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THE MINERALOGY OF PERALUMINOUS GRANITES: A REVIEW

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ABSTRACT

Peraluminous granitoid rocks are defined chemically as those in which the ratio (molecular proportions) $Al_2O_3/(CaO + Na_2O + K_2O)$ ($\equiv$ A/CNK) > 1. Mineralogically these granites consist of quartz + two feldspars + one or more characteristic phases such as biotite, muscovite, garnet, cordierite, the aluminosilicates Al_2SiO_5, mullite, topaz, tourmaline, spinel and corundum. The factors that control the appearance of these phases are temperature, pressure, P_{fluid}/P_{total}, and compositional variables such as $f(O_2)$, $f(CO_2)$, $f(HCl)$, $f(HF)$, $\Sigma(MgO + FeO)$, MgO/FeO and minor elements (B, Be, Mn, Li, Cs, *etc.*). Various origins for each of the characteristic phases have been proposed for different batholiths. A genetic classification of these phases is proposed as follows: 1. exogenic: (a) restite, (b) xenocrysts from high-level country rock; 2. endogenic: (a) nucleation from silicate melt with A/CNK > 1, (b) nucleation from water-saturated, alkali-depleted silicate melt with A/CNK ≫ 1, (c) metasomatic replacement of silicate rock by vapor phase, (d) nucleation directly from aqueous fluid. Rocks formed by processes in categories 1(a), 1(b) and 2(a) are termed "normal peraluminous" because they involve only melt-phase compositions that can be reached by silicate melt – crystal equilibria. Those in the remaining categories, 2(b), 2(c) and 2(d), are termed "hyperaluminous" because they represent melt or whole-rock compositions with A/CNK ≫ 1 that result from the interaction of fluids with silicate melts or solid rocks.

Keywords: peraluminous granitoid rocks, biotite, muscovite, garnet, cordierite, andalusite, sillimanite, tourmaline, topaz, mullite, spinel, corundum, mineralogy, mineral chemistry.

SOMMAIRE

Un granite est considéré comme peralumineux quand sa composition montre un rapport molaire $Al_2O_3/(CaO + Na_2O + K_2O)$ ($\equiv$ A/CNK) supérieur à l'unité. De tels granites contiennent quartz + deux feldspaths + au moins un des minéraux caractéristiques suivants: biotite, muscovite, grenat, cordiérite, aluminosilicates (Al_2SiO_5), mullite, topaze, tourmaline, spinelle, corindon. La cristallisation de ces minéraux dépend des variables suivantes: température, pression, rapport P_{fluide}/P_{totale}, $f(O_2)$, $f(CO_2)$, $f(HCl)$, $f(HF)$, $\Sigma(MgO + FeO)$, MgO/FeO, et concentration d'éléments accessoires (B, Be, Mn, Li, Cs, *etc.*). Plus d'une origine a été proposée pour chacune des phases caractéristiques dans divers batholithes. Nous proposons ici une classification génétique de ces phases: (1) Exogène, dérivée soit (a) d'une restite ou (b) des roches encaissante à faible profondeur (xénocristaux); (2) Endogène, par (a) nucléation dans un magma silicaté à A/CNK > 1, (b) nucléation dans un magma saturé en eau, appauvri en alcalins, à A/CNK ≫ 1, (c) remplacement métasomatique dû à une interaction de la roche silicatée avec une phase gazeuse, ou (d) nucléation à partir de la phase aqueuse. On considère comme hyperalumineuses normales les roches des catégories 1(a), 1(b) et 2(a) parce qu'elles impliquent des compositions magmatiques qui résultent d'équilibre entre magma et phases cristallines. Les roches des autres catégories [2(b), 2(c), 2(d)] sont considérées comme hyperalumineuses anormales parce qu'elles représentent des magmas ou des lithologies à A/CNK ≫ 1, qui proviennent de l'interaction d'un magma ou d'une roche avec une phase fluide.

(Traduit par la Rédaction)

Mots-clés: roche granitoïde hyperalumineuse, biotite, muscovite, grenat, cordiérite, andalousite, sillimanite, tourmaline, topaze, mullite, spinelle, corindon, minéralogie, chimie minérale.

INTRODUCTION

Granitoid rocks are normally classified on the basis of their relative modal proportions of quartz, alkali feldspar and plagioclase (Streckeisen 1976). Chemical classification systems are also in use; Shand (1927) advocated one based on the principle of alumina saturation. The ratio A/CNK ($\equiv$ moles Al_2O_3)/[moles($CaO + Na_2O + K_2O$)] is unity in feldspar, and represents an alumina-saturated condition for the bulk composition of a rock containing only quartz and two feldspars. Shand proposed four subdivisions: peralkaline (A < NK), metaluminous (CNK > A > NK), subaluminous (A = CNK) and peraluminous (A > CNK). In response to these changes in bulk chemical compositions, granitoid rocks will contain distinctive suites of minor minerals. This paper deals with those unique mineralogical expressions of peraluminous granitoid rocks, in which the excess Al raises the status of some accessory minerals to that of essential minerals (Table 1).

Peraluminous granites with A/CNK > 1.1 have also been classified genetically as S-type granites (Chappell & White 1974). However, since such peraluminous granites may represent

TABLE 1. CHARACTERISTIC MINERALS OF PERALUMINOUS GRANITES

Mineral		Formula	A/CNK	A*
Biotite	Annite	$K_2Fe_6Al_2Si_6O_{20}(OH)_4$	1.0	0.0
	Siderophyllite	$K_2(Fe_5Al)(Al_3Si_5O_{20})(OH)_4$	2.0	0.17
Muscovite		$K_2Al_4(Si_6Al_2O_{20})(OH)_4$	1.5	1.0
Phengite		$K_2(Fe, Mg)Al_3(AlSi_7O_{20})(OH, F)_4$	4.0	0.6
Aluminosilicates		Al_2SiO_5	∞	1.0
Cordierite		$(Mg, Fe)_2Al_4Si_5O_{18}$	∞	0.5
Garnet		$(Ca, Mg, Mn, Fe)_3Al_2Si_3O_{12}$	∞	0.25
Staurolite (?)		$2FeO \cdot 4.5Al_2O_3 \cdot 7.5SiO_2$	∞	0.69
Mullite		$3Al_2O_3 \cdot 2SiO_2$	∞	1.0
Topaz		$Al_2SiO_4(OH, F)$	∞	1.0
Tourmaline		$Na(Mg, Fe)_3Al_6(OH)_4(BO_3)_3Si_6O_{18}$	6.0	0.5
Spinel		$(Fe, Zn)Al_2O_4$	∞	0.5
Corundum		Al_2O_3	∞	1.0

* $A = Al_2O_3 - K_2O/(Al_2O_3 - K_2O + MgO + FeO)$

a point of convergence of evolutionary processes as diverse as fractional crystallization of metaluminous magma (Cawthorn *et al.* 1976), removal of alkalis as complexes soluble in a volatile phase from Al-saturated magmas, anatexis of pelitic source-material (White & Chappell 1977) and contamination by pelitic country rocks, terms with genetic implications should be rejected in favor of the noncommital term "peraluminous".

The unique suite of minerals that is produced in a given granitoid rock is a function of the bulk composition of the magma, the degree of Al oversaturation, T, P, $a(H_2O)$, $f(O_2)$, $f(HF)$, $MgO + FeO$, MgO/FeO and concentrations of certain trace elements such as B, Be, Cs, F, *etc.* Since the appearance of peraluminous minerals may be sensitive to all these factors, correct interpretation of their textures, parageneses, reaction relationships, chemistry, zonation, distribution coefficients, geothermometry, geobarometry and phase equilibria can be used to help define the magmatic and subsolidus crystallization histories of granitoid rocks. The problems are very complex, and undoubtedly more information remains to be extracted from this characteristic suite of minerals. This paper offers a review of the occurrence of those mineral phases that characterize peraluminous granites and a genetic framework into which they can be placed.

ABBREVIATIONS

The following set of abbreviations is used throughout this paper:

Als	aluminosilicate
And	andalusite
Bio	biotite
Cdt	cordierite
Cel	celadonite
Cor	corundum
Crs	cristobalite
Est	eastonite
Gnt	garnet
Ksp	K-feldspar
Kyn	kyanite
Lct	leucite
Liq	silicate liquid
Mul	mullite
Mus	muscovite
Phl	phlogopite
Phn	phengite
Pla	plagioclase
Qtz	quartz
Sid	siderophyllite
Sil	sillimanite
Spn	spinel
Stl	staurolite
Top	topaz
Tor	tourmaline
Trd	tridymite
Vap	water vapor

CHARACTERISTIC MINERALS OF PERALUMINOUS GRANITOID ROCKS

The list of common characteristic minerals of peraluminous granitoid rocks (Table 1) shows their stoichiometric formulas, their A/CNK values (largely reflecting variation in A/CNK in the host rock), and their *A* parameter [= $(Al_2O_3 - K_2O)/(Al_2O_3 - K_2O + MgO + FeO)$] of the AFM projection (Barker 1961). Each of these minerals will be discussed below in terms of their mode of occurrence, chemistry and presumed origin.

TABLE 2. POSSIBLE ORIGINS FOR CHARACTERISTIC MINERALS OF PERALUMINOUS GRANITES

1. SOLID-MELT REACTIONS
   (a) Primary crystallization from a silicate melt
   (b) Normal peritectic relationships during differentiation
   (c) Reactions with xenoliths
2. SOLID-SOLID REACTIONS
   (a) In thermal aureoles → xenoliths → xenocrysts
   (b) In subsolidus granites
3. REACTIONS OF AQUEOUS FLUID WITH MELTS OR SOLIDS
   (a) Modification of bulk composition of silicate melt (followed by 1(a))
   (b) Direct precipitation from fluid
   (c) Metasomatic replacement

CRITERIA FOR DETERMINING THE ORIGIN OF THE CHARACTERISTIC MINERALS

The characteristic minerals can originate in a wide variety of ways that involve reactions between solids, melts and fluids (Table 2). The techniques by which the origin of a given phase can be determined are reasonably well known, and the critical types of observations are listed below for convenience. In general, some combination of textural, chemical and phase-equilibrium determinations is the basis on which such decisions regarding origin are made.

Textures: What is the size of the mineral relative to other phases in the host rock ? Usually in equigranular rocks the characteristic phase has originated by the same mechanism as the other

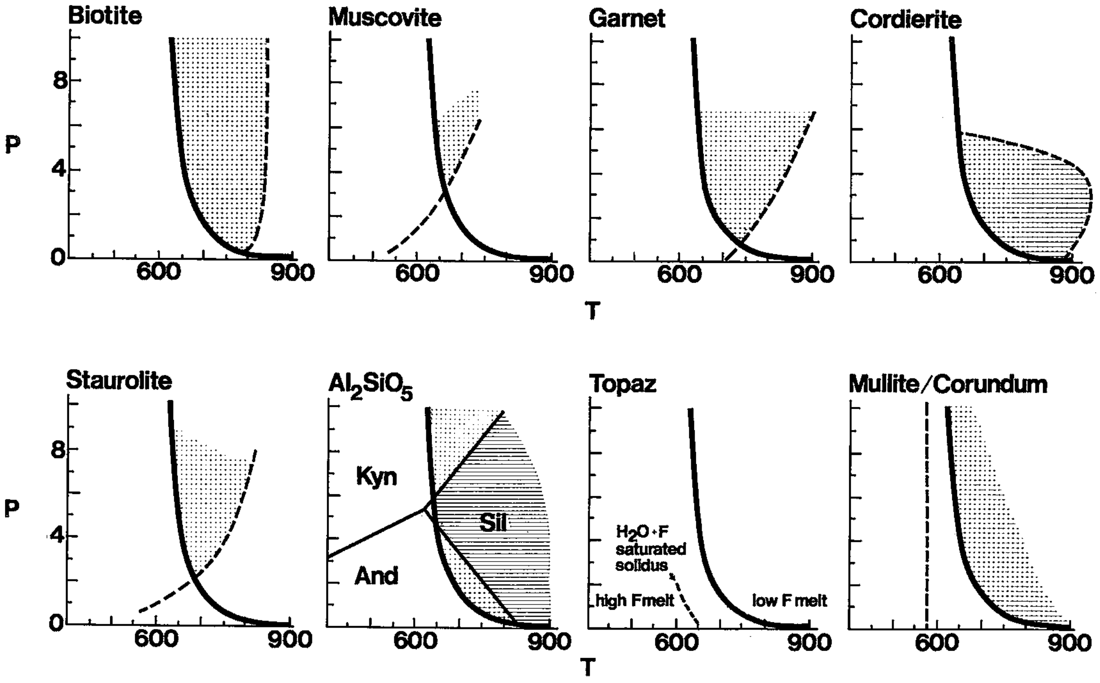

FIG. 1. Simplified P–T stability fields for some of the phases found in peraluminous granites and their relationship to the field of silicate melts. Overlap of the two fields (shaded) indicates that a primary magmatic origin for that phase is possible; however, the positions of all stability curves, including that of the granite solidus, are sensitive to many chemical parameters. The result may be to expand, contract or even eliminate the areas of overlap in the natural system. Sources of stability curves: biotite, Clemens & Wall 1981; muscovite, Althaus *et al.* 1970; cordierite, Clemens & Wall 1981; staurolite, Richardson 1968; aluminosilicates, Richardson *et al.* 1969; topaz, Glyuk & Anfilogov 1973; mullite and corundum, Roy & Osborn 1954.

minerals. What is the grain shape? Euhedral grains have almost certainly been in equilibrium with the silicate melt. What is the relationship to other grains? Poikilitic textures, reaction relationships and replacement textures should be considered directly relevant.

Chemistry: What is the composition of the mineral? Does it have a composition compatible with attainment of equilibrium with the other phases? Is the mineral zoned? Is the zoning normal or reversed (*e.g.*, in the case of garnets)? Is the zoning continuous or discontinuous? Discontinuous zoning may indicate two very different mechanisms or environments of growth.

Phase equilibria: How does the experimentally determined stability field for the mineral compare with that of a water-saturated silicate melt? The overlap of mineral and silicate-melt stability fields (Fig. 1) suggests the possibility of, but does not demand, a magmatic origin for that mineral. Only when overlap can be shown in P–T–X space (where X represents all the compositional variables listed in the Introduction) can the origin be clearly determined in terms of phase equilibria. Are K_D values with other phases compatible with attainment of equilibrium? Do mineral-chemistry geothermometers or geobarometers indicate reasonable conditions for emplacement? Do they agree with P–T conditions inferred for the country rocks? Are there any fluid inclusions? If so, what are the filling temperature and the composition of the fluid?

Not all of these criteria can be determined for any given phase, but the more data that can be collected, the greater will be the confidence in the deduced origin.

Biotite

Biotite is perhaps the most common mineralogical sink for excess alumina in granitoid rocks regardless of the A/CNK value in the host rock. It occurs as xenocrysts, early phenocrysts or microphenocrysts and as late groundmass varieties. Only in some leucogranitic and pegmatitic associations does it become rare.

The compositions are highly variable, but in all cases they are more aluminous than members of the phlogopite–annite join (Fig. 2). Studies by de Albuquerque (1973). Eugster & Wones (1962). Guidotti *et al.* (1975) and Clarke *et al.*

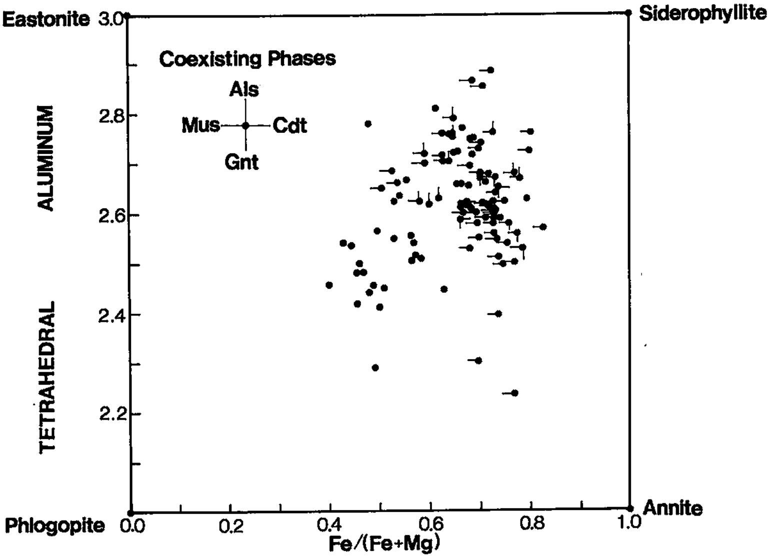

FIG. 2. Biotite compositions from peraluminous granitoid rocks. In more basic rocks such as tonalites and granodiorites, the biotites have low Fe/(Fe + Mg) ratios and no coexisting AFM phases. More highly differentiated rocks contain biotites with higher Fe/(Fe + Mg) ratios attributable to crystal–liquid fractionation processes in the magma and higher eastonite–siderophyllite components attributable to coexistence with other AFM phases. Sources of data: de Albuquerque 1973, Allan 1980, Best *et al.* 1974, Farley 1979, Guidotti *et al.* 1975, Harris 1974, Joyce 1973, Leake 1967, MacDonald 1979, Miller *et al.* 1981, Neiva 1974, Phillips *et al.* 1981, Saavedra 1978, Stallard 1975.

(1976) have shown conclusively that biotite compositions are highly sensitive to $f(O_2)$ conditions prevailing during crystallization and to mineral assemblage. In general, aluminum in both tetrahedral and octahedral positions increases as the coexisting minerals themselves have higher values of A/CNK.

Biotite is most commonly of magmatic origin, although some may occur as mafic remnants from the source region (White & Chappell 1977, Lorenzoni *et al.* 1979) or as refractory mafic schlieren resulting from the assimilation of country rocks.

Muscovite

The unfortunate term "two-mica", which is used almost synonymously with "peraluminous", alludes to the very common appearance of muscovite in alumina-oversaturated granitoid rocks. Muscovite can occur as genuine phenocrysts (Best *et al.* 1974), as euhedral books in the groundmass of granites and aplites, as large books in pegmatites, as texturally obvious breakdown products of feldspars, biotite, aluminosilicates and corundum in rocks affected by fluids and as an important constituent of greisens. (If feldspar alone is breaking down, the appearance of white mica does not necessarily mean A/CNK > 1 in the original host rock; therefore the terms "two-mica" and "peraluminous" are not synonymous.)

Pure end-member muscovite $K_2Al_4(Si_6Al_2O_{20})(OH)_4$ does not occur in any of the granite associations; instead, there is solid solution towards paragonite $Na_2Al_4(Si_6Al_2O_{20})(OH)_4$ and celadonite $K_2Al_2(Fe,Mg)_2(Si_8O_{20})(OH)_4$. Intermediate members of the muscovite–celadonite series are termed phengites. Phengite compositions (Fig. 3) are sensitive to both temperature and mineral association (Guidotti 1978).

As indicated in Figure 1, "muscovite" can have a magmatic origin at relatively high pressures (but see Miller *et al.* 1981), or can originate at subsolidus temperatures at lower pressures through reactions such as And + Ksp + Vap → Mus + Qtz. Muscovite is thus one of the most problematic minerals of peraluminous

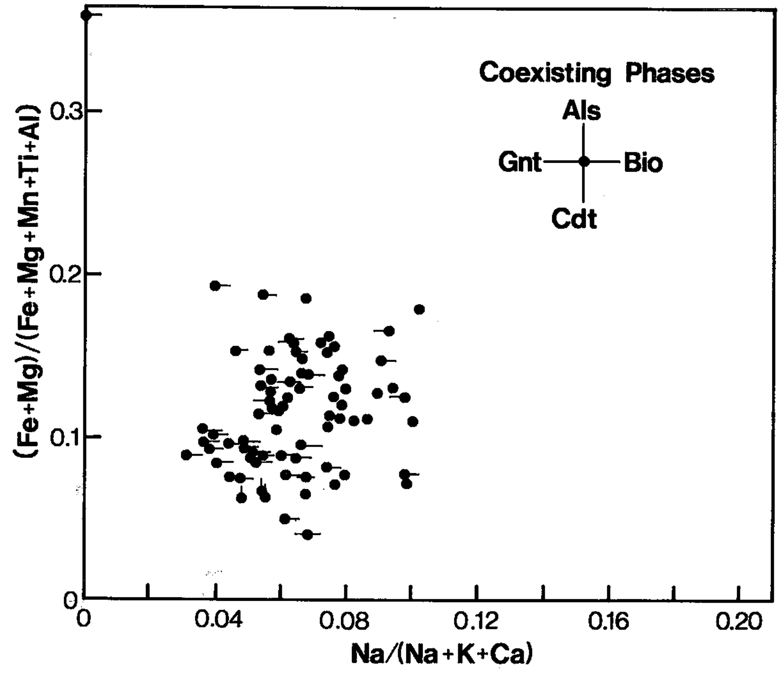

FIG. 3. Muscovite compositions from peraluminous granitoid rocks. Most analyses have (Fe + Mg)/(Fe + Mg + Mn + Ti + Al^{vi}) < 0.2 and Na/(Na + K + Ca) < 0.1, but no clear pattern of compositional control by paragenesis can be determined from these data. Sources of data: Best *et al.* 1974, Farley 1979, Guidotti 1978, Harris 1974, Joyce 1973, Miller *et al.* 1981, Neiva 1974, 1975a, b, Saavedra 1978.

granitoid rocks, since there are few clear textural or chemical means of distinguishing these two modes of origin (Saavedra 1978). Clarke *et al.* (1976) even suggested that most muscovite in peraluminous granitoids could be attributed to the subsolidus breakdown of aluminosilicates, since the above reaction would be virtually unavoidable provided aluminosilicates were more common constituents of peraluminous granitoids than are currently believed.

Garnet

Of all the characteristic minerals in peraluminous granites, garnet has received the most attention and yet remains one of the least understood phases. The confusion arises from the variety of textural relations displayed, the complex patterns of chemical zonation, and the rather misleading and often inappropriate experimental work. For example, Allan & Clarke (1981) have

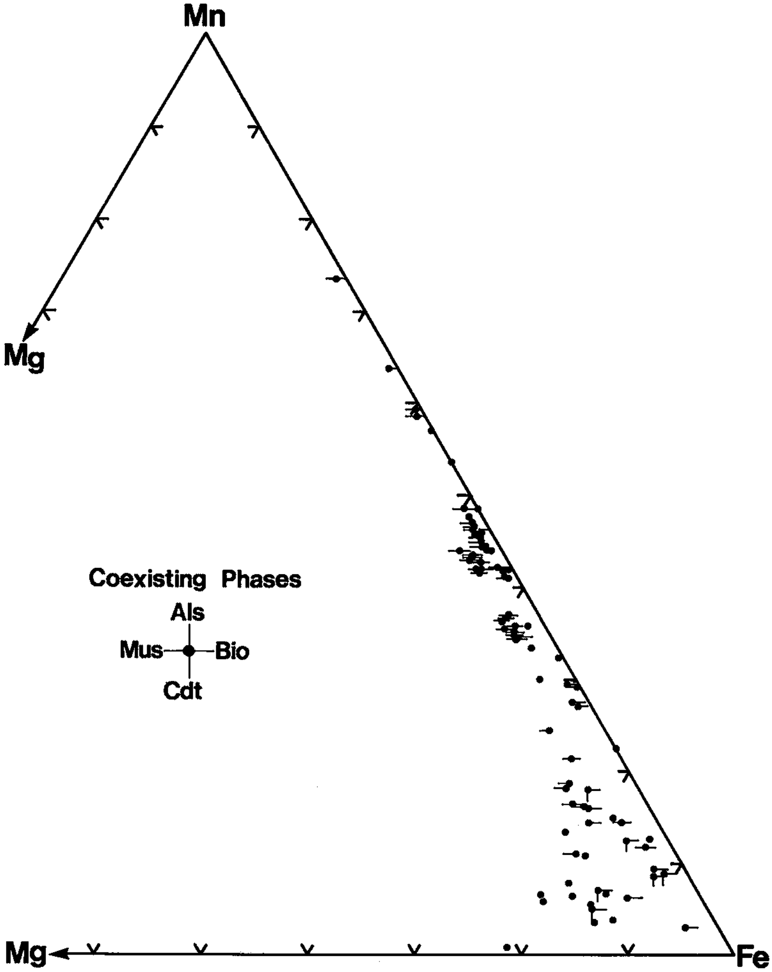

FIG. 4. Garnet compositions from peraluminous granitoid rocks. Most garnets are essentially almandine–spessartine solid solutions, those coexisting with cordierite being rich in Fe and those coexisting with one or two micas having highly variable Fe/Mn ratios. Sources of data: Allan 1980, Brammall & Harwood 1923, Brammall & Bracewell 1936, Hall 1965, Harris 1974, Joyce 1973, Leake 1967, López Ruiz & García Cacho 1975, MacDonald 1979, Miller & Stoddard 1978, Neiva 1975a, Phillips *et al.* 1981, Rub *et al.* 1977.

described, from the granodiorites of the South Mountain batholith, garnets with reaction rims of biotite, garnets overgrowing biotite, euhedral garnets free of any reaction relations, low-Mn garnets, high-Mn garnets, garnets with normal zoning (Ca–Mn cores, Fe–Mg rims) and garnets with reverse zoning. Similar observations are reported from many other localities; clearly, no single, simple interpretation is possible for such complex assemblages.

Garnets from peraluminous granites can have highly variable compositions, with most of the variation occurring in the ratio of Mn:Fe:Mg (Fig. 4). As explained by Leake (1967), zoning in garnet is an intrinsic feature of the growth mechanism and is controlled by the rate at which the necessary constituents can diffuse to, and within, the growing crystal. Normal zoning is generally attributed to growth in a metamorphic environment during prograde conditions. Reverse zoning may indicate metamorphic growth during retrograde conditions (*i.e.*, the garnet can continue to grow only if it increases in Mn content) or, in the case of igneous garnets, during an increase of $Mn/(Mn + Fe^{2+} + Mg)$ during falling temperature and fractional crystallization. It is also important to note that the ratio $Mn/(Mn + Fe^{2+} + Mg)$ may increase rapidly in aplite–pegmatite systems, where formation and dissociation of an aqueous fluid phase can cause the reaction $2FeO + \frac{1}{2}O_2 \rightarrow Fe_2O_3$. This may account for the prevalence of garnet-bearing aplite–pegmatite dykes in otherwise garnet-free granitic plutons.

The mechanisms that seem to be important in the appearance of garnet in granitoid rocks are (1) derivation as a refractory restite phase from the zone of partial melting (White & Chappell 1977); (2) derivation as refractory xenocrysts from porphyroblasts in high-level pelitic country rocks (Allan & Clarke 1981); (3) generation in the marginal facies of a granite body as a result of reaction between magma and pelitic xenoliths rich in Al and Mn relative to the melt (Jamieson 1974, Allan & Clarke 1981); (4) nucleation directly from the silicate melt, especially in aplites and pegmatites, as late-stage differentiation increases the $Mn/(Mn + Fe^{2+} + Mg)$ ratio sufficiently to stabilize spessartine-rich garnet (Hall 1965, Joyce 1973, Green 1976, 1977, Foord 1977, Allan 1980); (5) reaction between early-formed phases and silicate melt, for example, Liq + Bio → Gnt + Mus (Miller & Stoddard 1978) or Liq + Als + Bio → Gnt (Abbott & Clarke 1979).

Only a very thorough textural and chemical

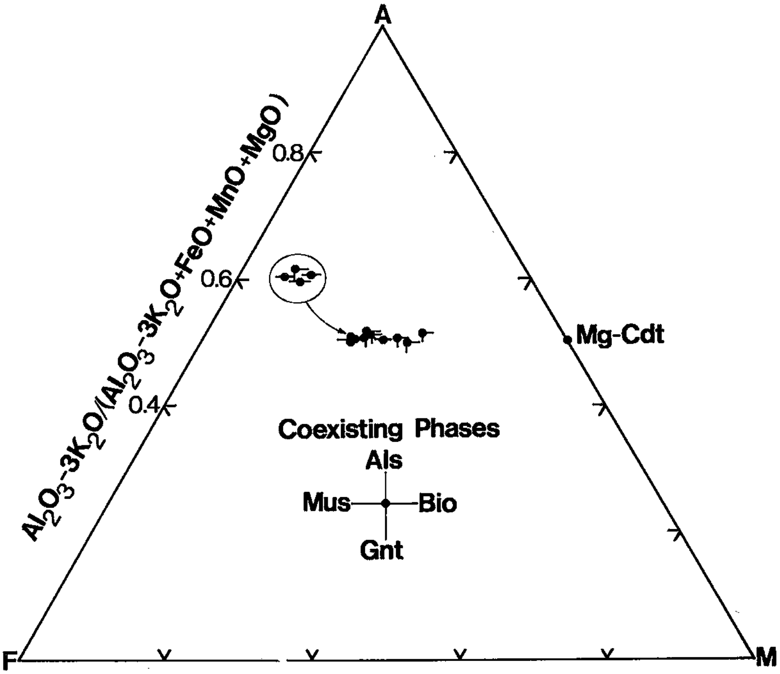

FIG. 5. AFM projection of cordierite compositions from peraluminous granitoid rocks. Sources of data: Flood & Shaw 1975, Phillips *et al.* 1981.

investigation will permit clear discrimination among these competing models.

Cordierite

Cordierite has been described from coarse-grained granitoid rocks (Brammall & Harwood 1923, Jones & MacMichael 1975, Flood & Shaw 1975), fine-grained aplites (Phillips *et al.* 1981) and coarse-grained pegmatites (Heinrich 1955). It is frequently found associated with biotite, garnet and aluminosilicates in these various granitoid rocks and may be related to them through a variety of reaction relationships (Abbott & Clarke 1979).

Cordierites are usually unzoned or weakly zoned, and have compositions ranging from Mg/(Mg + Fe) = 0.60 to 0.39 (Fig. 5). As with the garnets described above, there may be more than one generation of cordierite in a given rock, and it should be obvious from distribution coefficients with other AFM phases which cordierites are in mutual equilibrium and which represent foreign grains.

Experimental work (Fig. 1) shows conclusively that cordierite is expected to be a stable phase at the conditions prevailing during granite magmatism (Green 1976, Lee & Holdaway 1977, Clemens & Wall 1981). Because cordierite is an extremely common phase in thermal aureoles in pelitic rocks, it epitomizes the classic problem of magmatic *versus* thermal metamorphic origin. To determine the origin of the cordierite in any particular cordierite-bearing pluton requires using the complete arsenal of textural, chemical and phase-equilibrium weapons. Flood & Shaw (1975) have considered the full range of possibilities for cordierites in the New England batholith, Australia, and have opted for a restite origin, *i.e.*, derivation from the zone of partial melting where the granite melt was produced. The problem of complete disaggregation of the cordierites from the refractory residuum to form individual crystals was not considered, but Morin & Turnock (1975) have described an intermediate case from Manitoba where the cordierite is found in clots with other refractory phases such as sillimanite and biotite. Phillips *et al.* (1981) favored a magmatic origin for the cordierite in the Strathbogie batholith, which is in the same plutonic zone as the New England batholith. Clearly, the problem is not a simple one.

The occurrence of cordierite in aplites remote from the contact metamorphic aureole is fairly convincing evidence in favor of a magmatic origin, although M.A. MacDonald (pers. comm. 1980) has found large poikilitic cordierites with inclusions only of quartz in a fine-grained, sugary-textured aplite in the Musquodoboit pluton. The occurrence of very large (up to 1 m in length) cordierite crystals in pegmatite (Heinrich 1955) suggests an important role for the supercritical aqueous fluid, but perhaps more as an agent in modifying the silicate-melt composition, reducing viscosity and promoting diffusion, than as a medium of precipitation.

Aluminosilicates

Before phase relations in the system Al_2SiO_5 were known, and before aluminosilicate-producing reactions involving garnet, cordierite and alkali feldspar were studied, it was largely believed that andalusite, sillimanite and kyanite in granitic rocks were derived as xenocrysts from pelitic hornfelses of contact aureoles. Undoubtedly this kind of interpretation is still appropriate in some cases (*e.g.*, Lorenzoni *et al.* 1979), but careful examination of textures and application of phase relationships has led to a more general acceptance of magmatic origins for these phases in granitoid plutons. Heinrich (1955) has cited a number of examples of aluminosilicates occurring with cordierite in pegmatites; in these cases, the presence of an aqueous fluid phase may be a significant factor in the appearance of the aluminosilicates.

Andalusite: The case for direct crystallization of andalusite from the magma of the South Mountain batholith (Clarke *et al.* 1976) rests on its textural features (euhedral and same grain size as other phases in an aplitic host) and remoteness from aureole rocks (the aplite dykes occur in the interior of the batholith, away from pelitic aureole rocks). In the nearby Musquodoboit batholith, Abbott & Clarke (1979) have recognized a second mode of formation from the reaction Liq + Cdt → And + Bio. Phillips *et al.* (1981) have also discussed the possible origin of aluminosilicates by reactions of the type Gnt/Cdt + Ksp + H_2O → Bio + Als + Qtz.

Sillimanite, kyanite: Arguments developed for andalusite apply equally well to sillimanite and kyanite, given, of course, that each polymorph has its own unique P–T significance. Kyanite is rare in granitoid rocks (Ball 1923, Heinrich 1955), but examples of sillimanite are numerous. Sillimanite occurring as clusters of fibrolite in association with mafic schlieren probably has a metamorphic origin (Harris 1974, Guidotti

1978, Lorenzoni *et al.* 1979). Many other examples of sillimanite have been interpreted as a primary magmatic product even where sillimanite-bearing xenoliths are known to occur (de Albuquerque 1971).

The presence of Al_2SiO_5 polymorphs in granitoid rocks is a valuable aid in determining the ascent and cooling (P and T) history of the magma; their occurrence deserves greater attention.

Staurolite

As can be seen from Figure 1, the considerable field of P–T overlap between the stability field of staurolite and that of granitic magmas suggests that staurolite should be possible as a magmatic phase in peraluminous granitic melts. However, no examples of its occurrence have been described in the literature. This phenomenon illustrates very well that overlap in simple P–T space is not a reliable indication of the behavior of a given phase in the much more complex P–T–X hyperspace. For the compositions of peraluminous granites involved, biotite and garnet invariably are favored over staurolite.

Tourmaline

Tourmaline is a common minor phase of peraluminous granites, pegmatites and greisen zones. Its composition is generally close to the schorl end-member $Na(Fe,Mn)_3Al_6B_3Si_6O_{27}(OH,F)_4$ although elbaitic types $Na(Li,Al)_3Al_6B_3Si_6O_{27}(OH,F)_4$ are also found in greisen zones. Neiva (1974) has reported a large number of chemical analyses of tourmalines from northern Portugal and has related their change in composition to magmatic fractionation processes. Chorlton & Martin (1978) have produced experimental evidence to show that the presence of boron significantly lowers the temperature of the water-saturated granite solidus. They also showed that the partitioning of boron between silicate melt and vapor phase was a function of the co-ordination number of boron (B^{IV} and B^{III}, respectively) and that the co-ordination number was in turn a function of the T–P–X environment.

Although most tourmalines in peraluminous granites give a strong indication of having a subsolidus metasomatic replacement origin of the type $5\ Ksp + 1\ Alb + 3\ B^{3+} + 3\ Fe^{2+} + 7\ H_2O \rightarrow 1\ Tor + 12\ SiO_2 + 5\ K^+ + 3\ H^+$ (Němec 1975, Gentle 1977, Lister 1978), the work of Chorlton & Martin (1978) demonstrates the possibility of magmatic generation of tourmaline to explain the occurrence of isolated tourmaline crystals (Brammall & Harwood 1925, Foord 1977, Phillips *et al.* 1981) in some granites, apparently without reaction relationships and remote from the effects of hydrothermal activity. Tourmaline straddles the boundary between magmatic and nonmagmatic origins, but appears to be more prevalent under the latter conditions.

Topaz

Topaz is often found in late-stage, leucocratic peraluminous granitoid compositions. It is usually associated with high concentrations of Li, Cs and Be, in addition to F, and with such minerals as lepidolite, fluorapatite, amblygonite, spodumene, pollucite, *etc.* Topaz is generally accepted to be the product of reaction between high-temperature hydrothermal fluids and early-formed silicates, *e.g.*, $KAlSi_3O_8 + 4NaAlSi_3O_8 + 4HF \rightleftarrows Al_2SiO_4F_2 + KAl_3Si_3O_{10}F_2 + 9SiO_2 + 2Na_2SiO_3 + 3H_2O$ (Kovalenko 1973) or $Mus + Qtz + 2HF \rightarrow Top + Ksp + 2H_2O$.

Recently, however, this rather restricted concept has been expanded to include the full range of conditions, from magmatic [at temperatures from 1000°C down to a eutectic (Qtz + Ksp + Alb + Top + Liq + Mus) at 550°C] to pegmatitic, to high-temperature hydrothermal (greisenization) and even to deposition from hydrothermal fluids at temperatures less than 300°C (Naumov *et al.* 1977, Saavedra 1978). The strongest case yet made for a magmatic origin is that for the topazites (quartz + topaz rocks) of the New England batholith. Eadington & Nashar (1978) have examined both solid and fluid inclusions in topaz crystals and have deduced a crystallization range of 850 to 570°C. The fluid inclusions are brines with 57 wt. % NaCl equivalent, perhaps revealing the mechanism by which the silicate melt was able to crystallize topaz; *i.e.*, CaO, Na_2O and K_2O were stripped from the silicate melt as chloride complexes in an aqueous solution, whereas Al and F became concentrated in the silicate melt until saturation in topaz was reached.

Since topaz is not on the normal "liquidus line-of-descent" for silicate melt compositions, it is concluded that topaz appears through the operation of fluid phases, whether under magmatic or subsolidus conditions.

Mullite

There are no modern references to the occurrence of mullite in natural granitoid rocks, and yet with a composition between the alumino-

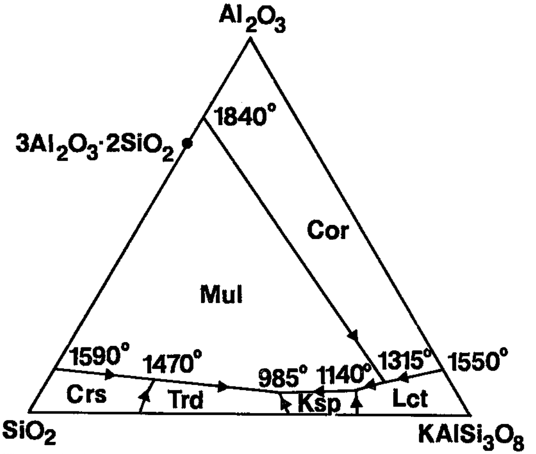

FIG. 6. Liquidus phase relations in the system $KAlSi_3O_8 - SiO_2 - Al_2O_3$ at 1 atm pressure (after Schairer & Bowen 1955).

silicates and corundum, both of which are found in peraluminous granites, and with a suitable stability field (Fig. 1), it should be expected to occur. Part of the failure to recognize mullite may lie in the fact that its habit and optical properties are similar to those of sillimanite.

Phase relations in the system $K_2O - Al_2O_3 - SiO_2$ (Fig. 6) show a "Qtz" + Ksp + Mul + Liq invariant point ($\hat{\equiv}$ mullite haplogranite) but at a temperature only 5C° lower than the alumina-saturated haplogranite minimum (Schairer & Bowen 1955). Pugin & Soldatov (1976) have suggested that mullite may appear as a result of incongruent melting of sodic plagioclase; however, it is likely that failure to contain sodium in these experiments is responsible for the appearance of mullite. Sodium loss in these experiments may approximate the natural conditions under which mullite may form, *i.e.*, by escape of alkalis from normal peraluminous melts.

Spinel

Little detailed work has been done specifically on the occurrence of aluminous spinel phases in granitoid rocks. Most of those described have high Fe (hercynite) contents, *e.g.*, the Dartmoor granite (Brammall & Harwood 1923), or high Zn (gahnite) contents, as in the Himalaya pegmatite where the spinels are primary magmatic (Foord 1977) or the South Mountain batholith where hercynite–gahnite spinels occur in greisenized aplite and pegmatite (Farley 1979). Richardson (1968) suggested that hercynitic spinels could form from the oxidation of Fe-cordierite.

Spinels are significant in being one of only two non-silicate mineralogical expressions of peraluminous chemistry in granitoid rocks. Their stability is favored by high T, low $a(SiO_2)$, high fO_2 and a high concentration of zinc, but precise petrogenetic interpretations using spinels have yet to be attempted.

Corundum

Corundum is the ultimate mineralogical expression of alumina oversaturation in granitoid rocks. It normally occurs in pegmatites in which the corundum is generally described as being in textural equilibrium with other phases in the rock, such as andalusite, muscovite and quartz.

Some experimental work (Carr 1968) and thermodynamic data do not favor a stability field for Qtz + Cor. Phase relations in the simple system $KAlSi_3O_8 - SiO_2 - Al_2O_3$ (Fig. 6) show no down-temperature liquid path from mildly peraluminous silicate melts to those that would crystallize corundum; furthermore, Dimitriadis (1978) showed that the primary phase field of alumina oversaturation is reached at very low concentrations of excess Al_2O_3. Nevertheless, Blencoe (1974) reported the existence of an invariant point involving a haplogranite mineral assemblage plus corundum and silicate liquid, which he interprets as demonstrating the possible magmatic origin of corundum. It is not clear whether the frequent natural assemblage of Qtz + Cor represents a stable or metastable association.

Explanations for the origin of corundum itself vary greatly, but all involve specific modification of the original silicate melt or its products. Findlay (1907) and Wilson (1975) favored contamination by highly aluminous pelitic rocks. Rose (1957) accounted for inclusions of corundum in clots of andalusite and muscovite in a pegmatite by the reaction $6And + 2H_2O + K_2O \rightarrow 2Mus + 3Cor$. Rossovsky & Konovalenko (1977) appealed to desilication of the pegmatitic fluid by highly silica-undersaturated wall rocks. The pegmatitic association of corundum suggests the important role of fluids, whether to remove alkalis to raise A/CNK in the silicate melt fraction or, as suggested by Rose (1957), to promote subsolidus hydrothermal alteration.

Other phases

The phases discussed above are the common mineralogical manifestations of normal alumina-oversaturated bulk compositions, with nothing more exotic than minor amounts of B, F and Zn to account for even the more unusual phases. However, in late-stage leucogranites, many other elements, including Li, Be, P, Cs, *etc.*, may become strongly enriched and combine with excess Al to form phases such as spodumene $LiAlSi_2O_6$, lepidolite $KLi_2AlSi_4O_{10}(OH)_2$, beryl $Be_3Al_2Si_6O_{18}$, pollucite $(Cs,Na)_2(Al_2Si_4)O_{12} \cdot xH_2O$, amblygonite $LiAlPO_4(F,OH)$ and others.

It is at least debatable whether, at very late stages of evolution of a granite when the operation of a fluid phase is so important, these phases are characteristic only of peraluminous granites. It may be that convergence operates once again, with peraluminous, calc-alkaline and even peralkaline granitoid rocks all becoming highly alumina-oversaturated. Such a discussion is beyond the scope of the present work.

Summary and conclusions

Detailed phase-relations for some characteristic minerals of peraluminous granites have been discussed by Thompson & Algor (1977), Thompson & Tracy (1979) and Clemens & Wall (1981). Abbott & Clarke (1979) presented a large number of hypothetical liquidus paths for peraluminous melts in an expanded system including Fe and Mg, in which the precise topology for a given pluton was determined to be largely a function of pressure, temperature,

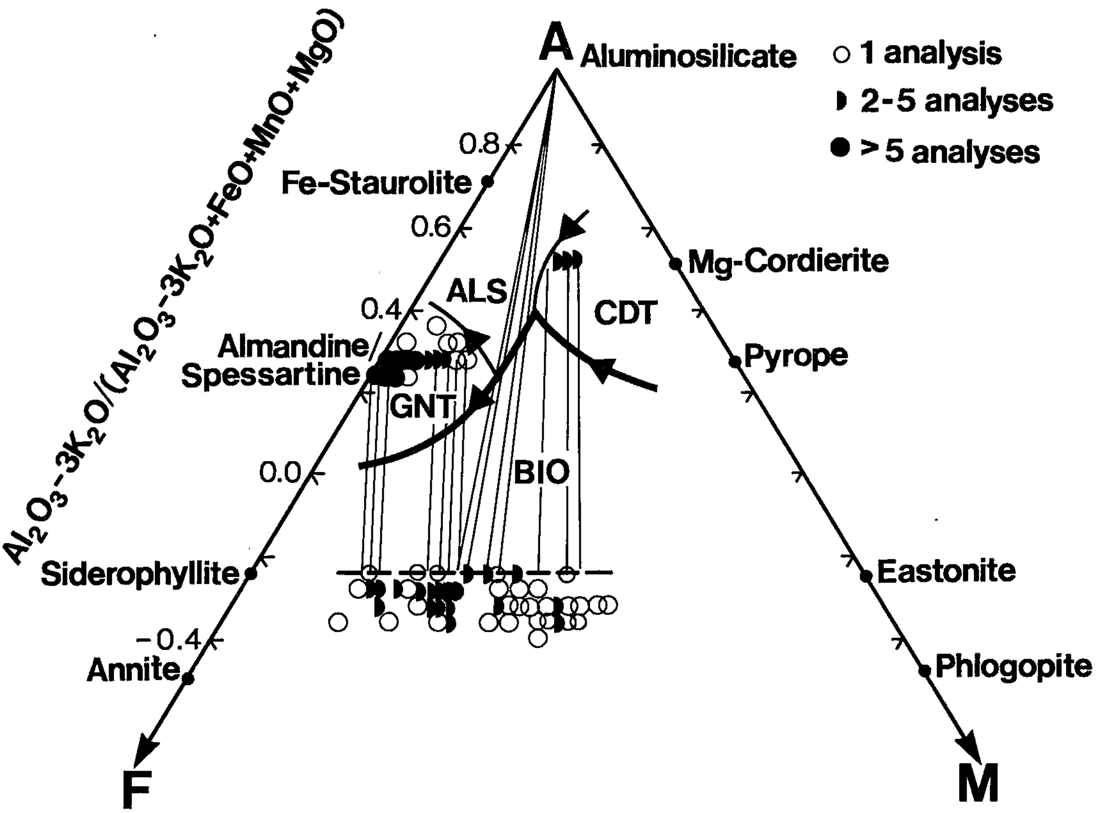

FIG. 7. Summary AFM projection from muscovite for the occurrence of primary magmatic biotite, garnet, aluminosilicate and cordierite in normal peraluminous granites. The tielines and cotectic paths are consistent with general trends of evolution in natural magmas. Many normal peraluminous melts have bulk compositions that project in the primary phase field of biotite and, through fractional crystallization, are joined by cordierite, aluminosilicates or garnet [depending on $(Fe + Mn)/(Fe + Mn + Mg$ ratio] along the heavy cotectic line. Biotite-free corderite–aluminosilicate, garnet–aluminosilicate or garnet–staurolite granites are rare or unknown. For other possible topologies, see Abbott & Clarke (1979).

TABLE 3. SUMMARY OF THE MODES OF OCCURRENCE OF COMMON CHARACTERISTIC MINERALS OF PERALUMINOUS GRANITES

			Biotite	Muscovite	Garnet	Cordierite	Alumino-silicates	Tourmaline	Topaz	Mullite	Spinel	Corundum
1. EXOGENIC	(a) restite	NORMAL PERALUMINOUS	x	x	x	x	x					
	(b) xenocrysts from high-level country rock		x	x	x	x	x					x
2. ENDOGENIC	(a) nucleated from silicate melt with A/CNK > 1		x	x	x	x	x	x			x	
	(b) nucleated from water-saturated, alkali-depleted silicate melt (A/CNK >> 1)	HYPERALUMINOUS		x	x		x	x	x	x	x	x
	(c) vapor phase-silicate rock metasomatic replacement			x	x			x	x	x	x	
	(d) nucleated directly from aqueous fluid			x	x			x				

activity of water and Fe:Mg:Mn proportions. These parameters govern the sequence of crystallizing phases and their possible reaction relationships. Figure 7 is a combined projection for the "AFM" phases shown in Figures 2, 4 and 5. The diagram shows the range of compositions of "AFM" minerals and their frequency of occurrence. Superimposed on the diagram is a general series of tie lines and a general liquidus topology. There are two features apparent from this diagram: (1) The conditions under which Liq + Als + Bio are stable are not always met, *i.e.*, the primary phase fields of cordierite and garnet may share a common boundary. (2) The cotectic path for peraluminous granites invariably passes between garnet and biotite compositions, rather than passing to the alumina-rich side of the garnet field where a field for staurolite might be encountered.

It is clear from the above descriptions that the specific mineralogical expressions of peraluminous bulk-compositions of granitoid rocks can originate in a variety of ways. Even a single phase in a single pluton may have more than one mode of origin, especially when P–T–*X* conditions on both sides of an intrusive contact may be so similar that porphyroblasts of aluminosilicates, garnet and cordierite from the thermal aureole, if incorporated as xenoliths, may be stable in the silicate melt. Only through careful textural, chemical and phase-equilibrium studies of the specific occurrence can its origin be determined.

Table 3 summarizes the various modes of origin that have been proposed, correctly or incorrectly, for the characteristic minerals of peraluminous granites. There are two main genetic subdivisions: exogenic (origin outside the igneous domain) and endogenic (origin as a result of internal processes within the silicate melt, or silicate melt – aqueous fluid system). These genetic subdivisions lead to two broad bulk-composition types of granitoid rocks, namely, those in which the A/CNK ratio is determined by silicate melt – silicate solid equilibria (here termed "*normal peraluminous*"), and those in which the A/CNK of the silicate melt phase (and hence later bulk-rock composition) is greatly increased by the evolution of a vapor phase rich in alkalis (here termed "*hyperaluminous*"). Normal peraluminous granitic magmas can only crystallize those phases that lie on the liquid line-of-descent for natural bulk-compositions such as that shown in Figure 7, and A/CNK ratios in melts and bulk rocks will be close to unity. Hyperaluminous granitic intrusions may be accompanied by alkali metasomatic aureoles such as occur at the Harding mine (Jahns & Ewing 1977); they may crystallize directly other phases such as mullite, topaz and corundum, or form such phases through the operation of a fluid on the silicate melt or solid rocks. The A/CNK ratio in the bulk rock is higher than that which could be reached during normal silicate melt – silicate crystal differentiation.

As can be seen from Table 3, there is no simple, universally applicable origin for a specific characteristic mineral. Each must be worked out on its own merit, and the reward of such painstaking work will be a greater understanding of the origin and evolution of the granitoid rocks that contain them.

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