

ORIGIN AND CRYSTALLIZATION OF SOME PERALUMINOUS (S-TYPE) GRANITIC MAGMAS

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ABSTRACT

Our experiments (700–900°C, 1–7 kbar, $a(\text{H}_2\text{O}) = 0.1\text{--}1.0$, $f(\text{O}_2) \leq \text{QFM}$, seeded glass starting materials) were carried out on four compositions from central Victorian S-type volcanic-plutonic suites. Biotite saturation curves have steep dP/dT slopes at 800–850°C for melt water-contents of 2–10 wt.%. Early crystallization of F-poor biotite with plagioclase (up to 3 kbar) and quartz (at higher pressures) requires water contents > 2.5 wt. % in the melt. Orthopyroxene has a wide crystallization field; its absence from most granitic rocks indicates relatively low initial magmatic temperatures or, more likely, reaction to biotite with falling T. Cordierite crystallizes near the liquidus at $P = 1\text{--}2$ kbar but does not develop above the solidus at $P > 5$ kbar. Almandine-rich garnet becomes a near-liquidus phase at $P \geq 4\text{--}5$ kbar. Integration of geothermometry and barometry calculations and crystallization sequences with the experimental data indicates melt water-contents of 3–5 wt. %, with early-crystallizing phases (garnet, biotite, orthopyroxene, plagioclase) present at $T \leq 850^\circ\text{C}$, $P \geq 4$ kbar. These relatively high-temperature, H_2O -undersaturated S-type magmas originated by partial fusion of mildly peraluminous materials under granulite-facies conditions. Calculated dehydration and melting equilibria model the partial fusion of biotite- and muscovite-bearing assemblages for $a(\text{H}_2\text{O}) < 1.0$. Together with 5-kbar crystallization experiments, these data are used to estimate anatexis conditions and the chemical features of melts derived from aluminous high-grade metamorphic rocks. Low-temperature, H_2O -rich melts generated by muscovite breakdown have compositions near the "granite minimum". Higher-temperature melts, formed by partial fusion of biotite-bearing assemblages, are strongly water-undersaturated and depart markedly from "minimum" compositions. Source-related thermal and chemical characteristics of peraluminous magmas are reflected in the emplacement mechanisms, chemistry and mineralogy of S-type granitoid rocks.

Keywords: peraluminous granitoid rocks, granite petrogenesis, phase relations, experimental studies, hydrous magmas, anatexis, high-grade metamorphism, S-type granites, mineralogy, biotite, crystallization, Victoria, Australia.

SOMMAIRE

Nos expériences (700 à 900°C, 1 à 7 kbar, $a(\text{H}_2\text{O})$ de 0.1 à 1.0, $f(\text{O}_2) \leq \text{QFM}$, compositions de

départ vitreuses, en présence de germes cristallins) ont porté sur quatre roches tirées de suites volcaniques et plutoniques de types S du Victoria central (Australie). Les courbes de saturation de la biotite se redressent fortement entre 800 et 850°C pour des quantités d'eau dans le bain de fusion allant de 2 à 10% (en poids). La cristallisation hâtive de la biotite déficiente en fluor en présence du plagioclase (jusqu'à 3 kbar) et du quartz (à pressions plus élevées requiert des teneurs en eau de plus de 2.5%. L'orthopyroxène possède un domaine de cristallisation étendu; son absence dans la plupart des roches granitoïdes indique des températures magmatiques initiales relativement basses ou une réaction orthopyroxène-biotite avec chute de T. La cordiérite apparaît près du liquidus à 1 ou 2 kbar, mais seulement en dessous pour $P > 5$ kbar. Un grenat riche en almandin s'approche du liquidus pour $P \geq 4$ ou 5 kbar. À la lumière des données expérimentales, des séquences de cristallisation et de calculs géothermométriques et géobarométriques, les roches étudiées seraient issues d'un magma contenant entre 3 et 5% d'eau en présence des premières phases cristallines (grenat, biotite, orthopyroxène, plagioclase) à $T \leq 850^\circ\text{C}$, $P \geq 4$ kbar. Ces magmas de type S, de température relativement élevée et sous-saturés en eau, auraient pris naissance par fusion partielle de matériaux légèrement hyperalumineux dans des conditions de facies granulite. On se sert d'équilibres calculés de réactions de déshydratation et de fusion pour illustrer la fusion partielle d'assemblages à biotite ou à muscovite pour $a(\text{H}_2\text{O}) < 1.0$. Ces calculs, et les résultats d'expériences à 5 kbar, permettent de préciser les conditions d'anatexis et le chimisme des liquides provenant de la fusion de roches alumineuses fortement métamorphisées. Les magmas de basse température, riches en H_2O et formés par la destruction de la muscovite, ont des compositions proches du "minimum" pour un granite. Les magmas de température plus élevée, formés aux dépens d'assemblages à biotite, sont fortement sous-saturés en eau et s'écartent notablement de la composition du "minimum". Les caractéristiques thermiques et chimiques de magmas hyperalumineux qui en reflètent la source sont mises en évidence dans les mécanismes de mise en place, le chimisme et la minéralogie de roches granitoïdes de type S.

(Traduit par la Rédaction)

Mots-clés: roches granitoïdes hyperalumineuses, pétrogenèse, relations de phase, étude expérimentale, magma hydraté, anatexis, facies métamorphique granulite, granite de type S, minéralogie, biotite, cristallisation, Victoria, Australie.

INTRODUCTION

Peraluminous granitoid rocks occur as syn- and post-tectonic bodies in metamorphic belts and post-tectonic high-level plutonic-volcanic associations. Extensive peraluminous magmatism is restricted to regions of highly evolved continental crust, for example, the Tasman Mobile Belt of eastern Australia. The southern portion of this belt contains accumulations of marine sedimentary and volcanic rocks of Cambrian to Mid-Devonian age. Crustal thicknesses of 35 to 45 km in this region indicate that much of the belt is underlain by Cambrian or older basement rocks. Paleozoic granitoid rocks comprise over 30% of exposures and range in age from Ordovician to Late Devonian in age. The older batholiths are commonly developed in regional metamorphic belts, although contact-aureole types are also represented (White *et al.* 1974). Younger granitoid bodies are generally massive, discordant, high-level batholiths. In central Victoria and elsewhere, felsic volcanic complexes of similar age and mineralogy to the granites were emplaced mostly as voluminous ash-flow-tuff sequences in large subsidence structures. Both peraluminous and metaluminous granitoid and volcanic rocks are present, and some complexes contain both types. In the southeastern Australian context, Chappell & White (1974) recognized widespread associations of peraluminous granitic rocks characterized by high K/Na ratios, high initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, high SiO_2 content (66–76 wt. %) and metasedimentary inclusions. These granites were designated "S-type" to infer metasedimentary magma sources, and were distinguished from the metaluminous I-type rocks.

The geochemical characteristics of peraluminous granites control their mineralogy. Biotite is the most common ferromagnesian phase and may be accompanied by combinations of garnet, cordierite, muscovite and Al_2SiO_5 phases (Chappell & White 1974, Clarke 1981). Peraluminous volcanic rocks and, more rarely, granites, may contain orthopyroxene or fayalite or both (Birch *et al.* 1977). Ilmenite is the opaque phase characteristic of S-type rocks. Mineralogical variations reflect bulk-composition control and the $\text{P-T-f}(\text{H}_2\text{O})\text{-f}(\text{F}_2)\text{-f}(\text{O}_2)$ history of magmas during their ascent and emplacement (Clemens *et al.* 1979, Abbott & Clarke 1979). White & Chappell (1977), Flood & Shaw (1975) and Green (1976) have implied that some aspects of the mineralogy of S-type granites are inherited from the magma's source rocks. White & Chappell suggested that chemical variation

in some S-type suites may be due largely to separation of the crystalline residual restite and the granitic liquid components of the magmas.

The observed variation in mineral paragenesis within and among peraluminous granitoid suites reflects in part the diversity of physicochemical conditions during magma evolution. However, few experimental phase-equilibrium data (*e.g.*, Green 1976, 1977, Huang & Wyllie 1973) are available for peraluminous compositions; the complexity of these systems makes them difficult to model quantitatively (see, however, Abbott & Clarke 1979). In this paper we present our experimental determination of the phase relations of a natural peraluminous composition at controlled $f(\text{O}_2)$ over a broad range of $\text{P-T-X}(\text{H}_2\text{O})^{\text{melt}}$ values. These data, in conjunction with geothermometry, barometry and crystallization sequences for the natural rocks, are used to assess the crystallization history of some S-type plutonic and volcanic associations from central Victoria.

The inferred early-magmatic assemblages and conditions place strong constraints on the conditions in, and the composition of, the source region for these granites. S-type peraluminous magmas may, however, be derived from a wide range of source environments. On the basis of high-pressure crystallization experiments and dehydration-melting and phase-equilibria models, we examined the nature of probable anatectic reactions and the characteristics of the partial melts. The source-region-related thermal and chemical properties of the magmas exert a strong influence on their ascent and emplacement history and their mineralogical and chemical evolution. The inferred character of and conditions in S-type source-regions afford additional insight into the tectonic evolution of the continental crust.

EXPERIMENTAL STUDIES

Starting materials and methods

Four natural compositions in the range 69–75% SiO_2 were studied experimentally. These were selected from central Victorian, Late Devonian plutonic-volcanic complexes with S-type affinities. The geological setting, petrology and geochemistry of these complexes have been described by Birch *et al.* (1977), Clemens *et al.* (1979) and Clemens & Wall (in prep.). The phase relations of the three less silicic rocks are similar; we will present detailed results for one of these, a garnet-cordierite-biotite granite (889), representative of a major mafic variant

TABLE 1. CHEMICAL AND MODAL DATA FOR THE STARTING MATERIAL USED IN THE MELTING EXPERIMENTS

SiO ₂	70.46	Mg/(Mg + Fe)	.44
TiO ₂	.59	NORMATIVE C	2.41
Al ₂ O ₃	14.92	Q*	34
FeO	3.51	OR	31
MnO	-	AB	24
MgO	1.54	AN	11
CaO	1.92	*Q + OR + AB + AN =	100
Na ₂ O	2.50	MAFIC MINERALOGY (MODAL)	
K ₂ O	4.53	Garnet + Cordierite + Biotite	

in the Strathbogie batholith (Phillips *et al.* 1981). Chemical and modal data for the starting material are given in Table 1. The rock contains approximately 50 vol. % euhedral phenocrysts of quartz (β habit), 10-mm-sized, normally zoned plagioclase crystals (An₄₄₋₁₇), large (5 mm) biotite booklets, almandine-rich garnet (10 mm) rimmed by cordierite, and large cordierite crystals. Orthoclase phenocrysts (~ 10 mm) are present locally. The groundmass comprises fine grained (0.5–1.5 mm) quartz, orthoclase, oligoclase, minor biotite and cordierite. Accessory phases include ilmenite, pyrrhotite, zircon, apatite, monazite and xenotime.

Strathbogie granite 889 was chosen for study in view of its well-established crystallization sequence (Clemens *et al.* 1979, Phillips *et al.* 1981), and the widespread development of similar cordierite–biotite granites. As 889 is a relatively SiO₂-poor, calcic composition, it could represent a restite-rich rock. The paucity of inclusions and absence of other petrographic features that could be ascribed to a restite origin suggest that 889 bears little or no restite component. It is probable, however, that the rock contains an accumulation of early magmatic phases (garnet, biotite, plagioclase and quartz). Thus, the experimental starting material may not represent a "primary" melt composition; this is a problem endemic to most experimental studies of igneous rock series (O'Hara 1968). Although the experimentally determined stability curves for early-crystallized phases may be biased towards higher T or lower $a(\text{H}_2\text{O})$, the experimental data indicate the conditions under which the granitic melts are saturated with these phases.

Anhydrous starting glasses were seeded with 1% natural cordierite and almandine. Seeding was necessary to overcome the apparent difficulty in nucleating garnet and cordierite. Some

near-liquidus experiments were conducted on rock powder (< 10 μm grain size). The crystallization experiments were carried out in large-volume, internally heated apparatus with argon pressures of 1–7 kbar ($P = \pm 0.05$ kbar) and temperatures in the range 700–900°C ($T = \pm 10^\circ\text{C}$), and with run times of 100–500 hours. Water activities [$a(\text{H}_2\text{O}) = 0.1\text{--}1.0$] were controlled by a CO₂–H₂O vapor phase, generated in the Ag₇₀Pd₃₀ capsules by silver oxalate–water mixtures. (The activity of H₂O at any P and T is here defined as $f(\text{H}_2\text{O})/f^\circ(\text{H}_2\text{O})$, where $f(\text{H}_2\text{O})$ is the fugacity of H₂O in the system at P and T and $f^\circ(\text{H}_2\text{O})$ is the fugacity of pure H₂O at the same P and T.) Fluid/rock mass ratios were ≥ 1 [for $a(\text{H}_2\text{O}) < 1$]; water contents of the melt were calculated using the models of Burnham (1975, 1979). Adjustments were made for H₂O dissolved in the melts and the proportion of crystalline materials present. These corrections and the probable limited deviations from ideality of H₂O in CO₂–H₂O mixtures lead to maximum uncertainties of ± 0.5 wt. % H₂O (melt). Hydrogen fugacity was controlled, in all runs, by a modified Shaw membrane to give $f(\text{O}_2)$ in the range QFM to QFM – 0.5 log $f(\text{O}_2)$. Phase boundaries proved insensitive to $f(\text{O}_2)$ in this range, chosen to match the reduced character of, and inferred low $f(\text{O}_2)$ in, the natural magmas (Clemens *et al.* 1979, Phillips *et al.* 1981). Runs were quenched isobarically; products (Appendix) were examined optically, by X-ray powder diffraction and by an electron microprobe and an SEM equipped with solid-state X-ray detectors. Owing to the fine grain-sizes of experimental products, reliable microprobe analyses of crystalline phases could be obtained only for high-temperature, near-liquidus experiments. Quench crystallization appeared to be restricted to the development of rare, small aggregates of a mica phase, readily distinguishable from the primary biotite. Quench glasses are homogeneous on the 20 μm scale. For near-liquidus runs at $T \approx 850^\circ\text{C}$, iron loss was limited to about 10 wt. % of total iron.

It is difficult to demonstrate the approach of the experimental products to equilibrium, especially for these complex compositions at relatively low temperatures (Piwinski & Martin 1970, Johannes 1978). The presence of crystalline seed material in most runs should have negated nucleation difficulties. Phase boundaries were located, in part, by overgrowth on and corrosion or disappearance of seeds. High-temperature, near-liquidus experiments on fine grained rock powder in all cases gave phase

relations and proportions similar to those in experiments with seeded glass materials. Phase boundaries (with the possible exception of orthopyroxene) are smooth in P - T - $X(\text{H}_2\text{O})^{\text{melt}}$ plots and vary predictably with these parameters. Microprobe analyses of some near-liquidus ferromagnesian assemblages gave reasonable Fe/Mg partition values, although orthopyroxene grains are zoned and have somewhat variable Al contents. Not all the reactions were reversed; hence the phase boundaries should be judged accordingly. Further runs on compositions synthesized to give high proportions of liquid saturated with small amounts of crystalline material are underway to demonstrate reversibility.

Experimental results

Detailed results of the experiments on the

Strathbogie rock are given in the Appendix along with a description of the phases and methods of identification. Figures 1, 2 and 3 show an interpretation of experimental phase relations for the Strathbogie granite composition at 1, 2 and 5 kbar, respectively, as a function of temperature and $X(\text{H}_2\text{O})^{\text{fluid}}$ (= moles H_2O in fluid/total moles $\text{H}_2\text{O} + \text{CO}_2$ in fluid) or water content of melt. Each of these diagrams represents about 20 experimental points. The 1- and 2-kbar data are useful for assessing the phase relations at the stage of emplacement. The 5-kbar diagram models early, deep-seated chemical processes.

Predictably, the reaction boundaries of the tectosilicates sweep to high temperatures with decreasing $a(\text{H}_2\text{O})$ at constant pressure. At $P < 3$ or 4 kbar, plagioclase exhibits the highest-temperature saturation boundary of these phases, whereas quartz assumes this position at

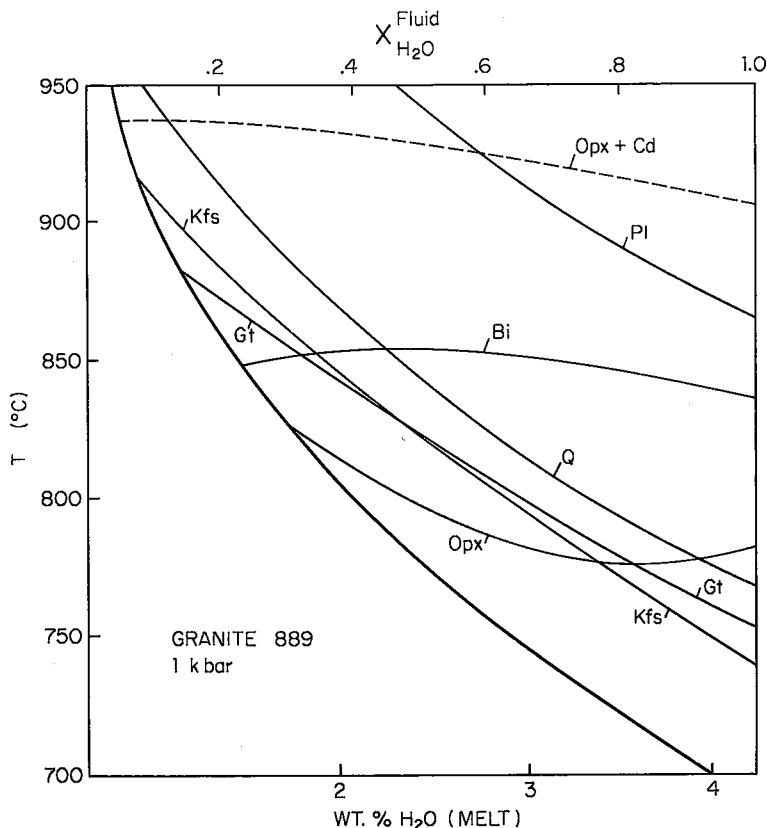


FIG. 1. Temperature versus $X(\text{H}_2\text{O})^{\text{fluid}}$ /wt. % H_2O (melt) diagram for the Strathbogie granite composition at 1 kbar for $\log f(\text{O}_2)$ in the range (QFM to QFM - 0.5). Q quartz, Kfs alkali feldspar, Pl plagioclase, Bi biotite, Cd cordierite, Gt garnet, Opx orthopyroxene.

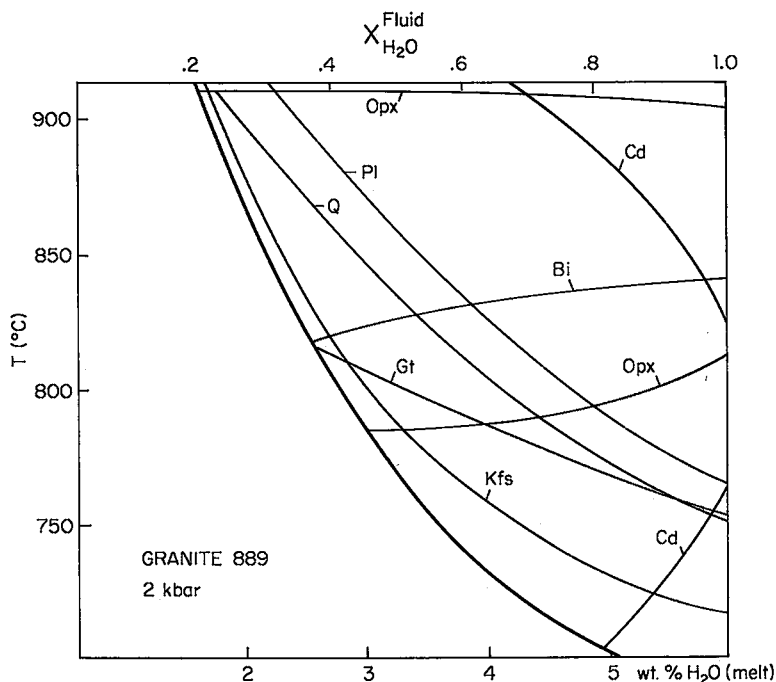


FIG. 2. Temperature versus $X(\text{H}_2\text{O})^{\text{fluid}}/\text{wt. \% H}_2\text{O}$ (melt) diagram for the Strathbogie granite composition at 2 kbar for $\log f(\text{O}_2)$ in the range (QFM to QFM - 0.5).

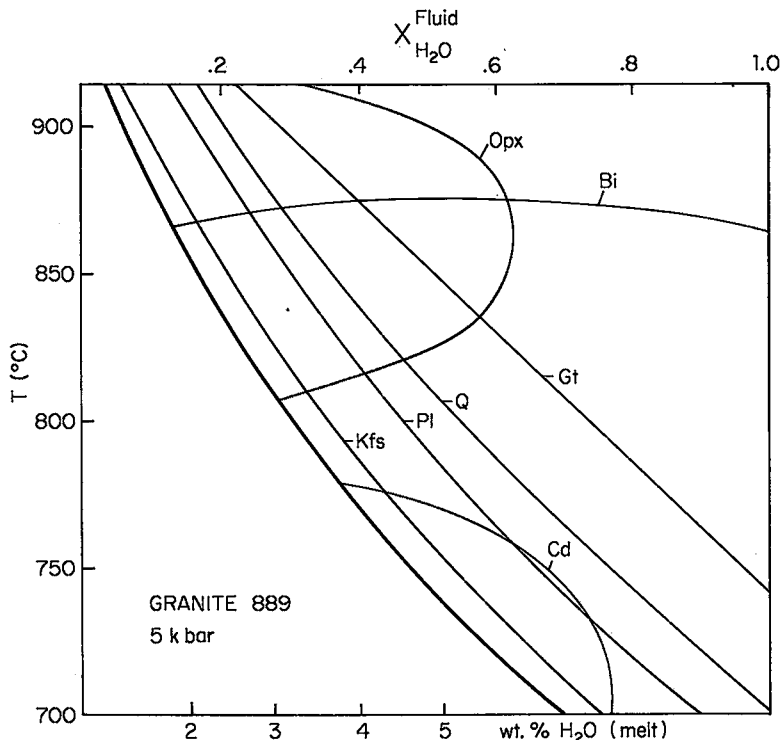


FIG. 3. Temperature versus $X(\text{H}_2\text{O})^{\text{fluid}}/\text{wt. \% H}_2\text{O}$ (melt) phase diagram for the Strathbogie granite composition at 5 kbar for $\log f(\text{O}_2)$ in the range (QFM to QFM - 0.5).

higher pressures. K-feldspar exhibits the lowest-temperature saturation boundary, although the bulk composition is still about 60% liquid at this phase boundary. Solidus loci are subject to large uncertainties.

Of the ferromagnesian phases, ilmenite is stable to the highest temperatures ($T > 900^{\circ}\text{C}$). Orthopyroxene has a broad field of crystallization ($780\text{--}900^{\circ}\text{C}$) at low pressure, which contracts markedly at high pressure to a region around 800°C . At 1–2 kbar, cordierite crystallizes over a wide range of temperatures and melt water-contents. At 5 kbar, cordierite crystallization is restricted to low-temperature, near-solidus conditions. The saturation boundary of garnet at low pressure occurs at relatively low temperatures and high $X(\text{H}_2\text{O})^{\text{fluid}}$. Garnets developed at 1 and 2 kbar are relatively spessartine-enriched, averaging $\text{Alm}_{70}\text{Sp}_{10}\text{Py}_8\text{Gr}_2$. At $P \geq 5$ kbar, garnets crystallize over a wide T – $a(\text{H}_2\text{O})$ range. Near-liquidus 5-kbar garnets are around $\text{Alm}_{80}\text{Sp}_7\text{Py}_{10}\text{Gr}_3$. Hercynite-rich spinels formed in some runs at $T \geq 800^{\circ}\text{C}$ where compositions were seeded with sillimanite.

Biotite saturation boundaries (T in the range $800\text{--}900^{\circ}\text{C}$) have relatively flat $[\partial T/\partial a(\text{H}_2\text{O})]$ (P) slopes, especially at 1 or 2 kbar. At $P \geq 5$ kbar, the biotite saturation boundaries occur at $\geq 850^{\circ}\text{C}$ and have more pronounced curvature. The biotite phase boundaries proved insensitive to $f(\text{O}_2)$ in the range QFM to QFM – $0.5 \log f(\text{O}_2)$. Electron-microprobe analyses indicate that near-liquidus biotites are relatively aluminous and Ti-rich. It should be noted that the Strathbogie bulk composition is fluorine-

poor. The synthetic biotites contain less than 0.5 wt. % fluorine. In compositions similar to the Strathbogie material but somewhat fluorine-enriched, the biotite saturation-boundary would occur at a higher temperature.

As an example of the H_2O -undersaturated relations of the Strathbogie rock, Figure 4 shows the P – T projection of phase boundaries for the 4 wt. % H_2O (melt) isopleth. Above 1 kbar, the saturation boundaries of all phases except cordierite have positive dP/dT slopes, with quartz and garnet exhibiting the shallowest positive slopes. The crystallization field of orthopyroxene envelops the biotite boundary and narrows markedly at high pressure. Restriction of the cordierite field to below 6 kbar is apparent. For $P > 2$ kbar and melt water-contents in the range 2 to 6 wt. %, the relative positions of the phase boundaries of anhydrous silicates change little, although the orthopyroxene field is considerably expanded at lower melt water-contents. For water contents ≤ 2 wt. % H_2O , biotite is a near-solidus phase, in strong contrast to its crystallization near the liquidus for 4 to 6 wt. % H_2O (melt).

Discussion of the experimentally determined phase relations

The relative locations of plagioclase, K-feldspar and quartz saturation boundaries broadly agree with relations in haplogranitic systems and thermodynamic models. Although the Strathbogie composition is relatively potassic, the plagioclase saturation boundary occurs at a relatively high temperature owing to the high Ca/Na of the rock. The broadening of the crystallization field of quartz at high pressures has been observed in haplogranitic systems for $a(\text{H}_2\text{O}) = 1.0$ (Luth 1976) and can be predicted from the thermodynamic models of Burnham (1979). This behavior is consistent with the fact that the source rocks are likely to be quartz-saturated. It is difficult to compare our results directly with those of Whitney (1975) for his R1 composition (which most closely resembles the Strathbogie rock), owing to differing methods of determining melt water-contents. However, it appears that for similar melt water-contents our plagioclase boundaries occur at lower temperatures; also, the quartz crystallization field is broader and the K-feldspar field narrower in our experiments. The higher-temperature quartz boundary in our experimental results reflects the higher normative quartz content of our starting composition. Calculations using the melt activity-com-

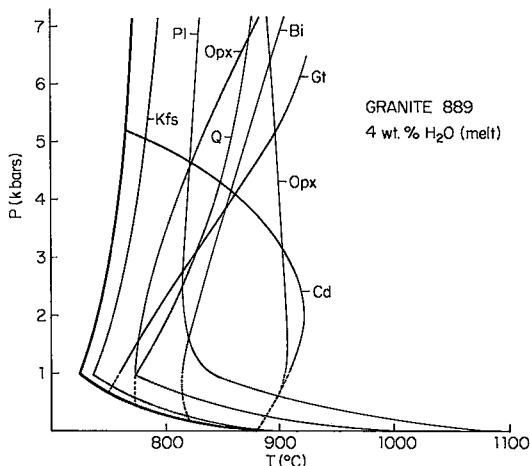


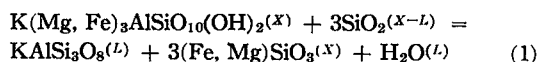
FIG. 4. Pressure–temperature projection of phase boundaries for the Strathbogie composition with 4 wt. % H_2O in the melt.

position models of Burnham (1979) suggest that our plagioclase boundary is in reasonable agreement with the data for simple anorthite-quartz melting. We suggest that the position of Whitney's plagioclase saturation boundary reflects the kind of kinetic problems encountered by Johannes (1978, 1980) and that these difficulties are less pronounced in compositionally more complex systems.

Turning to the ferromagnesian phases, the diminution of the crystallization field of cordierite with increasing pressure may be predicted from its subsolidus relations (Hensen & Green 1973), as was discussed by Abbott & Clarke (1979). Cordierite with $Fe/(Fe + Mg) \approx 0.5$, such as is commonly observed in S-type granites, is thus highly unlikely to be a restate phase (*cf.* Flood & Shaw 1975), although more magnesian cordierites could occur in source-region residual materials. The experimental phase relations support the contention of Phillips *et al.* (1981) that, in Victorian granitoid rocks, cordierite crystallization occurred mainly at low pressures.

The development of almandine-rich garnet at pressures as low as 1 kbar is not in accord with Green's (1976) contention that garnetiferous granites necessarily derive from $P > 5$ kbar. Green's experimental charges were mostly saturated with Al_2SiO_5 , and thus the garnet field was restricted to high pressures by the stability of cordierite below 4 to 5 kbar. The relatively high $Fe/(Fe + Mg)$ ratio of the Strathbogie composition also enhances the stability of almandine-rich garnet relative to cordierite-hypersthene or cordierite-biotite. Furthermore, the low-pressure garnets are somewhat Mn-enriched (see also Green 1977). In this connection, near-solidus, almandine-rich garnet was predicted by Abbott & Clarke (1979) on topological grounds (their reaction 14). It is suggested, however, that the lower $Fe/(Fe + Mg)$ ratios and high excess Al_2O_3 of some S-type magmas may prevent the crystallization of garnet at low pressures.

The fact that the crystallization field of orthopyroxene envelops that of biotite suggests that orthopyroxene may appear as the result of incongruent melting of biotite:



The low-temperature boundary of the orthopyroxene crystallization field occurs between 780 and 800°C (1–2 kbar) in the three compositions studied. Naney (1978) found similar temperatures (750–800°C at 2 kbar) for this

phase boundary in two other compositions. Granitoid rocks lacking orthopyroxene may have crystallized from relatively low-temperature magmas. Alternatively, the absence of orthopyroxene may result from complete reaction with the magma to form biotite during crystallization at temperatures below 780°C. Clemens & Wall (in prep.) noted that orthopyroxene occurs in the central Victorian volcanic rocks because reaction to biotite was halted by eruption. For $P = 2$ kbar, the Strathbogie biotite has a phase boundary similar in shape (in $T-X(H_2O)^{fluid}$ projection) but is about 50°C lower than that reported by Maaløe & Wyllie (1975). The biotite-out curves of Sykes & Holloway (1978) and Naney (1978) are steeper in the $T-a(H_2O)$ projection and straddle our own. Differences in experimental conditions, techniques and bulk compositions may explain these discrepancies. Higher $f(O_2)$ ($\sim Ni-NiO$) in the experiments of Maaløe & Wyllie, Sykes & Holloway and Naney and the lower $Fe/(Fe + Mg)$ ratios of their experimental materials may result in their biotites being more magnesian and thus stable to higher temperatures. The relatively high $a(SiO_2)$ over much of the experimental crystallization range of the Strathbogie granite tends to decrease biotite stability, although this effect may be offset by high $a(KAlSi_3O_8)$ in the K-rich Strathbogie composition (see reaction 1). Indeed, if the biotite reaction has a stoichiometry similar to that of reaction 1, calculations using the melt activity – composition models of Burnham (1979) show that the relatively flat $[\partial T/\partial a(H_2O)](P)$ slope of the biotite liquidus is attributable to the competing effects of the activities of SiO_2 , $KAlSi_3O_8$ and H_2O components. The contrasting behavior of biotite and hornblende saturation boundaries in felsic compositions as a function of $f(H_2O)$ (Sykes & Holloway 1978) reflects the differences in stoichiometry of the melting reactions for these phases.

Our experimental results suggest that biotite can coexist with peraluminous melts over a wide range of temperatures and melt water-contents. Early crystallization of biotite seems to be characteristic of S-type rocks, even for relatively high-temperature magmas (*e.g.*, Clemens *et al.* 1979). Late crystallization of biotite in many I-type granitoid rocks (*e.g.*, Bateman & Nokleberg 1978) may reflect the lower $a(KAlSi_3O_8)$ early in their crystallization histories and also the decrease in $a([Mg,Fe]SiO_3)$ due to the presence of hornblende. We also suggest that the substitution $Ti^{4+} + 2O^{2-} = (Fe, Mg)^{2+} + 2OH^-$ (Bohlen *et al.* 1980b) is extensive in S-

type biotites and that the lowered OH^- content leads to an increase in the stability of these micas. The synthetic biotites are probably Al-Si disordered; the entropy contribution from disordering would serve to stabilize disordered biotite relative to ordered variants. The importance of Al-Si ordering effects on the stability of natural biotites in the magmatic environment are, however, unknown.

F-OH substitution may markedly increase the stability of biotite relative to anhydrous crystalline assemblages (e.g., Munoz & Luddington 1974) and probably also to melts (Anfilogov *et al.* 1977). Hence, the saturation boundary for fluorine-enriched biotite would occur at higher temperatures than for our experimental data; the crystallization field of orthopyroxene would be smaller. Increasing $f(\text{HF})$ would also expand the field of coexisting biotite- Al_2SiO_5 in granitic magmas at the expense of assemblages containing cordierite and garnet. Topaz would develop at relatively high $f(\text{HF})$.

APPLICATIONS OF THE EXPERIMENTAL RESULTS

The experimental data may be combined with crystallization sequences, geothermometry and barometry for the central Victorian suites to clarify the ascent and emplacement history of the parent magmas.

P-T-X(H₂O) conditions of magma generation

Mutual inclusion relationships among phenocryst phases and modal variation in phenocryst suites and cognate xenoliths were utilized by Phillips *et al.* (1981) and Clemens & Wall (in prep.) to characterize the crystallization sequences in central Victorian granites and rhyodacites. They inferred that quartz, garnet, orthopyroxene and calcic plagioclase were the earliest crystallizing phases. Biotite saturation also occurred at an early stage, followed by cordierite, the latter phase forming largely at low pressures. K-feldspar was the last phase to begin crystallizing. A similar paragenetic sequence was inferred for the Strathbogie granite, although unaltered hypersthene was not observed. Application of several geothermometers to phenocryst assemblages in the volcanic rocks (Clemens *et al.* 1979) indicates the beginning of crystallization in the range 800–850°C, compatible with the experimentally determined crystallization field of orthopyroxene. Early-crystallized plagioclase-garnet-Ca-poor pyroxene \pm fayalite assemblages formed at pressures of > 3–4 kbar. Some volcanic rocks contain almandine-rutile as-

semblages, indicating pressures of \geq 7–8 kbar.

If the above estimates of early magmatic conditions (800–850°C, 4–7 kbar) for the central Victorian rocks are accepted, then their sequences of crystallization are best matched around 5 kbar for melt water-contents of 3–5 wt. % (Fig. 3). For similar P-T conditions and 2% water in the melt, the Strathbogie composition would be near its solidus even if magma temperatures were above 850°C and biotite could only crystallize late in the solidification history. Water contents much above 5 wt. % would not permit the melt to coexist with quartz for the early magmatic P-T range. Such a situation seems unlikely in view of the probable quartz-saturated nature of the source materials of the magma. Our melt water-contents estimates are considerably higher than the 1–2% magma water-contents inferred by Maaløe & Wyllie (1975) for the Bohaus granite, in which, unlike most S-types, biotite appears late in the crystallization history. The apparent discrepancy is reduced if some account is taken of the proportion of crystalline material when inferring the melt water-content in their runs. Our inferred melt water-contents are also considerably higher than those suggested by Wyllie *et al.* (1976) for batholith-forming I-type granitic magmas.

Ascent and emplacement of magmas

We shall now consider the ascent and emplacement of the central Victorian S-type magmas in the light of their crystallization histories. Taking a melt water-content of 4 wt. % as representative, an adiabatic ascent path (for vapor-absent conditions) would have an average gradient of 3°C/kbar. This gradient is steeper than those of the silicate saturation boundaries (Fig. 4) and would lead to partial resorption or reverse zoning of crystalline phases. However, petrographic evidence, such as 1) the presence of quartz throughout much of the magma crystallization history, 2) the normal (though slightly oscillatory) zoning of plagioclase and also 3) the range of pressure deduced for garnet development throughout ascent, implies that these relatively viscous magmas cooled along a shallower dP/dT gradient than for adiabatic ascent (*cf.*, Marsh 1978).

Birch & Gleadow (1974) and Birch *et al.* (1977) suggested that extensive magmatic differentiation produced a vertically zoned subvolcanic magma chamber below the Cerberean cauldron complex (central Victoria). On the basis of reaction relations of early-crystallized phases and geothermometry-barometry, Clemens

& Wall (in prep.) arrived at a similar conclusion for the Violet Town volcanic suite. Pre-eruption temperatures of around 750°C may be inferred from the phase diagram (Fig. 2), as the rhyodacite magmas attained K-feldspar saturation and were, on average, about 60% crystalline when erupted. For our inferred initial magma, water-contents of ~ 4 wt. %, cooling and crystallization of the system at 1–2 kbar would have brought about water saturation and second boiling. The volume change and pressure increase during vapor evolution must have contributed to the explosive character of the eruptions (Burnham 1979), resulting in magma emplacement as ignimbrite sheets. Holloway (1976) has pointed out that a small amount of CO₂ dissolved in the melt at high pressure may result in vapor evolution at pressures well above those for vapor saturation of CO₂-free magma. However, Holloway's model is still consistent with exsolution of the bulk of the melt H₂O at low pressures.

Chemical evolution of the magmas

We shall now examine some of the implications of the experimental studies for the chemical evolution of the central Victorian peraluminous magmas. Given our estimates of melt water-contents (~ 4 wt. %) for early magmatic conditions of $P > 5$ kbar and T in the range 800–850°C, compositions such as the Strathogie granite experimental material were more than 85% liquid at this stage. This conclusion is consistent with the contention of Clemens & Wall (1979) that the central Victorian magmas contained only a small proportion of xenolithic and xenocrystic materials that have petrographic or chemical characteristics (White & Chappell 1977) indicative of a restite origin. Restite-controlled differentiation in the central Victorian suites can, therefore, account for only a limited portion of the chemical variation. Under the above conditions, minerals compatible with the melts, whether primary or restitic, include garnet, orthopyroxene, relatively calcic plagioclase, biotite, ilmenite and quartz. The experimentally observed persistence of these phases to temperatures above the saturation boundary of quartz may indeed reflect the presence of some of these phases as restites. The presence of a restite component would result in the magmas having higher Fe, Mg, Ti and Ca contents and lower SiO₂ contents than the melt alone.

Microprobe analyses of 5-kbar run products indicate that Fe/(Fe + Mg) of phases have the following approximate relative values: ilmenite

(0.9) > garnet (0.8) > glass (0.7) (\approx melt) > orthopyroxene (0.6) > biotite (0.5) > cordierite (0.4). Plagioclase is approximately An₄₆. Fractionation of garnet could lead to a decrease in the Fe/(Fe + Mg) of residual liquids, whereas cordierite fractionation would have the opposite effect. For compositions similar to our experimental materials, the relatively aluminous character of the biotite precludes crystallization of more than a few percent garnet. Thus fractionation of garnet (at high pressure) could have only minor effects on major-element chemistry, although heavy REE in residual liquids could be markedly depleted. Cordierite crystallization is limited largely to low pressures. Thus, cordierite fractionation would favor late crystallization of almandine-rich garnet. Fractionation of orthopyroxene and biotite should result in a less pronounced increase in the Fe/(Fe + Mg) of the remaining melts, in keeping with the observed trends for central Victorian rocks (*e.g.*, Clemens *et al.* 1979). The Ti-rich character of the biotites, in conjunction with ilmenite, can account for the observed depletion of TiO₂ with increased SiO₂ content. Fractionation of plagioclase may be inferred to cause a decrease in Ca/(Na + K) in the more SiO₂-rich rocks in the suite.

Processes that lead to fractionation by the separation of melt and crystals (*e.g.*, crystal settling, variation in flow regimes of magma and inward crystallization of magma bodies) lead to accumulation of crystalline material. Rocks formed by the solidification of such crystal-enriched magma will have chemical characteristics reflecting their "cumulate" nature (McCarthy & Hasty 1976, McCarthy & Groves 1979). The starting materials used in this study (all relatively SiO₂-poor compositions) may have contained accumulations of early-crystallized phases. The experimental phase boundaries may thus be biased towards high temperature or high water-contents of the melt, reflecting the presence of refractory "cumulate" material.

Peraluminous melts may result from fractionation of diopside-normative phases from metaluminous magmas (*e.g.*, hornblende: Cawthorn & Brown 1976). Green (1978) argued that this hypothesis cannot account for many of the characteristics of S-type magmas, but Cramer (1979) has suggested that hornblende fractionation contributed to the development of slightly peraluminous rocks in the Mount Dandenong igneous complex, central Victoria. We observed no evidence of either clinopyroxene or hornblende among early-crystallized material

S-TYPE MAGMA SOURCES

or xenoliths in the rocks under consideration here. Neither amphibole nor clinopyroxene crystallized in our experiments. This seems unlikely to reflect nucleation problems with these phases, which appear to grow readily in a wide range of less aluminous, intermediate to acid compositions (I.A. Nicholls, pers. comm. 1979). Although hornblende could not form in equilibrium with the low-pressure, cordierite-bearing magmatic assemblages, it is conceivable that it could coexist with orthopyroxene-plagioclase (\pm garnet) assemblages at higher pressures. It remains to be ascertained whether hornblende is stable with relatively Ca-poor garnet at $T > 800^\circ\text{C}$ for bulk compositions having high $a(\text{SiO}_2)$. The presence of Ca-poor garnet precludes the presence of calcic pyroxenes in the early magmatic environment inferred for the central Victorian associations.

Inferences from melt-crystal-saturation relations

The P-T- $X(\text{H}_2\text{O})$ relations inferred from the early crystallization history of the central Victorian S-type magmas place strong constraints on the nature of, and conditions in, the source regions of the magma. In particular, muscovite-bearing assemblages cannot have been present at the high temperatures inferred for magma genesis. Partial-melting reactions involving muscovite produce liquids with water contents (>8 wt. %) markedly in excess of those suggested for the Victorian magmas. In addition, since these magmas attained K-feldspar saturation late in their crystallization history, we would conclude that K-feldspar may not have been a residual phase in the source region. This could imply relatively K_2O -poor source rocks in which K_2O was mainly accommodated in biotite. Elimination of K-feldspar is unlikely during partial melting of typical pelitic rocks owing to their high K_2O contents and $\text{K}/(\text{Na} + \text{Ca})$ ratios. Such highly aluminous rocks would commonly contain an Al_2SiO_5 phase at high metamorphic grade.

In view of the common occurrence of gneissic pelitic xenoliths in S-type granites and the widely held hypothesis that S-type magmas are derived from such parental materials, the melts should be chemically compatible with Al_2SiO_5 or other highly aluminous assemblages. For a given set of P-T- $f(\text{H}_2\text{O})$ conditions and in the presence of quartz, $a(\text{Al}_2\text{O}_3)$ is maximized, at low $a(\text{HF})$, by Al_2SiO_5 ; coexisting melts should exhibit the most peraluminous character. To examine those relationships we seeded the experimental compositions with finely ground sillimanite and quartz. Experiments aiming at saturation with these phases were conducted at 5 kbar, T between 750 and 850°C and for a range of $a(\text{H}_2\text{O})$.

In all runs, sillimanite seeds were partly or completely eliminated, and abundant almandine-rich garnet nucleated and grew. Biotite-quartz assemblages in run products were restricted to below 800°C, and hypersthene-sillimanite assemblages were not observed. The normative corundum content of glasses is given in Figure 5 as a function of $X(\text{H}_2\text{O})^{\text{fluid}}$ and temperature, for $P = 5$ kbar and for both sillimanite-saturated and -undersaturated assemblages. Initial attempts at electron-microprobe analysis of the quenched glasses demonstrated their apparent depletion in Na and Mg. There-

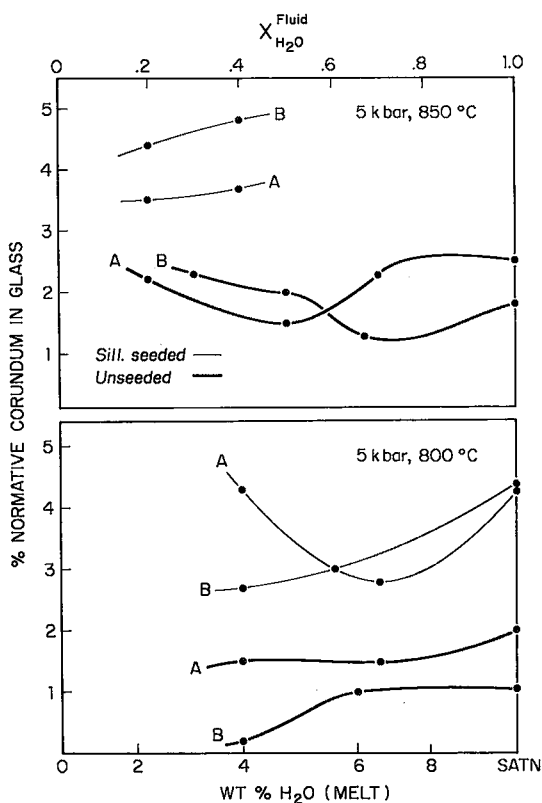


FIG. 5. Weight percent normative corundum in sillimanite-saturated (dashed curves) and -undersaturated (continuous curves) experimental glasses as a function of $X(\text{H}_2\text{O})^{\text{fluid}}/\text{wt. \% H}_2\text{O}$ (melt) at 5 kbar. A and B refer to the Strathbogie granite and Lake Mountain rhyodacite starting materials, respectively.

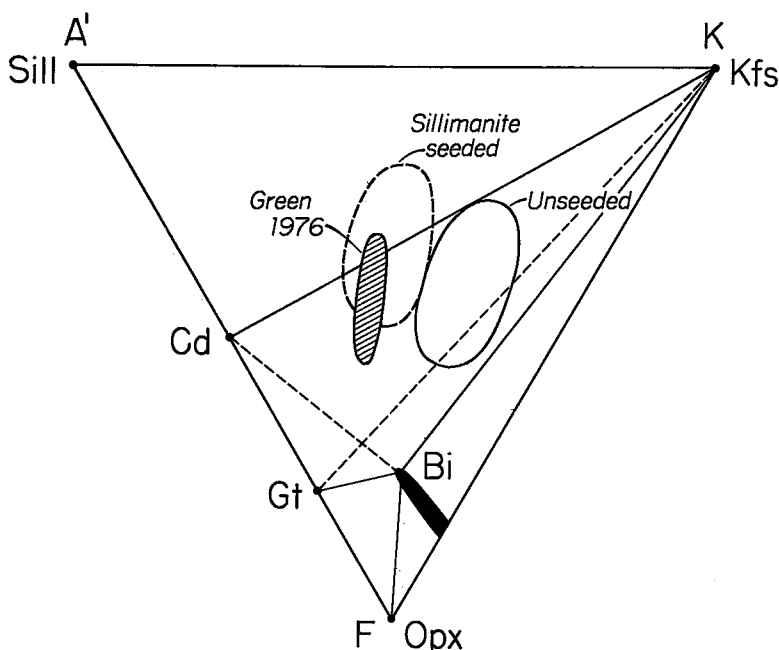


FIG. 6. Compositions of experimental glasses (5 kbar, 800 and 850°C) on A'KF projection. A' = $(\text{Al}_2\text{O}_3\text{--Na}_2\text{O--K}_2\text{O--CaO})$, K = K_2O , F = $(\text{FeO} + \text{MgO})$. Compositions of glasses from Green's (1976) melting experiments on a pelitic material are also plotted. Data are plotted on a molar basis.

fore, glasses of similar H_2O content were used as secondary standards to normalize the analytical data. Although the absolute normative corundum values of the glasses are subject to considerable error, the data in Figure 6 demonstrate that the normative corundum content of the sillimanite-bearing glasses is considerably higher than that of the unseeded runs. Normative corundum increased with both temperature and melt water-content. The normative corundum contents for sillimanite-saturated glasses given in Figure 6 are markedly lower than those calculated from Green's (1976) data. This reflects the lower temperature of our experimental runs and perhaps sodium depletion during Green's electron-microprobe analyses.

Figure 6 shows an A'KF projection of glass compositions respectively saturated and undersaturated with Al_2SiO_5 . These data indicate that melts initially saturated with Al_2SiO_5 at 5 kbar and 750–850°C must, on separation from their parent rocks, eventually crystallize 5 to 10 modal % (relative) garnet, cordierite and even some Al_2SiO_5 . Peraluminous melts that are undersaturated with respect to Al_2SiO_5 may crystallize biotite and hypersthene with or without garnet and cordierite. Normative corun-

dum data for southeastern Australian S-type granitoid rocks and the experimental glasses are plotted in Figure 7. This diagram highlights the differences between magmas undersaturated in Al_2SiO_5 , those that may have coexisted with sillimanite, and rocks that are so peraluminous that, in the absence of alteration effects, they may be presumed to contain abundant Al-rich restite material or an accumulation of peraluminous crystalline phases.

The moderately peraluminous character of the central Victorian magmas may reflect their derivation from source regions that were mainly undersaturated in Al_2SiO_5 . Some metapelitic rocks were probably present, accounting for the scarce gneissic pelitic xenoliths. Appropriate source rocks with assemblages compatible with the magmas (e.g., quartz-plagioclase-biotite-garnet-orthopyroxene) may have been somewhat immature quartzofeldspathic sediments with compositions broadly akin to mildly weathered dacitic materials. Huge volumes of such lithologies occur in some continental margins or highly evolved arc systems (e.g., New Zealand, North Island). In the central Victorian province, large volumes of source rocks appropriate for the granitic complexes are not

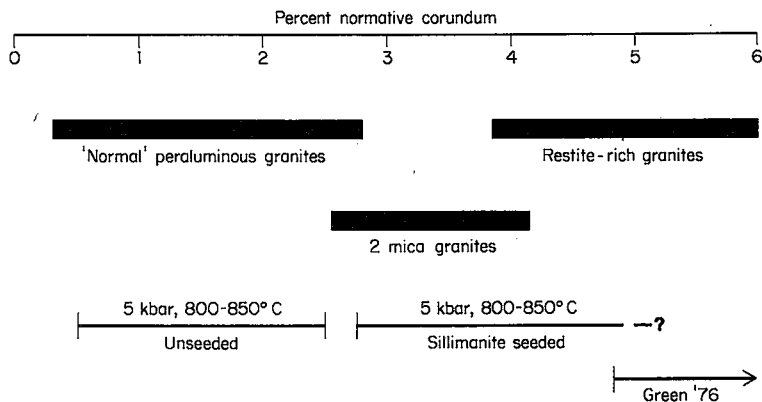


FIG. 7. Comparison of the normative corundum contents of some south-eastern Australian S-type granitoid rocks with those of experimental glasses.

presently exposed. Hence we may infer that thick Cambrian or Late Proterozoic sections may underlie the Paleozoic rocks (see Rutland 1973, 1979, Compston & Chappell 1979, Wyborn & Chappell 1979). These rocks apparently did not attain high-grade metamorphic conditions compatible with extensive partial fusion until Devonian times.

The P - T - $a(\text{H}_2\text{O})$ conditions inferred for the early history of the central Victorian magmas require partial melting of their source region under low- to medium-pressure granulite-facies conditions. In view of the 40+ km thickness of continental crust beneath central Victoria (Finlayson 1979), the high geothermal gradient required to effect partial melting requires uppermost mantle or lower crustal high-temperature heat sources (Packham 1979, Wells 1980). Emplacement of mafic magma is the most plausible source for the heat required (Zeck 1970, Presnall & Bateman 1973).

The role of the H_2O component in partial melting

Quartz- and feldspar-bearing rocks are abundant in most high-grade metamorphic terrains and provide ample silicate component for granitic partial melts. Burnham (1967), Fyfe (1969) and others have suggested that the availability of H_2O in anatexis environments will control the degree of partial melting and the nature of the melting reactions. These factors will, in turn, influence the chemical character of the melts and residual crystalline materials. There are two main possibilities for the source of the H_2O component essential for the development of hydrous granitic melts. (1) An aqueous

fluid may be present (at least at the onset of melting) in the source region. This situation may be common under upper-amphibolite-facies conditions, where $a(\text{H}_2\text{O})$ appears to be high (Tracy 1978, Bohlen & Essene 1979, Phillips & Wall 1979). However, most granitic magmas are H_2O -undersaturated (from early in their history), and an aqueous fluid coexisting with the melt would necessarily contain other components, which would reduce $a(\text{H}_2\text{O})$. CO_2 and CH_4 are the only components likely to be present in sufficient quantities to markedly reduce $a(\text{H}_2\text{O})$. In the case of S-type source-regions, C-bearing components could be derived from metasedimentary graphite, the presence of which would explain the highly reduced character of most S-type magmas. Deep crustal or mantle sources could also furnish C-bearing components for the source region (Holloway 1976, Schuiling & Kreulen 1979). (2) Most or all of the H_2O required for partial melting may be derived from the breakdown of crystalline hydrates (Fyfe 1969). Anatexis could thus occur under vapor-absent conditions.

It seems unlikely (to us) that large volumes of aqueous fluids are present during high-grade regional metamorphism. In view of the low porosities of high-grade metamorphic rocks, it is difficult to envisage a reservoir for these fluids. Further, the character of high-grade metamorphic assemblages suggests internal control of $f(\text{H}_2\text{O})$ (Fyfe 1973). Therefore, in discussing partial melting of possible S-type source-materials, we place particular emphasis on vapor-absent conditions. Eggler (1973) noted that such reactions are a special case of hydrate stability at reduced $a(\text{H}_2\text{O})$. However, in a situation in which only a small amount of fluid

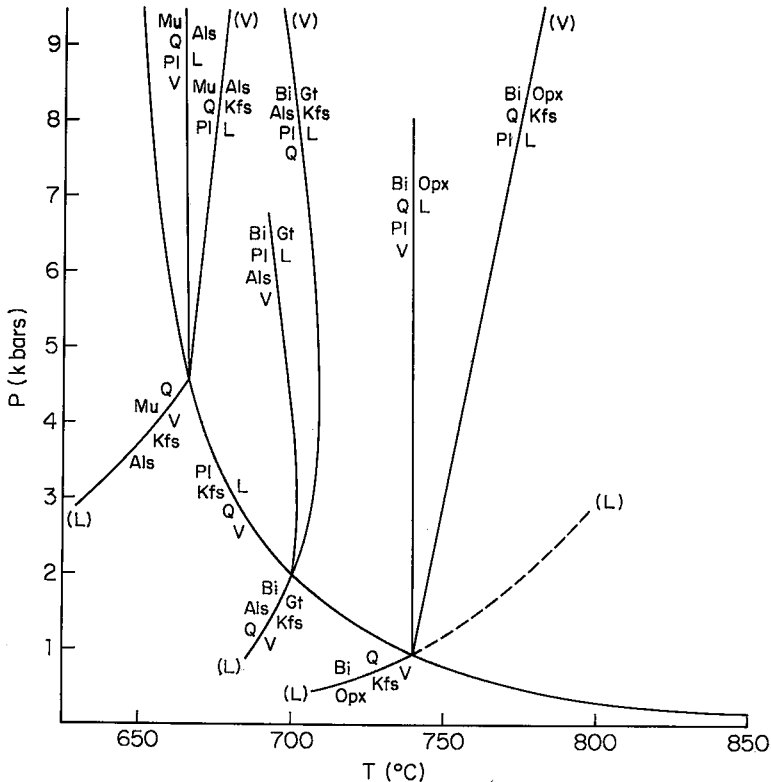


FIG. 8. P-T projection of selected equilibria involving muscovite, biotite and quartzofeldspathic melts \pm vapor. P-T loci are approximate, and portions of some curves are metastable. Fe/(Fe + Mg) ratio for biotite = 0.4. Abbreviations: Q quartz, Ab plagioclase (An_{20}), Or orthoclase or sanidine. Ms muscovite, Als Al_2SiO_5 , Sill sillimanite, Bi biotite, Gt garnet, Opx orthopyroxene, L silicate liquid, V aqueous vapor or fluid phase. For each set of reactions, the curves are labeled according to which phase is absent from the particular reaction.

is present, assemblages containing a crystalline hydrate may control $X(H_2O)^{fluid}$, with much of the melting of the hydrate occurring over a relatively narrow interval of temperature, around that of vapor-absent melting (Fig. 8).

Model partial melting reactions

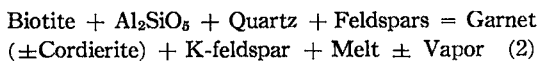
The major crystalline hydrates stable in peraluminous compositions under high-grade metamorphic conditions are muscovite, biotite and Ca-poor amphiboles. In examining the conditions of partial melting and the character of the products, we shall not consider reactions involving aluminous orthoamphiboles (*cf.*, Grant 1973). The stability relations of such amphiboles are poorly understood, and they rarely coexist with K-feldspar. In the absence of direct experimental determinations, the melting reactions depicted in Figure 8 were cal-

culated from subsolidus dehydration equilibria together with crystal-liquid-vapor equilibria in haplogranitic systems (Luth 1976). The latter were extrapolated to $a(H_2O) < 1$ using Burnham's (1979) models for melt activity-composition relations. Reactions involving biotite are drastically oversimplified owing to the difficulties of satisfying both mass-balance and mass-action requirements in systems with complex crystalline and melt solutions. However, our aim here is simply to illustrate the broad range of physicochemical conditions under which peraluminous (specifically S-type) melts and residual crystalline assemblages may develop. The reader is referred to the works of Grant (1973) and Abbott & Clarke (1979) for further examination of the chemography of melting relations in peraluminous systems.

Figure 8 illustrates that for pressures in the

range of 4 to 10 kbar, partial melting of muscovite-quartz-alkali-feldspar assemblages occurs at temperatures below about 700°C (Thompson & Algor 1977, Thompson & Tracy 1979). The relatively low-temperature granitic melts are extremely H₂O-rich (≥ 8 wt. % H₂O, even for vapor-absent melting) and should contain only small concentrations of components such as CaO, MgO and TiO₂. However, the high $X(\text{H}_2\text{O})^{\text{melt}}$ and saturation with Al₂SiO₅ should result in strongly peraluminous melts. Thompson & Tracy (1977) have noted normative corundum contents of 7 wt. % in quenched glassy products of partial melting of muscovite. Such high alumina contents require that the melt crystallize some combination of highly aluminous phases (muscovite, Al₂SiO₅ and garnet \pm cordierite). Solidification of these low-temperature, H₂O-rich melts would generally occur at $P \geq 3$ kbar (Burnham 1967, Harris *et al.* 1970) provided the melts migrated from their sources before appreciable increases in temperature over that of initial melting. Thus, muscovite-garnet leucogranites emplaced in regional metamorphic belts, particularly those in kyanite-sillimanite terrains, may be derived by partial melting of muscovite-bearing metasedimentary sources. Such an origin cannot, however, be applied to the high-level, S-type batholiths that are common in southeastern Australia and other terrains.

At temperatures above the muscovite-quartz stability field, partial melting in strongly peraluminous compositions will commonly involve biotite, Al₂SiO₅, garnet, cordierite, quartz and feldspars. We shall consider one set of the possible equilibria:

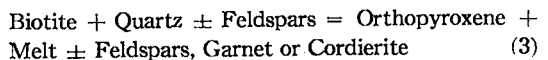


Such reactions characteristically take place in pelitic compositions under uppermost-amphibolite- or lower-granulite-facies conditions (Froese 1973, Bohlen & Essene 1979, Phillips & Wall 1979). Cordierite would be an important residual phase in typical pelitic compositions at low pressures, whereas garnet development would be favored by higher pressures and higher Fe/(Fe + Mg) ratios. In Figure 8, equilibria involving garnet were calculated from the experimental data of Richardson (1968), Weisbrod (1973), Holdaway & Lee (1977) and Ferry & Spear (1978), together with quartz-feldspar-melt equilibria as discussed above. Uncertainties in the positions of the phase boundaries are large, and the equilibria are metastable with respect to cordierite-bearing assemblages over much of

their P-T loci below 5 kbar, as well as being polyvariant.

Taking a temperature of 700–750°C and pressures in the 5- to 8-kbar range as a first approximation to vapor-absent melting conditions for our model pelite with biotite and Al₂SiO₅, we may infer H₂O contents of 6–8 wt. % for the derived granitic melts. The magmas thus derived are markedly water-undersaturated and may rise to high crustal levels. In view of the high K/(Na + Ca) ratios of typical pelites, K-feldspar would probably coexist with sillimanite, garnet and cordierite in the crystalline residues. The melt fractions would generally have high K/Na ratios, high normative orthoclase and quartz, low normative anorthite and relatively high normative corundum (owing to sillimanite saturation). The latter characteristics may result in crystallization of several percent garnet or cordierite (or both) from the melt (Fig. 7), even after separation from restite materials. Two-mica granites could be formed by cooling and subsequent crystallization of such magmas at moderate depths. Although the melts may depart appreciably from the lowest-temperature melting composition in granitic systems, microprobe analyses of glasses saturated with sillimanite, quartz, garnet and plagioclase quenched from 750°C indicate low contents of refractory components (Ti, Ca, Mg, *etc.*).

During high-grade metamorphism, biotite may persist to considerably higher temperatures in less aluminous compositions than those considered above. Biotite dehydration reactions in these less aluminous rocks will commonly result in the development of orthopyroxene (\pm garnet, cordierite) and a silicate liquid (Grant 1973):



It should be noted, however, that under granulite-facies P-T conditions, orthopyroxene-Al₂SiO₅ assemblages may be stable. The following discussion applies only to partial melting at pressures below this. In modeling biotite-orthopyroxene-melt equilibria (Fig. 8), we used the experimental data of Eugster & Wones (1962) and Wones *et al.* (1971) for Fe-biotite oxidation reactions together with the results of Bohlen *et al.* (1980a) and Hewitt (1978). Estimated entropy functions for Al-Si-disordered annite and Fe-Mg distribution coefficients for natural biotite-orthopyroxene pairs were also utilized. The calculated biotite-quartz-orthopyroxene dehydration equilibrium is as much as 150°C below an extrapolation of the curve of Wones &

Dodge (1977). (After the completion of this paper, the work of Hoffer & Grant (1980) became available to us. Their experimentally determined biotite–orthopyroxene–melt reactions lie approximately 30°C below our calculated curves.) Despite our approximations and considerable uncertainties in the P – T – $a(\text{H}_2\text{O})$ locations of reactions for complex biotites, several useful conclusions can be drawn from the phase relations in Figure 8.

As the stability of biotite–quartz is commonly limited by orthopyroxene–feldspar assemblages, these reactions produce the highest-temperature peraluminous partial melts for which biotite provides the H_2O . Thus we contend that substantial quantities of “primary” peraluminous magmas are unlikely to develop above biotite–orthopyroxene breakdown temperatures. The highest-temperature voluminous S-type magmas will thus have initial temperatures in the range 800–850°C, somewhat higher than those indicated on Figure 8; allowance should be made for stabilization of biotite by substitutions such as $\text{Ti}^{\text{VI}}\text{O}_2 = (\text{FeMg})^{\text{VI}}(\text{OH})_2$. Total depletion of the source rocks in quartz would also allow higher melting temperatures. Increasing F–OH substitution in biotite, concomitant with a decrease in biotite abundance, may markedly widen the stability field of this phase. The small amount of liquid produced by partial fusion of an assemblage containing F-enriched biotite may exhibit the characteristics of White’s (1979) R-type magmas. Where C–O–H fluids are present, anatexis of peraluminous rocks could produce magmas at temperatures above those for biotite stability. Peraluminous charnockites could develop by this mechanism.

For F-poor biotite–orthopyroxene partial melting [$a(\text{SiO}_2) = 1$] in the range 5–8 kbar, however, granitic liquids have H_2O contents of 3–5 wt. %. These magmas are thus strongly water-undersaturated and can rise to high crustal levels or be emplaced as volcanic suites. The weakly to moderately peraluminous melts may depart strongly from “near-minimum” compositions. Table 2 shows the major-element chemistry of some quenched experimental liquids coexisting with assemblages involving plagioclase, quartz, orthopyroxene and biotite at 5 kbar, 750–850°C. The relatively low SiO_2 content and the enrichment in more refractory components (*i.e.*, Ca, Ti, Fe, Mg) of these melts, compared with “near-minimum” compositions, are evident. These data give an idea of the chemistry of peraluminous melts derived from biotite–orthopyroxene reactions under granulite-facies conditions.

TABLE 2. NORMALIZED ELECTRON-MICROPROBE DATA FOR GLASSES PRODUCED IN 5-KBAR EXPERIMENTS ON THE RHYODACITE COMPOSITION

	750°C (wt. % $\text{H}_2\text{O}(\text{meit})=7.5$)	800°C (4%)	850°C (5%)
SiO_2	74.88	72.19	72.18
TiO_2	0.13	.17	.38
Al_2O_3	14.91	15.46	14.45
FeO	1.15	2.85	3.14
MnO	-	.05	-
MgO	0.45	.44	0.92
CaO	2.61	2.53	2.60
Na_2O	2.60	2.28	2.58
K_2O	3.27	4.03	3.77
Crystalline phases	Q, Bt, Ilm	Q, Pl, Bt, Opx, Ilm	Q, Bt, Opx, Ilm

Additional chemical evidence

Two further characteristics, taken together, may be diagnostic of magmas derived by partial melting of pelitic or psammopelitic source rocks. Their $\delta^{18}\text{O}$ should be high, reflecting the clay-rich character of the parent sediments (O’Neil *et al.* 1977). The rubidium-rich nature of the sediments could also result in magmas exhibiting a high initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (Chappell & White 1974). Marked *HREE* depletion in the melt could be associated with restitic garnet, but if cordierite is the major ferromagnesian restite mineral, little *HREE* fractionation will occur during partial melting. To account for the lack of *HREE* depletion in some suites of Australian S-type granitoid rocks, White (1979) has suggested that osumilite $(\text{K,Na,Ca})(\text{MgFe}^{2+})_2(\text{Al,Fe}^{3+},\text{Fe}^{2+})_3(\text{Si,Al})_{12}\text{O}_{30}\cdot\text{H}_2\text{O}$ may be a residual phase from partial melting of the pelitic source. We contend that White’s hypothesis is untenable since biotite, the inferred source of H_2O for the development of many S-type magmas, is probably largely eliminated by partial melting reactions in highly aluminous compositions at temperatures well below the osumilite stability field.

Typically, pelites and psammopelites contain > 30% biotite at metamorphic grades below the onset of partial melting by reactions similar to (2). From simple mass-balance calculations involving the “ H_2O ” content of biotite and the melts produced by reaction (2), we may infer that the degree of partial melting of the source rocks will commonly exceed 15–20% (by mass). These figures approach the values at which van der Molen & Patterson (1979) measured a drop in the strength of partially melted rocks and may even reach the critical melt fraction (30–35 vol. %) for suspension-like

behavior. In view of the decrease in density on melting, parts of the pelitic source-region could become gravitationally unstable and rise in diapir-like fashion. The high proportion of restite materials inferred in some apparently metasediment-derived granites (Chappell & White 1976) may result largely from such processes.

We submit that a number of southeastern Australian S-type granitoid suites are ultimately derived from broadly pelitic sources by partial melting reactions similar to reaction (2). Specific examples include the Deddick granodiorite (Porritt 1977, Nicholls & Wall, in prep.), the Bethanga gneiss (Kerber 1978) and the Cooma gneiss (Pidgeon & Compston 1965). Numerous alkali- and silica-depleted pelitic inclusions in the Deddick and Bethanga rocks contain garnet-sillimanite (\pm cordierite, K-feldspar, biotite) assemblages indicating equilibration around 5 kbar and 700–750°C, conditions presumably representative of the magma source-region. This supports the above contentions. Some S-type granitoid bodies from the Kosciusko batholith (Chappell & White 1976, Hine *et al.* 1978) probably belong to this "pelite-derived" group, as perhaps does the South Mountain batholith, Nova Scotia (McKenzie & Clarke 1975, Abbott & Clarke 1979).

Bulk compositions that could yield granitic melts by reactions such as (3) would include relatively immature quartzofeldspathic greywackes, psammopelites and altered intermediate to acid volcanoclastic rocks. Owing to the lack of strong Rb enrichment in these rocks, the magmas may have a moderate initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and their $\delta^{18}\text{O}$ may be closer to typical igneous values than magmas derived from originally clay-rich metasediments. The amount of biotite in mildly peraluminous quartzofeldspathic materials is typically less than 25%. Thus, where biotite is the H_2O source for the magmas, relatively low degrees of partial melting (10–20%) occur in the source region. Owing to the nature of the mechanisms involved in the extraction of such small proportions of partial melt from their crystalline host (Turcotte & Ahern 1978), the magmas would be unlikely to contain significant restite in the form of rock fragments or crystals. Residual assemblages would generally involve orthopyroxene-plagioclase-quartz \pm garnet, cordierite, K-feldspar, typical of rocks in some granulite-facies, supracrustal terrains. The general chemistry and inferred early P–T– $a(\text{H}_2\text{O})$ history of the central Victorian suites are compatible with their origin by biotite-orthopyroxene partial-melting

reactions in mildly peraluminous rocks under granulite-facies conditions.

Although the above considerations shed some light on the development of S-type magma suites, it should be noted that batch or progressive partial fusion may result in marked changes in the stoichiometry and chemography of melting reactions (Abbott & Clarke 1979). Furthermore, we have discussed only a few single-step reactions from the plethora of possibilities in peraluminous compositions. In view of the probable heterogeneity of S-type source regions, mixing of different melt fractions may occur and is likely to be further complicated by magma mixing during ascent and emplacement (Eichelberger 1974). Significant interaction between the magmas and their wall rocks may occur during ascent, and assimilation may be promoted by the heats of crystallization and cooling. A wide range of approaches and data is needed to evaluate these effects before inferences of source materials can be other than crude approximations.

CONCLUSIONS

Magmas with broadly S-type chemical characteristics may be derived by partial fusion of strongly to weakly peraluminous source rocks ranging, respectively, from pelites or psammopelites to relatively immature, quartzofeldspathic sediments or even altered igneous materials. Reactions involving biotite-bearing assemblages under vapor-absent or vapor-present conditions are inferred to be of major importance in partial fusion events producing S-type magmas. The variety of possible source rocks, coupled with the range of partial melting conditions (a function primarily of the geothermal gradient and behavior of volatile components) result in peraluminous melts spanning a broad spectrum of chemical compositions, thermophysical characteristics and transport properties. These source-related features strongly influence the ascent and emplacement history of the magmas.

S-type magmas, in the main, are lower in temperature and have higher water contents than many I-type magmas of the granodiorite-tonalite association, reflecting the lower stability ranges of biotite in quartz-bearing peraluminous rocks relative to that of hornblende in the intermediate to mafic source-materials of I-type magmas. At temperatures up to 800°C and for a wide range of melt water-contents, Ti-rich biotite is the main ferromagnesian phase that

crystallizes from moderately peraluminous magmas. Orthopyroxene development is characteristic of relatively high-temperature crystallization of such compositions. We may infer, from the chemistry of experimental liquids coexisting with a range of "restite" assemblages, that highly aluminous phases, notably garnet and cordierite, must crystallize from peraluminous magmas even on separation from their source materials. Garnet crystallization is favored by high pressures and high Fe/(Fe + Mg) ratios for the melt, whereas cordierite crystallizes mostly at low pressures (> 3 kbar). Fractionation of early-formed biotite and plagioclase can explain the chemical trends observed in many S-type suites. The relative contribution of fractional crystallization and "restite unmixing" (White & Chappell 1977) needs to be carefully assessed in view of the "non-minimum" compositions of many natural S-type granites and also of the experimental liquids coexisting with assemblages likely in S-type source-regions.

Although most S-type magmas are developed in intracontinental orogenic zones [Pitcher's (1979) "hercynotype" association], the relatively high-temperature, water-undersaturated character and high-level emplacement of some peraluminous magmas resemble Pitcher's "andino-type" associations. The high geothermal gradient required to produce these magmas at moderate crustal depths may result from underplating or intraplating of crust by mantle-derived material. The marked tectonic thickening of metasedimentary sequences in orogens related to continental collision (as inferred by Pitcher for his "hercynotype" case) need not be an essential feature of primary peraluminous magma genesis. Whereas further clarification of the origin and evolution of the voluminous I-type or andino-type associations is much needed, the S-type granitoid rocks can provide valuable insight into the tectonic, thermal and chemical evolution of Earth's continental crust.

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APPENDIX

Description and identification of experimental products

Quartz, where present, occurs as roundish, ~ 50 μm crystals that were readily identified optically.

Feldspar forms extremely fine-grained, feathery intergrowths at near-solidus conditions, but tabular crystals up to 20 μm long in near-liquidus experiments. Identification of the feldspars was principally by X-ray powder diffractometry. Plagioclase was confirmed by the presence of $\bar{2}01$, $\bar{1}30$, 111 , $\bar{2}02$ and 002 reflections. Some orthopyroxene peaks fall in positions similar to those of plagioclase, but the presence of a $\bar{1}30$ reflection was used to confirm the presence of plagioclase when orthopyroxene was known to be present. The presence of K-feldspar was chiefly recognized by the occurrence of $\bar{2}01$, $\bar{1}12$ and 040 reflections for this phase.

In near-solidus experiments, biotite forms extremely fine-grained aggregates with the other phases. Under conditions where more than 5% liquid is present, biotite forms pale brown, pleochroic and highly birefringent platelets up to 20 μm across. Where optical identification was difficult, the presence of a low-angle, basal X-ray reflection was diagnostic.

Orthopyroxene forms colorless to pale greenish untwinned needles and prisms 2-100 μm long with straight extinction and high relief.

Cordierite was not identified optically in near-solidus experiments but was detected by the presence of the 100 X-ray reflection. Near-liquidus cordierite forms 10 μm rectangular prisms with low relief and very low birefringence.

Euhedral overgrowths on garnet seeds were taken as evidence of garnet stability. Corrosion and disappearance of garnet seeds were interpreted as indicating instability of garnet.

TABLE OF EXPERIMENTAL RESULTS FOR GRANITE 889

Pressure (bar)	Temperature (°C)	fluid X_{H_2O}	Phases present
1039	748	.70	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
		.80	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
		.90	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
1045	804	1.00	Q, Pl, Bi, Gt, Cd, Ilm
		.40	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
		.60	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
1064	851	.80	Q, Pl, Bi, Opx, Cd, Ilm
		1.00	Q, Pl, Bi, Opx, Cd, Ilm
		.40	Q, Pl, Bi, Opx, Cd, Ilm
1027	883	.60	Pl, Opx, Cd, Ilm
		.80	Pl, Opx, Cd, Ilm
		1.00	Pl, Opx, Cd, Ilm
2037	752	.30	Q, Pl, Opx, Cd, Ilm
		.40	Pl, Opx, Cd, Ilm
		.50	Pl, Opx, Cd, Ilm
2075	775	.70	Pl, Opx, Cd, Ilm
		1.00	Opx, Cd, Ilm
		.49	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
2051	799	.70	Q, Pl, Bi, Gt, Cd, Ilm
		.90	Q, Pl, Bi, Opx, Gt, Cd, Ilm
		1.00	Q, Pl, Bi, Gt, Cd, Ilm
2045	838	.60	Q, Pl, Bi, Gt, Cd, Ilm
		.80	Q, Pl, Bi, Cd, Ilm
		.90	Pl, Bi, Cd, Ilm
2043	905	1.00	Bi, Ilm
		.50*	Q, Pl, Bi, Opx, Gt, Ilm
		.70*	Pl, Bi, Opx, Ilm
5063	704	1.00*	Bi, Ilm
		.20*	Q, Pl, Kfs, Opx, Ilm
		.50*	Pl, Opx, Ilm
5052	751	.70*	Opx, Ilm
		.10	Q, Pl, Opx, Cd, Ilm
		.20	Q, Pl, Opx, Cd, Ilm
5066	855	.30	Pl, Opx, Cd, Ilm
		.40	Opx, Cd, Ilm
		.70	Opx, Cd, Ilm
5068	902	1.00	Opx, Ilm
		.70	Q, Pl, Kfs, Bi, Gt, Cd, Ilm
		.90	Q, Bi, Gt, Ilm
5013	801	1.00	Bi, Gt, Ilm
		.60	Q, Pl, Bi, Gt, Ilm
		1.00	Bi, Ilm
5011	751	.40	Q, Pl, Bi, Gt, Ilm
		.70	Bi, Opx, Gt, Ilm
		1.00	Bi, Ilm
5013	801	.40	Q, Pl, Bi, Gt, Ilm
		.70	Bi, Opx, Gt, Ilm
		1.00	Bi, Ilm
5066	855	.30	Q, Pl, Bi, Opx, Gt, Ilm
		.50	Bi, Opx, Gt, Ilm
		.70	Bi, Ilm
5068	902	1.00	Bi, Ilm
		.25	Q, Opx, Gt, Ilm
		.35	Q, Opx, Gt, Ilm
5068	902	.45	Opx, Ilm
		1.00	Ilm

N.B. All experimental products contained glass in addition to the crystalline phases noted.

* Starting glasses for these experiments were not seeded with cordierite.

Near the solidus, ilmenite forms a fine, opaque dust throughout the charge. Where substantial amounts of liquid were present, ilmenite grew as purple, hexagonal platelets up to 10 μm across.

Construction of the $T-X(H_2O)$ isobaric diagrams

The combination of optical and X-ray methods generally allowed detection of 3–5 vol. % of phases, as confirmed by the addition of finely ground material to the run products. In the cases where more than 60% glass was present, optical examination of grain mounts and polished thin sections permitted detection of < 1 vol. % of phases. The interpolation of reaction boundaries between experimental points was based on estimates of the amount of the relevant phase present at surrounding experimental points.