# On the origin of graphic granite

# PHILIP M. FENN

Sullivan Park FR-01-8, Corning Glass Works, Corning, New York 14831

#### ABSTRACT

For over a century, the origin of the quartz-feldspar intergrowth known as graphic granite has been debated in the literature. These textures have been produced experimentally during a study of the phase equilibria and nucleation and growth characteristics of granitic pegmatites. Analysis of temperature, pressure, and compositional variables suggests that the texture is produced by the simultaneous growth of quartz and feldspar in a kinetically driven, nonequilibrium situation. The growing interface of the host phase, a sodic alkali feldspar, is degraded from planar to cellular by the development of a SiO<sub>2</sub>- (and probably H<sub>2</sub>O-) enriched boundary layer. Between the cell boundaries, the SiO<sub>2</sub> content of the residual liquid achieves a level of supersaturation that allows quartz to nucleate and grow along with the feldspar. The bulk composition may well be on a cotectic surface, but under this mechanism it is not necessary. The development of this texture is dependent upon local kinetics at the interface rather than being solely tied to the thermochemical equilibrium of the bulk composition.

# INTRODUCTION

The coarse intergrowth of quartz in a potassic alkali or sodic plagioclase feldspar host that displays runic or cuneiform texture is commonly referred to as graphic granite. Found predominantly in granitic pegmatites, graphic granite is volumetrically insignificant in the family of igneous rocks, but the process involved in the development of this texture may give considerable insight into the crystallization of granitic rocks. In spite of this low abundance, graphic granites have been the topic of considerable debate in the literature. Not only is their origin in question but also the existence of a crystallographic relationship between the two phases. It is beyond the scope of this paper to engage in any detailed description of the texture or a review of the literature; the reader is referred to the excellent discussion in Smith's (1974) treatise on the feldspar minerals.

The only previous experimental work pertinent to the origin of this intergrowth is that of Schloemer (1962, translated in 1964). In a study of the hydrothermal devitrification of glasses in the K<sub>2</sub>O-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system, he produced textures that are suggestive of graphic granite, e.g., his Figure 49a. Schloemer proposed that these textures were a eutectic structure resulting from the simultaneous growth of orthoclase and quartz. His P-T conditions and bulk compositions do not correspond to liquidus cotectics in published phase equilibria of this system (Tuttle and Bowen, 1958); thus, in the strict sense his textures cannot be called eutectic. In another series of experiments, he demonstrated the feasibility of the infiltration and replacement of orthoclase by quartz under hydrothermal conditions. Thus both proposed models for the origin of graphic granite were demonstrated but neither in such a manner as to exclude the other from future consideration and argument.

#### **EXPERIMENTS**

The pegmatite samples used as starting materials for the experiments described in this paper were provided by the late R. H. Jahns. They are splits of the samples used by Burnham and Jahns (1962), Jahns and Burnham (1969), and Vaughan (1963). The samples are composites made up of a bulk sample of a pegmatitic pod from Chalk Mountain in the Spruce Pine district, North Carolina, and from diamond drill cores of the Harding pegmatite in Taos County, New Mexico. These composites were prepared to represent estimates of the bulk compositions of the parent pegmatitic magmas. The bulk compositions and normative mineral contents expressed in the haplogranodiorite tetrahedron are listed in Table 1. Details of the phase equilibria of these samples at 5000 bars are reproduced in Figures 1 and 2.

The Spruce Pine and Harding pegmatite samples were ground to an average grain size of 5  $\mu$ m, loaded into gold or platinum capsules with a measured amount of distilled, deionized water, and finally sealed and placed in an internally heated pressure vessel. The samples were homogenized in the liquid or liquid plus aqueous vapor phase field at 900°C for a period of 72 h, then the temperature and pressure were rapidly readjusted to the nucleation and growth conditions. All of the experiments reported in this paper were performed at a confining pressure of 5 kbar. After a predetermined period, the runs were rapidly quenched to ambient conditions, the capsules opened, and thin sections prepared for petrographic analysis.

#### RESULTS

During the examination of the run products, it was noted that under restricted conditions of temperature and/ or water content, intergrowths of quartz and feldspar resembling graphic granite were produced. Figure 3 illus-

	Hard- ing	Spruce Pine
sio <sub>2</sub>	75.24	73.79
TiO <sub>2</sub>	0.05	0.05
A1203	14.42	15.11
Fe203	0.14	0.26
FeO	0.35	0.16
MnO	0.18	0.05
MgO	0.01	0.07
CaO	0.20	0.97
Na <sub>2</sub> 0	4.23	4.71
к <sub>2</sub> 0	2.74	4.02
P205	0.13	0.01
F	0.64	0.01
co <sub>2</sub>	0.03	0.02
Total	98.36	99.23
Ab	30.38	41.0
Or	18.19	24.48
An	0.00	4.69
Q	51.43	29.70

Table 1. Compositions of starting materials and normative mineralogy

trates a typical assemblage observed in the Spruce Pine samples. The capsule wall is in the upper right portion of the photograph, and two distinct morphologies of sodic plagioclase are observed: single laths and the complex intergrowths seen near the center. Figure 4 illustrates that these are formed by the intergrowth of two single crystals; the external morphology and the differences in index of refraction indicate that the host phase is a sodic plagioclase and the internal phase is quartz. Note that the intergrowth is best developed in feldspar crystals that grow away from the capsule walls, surrounded by bulk melt. The relative ease with which the feldspars nucleate on the walls of the capsule is not pertinent to the topic of this paper, but it may have application to the growth of large tapered feldspars from the hanging wall of many granitic pegmatite dikes (Jahns, 1953).

The growth conditions of the experiment illustrated in Figures 3 and 4 indicate that the initial undercooling of the plagioclase was 165°C and that of quartz 15°C. This places the bulk composition on the feldspar-quartz cotectic, but it should be noted that there is no free quartz



Fig. 1. Phase equilibria of Harding pegmatite composition at 5 kbar as a function of temperature and H<sub>2</sub>O added to capsule. L = liquid, V = aqueous vapor, Q = quartz ( $\alpha$ - or  $\beta$ -polymorphs), Ab = sodic alkali feldspar, Kf = potassic alkali feldspar, Mu = muscovite, Sp = Spodumene. Because beryl was present in all charges up to liquidus conditions, it was impossible to determine whether quartz or beryl is the liquidus phase. Location of solidus is approximate but is believed to be within 10°C of location shown.

in the sample. In the experiment shown in Figure 5, free quartz and feldspar are visible, but graphic intergrowths are not found under these conditions. Here an extra 50°C of undercooling has allowed the nucleation of both phases, but the feldspar grows initially as finely-bladed, dense spherulites along the capsule wall and the quartz as coarse dendrites. Figure 6 illustrates the results of a nucleation and growth experiment on the Harding composition. This bulk composition lies in the liquidus field of beta quartz under the P-T conditions of the experiment, and the presence of individual dendritic quartz crystals scattered throughout the glass confirms this. The graphic intergrowth displays the external morphology of a feldspar, and there is no indication of the growth of feldspar around any of the pre-existing quartz dendrites. This demonstrates that the feldspar (nearly pure albite in this system) is the host phase for the intergrowth and must begin to grow before any graphic texture can develop. Under these circumstances, the bulk composition is on the feldspar-



Fig. 3. Spruce Pine pegmatite plus 4.5 wt% H<sub>2</sub>O held at 750°C for 96 h. The capsule wall was in the upper left corner. Scale bar = 1.0 mm; cross-polarized light.

Fig. 2. Phase equilibria for the Spruce Pine pegmatite composition. Phase identification as in Fig. 1 except Pl = sodic plagioclase feldspar (probably an oligoclase).

quartz cotectic, but even though this is true, the texture is not developed everywhere in the charge as evidenced by the separate quartz dendrites and euhedral albite crystals in near-juxtaposition.

## DISCUSSION

An evaluation of the literature concerning origin of graphic granites indicates that there are two basic models for the formation of the texture: the replacement of portions of a pre-existing feldspar crystal by quartz or the simultaneous crystallization of quartz and feldspar. Most of the recent literature accepts the concept of simultaneous crystallization, and the subject of current debate has shifted to the role of eutectic or cotectic crystallization in the development of the texture. The similarity of the texture to the eutectic structures developed in metallic systems is compelling, but the lack of a strong correlation between the available analyses of graphic granites and the experimentally determined liquidus and solidus surfaces suggests that the origin is not so simple (Barker, 1970). The data presented in this study suggest an origin involving simultaneous crystallization driven by the kinetics of crystal growth and diffusion. The development of the graphic texture commences with growth of a feldspar crystal under



Fig. 4. Same experiment as Fig. 1 showing enlarged view of quartz-feldspar intergrowth. Scale bar = 0.1 mm; cross-polarized light. Note the rods and ends of rods in other crystals.



Fig. 5. Spruce Pine pegmatite plus 4.5 wt%  $H_2O$  held at 700°C for 240 h. Capsule wall was at right edge of photomicrograph. The large dendrite at the center is beta quartz, and feldspar appears as dense spherulites at the capsule walls and as thin lath-shaped crystals around the base of the dendrite. Scale bar = 0.1 mm; cross-polarized light.

conditions that stabilize a planar-growth interface; that is, a temperature and composition that yield a low undercooling relative to the upper stability limit of the feldspar. If the conditions are appropriate, the rate of advance of the crystal-liquid interface will exceed the rate at which nonfeldspar components, specifically H<sub>2</sub>O and excess SiO<sub>2</sub>, are able to diffuse away into the bulk melt. Under these conditions, the relative undercooling at the growth interface will decrease as the composition of the interface liquid moves toward one with a lower liquidus temperature, eventually approaching equilibrium with the feldspar (zero undercooling). The rate of advance of the interface will be slowed by this local decrease in undercooling, but if a perturbation develops that projects ahead of the interface into melt compositions more closely resembling the bulk composition, the tip of this perturbation will experience a relatively greater undercooling and thus may grow forward at a higher rate. This leads to the breakdown of the planar interface and may initiate the formation of a cellular interface (Tiller and Rutter, 1956). During the growth of a cellular interface, the excess components,  $SiO_2$  and H<sub>2</sub>O, are rejected not only from the tip of the growing perturbation but also from the sides. Within the grooves between growing perturbations, the concentration of SiO<sub>2</sub> and H<sub>2</sub>O will build up faster than at the tip. In metal alloy systems, this lateral segregation at the cell boundaries has been shown to yield a fifteenfold increase in solute concentration (Biloni and Bolling, 1963). In the growth of a feldspar from a granitic melt, the concentrations of  $SiO_2$ and H<sub>2</sub>O in the cell boundary regions may approach or even exceed the saturation levels of either or both components. If enough supersaturation is developed in the grooves, the nucleation of vapor bubbles or quartz is possible even though the bulk composition of the system might suggest that neither phase is stable. The nonequilibrium supersaturation of H<sub>2</sub>O at the interface of a growing crystal has been demonstrated experimentally and reported by Fenn and Luth (1973).



Fig. 6. Harding pegmatite plus 6.0 wt%  $H_2O$  held at 750°C for 168 h. Capsule wall was across top of photomicrograph. Fine dendritic crystals are beta quartz, and the large intergrowth is made up of quartz in an albite lath. Undercoolings of the two phases are beta quartz-137°C and albite-60°C. Scale bar = 0.1 mm; cross-polarized light.

In the experiment shown in Figures 3 and 4, the low degree of undercooling (15°C) has apparently prohibited the nucleation of quartz except in the graphic intergrowth where a higher degree of supersaturation has allowed its nucleation and subsequent growth. Once the quartz nucleates and begins to grow, the coupled growth of the two phases may increase the growth rate of the aggregate above that of either of the components (e.g., Carstens, 1983). This would explain why the intergrowths shown in the photomicrographs are considerably larger than either of the single phases in the same charge.

The local environment at the interface of the growing feldspar crystal must create the conditions necessary for the transition from a planar- to a cellular-interface morphology and thus permit the formation of the intergrowth. The formation of a planar interface on feldspars grown from granitic melts has been shown to occur at low to moderate undercoolings (Fenn, 1977; Swanson, 1977). Coupled with this restriction on proximity to the feldspar liquidus, there is also the requirement that the growth rate must be high relative to the diffusivity of silica in the residual melt. In the previously mentioned studies on the nucleation and growth of feldspars, the data suggest that as the H<sub>2</sub>O content of the system increases, the maximum growth rate shifts to lower undercoolings. This trend raises the possibility that the development of graphic textures is enhanced by high H<sub>2</sub>O contents and may help to explain the association of graphic granites and granitic pegmatites.



Fig. 7. Same compositon as Figure 6 but held at 750°C for 48 h. The albite lath is intergrown with quartz and surrounded at its base by dendrites of beta quartz. The capsule wall was to the right. Scale bar = 0.1 mm; cross-polarized light.

Another feature of the proposed mechanism concerns the time required to form the necessary interface enrichment and its relationship to the anisotropic nature of the feldspar lattice and the variation of growth rate along the crystallographic directions. Because finite time is necessary to build up the boundary layer, the center of the feldspar crystal should be free of intergrowths, as has been described in the literature (see Smith, 1974, p. 601–602). Also because the growth rates of the alkali feldspars depend upon the crystallographic direction (Fenn, 1977), the extent of boundary layer segregation will also depend upon the direction of growth. Thus, the appearance of the intergrowth will vary with the orientation of the observed section. This feature has also been documented in the literature and in the experiments reported in this study. Figure 7 illustrates a feldspar lath showing a sharp boundary between the nonintegrown core and the graphic zone.

## CONCLUSIONS

The origin of the intergrowth of quartz and alkali feldspar known as graphic granite has been shown to be the result of simultaneous growth of the two phases under conditions that favor the planar growth of the feldspar host. An imbalance between the growth rate of the feldspar and the diffusivity of silica in the bulk melt creates a silicaenriched boundary layer that in turn causes the interface to degrade from planar to cellular. Upon further growth of this cellular interface, the grooves between adjoining cells are greatly enriched in SiO<sub>2</sub>, and this enrichment causes the nucleation and growth of quartz along with the feldspar. This simultaneous growth was not caused by classical eutectic crystallization but by the kinetic phenomena in the boundary layer adjacent to the interface of the growing crystal.

#### **ACKNOWLEDGMENTS**

The author deeply regrets the passing of two good friends and colleagues, Dick Jahns and Peter Gordon, both of whom have made major contributions to this study. It is unfortunate that the publication of these results must come under such circumstances. His unparalleled teaching ability, boundless enthusiasm, and the ability to pass on this enthusiasm made Dick Jahns one of the most respected geologists of his time. Peter Gordon's skill in the design, fabrication, and maintenance of experimental equipment made this and dozens of other studies possible.

The experiments were performed in the Tuttle-Jahns Laboratory for Experimental Petrology at Stanford University. The support of the National Science Foundation through grant EAR 76-22688 is gratefully acknowledged.

#### References

- Barker, D.S. (1970) Composition of granophyre, myrmekite, and graphic granite. Geological Society of America Bulletin, 81, 3339–3350.
- Biloni, H., and Bolling, G.F. (1963) A metallographic study of solute segregation during controlled solidification in lead-tin alloys. Metallurgical Society of AIME Transactions, 227, 1351– 1360.

- Burnham, C.W., and Jahns, R.H. (1962) A method for determining the solubility of water in silicate melts. American Journal of Science, 260, 721–745.
- Carstens, H. (1983) Simultaneous crystallization of quartz-feldspar intergrowths from granitic magmas. Geology, 11, 339– 341.
- Fenn, P.M. (1977) The nucleation and growth of aklaki feldspars from hydrous melts. Canadian Mineralogist, 15, 135–161.
- (1979) On the origin of graphic intergrowths. (abs.) Geological Society of America Abstracts with Programs, 11, 424.
- Fenn, P.M., and Luth, W.C. (1973) Hazards in the interpretation of primary fluid inclusions in magmatic minerals. (abs.) Geological Society of America Abstracts with Programs, 5, 617.
- Jahns, R.H. (1953) The genesis of pegmatites. I. Occurrence and origin of giant crystals. American Mineralogist, 38, 563–598.
- Jahns, R.H., and Burnham, C.W. (1969) Experimental studies of pegmatite genesis: I. A model for the derivation and crystallization of granitic pegmatites. Economic Geology, 64, 843– 864.
- Schloemer, H. (1962) Hydrothermal-synthetische gemeinsame Kristallisation von Orthoklas und Quarz, I. and II. (In German) Radex-Rundschau, 3, 133–156; 4, 157–173. (transl. Geochemistry International, 3, 578–598, 599–612, 1964).
- Smith, J.V. (1974) Feldspar minerals, vol. 2, chemical and textural properties. Springer-Verlag, New York.
- Swanson, S.E. (1977) Relation of nucleation and crystal-growth rate to the development of granitic textures. American Mineralogist, 62, 966–978.
- Tiller, W.A., and Rutter, J.W. (1956) The effect of growth conditions upon the solidification of a binary alloy. Canadian Journal of Physics, 34, 96–121.
- Tuttle, O.F., and Bowen, N.L. (1958) Origin of granite in light of experimental studies in the system NaAlSi<sub>3</sub>O<sub>8</sub>-KAlSi<sub>3</sub>O<sub>8</sub>-SiO<sub>2</sub>-H<sub>2</sub>O. Geological Society of America Memoir 74.
- Vaughan, D.E.W. (1963) The crystallization ranges of the Spruce Pine and Harding pegmatites. M.S. thesis, Pennsylvania State University, University Park.

MANUSCRIPT RECEIVED MARCH 6, 1985 MANUSCRIPT ACCEPTED NOVEMBER 5, 1985