Chemical zoning of white micas: A record of fluid infiltration in the Oughterard granite, western Ireland

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Abstract

Backscattered electron imaging and electron microprobe analysis of white micas from the Caledonian Oughterard granite in Connemara, western Ireland, reveal chemical zoning generated during a complex and prolonged history of alteration. Three main stages of white mica formation are identified in this granite. Al- and Ti-rich magmatic muscovite was initially overgrown by mica of celadonitic composition formed during early fluid infiltration and retrogression of biotite. Both mica types were subsequently affected by a series of discrete low-temperature alteration events related to further episodes of fluid influx. A variety of zones of Fe-poor, Si-rich mica were produced with textures pointing to both localized internal and marginal alteration of the existing micas.

Introduction

Many workers have addressed the problem of how to determine whether the white micas found in granites are of primary, igneous origin or formed as a result of later deuteric or hydrothermal processes (cf. Speer, 1984, and references therein). The distinction is an important one, as the presence of muscovite in igneous rocks has been used both as an indicator that the magma was peraluminous (Miller et al., 1981), and as a means of estimating the conditions under which it crystalized (Zen, 1988). Various textural criteria have been proposed (Miller et al., 1981) that enable igneous muscovite to be identified, and this mineral has been found to exhibit systematic optical (Roycroft, 1991) and chemical zoning (Meunier and Velde, 1982). Detailed studies have also shown that several generations of white micas of different origin and composition can occur in granitic rocks (Meunier and Velde, 1982; Monier et al., 1984) but in those cases neither the textural nor the chemical criteria for distinguishing between igneous and secondary micas are diagnostic (Speer, 1984, p. 326).

We have examined white micas from three samples of a granitoid from western Ireland and report significant chemical variations in these minerals that may be linked to a complex postmagmatic history. Such variations in mica chemistry are probably common in granites and allow important constraints to be placed on fluid movements through the crust. They also have far-reaching implications for geochemical and geochronological studies.

Geologic setting

The Oughterard granite in Connemara, western Ireland (Bradshaw et al., 1969; Leake et al., 1981), consists of three irregular bodies (Fig. 1). These bodies are probably the surface expression of a single pluton at depth, together with many small satellite intrusions. The Dalradian metasedimentary country rocks are of late Proterozoic age and were intensely deformed and metamorphosed to upper amphibolite facies (Yardley et al., 1980, 1987) before the granite was emplaced. They have been affected by three major folding events (D2–D4) (Tanner and Shackleton, 1979; Leake and Tanner, 1994) and the Oughterard granite was intruded at about the time of formation of the D4 Connemara antiform. The region was subsequently intruded by late Caledonian granites (Fig. 1 of this paper; Leggo et al., 1966), and the country rocks were affected by pervasive retrogression (Jenkin et al., 1992).

The Oughterard granite has a characteristic pale gray color, is generally aphyric, and is typically relatively fine grained (<3 mm). It consists of quartz, oligoclase, and microperthitic microcline, with minor amounts of biotite and muscovite; it also includes a rare hornblende-bearing variety (Bradshaw et al., 1969). White mica and biotite vary in abundance, and alteration of biotite to chlorite is common. Plagioclase is compositionally zoned and clouded with sericite. The granite lacks a penetrative tectonic fabric but is locally strongly sheared, and chlorite-coated fractures are ubiquitous. It may contain a weak foliation. In thin section, cleavage planes in white mica and albite twins in plagioclase are commonly slightly kinked, and quartz develops subgrains. The Oughterard granite lacks a chilled margin and appears to have been intruded into rocks that had not completely cooled from peak metamorphic temperatures attained during D3. Its absolute age is not well known, despite numerous Rb-Sr and K-Ar isotopic dating studies involving whole rock and mineral analyses (Leggo et al., 1966; Moorbath et al., 1968; Leake, 1978; Kennan et al., 1987). These have yielded a variety of age estimates, and the topic remains controversial: together with Leake (1988), we favor an

age of 460–475 Ma, whereas Kennan et al. (1987, 1989) proposed that the Oughterard granite belongs to the suite of Caledonian granites dated at 400 Ma.

Samples have been studied from the three main granite masses and some of the satellite bodies, including a small intrusion exposed in a new cutting 100 m long immediately south of the fish hatchery at Lough Derryclare (loc. A, Fig. 1; Leake et al., 1981, grid ref. 831482). The latter intrusion is identical in appearance and petrography to other exposures of the Oughterard granite. Three samples of granite selected to represent a variety of alteration textures are described in detail below. They are representative of the variations shown by a number of other samples that we have examined from different parts of the Oughterard granite and comprise samples in which (1) biotite is unaffected by alteration (C1825); (2) biotite has been substantially altered to chlorite (C1878); and (3) muscovite is abundant, and the granite shows extensive alteration (C1888).

Sample localities are shown in Figure 1; two, C1825 and C1888, are from the small body of granite near Lough Derryclare (loc. A), collected 53 m apart, and the other, C1878, is from the south side of Lough Bofin (loc. B). The micas in these samples have been investigated to determine the chemical variations both within and between grains.

TECHNIQUES

Following petrographic examination, C-coated, polished thin sections were analyzed by backscattered electron imaging on the scanning electron microscope. This
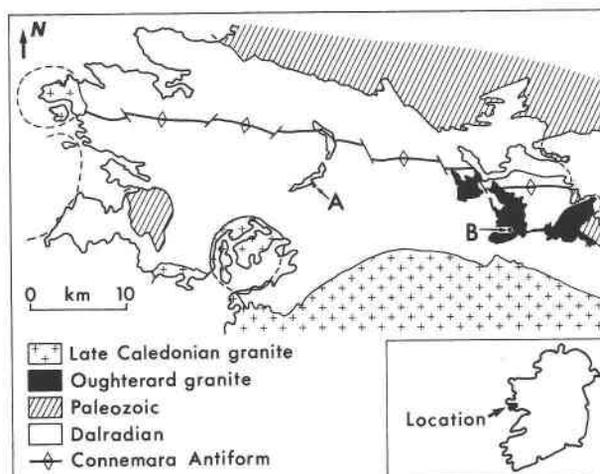

Fig. 1. Outline geology of Connemara, western Ireland (see inset), showing the locations (A, B) of the samples described in this paper.

technique highlights differences in mean atomic number (m.a.n.) and successfully detects some of the chemical variations found within the populations of white micas, although other major chemical variations may be hidden from this technique if the m.a.n. is nearly constant. Chemical variations within individual white micas were further characterized by electron microprobe analysis (Table 1) and high-resolution X-ray distribution maps showing the abundance of individual elements in areas of particular interest. Variations in abundance of ele-

TABLE 1. Representative electron microprobe analyses of white micas

Sample	C1825	C1878		C1888					
		Core A	Rim B	Grp 1 A	Grp 2 B	Grp 3 C	Grp 4		Grp 5 F
							D	E	
SiO$_2$	44.47	44.63	46.27	44.80	47.81	49.11	48.13	48.22	50.82
Al$_2$O$_3$	31.21	32.70	30.61	32.36	28.41	27.81	29.20	28.45	28.41
TiO$_2$	1.56	0.52	0.43	0.85	0.30	0.48	0.44	0.26	0.11
FeO	3.79	3.78	3.55	4.04	4.07	3.64	3.26	3.89	2.50
MnO	0.05	0.07	0.05	0.05	0.02	0.10	0.01	0.08	0.06
MgO	0.93	0.75	1.21	0.95	2.45	2.37	1.97	2.28	2.32
CaO	—	0.01	0.21	—	0.02	—	0.02	0.07	0.06
Na$_2$O	0.51	0.55	0.79	0.76	0.20	0.19	0.13	0.20	0.06
K$_2$O	10.52	10.37	10.30	10.54	10.73	10.56	10.70	10.75	10.58
	93.04	93.38	93.42	94.35	94.01	94.26	93.86	94.20	94.92
Si	6.18	6.16	6.37	6.15	6.55	6.68	6.56	6.59	6.79
Al	1.82	1.84	1.63	1.85	1.45	1.32	1.44	1.41	1.21
Al	3.29	3.58	3.34	3.38	3.13	3.14	3.25	3.17	3.26
Ti	0.16	0.05	0.05	0.09	0.03	0.05	0.05	0.03	0.01
Fe^{2+}	0.44	0.44	0.41	0.46	0.47	0.41	0.37	0.44	0.28
Mn	0.01	0.01	0.01	0.01	0.00	0.01	0.00	0.01	0.01
Mg	0.19	0.15	0.25	0.19	0.50	0.48	0.40	0.46	0.46
Total (oct)	4.09	4.23	4.06	4.13	4.13	4.02	4.07	4.11	4.09
Ca	—	0.00	0.03	—	0.00	—	0.00	0.01	0.01
Na	0.14	0.15	0.21	0.20	0.05	0.05	0.03	0.05	0.02
K	1.86	1.83	1.81	1.84	1.88	1.83	1.86	1.87	1.80
Total (alk)	2.00	1.98	2.05	2.04	1.93	1.88	1.89	1.93	1.83
Total cations	14.09	14.11	14.11	14.17	14.06	13.97	13.96	14.04	13.85

Note: cations per formula unit based on 22 O atoms; Fe$_{tot}$ = Fe^{2+}. C1825 = magmatic mica; C1878, A = magmatic mica, B = overgrowth on C1878A; C1888, A = magmatic mica, B = main overgrowth, C = structurally controlled alteration, D = diffuse alteration, E = halo adjacent to D, F = late marginal overgrowth.

ments with an atomic number lower than that of Na contribute to the backscattered electron images; however, these variations could not be identified using the electron microprobe. The backscattered electron images are commonly dominated by variations in Fe distribution because of its high atomic number relative to that of other major-element constituents of the white micas. Variations in the oxidation state of Fe are also hidden in these images, unless coupled charge balance substitutions result in significant atomic number variation. However, major changes in Fe^{2+}/Fe^{3+} seem unlikely because of the generally similar levels of octahedral site occupancy (Table 1).

PETROGRAPHY AND WHITE MICA CHEMISTRY

Sample C1825

This fine-grained gray, slightly foliated granite (typically 0.5–1 mm) has a mafic mineral assemblage dominated by biotite. Minor alteration of the granite is marked by chloritization of biotite (with associated rutile needles) and sericitization of feldspar, typically adjacent to discrete fractures, some of which may contain aggregates of white mica and minor calcite. Biotite is largely unaltered, and the quartz and feldspar crystals are less strained than is normal for the Oughterard granite. The granite is dominated by zoned subhedral oligoclase crystals (up to 1 mm) that have thin albite rims. It has abundant quartz and minor amounts of potassium feldspar. The white micas are Al-rich muscovite (Table 1), show no clear chemical variations either between or within grains, and are believed to be of magmatic origin.

Sample C1878

This pink, medium-grained, quartz-rich granite (typically 1–2 mm) has a weak foliation and contains similar proportions of potassium feldspar and plagioclase. The potassium feldspar occurs as large phenocrysts (up to 2 mm), zoned in Ba, which may enclose smaller euhedral plagioclase (of 1 mm). Alteration is pervasive, with biotite almost completely replaced by chlorite, rutile, epidote, and opaque minerals. Feldspars, especially the cores, are strongly sericitized to greenish plagioclase, and potassium feldspar commonly develops a red-pink color, which is especially intense close to late fractures. White micas are abundant and have commonly developed an irregular overgrowth of Fe- and Si-rich celadonitic mica (ca. 50 μm thick) on a core of euhedral Al-rich muscovite (ca. 600–300 μm) (Table 1). This texture indicates postmagmatic growth of white mica on original magmatic grains (cf. Speer, 1984).

Sample C1888

This sample was a medium-grained granite (typically 2–3 mm) but has been intensely altered to a gray-green rock containing minor amounts of potassium feldspar and quartz, abundant albite, and large white micas (up to 3 mm). The feldspars are sericitized and have deformed

albite twinning. Strongly twinned calcite is common, forming both as early veins and also filling later cavities. Chlorite is present in small quantities, apparently after biotite. The white micas have undulose extinction and are affected by variably oriented, open-kink folds. They contain complex chemical variations (Figs. 2, 3, 4, 5). Five main types of white micas have been identified in this sample:

1. Large books of Si-poor muscovite (Fig. 2a), which are abundant, are heavily kinked, and probably represent magmatic grains. These micas have a composition typical of magmatic muscovite (Speer, 1984), being Al-, Ti-, and Na-rich and poor in celadonitic component (Table 1). Some variation in m.a.n. appears to be controlled by the location of kink folds (Fig. 2a). However, electron microprobe major-element analyses show no correlation with this m.a.n. variation, and different volatile contents of the micas (e.g., H_2O, F) could be responsible, with the differences presumably established during deformation.

2. Overgrowths of coarse-grained Si-rich muscovite (Fig. 2a) form on the Group 1 micas. The overgrowths are large (typically 0.5 mm) and deformed and occur as discrete grains forming a partial overgrowth attached to the sides and ends of existing mica books. These celadonitic overgrowths are Al-poor (Fig. 2c) and Mg- and Si-rich (Figs. 2b, 3), compared with the larger magmatic grains. They have a high Mg/Fe ratio (Table 1) and are generally of similar composition to the celadonitic overgrowths in sample C1878. Thus, despite their large size, they are likely to represent postmagmatic growth. The contacts between these overgrowths and their host grains are typically well defined and often display crystallographic continuity (Fig. 4). However, such contacts may also be partially obscured by later alteration focused along the edge of the overgrowth (Fig. 5d).

3. Internal alteration in structurally defined zones (Figs. 4, 5) produces an Fe-poor mica (Table 1) of low m.a.n. in localized patches within existing Group 1 and 2 micas. These patches are commonly bounded by cleavage surfaces and are often developed in parts of the early micas containing fractures and kinks. Some of the micas displaying this structurally controlled type of alteration contain slight variations in their internal chemistry (Fig. 5b, z on Fig. 5d), with pockets of higher Al and lower Fe and Mg contents that, because of their more extreme composition, may reflect zones of more intense alteration.

4. Internal alteration of a diffuse nature (Figs. 4, 5) produces micas that are chemically similar to the Group 3 micas, although typically of slightly more extreme composition (i.e., Fe-poor) (Table 1). These micas are associated with an Fe-rich halo produced adjacent to altered zones (x and y on Fig. 4). The zones of structurally controlled type 3 alteration generally do not develop Fe-rich halos, suggesting that Fe may have been less mobile during this, possibly lower temperature, alteration event, although all the alteration must have taken place after the formation of the kink structures. The patchy type 4 alteration occurs within both Group 1 and Group 2 micas,
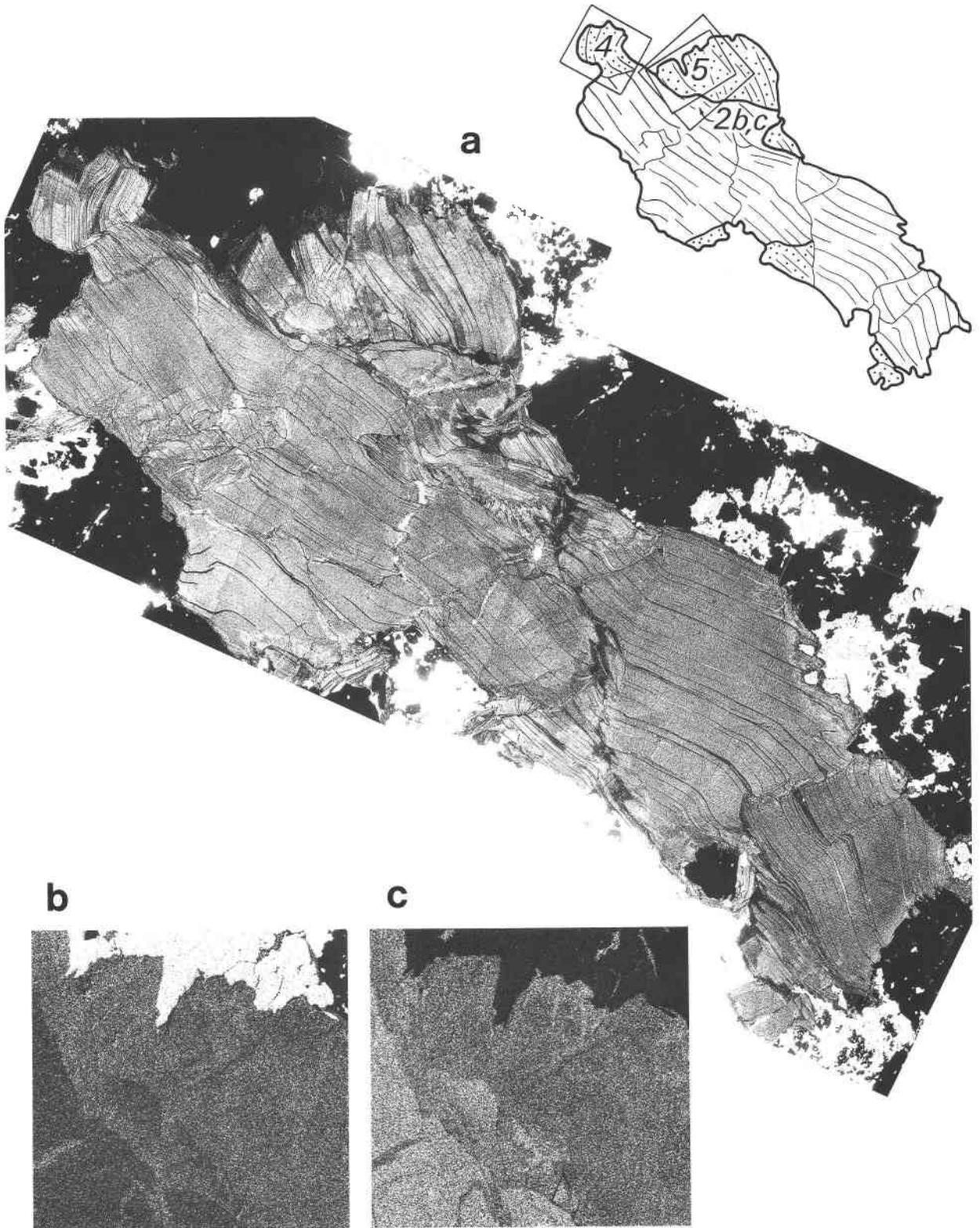
Fig. 2. (a) Backscattered electron image of white mica (3.5 mm long) from sample C1888, showing overgrowths on an original magmatic grain. The overgrowths are best developed at the top left side of the original grain and are shown in greater detail in Figs. 4 and 5. Inset (top right) shows location of late overgrowths (stippled). (b) High-resolution X-ray distribution map for Si, from contact between the largest overgrowth and the original grain (field of view 700 μm). (c) High-resolution X-ray distribution map for Al, from the same contact. Inset shows location of these and other figures.
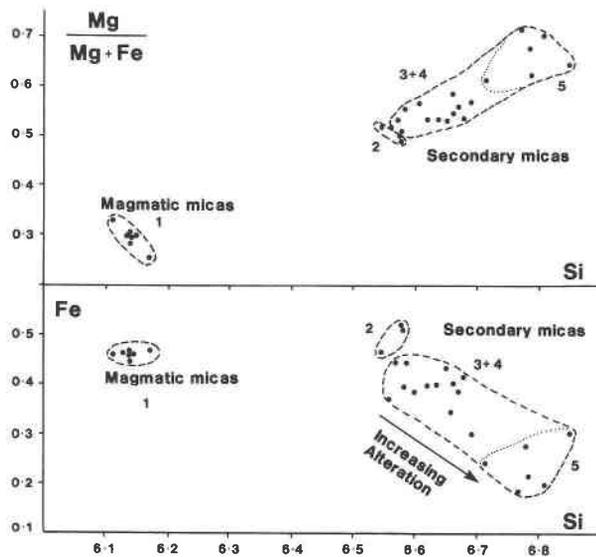
Fig. 3.   Chemical variation of magmatic (Group 1) and secondary (Groups 2–5) white micas, in cations per formula unit, from mica, shown in Figs. 2, 4, and 5 (C1888).

albeit often concentrated around zones of possible weakness, such as boundaries between Group 1 and Group 2 micas (Fig. 5). In such circumstances, a more prominent Fe-rich halo is developed on the magmatic grain (x on Fig. 4b), whereas in the Group 2 celdonite overgrowth the halo (y) appears diffuse, possibly because the altered mica is of a more similar composition to the host.

5. Marginal growth and alteration produces Fe-poor muscovite (Fig. 5a, 5c), which forms on, and replaces, Group 1 and Group 2 host micas as small zones of variable, but low, m.a.n. These zones of Fe-poor micas show weak, but regular, oscillatory zoning and are themselves often overgrown by small (50 μm) ragged shaped micas. Associated with this marginal alteration are internal zones, interpreted as alteration fronts and halos, of Fe-rich mica (Fig. 5c) superimposed on the existing micas. The Fe-poor zones are K-, Na-, and Ti-poor, have a high Mg/Fe ratio, and generally show high Si contents (Table 1). The adjacent Fe-rich "halos" show some minor depletion in Al and Si. All the late alteration effects tend to be most prominently developed within the Group 2 overgrowths, rather than within the original magmatic grains (Group 1).

Although texturally distinct, the three groups of late micas (3, 4, and 5) are chemically similar. However, the alteration appears to produce micas of progressively more extreme composition, such that the apparently later Group 4 and latest Group 5 micas are of increasingly Fe-poor composition (Fig. 3). The progressive nature of chemical alteration of the micas may support the argument that parts of the mica containing marginal alteration (Group 5) could represent zones of late alteration superimposed on the products of earlier periods of higher temperature alteration (e.g., Groups 3 and 4).

DISCUSSION

The variations in white mica chemistry preserved in sample C1888 may well be representative of those occurring in all highly altered micaceous igneous rocks. The celadonite overgrowths (Group 2 micas) on muscovite, of the type found in samples C1888 and C1878 (Table 1), may be characteristic of all two-mica granites that display some alteration of biotite. The magmatic micas show no evidence of growth zoning that could be equated with the optically zoned micas found in some granites (Roycroft, 1991). This indicates either that white micas of identical composition grew throughout the early crystallization history or that growth occurred at temperatures capable of eliminating zoning as a result of volume diffusion (Dodson, 1973).

The late micas are of two main types in the Oughterard granite, celadonite mica, (Group 2) and a relatively Fe-poor mica, which was produced during alteration of existing micas (Groups 3, 4, and 5) but also was associated with some new growth (Group 5). Late growth of a celadonite-rich mica has also been identified in metapelites (Dempster, 1992) and correlated with the retrogression of biotite. A similar control is probably operating in these granites, as there is a strong correlation between the presence of the overgrowths and the intensity of biotite alteration. Of particular note is the abundance of late (Group 2) mica overgrowths in C1888, the most altered sample. The granite contains minor secondary chlorite, which suggests that biotite was present, although substantial replacement by carbonate has occurred and so the original abundance of biotite is hard to assess. The alteration of feldspars may also produce mica, although it is as yet uncertain what role that plays in the formation of these white mica textures. Clearly, with relatively K-rich rocks such as granites, there may be many opportunities for postmagmatic growth of muscovite through interaction with late fluids. The growth of the late celadonite-rich overgrowths (Group 2) occurred prior to the prominent kinking of the micas and is thus a distinctly earlier phase of alteration than the development of the Fe-poor micas (Groups, 3, 4, and 5), which postdate the deformation.

The later alteration (Groups 3, 4, and 5) displayed by C1888 is also thought to be fluid related, because the patchy alteration is focused either along the margins of existing grains or around fractures or zones of late deformation within those grains. There have been several stages to this alteration process, each generating a distinctive texture of altered mica and affecting different parts of the mica structure. Thus there were probably many periods of fluid influx, and therefore mica compositions affected by later alteration may also have been altered by earlier events. The last, and probably lowest temperature, alteration appears to be the marginal effects (Group 5), which produced both the most extreme composition of mica (Table 1, Fig. 3) and the most noticeable halo of Fe. The formation of ragged late overgrowths, of similar composition to the altered mica, during or after the late marginal alteration events supports the argument that a degree of
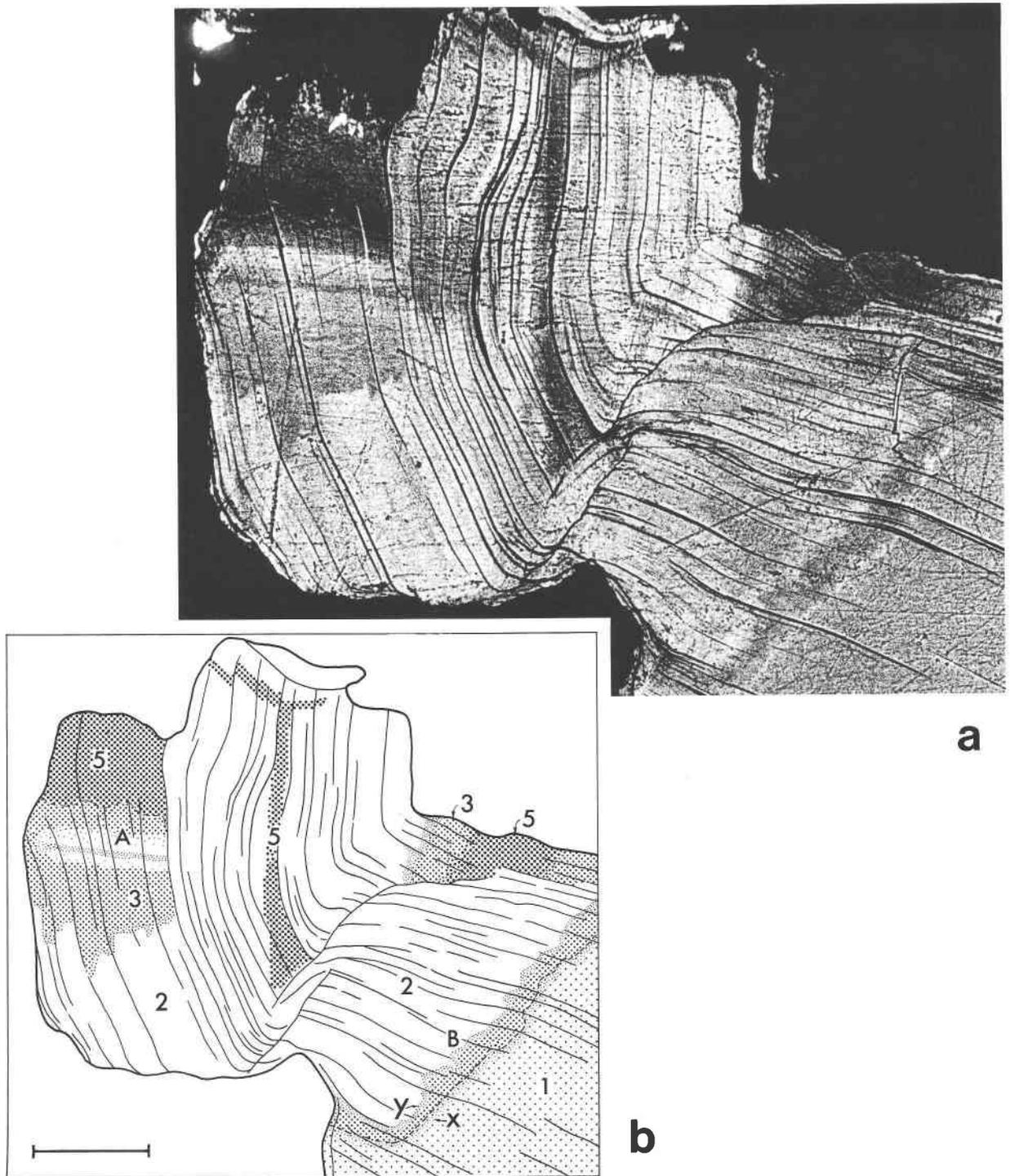
Fig. 4. (a) Backscattered electron image of an overgrowth (Group 2) on magmatic (Group 1) mica, with subsequent alteration (Groups 3–5) focused both at the margins and along the contact between the overgrowth and magmatic grains. See Fig. 2 inset for location. Sketch (b) summarizes various types of mica present; scale bar is 100 μm long. A, B, x, and y are referred to in the text.

chemical equilibrium was achieved. The chemistry of the altered micas is dominated by the loss of Fe and the increase in Al and Si and is similar to the chemical change during partial development of illite from muscovite (Velde, 1985). Although the late alteration occurred after

the deformation that may have allowed fluid access, the late alteration is most prominent in samples showing the greatest early alteration, meaning that ancient fluid pathways appear to have been reactivated.

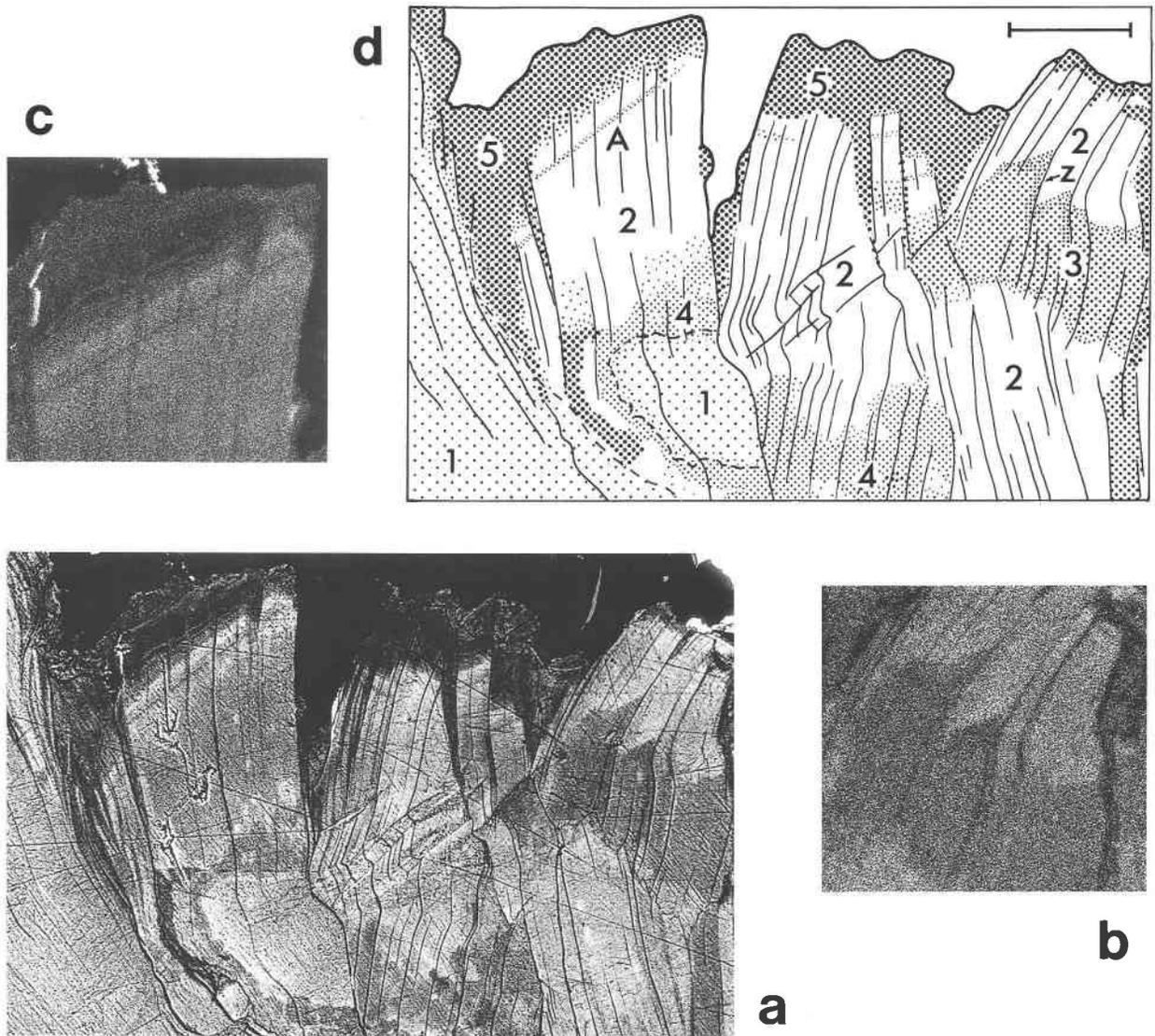The micas of different compositions provide con-

Fig. 5. (a) Backscattered electron image (field of view 600 μm) of part of the largest overgrowth (see Fig. 2, inset, for location). (b) Fe distribution map of internal Fe-poor structurally confined alteration; note small area of dark Fe-poor mica (see z on Fig. 5d) within generally altered area (field of view 180 μm); (c) Fe distribution map of marginal alteration and ragged Fe-poor overgrowth (field of view 180 μm). (d) Sketch summarizing the various types of mica present (Groups 1–5); scale bar is 100 μm long. A and z are referred to in the text.

straints on the movements of major elements within white micas. The sharp chemical boundaries separating the magmatic muscovite (Group 1) from its celadonitic overgrowths (Group 2) point to little diffusive reequilibration. The low-temperature areas of altered mica (Groups 3, 4, and 5) also preserve some sharp chemical gradients [especially in Fe (Fig. 5c)] but also contain more diffuse zones caused by differential movement of a variety of ions. This may reflect a combination of reaction and diffusion processes, with occasional patches of more intense internal alteration indicating some enhanced reaction possibly controlled by pooling of fluids by the mica structure (Fig. 5b). The complex chemical zoning produced

during the late alteration events (Groups 3, 4, and 5) appears, in part, to reflect a series of fronts produced during successive infiltration by different pulses of reaction within the host micas. These fronts are highlighted in the backscattered electron images by the relatively immobile Fe (Fig. 5c), which produces "halos" in both zones of marginal alteration (A in Figs. 4b, 5d) and zones of internal alteration (B in Fig. 4b).

The complex textures shown by the white micas in the Oughterard granite provide a wealth of information about alteration processes. Two very different types of alteration occur in radically different conditions. The earliest forms the coarse-grained celadonitic overgrowths (Group

2) and involves a major reconstitution of the mineral assemblage of the granite. This phase of alteration is thought likely to correlate with retrograde textures and replacement of biotite by chlorite in the Dalradian schists (Miller et al., 1991; Jenkin et al., 1992). The growth of these white micas may be associated with significant re-action-enhanced permeability, as the retrogression occurred prior to the deformation that caused the kinking of the mica. Later alteration produces many distinctive textural types of white mica (Groups 3, 4, and 5) and takes place after deformation, which may allow access for fluids but occurs in several discrete events rather than as a continuous process. The generally similar compositions of the micas that are produced by the latest events point to similar conditions prevailing during these episodes. Previous workers (Jenkin et al., 1992) have suggested that alteration in Connemara is a consequence of convective movement of meteoric waters, driven by the Galway granite. However, there were many stages to this alteration process and thus many potential sources of fluids. The crystallization of the Oughterard granite itself could be connected with fluid movements, and clearly the large Galway granite could have produced fluids at ca. 400 Ma, which might be partly responsible for the alteration. Additional fluid infiltration events could have occurred during the Carboniferous and Tertiary periods, associated with the intrusion of a few basic dikes (Mitchell and Mohr, 1986, 1987). Such complexities in the timing and conditions of fluid flow would be missed with studies on mineral separates, either being swamped by the signature of the dominant event or represented by a potentially misleading average. As yet such complications have only been observed within micas, which may provide a wealth of isotopic and geochemical data on the timing and conditions of infiltration and the sources of fluids. Other phases may preserve similarly complex records of these alteration events.

The parameters controlling the nature of fluid interaction with the crust can only be understood by characterizing the mineralogical changes that are produced during alteration. If a large-scale convective circulation of fluids was responsible for the alteration, then the late textures found in this granite argue against the continuous presence of fluids and rather for their occasional infiltration and subsequent reaction along zones of previous fluid activity. It may be that such patterns are controlled by periodic restrictions to the permeability as a consequence of either structural activity or the products of the alteration reactions themselves. Investigations of these textures over a larger scale may enable patterns of fluid flow to be determined.

SUMMARY

The development of white mica in the Oughterard granite occurred in three identifiable stages. A magmatic growth of muscovite was followed by the growth of a celadonitic mica, forming as a result of alteration of the existing magmatic mineral assemblage. The final development occurred in response to a series of relatively low-temperature hydrothermal alteration events. The complex white mica textures that have been observed are similar to those reported in other studies (e.g., Meunier and Velde, 1982), and we suggest that they are common in altered granites. Clearly granitoids may contain a variety of postmagmatic white micas, and so researchers undertaking studies based on mineral separates should be aware of the problems that this may cause, as such separates may be sampling minerals with a prolonged and diverse geological history. Under the circumstances, it is not surprising that Rb-Sr isotope dating of the Oughterard granite has so far proved inconclusive.

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