

ZONED PLAGIOCLASE AND THE BREAKDOWN OF HORNBLLENDE IN PYROXENE AMPHIBOLITES

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ABSTRACT

Two distinct varieties of pyroxene amphibolite from a small area within the hornblende-granulite facies terrane of the northwest Adirondacks, New York, contain hornblende + orthopyroxene + clinopyroxene + ilmenite + plagioclase. One variety (type A) has low bulk Fe and Ti, and contains magnetite in addition, whereas type B, with high Fe and Ti, carries garnet instead of magnetite. Compositions of ferromagnesian phases reflect rock chemistry, and exhibit an approach toward equilibrium behavior. Zoned plagioclase is very common in the amphibolites; it shows reverse zoning in type-A amphibolites, whereas in type B, it is normally zoned. The rim of plagioclase grains from type-A amphibolites approaches An_{70-80} , nearly equal to the Ca/Na ratio of the coexisting hornblende, whereas in type-B amphibolites, the rim is much more sodic (An_{35-45}). The textural and chemical evidence suggests that the plagioclase composition and type of zonation were controlled by local hornblende-breakdown reactions, which were themselves functions of bulk-rock chemistry.

Keywords: zoned plagioclase, amphibolite, Adirondack, granulite, hornblende breakdown.

SOMMAIRE

Deux variétés d'amphibolite à pyroxène provenant d'une région restreinte du socle métamorphique (facies granulite à hornblende) située au nord-ouest des montagnes Adirondack, dans l'état de New-York, contiennent l'assemblage hornblende + orthopyroxène + clinopyroxène + ilménite + plagioclase. La première variété (type A) a une faible teneur en Fe (total) et Ti et elle contient de la magnétite; l'autre (type B) a une forte teneur en Fe et Ti et elle contient du grenat au lieu de la magnétite. La composition des phases ferromagnésiennes reflète le chimisme de la roche et montre une tendance vers l'équilibre chimique. Les cristaux zonés de plagioclase sont très communs dans les amphibolites: dans le type A la zonation est inversée; elle est normale dans le type B. La bordure des grains de plagioclase des amphibolites de type A a une composition An_{70-80} , presque égale au rapport Ca/Na de la hornblende coexistante, tandis que dans les amphibolites de type B, cette bordure est beaucoup plus sodique (An_{35-45}). Les indications texturales et chimiques font penser que la composition du plagioclase et le type de zonation sont régis par les réactions de dissociation locale de la hornblende, réactions qui sont fonctions du chimisme de la roche.

(Traduit par la Rédaction)

Mots-clés: plagioclase zoné, amphibolite, montagnes Adirondack, granulite, dissociation de la hornblende.

INTRODUCTION

Chemically zoned plagioclase is apparently a relatively common constituent of metabasaltic rocks in the granulite facies. For example, it has been reported from Australia (Binns 1964, 1965), Guyana (Cannon 1966) and India (Leelanandam 1968). Significantly, both normal and reverse zoning have been reported, in some instances both from the same area (e.g., Cannon 1966). This is the case with plagioclase in pyroxene amphibolites from a small area in the northwestern lowlands of the Adirondack Mountains of New York. The purpose of this paper is to describe these crystals of zoned plagioclase and report on the petrography and geochemistry of the rocks in which they occur in order to offer a possible explanation for their origin.

GEOLOGICAL SETTING

The northwestern Adirondack lowlands are underlain by a complex of interstratified metasedimentary and metavolcanic rocks last deformed and recrystallized during Grenville time (approximately 1 Ga). These rocks constitute a northeast-trending belt that flanks the Adirondack highlands massif to the southeast. The lowlands belt varies in metamorphic grade from amphibolite facies in the southwest to hornblende granulite facies in the northeast (Engel & Engel 1958, 1960, 1962a). The pyroxene amphibolites that are the subject of this study are associated with a thick stratigraphic unit of regional extent composed dominantly of quartzfeldspathic to pelitic paragneiss (the "major paragneiss" of Engel & Engel). Samples examined were taken from the West Parishville synform, a fold located at the high-grade northeastern end of the lowlands belt (Fig. 1) near the town of Colton.

The gneiss in the area is a weakly foliated, medium-grained rock containing quartz, potassium feldspar, oligoclase-andesine, biotite, garnet, and rutile or ilmenite. Aluminous layers locally carry sillimanite \pm cordierite in addition; aluminum-poor layers locally carry hypersthene. The gneiss in the West Parishville synform is structurally underlain and surrounded by marble and calcsilicate rock. Peak metamorphic conditions in the area have been estimated at 725°C and 5 kbar P_{total} , with $P(H_2O)$ con-

TABLE 1. MODAL COMPOSITION OF PYROXENE AMPHIBOLITES

Specimen* Type	73-C-72 A	73-C-80 A	73-C-198 A	E & E, A A	73-C-71D B	73-C-160 B	74-C-248B B	E & E, B B
hornblende	30.3	59.8	38.8	31.3	23.4	38.4	36.1	16.6
plagioclase	33.8	26.5	39.8	35.6	44.3	34.2	28.4	27.9
clinopyroxene	22.9	3.2	5.3	19.2	4.2	6.6	22.6	}35.0
orthopyroxene	10.2	8.2	14.2	11.3	7.4	12.3	4.1	
garnet	—	—	—	—	13.7	3.7	5.1	11.4
quartz	—	—	—	0.1	—	0.5	0.5	—
opaques**	2.5	2.1	1.9	2.0	6.4	3.7	3.0	6.9
zircon	0.2	tr	—	—	0.2	—	—	—
apatite	0.1	0.2	tr	}0.3	0.4	0.5	0.2	}0.6
Total	100.0	100.0	100.0	99.8	100.0	99.9	100.0	98.4

* E & E, A is the average composition of "least-altered pyroxene amphibolite" from Colton (Engel & Engel 1962a, Table 3); E & E, B is their composition of a garnetiferous pyroxene amphibolite from Colton (their sample A9, Table 6). ** Opaque minerals are ilmenite + magnetite in type-A specimens, ilmenite alone in those of type B; a trace of sulfide may be present in these rocks. A minimum of 1000 points counted per specimen.

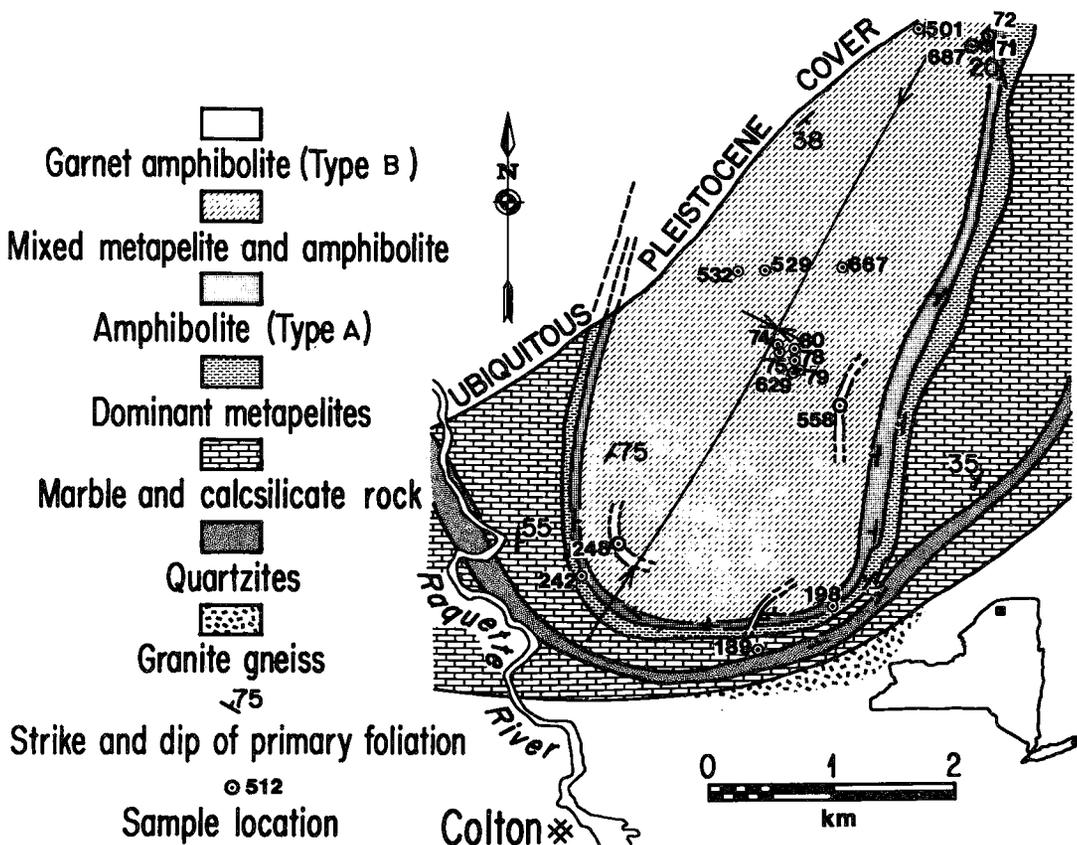


FIG. 1. Generalized geological map of the West Parishville synform near Colton, New York, with locations of specimens discussed in text.

siderably less than P_{total} (Stoddard 1980). There is no evidence for gradients of temperature or pressure in the area and, except within small late-stage shear zones, petrographic evidence for significant retrograde metamorphism or alteration is generally absent. In fact, most thin sections exhibit features suggestive of the attainment of equilibrium, with granoblastic textures and, commonly, 120° intersections of grains.

MINERALOGY AND GEOCHEMISTRY OF THE PYROXENE AMPHIBOLITES

Amphibolites are of two types. The first is a medium-grained, massive black pyroxene amphibolite interlayered with the paragneiss and occurring in layers ranging in thickness from about two cm to 200 m. Differences in mechanical properties of the amphibolite as compared with the paragneiss have

caused thin amphibolite layers to exhibit boudinage structures. The second metabasic lithology is coarser grained, with a lineation defined by hornblende porphyroblasts; it occurs in layers ranging from a few metres to about 50 m thick. Although such layers are largely concordant, they locally cross-cut other lithologies. Unique to this amphibolite is the common development of porphyroblasts of garnet and the rare occurrence of quartz; otherwise the two amphibolites seem to be mineralogically similar in hand specimen.

Thin-section petrographic analysis confirms the mineralogical differences between the two rock types. The amphibolites fall into two distinct groups on the basis of their mineral assemblages: (type A) hornblende + orthopyroxene + clinopyroxene + plagioclase feldspar + ilmenite + magnetite + apatite \pm biotite, and (type B) hornblende + orthopyroxene + clinopyroxene + plagioclase feld-

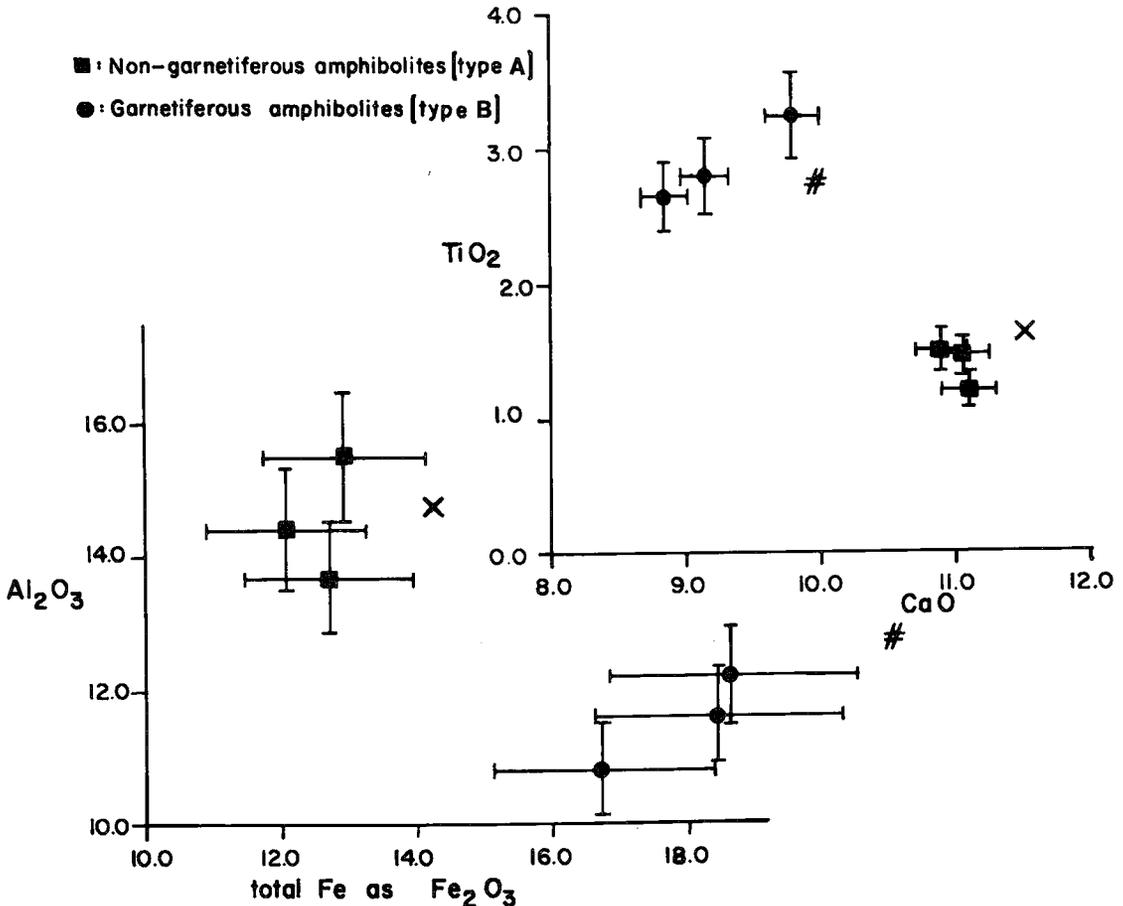


FIG. 2. Selected major oxides of pyroxene amphibolites plotted to show distinct compositional difference between types A (squares) and B (dots). Average composition of "least-altered pyroxene amphibolite" from Engel & Engel (1962a, p. 52) is represented by X and plots with type-A samples; that of their garnetiferous amphibolite (sample A9, Engel & Engel 1962a, p. 58), represented by #, plots with type-B samples.

spar + garnet + ilmenite + apatite ± quartz. Modal data (Table 1) indicate further that type-B amphibolites contain a greater abundance of Fe-Ti oxide minerals and apatite than do type-A amphibolites.

The foregoing observations suggest the presence of a bulk-composition difference between the two types of amphibolite. This hypothesis was tested by chemical analysis of bulk samples by X-ray fluorescence. Compositions of three specimens of each type

TABLE 2. CHEMICAL COMPOSITION OF PYROXENE AMPHIBOLITES

Specimen Type	73-C-72 A	73-C-198 A	75-C-687 A	E & E, A* A	73-C-71D B	73-C-160 B	75-C-558 B	E & E, B* B
SiO ₂	48.4	47.0	48.9	47.89	44.7	46.4	45.8	45.56
Al ₂ O ₃	13.8	15.6	14.4	14.63	11.6	12.2	11.7	12.86
Fe ₂ O ₃ **	13.61	13.82	12.89	14.27	19.67	20.04	17.96	21.06
MgO	5.78	5.84	6.56	7.41	4.31	4.46	4.34	5.46
CaO	10.95	11.01	10.79	11.54	9.11	8.78	9.71	10.01
MnO	0.20	0.20	0.22	0.25	0.26	0.33	0.26	0.35
TiO ₂	1.54	1.25	1.54	1.56	2.93	2.76	3.45	2.73
Na ₂ O	2.48	2.50	2.88	2.19	2.09	2.76	2.68	2.02
K ₂ O	0.65	0.69	0.71	0.58	1.07	0.29	0.54	0.41
P ₂ O ₅	0.18	0.11	0.12	0.14	0.36	0.36	0.60	0.29
Anhydrous Total	97.59	98.09	99.01	100.46	96.10	98.38	97.04	100.75

* E & E, A is the average of "least-altered pyroxene amphibolite" from Colton, by Engel & Engel (1962a, Table 3); E & E, B is their composition of a garnetiferous amphibolite from Colton (their sample A9, Table 6). ** total Fe as Fe₂O₃. Analyst: C.F. Miller. Analyses by X-ray fluorescence.

TABLE 3. COMPOSITIONS OF AMPHIBOLE, PYROXENE AND GARNET

Specimen Type Mineral	73-C-72 A hbl	73-C-198 A hbl	73-C-71D B hbl	74-C-248B B hbl	73-C-72 A opx	73-C-198 A opx	73-C-71D B opx	74-C-248B B opx
SiO ₂	42.39	41.20	38.56	42.45	51.27	50.28	47.61	51.00
Al ₂ O ₃	12.09	11.64	11.96	12.70	0.71	0.90	0.64	0.76
TiO ₂	1.95	2.51	1.92	2.07	na	0.26	0.05	na
FeO*	19.13	17.45	23.76	20.49	32.72	30.88	35.49	35.04
MnO	0.19	0.17	0.16	0.13	0.80	0.75	0.66	0.75
MgO	8.57	9.13	5.98	7.95	15.18	16.34	13.40	14.24
CaO	11.40	11.39	11.03	11.09	0.46	0.78	0.55	0.68
Na ₂ O	1.56	1.64	1.73	1.29	na	0.12	0.02	na
K ₂ O	1.59	1.72	1.68	1.29	na	na	na	na
total	98.87	96.85	96.78	99.90	101.14	100.31	98.42	102.47
Specimen Type Mineral	73-C-72 A cpx	73-C-198 A cpx	73-C-71D B cpx	74-C-248B B cpx	73-C-71D B gar	74-C-248B B gar		
SiO ₂	51.91	50.45	48.68	51.00	36.51	37.61		
Al ₂ O ₃	1.61	2.05	2.02	2.06	21.15	21.33		
TiO ₂	na	0.25	0.21	na	0.05	na		
FeO*	13.47	11.95	16.04	15.06	31.13	29.57		
MnO	0.30	0.31	0.26	0.31	1.87	1.39		
MgO	11.29	11.22	9.11	10.48	2.59	3.09		
CaO	21.61	21.85	21.03	20.93	7.26	7.57		
Na ₂ O	0.40	0.44	0.53	0.41	na	na		
total	100.59	98.52	97.88	100.25	100.56	100.56		

* total Fe as FeO. Abbreviations: hbl hornblende, opx orthopyroxene, cpx clinopyroxene, gar garnet, na not analyzed.

of amphibolite are presented in Table 2; pairs of oxides plotted in Figure 2 demonstrate that the amphibolites fall into two distinct groups with respect to major oxides: type-A amphibolites are enriched in Ca and Al, and impoverished in Fe and Ti with respect to type-B amphibolites.

Because of their consistent differences in composition, mineralogy, and field relationships, it is reasonable to suggest that the two varieties of amphibolite had distinct origins. Specifically, the concordant, nongarnetiferous amphibolites may have originated as basaltic flows, whereas the locally cross-cutting garnetiferous variety may represent slightly younger, Fe-enriched dykes and sills. For comparison, Tables 1 and 2 show compositions reported by Engel and Engel of average "least-altered pyroxene amphibolite" from Colton (Engel & Engel 1962a, p. 52), equivalent to type A of this study, and of a single garnetiferous pyroxene amphibolite, also from Colton (sample A9, Engel & Engel 1962a, p. 58), and equivalent to type B.

Coexisting phases in the amphibolites display systematic partitioning of elements (Table 3); furthermore, bulk chemistry exerts a strong effect on mineral composition and mineral assemblage. Both of these features can be seen in Figure 3; they provide evidence consistent with an approach to equilibrium. Iron-rich, garnetiferous amphibolites contain coexisting ferromagnesian silicates (hornblende, orthopyroxene and clinopyroxene) that are all most enriched in Fe; biotite-bearing, Mg-rich amphibolites have ferromagnesian phases that are poorest in Fe; two pyroxene amphibolites without biotite and garnet have bulk and mineral chemistry of intermediate value of $Mg/(Mg + Fe)$.

These ideas contrast with hypotheses of Buddington (1963, 1966) and Engel & Engel (1962a) that prograde garnet is not stable in amphibolite near Colton. The present study tends to support the hypothesis of De Waard (1965), that rock composition is the major controlling factor with regard to garnet stability.

PLAGIOCLASE IN THE AMPHIBOLITES

Plagioclase is a major constituent of all pyroxene amphibolites. It occurs commonly as anhedral, roughly equidimensional grains in the interstices between pyroxenes and hornblende. The mineral is exceedingly clear and free of alteration, but inclusions of hornblende and apatite are common. Albite twinning is invariably present, and generally extinction of the lamellae is crisp and distinct. Optical zoning is common (Fig. 4).

Plagioclase compositions range from An_{34} to An_{77} , the most common values being in the ranges An_{45-50} and An_{63-65} (Table 4). The maximum difference between core and rim compositions is 13% An component. Both normal (Na-rich rim) and reverse types (Ca-rich rim) of chemical zonation are present; the particular type displayed within a given thin section is consistent and a function of the mineral assemblage. Zoned plagioclase from nongarnetiferous amphibolites shows reverse zonation; from garnetiferous amphibolites, zoned plagioclase displays normal zonation. A few plagioclase grains studied show complex profiles. Figure 5 is a diagram of composition profiles with examples of A) reverse zonation, B) normal zonation, and C) complex zonation.

There is no reason to suspect that the rims of

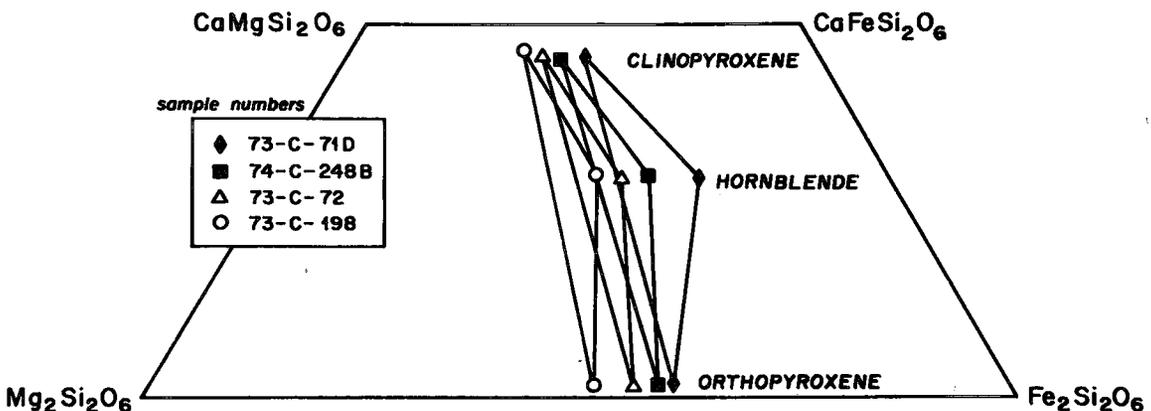


FIG. 3. Composition of coexisting pyroxenes and hornblende from two specimens of each type of amphibolite. The two Fe-rich assemblages also coexist with garnet. Locally, Mg-rich assemblages (not plotted) carry biotite. Note the consistent partitioning of Fe and Mg as well as the correlation between mineral and bulk-rock compositions (*cf.* Table 2). Hornblende compositions, plotted here in terms of pyroxene components, do not actually lie in the pyroxene plane.

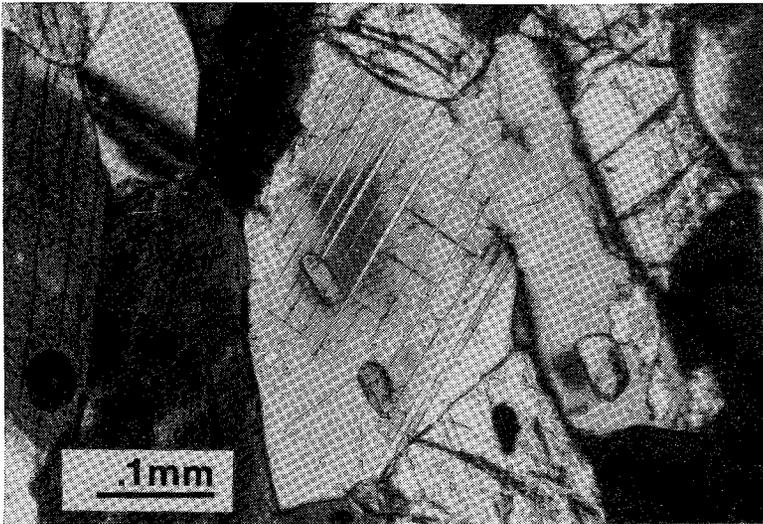


FIG. 4. Photomicrograph, under crossed polars, of optically zoned grain of plagioclase in a specimen of type-A amphibolite (sample 73-C-80). Composition ranges from about An_{65} (core) to An_{80} (rim). Midway between core and rim, region of exsolution lamellae probably represents unmixing of the Huttenlocher type (Smith 1974).

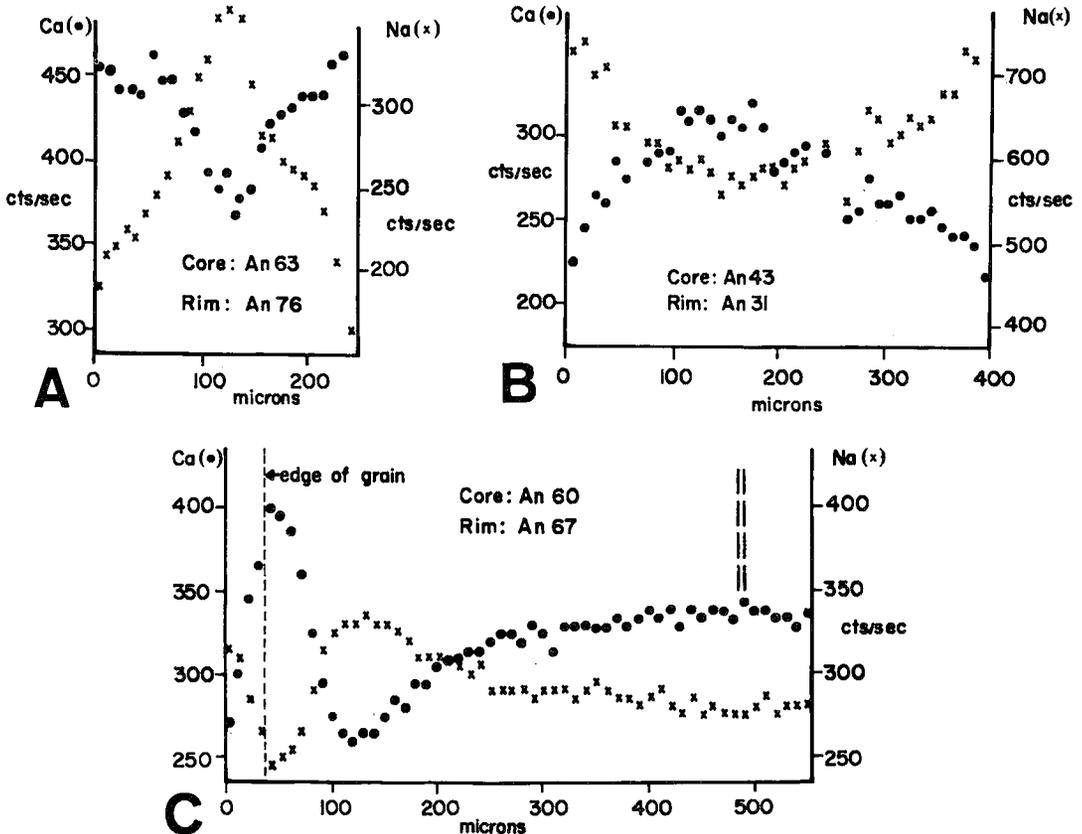


FIG. 5. Representative profiles by microprobe across optically zoned plagioclase from a sample of type-A amphibolite (A) and type-B amphibolite (B). A few samples have plagioclase that exhibits more complex profiles (C), suggesting that more than one episode of reaction may have occurred. Dashed double vertical line in C locates centre of grain.

TABLE 4. REPRESENTATIVE COMPOSITIONS OF PLAGIOCLASE

Specimen	73-C-80	73-C-80	73-C-198	73-C-198	73-C-160	73-C-160	74-C-248B	73-C-248B
Type	A	A	A	A	B	B	B	B
Grain	G	G	W	W	C	C	A	A
Core/rim	rim	core	rim	core	rim	core	rim	core
SiO ₂	48.50	51.42	51.14	53.52	59.47	57.73	54.53	53.16
Al ₂ O ₃	32.47	30.77	31.59	29.57	26.20	27.44	28.28	28.91
CaO	15.73	13.15	13.77	12.65	7.63	9.52	10.28	10.98
FeO	0.46	0.18	0.23	0.13	0.22	0.11	0.22	0.11
Na ₂ O	2.90	4.42	3.99	4.65	7.86	6.78	6.19	5.73
K ₂ O	0.11	0.19	0.16	0.20	0.22	0.20	0.21	0.19
total	100.17	100.13	100.88	100.72	101.60	101.78	99.71	99.08
Cations on the basis of eight atoms of oxygen								
Si	2.223	2.339	2.311	2.410	2.623	2.552	2.473	2.429
Al	1.755	1.650	1.683	1.571	1.363	1.430	1.512	1.558
Fe ³⁺	0.018	0.007	0.009	0.005	0.008	0.004	0.008	0.004
Ca	0.773	0.641	0.667	0.610	0.361	0.451	0.499	0.538
Na	0.258	0.390	0.347	0.406	0.672	0.581	0.544	0.508
K	0.006	0.011	0.009	0.012	0.012	0.011	0.012	0.011
Molecular percent of feldspar end-members								
Ab	24.9	37.4	34.0	39.5	64.3	55.7	51.6	48.1
An	74.5	61.5	65.1	59.3	34.5	43.2	47.3	50.9
Or	0.6	1.1	0.9	1.2	1.1	1.1	1.1	1.0

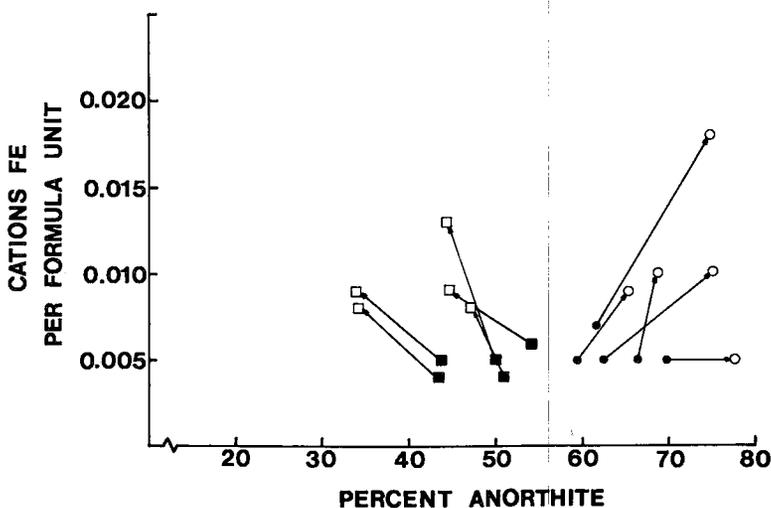


FIG. 6. An content of plagioclase plotted against number of Fe cations (calculated as Fe³⁺) per formula unit. Note increase in Fe at rim, regardless of type of zoning. Circles: plagioclase from type-A amphibolite; squares: plagioclase from type-B amphibolite. Closed symbols represent the core, and open symbols, the rim.

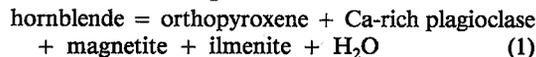
reversely zoned plagioclase in type-A amphibolites grew under conditions different from those of normally zoned plagioclase in type-B amphibolites. In fact, their textural similarities and their relationships to bulk chemistry suggest that they both formed under the same high-temperature conditions. Further support for this argument comes from the fact that Fe is consistently enriched in the rim of plagioclase grains, regardless of type of zonation (Fig. 6). The solubility of Fe in plagioclase increases with temperature and oxygen fugacity (?) (Smith 1974), hence the rims of both normally and reversely zoned grains probably formed under the same conditions at or near the peak of metamorphism.

HORNBLLENDE BREAKDOWN

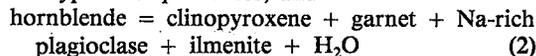
Petrographic evidence in rocks of the present study (for example, hornblende inclusions within its presumed breakdown-products, symplectic intergrowths of breakdown products in interstices between adjacent hornblende grains) and regional studies in the Adirondacks and elsewhere (*e.g.*, Engel *et al.* 1964, Binns 1964) have shown that hornblende dehydrates within the granulite facies by means of continuous or discontinuous reactions; these produce clinopyroxene or orthopyroxene (or both) with or without garnet or biotite. Field (Engel & Engel 1962b, Binns 1965) and laboratory studies (Binns 1969, Spear 1981) have shown that modal concentrations of phases vary systematically over the interval in which hornblende is progressively eliminated, and that the chemical composition of hornblende and of coexisting phases also varies in a continuous fashion. However, in the present study, direct evidence for continuous breakdown of hornblende is lacking, because the composition of hornblende grains in a given specimen is constant within the limits of analytical uncertainty. Hornblende inclusions within the mineral grains that were produced by hornblende breakdown are identical in composition to hornblende from the matrix of the same rock. Also, the total range of hornblende compositions reflects bulk chemistry. Finally, ferromagnesian silicates are unzoned.

Type-A amphibolites have a low value of Fe/Mg and reversely zoned grains of plagioclase whose rim approaches An₇₀₋₈₀ in composition; type-B amphibolites have a high value of Fe/Mg and normally zoned grains of plagioclase with a rim approaching An₃₅₋₄₅. Plagioclase rims and hornblende in type-A amphibolites have a similar value of Ca/Na; in type-B amphibolites, the Ca/Na value of hornblende is much greater than that of the plagioclase rims. These observations may be explained by envisioning hornblende breaking down discontinuously to produce plagioclase plus various combinations of ferromagnesian silicates

(orthopyroxene, clinopyroxene, Ca-bearing garnet). Thus a strong influence is exerted on the composition of the plagioclase produced. Two end-member model reactions might be:

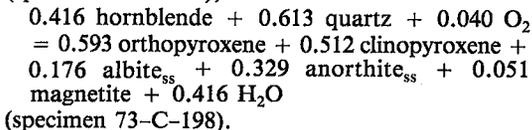
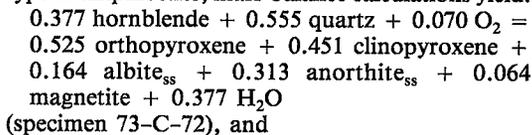


for type-A amphibolites, and

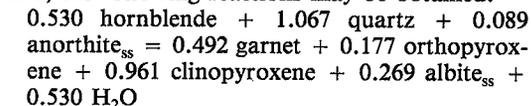


for type-B amphibolites. Reactions that produce two pyroxenes would presumably yield plagioclase of intermediate composition.

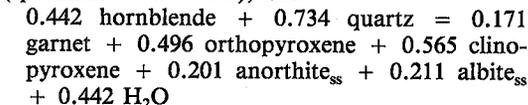
In fact, hypothetical hornblende-breakdown reactions may be written for each amphibolite assemblage, based upon the compositions of the coexisting ferromagnesian silicates in each specimen (Table 3), if one assumes (1) that the minor components TiO₂, MnO and K₂O can be safely ignored, (2) that excess SiO₂ was available, and (3) that excess FeO produced by hornblende breakdown could be oxidized to yield magnetite. Because there is an unknown amount of Fe³⁺ in the hornblende (and garnet), none of the actual reactions needs to have been oxidizing. Presumably the plagioclase component produced in each of the following reactions would go into solid solution in pre-existing plagioclase grains and would thus be reflected in the rim compositions of Table 4. For two specimens of type-A amphibolite, mass-balance calculations yield:



The plagioclase produced by these reactions corresponds to An_{65.6} and An_{65.1}, respectively (*cf.* Table 4). For two garnet-bearing type-B amphibolites, the following reactions may be obtained:



(specimen 73-C-71CD), and



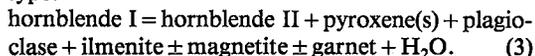
(specimen 74-C-248B).

The latter reaction produces plagioclase of composition An_{48.8} (*cf.* Table 4), whereas the former actually consumes the anorthite component while yielding the albite component. Table 5 summarizes the relationships.

TABLE 5. AMPHIBOLITE CHARACTERISTICS AND INFERRED HORNBLLENDE-BREAKDOWN REACTIONS

Amphibolite type	Examples	Magnetite or garnet	Plagioclase rim	Plagioclase zoning	Bulk Fe/Mg	Mol. Ca/(Ca+Na)	Inferred reaction
A	72, 80, 198	magnetite	An60-80	reverse	low	hbl<plag. rim	(1)
B	71D, 160, 248B	garnet	An35-50	normal	high	hbl>plag. rim	(2)

Whether or not these hypothetical reactions actually took place is not important; they are intended to illustrate quantitatively the consistent relationship between mineral assemblage and type of zoning, and further to suggest that the actual composition of plagioclase component produced in hornblende breakdown-reactions may be very sensitive to the compositions of coexisting minerals. Although the foregoing reactions are discontinuous and pseudo-univariant, the actual reactions that took place in the rocks were probably continuous reactions of the type:



CONCLUSIONS

Reversely zoned plagioclase in metamorphic rocks is not necessarily the result of retrograde reactions. Although it is true that plagioclase commonly becomes more calcic with increasing temperature within the amphibolite facies (e.g., De Waard 1959, Crawford 1966, Cooper 1972), this is not always true in the granulite facies. Bulk-rock composition in addition to P and T may determine mineral reactions that produce a plagioclase component of a particular An content (see also Misch 1968). Strict attention should always be paid to the actual reactions that produce the chemical changes in plagioclase.

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