

REFRACTIVE INDEX FIELD DIAGRAMS AND MINERAL
RELATIONSHIPS FOR COEXISTING
FERROMAGNESIAN SILICATES

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ABSTRACT

Over 1600 refractive indices of coexisting ferromagnesian silicates have been used to construct field diagrams by plotting γ of one mineral against that of another. Fields for 2, 3, 4, 5 and 6 coexisting minerals are outlined for definite ranges of indices of each mineral. A separation of igneous and metamorphic rock types is apparent for minerals plotting in the low index range. This appears to bear out the occurrence of a partitioning of Mg and Fe atoms between coexisting ferromagnesian phases and to support the conclusion that the partitioning is a function of the conditions of formation.

INTRODUCTION

Since there is a relationship between the chemical composition of coexisting minerals and the conditions of formation of the assemblage, it seems reasonable to inquire whether there is a corresponding relationship between the more easily obtained refractive indices of coexisting phases and the conditions of formation of plutonic and volcanic rocks. Because the correlation of refractive index and chemical composition is far from simple, this relationship will not be exact, but it might provide a sufficiently useful and simple device for obtaining a first approximation diagram of index and environmental conditions. It was clearly recognized that in making this assumption we were treating a large number of variables in terms of only a small number of parameters, but it was hoped that some significant results might show through the "noise" in the wide scatter resulting from such a procedure.

We have explored a method of investigation or a postulate and obtained both positive and negative results. These results should be made available since variations of the method used have been employed by geologists for different purposes in the past and are being used with increasing frequency.

The composition of coexisting minerals from various types of magmatic rocks should differ from those of rocks formed at other temperatures and pressures, *e.g.*, reconstituted rocks such as are produced by certain kinds of metamorphism, and metasomatic rocks. As far as the ferromagnesian silicates are concerned, a composition variable is the Fe-Mg content, and to a first approximation this controls in large part

the variation in refractive index of ferromagnesian silicates. Therefore, it should be interesting to investigate, phenomenologically and in an exploratory fashion, the relationships that exist with respect to the refractive indices of coexisting ferromagnesian silicates in selected lithologic types, as well as rocks distinguished on the basis of origin, *e.g.*, volcanic and plutonic magmatic rocks.

On the diagrams showing distribution of rock types, all granitic rocks and charnockites are plotted as igneous, although some are undoubtedly metamorphic or metasomatic in origin. Since there are few generally acceptable criteria for the origin of certain igneous and metamorphic rocks, and since igneous and metamorphic processes may converge in deep-seated orogenic regions, the origin of these rocks remains controversial. By plotting all such rocks as igneous, it is impossible to make a meaningful separation of all igneous and metamorphic rocks on the diagrams. Therefore, on separate diagrams (not shown) we plotted isochemical rock series that are generally accepted as being igneous, and series acknowledged to be metamorphic in an attempt to obtain separate fields. However, the number of points obtained from data in the literature was insufficient to produce usable refractive index field diagrams.

Rocks included in this study are phyllites, schists, gneisses, granulites, amphibolites, and hornfels and the normal calc-alkaline igneous suite, including intrusive and extrusive types. Marbles, calcite-bearing gneisses, and Na-rich rocks such as sodalite-, nepheline, and aegerine-bearing syenites have been excluded. Data on coexisting minerals in alkaline rocks are scarce and were not included. The choice of rocks was made solely on the basis of availability of specimens in museum and university collections and on data from the literature. In order to achieve adequate coverage on the field diagrams, as many of the common igneous and metamorphic rocks as possible were selected, with the exceptions noted above. In the interest of completing the study, and after plotting the data, we searched for specimens which represented Mg-rich or Fe-rich environments in order to fill gaps in the diagrams. Approximately two-thirds of the indices recorded in this paper were determined by the authors; the remaining one-third were obtained from the literature. Useful data from the literature are limited because relatively few authors give the number, kind, and indices of coexisting phases from the same specimen and this is essential to our study.

The problem of just what constitutes *coexisting* minerals is a complex one. We have had to assume that the present assemblages of coexisting minerals represent the assemblages of the "original" rocks. Secondarily altered rocks were omitted from this study whenever this could be determined. However, some authors did not indicate whether biotite and horn-

blende were primary or secondary. Since it was impossible to know which data to omit, all were plotted. This means that boundary lines on the diagrams are approximate.

Only rocks containing one or more of the following minerals were investigated: biotite, hornblende, clinopyroxene, and orthopyroxene. Biotite as used includes common biotite, phlogopite, siderophyllite, and lepidomelane. Hornblende includes common hornblende, edenite, pargasite, basaltic hornblende, and hastingsite, but excludes crossite, riebeckite, arfvedsonite, tremolite-actinolite, cummingtonite-grunerite, richterite, glaucophane, and anthophyllite. Tremolite-actinolite, cummingtonite-grunerite and anthophyllite are plotted as separate phases. Clinopyroxene includes diopside-hedenbergite, salite, diallage, leucaugite, augite, ferroaugite, aegirite-augite, and omphacite, but excludes clinoenstatite, schefferite, acmite (aegirite), and pigeonite. Pigeonite is plotted as a separate phase. Orthopyroxene includes enstatite, hypersthene, bronzite, and ferrosilite.

Refractive index field diagrams presented here show the range of indices of biotite, hornblende, clinopyroxene, or orthopyroxene when: (1) each occurs in a rock by itself; (2) they coexist in rocks in various combinations; and (3) they coexist in various combinations with other ferromagnesian silicates. The diagrams also show how these ranges of indices vary from one rock type to another. The petrologic significance of the diagrams cannot be determined until extensive physical-chemical phase studies have been made.

PREVIOUS INVESTIGATIONS

In the past few geologists utilized plots of refractive indices of one mineral against another; Wiseman (1934) may have been the first to do so. Recently, however, such diagrams have been utilized to a greater extent. Seki (1957), Seki and Yamasake (1957), Hashimoto (1957), Miyashiro (1958), Shido (1958), and Wilcox and Poldervaart (1958) show that indices of one mineral have a linear relationship with respect to indices of another mineral. Chinner (1962) plotted γ of cordierite against β of biotite and showed that garnet coexists with cordierite and biotite only when biotite has β greater than 1.660 and when cordierite has γ greater than 1.556.

Other authors have plotted mineral indices against different variables. Tsuboi (1933) plotted refractive indices of biotite and hornblende against An percentage of plagioclase, and Taneda (1942) and Ota (1952) plotted β of orthopyroxene and clinopyroxene against An percentage in coexisting plagioclase. Tsuboi showed that "biotites and hornblendes associated with more calcic (earlier) plagioclases have lower refractive indices, and

those associated with more sodic (later) plagioclases have higher refractive indices." Also, that "biotites in the granites that contain hornblendes have higher refractive indices than biotites in the granites that lack hornblendes." Taneda pointed out that orthopyroxene indices are higher in hornblende andesites than in pyroxene andesites, and Ota showed that indices of hypersthene and augite decrease as the An percentage of plagioclase increases. Yoshizawa (1952) plotted γ of biotite and hornblende against the orthoferrosilite molecular percentage of orthopyroxene. Parras (1958) and Simonen (1960) plotted indices of biotite, hornblende, clinopyroxene, and garnet against Si percentage in rocks and showed changes in mineralogical and chemical composition with changes in rock type. Simonen also showed that biotite indices tend to rise with increasing Si percentage of the host rock. Oki (1961) plotted γ of biotite against An percentage of plagioclase and demonstrated an increase in An percentage with an increase in biotite index. The literature also contains many diagrams on which indices of minerals are plotted against chemical composition (as compiled in Winchell and Winchell, 1951).

METHOD USED

The oil immersion technique was used with both sodium and white light. Precision for the determinations, personally made and from the literature, ranges from ± 0.005 to ± 0.003 ; the overall precision of plotted data is probably near ± 0.003 .

No effort was made to distinguish between β and γ of biotite, since the values are usually within 0.003 of each other. γ was read directly from basal cleavage flakes immersed in oil. Hornblende γ' was obtained on {100} cleavage fragments, recording the highest index of all the grains immersed in the oil. A similar technique was used for clinopyroxene. Determinations of true γ for hornblende and clinopyroxene were not attempted because of the length of time required to find a properly oriented cleavage fragment and because γ' values were systematic and repeatable. Orthopyroxene γ was read directly from cleavage fragments showing parallel extinction.

REFRACTIVE INDEX FIELD DIAGRAMS

Figure 1 is a plot of γ of biotite against γ' of hornblende. All biotite-hornblende pairs are from rocks containing biotite and hornblende as sole ferromagnesian silicates or coexisting with various combinations of muscovite, cummingtonite, clinopyroxene, orthopyroxene, garnet and/or olivine. The diagram shows that biotite coexists with hornblende over a broad range of indices, from 1.576 to 1.695 for γ of biotite and from 1.640 to 1.721 for γ' of hornblende. The data fall in a wide belt with a positive slope of about 45 degrees; in general, as biotite γ increases γ' of horn-

blende also increases. Since refractive indices can be used as an approximate indication of the Fe^{2+} content of biotite and hornblende, as the Fe^{2+} content of biotite increases the Fe^{2+} content of hornblende also increases.

Biotite coexisting with hornblende alone plots over the entire range of indices for biotite and hornblende. For each addition of a coexisting silicate in the order muscovite, clinopyroxene, orthopyroxene, garnet, cummingtonite, olivine, the field in which the data fall becomes smaller and shifts steadily upward and to the right. That is, biotite and hornblende coexisting with clinopyroxene or muscovite fall in Field III which extends from the lower lines of demarcation to the upper part of the figure; points in this field plot within the limits 1.600 to 1.695 for biotite γ and 1.663 to 1.721 for hornblende γ' . Biotite and hornblende coexisting with orthopyroxene or with both clinopyroxene and orthopyroxene fall within essentially the same limits for hornblende (1.659 to 1.721) but at a slightly higher position for biotite (1.618 to 1.695). The first appearance of garnet with biotite and hornblende or in combination with orthopyroxene and/or clinopyroxene is at a still higher position for biotite (1.631 to 1.695) and for hornblende (1.674 to 1.721). The first appearance of cummingtonite or olivine is at an even higher position for biotite (1.642 to 1.695) and for hornblende (1.675 to 1.720). Data for cummingtonite are limited and further information may alter the position of the field boundary.

Thus, from the lower left corner of Fig. 1 to the upper right there are fields in which 2, then 3, 4, 5 and 6 coexisting ferromagnesian silicates are possible. Each field extends from where it makes its first appearance to the top of the figure but has been encroached upon by the next higher field. Modal analyses indicate that for rocks containing clinopyroxene and falling in the 3-mineral field near the boundary with the 2-mineral field the clinopyroxene occurs in trace amounts only. At higher biotite and hornblende indices the modal percentage of clinopyroxene increases. This general relationship seems to hold for the first appearance of other minerals near the field boundaries on all of the figures.

The minimum assemblage in the field sequence would be one mineral, biotite or hornblende. Figure 7 shows the range of γ of biotite and hornblende in rocks containing biotite or hornblende as the only ferromagnesian silicate. Figure 7 in combination with Fig. 1 show the complete succession of 1 through 6 ferromagnesian silicates.

On the figures dashed lines indicating coexistence fields or the first appearance of a mineral should not be considered to be fixed. As more data become available the positions of these lines may be shifted to increase the size of a field.

Figure 2 is a plot of the same data presented in Fig. 1, but the symbols

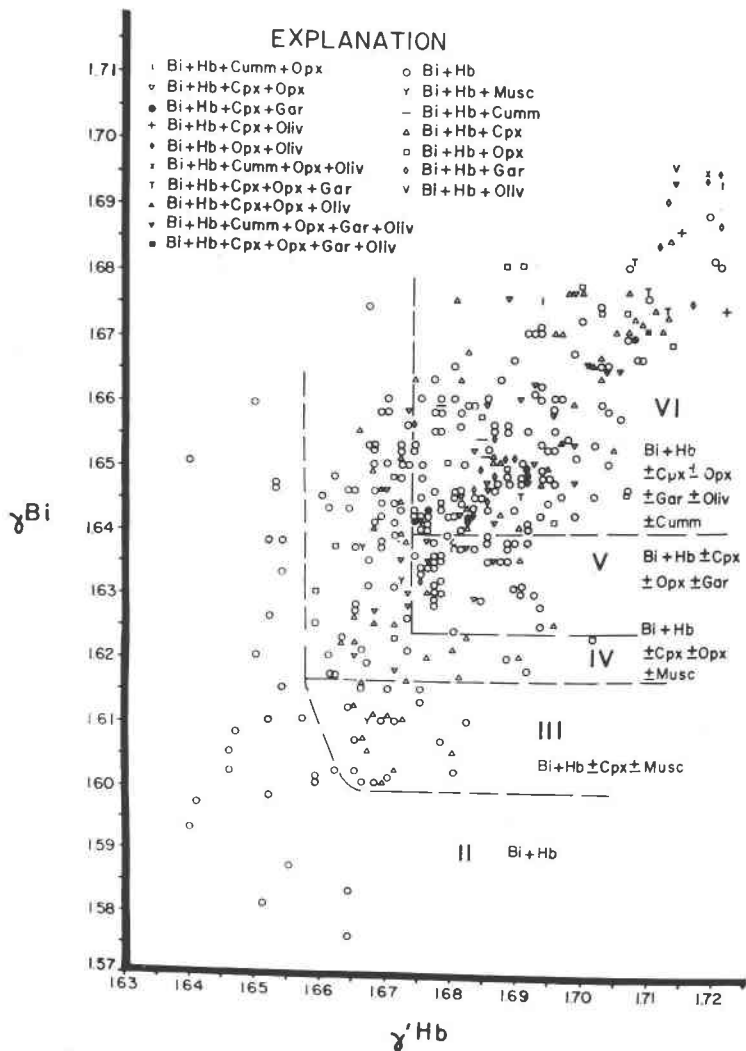


FIG. 1. Refractive index field diagram for biotite (γ)—hornblende (γ').

The explanation of the abbreviations on the figures is as follows: Bi, biotite; Hb, hornblende; Cpx, clinopyroxene; Opx, orthopyroxene; Gar, garnet; Oliv, olivine; Musc, muscovite; Chlor, chlorite; Act, actinolite; Trem, tremolite; Cumm, cummingtonite; Pig, or Pigeon, pigeonite; Sapph, sapphirine; Cord, cordierite; Anth, anthophyllite; Staur, stauriolite; Epid, epidote. Roman numerals indicate total number of ferromagnesian silicates that may coexist in a rock plotted in a field. Symbols on the Refractive Index Field Diagrams represent the minerals, plotted on the ordinate and the abscissa, in rocks in which they are the only coexisting ferromagnesian silicates or in which they occur in combination with other ferromagnesian silicates. (*continued on next page*)

correspond to the rock types from which the data for Fig. 1 were obtained. Figure 2 reveals that biotite and hornblende indices from igneous rocks as compared with those from metamorphic rocks fall in separate but overlapping fields. Biotite with γ less than 1.625 does not coexist with hornblende in any type of igneous rock. In igneous rocks the Fe/Fe+Mg ratio in biotite to the Fe/Fe+Mg ratio in hornblende appears to be higher than it is in metamorphic rocks. Thus, data for metamorphic rocks fall in an area with a somewhat steeper slope than the area containing igneous rock data. Yamada (1953) plotted indices of biotite against Fe²⁺ and Fe³⁺ content and showed that biotites from igneous rocks have higher Fe³⁺ content than biotites from metamorphic rocks. This probably helps to explain the separation of igneous and metamorphic rocks into two areas on the refractive index diagrams.

In order to determine the apparent trend of the igneous and metamorphic data, an analysis of variance for the multiple linear regression of the refractive indices was programmed separately on an IBM 7094 computer for each of the two major rock types. This gave two intersecting regression lines for each rock type indicating the relative dependence of one mineral index on that of the other. When this was completed, the reduced major axis of curve fitting for the intersecting regression lines for each rock type was calculated and plotted on Fig. 2, 4, 6, 8, 10 and 12. On Fig. 2, these two curves show that the igneous rock data, on the average, plot in an area trending with a gentler slope than the metamorphic data.

In the igneous area, from low indices to high, there is no separation of rock types; all varieties plot over almost the entire range. However, at right angles to the trend of the igneous area there does seem to be a gradational change in rock types. That is, near the metamorphic field occur adamellites and granites associated with charnockites; charnockites are in the center of the igneous-metamorphic overlap area. Further into the igneous field syenites, monzonites, granodiorites, and diorites make their appearance followed by gabbros, norites, anorthosites,

Following are the correlation coefficients for Figs. 2, 4, 6, 9, 11 and 13:

Figure	Metamorphic Data	Igneous Data
2	.7760*	.6076*
4	.0095 ns	.7147*
6	.3854*	.5303*
9	.8332*	.4401*
11	.7670*	.6939*
13	.7241*	.7996*

ns=not significant.

*=significant at 95% confidence level or greater.

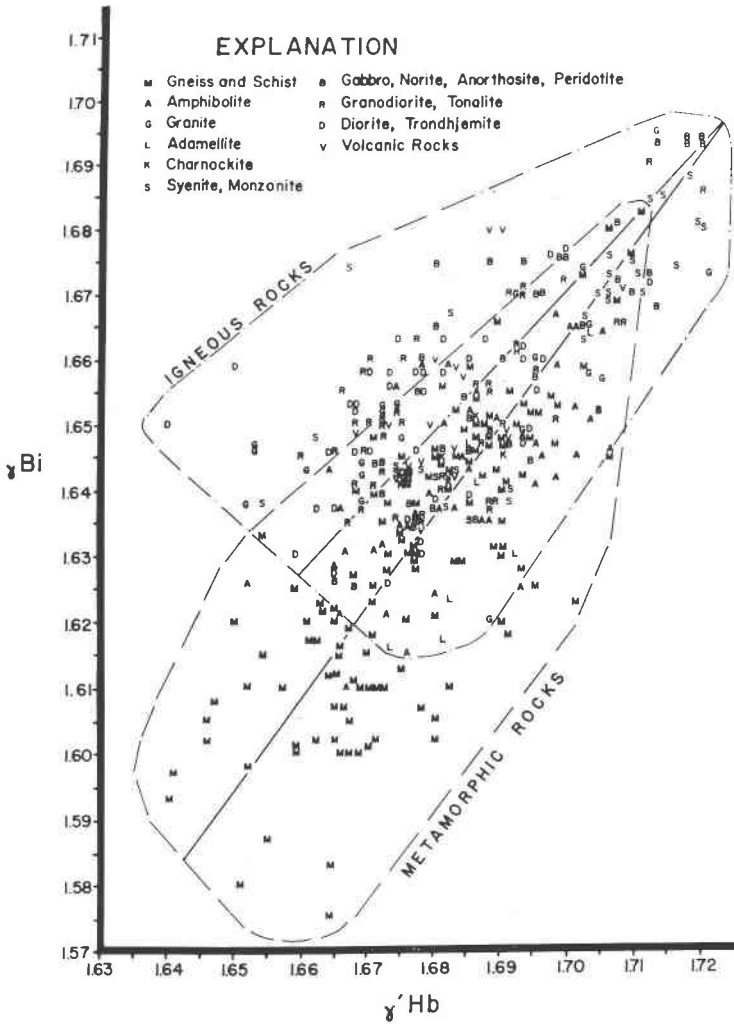


FIG. 2. Rock types from which data on Fig. 1 were obtained.
Straight lines indicate statistical trend of data.

peridotites, and by volcanic rocks. The indices of volcanic hornblendes and biotites may be high because of oxidation during extrusion. Granites, grandiorites, syenites, and diorites are present over the entire igneous area. In the upper right portion of the diagram the rocks are predominantly ferrogabbros and Na-rich syenites. Amphibolites fall for the most part in the area of overlap. Many of the plutonic and volcanic rocks described in the literature as either meta-igneous or metasomatic fall in the area of overlap; metasomatic rocks include gneisses, schists, and

hornfels. Meta-igneous plutonic rocks include those with distinct igneous textures and fabric which show retrograde changes, *e.g.*, clinopyroxene to hornblende, or the formation of secondary garnet. Some volcanic rocks containing lepidomelane or annite ($\gamma=1.730$) and oxyhornblende ($\gamma=1.730$) fall above the graph.

Figure 3 is a plot of γ' of hornblende against γ' of clinopyroxene. Data presented are from rocks containing hornblende and clinopyroxene plus or minus various combinations of biotite, orthopyroxene, garnet, and olivine. The data fall nearly uniformly over the entire figure, indicating a wide range of conditions under which hornblende can coexist with clinopyroxene. However, definite fields of coexisting ferromagnesian silicates are present. Coats (1952) reports a hornblende andesite which contains hornblende ($\gamma=1.743$), clinopyroxene ($\alpha=1.68$), and olivine. In order to plot these data and include this combination of minerals we estimated the clinopyroxene γ' to be 1.710.

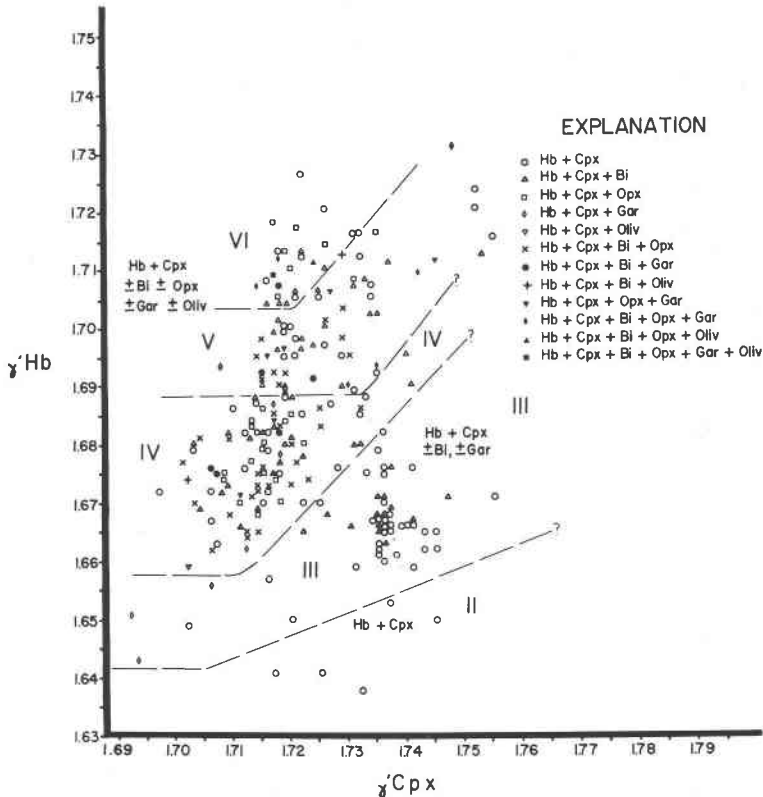


FIG. 3. Refractive index field diagram for hornblende (γ')—clinopyroxene (γ').

The ordered appearance of orthopyroxene, garnet, and olivine on Fig. 1 is not as distinct on Fig. 3, and boundary lines separating fields have not been drawn on Fig. 3. Nevertheless, from lower right to upper left fields of 2, 3, 4, 5, and 6 minerals occur. In the lower right corner, data for clinopyroxene with γ' greater than 1.755 are not shown because such clinopyroxene is usually the only ferromagnesian silicate in metamorphic rocks. Because this figure is a plot of hornblende indices against clinopyroxene indices, these data were added to Figure 7. However, the lower right area on Fig. 3, above clinopyroxene γ' 1.755, is the field of 1 mineral and completes the sequence of 6 minerals.

In igneous rocks clinopyroxene with γ' greater than 1.755 may coexist with hornblende, but in such rocks, both clinopyroxene and hornblende are Na- and Fe-rich and fall in the upper right portion of Fig. 3. In igneous rocks containing no hornblende, clinopyroxene (aegirite or titaniferous augite) with γ' greater than 1.755 may also occur alone (Fig. 7) or in combination with orthopyroxene, garnet, or olivine, but these combinations can not be plotted on Fig. 3 because hornblende is absent.

Data plotted on Fig. 3 are replotted on Fig. 4 with symbols indicating the rock types from which hornblende and clinopyroxene indices were obtained. A separation between metamorphic and igneous rocks is apparent. Also, the data from metamorphic rocks trend from lower right to upper left, whereas igneous rock data trend from lower left to upper right. Hornblende with γ' less than 1.633 does not coexist with clinopyroxene in igneous rocks. The position of the igneous area suggests that hornblende is enriched in iron relative to clinopyroxene, whereas the area of heaviest concentration of metamorphic rocks (lower right) suggests that clinopyroxene in these rocks is enriched in iron relative to hornblende.

Igneous rocks plotted on Fig. 4 are principally diorites, gabbros, norites, charnockites, adamellites, anorthosites, pyroxenites and syenites; granites are absent, and granodiorites are rare. Syenites may show high indices for both hornblende and clinopyroxene. Eclogites, containing omphacite, fall in the metamorphic field just below the lower end of the igneous area, however, the hornblende in the eclogite is probably secondary. Also, some charnockites and adamellites fall in the igneous field near the boundary between the igneous and metamorphic fields.

Figure 5 is a plot of γ of biotite against γ' of clinopyroxene. The data are from rocks containing biotite and clinopyroxene with or without tremolite, cummingtonite, hornblende, orthopyroxene, garnet, pigeonite and olivine. As in Fig. 3, the fields of orthopyroxene, garnet, pigeonite and olivine overlap, and no attempt has been made to separate them, although there does seem to be a sequence in their appearance and rela-

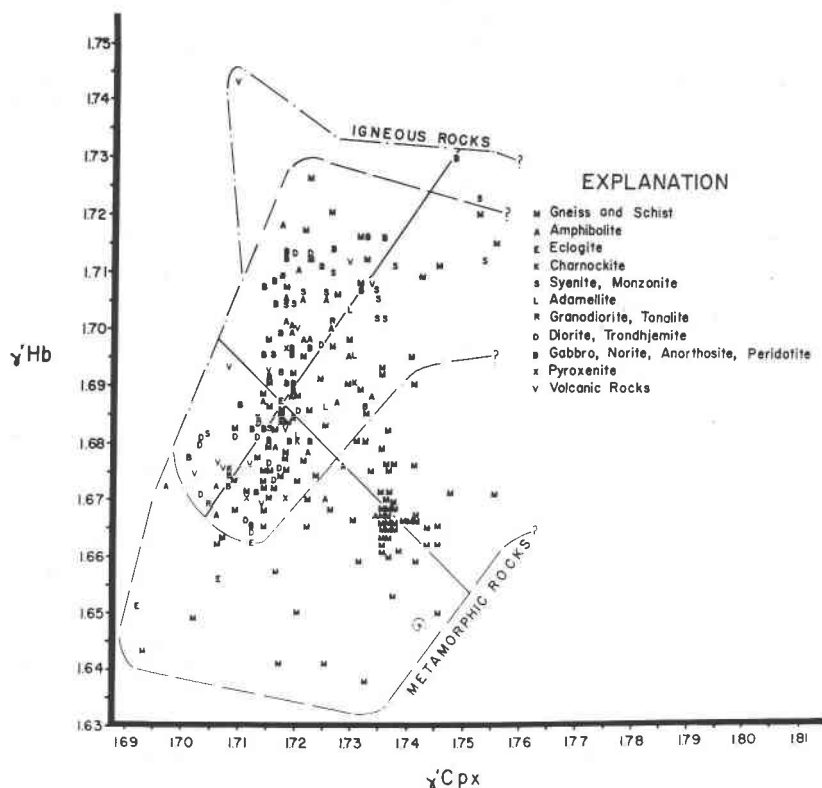


FIG. 4. Rock types from which data on Fig. 3 were obtained. Question marks refer to areas which contain Na-rich pyroxenes (aegirites for which there are no data). Straight lines indicate statistical trend of data.

tive abundance. Clinopyroxene occurs in rocks as the only ferromagnesian silicate where clinopyroxene γ' are higher than 1.755. This provides a field for 1 mineral in the upper part of the figure to complete the sequence.

Figure 6 is a plot of rock types from which biotite and clinopyroxene indices were obtained for Fig. 5. Igneous and metamorphic rocks fall in the left portion, whereas only metamorphic rocks are found in the lower right. Trends of the igneous and metamorphic data are similar to those on Fig. 4. Usually the igneous rocks containing coexisting biotite and clinopyroxene are gabbros, diorites or syenites, but a few granodiorites, norites, and monzonites also contain these minerals. Syenites, as in Fig. 2 and 4, fall where the indices are high. Volcanic rocks fall principally in the upper half of the diagram primarily because the pyroxenes are slightly sodic and have taken up more iron than normal or because of oxidation

during extrusion. Charnockites and some adamellites are found in the area of overlap. The separation of igneous and metamorphic fields in the lower right portion suggests that in igneous rock biotite tends to be enriched in iron relative to clinopyroxene, whereas in some metamorphic rocks clinopyroxene tends to be enriched in iron relative to biotite.

Figure 7 is in part a tabulation of γ of biotite from rocks in which biotite is the only ferromagnesian silicate and from rocks in which biotite coexists with muscovite, garnet, garnet plus muscovite, and with chlorite. For rocks in which biotite coexists with garnet, γ is greater than 1.613; where biotite coexists with muscovite, biotite γ is greater than 1.600. However, the muscovite in the biotite range from 1.600 to 1.620 is actually sericite, but above 1.620 biotite coexists with either sericite or muscovite. Biotite coexisting with sericite is found in schists and phyllites, whereas biotite coexisting with muscovite is found in both igneous and the higher grade metamorphic rocks.

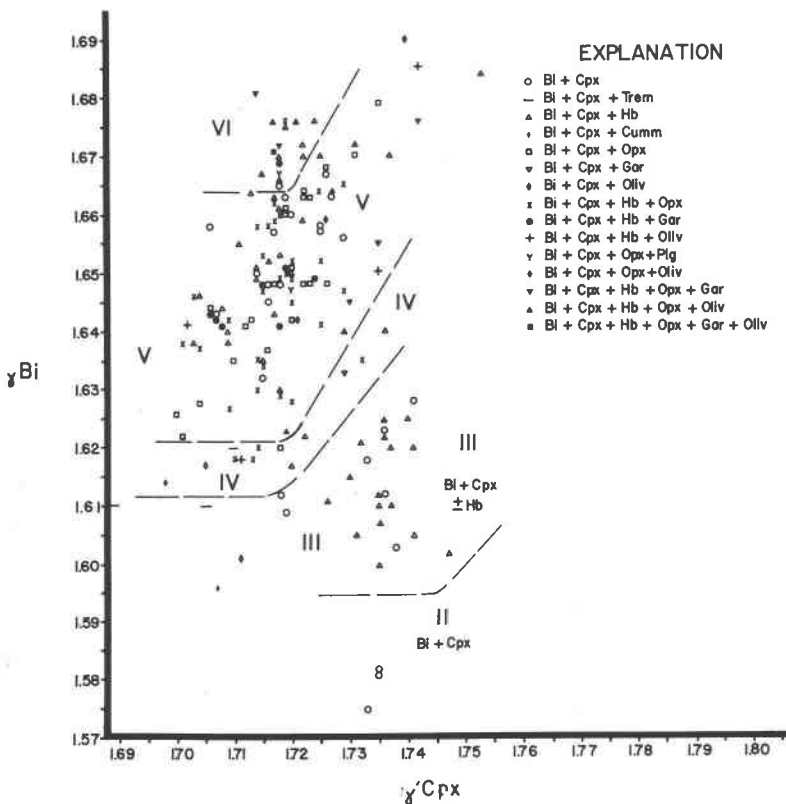


FIG. 5. Refractive index field diagram for biotite (γ)—clinopyroxene (γ').

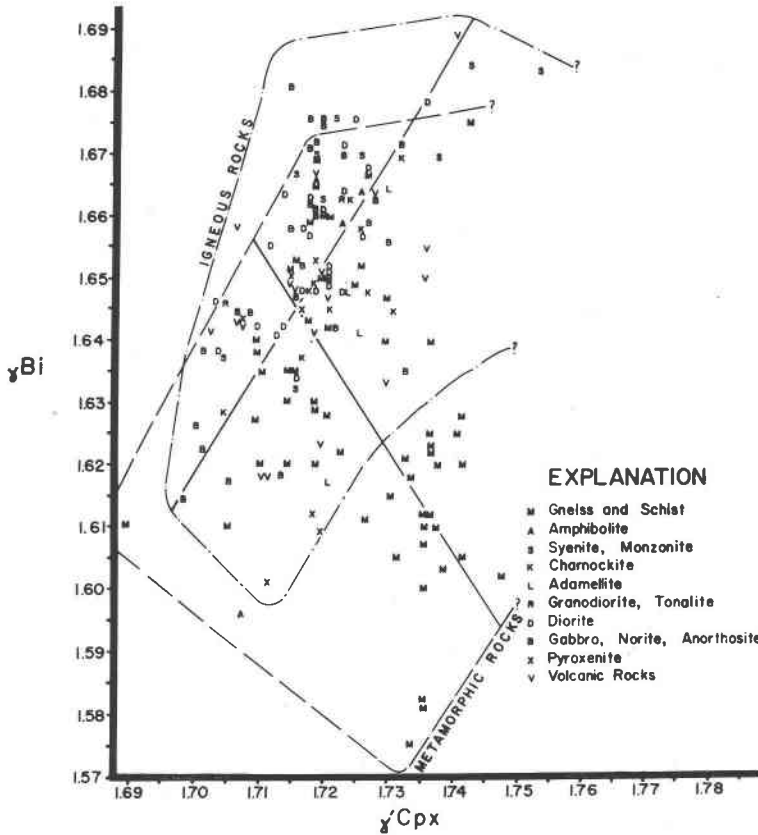


FIG. 6. Rock types from which data on Fig. 5 were obtained.
Straight lines indicate statistical trend of data.

The dominant igneous rock is granite. Of the igneous rocks, granites and syenites contain biotite with the highest γ ; basic volcanic rocks contain biotite with the lowest indices provided the biotite does not coexist with hornblende. Some schists and gneisses contain biotite with still lower indices. No biotite coexisting with garnet and/or muscovite in igneous rocks has indices less than 1.635. Thus, igneous rocks contain biotite with indices higher than those of biotite from low grade metamorphic rocks. In this connection, Iwao (1937) has described a garnet-bearing quartzose-biotite schist progressively replaced by granite aplite in which there is a steady rise in the refractive index of biotite from 1.633 in the schist to 1.657 in the granite.

Biotite indices from biotite-muscovite rocks may be very perplexing. Biotite flakes from a single 10-gram specimen often show large differ-

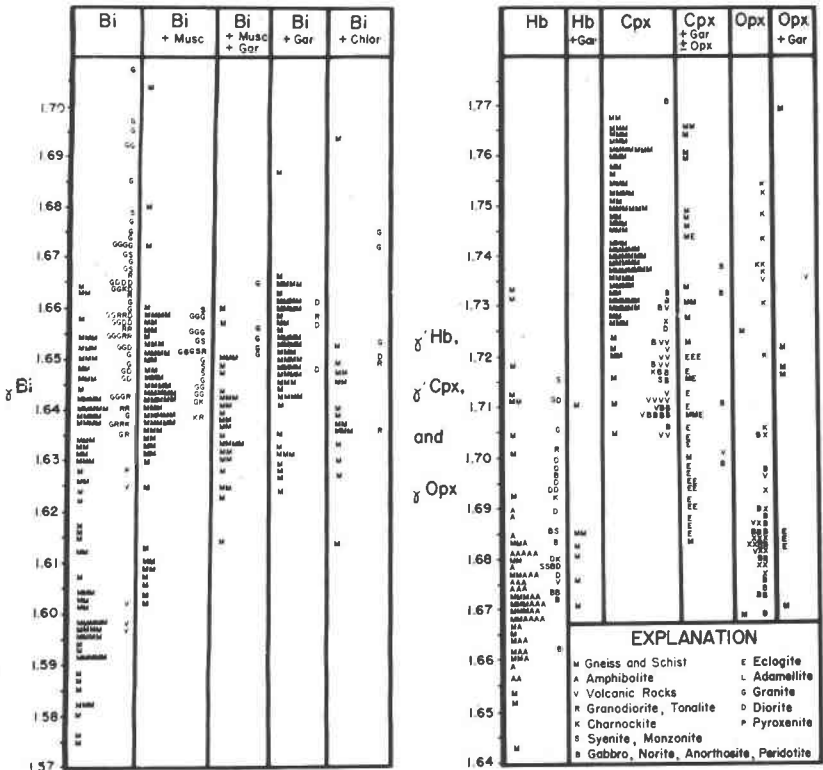


FIG. 7. Plot of biotite γ , hornblende γ , clinopyroxene γ , and orthopyroxene γ in which each of these is the only ferromagnesian silicate in the rock, and in which each coexists with other minerals.

ences in refractive indices when the rock contains both biotite and muscovite. This does not seem to be true when muscovite is absent and ferromagnesian silicates are present. A spread in γ of 0.038 has been observed, but generally the spread is not so great. Other investigators of similar rocks also report a range of indices for biotite.

Nockolds (1947) showed that biotite associated with muscovite is rich in aluminum. Perhaps variations in indices in a single specimen are due to different degrees of substitution of Fe^{3+} and Al^{3+} ions in the biotite structure. If these two ions are freely interchanged, variations in the chemistry of the rock may produce differences in refractive indices. Hall (1941) and others have shown that Fe^{3+} variations cause marked changes in refractive indices of biotite.

Figure 7 contains a tabulation of γ of hornblende, clinopyroxene, and orthopyroxene in rocks with these minerals as the only ferromagnesian

silicates and also in rocks in which they coexist with garnet. Only metamorphic rocks contain hornblende with γ' less than 1.663. It is interesting to note that hornblende in the more "basic" igneous rocks, diorite, gabbro, basic charnockite, basic syenite, and basic volcanic rocks may have indices that are lower than those of hornblende in "acid" igneous rocks such as granite, syenite and granodiorite.

For clinopyroxene alone, γ' ranges from 1.705 to 1.771. Clinopyroxene with γ' greater than 1.735 comes primarily from metamorphic rocks; some syenites have aegirine augite as the only ferromagnesian silicate with γ' of 1.758 and higher. One gabbro (Muir, 1951) has clinopyroxene (γ' 1.771) as the only ferromagnesian silicate, but this rock is associated with other gabbros in which titaniferous augite coexists with olivine. Many eclogites contain omphacite with extremely low γ' (1.685 to 1.710). If clinopyroxene has a higher γ' and coexists with garnet, the rock is generally an iron rich skarn. In general, if the index is below 1.705, and as low as 1.684, clinopyroxene coexists with orthopyroxene, garnet or olivine, but is seldom the only ferromagnesian silicate in the rock.

On Fig. 7 the rocks with orthopyroxene as the only ferromagnesian silicate are volcanic rocks, harzburgites, charnockites, norites, pyroxenites and gneisses. For the most part these rocks are igneous; few gneisses fit this category. In charnockites, norites, and some volcanic rocks orthopyroxene has high indices, whereas in pyroxenites, harzburgites, and also in other volcanic rocks the indices are low. Orthopyroxene coexisting with olivine has both low and high indices.

On Fig. 8 γ of biotite is plotted against γ of orthopyroxene. Symbols represent rocks with biotite and orthopyroxene or these minerals coexisting with various combinations of cummingtonite, hornblende, clinopyroxene, garnet, cordierite, sapphirine, pigeonite, and olivine.¹ Since many biotites with low indices that are associated with olivine are secondary and therefore not in equilibrium with olivine, the boundary line for the first appearance of olivine is uncertain and may be somewhat lower than shown.

γ of biotite and orthopyroxene coexisting with pigeonite and augite in volcanic rocks (data for 3 specimens; Walker and Poldervaart, 1940, 1941) fall in an area of high γ for biotite (1.645 to 1.670) and low γ for orthopyroxene (1.685 to 1.690). This is probably a disequilibrium assemblage.

¹ For two points on Figs. 8 and 9 γ of orthopyroxene were estimated. These are: Yoshizawa (1953), gabbro; biotite γ 1.654, clinopyroxene β 1.696, orthopyroxene β 1.712 (γ estimated as 1.715), and olivine β 1.746; Poldervaart and von Backstrom (1950), gabbro with biotite γ 1.660, hornblende (trace), clinopyroxene γ 1.718, orthopyroxene Of_{23} (γ estimated as 1.693), olivine Fa_{30} γ 1.713.

Although there is a suggestion in Figure 8 of successive fields of co-existing ferromagnesian silicates, we are not certain that this is so. Segnit (1957) reports a cordierite-sapphirine schist containing biotite and orthopyroxene. The indices are: biotite γ 1.570, orthopyroxene γ

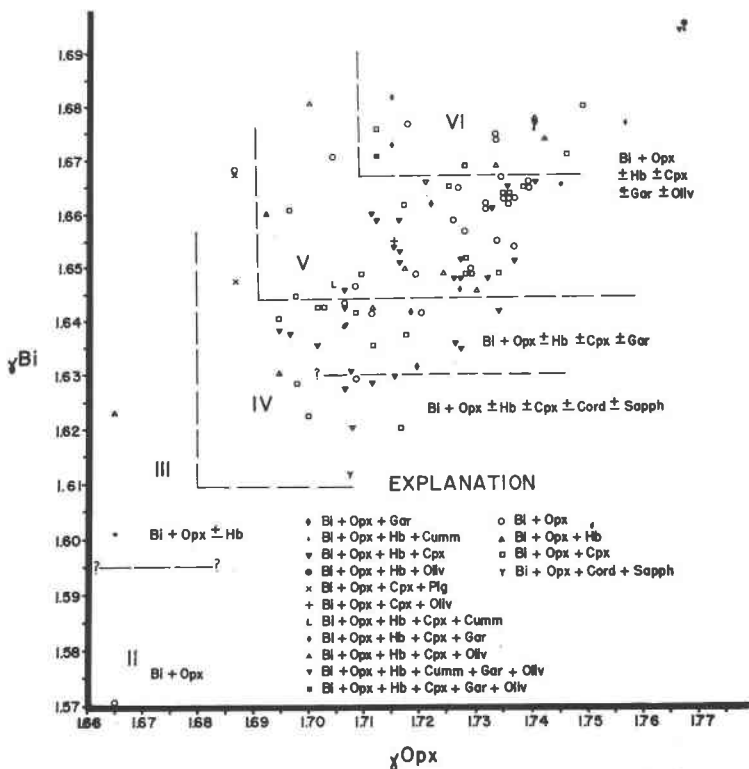


Fig. 8. Refractive index field diagram for biotite (γ)—orthopyroxene (γ).

1.665, cordierite γ 1.540, and sapphirine γ 1.710. Since we are uncertain from Segnit's description whether all four of the analyzed minerals coexist in the same specimen, these data have been plotted in the 2-mineral field for biotite and orthopyroxene. However, this field could be a 3- or 4-mineral field which would include cordierite plus or minus sapphirine, but exclude hornblende, clinopyroxene, garnet, and olivine. Barker (1964) also records γ data for minerals from a sapphirine-bearing rock as follows: biotite 1.612, orthopyroxene 1.708, cordierite 1.541 and sapphirine 1.727, but these data fall higher on Fig. 8 in the 4-mineral field. More data are needed to resolve this uncertainty.

Figure 9 is a plot of the same data shown in Fig. 8; symbols correspond

to rock types. Insufficient data are available to be certain that a separation between metamorphic and igneous rocks exists in the lower portion and consequently no fields have been outlined. However, there is a tendency for metamorphic rocks to fall in the lower right and igneous

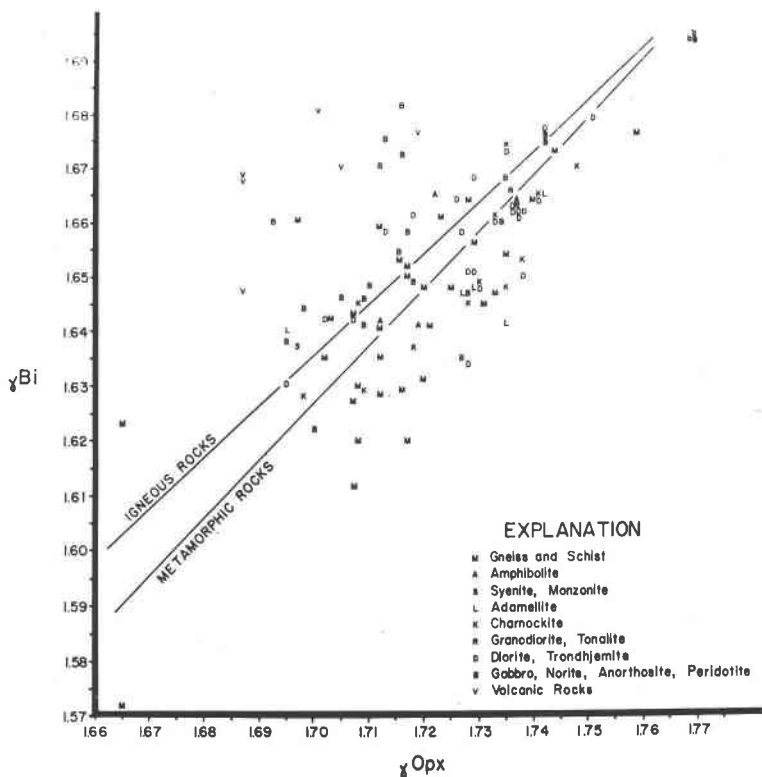


FIG. 9. Rock types from which data on Fig. 8 were obtained.

Straight lines indicate statistical trend of data.

rocks in the upper left, as in Figure 2. Volcanic rocks in particular plot in the upper left.

Figure 10 is a plot of γ' of hornblende against γ of orthopyroxene. Data are from rocks containing hornblende and orthopyroxene with or without combinations of biotite, cummingtonite, clinopyroxene, garnet and olivine.² In Fig. 10 there is a tendency to reverse the order of appearance

² For 3 points on Fig. 10 and 11 γ of orthopyroxene was estimated. These are: Buddington (1952), metagabbro; biotite (0.6%), hornblende γ 1.693, orthopyroxene En_{82} (est. 1.711), garnet (6.7%); biotite (0.2%), hornblende γ 1.696, orthopyroxene En_{56} (est. 1.720), garnet (4.0%); and biotite (0.9%), hornblende γ 1.676, orthopyroxene En_{71} (est. 1.701), garnet (11.0%).

of minerals. In the low index portion in the field of 3 coexisting minerals, hornblende and orthopyroxene coexist with biotite, clinopyroxene, or olivine (5 specimens). This is the first figure in which olivine falls in a region where the coexisting minerals have low indices. In the field of 4 coexisting minerals, hornblende and orthopyroxene coexist with garnet

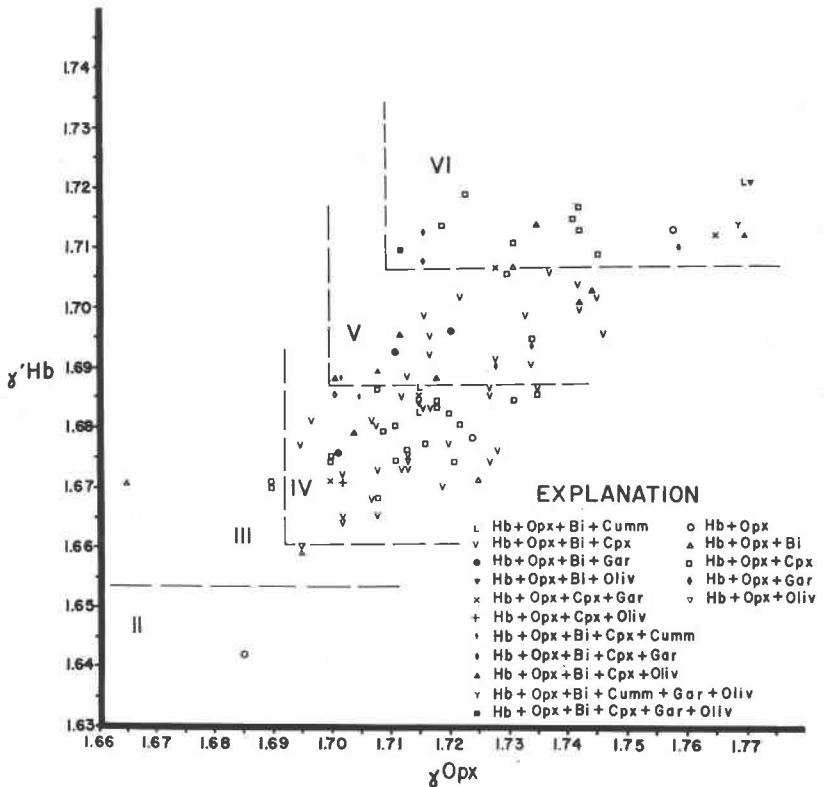


Fig. 10. Refractive index field diagram for hornblende (γ')—orthopyroxene (γ).

and biotite, clinopyroxene and biotite, or cummingtonite and biotite. In the field of 5 minerals, orthopyroxene and hornblende coexist with biotite, clinopyroxene and garnet or with biotite, clinopyroxene and olivine or with biotite, clinopyroxene and cummingtonite. Thus the fields of coexisting minerals are not represented by "characteristic" minerals but rather by the number of coexisting minerals present. Figure 10 may represent a transition to a sequence that is the reverse of the previous sequences. This situation is more apparent in Fig. 12 on which the indices of clinopyroxene and orthopyroxene are plotted.

On Fig. 11 symbols correspond to rock types from which the data for

Figure 10 were obtained. Figure 11 shows that the overlap of metamorphic and igneous rock fields is virtually complete, as in Fig. 9, and no separation of fields has been attempted. However, metamorphic rocks tend to plot in the lower right and igneous rocks in the upper left.

Figure 12 is a plot of γ' of clinopyroxene against γ of orthopyroxene.

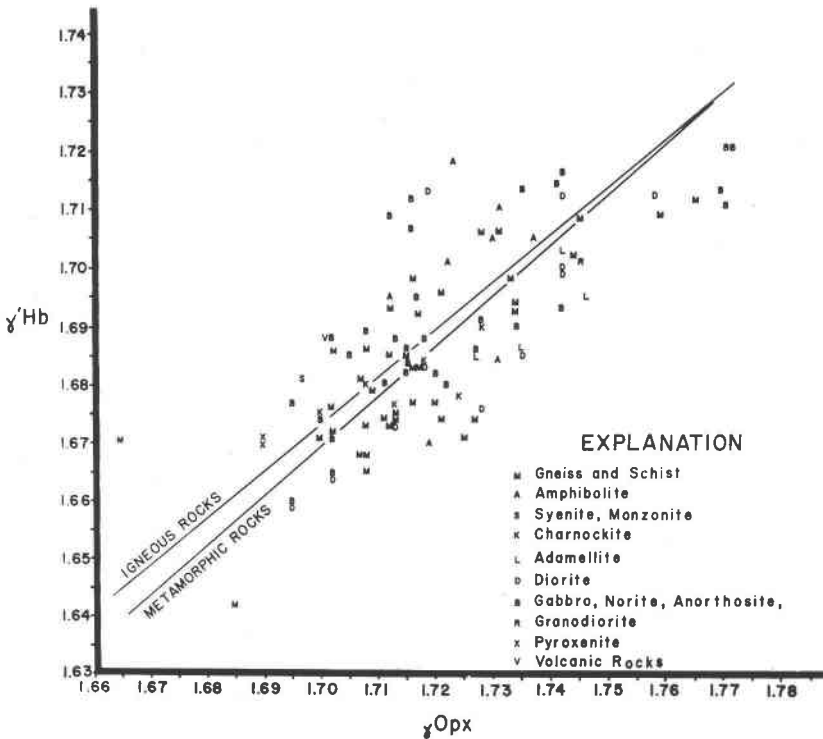


FIG. 11. Rock types from which data on Fig. 10 were obtained. Straight lines indicate statistical trend of data.

Data represent rocks containing clinopyroxene and orthopyroxene plus or minus combinations of biotite, hornblende, pigeonite, garnet, and olivine. The index of orthopyroxene seems to increase on the average about 2.5 times as fast as the clinopyroxene index. The sequence of coexisting minerals from lower left to upper right is apparent, but the first mineral to coexist with the pyroxenes in the low index portion is olivine rather than biotite. Biotite does not appear until 4 coexisting ferromagnesian silicates are present and it is more abundant in the field of 5 coexisting minerals. Whereas biotite and olivine occur in the same 3-mineral field on Fig. 10, this is not so on Fig. 12. Some garnet is found in the field of 3 coexisting minerals, but it is more abundant in the field

containing 4. In Fig. 12 the sequence of minerals from low to high indices is more sharply delineated than it is in Fig. 10.

Figure 13 is a plot of the data in Fig. 12 showing the rock types from which the latter were obtained. "Acidic rocks," charnockites, and adamellites are somewhat concentrated in the lower right portion, basic

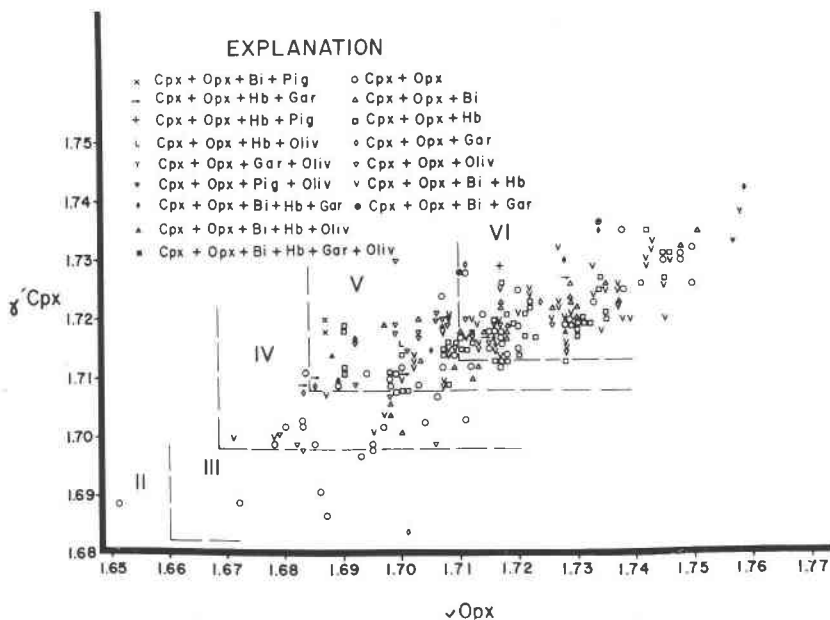


FIG. 12. Refractive index field diagram for clinopyroxene (γ')—orthopyroxene (γ).

rocks in the center, and volcanic rocks in the upper left. Pyroxenites, periodotites, and eclogites fall primarily in the lower left corner. No separation of metamorphic and igneous fields is practicable. Metamorphic rocks tend to plot in the upper left and igneous rocks in the lower right, but the difference between them is very slight.

In addition to the above figures, the writers have prepared refractive index field diagrams for the following combinations of minerals: oliv-cpx, oliv-opx, bi-musc, bi-gar, cpx-gar, bi-cord, bi-anth, anth-cord, act-chlor, hb-chlor, hb-gar, and hb-oliv. Data from the literature that can be used in the construction of these diagrams are limited, but when plotted, linear relationships become apparent.

DISCUSSION

Interpretation of Refractive Index Field Diagrams. If the data approximate equilibrium conditions, what physical and chemical conditions can

be interpreted from these diagrams? Many geologists have discussed the poor correlation between the refractive index of a mineral and its chemical composition. Biotite, hornblende, clinopyroxene, and garnet may show chemical variation at constant index, whereas orthopyroxene and

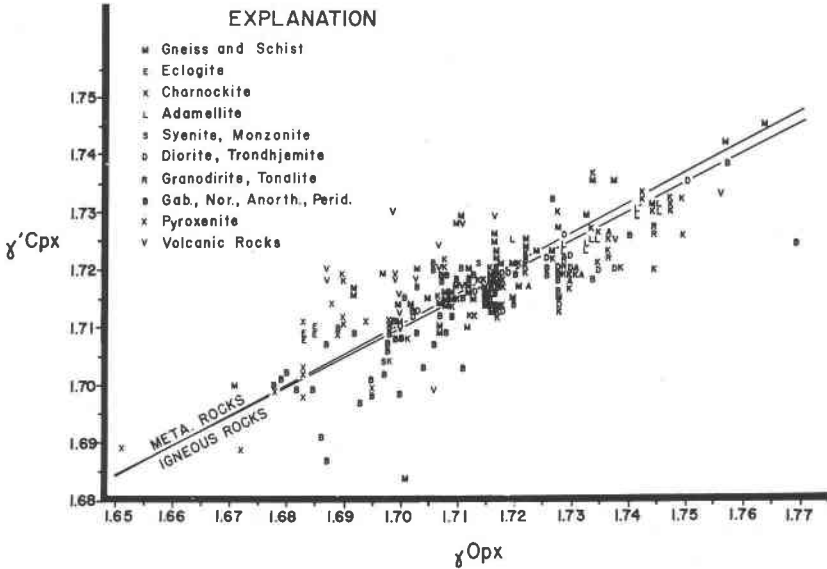


FIG. 13. Rock types from which data on Fig. 12 were obtained. Straight lines indicate statistical trend of data.

olivine show little or none. For example, 11 chemically analyzed biotite specimens from the literature, all with γ of 1.641, show the following ranges in chemical composition: FeO, 13.15–19.22%, Fe₂O₃, 1.14–3.75%, MgO, 8.25–14.65%, Al₂O₃, 13.96–19.79%, and TiO₂, 2.45–5.55%. Hess and Phillips (1940), Hall (1941), Taneda (1947 a, b), Hess (1949), Kuno (1954), Dudek (1954), Henriques (1957, 1958 a, b, c, d, e), and Parker (1961) have derived formulae or give other information to show how an increase in each of these oxides raises or lowers the refractive index of ferromagnesian silicates. These formulae and chemical analyses have little value, however, in interpreting the refractive index field diagrams, for, although the optically and chemically analyzed minerals are from a variety of geologic environments, they represent analyses of single minerals, and other ferromagnesian silicates that may have coexisted with them were not analyzed.

On Fig. 1, biotites, each with γ of 1.641, coexist with hornblendes ranging in index from about 1.640 to about 1.710. Because of this broad

range in indices, and the fact that coexisting minerals are derived from many types of igneous and metamorphic environments, it is not surprising that biotites in these rocks show a diversity of chemical composition even though each has γ of 1.641. A similar range in chemical variation is found for hornblendes at constant index. However, chemically analyzed coexisting biotite and hornblende for which optical data are given are sparse in the literature; this is also true of other coexisting ferromagnesian silicates.

Although ferromagnesian silicates with the same index may show a diversity in chemical analyses when selected from rocks of markedly different bulk chemical compositions and geologic environments, a number of investigators, *e.g.*, Hess and Phillips (1940), Hall (1941), Heinrich (1946), Taneda (1947 a, b), Hess (1949), Winchell and Winchell (1951), Yamada (1953), and Clavan *et al.* (1954), have shown that locally for rocks of one bulk composition, an increase in refractive index of a ferromagnesian silicate corresponds to an increase in Fe^{2+} and a decrease in Mg^{2+} . Hagner and Collins (1955) and Hagner *et al.* (1963), have also found this to be true.

The refractive index field diagrams illustrate a potential value of this method of plotting. When sufficient data on isochemical rock series are available, it should be possible to apply the mineralogic phase rule to refractive index field diagrams of such systems. In addition, abundant chemical and optical data for coexisting ferromagnesian silicate pairs would permit the use of these diagrams in interpreting the conditions of formation of rocks of unknown origin. The tendency for igneous rocks to fall in an area different from that for metamorphic rocks, may be a manifestation of the likelihood that in a plutonic environment subjected to descending temperature the sequence of the partition factors of Fe and Mg probably operates in the opposite direction from that in an environment of ascending temperature such as is the case in progressive regional metamorphism. Thus in a cooling magma olivine is early and Mg-rich whereas hornblende is late and relatively Fe-rich. In most cases successively later mafic silicates are more Fe-rich.

On the other hand, if the process is reversed and temperature increases, as under some metamorphic conditions, an Fe-rich biotite in a biotite schist should become unstable and progressively more Mg-rich. At the higher temperatures at which hornblende first begins to develop in the schist, the first crystals to form would be relatively Fe-rich. If the temperature of the schist increases slowly enough so that equilibrium is maintained, the hornblende might have a higher Fe/Fe+Mg ratio than the coexisting (Mg-rich) biotite whereas in igneous rocks in which

equilibrium is not maintained, the hornblende might have a lower Fe/Fe+Mg ratio than the biotite. This would account for the separation of metamorphic and igneous rock areas in the lower portion of Fig. 2. These same relationships should hold true for hornblende relative to clinopyroxene and biotite relative to clinopyroxene on Figs. 4 and 6. A suggestion of this separation is also present on Figs. 10 and 12, but the overlap of igneous and metamorphic areas is so large that chemical data are needed to confirm this.

Work by Heinrich (1946) on the chemical composition of biotite shows the biotite rich in Fe²⁺ is found in granites and pegmatites, biotite rich in Fe³⁺ and less rich in Fe²⁺ is found in intermediate rocks, and biotite poor in Fe²⁺ and Fe³⁺ and rich in Mg²⁺ (phlogopite) is found in peridotite and ultrabasic igneous rocks. Thus, the phlogopite and hornblende of ultrabasic igneous rocks should plot in the lower left corner of Fig. 2. But the data in Figs. 1 and 2 suggest that phlogopite will not be found coexisting with hornblende in ultrabasic igneous rocks unless these are metamorphosed. Fig. 2 shows that the lower left corner is occupied by metamorphic rocks such as biotite-hornblende gneisses and schists; biotite of igneous rocks has much higher indices. Figures 5 and 6 show phlogopite coexisting with Mg-rich clinopyroxene in ultrabasic igneous rocks (lower left corner). However, these rocks do not contain hornblende, although they may have tremolite, cummingtonite, enstatite, pigeonite, or olivine. Apparently phlogopite does not coexist with hornblende in igneous rocks. Occasionally an ultrabasic rock is described containing phlogopite and small needles of tremolite, cummingtonite, or an unidentified fibrous hornblende, but such rocks have been metamorphosed. Olivine and pyroxene in these rocks have been altered to hornblende and phlogopite, indicating that new equilibrium conditions were probably established.

In this investigation garnet has been studied from gneisses, eclogites, high-grade ultrabasic metamorphic rocks, and from certain granites and pegmatites, volcanic bombs, anorthosites, norites, charnockites and meta-ultrabasic igneous rocks. Rocks containing garnet seem to be (1) relatively Al-rich as in some eclogites or muscovite-sillimanite-kyanite-bearing varieties and (2) relatively Fe-rich (including an abundance of Fe³⁺) as indicated by the presence of staurolite or by the relatively high index of coexisting biotite, hornblende, clinopyroxene, or orthopyroxene.

In addition to the separation of rock types, the diagrams show a separation on the basis of total number of coexisting mineral phases. What temperature-pressure-composition conditions are required on Fig. 1 for the transition from the 2-mineral field to the 3-mineral field, and

from the 3 to the 4? What differences are there between the Fe/Fe+Mg ratios in the coexisting phases at different transition points along the boundary separating the mineral fields? At the same indices for biotite and hornblende, what is the difference in their Fe/Fe+Mg ratios when they are the only ferromagnesian silicates in comparison to when they coexist with additional ferromagnesian silicates? Because the data are lacking, we cannot answer these questions. Perhaps these diagrams in combination with other optical data, such as 2V or extinction angle, may provide a clue to a specific set of conditions for the formation of a given rock.

Cautions and Limitations in the Use of Refractive Index Field Diagrams. Rocks are dynamic systems subjected to continuous adjustment to changing environmental conditions. The rocks we have dealt with are assemblages of minerals that probably represent equilibrium conditions, but at what stage in their history?—at the time of formation of the “original” rock or later? The refractive indices of coexisting ferromagnesian silicates within these rocks are dependent on the partitioning of Fe²⁺ and Mg²⁺ ions, as well as on other ions (Ti⁴⁺, Al³⁺, Fe³⁺, Mn²⁺, Ca²⁺, Na¹⁺, and K¹⁺). The various partition coefficients, in turn, are complex functions of variables such as total pressure, several volatile partial pressures, temperature, composition of coexisting felsic phases, number and kinds of coexisting ferromagnesian silicates, bulk chemical composition of the rock, and prior geologic history. It is difficult or impossible to determine how metastable the original partition was and how likely it is that it was preserved. This presents the problem of what constitutes coexisting minerals and this, in turn, has a bearing on the interpretations made; it has received insufficient attention from petrologists and mineralogists. Nonetheless, we have had to assume, except where petrographic evidence is to the contrary, that the present assemblages of coexisting minerals represent the main assemblages and compositions of the “original” rocks.

The writers are thus aware that the interpretations given have been derived from parameters that are a function of several independent variables. Refractive index data on minerals with different chemical compositions from many localities have been plotted together, and generalizations arrived at from a study of minerals that formed under diverse geologic conditions must be subjected to careful check and revision as more information is accumulated. A major factor preventing more extensive interpretation of the refractive index field diagrams is the lack of knowledge about the conditions of formation and origin of some of the rocks. For many rocks, no information is given on the presence

or absence of accessory magnetite; the presence of magnetite might change the number of minerals characteristic of a given field. Dudek (1954) has pointed out that with increasing iron oxides in a rock there is a rapid increase in MgO in biotite, which would affect the biotite index. Also, in the interpretation of the figures the effect of the presence of felsic phases has not been taken into account. Modal percentages and variations in feldspar composition do seem to have some effect on the refractive indices of coexisting ferromagnesian silicates even though there is no *direct* relationship of index of mafics to that of felsics. Work by Tsuboi (1933), Taneda (1942), Ota (1952) and Oki (1961) suggests that this is true. Thus, the refractive index field diagrams are not intended to be used as true phase diagrams as are ACF ternary diagrams in which the whole rock can be represented, but rather as guides to the distribution of certain elements (Fe and Mg) between coexisting ferromagnesian silicates.

Omission from the diagrams of ferromagnesian silicates such as chlorite, epidote, clinozoisite, tremolite-actinolite, etc., because of paucity of data, raises the question of number of minerals in some of the fields shown in the diagrams. Would the addition of these minerals raise the number of minerals in fields II and III to that characteristic of fields V and VI? In other words, do these minerals coexist with biotite, hornblende, clinopyroxene, and orthopyroxene, and if so, do they coexist in the low index portion of Figs. 1, 3, 5, 8, 10 and 12 so that fields II and III are invalid? The data we have presented suggest that the fields are real in most cases. In the search for data it was found that most of the above mentioned minerals do not coexist with biotite plus hornblende, clinopyroxene, or orthopyroxene and therefore cannot be plotted on these figures. Those minerals that occasionally do coexist with these ferromagnesian silicates, such as chlorite, epidote, tremolite-actinolite, glaucophane and anthophyllite, do so only in such combinations that the sequence of fields III through VI is unchanged. That is, the kinds of coexisting phases within each field might vary, but not the total number or trend.

CONCLUSIONS

When the refractive index of one ferromagnesian silicate is plotted against that of another from rocks in which chemical equilibrium is approximated, fields for certain combinations of coexisting ferromagnesian silicates become apparent. These fields represent a type of coexistence diagram. The information gained from the diagrams is summarized below and is subject to the cautions and limitations discussed in the preceding section.

1. Fields of 2, 3, 4, 5 and 6 coexisting ferromagnesian silicate phases are established within definite ranges of indices for biotite, hornblende, clinopyroxene and orthopyroxene. The addition to the fields of other ferromagnesian silicates such as chlorite, epidote, clinozoisite, tremolite-actinolite, etc., does not seem to alter the total number of coexisting phases in a given field, except perhaps for cordierite and sapphirine.

2. Field boundaries suggest that there are definite compositional limits in the partitioning of Fe and Mg atoms between coexisting ferromagnesian silicates.

3. The separation of igneous and metamorphic areas on the lower portions of Figs. 2, 4 and 6 suggests that under disequilibrium conditions in igneous rocks the Fe/Fe+Mg ratios in coexisting phases increases in the order: olivine, pyroxene, hornblende, biotite, but under conditions approaching equilibrium in metamorphic rocks and with increasing grade the Fe/Fe+Mg ratio increases in the order: biotite, hornblende, clinopyroxene, orthopyroxene, garnet. Chemical data are needed to confirm this.

4. The refractive index field diagrams indicate a need for extensive chemical and optical studies of coexisting ferromagnesian silicates, particularly in isochemical systems, before further interpretations are possible.

5. In addition to the data presented, perhaps the most significant contribution of this study is the illumination it throws on particular problems that need further and more detailed investigation by field and petrographic methods, chemical analysis, and by experimental studies of phase equilibria in simplified and analogous artificial systems.

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An expanded bibliography of references from which refractive index data and other material pertinent to this paper were obtained has been deposited, along with certain refractive index field diagrams as Document No. 8508 with the American Documentation Institute, Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington 25, D. C. Copies may be secured by citing the document number, and remitting \$5.00 for photoprints or \$2.25 for 35 mm. microfilm. Advance payment is required.

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