

BIOTITES IN GRANITES, BIOTITES IN GNEISSES, AND THE STATUS OF BIOTITE AS A ONE-MINERAL ENVIRONMENT INDICATOR

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ABSTRACT

Some aspects of one-mineral geological environment indicators are discussed. Factors controlling the composition of biotite in different P, T, and chemical environments are reviewed. On the basis of theoretical considerations and empirical observation it is concluded that biotites in igneous granites may tend to be somewhat richer in iron than biotites from gneisses, but the relation is not strict, and the Fe/Mg or FeO/MgO/Al₂O₃ ratios in biotite do not provide unequivocal indications of the origin of biotite-bearing rocks.

Minerals as geological indicators

A common approach to the problem of temperature, pressure, and provenance of material in petrogenesis is the investigation of intracrystalline chemical variations in mineral phases from different types of rock. The observed differences are then generalized to establish universally valid indicators. Familiar instances are observations of high concentration of Mn and/or Ca in low-grade garnets (Miyashiro 1953, Atherton 1965, and others), high Mg in garnets from granulites and eclogites, high contents of Al and Ti in high-grade amphiboles (Harry 1950, Leake 1965).

An inherent setback in using this »empirical» approach is the difficulty to appreciate the influence of changing rock composition as contrasted to intracrystalline variations depending on temperature, pressure, fugacity of volatiles, etc. A typical and fairly well investigated »index mineral» is garnet, where the common observation of high Ca and Mn in low-grade garnets has been

reversed to propose a one-mineral geological thermometer (Nandi 1967, Li-Ping Tan 1968). An implication must then be that Mn (and Ca) either enter other minerals as temperature or pressure increase, or that there is wholesale loss of manganese from all rocks of higher facies. Difficulties of such an application of mineral composition are indicated by observations of garnets high in Ca and/or Mn in plutonics and in high-grade metamorphic rocks (Wright 1938, Clifford 1960, and others), and by studies of intermineral element distribution. Considering the particular case of Mn in garnet, the distribution of manganese between garnet and other common ferromagnesian silicates (Kretz 1959, Gorbatshev 1968, Saxena 1968 a and b, and others) suggests that Mn (and Ca) stabilize the garnet lattice in preference to biotite, amphibole, and cordierite. Low-grade garnets close to the lower stability limits of this mineral at any particular Fe/Mg will thus tend to form in rocks rich in Mn and Ca. Since from experiments

and studies of mineral associations garnet is on the high-grade side of reactions of the type:

biotite (or chlorite) + muscovite + quartz \rightleftharpoons garnet + K-feldspar + H₂O and

biotite + Al₂SiO₅ + quartz \rightleftharpoons garnet + K-feldspar + H₂O,

the absolute amount of garnet in any particular isochemical rock composition will tend to increase with increasing grade, the rate depending among other things on the Fe/Mg ratio and the activity of water. Consequently the attainment of higher metamorphic facies is accompanied by dilution of Mn in garnet and increase in Mg, which does not need imply that Mn is expelled from the garnet lattice. This is well in accord with observed changes of garnet composition in complex lithological sequences where variations of Mn and Mg in garnet with metamorphic grade are prominent, but differ between the various rock types as a function of total-rock composition (*e.g.* Velikoslavinsky 1965).

A considerably more rewarding approach than that of single-mineral indicators is the study of interphase element distribution, which is based on the fact that at equilibrium the chemical potential of a chemical species is the same in all coexisting phases. This provides a firm theoretical basis liable to treatment in strictly quantitative terms. Pioneered by Ramberg (1949), and Ramberg and De Vore (1951), numerous investigators have during the last decades shed light on crystal-chemical and environmental factors governing the distribution of elements between different coexisting mineral phases. Actually some useful »one-mineral indicators«, as for instance the contents of Ti in biotite or amphibole or magnetite, can be considered more strictly as cases of the intermineral distribution model.

Somewhat more evasive are indicators of »geological provenance« of chemical components which enter rocks and minerals. The basis of reasoning is here that certain geological processes have compositional or structural consequences of such a frequency and regularity as to make them

eligible for use as guides in the interpretation of petrogenetical problems. A common and long-pursued approach has been the observation of compositional parameters of minerals and rocks and their attribution to certain environments of formation. Considering the particular case of biotite, differences between groups of biotites from different types of rock have been observed by *a.o.* Heinrich (1946) and Rimšaite (1964), and have recently been proposed as means of discriminating between granites of »igneous« and »metamorphic« origin (Gokhale 1968). This potential employment of biotite as a one-mineral provenance indicator is entirely dependent on the regularity of the observed compositional variation, preferably augmented by the theoretical recognition of processes which exclude certain biotite compositions from products of either igneous crystallization or metamorphic-metasomatic granitization. Some aspects of the problem are discussed here.

Some theoretical considerations

While the Fe/Mg proportions of biotites may reflect the whole-rock Fe/Mg ratio there is a number of other factors which may have equal or greater importance. At equilibrium any rock will have the mineral composition corresponding to a minimum of free energy. The equilibrium distribution of elements between different mineral phases implies equality of chemical potential (μ) of a species i in all host minerals:

$$\mu_i^a = \mu_i^b = \mu_i^c \dots = \mu_i^n \quad (1)$$

where $a, b, c \dots n$ are mineral phases. The chemical potential of a chemical species is determined by concentration, temperature, and pressure according to:

$$\mu_i = s dT + v dP + RT \ln fX \quad (2)$$

where s is entropy, v specific volume, X concentration, f activity coefficient, and R the gas constant.

In minerals of variable composition other chemical components may alter the lattice dimensions and properties of bonding, and thus the lattice energetics and according to (2) and (1) the affinity of a mineral phase for a particular chemical species.

The equilibrium distribution of Fe and Mg between different coexisting minerals must conform with the requirement imposed by (1) and can be described by the distribution coefficient (K_D) defined as:

$$K_{D_{Fe}}^{ab} = \frac{X_{Fe}^a}{X_{Mg}^a} \frac{X_{Mg}^b}{X_{Fe}^b} = K_a \exp(-\Delta G_{(4)}/RT) \quad (3)$$

where X are the mole fractions in phases a and b , K_a is an activity coefficient term and $\Delta G_{(4)}$ is the increase of free energy for the exchange reaction



In case of ideal distribution the term K_a is equal to unity and the distribution coefficient is a constant for any constant combination of T and P . If the lattices of two coexisting mineral phases are crystal-chemically similar, the effect of changing P — T conditions will be more or less inefficient as a grade indicator. This is the case *e.g.* in coexisting biotite and Ca-amphibole where the response of iron-magnesium K_D to changing P and T is much weaker than its variation as a function of (Si, Al, and Ti-) substitutions in tetrahedral or octahedral sites.

To survey compositional and environmental factors influencing the composition of biotites in granites and metamorphic rocks we will next consider the distribution of Fe and Mg between biotite and other relevant minerals.

For the purpose of this paper K_D for coexisting biotite and Ca-amphibole can be regarded as being approximately unity, which implies that Ca-amphibole coexisting with biotite at equilibrium will have little effect on the biotite Fe/Mg parameter. Because M_4 sites in orthoamphibole

take Fe in preference to Mg, the distribution of iron is shifted somewhat in favor of that mineral. Nevertheless, the shift from unity is comparatively small, the distribution coefficient $K_{D_{Fe}}^{bl-a}$ being approximately 1.2—1.4 in the X_{Fe} concentration range considered here (Gorbatshev unpublished data).

Other important ferromagnesian minerals commonly coexisting with biotite are garnet, cordierite, and chlorite. Pyroxene and Fe—Mg spinels are of considerably less importance whether granites or granitization products are concerned. Staurolite and chlorite may occur in large quantities, but are for most relevant Fe/Mg proportions restricted to low-grade rocks. Since staurolite carries iron in 4-coordination, which is a type of lattice site not easily occupied by Mg in silicates, the distribution of Mg is anticipated to be strongly in favor of biotite as is confirmed by Green's (1963) and Hounslow and Moore's (1967) data, the latter giving $K_{D_{Fe}}^{staur-bl} \approx 4.0$.

Somewhat higher $K_{D_{Fe}}$ (4 to 6) have been found for coexisting Mg—Fe—Al spinel and biotite (Gorbatshev unpublished data). The variation of K_D with temperature has not been investigated, but may be expected to be prominent. Due to the uncommonness of spinel — which is often stabilized by small amounts of Zn — and for reasons of the restricted P — T field of staurolite occurrence, K_D variations with temperature are of little concern here. Data on biotite-chlorite pairs are scarce and not wholly unambiguous. Because of the general similarity of the biotite and chlorite lattices, K_D does probably not depart very much from unity which is in accord with the actinolite-chlorite data of Ramberg (1952). Extensive possibilities of intramineral element substitutions indicate susceptibility of $K_{D_{Fe}}^{bl-chl}$ to compositional influences.

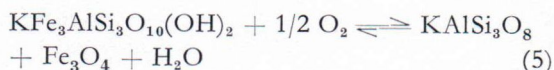
The distribution of Fe and Mg between biotite, cordierite, and garnet is well elaborated. Cordierite has long been known to be a more mag-

nesiophile mineral than garnet (Folinsbee 1941) and accordingly we find $K_{D_{Fe}}^{bl-cord} \approx 1.8$, and $K_{D_{Fe}}^{gt-bl}$ ranging between approximately 9 and 2 (Gorbatshev 1968, Saxena 1968 a, and numerous other papers). Because of the basically different crystal lattices of garnet and biotite we may anticipate a different response to changing temperature, which is confirmed by $K_{D_{Fe}}^{gt-bl}$ being approximately 9.0 in epidote-amphibolite and 5.6 in upper amphibolite to granulite facies (Saxena 1968 a and b). Studies of coexisting garnets and biotites in the Svecofennian basement of Finland and Sweden suggest the following data: transition epidote-amphibolite to amphibolite facies ~ 7 to 7.5, transition between andalusite and sillimanite grade at pressures of regional metamorphism in buried geosynclinal sediments (600°—630°C, Althaus 1967) ~ 6 , uppermost amphibolite- hornblende granulite facies 3—4, aggregation of veins in veined gneisses into independent pegmatite-granite bodies 3—3.5. The highest values yet recorded ($K_D \leq 2.5$ —3) are from intensely granite-veined gneisses and some granulites (Gorbatshev unpublished data, Kostyuk 1955, and inferences from Eskola 1952). The above data refer to garnets low in Ca and Mn. The Fe/Mg distribution between garnet and biotite is sensitive to variations of the Mn-concentration in garnet (Kretz 1959, Saxena 1968 b), to Al, Ti, and possibly the occupancy of octahedral layers in biotite, and may be somewhat non-ideal (Dahl 1969, Saxena 1969, Gorbatshev 1968). Except for cases of extremely Mn-rich garnet all these factors are numerically of less importance than the dependence on temperature, the effect of pressure being slight (Kretz 1963).

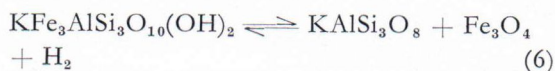
We conclude from the above that as far as the distribution of iron between biotite and coexisting ferromagnesian silicates is concerned, low-oxidized granite biotites are apt to represent closely the whole-rock silicate-iron/magnesium ratio, while the nature of coexisting mineral phases is of paramount importance in many

metamorphic rocks. In aluminous rocks the effects of garnet and staurolite combine to produce »too low« iron in biotites as compared to the anticipated biotite compositions in granitization products which have resulted from the dilution of Al by alkalis, but preserve the Fe/Mg ratio basically unchanged. This trend is to some extent counterbalanced or may even be outweighed by cordierite in extremely aluminous and rather magnesium-rich paragneisses. Naturally the numerical relation between the Fe/Mg composition of biotites and the whole-rock Fe/Mg ratio will depend on the absolute amounts of the different ferromagnesian phases, which are controlled not only by metamorphic grade (\sim temperature) and the Fe/Mg ratio in the rock, but also by the availability of *e.g.* Ca and Mn — which stabilize garnet — and H₂O and Ti which stabilize biotite and may increase the variance of the whole system.

In addition to rock bulk composition and the nature of coexisting phases the Fe/Mg ratio in silicates is highly sensitive to the oxidation potential of the environment. For the decomposition reaction of biotite:



which can also be written:



the condition of equilibrium is:

$$\Delta G_{(6)} = 0 = G_{KfsP} + G_{mt} + \mu_{H_2} - \mu_{ann} \quad (7)$$

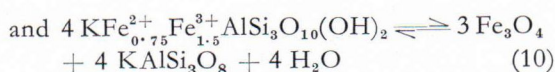
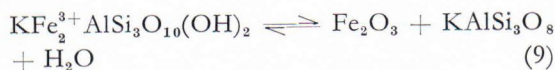
and since $\mu_{ann} = G_{ann} + RT \ln f_{ann} X_{ann}$,

we obtain by transforming:

$$X_{ann} = \frac{1}{f_{ann}} \exp \left(\frac{\Delta G_{(6)}^\circ + \mu_{H_2}}{RT} \right), \quad (8)$$

which implies that the mol fraction of annite in biotite solid solution is a function of temperature, pressure, and the chemical potential of H_2 . At constant temperature and pressure the oxidizing conditions of geological environments thus impose restrictions on the maximum possible iron contents of biotite. Exhaustive theoretical and experimental consideration of Fe-biotite stability is given by Wones and Eugster (1965).

High activity of H_2O promotes the formation of biotite and from reactions of the following type:



$$\text{we find } X_{\text{ferribi}} = \frac{1}{f_{\text{ferribi}}} \exp\left(-\frac{\Delta G^\circ_{(9)} + \mu_{H_2O}}{RT}\right) \quad (11)$$

$$\text{and } X_{\text{bi}_{(10)}} = \frac{1}{f_{\text{bi}_{(10)}}} \exp\left(-\frac{\Delta G^\circ_{(10)} + 4\mu_{H_2O}}{4RT}\right) \quad (12)$$

which informs us that the composition of biotite is a function of the chemical potential of H_2O .

For sulphidization we similarly find restrictions on maximum possible iron in silicates as functions of μ_{H_2S} , μ_{S_2} , and μ_{SO_2} .

Mineral data

To survey the parentage of biotites rich and intermediate in iron, which have been claimed to define the fields of igneous granites and metamorphic rocks, data from some recently analyzed or published groups of biotites from well-investigated rocks have been plotted in Figs. 1 and 2. From the diagrams in Gokhale's paper (1968) it is evident that the alleged main difference is in the Mg/Fe^{2+} ratio, but in order to allow direct comparisons with previously published data and

since iron in many analytical techniques is determined as total Fe, the mode of presentation adopted here is by the kind of diagrams employed by Nockolds (1947). With the possible exception of one sample (Grey Revsund, cf. Gorbatshev 1969) the parent granites of the biotites in Fig. 1 are considered to be »igneous», two of the groups — Råtan and Graversfors — being clearly crosscutting and somewhat later than the main phase of migmatization and associated folding in the concerned areas. Indeed, the Råtan granite is taken to be »older post-orogenic» in its regional context (Lundqvist 1967). The Ben Nevis granites studied by Haslam (1968) are part of a complex quartz-diorite — granite intrusion. The biotites of the Ben Nevis quartz-diorite are similar to the dark mica in the granite, but have not been plotted in Fig. 1 which is limited to rocks of the granite clan proper. The detached left-side pegmatite entry in Fig. 1 represents a rock associated with Grey Revsund granite (Gorbatshev 1969), while the other Svecofennian pegmatite (MgO-proportion in biotite 14.9) is a garnetiferous isolated lump in migmatized terrain. The rest of the pegmatites are Alpine (Schwander *et al.* 1968).

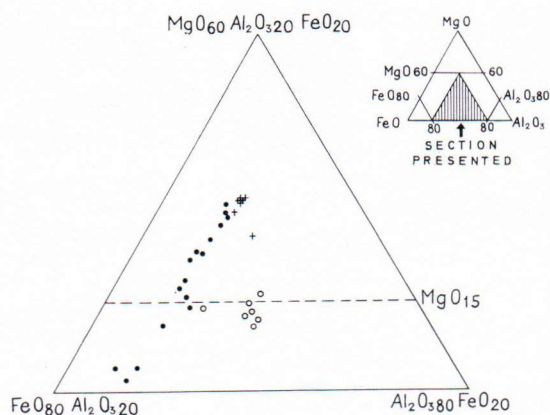


Fig. 1. $MgO-Al_2O_3-Fe_{\text{total}} = FeO$ plot of biotites from granites and pegmatites. Signs: crosses = granites and acid quartz-diorites from Ben Nevis (Haslam 1968), open circles = Alpine and Svecofennian pegmatites (Schwander *et al.* 1968, Gorbatshev unpublished), dots = granites (Gorbatshev 1969). The line at $MgO = 15$ delimits the magmatic field of Gokhale (1968).

Together the data plotted in Fig. 1 combine to show that some of the most characteristically »igneous» granites in the Svecofennian basement of Scandinavia carry biotites well inside the allegedly metamorphic field. Considering two other complexes of igneous granites, total-rock chemical analyzes of Finnish rapakivis suggest high Fe in their biotites, whereas the Dala granites of Sweden, while exhibiting $\text{MgO}/\text{MgO} + \text{FeO}$ ratios of 0.1–0.3 have $\text{Fe}^{3+}/\text{Fe}^{3+} + \text{Fe}^{2+}$ around or above 0.5 (Lundqvist 1967) and the biotites mentioned to occur in these rocks must consequently be highly magnesian. The inclusion of tonalites, quartz-diorites etc, would greatly increase the number of entries in the central and upper parts of Fig. 1.

Figure 2 is a plot of biotites from metamorphic rocks, most of them of clearly evidenced sedimentary derivation. While some of the biotites, particularly those from Moldanubian gneisses in the border area between Czechoslovakia and Germany, are associated with cordierite plus garnet and spinel, most of the Svecofennian biotites have been analyzed for studies of element distribution in garnet and biotite and are associated with garnet only. They may thus be anticipated

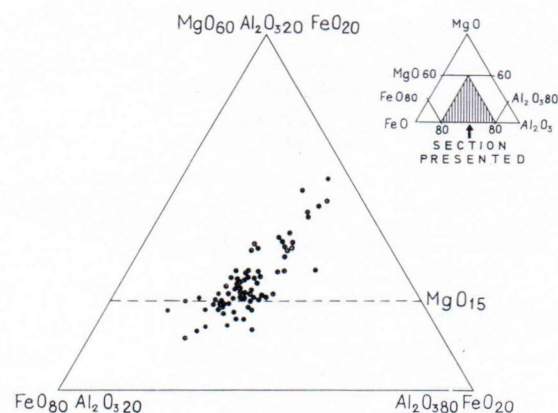


Fig. 2. $\text{MgO}-\text{Al}_2\text{O}_3-\text{Fe}_{\text{total}} = \text{FeO}$ plot of biotites from schists and gneisses. Sources: Butler (1967), Moine schists; Hounslo and Moore (1967), Grenville schists; Schwander, Hunziker, Stern (1968), Alpine schists and gneisses; Gorbatshev unpublished, Svecofennian and Moldanubian gneisses and schists. The line at $\text{MgO} = 15$ demarcates the magmatic field of Gokhale (1968).

to be less ferruginous than biotites in similar rocks below the »garnet-grade» or biotites produced during metasomatic addition of alkalis in granitization. To give the separation into igneous and metamorphic biotite fields the benefit of doubt, no analyzed biotites from Svecofennian leptyte-gneisses (acid metavolcanics and meta-arkoses) have been included. Generally these rocks have somewhat higher Fe/Mg ratios than schists, metagreywackes, and argillaceous paragneisses.

It must be emphasized here that neither Fig. 1 nor Fig. 2 purport to represent a statistically as unbiased as possible review of the compositional variations in biotite as depending on rock parentage. The aim is here not to investigate the averages and variances of biotites in different groups of rocks, but rather to consider the problem of whether there is an unequivocal separation into paragneiss and orthogneiss fields of biotite composition on the basis of MgO contents, which could provide a provenance indicator to be used without excessive reservations and hesitation. Biotites from rock groups not relevant for the tackling of this problem have been deliberately excluded. Plotting all available granites and all available paragneisses would demonstrate a statistically significant difference between the group averages. Again, it should be noted that the inclusion of all metamorphic rocks, para- as well as orthogneisses of volcanic and plutonic provenance, tends to obliterate the differences between biotites of metamorphic-metasomatic and igneous origin. Actually, a consideration of all metamorphic rocks is of greater interest when dealing with the general composition of granitization products than a restriction to metaargillites only.

Discussion and conclusions

Factors which control the composition of biotites have been discussed in the preceding sections. In anhydrous silicates coexisting with iron

oxides the $\text{Mg}/\text{Mg} + \text{Fe}^{2+}$ ratio permits the determination of univariant $p_{\text{O}_2}^*$ ($p^* = \text{fugacity}$) conditions. The controls of $\text{Mg}/\text{Mg} + \text{Fe}^{2+}$ in biotite are described by reaction (5), and the decisive factor is the relation between the chemical potentials of oxygen and water, which can be rewritten to give (8). Because chemical analyses indicate that biotite accommodates ferric iron as »oxybiotite» plus di-octahedral »ferri-mica» end-members, the $\text{Mg}/\text{Mg} + \text{Fe}_{\text{total}}$ ratio will vary as a function of water fugacity as suggested by equations of type (11) and (12). At high hydrogen fugacities all ferric iron may be accommodated in biotite (and other silicates) and the $\text{Mg}/\text{Mg} + \text{Fe}_{\text{total}}$ ratio will reflect rock bulk composition and the distribution of iron between silicate phases. The critical assemblage which permits the determination of univariant $p_{\text{H}_2}^* - T$ conditions of equilibrium from biotite compositions must thus include all the solid components of reaction (6). In other cases biotite composition only indicates lowest possible $p_{\text{H}_2}^*$ at known T .

Relations between the oxidation state of iron and X_{Mg} in whole-rock and biotite analyses of some granites are given in Table 1. All included biotites coexist with amphibole. Rocks of the Råtan group and granites L 55 and 15 558 in

addition contain magnetite (plus microcline and quartz). Disregarding possible effects of substitutions other than $\text{Mg} \rightleftharpoons \text{Fe}$, the Fe/Mg ratio of the ferromagnesian silicate phases in these samples is determined by $p_{\text{H}_2}^* - T$. In the other granites magnetite is absent or found in trace amounts in cracks, and may not be in equilibrium with the ferromagnesian silicates. As demonstrated previously, the occurrence of coexisting amphibole influences biotite composition but slightly, and the low-oxidized samples show close agreement between $\text{Mg}/\text{Fe}_{\text{total}}$ in biotite and bulk analyses. High Fe in biotite 15 299 is probably due to coexisting chlorite which takes somewhat more Mg than does biotite.

Reverting to the composition of biotites in specific groups of rocks, we infer from the averages compiled by Pettijohn (1957) that sedimentary differentiation is usually not very efficient in separating Fe and Mg in greywackes, subgreywackes, and shales, while arkoses and orthoquartzites are strongly enriched in iron. To some extent these differences may result from the composition of source-rocks and particularly the influx of ophiolitic-spilitic basic material into the provenance areas of the greywacke suite. In rocks where ferromagnesian silicates do not coexist with iron oxides the composition of biotites depends on rock composition and inter-mineral element distribution. The data in Fig. 2 demonstrate that $p_{\text{H}_2}^*$ is in many cases sufficiently

high to allow the formation of iron-rich biotites. We conclude that the formation of »pseudoigneous» plutonics by a metamorphic-metamorphic process which, as suggested by Peikert (1964), does not affect original differences in oxidation, must result in a range of different oxidation states and in biotites with different Fe/Mg ratios. As can be inferred from Fig. 2, some of these will be low in Mg. In the rocks investigated by Peikert (1964) R.I. (= refractive index) of biotite had been measured to determine the oxidation state *i.e.* more precisely $p_{\text{O}_2}^*/p_{\text{H}_2\text{O}}^*$ at known or estimated temperature. Here the

TABLE 1

Some total-rock and biotite parameters of analyzed granites.

Specimen nr. (Gorbatshev 1969)	Whole rock			Biotite
	Fe^{3+}	Mg	Mg	Mg
	$\text{Fe}^{3+} + \text{Fe}^{2+}$	$\text{Mg} + \text{Fe}_{\text{total}}$	$\text{Mg} + \text{Fe}^{2+}$	$\text{Mg} + \text{Fe}_{\text{total}}$
Råtan granite				
66/198	0.46	0.40	0.55	0.59
66/42	0.44	0.39	0.53	0.57
15286	0.45	0.38	0.52	0.55
66/122	0.36	0.34	0.44	0.46
66/104	0.37	0.34	0.45	0.45
66/87	0.42	0.32	0.44	0.51
Other granites				
15298	0.18	0.32	0.37	0.32
L 55	0.29	0.31	0.38	0.35
15558	0.21	0.26	0.30	0.25
15299	0.14	0.15	0.17	0.10
15293	0.13	0.09	0.11	0.10

strongest argument in favor of non-igneous formation is the areal pattern of R.I. distribution rather than the R.I. measure by itself. Further, Peikert's rocks are granodiorites, quartz-diorites, and quartz-monzonites, not granites *sensu stricto*, and the inclusion of igneous counterparts of such rocks in Fig. 1 would place most of them in the field alleged to be characteristic of metamorphic provenience. A good example are rocks of the well-investigated South California batholith (Larsen 1948, Larsen and Draisin 1948) which show enrichment of iron in biotites of late granites, but where the biotites of tonalites and granodiorites have $MgO \geq 20$ in terms of the diagram type employed in Figs. 1 and 2. Similar trends appear to prevail in the Early Svecofennian suites of differentiated plutonics in Fennoscandia and in members of the postorogenic Rapakivi suite. Incidentally biotite compositions in these rocks must imply that $p_{H_2}^*/p_{O_2}^*$ increases as differentiation proceeds. Again, there are groups of rhyolitic effusives and »intrusive» granites which have low bulk $Fe/Fe + Mg$ and thus necessarily must contain magnesian biotites, no matter what the oxidation state. Characteristic instances in Sweden are some »hällflinta» volcanics (Sundius 1923) and granites of Råtan, Filipstad, and Järna types. As mentioned previously the latter rocks behave clearly intrusively into their environment, even cutting across boundaries between diversely metamorphosed terranes allegedly belonging to different »orogenic belts». From the general homogeneity and the pattern of subordinate compositional variations, they are not metamorphic-metasomatic in the sense of having been produced *in situ*. It may be noted that these groups characteristically are poor in associated gabbroid differentiates. It is, however, outside the scope of this paper to discuss their origin beyond the statement of extreme improbability of *in situ* granitization. Undifferentiated synkinematic Late-Svecofennian granites associated with regional migmatization (the Stockholm group and allied types) are com-

monly reckoned as formed by a combination of granitization *in situ*, partial melting, and igneous intrusion. Since the numerous older analyses of these rocks usually do not differentiate between different modes of origin, they are of little value in the present context. From analyses in *e.g.* Stålhös' paper (1962) it can nevertheless be concluded that most varieties, regardless whether of granitization or locally intrusive »igneous» origin, are too high in Mg/Fe to qualify for the »igneous» field. The same also applies to isolated massives of similar granites intruding into low-grade terrains outside of the main belt of folding.

To sum up, biotites coexisting with other ferromagnesian minerals are valuable checks of equilibrium distribution of elements and can in some instances, *e.g.* in association with garnet or titanium oxides, be employed as geological thermometers. In mineral assemblages which include iron oxides the $Mg/Mg + Fe^{2+}$ parameter of biotites defines the univariant $p_{H_2}^*-T$ conditions of equilibrium. However, the chemical composition of biotites is no good general indicator of granite formation by either »igneous» intrusion or metamorphism. Exceptions in relation to most metamorphic terrains may be the extremely iron-rich late acid members of differentiated plutonic suites. The salient difference is here one of bulk X_{Fe} rather than state of oxidation. From studies of intermineral distribution of elements it is evident that granites cannot produce biotites which greatly exceed the rock Fe/Mg , no matter what the $p_{H_2}^*$ status. The defining characteristic is thus a matter of whole rock rather than biotite composition. While there may be differences between the average compositions of biotites from metamorphic rocks and from igneous granites, the comparison of these measures has little relevance in considering the critical area of biotites poor and intermediate in Mg . Most of the extremely magnesian metamorphic biotites stem from highly oxidized iron-formations, which are either quantitatively un-

important or uncontroversial from the point of view of granitization.

Basically, the problem of iron-magnesium relations in biotite can be subdivided to concern the Mg/Fe^{2+} and $\text{Mg}/\text{Fe}_{\text{total}}$ parameters, respectively. At low $p_{\text{H}_2}^*$ both will be functions of the oxidation state. At high $p_{\text{H}_2}^*$ the Mg/Fe^{2+} ratio will still be a measure of oxidation, while $\text{Mg}/\text{Fe}_{\text{total}}$ may become a function of rock composition and element distribution. If the critical difference between two rock groups is in the bulk contents of Fe and Mg, then $\text{Mg}/\text{Fe}_{\text{total}}$ will be the proper measure to investigate. However, for this purpose and to evaluate the amount of Al, bulk analyses of rock specimens are the rational approach because they provide more relevant information on these particular aspects and in most analytical procedures involve less effort than the separation and analysis of minerals. If

again, $p_{\text{H}_2}^*$ is assumed to be the critical factor, the proper measures are Fe^{2+}/Mg or $\text{Fe}^{2+}/\text{Fe}^{3+}$. Investigation of these parameters in minerals is of utmost importance for the understanding of geological processes. However, in the present case, differences in biotite oxidation state have not been claimed to be a critical point in distinguishing between granites of different origin (Diagram 1 in Gokhale 1968).

Acknowledgments — Numerous chemical data quoted in this paper have been obtained by analytical work on the Geoscan electron probe at the Dept. of Geology in Uppsala, sponsored financially by grants from Malmfonden (87) and Naturvetenskapliga Forskningsrådet (nr. 2392—17).

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Manuscript received, April 15, 1969.