SECTOR-ZONED ANNITE₈₅PHLOGOPITE₁₅ MICAS FROM THE MT. LINDSAY Sn–W–F(–Be) DEPOSIT, TASMANIA, AUSTRALIA

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ABSTRACT

Annite₈₅ phlogopite₁₅ mica from the Sn–W–Be deposit at Mt. Lindsay, Tasmania, Australia, is sector zoned. In each sector, the highest $(Mg+Ti)$ values (Ti $\simeq$ 1.0, Mg $\simeq$ 3.5 wt. %) occur near the dark brown centres of crystals; the $(Mg+Ti)$ values and the intensity of the brown color decrease outward from the centres. $(Mg+Ti)$ values in the (001) sectors are much lower than in the (010) or (100) sectors. The structural formulae are satisfied by filling the tetrahedral positions with Si, Ti and Al. A small amount of Al also enters the octahedral position with Fe^{2+}, Mn, Mg and Zn. The F:Cl:OH ratios vary, with OH$>>$F$>$Cl. The compositional zoning out from the cores is due to depletion of both Ti and Mg in the local environment. Chemically, mosaic equilibrium was present to a scale of less than one millimetre. The sector zoning is probably due to the retention of "protosite" differences. The environment in which the micas are found indicates that sector zoning is not necessarily related to rapid crystallization, as has been suggested for examples elsewhere.

Keywords: Mt. Lindsay, annite, sector zoning, Tasmania, skarn.

SOMMAIRE

Le mica que l'on trouve au gisement Sn–W–Be du mont Lindsay (Tasmanie, Australie), de composition annite₈₅phlogopite₁₅, montre une zonation en secteur. Dans chaque secteur, la concentration $(Mg+Ti)$ la plus élevée (Ti $\simeq$ 1.0, Mg $\simeq$ 3.5%) en poids) se trouve au centre brun foncé du cristal. La somme $(Mg+Ti)$ et l'intensité de la couleur brune diminuent graduellement vers l'extérieur du cristal. La somme $(Mg+Ti)$ dans le secteur (001) est de beaucoup inférieure à celle des secteurs (010) ou (100). La formule structurale est satisfaite par l'occupation des positions tétraédriques par Si, Ti et Al. L'excédent d'Al occupe une position octaédrique ainsi que Fe^{2+}, Mn, Mg et Zn. Les valeurs des rapports F:Cl:OH sont variables, avec OH$>>$F$>$Cl. La zonation vers la bordure résulterait d'un appauvrissement progressif local en Ti et en Mg du milieu de croissance. Un équilibre en mosaïque existe sur une échelle de moins d'un millimètre. La zonation en secteur résulterait de la préservation de différences dans les "protosites" pendant la croissance. L'étude du milieu de crois-

sance indique que ce type de zonation n'implique pas nécessairement un taux de croissance rapide, comme on l'a proposé dans d'autres cas.

(Traduit par la Rédaction)

Mots-clés: mont Lindsay, annite, zonation en secteur, Tasmanie, skarn.

INTRODUCTION

Compositional zoning within individual mineral grains is common in both igneous and metamorphic rocks; it may reflect (1) depletion or change in the supply of constituents during growth (*e.g.,* Hollister 1966), (2) reaction or homogenization after initial growth (*e.g.,* Kwak 1970, Atherton & Edmunds 1966) or (3) chemical inequalities produced during growth along different crystallographic directions (*e.g.,* Nakamura 1973, Hollister & Bence 1967, Hargraves *et al.* 1970). The third process produces "sector" (Hollister 1967) or "hourglass" (Wass 1973) zoning.

Sector zoning has been reported for staurolite (Hollister & Bence 1967), pyroxene (Nakamura & Coombs 1973, Wass 1973, Hargraves *et al.* 1970), melilite (Sahama 1976), plagioclase (Bryan 1972) and anorthoclase (Havette & Nativel 1977). The only sector-zoned mica reported is a ferri phlogopite (Velde & Yoder 1977). Unfortunately, the authors did not include sufficient analytical data to enable the difference between the sectors to be properly defined.

Fe-rich mica is common in the Sn–W–F–Be skarn deposits of Mt. Lindsay and also occurs in other similar deposits, such as in the skarns at Iron Mountain, New Mexico, U.S.A. The purpose of this study is to define the compositional parameters of the zoning and explain possible environmental or compositional limits on this unique example of sector zoning.

GEOLOGY OF THE OCCURRENCE AT MT. LINDSAY

The Mt. Lindsay deposits are located ap-

proximately 24 km northwest of Renison, Tasmania, Australia, on the southern flanks of Parsons Hood (Mt. Lindsay). Skarn has replaced a number of carbonate units of the (Cambrian) O'Brien Formation (Jessop 1969) up to 150 m normal to the contact of a phase of the (Devonian) Meredith granite. The skarn units are, on average, 50 m wide and are bounded by a monotonous succession of medium grained subgreywackes and grey shale.

A study of the skarn by the author is in progress. The "No. 2 anomaly" skarn, which has received most attention, shows a well-defined zonal relationship as a function of distance from the granite contact. Farthest from the contact, either unaltered (a) andradite–grossularite + vesuviante + calcite ± sphene or (b) magnetite + ilmenite + cassi-

terite + K-feldspar + quartz + calcite + siderite (± scheelite ± danalite ± fluorite) skarns occur; these become variously altered to a (c) amphibole + Sn–sphene + fluorite + Sn–babingtonite + Fe-mica assemblage. Nearest the contact, skarns (a) and (b) have been altered entirely, often leaving amphibole-filled pseudomorphs of vesuvianite and garnet.

The Fe-mica will be called annite, although $Fe_{total}/(Fe_{total}+Mg)$ indicates only about 85% annite. The mineral forms either as a reaction product of amphibole in assemblage (c) or by direct reaction in assemblages (a) and (b). In some places, especially nearest the pluton, reaction has been complete and the rock consists of 90% annite, with minor fluorite and quartz. All the annite is not obviously sector-zoned; the best examples are from assemblage (b), especially where annite borders carbonate grains.
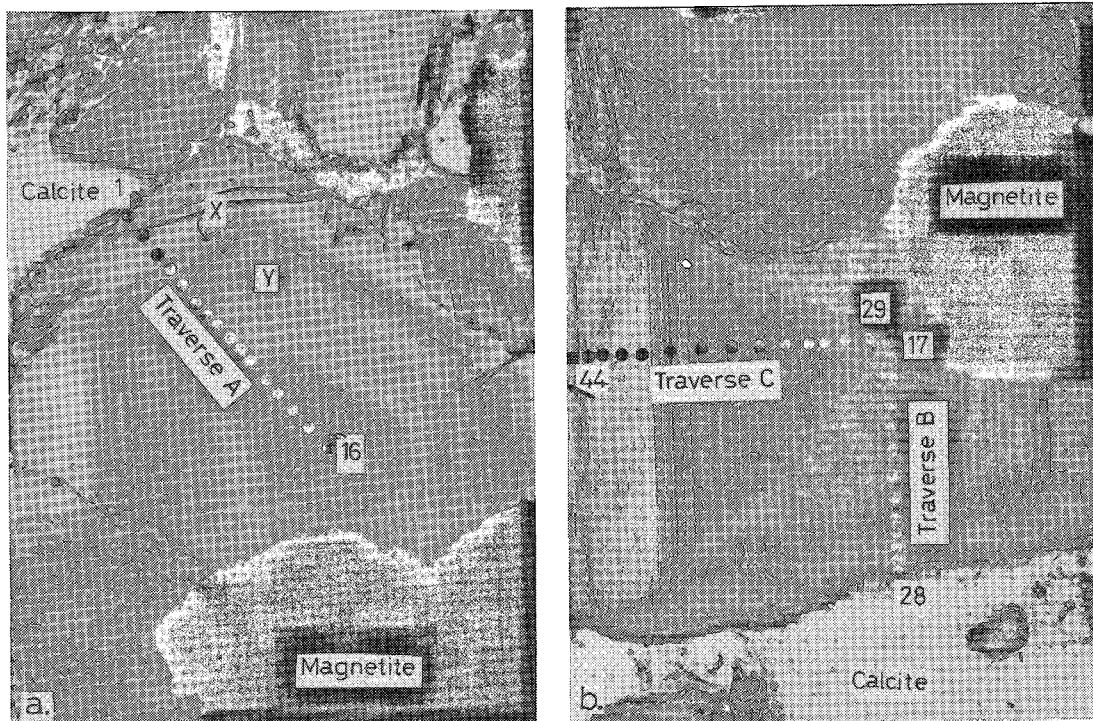

FIG. 1. Photomicrograph of sector-zoned mica (a) oriented normal to the Z axis, (b) parallel to the Z axis. The uniform light area in the centre of 1a is the [001] sector (see Fig. 2); the darker surrounding concentric area consists of the [110] and [010] sectors. The [110] and [010] sectors become progressively lighter towards the edge of the crystals where the grain does not occur in contact with grains of magnetite (black) or mica (grey). The color change from dark to light in these sectors is not entirely systematic, as lighter lamellae in a darker host do occur at points approximately two thirds of the distance from the centre (see Y). Also, there is a dramatic decrease in the darker color approximately three quarters of the distance from the centre (see X). Traverses A, B and C are 0.55, 0.58 and 0.55 mm long, respectively.
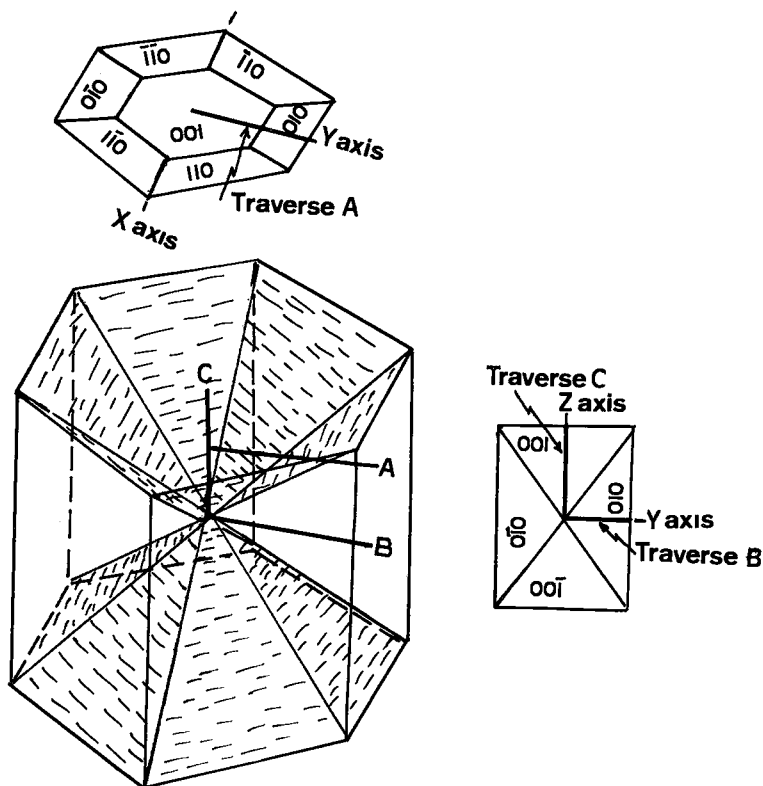
FIG. 2. The generalized relations of the sectors in the mica. The sectors are shown as two hexagonal pyramids whose apices meet at the centre of the crystal. The traverses are shown by the three dark lines within this perspective view and on the two sections. The sections represent views normal to the Z axis (traverse A) and parallel to the Z axis (traverses B and C).

MORPHOLOGY OF THE ZONING

The annite grains are much too fine to enable one to section a single crystal. However, the sector zoning appears identical in all of more than 100 polished thin sections studied. Typical examples of sector zoning observed on the (001) and (010–100) planes are shown in Figures 1a and 1b, and a diagrammatic representation of the zoning is shown in Figure 2.

In plane-polarized light, the limits of the sections are clearly shown by color differences. If grains are oriented with the Z axis approximately parallel to the plane of polarization of the microscope, the entire crystal is light green, and no sectors are evident. Normal to this, the sectors are clearly observable.

Sectors with triangular bases parallel to the (001) faces are dark brownish green near their

apices to medium green at the (001) face margins where the orientation of the crystal is parallel to the X or Y axes. Those having triangular bases parallel to (100) or (010) faces are dark brown near their apices and medium brown-green near their bases. Sections cut halfway between the centre and the (001) face (Fig. 1a) show a medium green core zone, a darker brown-green concentric zone and a lighter brown-green edge zone. No obvious differences were noted between sections normal to X or Y axes. The sector-zone relationships are obvious in most examples, but in others the boundaries between sectors may be irregular (e.g., Fig. 3a, b).

In all three point-analysis traverses shown in Figure 1, the color intensity and the degree of brown versus green decrease from crystal centres out to their edges. The outward change is more or less regular, but at approximately
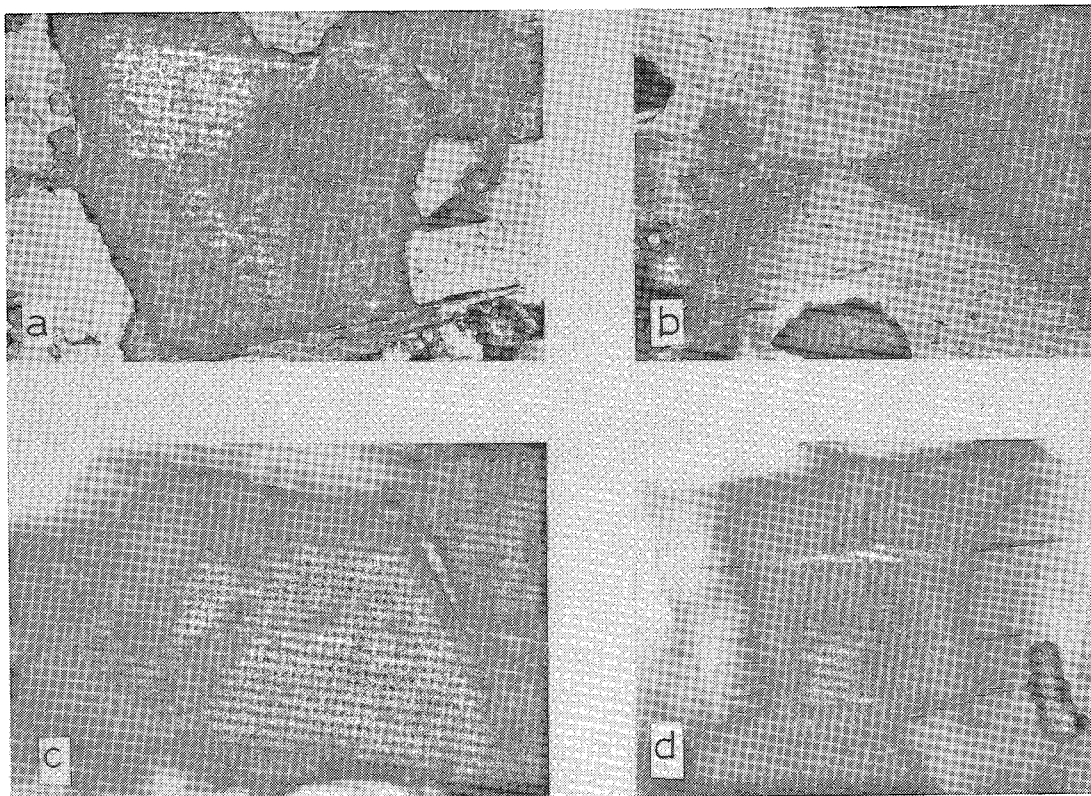
FIG. 3. Irregular sector-zoning of micas in the Mt. Lindsay deposit (a and b) and sector zoning in micas from the Little Hot Spot deposit, Iron Mountain skarn, New Mexico, U.S.A. (c and d). Figure 3c shows an example oriented normal to the Z axis; 3d shows an example parallel to the Z axis.

three quarters of the distance from the centre in Figure 1a there is a distinct decrease in both color intensity and degree of brown present (X in Fig. 1a). In addition, laminar areas of lighter color occur near the inner edge of the color change (Y in Fig. 1a), starting approximately two thirds of the way from the centre.

CHEMISTRY OF THE SECTORS

Continuous electron-microprobe spot analyses for Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, Cu, Sn, W, Zn, Cl and F were done along the traverses shown in Figure 1. The analytical methods and results of representative analyses are given in Table 1. The intervals chosen were so close that the edge of each analysis spot almost touched the next (see Fig. 1). The values of the elements that varied systematically (Mg, Ti and Fe) are shown graphically in Figure 4. Generally, (Mg + Ti) varies inversely with Fe (calculated here as FeO). The decrease in the

intensity of brown or brown-green near the centre of the crystals to light brown-green or green at the edge is reflected by decreasing (Mg + Ti). Lighter lamellae within darker brown-green zones (Y in Fig. 1a) have lower (Mg–Ti) contents. The values of all elements, including Ti, Mg, Fe, Cl and F, along a section parallel to the base of a sector (numbers 12 to 16, traverse A) are very similar. The color variations along such a section are also very slight. The two types of zoning seen optically, namely, sector zoning and the outward decrease of color intensity in particular sectors, correlate with a decrease in (Mg + Ti).

The outer edges of the crystals are lowest in (Mg + Ti), but (001) edges have compositions different from those on (010) or (100) edges (Fig. 4, traverses B and C). Variations in the oxidation state of Fe, not determinable here, may explain some of the unsystematic irregularities in the trends. Mn values are generally very constant (Table 1).

TABLE 1. REPRESENTATIVE ANALYSES OF ANNITE

	1	2	3	4	5	6
SiO2	34.56	35.29	35.80	36.04	34.65	34.82
TiO2	0.30	0.35	0.32	0.96	1.08	1.36
Al2O3	11.57	11.88	11.68	11.43	12.01	11.67
FeO	35.99	35.34	36.20	35.60	34.71	34.98
MnO	0.25	0.21	0.25	0.22	0.22	0.25
MgO	2.70	2.79	3.06	3.00	3.22	3.31
CaO	0.00	0.00	0.00	0.00	0.00	0.00
SnO	0.00	0.04	0.02	0.00	0.00	0.02
WO	0.00	0.00	0.00	0.02	0.00	0.04
Sn	0.14	0.09	0.01	0.10	0.04	0.06
Na2O	0.09	0.02	0.14	0.14	0.06	0.08
K2O	8.67	8.75	8.72	8.72	9.71	9.01
F	0.76	0.20	0.22	0.88	0.89	1.58
Cl	1.20	0.52	0.26	1.02	0.80	0.95
Anhydrous Total	96.23	96.50	96.96	98.13	96.89	98.24

Numbers of ions on the basis of 24 (O,OH,F,Cl)

	1	2	3	4	5	6
Si	5.64	5.71	5.85	5.98	5.72	5.74
Al	2.23	2.25	2.11	1.90	2.15	2.09
Ti	0.04	0.04	0.04	0.12	0.13	0.17
	7.91	8.00	8.00	8.00	8.00	8.00
Al	0.00	0.02	0.17	0.33	0.19	0.18
Fe+2	4.91	4.78	4.95	4.94	4.79	4.82
Mn	0.03	0.03	0.03	0.03	0.03	0.03
Mg	0.66	0.67	0.75	0.74	0.79	0.81
Sn	0.00	0.00	0.00	0.00	0.00	0.00
W	0.00	0.00	0.00	0.00	0.00	0.00
Zn	0.00	0.00	0.00	0.01	0.00	0.02
	5.60	5.49	5.87	6.06	5.80	5.86
Na	0.03	0.05	0.05	0.05	0.02	0.05
K	1.80	1.81	1.82	1.85	2.05	1.89
	1.83	1.86	1.87	1.90	2.07	1.94
F	0.39	0.10	0.11	0.45	0.45	0.82
Cl	0.33	0.14	0.07	0.29	0.22	0.26
OH	4.10	4.88	3.84	2.08	2.88	2.12
	4.82	5.12	4.12	2.83	3.56	3.20

	7	8	9	10	11	12
SiO2	35.09	34.75	34.94	34.31	34.33	35.21
TiO2	1.88	1.34	1.86	1.92	1.91	1.04
Al2O3	11.40	11.67	11.66	11.46	11.62	11.68
FeO	35.04	34.86	35.32	35.21	35.38	35.59
MnO	0.20	0.22	0.16	0.24	0.23	0.22
MgO	3.05	3.54	3.23	3.02	2.77	3.35
CaO	0.00	0.00	0.00	0.00	0.00	0.00
Sn	0.01	0.02	0.00	0.00	0.00	0.00
W	0.07	0.06	0.17	0.02	0.03	0.02
Zn	0.00	0.00	0.02	0.00	0.10	0.05
Na2O	0.11	0.02	0.08	0.11	0.09	0.08
K2O	9.22	9.14	9.30	9.24	9.23	9.15
F	0.58	0.82	0.52	0.81	0.76	0.89
Cl	1.18	1.04	1.25	1.22	1.14	0.74
Anhydrous Total	97.85	97.51	98.57	97.56	97.66	98.13

Numbers of ions on the basis of 24 (O,OH,F,Cl)

	7	8	9	10	11	12
Si	5.83	5.72	5.86	5.69	5.69	5.85
Al	1.93	2.12	1.91	2.07	2.07	2.02
Ti	0.24	0.16	0.23	0.24	0.24	0.13
	8.00	8.00	8.00	8.00	8.00	8.00
Al	0.30	0.15	0.39	0.17	0.20	0.27
Fe+2	4.87	4.80	4.95	4.88	4.90	4.95
Mn	0.03	0.03	0.02	0.03	0.03	0.03
Mg	0.75	0.87	0.81	0.74	0.68	0.83
Sn	0.00	0.00	0.00	0.00	0.00	0.00
W	0.50	0.00	0.01	0.00	0.00	0.00
Zn	0.00	0.00	0.00	0.00	0.01	0.01
	5.95	5.85	6.18	5.82	5.82	6.09
Na	0.04	0.00	0.03	0.04	0.03	0.03
K	1.95	1.92	1.99	1.95	1.95	1.94
	1.99	1.92	2.02	1.99	1.98	1.97
F	0.30	0.43	0.28	0.42	0.40	0.47
Cl	0.33	0.29	0.35	0.34	0.32	0.21
OH	2.49	2.88	1.88	2.73	2.81	2.42
	3.12	3.60	2.51	3.49	3.53	3.10

Analyses refer to traverse A and are similar to those of traverses B and C. Analyses were performed with a Jeol JXA-5A microprobe with computer control (Department of Geology, University of Melbourne); beam current 0.1 μA, accelerating potential 20 kV. Standards used were: Si, wollastonite and quartz; Al, corundum; Ti, rutile; Fe, Fe metal; K, potassium tantalite; Mn, Mn metal; Na, jadeite; Cl, fluorite; Sn, Sn metal; W, W metal; Zn, Zn metal; Cu, Cu metal; Mg, periclase; Ca, wollastonite. Data reduced with the program of Mason *et al.* (1969), as modified by A. K. Ferguson.

The X-ray-diffraction pattern of a bulk separate from a nearly pure sector-zoned annite rock having very similar compositions to that shown in Table 1 gave the following *d* values, listed in order of decreasing intensities: 3.3634 (10), 9.118 (5), 3.1865 (4), 2.5 (3), 2.6595 (2) and 5.0675 (2).

The micas have structural formulae that are usual for high Fe^{2+} micas (Table 1). All the tetrahedral sites are filled (to 8.00) by Si, Ti and Al, whereas octahedral sites are filled mainly by Fe, to a much lesser extent by Mn and Al, and rarely by Zn, to a total of nearly 6.00.

The total of interlayer cations (Na + K) generally is low ($\simeq$ 1.9); this is lower than that usually reported (*e.g.*, Deer *et al.* 1966). Whether some Ba, Rb, Cs or, possibly, oxonium (H_3O^+) is present is not known but remains a possibility. However, no peaks for Ba, Rb or Cs were noted on the Edax scan-system spectra associated with the microprobe. No Ca is present in any of the micas analyzed. The anionic population, namely involving Cl and F, is extremely variable and unsystematic, as can be seen in the representative analytical data in Table 1. Values at the same level of a sector (analyses 12–16, Fig. 4 or Table 1) are nearly constant, suggesting that the variable concentrations shown are real and not due to analytical error.

Analyses for Li were not undertaken, but it is unlikely that sufficient Li could be present to partly fill the octahedral position because (1) the totals (Table 1) are those to be expected from a normal mica analysis ($\simeq$ 96 wt. % anhydrous total), (2) the micas do not exhibit any color characteristics generally associated with Li-micas and (3) no high Li values are known in the skarns or surrounding granites.

DISCUSSION

Color and composition zoning have been reported by Wagner (1909), Métais *et al.* (1962), Rimšaite & Lachance (1966), Rimšaite (1969) and Velde & Yoder (1977) for micas from plutonic and volcanic rocks, but not from metamorphic rocks. Various substitution schemes have been suggested in the above studies, but generally Fe and Mg show opposite trends, modified by variations in Al and Ti (*e.g.*, Rimšaite 1969). For the present study, a similar scheme is inferred, namely, $Ti^{4+} + Mg^{2+} \rightleftarrows 2Fe^{3+}$ or $Ti^{3+} + Mg^{2+} \rightleftarrows Fe^{2+} + Fe^{3+}$. The distribution of both Ti and Mg from the centre of the crystals to the edge of each sector can best be explained by a depletion or Rayleigh fractionation model (Hollister 1966). In the skarn, annite forms as a reaction product in both primary type (a) and (b) skarns and secondary type (c) skarn. Both ilmenite and sphene are volumetrically less abundant and more erratically distributed than magnetite. Mg–Fe vesuvianite or Mg–Fe am-

FIG. 4. The variation of TiO₂, MgO and FeO *versus* distance along traverses A, B and C. The numbers refer to individual spot-analyses. Complete results of analyses for 1 to 16 are given in Table 1. Note that MgO and TiO₂ vary inversely to trends of Fe in most cases. In traverse C, however, near sample 44, TiO₂ and MgO show inverse relationships. Traverse 4a is of a grain located less than 2 mm from the grain on which traverses 4B and 4C were obtained.

phibole. The activity of Fe in the entire system would be fixed at a fairly constant value by the occurrence and even distribution of these Fe minerals, but the activity of Ti would be less well defined, especially if diffusion rates are limited, permeability is limited and Ti phases are depleted by reaction.

Mozaic equilibrium was present in the area of a single thin section during mica crystallization, as is indicated by the differences in composition of comparable grain edges (compare the edges of the two grains in the traverses in Fig. 4). The Ti content of the growing mica reflects the availability and the diffusion rates within a restricted area where an approach to equilibrium presumably occurred. This explains both the progressive outward decrease of Ti in the crystals and the abrupt decrease of Ti two thirds to three quarters of the dis-

tance from their centres. This decrease is interpreted as having occurred when ilmenite or sphene was consumed, thus leaving the Ti content in additional annite growth partly dependent on the Ti contents of reacting Fe–amphibole ($\simeq$ 0.30 wt. % TiO₂), magnetite ($\simeq$0.80%), vesuvianite ($\simeq$3.0–4.0%) or garnet ($\simeq$0.3–5.5%).

The Mg for the crystallizing mica is produced by reacting Mg–Fe vesuvianite, its alteration product, Mg–Fe amphibole or Mg-bearing calcite, which texturally can be seen to have reacted to form Ca-bearing, low-Mg siderite. The availability of Mg and Ti to the growing mica is thus dependent upon the reaction kinetics and diffusion rates of at least two reactions. This may explain some of the irregular (nonlinear) variations shown in Figure 4.

The origin of sector zoning has been discussed by a number of authors, including Hollister & Bence (1967), Hollister (1970), Nakamura (1973), Strong (1970), Hollister & Hargraves (1971) and Wass (1973). For sector zoning in igneous pyroxenes, Wass (1973) has suggested that growth of the crystal is faster than the ionic diffusion in the rapidly cooling melt. This creates inequalities: one face grows faster than another. The model is unlikely to be applicable to the present study because the crystallization of annite probably took place over a long period of time, during the cooling of the pluton, and fluid compositions should have equilibrated over the small distances involved within the area of a thin section (<1 mm). Hollister & Bence (1967) and Hollister (1970) similarly suggested that in sector zoning in staurolite from the Kwoiek (B.C.) contact-metamorphic zone, disequilibrium existed between the bulk composition of each sector and the matrix, possibly because of rapid growth. This is suggested to be the reason that sector zoning has not been reported in regionally metamorphosed terranes. In the above example, the explanation of the exact mechanism to produce the zoning is somewhat obscure, but is thought by Hollister to be related to the different lattice configurations at each face of staurolite.

Sector zoning is best accounted for by the protosite concept of Nakamura (1973). A protosite is defined as a partly formed structural site on a crystal surface. When a crystal grows, protosite is defined as a partly formed structural in equilibrium with the fluid phase. The geometry of each protosite on each growth surface differs, in variable degrees, from that of the corresponding true structural site. Where this possible deviation is greatest, the composition of that site can deviate most prominently from the equilibrium composition. Relating this theory to the present study, the (100) and (001) surfaces of the annites have the most flexible protosites; hence, with continuing growth, hopper-like (100) and (010) sectors form that accommodate more (Mg + Ti) than the (001) sites.

The following possibilities for the origin of sector zoning of minerals are suggested on the basis of this study and those cited from the literature: (1) Sector zoning is not necessarily related to extremely rapid cooling, such as found in volcanic environments. This is evident from the occurrences of sector-zoned staurolite reported by Hollister & Bence (1967) and from the annite of this study. Although the zoned annite reported here is unusual, it is not unique. Samples from the Little Hot Spot deposit (part of the Iron Mountain Fe–F–W–Sn–Be skarns, New Mexico), supplied to the author by Dr. D. Burt, show similar features (Fig. 4e, f). These occurrences do imply that extremely rapid cooling is not a necessary prerequisite for the formation or retention of sector or hourglass zoning in minerals, as has been implied by Wass (1973) and Velde & Yoder (1977). (2) High Ti contents may be needed for sector zoning. Chemically, sector zoning has been best documented in clinopyroxene, staurolite and, here, in annite. On a percentage basis, in each case, the element that shows the greatest difference between the (001) sectors and adjacent (010) and (100) sectors is titanium [note, for example, Fig. 4 in Hollister & Bence (1967), Table 11 in Wass (1973) and Table 1, this study]. (3) Sector zoning is perhaps related to the oxidation state of titanium. The darker brown colors of the annite in the high Ti areas are probably due to the presence of Ti in the Ti^{3+} state rather than Ti^{4+}, as pointed out by others (Wones 1967). If both Ti^{3+} and Ti^{4+} were present in the fluid phase associated with a growing crystal, the Ti^{3+} might be preferred on the (100) and (010) faces, whereas Ti^{4+} (and, perhaps, to a lesser extent, Ti^{3+}) is preferred on the (001) face. (4) Sector zoning in micas may be possible only in extreme end-member compositions. In micas, sector zoning has been reported from a Mg-rich example (Velde & Yoder 1977) and the Fe-rich type reported here. Obviously, more examples are needed. (5) Sector zoning in micas may be related to a specific geochemical environment. The example described by Velde & Yoder (1977) as well as more "normal" zoning reported by Rimšaite (1969) and others indicate that zoning in micas occurs in silica-poor environments and often in carbonate-rich environments. (6) Sector zoning in metamorphic minerals may be possible only in contact-metamorphic environments. It is noteworthy that all three examples of sector zoning in metamorphic minerals (staurolite in the Kwoiek area and the two examples cited here) are from examples of contact metamorphism and not from environments of regional metamorphism.

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