

GEOLOGICAL NOTES

SODIC AMPHIBOLES AS OXYGEN FUGACITY INDICATORS IN METAMORPHISM

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ABSTRACT

Analyses of sodic amphiboles from a blueschist metamorphic terrain of constant grade in the Tavşanlı area, Northwest Turkey, show that practically the whole range of sodic amphibole compositions is stable in the glaucophane-lawsonite facies. By analysing sodic amphiboles coexisting with magnetite and hematite, it has proved possible to plot for a constant pressure and temperature an oxygen fugacity isopleth. Through the analyses of sodic amphiboles and reference to the extended oxygen fugacity contours drawn on the Miyashiro compositional diagram, relative rock oxygen fugacities can be obtained. The oxygen fugacity isopleth corresponding to the magnetite/hematite buffer from two blueschist terrains with differing P-T regimes, namely Tavşanlı in Turkey and Shuksan in Cascades, lies along the same loci on the amphibole diagram indicating that the intrinsic fO_2 slope of sodic amphiboles in the fO_2 -T plane is approximately the same as the fO_2 slope of the magnetite/hematite buffer. Changes in the fO_2 during metamorphism may be recorded as zoning in sodic amphiboles, two examples of which are given.

INTRODUCTION

Sodic amphiboles are important as indicators of high pressure and low temperature conditions in metamorphism. They occur in a variety of rock types in the blueschist facies, becoming sporadic with decreasing P/T towards the greenschist facies. Among the major mineral species sodic amphiboles are unique in having independently variable ferric/ferrous ratios, which can have any value between zero and infinity, making their compositions highly sensitive to oxygen fugacity during metamorphism.

In this paper the relationship between the composition of sodic amphibole and oxygen fugacity is studied with the aim of using sodic amphibole compositions as oxygen fugacity indicators. The lack of reliable thermodynamic and experimental data for sodic amphiboles justifies an empirical approach, based on the

study of phase relations and mineral compositions from blueschists.

METAMORPHIC ENVIRONMENT AND PETROGRAPHY

In Northwest Turkey blueschists form an extensive and coherent metamorphic belt flanking the northern margins of the ultrabasic massifs along the Izmir-Ankara ophiolite zone (Brinkmann 1976, p. 84-87). Sodic amphibole compositions reported here come from a small area (6 by 18 km) in the district of Tavşanlı (Okay 1978). No change in grade is detectable across the sampled area. The dominant rock types in the region are metabasites and metacherts. In the metabasites the essential mineral assemblage is: sodic amphibole + lawsonite \pm sodic pyroxene \pm chlorite + phengite + sphene. Compositions of sodic pyroxenes from the metabasites are given in an earlier paper (Okay 1978). The common mineral assemblage in metacherts is: sodic amphibole + garnet + lawsonite + hematite + quartz + phengite, with sporadic epidote and sodic

¹ Manuscript received June 29, 1979; revised October 29, 1979.

[JOURNAL OF GEOLOGY, vol. 88, p. 225-232]

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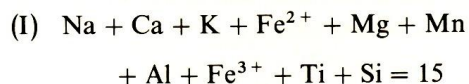
pyroxene. Phase relations of these rocks will be described in a separate paper.

After lawsonite, sodic amphibole is the most common and abundant mineral in the blueschists from the Tavşanlı area and occurs in all rock types in varying amounts. In metabasites, sodic amphibole together with lawsonite make up 80 to 90% of the mode; in metacherts it is the principal ferromagnesian mineral.

METHOD AND FERRIC/FERROUS RECALCULATION

The mineral compositions were determined for twelve elements (Na, Ca, K, Fe, Mg, Mn, V, Ni, Cr, Ti, Al, Si) using an electronprobe microanalyser with a Harwell Si(Li) detector and pulse processor (Statham 1976). The correction procedures are given by Sweatman and Long (1969). Olivine and jadeite standards were used throughout. The relative accuracy is estimated at 2% for more than 5 wt% of the element present.

The amount of ferric ion in sodic amphiboles is estimated according to the following assumptions.



The model assumes that the total anion charge is equal to 46-, corresponding to 22 oxygens and 2 hydroxyl groups and that there is no A-site occupancy. The total for alkali ions (Na + Ca + K) in the formula unit, calculated assuming 46- charge, lies between 1.90 and 2.10 suggesting very little if any A-site occupancy. This has also been shown by wet chemical analyses on separated samples from other blueschist terrains (Coleman and Papike 1968). In cases where Na + Ca > 2.08, the amount of ferric and ferrous iron was also calculated assuming $Fe^{2+} + Mg + Mn + Al + Fe^{3+} + Ti + Si = 13$ and an intermediate value between the two calculations is chosen. Consistency between the recalculated amphibole analyses is considered to be most important; in all recalculated sodic amphibole analyses $Fe^{3+} + Al$ per formula unit ranges between 1.80 to 2.00 and $Mg + Fe^{2+} + Mn$ between

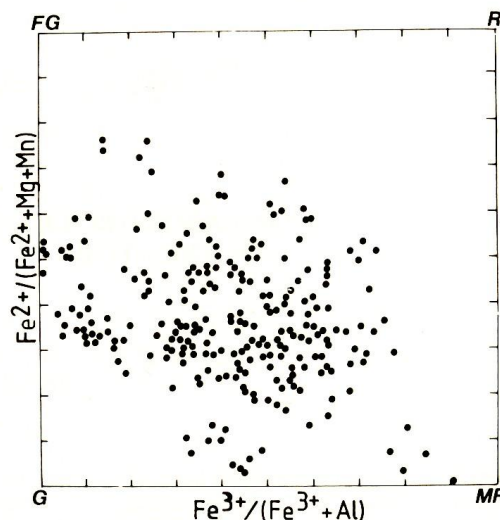


FIG. 1.—Compositions of sodic amphiboles from the glaucophane-lawsonite facies blueschists of Tavşanlı area, Northwest Turkey. G: glaucophane, FG: ferroglaucofane, R: riebeckite, MR: magnesio-riebeckite.

2.85 to 3.15, depending on the amount of Ca present. Calculations indicate that small variations in the total anion charge, due to the oxidation of the hydroxyl groups, will not have a strong effect on the ferric/ferrous ratio and are well within the compositional variation shown by most natural "homogeneous" sodic amphiboles in a single thin section. The results obtained using the above model compare well with the ferric/ferrous determinations done using wet chemical methods (Brown 1974, p. 334).

COMPOSITIONS OF SODIC AMPHIBOLES

243 complete electron microprobe analyses of sodic amphiboles from 47 Tavşanlı blueschist specimens are plotted on the diagram of Miyashiro (1957) in figure 1. A minimum of five, and for blueschists with zoned sodic amphiboles as many as fifteen analyses have been performed for each rock. The miscibility of the actinolite component in sodic amphibole in the Tavşanlı blueschists is limited and all sodic amphibole analyses plotted in figure 1 have Na/(Na + Ca) ratios in the formula unit greater than 0.9. Calcic amphibole was not observed either in metabasites or metacherts.

Sodic amphibole compositions are expected to extend towards riebeckite and ferro-

glaucophane end-members in ferrous-rich (stilpnomelane- or deerite-bearing) rocks, absent in the Tavşanlı area. Figure 1 suggests that most of the compositional field of sodic amphiboles with the possible exception of compositions close to the glaucophane end-member, is stable under the P-T conditions of the glaucophane-lawsonite facies.

SODIC AMPHIBOLES AS OXYGEN FUGACITY INDICATORS

The oxygen fugacity of an oxygen-bearing solid phase of variable composition, at a specified exact composition, is its intrinsic oxygen fugacity. It is a function of temperature and total pressure and is directly related to the chemical potential of oxygen within the phase of this specified composition (Sato 1972, p. 290). The concept of intrinsic fO_2 may easily be understood with reference to the free energy-composition diagram after Sato, shown in figure 2. In the diagram it is assumed that a metal forms a stable oxide MO with an appreciable range of composition. The chemical potential of oxygen, hence fO_2 , is determined by the exact composition of MO and vice versa. The fO_2 value, determined by a tangent to the free energy curve at point "a" is the intrinsic fO_2 of the oxide at composition A. At composition B, the intrinsic fO_2 of MO is equal to the equilibrium fO_2 of the three phase assemblage M-MO-gas. The relation between fO_2 and composition is unique in the case of a binary oxide. In oxide phases of more complex composition a range of compositions corresponds to each value of fO_2 , though each composition corresponds to only a single value of fO_2 . An important point is that the intrinsic oxygen fugacity is a quantity definable in terms of composition, temperature, and total pressure; it is independent of specific mineral assemblage and rock composition.

In natural assemblages minerals with widely variable ferric/ferrous ratios will be suitable for measuring intrinsic oxygen fugacities. The variable ferric/ferrous ratio and extensive compositional range shown by sodic amphiboles open the possibility of using their composition as oxygen fugacity indicators, provided the dependence of composition on fO_2 , T and P is known.

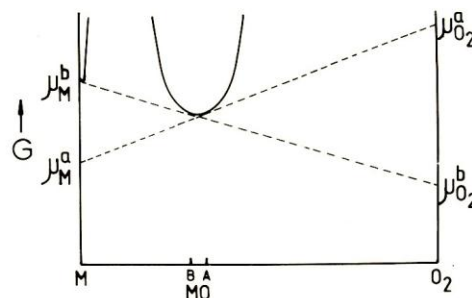


FIG. 2.—Free energy(G)-composition diagram for a hypothetical binary system M-O at constant temperature and pressure, modified after Sato (1972).

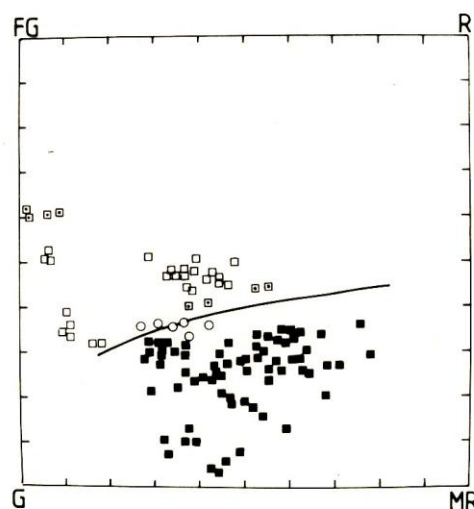


FIG. 3.—The compositions of sodic amphiboles from the Tavşanlı area coexisting with hematite (filled squares), hematite and magnetite (open circles), and magnetite (open squares). Sodic amphibole compositions from magnetite-bearing blueschists from Gümele are indicated by open squares with a dot. Abbreviations are as figure 1.

Sodic amphibole compositions from 16 iron-oxide bearing specimens from Tavşanlı and three from Gümele, an area of similar metamorphic grade 50 miles east of Tavşanlı (Lünel 1967), are plotted in Fig. 3 in relation to the coexisting iron-oxides. Iron-oxides were identified in polished sections under reflected light and their compositions were checked by electron microprobe. Hematites from the Tavşanlı blueschists contain low amounts of TiO_2 (up to 4 wt%), so that hematite and

magnetite depart insignificantly from the system Fe-O.

Sodic amphiboles coexisting with hematite and magnetite should have compositions controlled by the oxygen fugacity of this buffer. Although only a single specimen (K187, table 1) was found with this sub-assemblage, sodic amphiboles coexisting with hematite or magnetite bracket closely the compositions which define oxygen fugacities corresponding to the hematite/magnetite buffer (fig. 3).

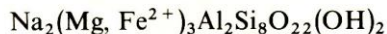
Oxygen fugacity contours.—A set of constant oxygen fugacity lines (oxygen fugacity isopleths) can be constructed in a qualitative way on the sodic amphibole compositional field (fig. 4). During the construction of the grid the following points have been considered:

1. Iron-free sodic amphibole with the end-member glaucophane composition



will be stable over the whole range of oxygen fugacities in geological environments. Consequently all oxygen fugacity isopleths will converge towards and pass through the glaucophane end-member (Muir Wood 1977).

2. Ferric-free



(glaucophane-ferroglaucophane) and ferrous-free $\text{Na}_2\text{Mg}_3(\text{Al, Fe}^{3+})_2\text{Si}_8\text{O}_{22}(\text{OH})_2$ (glaucophane-magnesioriebeckite) sodic amphiboles will be stable over a limited range of oxygen fugacities. The situation is analogous to other ferrous- or ferric-bearing minerals e.g. annite (Eugster and Wones 1962) or hematite.

3. A sodic amphibole of the general composition $\text{Na}_2(\text{Mg, Fe}^{2+})_3(\text{Al, Fe}^{3+})_2\text{Si}_8\text{O}_{22}(\text{OH})_2$ making up the bulk of the naturally occurring sodic amphiboles, will be stable only at a single value of oxygen fugacity at a given P-T.

4. The sodic amphibole compositions from hematite + magnetite assemblages, defining a constant oxygen fugacity provide a reference line in the construction of an isopleth set. These isopleths are assumed to be sub-parallel to this fixed reference line, converging towards the glaucophane end-member composition (fig. 4).

Figure 4 shows an empirical set of relative oxygen fugacities plotted on the Miyashiro

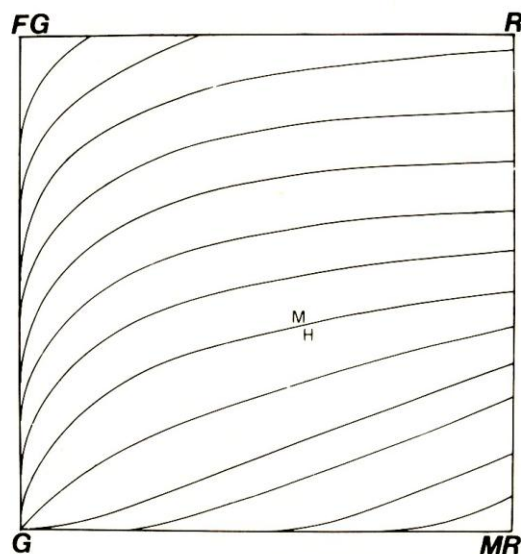


FIG. 4.—A set of oxygen fugacity isopleths for the P-T conditions of glaucophane-lawsonite facies metamorphism in the Tavşanlı area, Northwest Turkey. The isopleths represent increasing oxygen fugacities from ferroglaucophane to magnesioriebeckite. The line marked by the letters H and M corresponds to the $f\text{O}_2$ values of the hematite/magnetite buffer.

sodic amphibole diagram. The actual values of sodic amphibole intrinsic oxygen fugacities will be dependent on the pressure and temperature. The intrinsic $f\text{O}_2$ values for sodic amphiboles coexisting with magnetite + hematite in the Tavşanlı area can be calculated from the estimated P-T values of metamorphism (for $T = 325^\circ\text{C}$ and $P = 8\text{ kbars}$ $\log f\text{O}_2 = -27$, Okay in prep.). However other reference points for the intrinsic oxygen fugacity values of sodic amphiboles, allowing calibration of the isopleths in figure 4, are unavailable. Fayalite is not found in the blueschist facies and there is no information on the stability range of magnetite + quartz + sodic amphibole assemblage. Ernst's synthetic work on magnesioriebeckite and riebeckite (1960; 1962) must be considered suspect since he claims to have synthesized both of these end-members, representing widely differing oxygen fugacities, under a common magnetite/hematite buffer. Hoffmann (1972) found that natural ferroglaucophane is stable at 5 kbars and 350°C under the oxygen fugacities of the magnetite/wustite and magnetite, quartz/fayalite buffers but breaks down under the hematite/magnetite

buffer. No attempt was made to determine the upper oxygen fugacity stability limit of ferro-glaucophane. The absence of any reliable information prevents the quantification of oxygen fugacities in figure 4.

The overall pattern of the fO_2 isopleths in figure 4, constructed for the P-T conditions of the Tavşanlı glaucophane-lawsonite facies rocks, should remain the same for other blueschist terrains, although the actual oxygen fugacity values may differ. Using this set it is possible to abstract the relative oxygen fugacities in rocks, with differing bulk rock compositions and mineral assemblages, from their sodic amphibole compositions.

It is important to realize that sodic amphibole composition will be a function of oxygen fugacity and not of oxidation ratio, which is dependent on the modal amounts of iron-bearing minerals present in the rock. As shown in figure 4 sodic amphiboles with different ferric/ferrous ratios (oxidation ratios) can have the same oxygen fugacity.

It is apparent from figure 4 that sodic amphiboles and consequently blueschists at the glaucophane-lawsonite facies conditions are stable over a wide range of oxygen fugacities, corresponding to the hematite and magnetite stability fields. Any attempt to synthesize sodic amphiboles outside their oxygen fugacity stability limits, e.g. magnesioriebeckite at the magnetite/wustite buffer (Ernst 1960) or riebeckite at the hematite/magnetite buffer (Ernst 1962), will result in the oxidation or reduction of the iron, and reaction products which deviate from the sodic amphibole composition.

EFFECT OF PRESSURE AND TEMPERATURE ON THE INTRINSIC OXYGEN FUGACITIES OF SODIC AMPHIBOLES

The intrinsic oxygen fugacity of a sodic amphibole of fixed composition will be a positive function of total pressure and temperature. However the effect of pressure on oxygen fugacities is in general negligible, compared to the effect of temperature (Eugster and Wones 1962, p. 90-92), and pressure will be neglected in the following discussion. The temperature dependence of sodic amphibole intrinsic oxygen fuga-

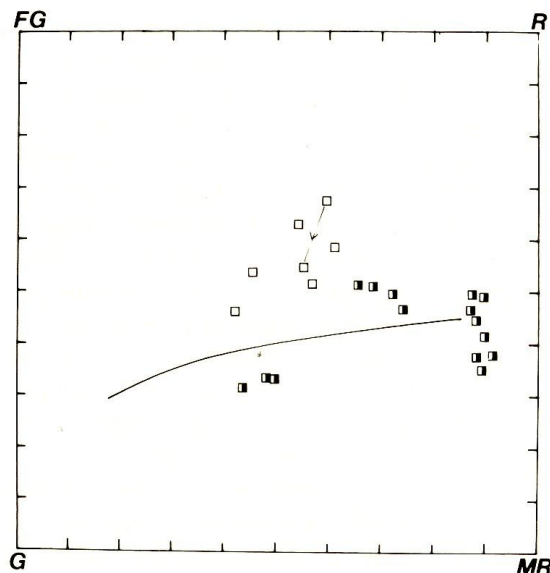


FIG. 5.—The compositions of sodic amphiboles from Shuksan coexisting with magnetite (open squares) and magnetite + hematite (half filled squares). The fugacity isopleth for hematite + magnetite bearing assemblages from Tavşanlı area is also indicated. The arrow points from core to rim compositions. Abbreviations are as figure 1.

city can be obtained qualitatively by comparing sodic amphibole compositions and coexisting iron-oxides from different blueschist terrains. The Shuksan metamorphic unit in the North Cascades of Washington is the only other blueschist terrain where sodic amphibole compositions have been studied in terms of coexisting iron-oxides (Brown 1974). The Shuksan metamorphism is transitional between blueschist and greenschist facies. Actinolite-albite-chlorite-epidote bearing greenschists are interlayered with blueschists which contain sodic amphibole and iron-oxide instead of actinolite (Brown 1974). The presence of interlayered greenschists and the absence of lawsonite in metabasites indicate that rocks from the Shuksan unit have been metamorphosed at higher temperatures than those from Northwest Turkey.

Sodic amphiboles from three Shuksan blueschists with coexisting magnetite and hematite (15, 27F and 77B) and one specimen with magnetite (18) were analysed. Twentytwo analyses from these four specimens are shown in figure 5 in relation to coexisting iron-oxides.

The two analysed metabasites from Shuksan (18, 77B) show a whole range from calcic to sodic amphibole compositions (Brown 1974, p. 335). To ensure comparability with sodic amphiboles from the Tavşanlı area only sodic amphiboles with $\text{Na}/(\text{Na} + \text{Ca})$ ratios greater than 0.85 per formula unit are plotted in figure 5. The sodic amphibole compositions from magnetite + hematite bearing assemblages from Shuksan show a relatively wide scatter. This may be due to the variable substitution of Ca for Na, Al for Si and the increasing occupation of the A-site. However the isopleth corresponding to the magnetite/hematite buffer from both terrains apparently lies along the same loci on the amphibole diagram indicating that the intrinsic oxygen fugacity slope of sodic amphibole in the $f\text{O}_2$ —temperature plane is approximately the same as the oxygen fugacity slope of the magnetite/hematite buffer.

OXYGEN FUGACITIES DURING THE METAMORPHISM

The discovery of several geological occurrences of closely interlayered metamorphic rocks showing distinct variations in the oxidation state (cf. Chinner 1960), has suggested that in general oxygen is a locally buffered component. The extensive spread of sodic amphibole compositions away from an oxygen fugacity isopleth in a single metamorphic terrain, such as the Tavşanlı area, also suggests that the oxygen fugacity was not externally controlled.

However, the lack of externally controlled uniform oxygen fugacities does not preclude changes in oxygen fugacity in individual rocks during metamorphism, that would be recorded as zoning in sodic amphiboles. Although the sodic amphiboles from the majority of blueschists from the Tavşanlı area show a small scatter of analyses, two examples of large scale zoning have been found that are best explained by changes in the oxygen fugacity during metamorphism:

Specimen K232/1 from Tavşanlı consists of phengite bands with small scattered idioblastic grains of garnet, tabular crystals of lawsonite, prismatic sodic amphibole and rare hematite platelets, all set on a quartz mosaic. Apatite and small wedge-shaped epidote grains also occur in

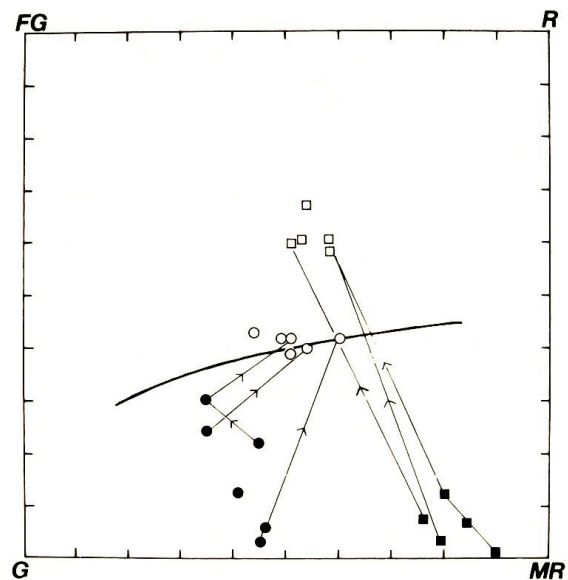


FIG. 6.—Compositions of zoned sodic amphiboles from the specimens K232/1 (circles) and K184 (squares) from Tavşanlı, Turkey; filled symbols: core compositions, open symbols: rim compositions. Lines join compositions from single grains. Abbreviations are as figure 1.

small quantities. Most sodic amphibole grains in the section show distinct zoning with pale blue to colorless cores and dark blue rims. The compositions of the cores varies from grain to grain whereas the rim compositions are more uniform (fig. 6; table 1). Sodic amphiboles of "rim" composition can be seen growing around small hematite laths, which often have sutured edges.

Sodic amphibole rim compositions from K232/1 fall around the magnetite/hematite isopleth on the Miyashiro diagram (fig. 6). This strongly suggests that the incoming fluid, with $f\text{O}_2$ values well within the magnetite stability field, was buffered by the hematite in the rock at the magnetite/hematite buffer values and caused an alteration of the sodic amphibole compositions accordingly. In this case sodic amphibole compositions could not move into the magnetite field without the resorption of all the hematite in the rock (cf. Greenwood 1975). It is important to note that the fluid, although buffered by hematite, caused a change in the sodic amphibole composition. This example illustrates that the relationship between rock and fluid during metamorphism is not a simple

TABLE 1
ELECTRON MICROPROBE ANALYSES OF SODIC
AMPHIBOLES

	K187	K232/1 core	K232/1 rim	K184 core	K184 rim
SiO ₂	57.11	58.12	56.95	56.99	55.58
Al ₂ O ₃	7.64	7.16	5.88	2.79	4.71
FeO*	15.55	11.52	18.39	14.21	23.43
MgO	9.13	11.95	8.11	12.98	5.48
MnO	0.20	0.26	0.12	1.15	0.23
CaO	0.87	0.69	0.26	0.37	0.00
Na ₂ O	6.84	7.28	7.22	7.51	7.56
Total	97.34	96.98	96.93	96.00	96.99
Number of cations per 23 oxygens					
Si	7.99	7.99	8.08	7.95	8.03
Al ^{iv}	0.01	0.01	0.00	0.05	0.00
Al ^{vi}	1.25	1.15	0.98	0.41	0.80
Fe ³⁺	0.70	0.80	0.95	1.56	1.13
Fe ²⁺	1.13	0.53	1.23	0.10	1.70
Mg	1.90	2.45	1.71	2.70	1.18
Mn	0.02	0.03	0.01	0.14	0.03
Ca	0.13	0.10	0.04	0.06	0.00
Na	1.86	1.94	1.99	2.03	2.12

Notes: (1) * Total iron given as FeO

(2) Fe³⁺ recalculated (see text).

(3) K, V, Ni, Cr, Ti below detection limits (< 0.05 wt.%)

control of one by the other but rather a continuous and variable interaction between the two.

In the second example, illustrating reduction during the metamorphism, the process has gone nearer to completion. Specimen K184 from Tavşanlı is a metachert consisting of sodic amphibole, sodic pyroxene, garnet and quartz. Over 90% of sodic amphibole grains have a uniform ferrocrossite composition. However, a few sodic amphibole grains in the section have colorless cores separated by a sharp contact from the dark blue rims (fig. 7). The compositions of the core and the rim (fig. 6; table 1) indicate strong reduction during the later stages metamorphism. The original assemblage must have been highly oxidized with almost all the iron in the ferric state. The uniform garnet compositions (Spess₉₀ Alm₁ Gr₉) with very low almandine contents and sodic pyroxene compositions (Ac₆₅Jd₂₂Di₁₃) with no hedenbergite components are also characteristic of highly oxidized assemblages. Thus the cores and not

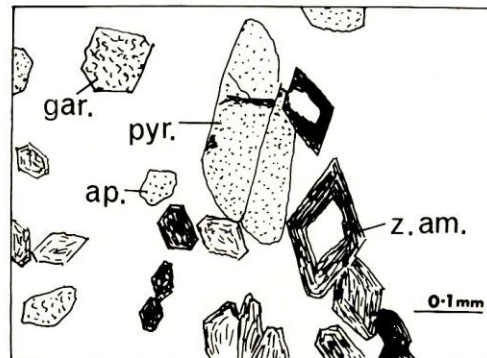
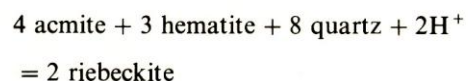


FIG. 7.—Drawing from a photomicrograph of the specimen K184 illustrating zoned sodic amphibole (z.am.) coexisting with garnet (gar.), sodic pyroxene (pyr) and apatite (ap.) all set on quartz.

the rims of sodic amphibole are in equilibrium with garnet and sodic pyroxene. This unusual reversal of textural equilibrium is possibly due to the greater susceptibility of hydrous sodic amphibole to changing oxygen fugacities.

It is noteworthy that in the both of these cases reduction did not simply involve transformation of hematite to magnetite, rather it led to the further crystallization of hydrous silicate phases. This implies that the redox reactions in these examples involved the influx of hydrous fluids. A possible reaction involving the acmite component in sodic pyroxene and the riebeckite component in sodic amphibole is:



Reduction has accompanied hydration; hematite reacting to sodic amphibole rather than forming magnetite. Such reactions suggest the presence of local, mobile, reducing fluids. Under such circumstances oxygen fugacities need not remain constant during metamorphism.

CONCLUDING REMARKS

Sodic amphibole compositions provide a direct means of evaluating and comparing relative oxygen fugacities in blueschist terrains. Published sodic amphibole analyses (Ernst et al. 1970, p. 145; Coleman and Papike 1968, p. 111) indicate that most blueschists have recrystallized in the magnetite stability field,

although there are some interesting regional differences. Metacherts from the Franciscan formation in California contain riebeckitic sodic amphibole coexisting with stilpnomelane (Ernst et al. 1970, p. 200; Coleman and Papike 1968) whereas metacherts from Northwest Turkey contain magnesiochloritoid and hematite. As is clear from these sodic amphibole compositions, this difference between the two chemically similar rock types is largely due to the higher oxygen fugacities during the metamorphism of the Northwest Turkish metacherts. The different oxidation states are probably related to the different depositional environments of the original cherts. Metabasites from both terrains have been metamorphosed under similar oxygen fugacities.

The use of sodic amphiboles as oxygen fugacity indicators is limited toward higher pressures by the restriction of sodic amphibole compositions toward the glaucophane, where the evaluation of fO_2 becomes vague due to limited data and the convergence of fO_2 lines towards glaucophane end-member. The increasing calcic amphibole component with higher T/P provides a further complexity to the elucidation of oxygen fugacities in the intermediate blueschist-greenschist terrains.

ACKNOWLEDGMENTS.—I thank T. J. B. Holland, R. Muir Wood, I. Hutcheon, and E. H. Brown for reviewing the manuscript; E. H. Brown also kindly supplied the blueschist specimens from Shuksan. This work was carried out while in receipt of a grant from the Mineral Research and Exploration Institute of Turkey (MTA) which is gratefully acknowledged.

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