METASOMATICALLY ALTERED AMPHIBOLITE INCLUSIONS IN ZONED GRANITIC–TONALITIC PEGMATITE NEAR CHICOUTIMI, QUEBEC

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ABSTRACT

Amphibolite inclusions in felsic pegmatite in the central Grenville Province near Chicoutimi, Quebec, are bordered by an alteration rim comprising an inner reaction-zone of biotite, andesine, quartz, and accessory apatite, and an outer zone of coarse biotite, quartz and albite, with accessory apatite and zircon. The interface between the inner and outer reaction-zones is interpreted as a compositional boundary that coincides with the original margin of the amphibolite inclusions. In this model, the outer reaction-zone acted as a sink for excess Mg and Fe liberated from the inner reaction-zone; other elements were provided by the metasomatic fluid. On the scale of both reaction zones, metasomatism involved the net introduction of K, P and volatiles, and the removal of Ca and Mn.

Keywords: Grenville Province, metasomatism, amphibolite, pegmatite, isocon diagram.

SOMMAIRE

Des enclaves d'amphibolite prélevées d'une pegmatite felsique de la région de Chicoutimi (Québec), dans la partie centrale de la province du Grenville, sont entourées d'une zone de réaction. L'assemblage biotite + andésine + quartz + apatite accessoire caractérise la partie interne de la zone, tandis que sa partie externe contient l'assemblage biotite (en gros cristaux) + quartz + albite, avec apatite et zircon accessoires. L'interface entre les deux parties de la zone de réaction serait l'expression de la discontinuité originelle en composition entre pegmatite et amphibolite. Selon ce schéma, la partie externe aurait piégé le surplus de Mg et de Fe libérés de la partie interne. Les autres éléments constitutifs auraient été apportés par le fluide métasomatisant. À l'échelle des deux parties de la zone de réaction, il y a eu apport de K, P et des éléments volatils, et élimination de Ca et de Mn.

(Traduit par la Rédaction)

Mots-clés: métasomatose, amphibolite, pegmatite, diagramme d'isoconcentrations, Chicoutimi, province du Grenville.

INTRODUCTION

Wallrock adjacent to granitoid rocks and associated pegmatite commonly shows signs of metasomatic alteration. Such alteration, typically involving the introduction or remobilization of volatiles, alkalis and Ca, and, in some instances, of relatively immobile elements such as Al (Kanaris-Sotirious & Angus 1979, Williams 1987), may be manifested by changes in the bulk and modal composition of the wallrock. This study describes features resulting from the metasomatism of amphibolite inclusions in felsic pegmatite from the central Grenville Province of Quebec. The amphibolite inclusions are of interest because the metasomatic alteration is zonal, reflecting not only net compositional changes associated with metasomatism (principally a gain of K, P and volatiles, and a loss of Ca and Mn), but also the mobility of other elements (Mg, Fe and possibly Na) between the zones.

The pegmatite considered here intruded mafic gneiss (amphibolite) associated with pelitic and granitic gneiss of the central Grenville Province near Chicoutimi, Quebec. The regional geology of the Chicoutimi area was described by Roy *et al.* (1986), who related the pegmatite veins to diverse Grenvillian granitoid bodies that cut the gneiss complex.

The pegmatite outcrops along the north shore of the Saguenay River near St. Fulgence, approximately 15 km east of Chicoutimi. The pegmatite is 2 m wide, with a rose-colored granitic core and green-grey tonalitic borders. The pegmatite intruded medium-grained (~ 0.3 mm) amphibolite. Inclusions of amphibolite ranging from 5 to 50 cm in diameter occur within the tonalitic margin of the dyke.

LITHOLOGY

The amphibolite away from the pegmatite is medium grained and homogeneous. It contains green hornblende, calcic andesine, quartz and ilmenite ($\pm$ magnetite), with accessory biotite and traces of apatite. Some plagioclase near the pegmatite shows reverse zoning (core: An_{47}; rim: An_{72}). The amphibolite protolith is preserved in the core of all but the smallest inclusions. The smallest inclusions have been completely altered.

Amphibolite in contact with pegmatite has developed an inner reaction-zone of medium-grained andesine (An_{41}), biotite, quartz, ilmenite and accessory apatite, and an outer zone of coarse-grained biotite, quartz, and accessory ilmenite, albite (An_0), zircon and apatite. The inner zone is 1 to 3 cm wide. The outer zone is somewhat irregular, being up to 8 cm wide, with cuspate wisps of biotite extending into the pegmatite (Fig. 1). Modes and compositions

FIG. 1. Recrystallized amphibolite enclaves in tonalitic pegmatite, central Grenville Province, St. Fulgence, Quebec. Note the coarse, biotite-rich outer reaction-zone (dark grey) and the finer-grained, mesocratic inner reaction-zone (light grey) that envelops unaltered amphibolite (medium grey). Pen is 14 cm long.

of minerals of the amphibolite and the alteration rim are listed in Tables 1 and 2, respectively.

The amphibole in the amphibolite is tschermakitic hornblende (Table 2). The hornblende coexists with biotite of higher X_{Mg} [$= Mg/(Mg + Fe_t)$] ($X_{Mg} = 0.53$) than biotite in both reaction zones ($X_{Mg} = 0.45$). Compositionally similar biotite ($X_{Mg} = 0.41$) occurs in the tonalitic pegmatite. Plagioclase in the pegmatite is oligoclase (An_{24}).

GEOCHEMISTRY

Mineral analyses were performed on a JEOL JXA–50A wavelength-dispersion electron-probe microanalyzer at the Department of Earth Sciences, Memorial University of Newfoundland, using a beam current of 22 nA and an accelerating voltage of 15 kV. Counts were made for 30 s or to a maximum of 60,000. Bence–Albee corrections were employed in data reduction. Results of mineral analyses are presented in Table 2. Amphibole composi-

tions were recalculated to 23 oxygen atoms, with all cations except Ca, Na and K being normalized to 13, after which Fe^{3+} was estimated to achieve charge balance.

The amphibolite, reaction zones, and pegmatite were analyzed for major elements by atomic absorption spectrometry using a Perkin-Elmer digitized

TABLE 1.	MODAL COMPOSITION OF AMPHIBOLITE AND THE ALTERATION RIM (VOLUME %)		
	1	2	3
Bt	1	30	81
Qtz	9	10	18
Pl	31	56	tr
Hbl	53		
Ilm (±Mag)	6	4	1
Ap	tr	tr	tr
Zrn			tr

1 amphibolite within 2 cm of alteration rim; 2 inner reaction zone; 3 outer reaction zone. Point count total= 1000 per domain; values are rounded to nearest 1% (tr trace).

TABLE 2. CHEMICAL COMPOSITION OF SELECTED MINERALS FROM THE AMPHIBOLITE, ALTERATION RIM AND PEGMATITE

	Amphibolite					Inner reaction zone			Outer reaction zone			Pegmatite	
	Bt	Hbl*	Pl	Ilm	Mag	Bt	Pl	Ilm	Bt	Pl	Ilm	Bt	Pl
SiO2	35.94	41.60	56.10			37.50	57.44		36.41	68.76		38.01	63.09
TiO2	3.27	.95		44.84	.11	3.49		46.32	3.73		48.16	3.46	
Al2O3	16.64	12.25	28.39		.40	16.01	26.19		16.17	19.10		16.03	24.95
FeO	20.00	18.62	.07	50.30	94.78	21.33	.10	47.92	20.79	.10	49.43	22.86	.06
MnO	.11	.41		1.52	.04	.17		1.79	.24		2.88	.29	
MgO	12.41	9.10		.19		10.09		.16	9.36		.09	8.98	
CaO		11.03	9.80				8.46			.13			5.12
Na2O	.07	1.29	5.96			.10	6.62		.22	11.79		.05	8.77
K2O	9.52	.97	.14			9.05	.14		9.05	.03		8.73	.36
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	97.9	96.2	100.5	96.9	95.3	97.7	98.9	96.2	95.9	99.9	100.6	98.4	99.9
oxygen basis	22	23	8	3	4	22	8	3	22	8	3	22	8
Si	5.354	6.402	2.510			5.595	2.598		5.541	3.006		5.653	2.744
^{IV}Al	2.646	1.598	1.497		.017	2.405	1.397		2.459	.984		2.347	1.264
^{VI}Al	.276	.624				.411			.442			.463	
Ti	.366	.110		.909	.002	.392		.936	.427		.932	.387	
Fe**	2.492	2.396	.003	1.135	2.962	2.662	.004	1.077	2.646	.004	1.064	2.843	.003
Mn	.014	.053		.034	.001	.021		.040	.031		.063	.037	
Mg	2.755	2.087		.007		2.244		.006	2.123		.003	1.990	
Ca		1.819	.470				.410			.006			.237
Na	.020	.385	.517			.029	.581		.065	1.000		.014	.736
K	1.809	.190	.008			1.723	.008		1.757	.002		1.656	.018

*   hornblende formula: $(Na^A_{.16} K_{.19}) (Ca_{1.78} Na^{M4}_{.22}) (Mg_{2.04} Fe^{2+}_{1.41} {}^{VI}Al_{.45} Fe^{3+}_{.94}) (Si_{6.27} {}^{IV}Al_{1.73}) O_{22}(OH)_2$

** all Fe as Fe^{2+}

TABLE 3.   MAJOR ELEMENT GEOCHEMISTRY OF THE AMPHIBOLITE, ALTERATION RIM, AND PEGMATITE

	Amphibolite protolith						Reaction zones		Pegmatite		
	1.	2.	3.	4.	s*	5.	Inner	Outer	6.	7.	8.
SiO2	50.7	48.9	50.4	50.0	.79	50.3	55.0	41.8	65.3	71.2	84.8
TiO2	2.93	2.97	2.80	2.90	.07	2.84	3.49	3.72	.06	tr	tr
Al2O3	12.5	12.4	12.3	12.4	.08	12.3	14.1	13.1	18.5	14.7	8.00
Fe2O3	17.80	17.88	17.42	17.70	.20	17.26	13.49	22.47	1.08	.29	.34
MnO	.33	.33	.29	.32	.02	.31	.22	.21	.03	tr	.01
MgO	4.92	4.95	4.85	4.91	.04	4.61	3.55	8.83	.31	.09	.10
CaO	7.95	8.04	8.23	8.07	.12	7.29	3.52	.87	5.79	.27	.25
Na2O	1.39	1.33	1.26	1.33	.05	1.47	2.05	.19	5.24	2.22	1.30
K2O	.93	.94	.84	.90	.04	1.26	3.02	7.20	.99	9.48	4.70
P2O5	.33	.32	.32	.32	.00	.34	.41	.43	1.18	.04	.02
LOI	.49	.34	.44	.42	.06	.59	.45	1.04	.54	.40	.37
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	100.3	98.4	99.2	99.3		98.6	99.3	99.9	99.0	98.7	100.0
n =	1	1	1	3		1	2	2	1	1	1
density (g/cm³)	3.07	3.07	3.07	3.07		3.07	2.82	2.98	not determined		

1 amphibolite 2-4 cm from alteration rim; 2 amphibolite 4-6 cm from alteration rim; 3 amphibolite 6-8 cm from alteration rim; 4 mean amphibolite composition 2-8 cm (analyses 1 to 3) from alteration rim (*s = one standard deviation); 5 amphibolite composition within 2 cm of alteration rim; 6 tonalitic pegmatite; 7 granitic pegmatite; 8 quartz-rich granitic pegmatite. All Fe as Fe2O3. LOI-loss on ignition.   *s = one standard deviation   n = number of analyses

spectrometer. P_2O_5 was determined by colorimetry. Estimated analytical errors based on replicate analyses of standards are less than 5% (relative).

Compositions of four slabs of amphibolite, in 2-cm-wide segments up to 8 cm from the adjacent reaction-zone, are listed (Table 3) along with data for the reaction zones and the pegmatite. Amphibolite within 2 cm of the alteration rim shows some compositional differences [e.g., higher K and loss on ignition (LOI)], which are attributed to metasomatism (see below). Consequently, the protolith is considered to be the least-altered amphibolite core (i.e., > 2 cm from the alteration rim) of the inclusions. The bulk-rock data reflect the distinctive mineral assemblages comprising the reaction zones.

The analytical data are used to evaluate two aspects of the metasomatic history of the amphibolite: (1) mass, volume and compositional changes

accompanying metasomatism of the amphibolite; and (2) the significance of the interface between the inner and outer reaction-zones. Finally, the mineralogy of the altered amphibolite is compared with experimental data on the metasomatic breakdown of hornblende in mafic rocks.

CHANGES IN MASS, VOLUME AND MAJOR-ELEMENT CONCENTRATIONS

Mass, volume and compositional changes accompanying metasomatism of the amphibolite have been evaluated by the isocon method of Grant (1986). By this approach, composition–volume relations derived by Gresens (1967) are expressed as linear equations that compare the concentration of different components ("C_i") in the least-altered amphibolite (C_i^o) and its altered equivalent (C_i^a). The isocon, a line of equal elemental concentration, has a slope equal to the ratio of the mass of the component in the protolith to that in the altered rock. Compared with the least-altered amphibolite, concentration changes of various elements may be expressed relative to model isocons (based on constant mass, volume or a chosen element) or to a best-fit isocon defined by a linear array of elements in C_i^o–C_i^a space.

Reconstruction of the metasomatic history of the amphibolite requires an evaluation of the process(es) of metasomatic exchange near the amphibolite – pegmatitic melt interface, and the identification of relatively immobile components against which more mobile elements may be compared.

The available data provide little information on the mechanism of element transfer. For the inner zone, it is assumed that metasomatism involved the isobaric diffusion of migrant components through an intergranular metasomatic fluid in equilibrium with the nearby mineral assemblage. Relaxation of thermal gradients near the inclusion – pegmatitic melt interface would be enhanced by convective or advective heat-flow through the fluid, so that metasomatism would tend toward isothermal diffusion.

Evaluation of the relative mobility of different elements during alteration also is problematic. Although Al and Ti are commonly considered to be relatively immobile during metasomatism, contrasting Al/Ti values between the least-altered amphibolite (Al/Ti = 3.76) and the inner and outer reaction-zones (Al/Ti = 3.56 and 3.10, respectively) suggest that one or both of these components was (were) mobile during alteration (cf. Lesher et al. 1986). The available data do not indicate which element was less mobile. For illustrative purposes, Al will be used as a reference component. The mobility of other elements in both reaction zones is therefore described by assuming constant aluminum; the results based on the assumptions of constant mass and constant volume are offered for comparison. In addition, a best-fit isocon for the inner reaction-zone is constrained by data for Al, P, Ti and Si. This best-fit isocon represents a suitable and pragmatic compromise between the assumptions of constant Ti and constant Al.

Concentration changes between the least-altered amphibolite and the reaction zones are listed for model isocons in Table 4, and depicted graphically in Figure 2. These data tacitly assume that both reaction zones were directly derived from the amphibolite protolith (i.e., that the outermost margin of the reaction rim coincides with the original border of the amphibolite inclusions). This assumption is evaluated below, where it is proposed that the boundary between the inner and outer reaction-zones coincided

TABLE 4.   CONCENTRATION CHANGES ($\Delta C_i/C_i^o$) IN ALTERED AMPHIBOLITE, ASSUMING CONSTANT ALUMINA, MASS AND VOLUME

Constant:	Inner reaction zone			Outer reaction zone			Weighted mean alteration rim		
	Al_2O_3	mass	vol.	Al_2O_3	mass	vol.	Al_2O_3	mass	vol.
CaO	-0.62	-0.56	-0.60	-0.90	-0.90	-0.90	-0.71	-0.67	-0.70
MnO	-0.40	-0.31	-0.37	-0.38	-0.34	-0.36	-0.38	-0.31	-0.36
MgO	-0.36	-0.28	-0.34	0.70	0.80	0.75	-0.03	-0.08	0.01
Fe_2O_3	-0.33	-0.24	-0.30	0.20	0.27	0.23	-0.16	-0.07	-0.13
SiO_2	-0.03	0.10	0.01	-0.21	-0.16	-0.19	-0.09	0.01	-0.05
Al_2O_3	-	0.14	0.05	-	0.06	0.02	-	0.11	0.04
TiO_2	0.06	0.20	0.11	0.21	0.28	0.25	0.11	0.23	0.15
P_2O_5	0.13	0.28	0.18	0.27	0.34	0.30	0.18	0.31	0.23
Na_2O	0.36	0.54	0.42	-0.86	-0.86	-0.86	-0.03	0.08	0.01
K_2O	1.95	2.36	2.08	6.57	7.00	6.77	3.42	3.91	3.59
LOI	-0.06	0.07	-0.02	1.34	1.48	1.40	0.39	0.55	0.45

Data compare compositional differences between the least altered amphibolite and the inner, outer and mean reaction zones.
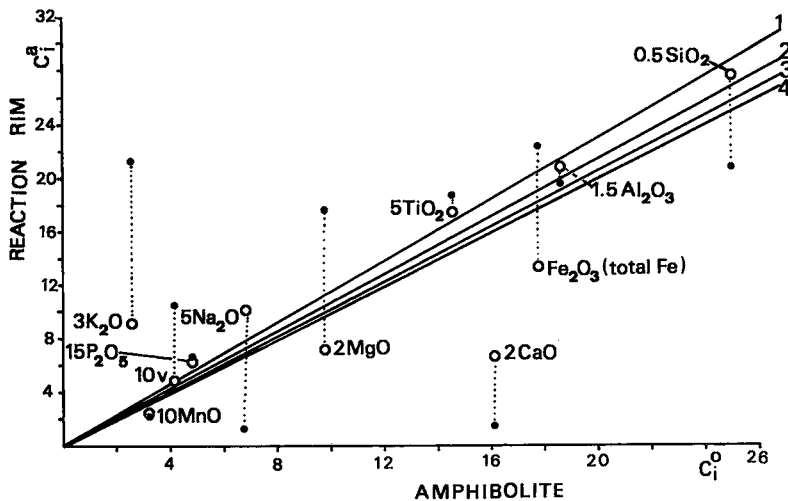
FIG. 2. Isocon diagram that compares the bulk composition of least-altered amphibolite (anal. 4, Table 3) and the reaction rim (open circles: inner reaction-zone; closed circles: outer reaction-zone). Components are scaled as indicated. (1) Best-fir isocon (inner zone); (2) constant-volume isocon (inner zone); (3) constant-volume isocon (outer zone); (4) constant-mass isocon. Volatiles (LOI) denoted by "v".

with the amphibolite – pegmatitic melt interface rather than with a reaction boundary. The composition of the least-altered amphibolite nonetheless provides a suitable reference for a determination of concentration changes accompanying metasomatism, and for an evaluation of the extent to which mobile species liberated during metasomatism contributed to the formation of the reaction zones.

For the inner reaction-zone, data for Al, P, Ti and Si plot as a quasilinear array defining a best-fit isocon with a slope of about 1.15, corresponding to a 13% mass loss. The ratio of densities of the amphibolite and the inner reaction zone is 1.09, implying a volume loss of about 5%. Mobile species plot away from the best-fit isocon. It is apparent from both the best-fit and the three model isocons that the formation of the inner reaction-zone involved a 24–40% decrease in Mn, Mg and Fe, a 55–60% decrease in Ca, a 35–55% increase in Na, and an increase of about 200% in K. The volatile (LOI) datum lies on or close to the best-fit isocon and the three model isocons, which implies that recrystallization within the inner zone was not accompanied by a significant net change in the concentration of volatiles.

As noted above, the interface between the inner and outer zones may coincide with the original boundary of the inclusion. This inference implies that the outer zone grew outward into the pegmatitic melt. The extent to which ions released from the inner zone were consumed by growth of the outer zone may be evaluated by comparing the composition of outer zone to the least-altered amphibolite. However, since it is apparent that the outer zone did not directly replace the amphibolite (see below), the following discussion provides only a semiquantitative portrayal of the behavior of different elements during metasomatism.

Compared with geochemical trends defined for the inner zone, the outer reaction-zone shows greater loss of Ca, and greater gain of K. Similar behavior of Mn, Ti, and P, and reversals in the behavior of Na, Mg and Fe, also are apparent. The data do not readily define a best-fit isocon. However, similar geochemical trends are apparent regardless of which model isocon is considered (Table 4). Compared with the least-altered amphibolite, Ca and Na decreased by 85–90%, Mn decreased by 35%, Ti, P and Fe increased by 20–30%, Mg increased by 70–80%, and K increased by 650–700%. LOI values are up to 140% higher, suggesting a substantial influx of volatiles.

Patterns of concentration changes similar to those outlined above emerge if Ti is assumed to be constant for both reaction zones, although, with this assumption, the outer reaction-zone shows no appreciable change in Fe, and a diminished apparent gain of Mg, compared with the other model isocons.

Although most components were confined to the alteration rim, the available data point to significant interzone elemental migration. For example, con-
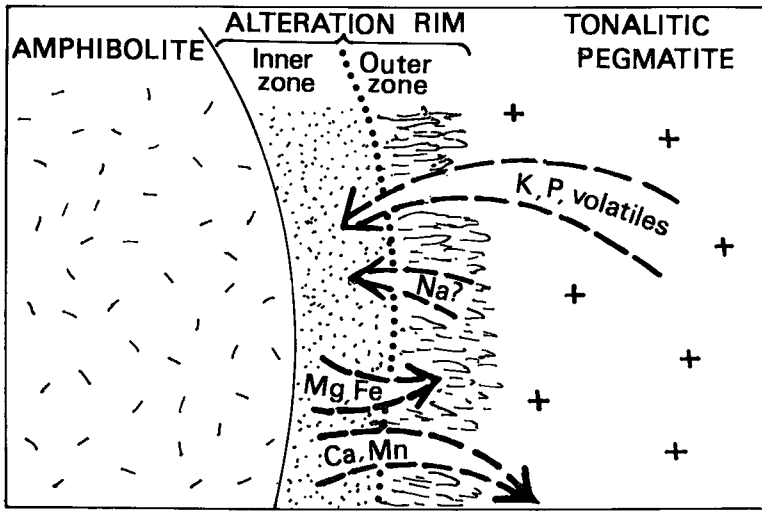
FIG. 3. Interpretative sketch depicting metasomatic exchange near the amphibolite
– pegmatitic melt interface. Dotted line represents the original margin of the
amphibolite inclusion.

trasts in the behavior of Mg and Fe (Fig. 2) between
the reaction zones identify the inner zone as a likely
source of these elements during the development of
the outer zone. Similarly, the isocon diagrams do not
indicate an external source for Na, which is concen-
trated in the inner zone, and depleted in the outer
zone. Sodium (+ substantial Ca) liberated by the
breakdown of relatively calcic plagioclase in or near
the outer zone (i.e., oligoclase in the pegmatite?) may
have contributed to the growth of intermediate ande-
sine in the inner zone. Thus, alteration of the
amphibolite involved elemental mobility between the
reaction zones as well as the net introduction of K,
P and volatiles, and the loss of Ca and Mn.

The net effect of metasomatism may be evaluated
by comparing the amphibolite and the combined
composition of both reaction zones, weighted to
reflect their relative volumes as estimated from the
two-dimensional outcrop surface (cf. Fig. 1). Using
a weighting ratio of 1.5:1 (inner zone:outer zone),
and assuming constant aluminum, it is apparent that
metasomatism principally involved the introduction
of K (+340%), P (+18%) and volatiles (40%
increase in LOI), and the removal of Ca (–70%) and
Mn (–38%), with a mass loss of 10% (Table 4).

It is unlikely that the Ca liberated by hornblende
and plagioclase breakdown was taken up as calcic
rims on plagioclase in the amphibolite, since the
bulk-rock data indicate that amphibolite within 2 cm
of the reaction rim contains less Ca than the central
amphibolite core of the inclusion. Therefore calcium
was lost from the system.

SIGNIFICANCE OF THE BOUNDARY BETWEEN INNER
AND OUTER REACTION-ZONES

The boundary between the inner and outer
reaction-zones may be interpreted either as a reac-
tion boundary or as the original amphibolite – peg-
matitic melt interface. Distinction between these pos-
sibilities focuses on the mineralogical similarity of
both reaction zones. As pointed out by C.T. Foster
(pers. comm. 1988), this similarity ensures that
Gibbs–Duhem constraints are the same in each
domain, which thereby places constraints on element
mobility within the alteration rim.

Although their modes differ (Table 1), both reac-
tion zones contain biotite, plagioclase, quartz,
ilmenite and apatite. Consequently, the Gibbs–
Duhem constraints provided by these minerals are
the same in both zones. Assuming local equilibrium,
the boundary between the zones may thus be inter-
preted as a compositional boundary that acted as an
inert marker (Brady 1975, Foster 1981). In this
model, the inner reaction-zone is interpreted to have
formed directly from the amphibolite, whereas the
outer zone grew outward from the amphibolite – peg-
matitic melt interface by consuming mobile compo-
nents from both the inner reaction-zone and the peg-
matitic melt.

The presence of quartz and ilmenite in the
amphibolite and both reaction zones merits some dis-
cussion. The Gibbs–Duhem constraints on quartz fix
the chemical potential of silica in all domains in the
rock. Consequently, the silica-fixed reference frame

is an inert marker, and a constant-silica isocon may therefore be used to evaluate the behavior of Fe and Ti in the inner and outer reaction-zones.

Owing to the presence of ilmenite in all domains in the rock, the Gibbs–Duhem constraint on ilmenite requires that the transport of Fe and Ti be in opposite directions in an inert-marker reference frame. With reference to the constant-silica isocon, this requirement is satisfied only for the inner reaction-zone. Therefore, it provides supplementary evidence that the outer zone nucleated on the amphibolite – pegmatitic melt interface.

COMPARISON WITH EXPERIMENTAL DATA

The mineralogy of the alteration rim differs from that anticipated by some experimental studies of hornblende breakdown. For example, hornblende in the presence of K_2SO_4–H_2SO_4-bearing aqueous fluids yields biotite, anhydrite and quartz, and primary igneous biotite re-equilibrates to a lower Ti and more magnesian composition (Brimhall *et al.* 1985). Relatively Fe- and Ti-depleted biotite also is characteristic of hydrothermally altered rocks derived from granitic rocks in porphyry copper deposits (Moore & Czamanske 1973, Brimhall *et al.* 1985).

This finding contrasts with the relatively ferroan composition of biotite in the alteration rim of the St. Fulgence amphibolite inclusions. Since bulk Fe/Mg was nearly constant during metasomatism (at least on the scale of the combined reaction-zones: Table 4), the relatively ferroan composition of the secondary biotite is attributed to reduced $f(O_2)$ [or reduced $f(S_2)$] in the metasomatic fluid.

CONCLUSIONS

Alteration zones that mantle amphibolite inclusions in tonalitic pegmatite at St. Fulgence were developed by the breakdown of hornblende and calcic plagioclase to biotite, quartz, and relatively sodic plagioclase. Compared with the least-altered amphibolite, alteration in the combined inner and outer reaction-zones involved a net increase in K, P and volatiles, and a net loss of Ca and Mn.

The mineralogical similarity of the inner and outer reaction-zones suggests that the interface between them coincides with the original margin of the amphibolite inclusion. The inner zone developed by alkali metasomatism of the amphibolite protolith. The outer zone acted as a sink for Mg and Fe liberated from the inner zone; other components were apparently provided by the metasomatic fluid.

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