

# CORRELATION OF PHYSICAL PROPERTIES AND CHEMICAL COMPOSITION IN THE PLAGIO- CLASE, OLIVINE, AND ORTHO- PYROXENE SERIES

ARIE POLDERVAART\*

## ABSTRACT

Physical properties of the plagioclase, olivine, and orthopyroxene series are plotted against chemical composition in a series of diagrams. The limitations of the various methods used to determine compositions of rock-forming minerals by optical means are discussed in detail, and lines of further work on the three mineral series are noted. A plea is made for the use of molecular percentages and for standard nomenclature in these, and other mineral groups.

## INTRODUCTION

For some years the author has collected data on common rock-forming minerals published in the literature. The present paper presents this information in several diagrams pertaining to the three best-known solid solution series; albite-anorthite, forsterite-fayalite, and enstatite-orthoferrosilite.

The assessment of the chemical composition of a mineral by the determination of a set of optical properties is one of the chief aims of the petrographer. Frequently he also requires further information; concerning the temperature of crystallization of the mineral, its composition in terms of oxides as well as of mineral molecules, and other physical properties in addition to those already measured, but which cannot be determined in a single rock slice. This information is scattered throughout the literature, and is often not presented in a uniform manner, necessitating cumbersome calculations. The present diagrams bring together all information required in routine petrographic procedure. They have been checked against modern analytical data, but to avoid overcrowding, the analyses have not been plotted on the curves.

The nomenclature of the plagioclase series on a decimal, *molecular* percentage basis (Calkins, 1917) is now accepted by most petrologists, but the same cannot be said of other mineral series. There is still much confusion in mineral nomenclature, enhanced by the fact that compositions are indiscriminately expressed as weight or as molecular percentages.

In an attempt to apply Calkins' decimal, molecular percentage nomenclature also to other mineral series, Deer and Wager (1939) proposed a similar classification for the olivine series, while the author (1947) sub-

\* Geological Survey, Lobatsi, Bechuanaland Protectorate.

sequently extended this system of nomenclature to the orthopyroxene series. The writer repeats his plea (1947, 1949, also *vide* Benson, 1944) for standard nomenclature and the use of molecular percentages in expressing mineral compositions. A scale has been included in the present diagrams, whereby molecular percentages are converted to weight percentages, for use in modal calculations.

Mineral compositions in the plagioclase, olivine, and orthopyroxene series are normally determined optically with an accuracy of  $\pm 2$  per cent, but this can be increased by measuring several crystals and taking mean values. Much depends on the purity of the mineral in question, and on the methods employed in assessing its optical constants. It is always best to use two unrelated methods; for instance, to determine for olivines and orthopyroxenes both the optic axial angle and one of the refractive indices. If such measurements are repeated several times on different crystals, and the results are found to agree closely, both with one another and with the values given by the corresponding curves, the derived composition can be accepted with confidence. Anomalous values may be due to incorrect measurements, presence of impurities, strain, zoning, and other causes, and should be studied in greater detail, by the determination of other physical constants, and by close observation of the natural habitat of the mineral. If possible this should be followed by its isolation and chemical analysis.

#### PLAGIOCLASE SERIES

Probably no other mineral series has been studied in such detail as the plagioclase series. Yet there remains ample scope for further work, in particular in defining the influence of admixtures and of temperature on the optical properties of the series. The plagioclase feldspars really belong to a ternary system with components albite, anorthite, and orthoclase, but it has become customary to consider them as a simple binary system. Yet correlations of optical properties and chemical composition in a triangular diagram would be of great value (Alling, 1936), while the co-existence of plagioclase feldspars and potash-soda feldspars (Niggli, 1941) is another field of study requiring further work. Yet another problem is the association of plagioclase and quartz in myrmekitic and other intergrowths (Drescher-Kaden, 1948; Poldervaart and von Backström, 1949). The thermal relations of the albite-anorthite series have been studied by Bowen (1913), and his classic thermal diagram is probably the best known in petrology.

A large number of methods have been devised to correlate physical properties with chemical composition. Since plagioclase occurs in nearly every igneous rock and in many metamorphic and sedimentary rocks,



the determination of the An content of the feldspar is one of the most essential and common parts of petrological procedure. Yet many problems remain in determining compositions of members of the series by optical means. It is seldom realized how difficult is the correct determination of the average plagioclase composition in a rock in which the feldspar is strongly zoned (Wenk, 1945). Merely taking the mean of extreme values determined in one or more crystals rarely provides the correct result, unless a large number of crystals is examined. Determination of the mean index of refraction by comparison of a large number of grains orientated at random is less time-robbing, but likewise not very accurate. Probably the best procedure in this case is to ascertain the mean specific gravity by floating the crush in a liquid, of which the density is next determined.

The influence of admixtures on the optical properties of plagioclase feldspars is largely unknown. The most common of these are  $K_2O$ ,  $Fe_2O_3$ ,  $BaO$ , and  $SrO$ . An Or content up to 10 per cent apparently does not affect the physical constants of the plagioclase, but with more than 10 per cent Or the indicatrix is changed (Chudoba and Engeld, 1937).  $Fe_2O_3$  also occurs in plagioclase to a limited extent (Faust, 1936; Ramberg, 1949), apparently replacing  $Al_2O_3$ , but its influence on the optical properties is unknown.  $BaO$  and  $SrO$  also enter the molecule, but their influence is equally uncertain. Excessive  $Al_2O_3$  apparently distorts the ionic structure and results in anomalous optical properties (Perrier, 1930; Beljankin, 1931). Temperature also appears to affect the indicatrix, and different curves have been drawn for plagioclase of effusive and deep-seated rocks (Barber, 1936; Köhler, 1942; Scholler, 1942), though presumably there are feldspars which fit neither curves, having crystallized at intermediate temperatures and pressures.

Methods used to ascertain sets of optical properties, which in turn are correlated with composition, may be divided into two classes; universal stage methods and refractive index methods. Some workers prefer the former to the latter, but Wenk (1945) has shown that there is no valid reason for such preference. Each case should be considered on its own merits, and a method employed which is of the required accuracy, the most easily applied, and the least time consuming. Slightly altered or clouded crystals are best determined by universal stage methods. In zoned crystals refractive index methods tend to emphasize lower An values, since the edges of cleavage fragments are employed, while universal stage methods emphasize higher An values, as crystal cores are most conveniently used (Wenk, 1945). Some methods are more cumbersome and require more time than others. Since in one section there are

generally scores of plagioclase crystals, it is usually more profitable to select suitably orientated crystals, and to apply the easier and speedier method. Again, some methods are more accurate than others. It is good policy to aim at maximum accuracy, but there are often natural limitations, when less accurate methods may be employed successfully.

Refractive index methods may be divided into: (1) the ordinary immersion method using sodium light, (2) the single variation method, and (3) the double variation method; the three methods being of increasing accuracy in the order stated. The double variation method, perfected by Emmons (1929), is undoubtedly the most accurate, but it requires elaborate and expensive apparatus and consumes more time than the other two methods. Hence its application will remain limited to the better equipped laboratories, and to instances where such high accuracy is required. The single variation method, as applied by Tsuboi (1923, 1934) to the plagioclase series, is for most purposes the most accurate and convenient method. Yet the ordinary immersion method is the most commonly used. With practice it is more rapid than either the single- or the double variation method, while its accuracy is sufficient for normal purposes. In using either method, it is well to note that Tsuboi's curve for the lower index of (001) cleavage fragments does not differ appreciably from that of (010) cleavage fragments, nor from the  $\alpha$  index curve. Thus it is normally sufficiently accurate to determine the lower index of either (001) or (010) cleavage fragments, and to plot this value on the  $\alpha$  curve, in order to ascertain the composition of the plagioclase. The paucity of data on plagioclase  $An_{60-80}$  should be noted. More data in this composition range are required to check the present curves.

Universal stage methods may be divided into: (1) the Rittmann zone method (1929), and (2) the Fedorow method. Both methods have been discussed in an admirable manner by Emmons (1943). The Rittmann zone method, more rapid and convenient than the Fedorow method, is best applied to plagioclase with less than 60 per cent An, while the Fedorow method gives the best results for plagioclase with more than 60 per cent An. The two methods are complementary.

Maximum extinction angles on albite twin lamellae in oriented sections provide an accurate and convenient means of determining compositions of plagioclase with less than 50 per cent An. Extinction angles on combined Carlsbad-albite twins are also very accurate, but normally such twins are somewhat rare. Extinction angles on (001) and on (010), and the "angle of the rhombic section" appear to be less accurate. The optic axial angle of plagioclase with more than 60 per cent An also varies consistently enough to form a means of determining its composition.

The specific gravity, hardness (Holquist, 1914), and width of albite twin lamellae (Donnay, 1940), have also been correlated with the composition of the plagioclase feldspars.

The present diagram includes the following curves:

1. Liquidus-solidus.
2. Specific gravity of crystals and corresponding glass.
3. Molal and latent heat of melting.
4. Weight percentages of  $\text{Na}_2\text{O}$ ,  $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{SiO}_2$ .
5. Optic axial angle.
6. Refractive indices and birefringence.
7. Extinction angles on (001) and on (010).
8. "Angle of the rhombic section."
9. Maximum extinction angle of albite twin lamellae.
10. Extinction angles of combined Carlsbad-albite twins.

For other curves and stereograms, applied in universal stage methods, reference is made to Emmons (1943), or Winchell (1946).

#### OLIVINE SERIES

The composition of most naturally occurring olivines may be expressed in terms of  $\text{Mg}_2\text{SiO}_4$  (forsterite) and  $\text{Fe}_2\text{SiO}_4$  (fayalite). This solid solution series has been studied thoroughly, and variations of physical properties with composition are known with accuracy. Deer and Wager (1939) have shown that the results obtained by Bowen and Schairer (1935) in their study of the system  $\text{MgO-FeO-SiO}_2$  are faithfully reproduced in natural olivines.

Generally it is most convenient to find the composition of olivine in a rock from the determination of one or more of the refractive indices. Yet it is advisable to determine the optic axial angle as well. This second determination forms a good check on the first, while zoned olivines (Tomkeieff, 1939) are more easily identified and measured by this means. Frequently olivines from the same rock show a range in composition, though individual crystals are unzoned. Both this tendency and zoning are most pronounced in olivines of intermediate composition. In the range between  $\text{Fa}_{40}$  and  $\text{Fa}_{80}$ , a gap in the crystallization sequence of the series may be encountered, especially in strongly differentiated intrusions. This is in accord with Bowen and Schairer's results, but it does not imply the existence of a gap in the miscibility curve of the series.

The chief admixtures of natural olivines are  $\text{Fe}_2\text{O}_3$ ,  $\text{NiO}$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{MnO}$ , and  $\text{CaO}$ . The first three oxides tend to be associated with magnesian olivines, while the last two are often more prominent in the iron-rich members of the series. For the purpose of calculating compositions of olivines belonging to the forsterite-fayalite series,  $\text{MnO}$  is generally added to  $\text{FeO}$ , while other admixtures are neglected.



Part of the  $\text{Fe}_2\text{O}_3$ , NiO, and  $\text{Cr}_2\text{O}_3$  shown in analyses is probably present as small inclusions of chrome-magnetite or picotite, in the form of minute octahedra, dendrites, or thin plates and wedges parallel to (001) or (100) of the olivine host. Part is undoubtedly dissolved in the molecule, and Bowen and Schairer (1935) have found that small amounts of  $\text{Fe}_2\text{O}_3$  remained in their melts in equilibrium with metallic iron, being incapable of further reduction.

The amounts of MnO and CaO found in natural olivines are generally small. Yet both forsterite and fayalite form solid solution series with tephroite,  $\text{Mn}_2\text{SiO}_4$ , the intermediate members being known respectively as picrotephroite and knebelite. Solid solution series probably also exist between forsterite and monticellite,  $\text{MgCaSiO}_4$  (Ferguson and Merwin, 1919), fayalite and the double salt  $\text{FeCaSiO}_4$  (Bowen, Schairer, and Posnjak, 1933), and tephroite and glaucochroite,  $\text{MnCaSiO}_4$ . Apparently monticellite,  $\text{FeCaSiO}_4$ , and glaucochroite are miscible in all proportions. Selected data of these rare olivines are given in Table 1.

TABLE 1. DATA FOR MANGANESE AND CALCIUM OLIVINES

Index	1	2	3	4	5	6	7
$\text{SiO}_2$	31.39	32.95	29.94	31.48	37.36	36.67	34.80
FeO	—	—	46.88	—	1.40	7.57	4.11
MnO	65.34	46.99	18.83	38.00	.04	.17	13.39
MgO	3.15	18.11	3.01	—	24.90	21.11	17.65
CaO	—	.71	—	28.95	33.08	32.56	27.38
rest	—	1.05	1.14	1.74	1.28	1.64	2.54
total	99.88	99.81	99.80	100.17	98.06	99.72	99.87
$\alpha$	1.759	1.711	1.805	1.686	1.641	1.654	1.663
$\beta$	1.786	1.727	1.838	1.722	1.649	1.664	1.674
$\gamma$	1.797	1.740	1.847	1.735	1.655	1.674	1.680
$2V (-)$	$65^\circ$	$85^\circ$	$54^\circ$	$61^\circ$	—	$82^\circ$	$75^\circ$

1. Tephroite (Magnusson, 1918).
2. Picrotephroite (Magnusson, 1918).
3. Ironknebelite (Magnusson, 1918).
4. Glaucochroite (Gümbel, 1894).
5. Monticellite (Schaller, 1935).
6. Monticellite (Larsen, Hurlbut, Griggs, Buie, and Burgess, 1941).
7. Monticellite (Hallimond, 1921).

The present diagram includes the following curves:

1. Liquidus-solidus.
2. Specific gravity of crystals.
3. Weight percentages of MgO, FeO, and  $\text{SiO}_2$ .
4. True and apparent optic axial angle.
5. Refractive indices and birefringence.



At present the data are still too scarce to allow for accurate correlations of optical properties with chemical composition.

#### ORTHOPIYROXENE SERIES

Studies of correlations of physical properties and chemical composition in the orthopyroxene series have been published by Walls (1935), Henry (1935), Hess and Phillips (1940), Burri (1941), Kuno (1941), and the present writer (1947). The curves do not differ materially, hence the determination of the composition of orthopyroxenes by optical means appears to be reasonably accurate. Yet there remains scope for further studies on this mineral series.

Since most orthopyroxene cleavage fragments in a crush lie on a (110) face, the  $\gamma$  index can generally be determined with the greatest ease and accuracy. The optic axial angle may show anomalous variations. Hess and Phillips (1940) have found that in deformed (but not recrystallized) anorthosites and gabbros many orthopyroxenes have abnormally large optic axial angles. Intermediate orthopyroxenes of some metamorphic or igneous rocks may have optic angles as low as  $45^\circ$ , although  $51^\circ$  is generally accepted as the minimum optic axial angle of orthopyroxenes of this composition. These anomalous values may be the result of strain, or of the presence of impurities, notably  $Al_2O_3$ .

Kuno (1941) has found that the dispersion about X changes twice in the series; at about  $Of_{13}$  from  $r < v$  to  $r > v$ , and at about  $Of_{47}$  back again to  $r < v$ . Again the presence of impurities may well change the type of dispersion.

The phenomenon of pleochroism in orthopyroxenes is also not clearly understood. Yet it is known that pleochroism is unrelated to the Of content of the mineral.

One of the most intriguing fields of study concerns the co-existence of the orthorhombic and monoclinic pyroxenes; including those of low optic axial angle (pigeonites). Without doubt this problem is as important in petrogenesis as that of the co-existence of plagioclase and alkali feldspars. Bowen and Schairer's results (1935) show that enstatite-orthoferrosilite form a typical isodimorphous series with clinoenstatite-ferrosilite, the monoclinic series consisting of the high temperature modifications. Pigeonites are the natural representatives of the clinoenstatite-ferrosilite series, magmatic conditions apparently favoring the inclusion of small amounts of CaO in the molecule. Crystallization frequently starts with the formation of pigeonite, which on slow cooling inverts to orthopyroxene, the excess lime being expelled just before inversion in the form of a curious exsolution intergrowth. The intergrowth may be called *lamellar* when the optic plane of the clinopyroxene is parallel

ORTHOPYROXENE SERIES.

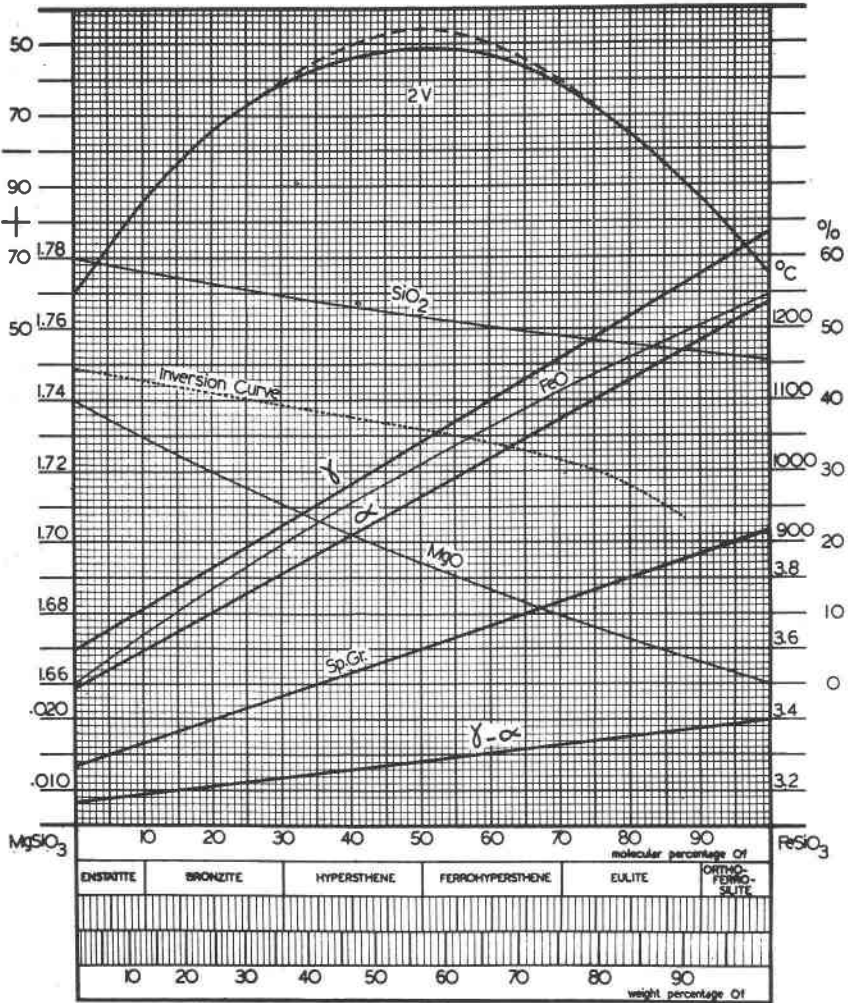


FIG. 3

to (100) of the orthopyroxene host, and *graphic* when it lacks orientation with respect to the host mineral. Many natural orthopyroxenes contain about 1.5 per cent CaO.\* The relationship of orthopyroxene to pigeonite is discussed in detail by Hess and Phillips (1938, 1940), Hess (1941), and the present writer (1947). Hess considers that in magmas two pyroxene phases are normally present between MgO:FeO ratios of approxi-

\* Hess believes that *all* natural orthopyroxenes contain about 1.5 per cent CaO.

mately 75:25 and 35:65, although laboratory investigations prove the existence of complete series of solid solutions between enstatite-diopside, enstatite-orthoferrosilite, and orthoferrosilite-hedenbergite, at the higher temperatures of the anhydrous melts. Guimarães (1946, 1948), on the other hand, collates the resorption of olivine, enstentization of clinopyroxene, and rhythmic zoning of plagioclase, with assimilation of quartz rich sedimentary material. The present writer disagrees with this theory.

The chief admixtures of natural orthopyroxenes are  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{MnO}$ , and  $\text{CaO}$ , while  $\text{NiO}$  and  $\text{Cr}_2\text{O}_3$  may also be present in magnesian orthopyroxenes. Little is known of the influence of these admixtures on the optical properties. In calculating compositions of orthopyroxenes,  $\text{MnO}$  is generally added to  $\text{FeO}$ , while other admixtures are neglected. However, calculations in terms of En, Wo, and Fs are to be preferred when comparing orthorhombic and monoclinic pyroxenes.

Analyses with high percentages of  $\text{Al}_2\text{O}_3$  are regarded by the writer with suspicion, as re-examination often proves the mineral to be orthorhombic amphibole. Thus "bidalotite" (Rao and Rao, 1937) has been shown by Rabbitt (1948) to be anthophyllite. Other modern analyses with unusually high  $\text{Al}_2\text{O}_3$  are given by Lokka (1943) and Rajagopalan (1946).

The present diagram includes the following curves:

1. Inversion curve.
2. Specific gravity of crystals.
3. Weight percentages of  $\text{MgO}$ ,  $\text{FeO}$ , and  $\text{SiO}_2$ .
4. Optic axial angle.
5. Refractive indices and birefringence.

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