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ISOTOPIC AND CHEMICAL CONSTRAINTS ON THE DEVELOPMENT OF PERALUMINOUS CALEDONIAN AND ACADIAN GRANITES

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ABSTRACT

Peraluminous magmas may be generated by a variety of mechanisms. Isotopic studies can provide strong constraints to be imposed on the processes involved. However, the equivocal nature of petrogenetic interpretations based on any one individual criterion requires the combined use of field, chemical, stable- and radiogenic-isotope data. Several plutonic suites in the Caledonides of Britain and the Acadian of Nova Scotia indicate a tendency for $(^{87}Sr/^{86}Sr)_i$ and $\delta^{18}O$ to increase during the development of peraluminous compositions. This observation can be adequately explained only by the assimilation or fusion of crustal rocks (which in some cases should be sedimentary), processes that best account for the generation of large volumes of peraluminous magma. The processes of vapor-phase transfer and fractional crystallization may be important in producing local variations in the ratio A/CNK.

Keywords: peraluminous granites, strontium isotopes, inherited zircons, oxygen isotopes, Caledonian, Acadian.

SOMMAIRE

Plusieurs mécanismes génétiques peuvent expliquer l'origine des magmas hyperalumineux. Les études isotopiques indiquent de puissantes contraintes à imposer sur les processus invoqués. Toutefois, vu l'ambiguïté des interprétations pétrogénétiques fondées sur un seul critère, il est essentiel de considérer à la fois les observations de terrain, les compositions et les données sur les isotopes stables et radiogéniques. Dans plusieurs associations plutoniques Calédoniennes (Grande-Bretagne) et Acadiennes (Nouvelle-Ecosse), le paramètre $\delta^{18}O$ et le rapport initial $^{87}Sr/^{86}Sr$ tendent à augmenter lors du développement des compositions hyperalumineuses. Cette observation ne peut que refléter l'assimilation ou la fusion du socle (d'origine sédimentaire dans certains cas), qui expliquerait aussi la formation de volumes importants de magmas hyperalumineux; quand bien même les processus de transfert *via* phase gazeuse ou cristallisation fractionnée pourraient rendre compte de certaines variations locales du rapport $Al_2O_3/(CaO + Na_2O + K_2O)$.

(Traduit par la Rédaction)

Mots-clés: granites hyperalumineux, isotopes de strontium, zircons hérités, isotopes d'oxygène, granites Calédoniens, granites Acadiens.

INTRODUCTION

Origins of peraluminous granites

Peraluminous granitic compositions may be generated by a number of diverse mechanisms including fractional crystallization, crustal anatexis and vapor-phase transfer. Isotopic and chemical data can place strong constraints on both the nature of the source region for the magma and the importance of processes such as assimilation and fractional crystallization in the evolution of the magma chemistry. In this paper we consider O, Sr and Pb (zircon) isotopic data for several calc–alkaline intrusive bodies of the Caledonian of Scotland and the Acadian of Nova Scotia, and their bearing on the development of peraluminous compositions. However, before doing so, it is useful to review prevalent models for the generation of peraluminous granitic magmas.

(*a*) *Melting of diopside-normative compositions*: Equilibrium fusion of crustal material of andesitic bulk composition may generate the calc–alkaline trend of magmas of the Sierra Nevada batholith (Presnall & Bateman 1973); if, as Cawthorn *et al.* (1976) pointed out, amphibole were a significant component of the residue, the melts might be corundum-normative. Experimental melting of basalts at 5 kbar P(H_2O) by Helz (1976) resulted in melt compositions that were consistently peraluminous. This was ascribed to the highly diopside-normative composition of the residual hornblendes. Helz also pointed out that at 10 kbar P(H_2O) and 1025°C, melts produced by anatexis in the system CaO–MgO–Al_2O_3–SiO_2 (Kushiro & Yoder 1972) were also peraluminous without the involvement of hornblende. This suggests that peraluminous melts may be characteristic of anatexis at high water-pressures in the presence of any phase containing normative diopside.

(*b*) *Fractional crystallization* of amphibole has been suggested as a possible mechanism for producing residual magmas that are peraluminous. Amphiboles have been shown to be important near-liquidus phases in basaltic and intermediate magmatic compositions (Cawthorn & O'Hara 1976). Ringwood (1974) suggested that fractional crystallization of amphiboles with appropriate Fe:Mg ratios may produce the calc-alkaline trend observed on AFM diagrams. Similarly, amphiboles stable in magmas of calc-alkaline composition are diopside-normative and thus should be capable of producing trends towards normative corundum with increasing SiO_2 (Cawthorn *et al.* 1976).

(*c*) *Anatectic melting* of peraluminous compositions, such as pelitic metasedimentary material, may result in peraluminous liquids rich in normative quartz, orthoclase and albite, together with residual minerals such as cordierite and garnet (White & Chappell 1977). Upon solidification, such a magma (melt plus restite) would have the peraluminous characteristic of an "S-type" granite [*i.e.*, one derived from a (meta-) sedimentary source] as defined by Chappell & White (1974). In this case, the peraluminous characteristic would be inherited from the source region by the melting of peraluminous phases and by the transport of refractory residual minerals.

(*d*) *Assimilation*: Contamination of a metaluminous magma by aluminous sediments during ascent or emplacement might give rise to chemical and mineralogical features similar to those found in granitic rocks generated by mechanism *c* above. However, the efficacy of such contamination by assimilation in bringing about *large-scale* changes in composition is uncertain.

(*e*) Vapor-phase transfer of alkalis from a metaluminous granitic liquid may cause the composition of the liquid to become peraluminous (Luth *et al.* 1964). Such mass transfer of alkalis is greatly facilitated by the presence of aqueous chloride solutions (Burnham 1967, 1979). Evidence for such a process comes from the presence of alkali halide salts in fluid inclusions in pegmatitic minerals (Roedder 1979) and the subsolvus nature of most peraluminous granites (Luth *et al.* 1964).

The relevance of isotope geochemistry

Radiogenic- and stable-isotope studies of igneous rocks are potentially powerful tools in defining the processes that may have been responsible for the production of peraluminous magmas. The $^{87}Sr/^{86}Sr$ ratio of a rock is related to the time-integrated effects of the Rb/Sr ratios of its components. Hence, the earth's continental crust, with its enhanced average Rb/Sr ratios, has developed higher average $^{87}Sr/^{86}Sr$ than the mantle. The effect of assimilation or melting of such crust, in general, is to add relatively radiogenic Sr to less-evolved, mantle-derived magmas. Oxygen-isotope ratios are fractionated to more ^{18}O-rich values during weathering, sedimentation and diagenesis (Savin & Epstein 1970). The ^{18}O-enriched isotopic signature of sedimentary and altered volcanic rocks will be passed on to their magmatic derivatives provided no other processes (*e.g.*, granulite-facies metamorphism or exchange with low-^{18}O meteoric waters) are involved. Clastic sedimentary rocks are commonly found to be depleted in ^{18}O during high-grade metamorphism, particularly at granulite-facies conditions (Shieh & Schwarcz 1974, Longstaffe & Schwarcz 1977). ^{18}O depletion, sometimes extreme, is a common feature of low-temperature interaction of granites (chiefly epizonal) with meteoric water (Taylor & Forester 1971). It is also to be expected in granites with melted or assimilated rocks that had previously undergone such exchange (Taylor 1977). Fractional crystallization should produce very little significant change, if any, in either $^{18}O/^{16}O$ ratios (Taylor & Silver 1978) or $^{87}Sr/^{86}Sr$, except (in the case of oxygen) in magmas of unusual and somewhat hypothetical composition. This is of major importance, since the process of assimilation, if accompanied by fractional crystallization, may produce little obvious change in the major element chemistry of a magma (Taylor 1980). This is not true, however, for trace elements such as Sr, Zr, Ba, Th and U which, together with $^{18}O/^{16}O$ and $(^{87}Sr/^{86}Sr)_i$ ratios, may be more sensitive indicators of such a process. U is strongly partitioned into zircon in granitic magmas; because zircon is a very resistant phase during partial melting, a Pb-isotopic memory may be carried in restite zircon if U-rich zircon were an abundant accessory phase in the source. Zircon is generally not abundant in basaltic and gabbroic rocks and, if present, has low U contents. Hence, inherited zircons are generally diagnostic of melting or assimilation of granitic or metasedimentary crust.

The five mechanisms (*a* to *e*) outlined above for the generation of peraluminous granites can be expected to produce a spectrum of isotopic effects. Melting of basaltic rocks (mechanism *a*) should yield magmas with low $^{87}Sr/^{86}Sr$ and low $^{18}O/^{16}O$ ratios unless the source basalt had undergone major low-temperature alteration (Muehlenbachs & Clayton 1972). However, little or no Pb-isotopic memory is to be expected in the zircons of rocks formed in such a manner. The generation of peraluminous compositions by fractional crystallization alone (mechanism *b*) will not produce any significant variation in $\delta^{18}O$ or $^{87}Sr/^{86}Sr$. Anatexis of most peraluminous pelitic sedimentary source material (mechanism *c*) will impart high $^{18}O/^{16}O$ and $^{87}Sr/^{86}Sr$ ratios to the melts unless the sediment has been metamorphosed to high grades. Similarly, contamination of a metaluminous magma by sediments (mechanism *d*) would be expected to produce a spectrum of correlated $^{18}O/^{16}O$ and $^{87}Sr/^{86}Sr$ ratios related to the degree of contamination and amount of cumulus phase removed from the melt. However, such correlations might also indicate a mixed igneous–sedimentary source, or magma-mixing processes. Vapor-phase transfer of alkalis (mechanism *e*) will not by itself cause major changes in $^{87}Sr/^{86}Sr$ or $^{18}O/^{16}O$ ratios. However, if the vapor phase is able to interact with country rocks and recirculate through the magma, some exchange is to be anticipated.

Clearly, the individual isotopic signatures do not always provide unequivocal evidence for the specific origins of magmas, and hence the combined investigation of isotopic, chemical, petrographic and geological characteristics is the most powerful approach by which to elucidate the possible roles of mechanisms *a* to *e* in the generation of a particular peraluminous magma. In this paper we take reasonably well-documented examples from the Caledonian of the British Isles and the Acadian of Nova Scotia to illustrate different kinds of chemical and isotopic variations in granites (*sensu lato*) bearing on the origin of peraluminous compositions in granites generally.

Terminology

Peraluminous rocks have been defined as having molar proportions of Al_2O_3 in excess of the combined molar proportions of Na_2O, K_2O and CaO (Shand 1943). Excess alumina is expressed as CIPW normative corundum, whereas alumina deficiency is reflected by normative diopside. Generally, we shall make use of the molar ratio $Al_2O_3/(CaO + Na_2O + K_2O)$ and refer to it as A/CNK. We shall also frequently refer to the initial $^{87}Sr/^{86}Sr$ ratio as $(^{87}Sr/^{86}Sr)_i$ and express the oxygen isotopic composition ($^{18}O/^{16}O$ ratio) in the familiar "δ" notation relative to standard mean ocean water (SMOW), as defined by the Snowbird Quartz (+16.20‰) and African Glass Sand (+9.60‰) standards.

SCOTTISH CALEDONIAN GRANITES

In the British Isles sector of the Caledonian fold belt there are abundant post-tectonic "granites" ranging in age from about 460 to about 390 *Ma* (Fig. 1). Early syntectonic granites (generally > 480 *Ma* old) are primarily migmatitic and sheeted complexes; we will exclude these from this discussion, though many are peraluminous. The post-tectonic granites display a variety of emplacement mechanisms, from forceful diapiric-like structures (apparently the majority) to the classical, passively emplaced cauldron-subsidence structures such as Glen Coe and Ben Nevis. This approach to classifying these granites by emplacement mechanism was used by Read (1961) in his important review of Caledonian magmatism. Subsequent studies have tended to follow this example or have adopted criteria such as apparent age (Pankhurst & Sutherland, in press) or geophysical expression (Brown & Locke 1979). No attempt to use compositional criteria has yet been made, presumably because of the rather sparse and sporadic nature of the available data. Inevitably some plutons have been thoroughly studied whereas others have been largely neglected. Consequently the quantity and quality of the data in the published literature over the past century are rather variable. The major oxide and isotopic data considered below have been gleaned from the literature. Analyses have been rejected only if it is quite clear that they do not adequately represent the described rock-type.

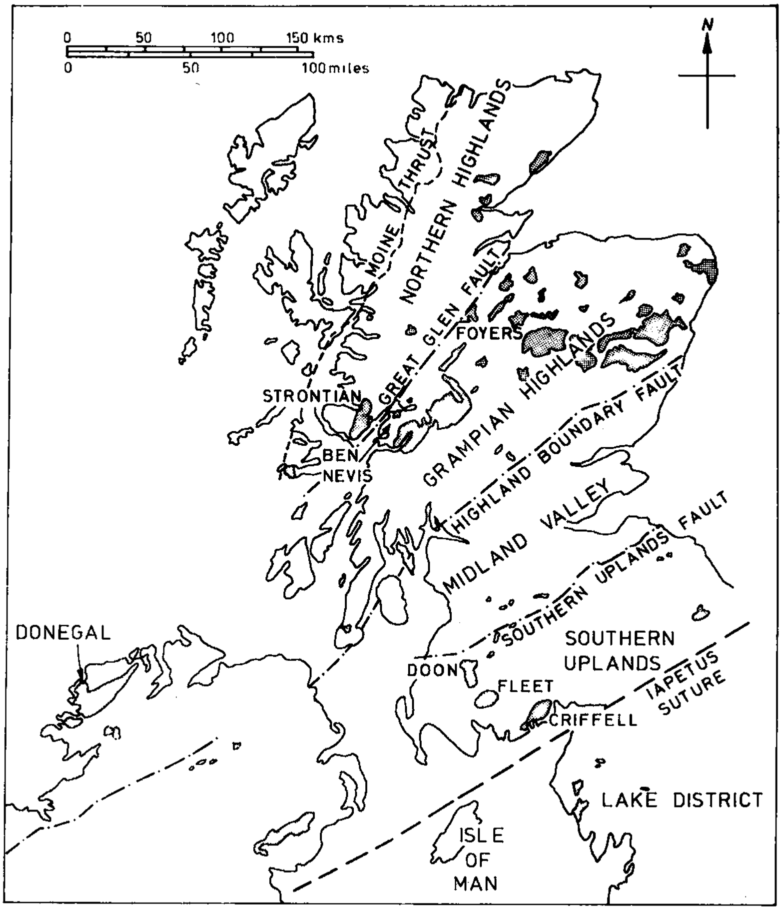

FIG. 1. Map of the northern part of the British Isles showing outlines of plutons, Caledonide zones, major faults and locations referred to in the text.

Northeastern Grampian region

The Dalradian of the northeastern Grampian region (Fig. 1) underwent Buchan-type (high-temperature) regional metamorphism *ca.* 500 *Ma* ago, and the rocks were cut by a number of (generally small) post-tectonic granites dated *ca.* 460 *Ma* (Pankhurst 1974, Pidgeon & Aftalion 1978). These granites are of particular interest because they represent the earliest examples thus far dated of Read's (1961) Newer Granites. The granites have high (^{87}Sr/^{86}Sr)$_i$ and ^{18}O/^{16}O ratios (> 0.71 and $\geq$ 10‰, respectively: Bell 1968, Pankhurst 1974, R.S. Harmon, unpubl. data).

The high (^{87}Sr/^{86}Sr)$_i$ and ^{18}O/^{16}O ratios indicate unambiguously that their genesis involved continental crust, local examples of which are known to have had high ^{87}Sr/^{86}Sr at this time (Bell 1968, Pankhurst 1974, Halliday *et al.* 1979). The presence of inherited zircons (Pidgeon & Aftalion 1978) indicates that this was more than just a selective contamination process, as proposed by Pankhurst (1969) for the slightly older Newer Gabbros. These 460-*Ma*-old intrusions are almost entirely granite (*sensu stricto*); they are commonly muscovite-bearing and lack amphibole. As there are no associated coeval basic intrusive bodies, with the exception of diorite at the Kennethmont complex, an origin by partial melting of the crust rather than assimilation has generally been accepted (Pankhurst & Sutherland, in press). Also included in this group is the Aberdeen granite, the exact age of which is uncertain but which shares the same characteristics of being a two-mica granite with inherited zircons, $\delta^{18}O > 10‰$ and $(^{87}Sr/^{86}Sr)_i > 0.71$ (Pidgeon & Aftalion 1978, Halliday *et al.* 1979, Harmon & Halliday 1980). The uncertainty in the exact age does not affect the conclusion that the $(^{87}Sr/^{86}Sr)_i$ is > 0.71.

The Hill of Fare granite contrasts with this group of granites in being passively emplaced (M. Munro, pers. comm. 1978), and younger than the other granites, with a Rb–Sr whole-rock age of 413 ± 3 *Ma* (Halliday *et al.* 1979). It also differs in that it is not muscovite-bearing, has a low $(^{87}Sr/^{86}Sr)_i$ of 0.7057 ± 3 (Halliday *et al.* 1979), a $\delta^{18}O$ value of $< 10‰$ (Harmon & Halliday 1980) and little or no trace of an inherited zircon component (Pidgeon & Aftalion 1978). These differences require a different source. Whereas the 460-*Ma*-old suite was likely generated by partial fusion of metasedimentary crust, the *ca.* 415-*Ma*-old granites appear to have been derived from melting at lower levels in the crust–mantle section.

In Figure 2 the A/CNK ratios are plotted *versus* SiO_2 for those granites for which chemical data are available. All of the *ca.* 460-*Ma*-old granites and the Aberdeen granite are moderately to strongly peraluminous. By contrast, the Hill of Fare granite does not have A/CNK ratios greater than 1.12, and one sample is metaluminous, despite evidence for strong fractionation as judged by the Rb/Sr ratios (between 1 and 15: Halliday *et al.* 1979). The available isotopic and chemical data combine to suggest strongly that mechanism *c*, melting of peraluminous metasediment, is the most likely direct cause of the high A/CNK ratios of the 460-*Ma*-old granites and the Aberdeen granite. Melting of altered basalt might explain the isotopic compositions but could not account for the inherited zircons. Fractional crystallization and assimilation are not compatible with the absence of evidence for more basic and metaluminous compositions. Vapor-phase transport, on its own, even if involving exchange with country rocks, will not account for the inherited zircons. In addition, there is no evidence for significant late-stage fluid activity in these intrusive bodies. However, melting of peraluminous Precambrian metasedimentary rocks is compatible with all isotopic and chemical data.

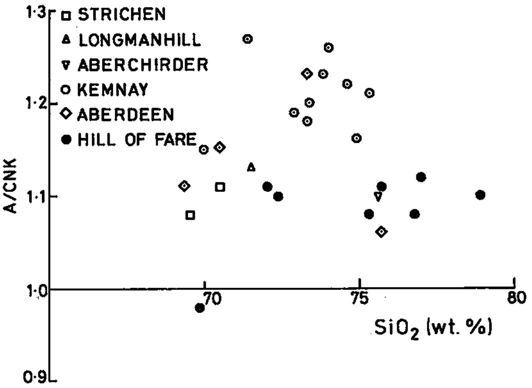

FIG. 2. Plot of the molar ratio $(Al_2O_3)/(CaO + Na_2O + K_2O)$ *versus* SiO_2 (wt. %) for north-eastern Grampian Highland granites. Most of the information for Aberdeen, Kemnay and Hill of Fare is derived from data of Walsworth-Bell (1974).

The Strontian and Foyers complexes

The Strontian and Foyers complexes are forcefully emplaced (Newer) granites in the scheme of Read (1961). They are of interest

TABLE 1. COMPARISON OF STRONTIAN COMPLEX WITH FOYERS COMPLEX

	Strontian	Foyers
Rock types	Zoned and composite chiefly mafic to felsic granodiorite	Zoned and composite tonalite to granodiorite to granite
Inherited zircons	Absent from earlier mafic portions, present in later felsic portions	Present in earlier tonalitic portion. Not determined in granodiorite or granite
$(^{87}Sr/^{86}Sr)_i$	Spectrum from 0.7052 to 0.7072 increasing with Rb/Sr and SiO_2	Tonalite and granodiorite exhibit spectrum from 0.7060 to 0.7081 increasing with Rb/Sr and SiO_2. Granite has uniform value of ~ 0.7045
$(\delta^{18}O)_{SMOW}$	Increasing with $(^{87}Sr/^{86}Sr)_i$ from 7.1 to 8.5‰ (three samples)	Tonalite 8.5‰ (one sample)
A/CNK	0.83 to 1.07 increasing with SiO_2	0.88 = lowest for tonalite increasing with SiO_2 to 1.02 in granodiorite. Granites are uniformly ~ 0.97 despite higher SiO_2

as a pair because Kennedy (1946) suggested, on the basis of their apparent petrographic similarities, that they were once a single complex; he postulated that they had been split apart by about 100 km of sinistral movement along the Great Glen fault (Fig. 1).

A summary of the isotopic and other related data for these two complexes is given in Table 1. Most of the Sr-isotopic analyses are those of Pankhurst (1979). In both cases the intrusions have been described as composite, comprising tonalite, granodiorite and granite phases, from margin to centre, approximately. However, whereas the modal data for Foyers (Marston 1971) fit these lithologies in the scheme of Streckeisen (1976), the rocks of Strontian (Sabine 1963) are all granodiorites, according to this classification. Nevertheless, the isotope systematics and composite nature of the Strontian complex make it convenient to retain the old terminology.

The Strontian complex has been dated by granodiorite and an amphibolite xenolith in U–Pb analyses of zircons from the tonalite, the the tonalite, which yield a combined age of 435 ± 10 *Ma* (Pidgeon & Aftalion 1978, Halliday *et al.* 1979). Whereas there is no isotopic indication of an inherited zircon component in these phases, the central granite at Strontian contains a marked Pb-isotopic memory of inherited Precambrian zircons (Halliday *et al.* 1979). The Rb–Sr isotopic data for Strontian define an apparent isochron age of ~ 1200 *Ma*. This "pseudo-isochron" is due to a variation in (^{87}Sr/^{86}Sr)$_i$ correlating with Rb/Sr (and approximately with SiO_2), which suggests either some kind of mixing process between a ^{87}Sr-enriched crustal component and a low-^{87}Sr mantle – lower crustal component, or a variation inherited from a single isotopically heterogeneous source (Pankhurst 1977, 1979). In the latter case, the ^{87}Sr/^{86}Sr variation might result from the melting of an old, differentiated igneous complex at depth. Such a simple origin is precluded, however, by the 1.4‰ range in δ^{18}O values, which indicates that the O isotopes, and hence probably also the Sr isotopes, were never in equilibrium in the source. The difference in Pb-isotope systematics in the zircons is similarly difficult to explain in this manner. Hence, these results must reflect either a mixed source (*e.g.*, amphibolite overlain by metasediment) or an assimilation process of some kind. The inherited zircons can only be residual minerals from xenoliths or the source and therefore strongly constrain the process to melting and disaggregation of crustal rocks (Leake *et al.* 1980) rather than simply selective contamination by O and Sr carried in a fluid phase (*cf.*, Briqueu & Lancelot 1979).

In striking contrast to Strontian, the Rb–Sr data for the Foyers granite define a *ca.* 415 *Ma* isochron (*i.e.*, similar in age to Strontian) with a uniform initial ^{87}Sr/^{86}Sr ratio of ~ 0.7045 (Pankhurst 1979). However, the tonalite and granodiorite Rb–Sr data define a "pseudo-isochron" age of ~ 750 *Ma*. The Foyers complex also contrasts with the Strontian complex

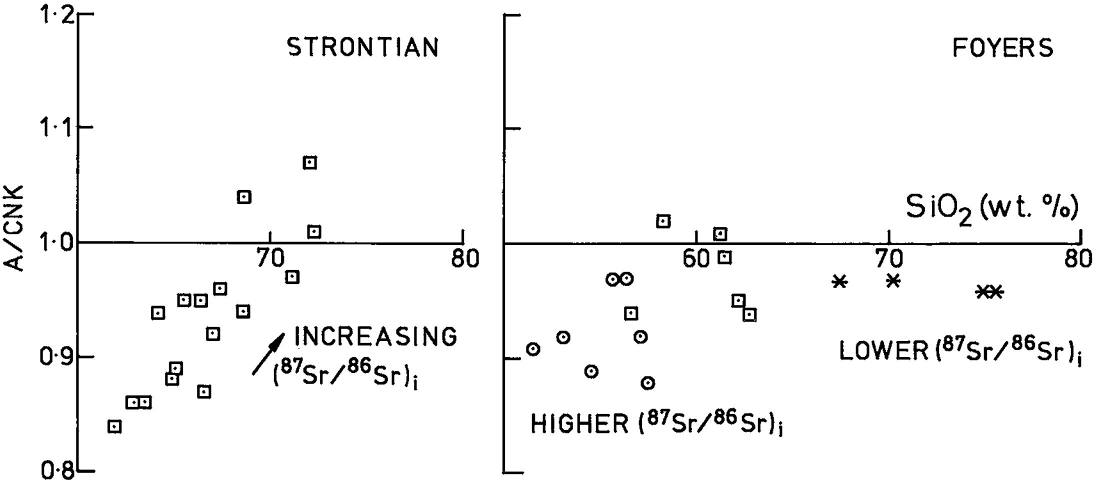

FIG. 3. Plots of the molar ratio $(Al_2O_3)/(CaO + Na_2O + K_2O)$ *versus* SiO_2 (wt. %) for Strontian and Foyers samples. Circles, tonalites; squares, granodiorites; stars, granites (nomenclature according to Streckeisen 1976).

in having a marked Pb-isotopic memory in the zircons of the tonalite (Pidgeon & Aftalion 1978). These data indicate that at Foyers it was the earlier tonalitic and granodioritic magmas that assimilated or were melted from crustal rocks with high $^{87}Sr/^{86}Sr$; the central granite shows no sign of contamination and was probably derived from a relatively primitive source in the lower crust.

Figure 3 is a plot of A/CNK *versus* SiO_2 for Strontian and Foyers. There is a relatively simple trend of increasing A/CNK with SiO_2 for Strontian, which is paralleled by an increase in initial $^{87}Sr/^{86}Sr$ and $^{18}O/^{16}O$ ratios and the change due to an inherited zircon population. (The results cannot be contoured because sample equivalence is not exact.) The data for Foyers are markedly different insofar as some of the granodiorites are peraluminous ($A/CNK > 1$) whereas the granites are not. Hence it appears that the major element chemistry reflects the degree of crustal interaction. That is to say, only the rocks that display an increase in initial $^{87}Sr/^{86}Sr$ with Rb/Sr and SiO_2, along with other evidence for crustal contamination, display the tendency for A/CNK to increase with SiO_2. The simplest explanation for this trend is that the high-$^{87}Sr/^{86}Sr$ Precambrian crust involved was peraluminous. The tendency for $\delta^{18}O$ to increase with $(^{87}Sr/^{86}Sr)_i$ at Strontian would suggest that this is metasedimentary crust, or its igneous derivative. The fact that the $\delta^{18}O$ values are relatively low (see Table 1) is compatible with the metasediment having previously undergone high-grade (*e.g.*, granulite-facies) metamorphism (Longstaffe & Schwarcz 1977), as has been predicted elsewhere for the crust underlying this region (Richardson & Powell 1976, Wells & Richardson 1979). None of the other models for the generation of peraluminous compositions fit the data as well as model *d*, the interaction of metaluminous magma with peraluminous crust, although the data could be explained by the melting of a composite peraluminous metasedimentary and metaluminous metaigneous source. Whatever the case, the development of peraluminous compositions appears to be due to melting or assimilation of metasedimentary crust or granitic crust derived therefrom.

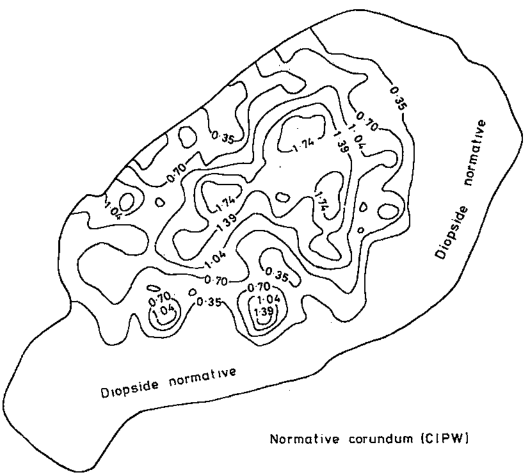

FIG. 4. Contour map of normative corundum in the Criffell pluton.

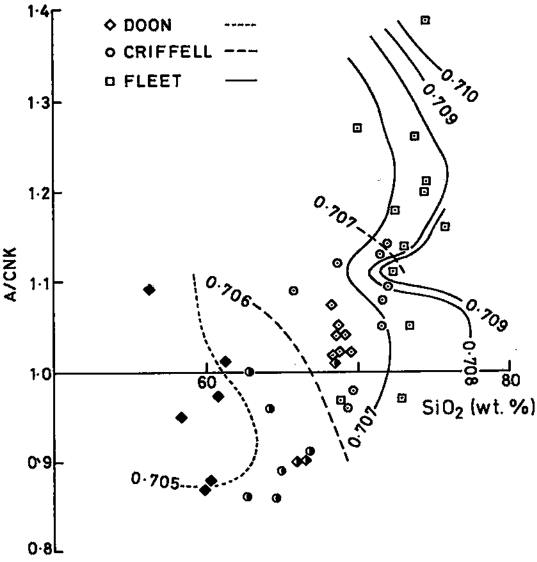

FIG. 5. Plot of the molar ratio $(Al_2O_3)/(CaO + Na_2O + K_2O)$ *versus* SiO_2 (wt. %) for Southern Uplands granites. Filled symbols, diorites; half-filled symbols, granodiorites; open symbols, granites. $(^{87}Sr/^{86}Sr)_i$ contours are plotted for each intrusion.

Southern Uplands

In southern Scotland, immediately north of the proposed site for the Iapetus suture in Britain (Fig. 1), late Caledonian plutons cut Lower Paleozoic flysch sedimentary rocks, forming a wedge likened to present-day accretionary prisms in trench–subduction-zone environments (Leggett *et al.* 1979). The three largest plutons, each 20 ± 2 km long, have been dated at 408 ± 2 *Ma* (Loch Doon), 397 ± 2 *Ma* (Criffell) and 392 ± 2 *Ma* (Fleet) by Rb–Sr mineral–

whole-rock isochrons (Halliday *et al.* 1980). This emplacement sequence is accompanied by a progression towards a higher average differentiation index DI (the sum of normative quartz, orthoclase and albite in SiO_2-oversaturated rocks: Thornton & Tuttle 1960), ($^{87}Sr/^{86}Sr)_i$ and $^{18}O/^{16}O$ ratios, a restriction of chemical variability and more peraluminous compositions. All three plutons are composite and normally zoned. At Loch Doon the lithology ranges from diorite to granite; at Criffell granodiorite and granite are present; Fleet is entirely granite. Within each pluton there is an inward progression towards higher DI, lower normative diopside, increased normative corundum, and higher ($^{87}Sr/^{86}Sr)_i$ and $\delta^{18}O$ values (Halliday *et al.* 1980). As illustrated in Figure 4, the chemical variability is particularly well established for Criffell, which has been studied in the most detail (Stephens & Halliday 1979, 1980).

Figure 5 shows the change in corundum–diopside normative compositions with SiO_2 in samples representative of the range of rock types for which Sr-isotope data are available. It can be seen that ($^{87}Sr/^{86}Sr)_i$ increases with increasing normative corundum and SiO_2 for Criffell and to a lesser extent for Fleet. The Doon data show little variation in ($^{87}Sr/^{86}Sr)_i$ or normative corundum. Some of the diorites are corundum-normative, apparently related to the abundance of biotite in these rocks; $\delta^{18}O$ values for the three plutons range from 7.8 to 11.8‰ and correlate with (^{87}/Sr/$^{86}Sr)_i$ and SiO_2 (Fig. 6).

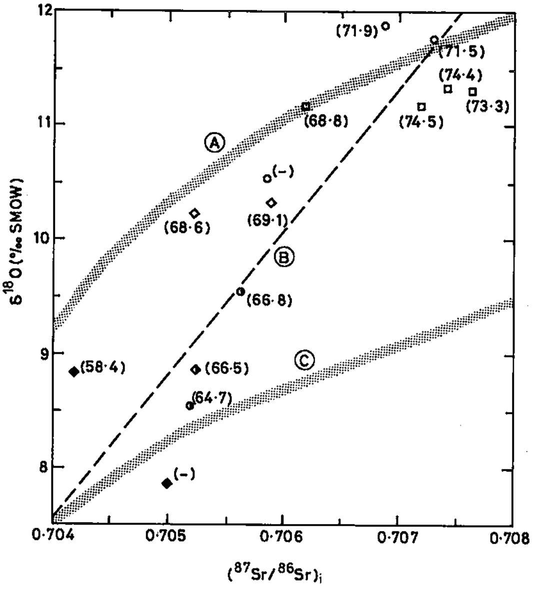

FIG. 6. Plot of $\delta^{18}O$ *versus* ($^{87}Sr/^{86}Sr)_i$ for Southern Uplands plutons (symbols as in Figure 5). SiO_2 (wt. %) contents, where known, are shown in brackets. Line B is the solution to a least-squares fit of the averaged diorite, granodiorite and granite data. See text for detailed explanation of curves A and C.

The isotope data and correlated O–Sr behavior illustrated in Figure 6 can only be explained in terms of interaction with continental crust; the high $^{18}O/^{16}O$ ratios suggest that this crustal material was low-grade and metasedimentary. Some indication of the process by which this crustal interaction occurred can be obtained from Figure 6. Here, following the modeling of Taylor (1980), model curves are shown to indicate the paths that would be followed (1) for the mixing of two appropriate end-member compositions with different Sr contents (A) and (2) for assimilation and fractional crystallization, where the ratios of magma to contaminant Sr content $[M/C]_{Sr}$ and cumulate to assimilated country rock (β) are both 5:1 (C). In both cases the end-member compositions are taken to be representative of the mantle or underplated lower crust ($\delta^{18}O$ = 6‰; $^{87}Sr/^{86}Sr$ = 0.703) and local Lower Paleozoic geosynclinal metasediments ($\delta^{18}O$ = 12.5‰, $^{87}Sr/^{86}Sr$ = 0.708). The straight line through the data (B) is the statistically significant (r^2 = 0.99) regression line through the data when averaged for each rock type (Halliday *et al.* 1980). From this modeling it is clear that assimilation and fractional crystallization are unlikely to have been the processes responsible for the observed O–Sr isotopic correlation unless the values of both $[M/C]_{Sr}$ and β are close to unity (Taylor 1980), or unless the process was simple bulk-mixing of two end-members of the same Sr content. Otherwise, the linear nature of the O–Sr correlation requires a more complicated evolution.

There are portions of each intrusion that display little variation in ($^{87}Sr/^{86}Sr)_i$ but are chemically variable, *e.g.*, in terms of Rb/Sr ratio and A/CNK. This has been explained by Halliday *et al.* (1980) in terms of fractional crystallization in a manner similar to that proposed by Vance (1961). Brown *et al.* (1979) have also suggested this for Loch Doon on the basis of chemical data. Hence, some of the variability in A/CNK may be due to this process. Note that in Figure 5, the ($^{87}Sr/^{86}Sr)_i$ contours for Criffell do not correspond to those of Fleet. In particular, the 0.707 contours are nearly perpendicular. There is very little evidence of late-stage fluids in the Loch Doon and Criffell plutons (*e.g.*, aplites and pegmatites are rare). However, at Fleet aplites and pegmatites are relatively common; Parslow (1971) concluded that felsic mineral distributions in the pluton were controlled by late- or postmagmatic processes. Movement of alkalis *via* a vapor phase would be an explanation for the tendency of the $(^{87}Sr/^{86}Sr)_i$ contours for Fleet to be somewhat irregular and near-vertical in Figure 5. However, explanation of the overall high A/CNK for Fleet in terms of vapor-phase transfer, rather than assimilation or melting of sediment, does not extend to the high $(^{87}Sr/^{86}Sr)_i$ and $\delta^{18}O$ values. As with the A/CNK ratios, the $(^{87}Sr/^{86}Sr)_i$ and $\delta^{18}O$ values are high overall and, as in the other plutons, there is a tendency for these parameters to increase toward the geographical centre of the pluton. There is little evidence of contact reactions that would suggest that the magma was not a closed system with respect to the immediate country rocks. Thus, it is doubtful that the overall high A/CNK, $(^{87}Sr/^{86}Sr)_i$ and $\delta^{18}O$ values at Fleet reflect vapor-phase transfer. However, all the data are compatible with these aspects as the direct result of melting of sedimentary material. Nonetheless, vapor-phase transfer could be a viable explanation for the wide variation in A/CNK for a given $(^{87}Sr/^{86}Sr)_i$ at Fleet.

THE SOUTH MOUTAIN BATHOLITH, NOVA SCOTIA

The South Mountain batholith is a post-tectonic composite batholith, chiefly of biotite granodiorite but with subsequent intrusions of adamellite and porphyry; the granitoid rocks were emplaced into the Meguma metasedimentary sequence (McKenzie & Clarke 1975, Clarke *et al.* 1976). Mineral and whole-rock Rb–Sr data for the intrusive phases define dates as follows: biotite granodiorite 372 ± 2 *Ma*, adamellite 364 ± 1 *Ma*, porphyry 361 ± 1 *Ma* (the latter two are those of the New Ross–Vaughan complex) (Clarke & Halliday 1980). The intrusion is of particular interest in the context of this study because it has been examined in detail chemically and isotopically and also because it is a large intrusion of peraluminous granite magma.

The intrusive phases generally have 1–4% normative corundum and between 67 and 77% SiO_2. There is a general tendency for the intrusions to become (temporally) more peraluminous and siliceous, although granodiorites from the contact with Meguma metasediments have slightly lower SiO_2 and higher normative corundum than some rocks further into the intrusion. The initial $^{87}Sr/^{86}Sr$ increases from the granodiorite (average 0.7081) to the adamellites (0.7094 ± 4) and porphyries (0.710 ± 1), an increase that can be explained in terms of variations in Rb/Sr produced by closed-system fractional crystallization over the eleven-million-year crystallization interval (Clarke & Halliday 1980). These values are lower than the $^{87}Sr/^{86}Sr$ of the Meguma sediments at the time of emplacement. However, granodiorites from near the contact with the country rock have higher $(^{87}Sr/^{86}Sr)_i$, suggesting contamination with Meguma metasedimentary units. Oxygen-isotope data (Longstaffe *et al.* 1980, Harmon & Halliday, unpubl. results) indicate involvement of metasediments in the magma.

Longstaffe *et al.* (1980) obtained high whole-rock magmatic $\delta^{18}O$ values (10.1 to 12.0‰) for the various phases of the South Mountain batholith. The surrounding Meguma formation was found to have a range of $\delta^{18}O$ from 9.0 to 13.3‰, with no apparent dependence on metamorphic grade. The values for sediments and magmas being similar, Longstaffe *et al.* concluded that the magmas were the product of metasediment melting rather than assimilation of, or exchange with, sediment. This conclusion is supported by general features of the batholith, namely, its size (the largest in the Appalachians), its uniform peraluminous composition (unlike Strontian, Foyers or the Southern Uplands granites) and the lack of more basic intrusions associated with its emplacement. This conclusion suggests that the magmas were not low-degree partial melts and not differentiates of basic magma. The combined information indicates that the source was either sedimentary (but not entirely Meguma) or a mixed sedimentary–igneous source and that the peraluminous nature of the magmas is a direct result of sediment melting. Since granulite-facies metamorphism appears to be associated with a lowering of $\delta^{18}O$ values (*e.g.*, Shieh 1974), the metasediment involved was not high-grade basement; therefore the Meguma appears to be the most likely candidate. In this respect it is of interest that Longstaffe *et al.* (1980) found lower $^{18}O/^{16}O$ ratios in the smaller plutons to the south of the South Mountain batholith, where the country rocks are at a higher grade. They attributed this to the melting at depth of sediment that had undergone high-grade metamorphism and exchanged oxygen with a low-^{18}O reservoir.

IMPLICATIONS FOR THE GENESIS OF PERALUMINOUS MAGMAS

From studies of the above-mentioned granites, it is clear that the development of strongly peraluminous compositions is frequently paralleled by isotopic evidence for melting and assimilation of crustal rocks. In some instances, for example, the Southern Uplands granites and the South Mountain batholith, these crustal rocks are quite clearly metasedimentary, as indicated by the high-^{18}O nature of the most evolved granites. In the case of Strontian and Foyers, the oxygen-isotope signatures suggest that if metasediments were the crustal material involved, it would already have undergone high-grade metamorphism.

In the case of the Southern Uplands granites, there is evidence for major changes in chemistry with little or no change in (^{87}Sr/^{86}Sr)$_i$. This is most reasonably explained by high-level fractional crystallization, restite separation or (in the case of Fleet) perhaps vapor-phase transfer. However, the overall strong tendency for A/CNK to increase with (^{87}Sr/^{86}Sr)$_i$ and δ^{18}O in these granites is powerful evidence for the importance of sediment melting or assimilation (or both) in the development of peraluminous compositions. However, modeling the Sr- and O-isotope data in the manner proposed by Taylor (1980) suggests that the process was not simply one of combined assimilation and fractional crystallization. The data for the South Mountain batholith also suggest a direct causal relationship between sediment melting and peraluminous compositions. Similarly, the tendency towards high A/CNK in the Strontian and Foyers complexes is matched by isotopic evidence that unambiguously indicates crustal assimilation or melting (or both).

We find some previously considered models for the origins of peraluminous magmas unable to explain all the observations. Amphibole fractionation (Cawthorn *et al.* 1976) does not account for the overall tendency of increased normative corundum with increased (^{87}Sr/^{86}Sr)$_i$ and, in some instances, δ^{18}O. We consider it probable that amphibole fractionation is a rather minor mechanism for generating large volumes of peraluminous magma. On the other hand, it is abundantly clear that the criterion of $>$ 1% normative corundum as diagnostic of "S-type" granites (Chappell & White 1974) is not unambiguously applicable, because it is possible to obtain magmas of this composition without invoking incorporation of sediment, as outlined in the Introduction. It is also clear that the (^{87}Sr/^{86}Sr)$_i$ ratio depends on the ratio Rb/Sr and the age of the source rather than its possible sedimentary nature, and that the O-isotopic composition of a metasediment depends on metamorphic grade. Hence, these criteria alone cannot in the first instance prove the involvement of sediment and in the second instance negate that possibility (*cf.*, Chappell & White 1974, O'Neil & Chappell 1977).

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