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Amphiboles and biotite in relation to the stages of metamorphism in granogabbro

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SUMMARY. The Bratteggdalen granogabbro, a relict mass in the Caledonian amphibolites of Vestspitsbergen, shows a series of amphibole development reflecting its thermal history. A brown Ti-rich hornblende occurs as a late magmatic mantling of pyroxene. Low-grade metamorphism (greenschist facies) was accompanied by the development of actinolite from pyroxene; subsequent amphibolite facies metamorphism saw the development of an alkali-rich, blue-green hornblende. The chemical changes in the amphiboles are a succinct illustration of those found in progressive regional metamorphism.

THE Pre-Cambrian volcanic and subvolcanic Eimfjellet formation, part of the Hecla Hoek succession in the Hornsund area of Vestspitsbergen, was metamorphosed during the Caledonian Orogeny. The metamorphosed granogabbro of Bratteggdalen, an interesting mineralogical and textural relict within the amphibolites, was previously described by Smulikowski (1965).

An ophitic texture in the rock, with oligoclase-andesine plagioclase, amphiboles, and biotite idiomorphic against the typically xenomorphic quartz and chequer-albite appears to be inherited from the original primary igneous texture. Microscopic study of relicts, reaction-rims. and other mineral interrelations has enabled the reconstruction of the metamorphic history of the rock; in fig. 1 this is shown in diagrammatic form.

Pyroxene relicts occur in two ways: as small, generally well-preserved grains enclosed within greenish-brown hornblende (α olive, β greenish-brown, γ dirty green, low birefringence, γ: [001] 11°; called here *brown hornblende*), or as irregular pyroxene grains being replaced by aggregates of actinolite (colourless to pale green, high birefringence, γ: [001] 15°). Blue hornblende (blue to bluish-green, medium birefringence, γ:[001] 15°) forms rims on both the brown hornblende and the actinolite. As shown in fig. 1 the brown hornblende is considered to be a primary, magmatic mineral, while the actinolite belongs to an early stage of metamorphism, and the blue hornblende to the main, somewhat later stage.

Primary biotite is developed in form of large homogeneous, dark reddish-brown plates (2V = 12°) with sagenite net and sphene rims. Some of the plates display lighter colouration near the margins. Small flakes of lighter biotite also occur, most of them associated with bigger biotite plates. However, it cannot be excluded that some small-flake biotite is the result of biotitization of amphiboles.

Ilmenite ($Ti_{2\cdot0}Fe_{1\cdot9}Mn_{0\cdot1}O_{6\cdot0}$); grains have always rims of sphene:

$$[(Ca_{3\cdot45}Na_{0\cdot01}K_{0\cdot04}Mn_{0\cdot01})(Ti_{3\cdot34}Fe_{0\cdot20}Mg_{0\cdot03})(Si_{3\cdot55}Al_{0\cdot25})O_{16\cdot00}(OH)_{4\cdot00}].$$

Light minerals in the rock are represented by two kinds of plagioclase and quartz. The oligoclase-andesine grains are rich in small grains of epidote, of composition corresponding to the intermediate members of clinozoisite–epidote series. A relict of

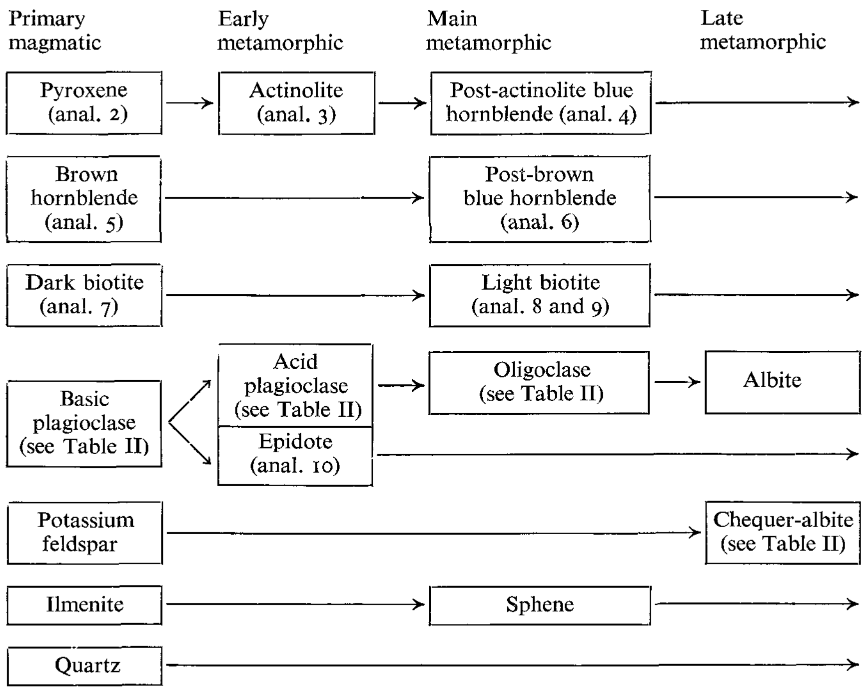

FIG. 1. Schematic representation of mineral alterations in granogabbro and stages of metamorphic development (analyses see Table I).

the original zonal structure of plagioclase can be distinguished in some grains. Epidote is often concentrated at the central, originally most anorthite-rich part. The highest determined An content in plagioclase is 44 %, but in the saussuritized centres of the grains it must have been still higher. An accurate calculation of the original composition of plagioclases is not possible, but 55 to 60 % An is a not unreasonable estimate (see Table II).

Another kind of plagioclase is represented by albite with minute, irregular albite twinning, so called 'Chequer-albite'. This mineral fills the interstices together with quartz between the partly idiomorphic oligoclase-andesine laths and dark minerals. In the primary magmatic granogabbro they must have been occupied not by plagioclase but by another mineral, most probably potassium feldspar, later totally replaced by albite during the metamorphism (Smulikowski, 1965). Along the boundary of the oligoclase-andesine grains against the chequer-albite narrow, clear, untwinned rims of albite-oligoclase composition have been developed and very subtle myrmekite can often be observed in them.

In Table III an attempt has been made to calculate the primary gabbro modal composition from the present mode (col. 1). In this calculation it is assumed that metamorphic transformations occurred at constant volume and that all actinolite was originally pyroxene. Blue hornblende, developed as single grains, was counted as either brown hornblende or as actinolite, in proportion to the content of the latter minerals in the rock; 'total' actinolite was then counted as pyroxene. Similarly secondary biotite was divided between brown hornblende, actinolite, and primary biotite.

The graph, fig. 2, presents numbers of ions in the formulae listed in Table I. The pyroxene formula is presented on the graph as calculated on the basis of 24 (O), to be compared with the amphiboles. The amphibole formula $A^{xii}_{0-1}\ X^{viii}_{2}\ Y^{vi}_{5}\ Z^{iv}_{8}\ O_{22}$ $(OH, O, F)_2$ (Ernst, 1968) and biotite formula $X_2\ Y_{4-6}\ Z_8\ O_{20}\ (OH, F)_4$ (Deer *et al.*, 1962) have been used for calculation. The Fe^{2+}/Fe^{3+} ratio and H_2O^+ could not be determined in the probe and the data in the formulae would be affected by it, especially Al^{vi}, and Mg^{viii} and Na values for amphiboles. The more important ratios are listed in Table I and plotted on the graph (fig. 3). The minerals are arranged in genetically related groups in which various minerals correspond to different stages of metamorphic development.

Discussion and conclusions

The study of the rock texture and the composition of minerals, e.g. zonal amphiboles, varying composition of the blue hornblende in relation to the original minerals, non-homogeneous biotite, relicts of pyroxene, sphene rims on ilmenite, two kinds of plagioclase, one of them zonal, clearly show that the rock as a whole did not attain equilibrium; possibly only local equilibria existed.

In the granogabbro in question the succession of minerals and their compositional modifications allows inference of the petrologic history of the rock. Thus the direction of changes of composition of minerals (first of all amphiboles) in relation to the stages of metamorphism, which can be distinguished on the basis of textural and structural features, can throw some light on the conditions of metamorphism.

Primary magmatic rock. Pyroxene (augite Mg 31·0, Fe 24·5, Ca 44·5), brown hornblende rich in Ti and alkalis, dark Ti-rich biotite, ilmenite, zonal andesine-labradorite plagioclase, potassium feldspar, and quartz, represent an assemblage of a possibly subvolcanic rock of granogabbro composition. The high Ti content in brown hornblende is in agreement with the opinion (Leake, 1965), that high Ti is an indicator of high temperature and is responsible for brown tints in amphibole. The high Na+K content in brown hornblende fits well with it and indicates the high temperature of formation (Bard, 1970; Engel and Engel, 1962).

Early stage of metamorphism. Very little can be deduced about the replacement of pyroxene by actinolite. The Ti content in actinolite is even lower than that in the replaced pyroxene. This replacement could represent a low temperature, autometamorphic uralitization, or, more probably, a metamorphic process at the beginning of

TABLE I. *Chemical analysis of granogabbro and electron-probe analyses of minerals*

	1	2	3	4	5	6	7	8	9	10
Na_2O	3·41	0·26	0·8	0·79	1·33	1·59	0·12	0·09	0·10	0·05
MgO	4·92	10·64	11·67	10·64	9·28	5·33	8·52	9·19	8·63	0·01
Al_2O_3	14·88	0·42	0·73	4·59	7·09	14·93	14·71	15·01	15·99	27·39
SiO_2	51·85	51·68	53·43	48·72	44·58	40·46	35·97	36·12	35·29	38·44
K_2O	2·45	0·03	0·04	0·40	0·94	0·94	9·26	8·79	9·22	0·04
CaO	8·99	21·28	12·22	11·29	10·76	11·39	0·02	0·07	0·04	23·27
TiO_2	1·55	0·09	0·03	0·29	1·78	0·31	2·88	1·93	1·66	0·04
MnO	0·23	0·44	0·36	0·39	0·31	0·32	0·18	0·18	0·16	0·17
Fe_2O_3	2·84 } 10·57	—	—	—	—	—	—	—	—	8·04‡
FeO	8·01 }	15·03*	19·69*	20·15*	21·10*	21·86*	23·62*	23·19*	23·44*	—
Total	100·37†	99·87	98·25	97·26	97·17	97·13	95·28	94·57	94·53	97·45
FeO/MgO*	4·10	1·41	1·69	1·89	2·27	4·10	2·77	2·52	2·72	—
Al_2O_3/SiO_2	0·29	0·01	0·01	0·09	0·16	0·37	0·41	0·42	0·45	—
K_2O/Na_2O	0·72	0·12	0·50	0·51	0·71	0·59	77·17	97·66	92·20	—
(Na+K)/(Na+K+Ca)	0·415	0·014	0·018	0·103	0·188	0·187	—	—	—	—
$100Al^{vi}/Si^{iv}$	—	0·30	0·38	2·49	2·17	15·73	5·81	7·37	9·21	—

	2		3	4	5	6		7	8	9		10
Number of ions on the basis of *n* oxygen:												
n	6		23	23	23	23		22	22	22		12½
Si	1·987		7·901	7·365	6·863	6·261		5·615	5·648	5·547	Si	3·009
Al^{iv}	0·013		0·099	0·635	1·137	1·739		2·385	2·352	2·453	Al	—
Z	2·00		8·00	8·00	8·00	8·00		8·00	8·00	8·00	Σ	3·01
Al^{iv}	0·006		0·030	0·183	0·149	0·895		0·323	0·416	0·511	Al	2·527
Ti	0·003		0·004	0·034	0·206	0·037		0·338	0·228	0·196	Fe^{3+}	0·474
Fe*	0·484		2·436	2·548	2·715	2·829		3·084	3·032	3·083	Mn	0·001
Mn	0·015		0·045	0·051	0·041	0·043		0·025	0·024	0·022	Σ	3·01
Mg	0·610		2·485	2·184	1·889	1·196		1·982	2·143	2·072	Mg	0·001
		Y	5·000	5·000	5·000	5·000		5·75	5·84	5·83	Ti	0·002
		Mg	0·087	0·215	0·238	0·034		—	—	—	Ca	1·951
Ca	0·877		1·936	1·829	1·773	1·888		0·003	0·011	0·008	Σ	1·95
Na	0·020		—	—	—	0·078		—	—	—		
		X	2·023	2·044	2·011	2·000						
		Na	0·024	0·233	0·397	0·399		0·037	0·028	0·032		
K	0·002		0·008	0·078	0·185	0·185		1·845	1·753	1·849		
YX	2·02	*A*	0·032	0·311	0·582	0·584	*X*	1·89	1·79	1·89		

* Total Fe calculated as FeO or Fe^{2+}. † Including 0·12 % P_2O_5, 0·10 % H_2O-, 1·02 % H_2O+; analysed by W. Narebski in Smulikowski, 1965.
‡ Total Fe calculated as Fe_2O_3.

1. Bulk composition of the rock (granogabbro).
2. Pyroxene.
3. Actinolite.
4. Post-actinolite, blue hornblende.
5. Brown hornblende.
6. Post-brown, blue hornblende.
7. Dark biotite, central.
8. Light biotite, marginal.
9. Light biotite, small flakes.
10. Epidote in plagioclase.

TABLE II. *Composition of plagioclases in granogabbro (electron-probe determinations)*

	Ab	An	Or	Total
1a Elongated grain, in its central part rich in clinozoizite inclusions. Analysed spot in central part surrounded by epidote.	81·9 %	14·4 %	0·8 %	97·1 %
1b The same grain, marginal part free from epidote inclusions.	76·1	24·8	0·7	101·6
2a Zonal grain with epidote aggregate in the centre. Most central zone.	54·4	44·4	0·5	99·3
2b The same grain. Intermediate zone.	65·7	33·8	0·6	100·1
2c The same grain. Marginal zone.	75·6	25·3	0·5	101·4
3a Chequer-albite. Big homogeneous grain.	93·8	5·9	0·3	100·0
3b The same grain, another spot.	93·5	5·6	0·3	99·4

TABLE III. *Mineral composition of granogabbro (in volume per cent)*

Actual composition (average of 2 samples)				Supposed composition of primary magmatic rock		
Quartz		7·1	46·5 (Quartz + Chequer-albite + Oligoclase-andesine+epidote)	7·1	46·5 (Quartz + Potassium feldspar + Basic plagioclase)	Quartz
Chequer-albite		8·1		8·1		Potassium feldspar
Oligoclase-andesine+epidote		31·3		31·3		Basic plagioclase
Pyroxene		1·6	49·6 (Pyroxene to Blue hornblende)	27·5	49·6 (Pyroxene + Biotite + Brown hornblende)	Pyroxene
Dark biotite, primary	9·1	19·5 (Dark + Light biotite)		11·9		Biotite
Light biotite in small flakes	10·4					
Brown hornblende		6·6		10·2		Brown hornblende
Actinolite		17·6				
Blue hornblende in rims on actinolite	1·8	4·3 (Blue hornblende total)				
Blue hornblende in rims on brown hornblende	1·1					
Blue hornblende in separate grains	1·4					
Ilmenite		1·6	3·9 (Ilmenite + Sphene + Apatite + Zircon)	2·3	3·9 (Ilmenite + Apatite + Zircon)	Ilmenite
Sphene		0·7				
Apatite		1·6		1·6		Apatite
Zircon		n.d.		n.d.		Zircon

prograde sequence, corresponding to the greenschist facies. Brown hornblende and biotite were resistant to this low-grade change. Plagioclase might have been saussuritized in this stage. This would explain the lower An content in the epidote-rich centres of some grains than in their marginal parts, as the marginal parts were later recrystallized during the main stage of metamorphism, at higher *PT* conditions.

Main stage of metamorphism. Increasing metamorphic grade was accompanied by the formation of blue hornblende as rims on the already existing amphiboles, i.e. actinolite and brown hornblende.

The blue hornblende rimming actinolite differs considerably from that rimming the brown hornblende. This is not only the higher alkali content together with the lower

Ti content responsible for the bluish colouration of the hornblende. Although oxidation ratio and H_2O^+ content can play an important role, they are not determinable in the electron probe. According to Seitsaari (1953) blue-green hornblende has a higher H_2O^+ content and higher Fe^{3+}/Fe^{2+} ratio than other hornblendes.

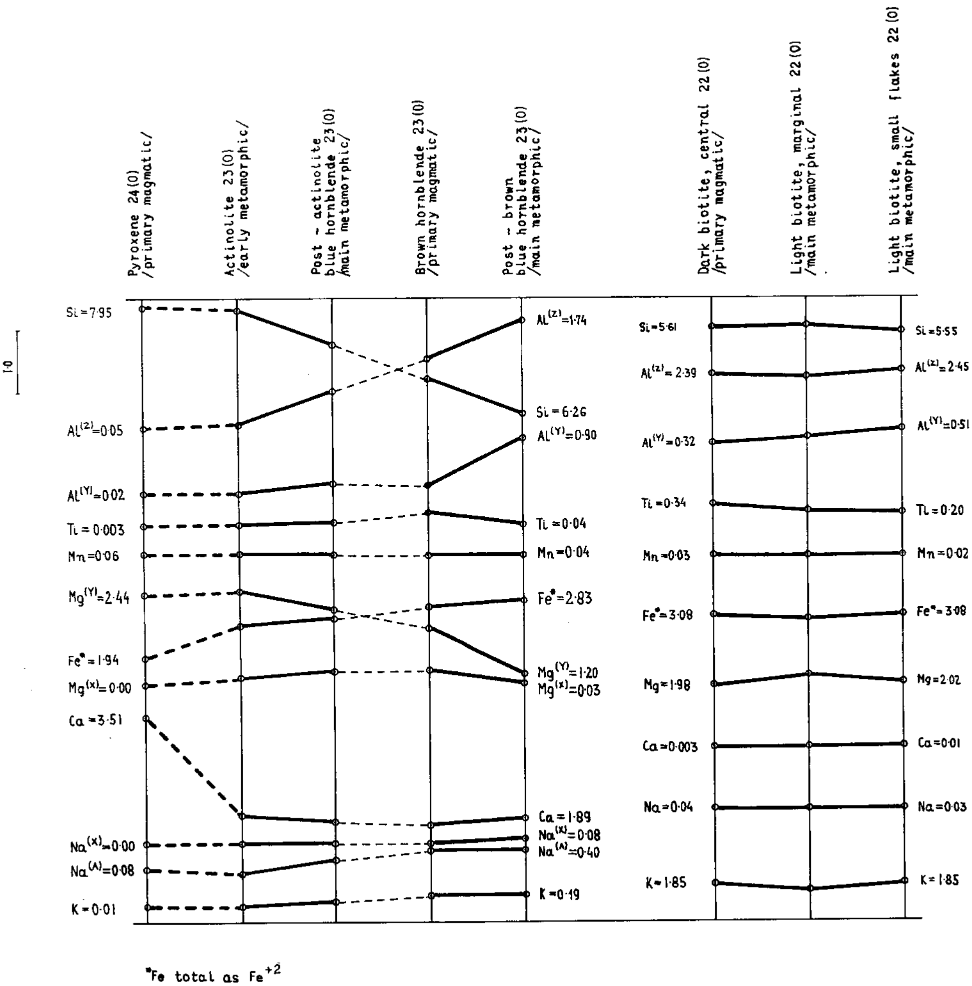

FIG. 2. Ions per *N* oxygen in successional minerals compared. Total Fe as Fe^{2+}.

The post-actinolite blue hornblende shows an increase of Ti, alkalis, Al^{vi}, and Al^{iv}, together with a decrease of Si, as compared with the actinolite; a slight increase of FeO/MgO ratio and a rather insignificant increase of K_2O/Na_2O ratio can be observed. The 'post-brown', blue hornblende, compared with the brown one, shows a remarkable increase of Al^{vi} and Al^{iv}, whereas the alkalis remain at almost the same level; the K_2O/Na_2O ratio slightly decreases; Ti decreases to the same level as in the post-actinolite blue hornblende.

It could be reasonably assumed that blue hornblende originated more or less simultaneously throughout the rock. The significant difference in composition in relation to the rimmed amphibole illustrates the local equilibria. The biggest difference is in the Al_2O_3/SiO_2 and Al^{vi}/Si^{iv} ratios and the MgO/FeO and (Na+K)/(Na+K+Ca)

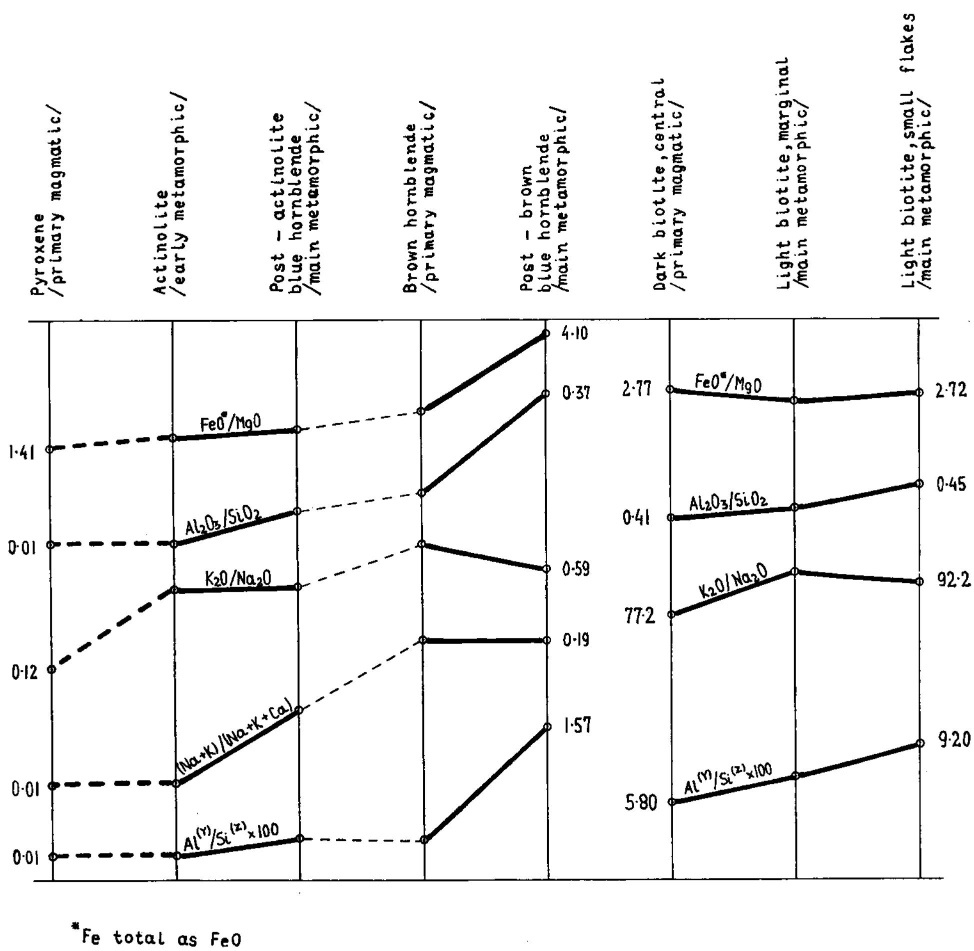

FIG. 3. Ions per N oxygen in successional minerals compared. Total Fe as Fe^{2+}.

ratios as well. Both the boundaries between brown hornblende and blue rims, as well as between actinolite and blue rims, are sharp. This seems to be the result of a rapid change of conditions, rather than being due to a compositional gap in the amphiboles.

The lighter variety of biotite present in the rock as aggregates of small flakes or as marginal parts of bigger plates with the dark primary variety in their centres would correspond probably to the main stage of metamorphism. The chemical changes in response to changing conditions are not well pronounced in biotite. The lighter variety differs from the dark original-magmatic one with higher Al_2O_3/SiO_2 ratio. This is illustrated even better by the Al^{vi}/Si^{iv} ratio. The Ti content in light biotite is significantly lower than in the dark one. The sphene rims on ilmenite were formed probably during the same stage of metamorphism.

The outermost epidote-free rims of plagioclase grains of oligoclase An_{25} composition would most probably correspond to the main stage of metamorphism. The original-magmatic plagioclase grains were zonal. In some of them this zonation could be preserved, while in the others the saussuritization took place in the early stage of metamorphism.

Oligoclase and blue hornblende would represent a typical assemblage of the lower part of amphibolite facies of prograde metamorphism.

Late stage of metamorphism. It is supposed that in this stage a metasomatic replacement of original potassium feldspar by the chequer-albite took place. Possibly the albite-oligoclase rims of plagioclase grains along the boundary against chequer-albite were formed simultaneously as a result of the albitization of the more basic oligoclase along the contact with the totally albitized potassium feldspar. The subtle myrmekite could be the relict of original myrmekite along the boundary potassium feldspar–plagioclase, but the possibility that it was connected with the process of albitization cannot be excluded. The albitization of potassium feldspar could not take place simultaneously with the formation of blue hornblende in the main stage of metamorphism, as the K_2O/Na_2O ratio would have dropped rapidly in the blue hornblende as compared with the brown one. The albitization probably took place as a late, separate event in low metamorphic conditions, the other minerals being unchanged.

It might be assumed that the primary, magmatic stage represents high-temperature, low-pressure conditions; the early stage would correspond to low-temperature, rather low-pressure; the main stage to moderate temperature and pressure; and the late stage again to low temperature and pressure conditions.

In the case described the Ti content in hornblende seems to be a sensitive indicator of physical conditions of formation, especially, as suggested by Leake (1965), of temperature. This would be the indicator working both ways—up, from actinolite to blue hornblende (rise of temperature and pressure), and down, from brown hornblende to blue hornblende (temperature down, pressure up).

The alkalis mentioned by Bard (1970) as a good prograde indicator show the increase from actinolite to blue hornblende, while they did not react to the change of conditions from brown hornblende to blue.

Non-systematic variation of Al/Si has been reported in prograde metamorphism (Engel and Engel, 1962; Bard, 1970), but generally a decrease of silica is observable with increase of metamorphic grade, especially in the lower grades of metamorphism (Leake, 1965; Miyashiro, 1967). The Al^{vi} is generally lower in igneous amphiboles than in metamorphic ones. Al^{vi} is supposed to be dependent on pressure (Leake, 1965) but Black (1973) pointed out that Al^{vi} is high in sodic amphiboles while it is low in co-existing calcic amphiboles. The increase of Al_2O_3/SiO_2 as well as Al^{vi} in amphiboles of the granogabbro in question, formed during the main stage of metamorphism (blue hornblende) is fairly clear both as compared with the primary magmatic stage (brown hornblende) and with the early stage of metamorphism (actinolite). The increase of Al in amphiboles in this case would be dependent rather on pressure than on temperature increase.

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