyses of graphic granitic pegmatite from 10 localities (Table 1) are presented in Figure 4a. Representative samples consisting of single feldspar crystals and their contained quartz intergrowths were analyzed. The samples varied from 2 to 5 kg depending on the grain size. They contain less than 1 vol.% of additional phases. This was confirmed by the normative mineralogy (>99 wt.% quartz, orthoclase, albite, and anorthite in the norm; Table 1). The low abundance of anorthite (0 to 1.7 wt.%) in the norm is a result of the analysis of perthitic microcline. Therefore, a separate plagioclase phase was not incorporated into the graphic texture. The system quartz – albite – orthoclase – H_2O (Fig. 4b) is used to illustrate the phase relations because of the considerable experimental work published on equilibria in the haplogranitic system. The quartz – K-feldspar intergrowths consistently fall on the feldspar side of the cotectic. This would also include the two samples of quartz–plagioclase intergrowths that are located in the quartz-saturated field if the calcic component of plagioclase were considered. There is no obvious relationship between the grain size of the graphic texture and composition with respect to the eutectic. The quartz–microcline intergrowths have an average quartz content of 27.0 ± 1.5 wt. % (n = 23).

Phase-equilibria considerations

The transition from a granitic melt saturated in quartz and feldspar to one saturated in feldspar alone is problematic. In order for the graphic texture to grow, it is necessary that equilibrium saturation in quartz not occur throughout the melt;

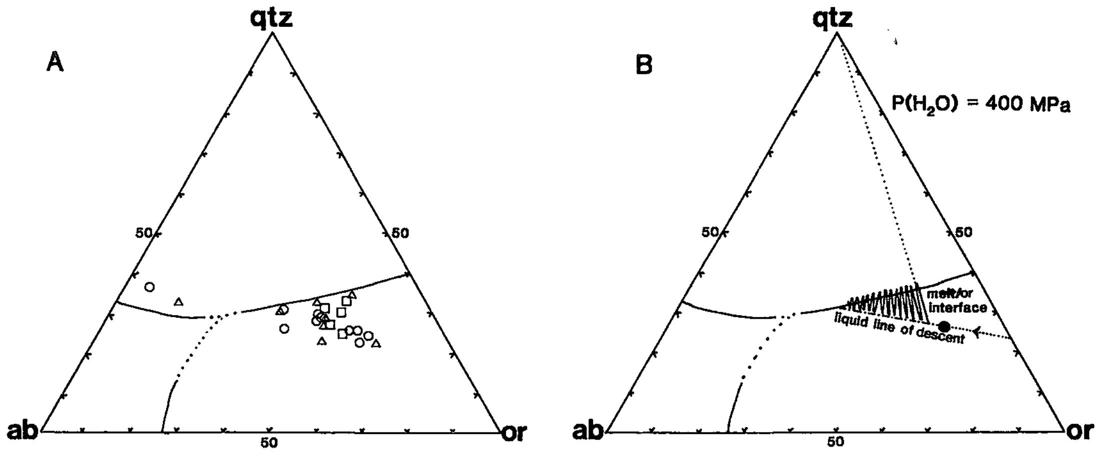

FIG. 4. A. The pseudoternary system quartz (Qtz) – albite (Ab) – K-feldspar (Or), showing the normative mineralogy of 23 samples of graphic granite pegmatite (this study). The squares represent coarse-grained, circles, medium-grained, and triangles, fine-grained samples. B. The estimated pseudoternary quartz (Qtz) – albite (Ab) – K-feldspar (Or) diagram with the phase boundaries for the haplogranite system at 400 MPa P(H_2O). The dot represents the approximate composition of the melt. The dashed-dotted line indicates the integrated evolution of the liquid over the course of crystallization of the graphic texture. The jagged line represents the composition of the boundary-layer melt at the crystal–melt interface.

therefore, the appropriate phase-equilibrium relationships must be considered.

At the approximate pressures and temperatures corresponding to pegmatite crystallization (H_2O-saturated), the quartz and alkali feldspar cotectic at 500 MPa (Luth *et al.* 1964) is located at 28% Qtz (eutectic, (650°C) to 36% Qtz (Qtz–Or pseudobinary eutectic, 735°C). At 300 MPa, the cotectic lies between 33% Qtz (eutectic, 665°C) to 42% Qtz (Qtz–Or pseudobinary eutectic, 745°C) (Tuttle & Bowen 1958). By interpolation, an alkali feldspar composition of Or_{70} at 400 MPa corresponds to 35% quartz along the cotectic. The minor amount of fluorine inferred to be present, based mainly on fluorite in the associated skarns and fluorine-rich biotite in the pegmatites, would reduce the liquidus and solidus temperatures in the Qtz – Or – Ab system and expand the primary field of quartz. At a pressure of 275 MPa, Wyllie & Tuttle (1961) noted a decrease in the volatile-saturated thermal minimum from 675° to 595°C with the addition of 4 wt.% fluorine. Manning (1981) reported a substantial shift to higher proportions of feldspar with the addition of fluorine at 100 MPa (volatile-saturated system). These observations suggest that similar shifts in the quartz–feldspar equilibria would occur at 400 MPa pressure with the addition of fluorine. The inferred concentration of fluorine in the granitic melts that crystallized to form the Grenville pegmatites is variable but is estimated to be less than 1 wt.%. The maximum effect on the solidus is, by analogy, on the order of 30°C, yielding a 600°C eutectic temperature, thus enhancing the stability field of quartz, mainly at the expense of albite.

In contrast, a decrease in the confining pressure can displace the quartz–orthoclase cotectic, enhancing the primary field of feldspar with respect to that of quartz. The occurrence of large alkali feldspar phenocrysts only does not preclude quartz saturation elsewhere in the granitic melt; however, the phase equilibria indicate that crystallization in the feldspar field is possible under specific conditions.

A MODEL OF DYNAMIC CRYSTALLIZATION FOR GRAPHIC INTERGROWTHS

The observation of lobate inner boundaries on feldspar-hosted quartz is critical (Figs. 2, 3). Rough boundaries arise during conditions of high supercooling (Woodruff 1973), when the supply of growth atoms is restricted by low diffusivities or depletion of crystallizing constituents in the boundary layer. This results in nonequilibrium growth. Protuberances on the crystal face become stabilized and grow (*e.g.*, Nittmann & Stanley 1986). We interpret the appearance of the lobate inner boundary as the onset of diffusion-limited conditions during feldspar growth. Feldspar-hosted quartz in graphic intergrowths has characteristic prism, arrow, and rod morphologies. Fenn (1977)

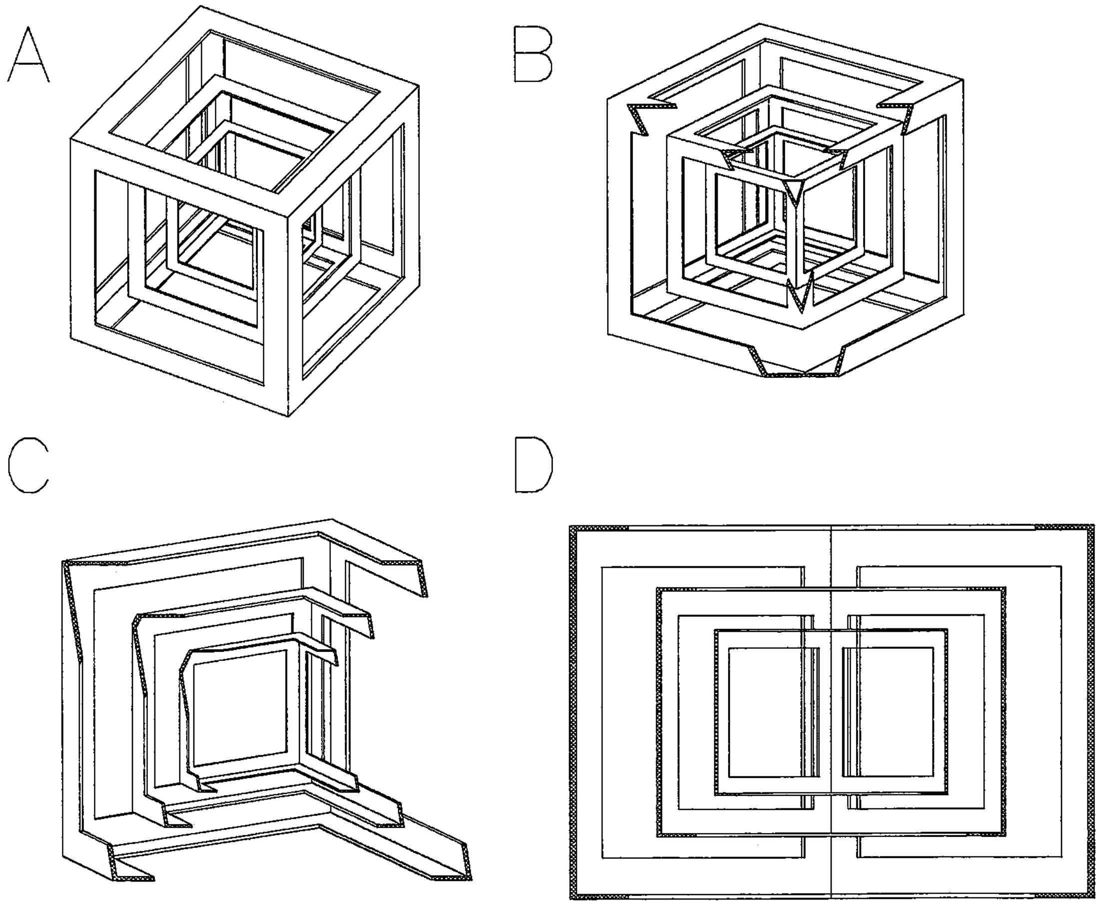

FIG. 5. Schematic three-dimensional drawings of feldspar showing three quartz intergrowths and illustrating possible two-dimensional morphologies of graphic texture (cross-hatched) A. Simple three-dimensional view of quartz (angles) inside host feldspar (space) B. and C. Oblique sections through quartz–feldspar intergrowth with two-dimensional view (cross-hatched) D. Two-dimensional view (110) of A with two-dimensional section (cross-hatched).

found that the growth morphology of feldspar changes from equilibrium planar to skeletal growth as a function of the degree of undercooling. Crystal corners and edges subtend a greater angle in the melt than planar crystal faces and preferentially grow because they have access to a greater volume of melt. The striking patterns of the graphic texture are the result of consistent angular relationships between the intergrown quartz and the host feldspar crystal imposed during growth. We interpret them to be due to preferential nucleation and growth of quartz along corners, edges and, to a lesser extent, faces of the alkali feldspar. The observed narrow arrows and prisms of quartz are two-dimensional sections of quartz that grew along edges of the feldspar, whereas rods represent two-dimensional slices of edge or surface growth of quartz on the host feldspar. Figure 5 is a schematic three-dimensional image of a graphic intergrowth of quartz and feldspar that illustrates the geometrical aspects of the growth. For simplicity of design, we used a cubic rather than a monoclinic feldspar host. Note that quartz (hachured) occurs along corners and edges of the feldspar; Figure 5 illustrates a few of the possible geometries of quartz in feldspar. The spacing between the three sequential episodes of quartz growth is exaggerated in order to exemplify the three-dimensional nature of the texture; it portrays only the large-scale morphologies and leaves out details on the irregular (inner) and regular (outer) surfaces.

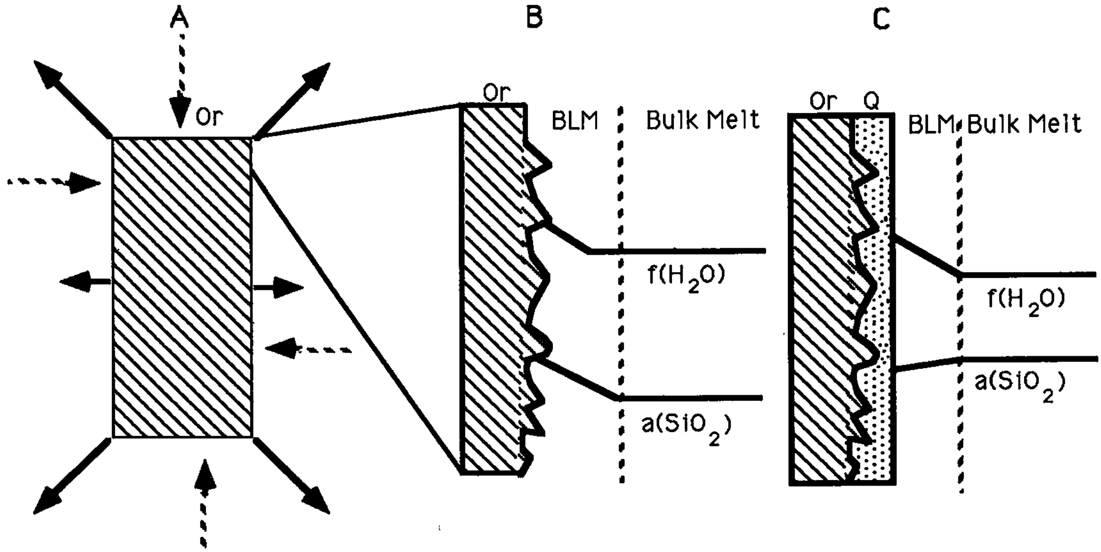

FIG. 6. A. Growth of alkali feldspar crystal. Shown schematically are the diffusion of rejected Si-species and H_2O (solid vectors) and the diffusion of Al-species added to the crystal (hachured vectors). B. Magnification of crystal edge at corner of (A), showing growth of protuberances (displaced diagonal hachure) to form rough boundary. The relative fugacity of water [$f(H_2O)$] and activity of silica [$a(SiO_2)$] are shown in the boundary-layer melt (BLM) and bulk melt. C. Crystallization of quartz has reduced $a(SiO_2)$, so that alkali feldspar crystallizes next.

As shown previously, growth of the graphic texture in pegmatites of the Grenville Province occurred at an approximate pressure of 400 MPa and a minimum melt temperature near 600°C, close to H_2O-saturated conditions. Figures 4b and 6 illustrate the dynamics of growth. Alkali feldspar was the first mineral on the liquidus. Growth of the feldspar in this melt required the rejection of H_2O and excess silica. An increase in dissolved volatiles alone should increase slightly the diffusivity of all feldspar components by decreasing the degree of polymerization of the melt (Dingwell 1986, Scarfe 1986). This has the effect of increasing rates of crystal growth. Fenn (1986) proposed that increased growth occurred in response to the increasing activity of water at the melt interface. Thus the net effect of the rejected H_2O is to catalyze feldspar growth in the immediate vicinity of the crystal. Initially, this process is self-propagating, because growth will cause rejection of H_2O, which increases the rate of growth, and so on. Depletion of Al-bearing species in the boundary layer (*i.e.*, Si enrichment) results from more rapid growth of feldspar and slow diffusion of Al-bearing species from the bulk melt into the boundary layer. Therefore, diffusion-limited growth of feldspar arises (Fig. 6). Thus edges and corners become sites of preferred growth, and the feldspar faces become rough. Eventually, SiO_2 builds up to saturation at these sites, and quartz is epitactically nucleated and grown on the feldspar structure. The growth of quartz depletes the boundary layer in Si (Fig. 6c) relative to Al, resulting in renewed crystallization of alkali feldspar.

DISCUSSION

The shape of quartz in graphic intergrowths of quartz and feldspar is a function of orientation of the feldspar (Fersman 1928, Simpson 1962, Smith 1974), as is observed here. Consistent orientations also have been observed by Fersman (1928), Simpson (1962) and Smith (1974). The detailed work by Heritsch & Holler (1960) and Simpson (1962) shows that quartz rods change shape along their length. Surprisingly, the irregular nature of the inner surface of the rod quartz had not previously been documented. This may be in part due to the lack of preservation of the irregular interface (*e.g.*, Smith 1974). Fersman (1928) found a 42° angle between the *c* axes of quartz and feldspar. Using X-ray and optical techniques, Wahlstrom (1939) found no particular orientation. However, using both X-ray and optical techniques, Heritsch (1953a, b), Heritsch & Holler (1960), Heritsch *et al.* (1962), Choudhury *et al.* (1965) and Bakumenko (1966a, b) confirmed several angular relationships.

We found consistent proportions of quartz to feldspar (28 wt.% normative quartz) in graphic pegmatites from the southwestern Grenville Province (Table 1). Similarly, Erdmannsdorfer (1942) found an average of 27 vol.% quartz and a range of 10 to 65 vol.% quartz in quartz–microcline intergrowths. Vogt (1931) found 25 vol.%, whereas de Schmid (1916) determined an average 28.9 wt.% quartz for the Lower St. Lawrence pegmatites. Černý (1971) found similar proportions of quartz to feldspar in a study of graphic pegmatites from Czechoslovakia. Mehnert (1968) argued that a quartz-to-feldspar ratio of 27:73 was not compatible with a melt on the quartz–feldspar cotectic. Within each data set, the volume proportion of quartz is approximately constant, indicative of a recurrent process.

The dynamics of the proposed model are driven by an interplay between crystal growth and diffusion rates of rejected solutes. The diffusivity of Si- and Al-bearing species is very low in low-temperature granitic systems (Hofmann 1980, Dunn 1986); however, the depolymerization of the melt by network modifiers (H_2O and HF) reduces viscosity and increases diffusion in volatile-rich granitic melts (Scarfe 1986). The diffusivity of H_2O in obsidian and granitic melt is between 10^{-8} and 10^{-9} cm^2/s at temperatures between 600°C and 800°C (Shaw 1974, Arzi 1978, Karsten *et al.* 1982). The effect of depolymerization of the network of Si(Al)–O tetrahedra and coordination with OH^- could considerably reduce the diffusivity of OH^- in the melt and increase the diffusivity of Si and Al because of their coherent behavior. The nucleation and growth of either quartz or feldspar at the interface may be too rapid for the rejected H_2O to diffuse into the volatile-undersaturated melt, and may create locally volatile-saturated conditions at the interface. Evidence of volatile saturation was found by Fenn & Luth (1973), who reported the presence of fluid inclusions in quartz, in experiments at H_2O-undersaturated conditions. The limiting factor in diffusion-controlled crystal growth is most likely the diffusion of Si away from the feldspar and diffusion of Al toward the feldspar interface, since the diffusivity of potassium and sodium is considerably greater (Hofmann 1980, Dunn 1986).

Swanson (1977), Fenn (1977) and Dowty (1980) have shown that nucleation and growth curves for feldspar shift toward the liquidus at higher volatile contents in the melt. The formation of epitactic quartz on feldspar may be indicative of a relatively low density of nuclei for quartz, which is expected at very low undercoolings. Heterogeneous nucleation in pegmatites, particularly along the walls, will initiate crystal growth at low degrees of supercooling. This inference was substantiated by Fenn (1986) and London *et al.* (1989), who found the graphic texture along the walls of their reaction vessels.

Perhaps the most unusual feature of the proposed model is its oscillatory nature. Therefore, the liquid line of descent (Fig. 4b) is distinctly different from the familiar equilibrium case. The graphic intergrowths are a nonequilibrium phenomenon. If the crystallization were equilibrium (*i.e.*, if the system was well mixed), one would observe a granitic texture produced by the crystallization of discrete crystals of alkali feldspar and quartz. It is now known that systems driven far from equilibrium may have an oscillatory behavior (*e.g.*, Nicolis & Prigogine 1989). Chemical oscillators produce striking spatial or temporal patterns that, in the opinion of Gray & Scott (1990), are due to an interaction of autocatalytic or thermal feedback with the diffusive transport of molecules in the system. The proposed model for graphic intergrowths identifies H_2O as the catalyst that changes the diffusion properties of the melt with crystallization. The corner and edge habit, and inner lobate boundaries of quartz, indicate that the growth of feldspar was diffusion-limited. Conversely, the euhedral outer boundaries of the quartz with the feldspar are indicative of a return to an equilibrium style of growth (Fenn 1977). Oscillations in far-from-equilibrium growth commonly are characterized by such growth, involving a transition from stable to unstable growth of one phase followed by an abrupt and stable growth of a different phase (*e.g.*, Gray & Scott 1990).

The common association of graphic and granophyric textures in granites and pegmatites indicates that understanding the nature of the magmatic–hydrothermal transition is key to the formation of these textures. There is an increasing body of experimental information on the phase relations in volatile-bearing granitic magmas (melt + crystals + vapor) (Luth & Tuttle 1969, Maaløe & Wyllie 1975, Whitney 1975, 1977, Naney 1983, Huang & Wyllie 1986, London *et al.* 1989). The crystallization history of granitic magmas may be significantly affected by the original volatile contents (amount and species).

Figure 7 illustrates in a qualitative manner the nature of the vapor-saturation surface in terms of pressure, temperature, and amount of H_2O for a melt of granitic composition. The crystallization of anhydrous silicates in the melt enriches the remainder of the melt in that component by diffusion of the volatile component away from the crystal–melt interface (Jahns & Burnham 1969, Fenn 1977; Fig. 7, curve A), to the point of achieving saturation in a volatile phase. Water-saturated conditions at the interface enhance feldspar growth and result in progressively decreas-

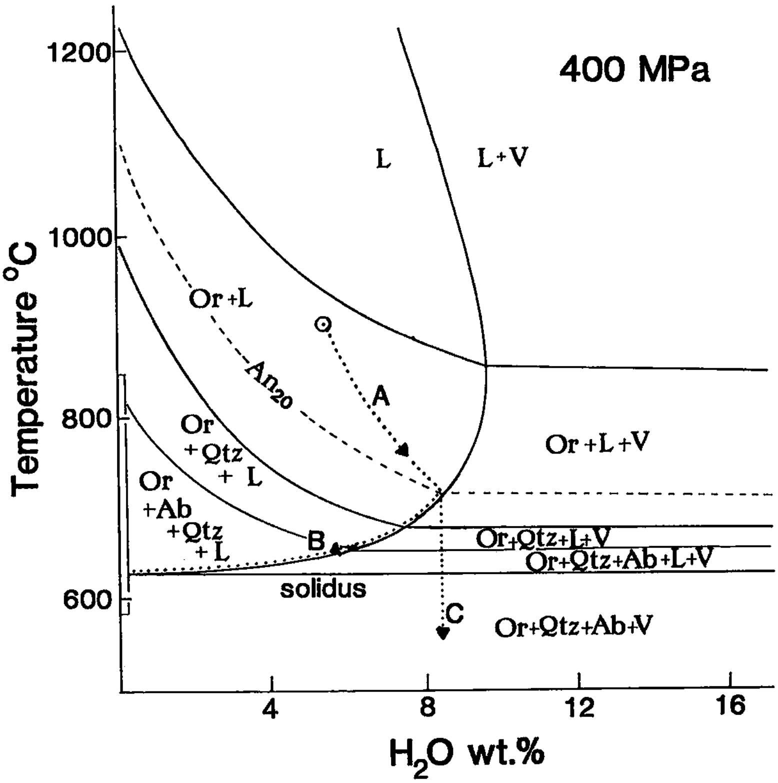

FIG. 7. Schematic representation of crystallization of a granitic melt and conditions of saturation in the haplogranite system at 400 MPa similar to the experimentally derived equilibria (Fenn 1986). The sequence of crystallization has been modified so that orthoclase apears on the liquidus before quartz, as explained in the text. Curve A is the bulk-liquid line of descent at volatile-undersaturated conditions due to crystallization of orthoclase and intermittent saturation of quartz; curve B is the path of crystallization at volatile-saturated conditions, with separation of the vapor phase, and curve C is the path of volatile-saturated crystallization for a fluid partially retained in the pegmatite into subsolidus conditions. Within the pegmatite, plagioclase compositions in the system are variable between albite and oligoclase.

ing activities of H_2O in the melt away from feldspar–melt interface. This in turn leads to a decrease in the diffusivity of cations away from the crystal. Therefore, factors affecting saturation and separation of a volatile phase in the melt are important, in particular geostatic changes in pressure arising from emplacement of the pegmatite and hydrofracturing associated with separation of the volatile phase (Fig. 7, curve B).

If the fluid is retained within the pegmatite because the competency of the rocks is not exceeded (Fig. 7, curve C), internal replacement of primary minerals and textures would occur (London *et al.* 1989).

CONCLUSIONS

The physical conditions within the U-, Mo-, *REE*-, and Nb-bearing granitic pegmatites of the Grenville Province were responsible for the formation of graphic quartz–feldspar intergrowths. The spatial distribution of the graphic texture in these pegmatite bodies, near the contact with the host rocks, suggests that the saturation of a volatile phase during crystallization is important to understanding their formation. The available whole-rock geochemical data, the volume proportions of quartz to K-feldspar, and phase-equilibrium data in the fluorine-bearing haplogranite system favor nearly simultaneous crystallization of quartz and K-feldspar from a melt approaching the cotectic toward quartz saturation. Close observation of the textures reveals an irregular quartz - K-feldspar boundary and indicates that quartz preferentially grew on the corners and edges of the feldspar. During feldspar growth, the area immediately surrounding alkali feldspar crystals became depleted in Al and enriched in Si. This resulted in growth being concentrated at crystal edges and corners and the production of rough crystal faces (feldspar protuberances). The buildup of rejected silica resulted in quartz saturation and epitactic nucleation. Growth of quartz depleted the silica concentrated in the boundary layer melt and restored feldspar-saturated growth. This process repeated and resulted in the oscillation of the melt composition in the boundary layer from quartz-oversaturated to quartz-undersaturated conditions, producing rhythmic quartz–feldspar intergrowths. The saturation of a volatile phase (H_2O) at the feldspar–melt interface may increase undercooling and further reduce diffusion of Si away from the feldspar.

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