## FURTHER STUDIES IN THE PYROXENE GROUP

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Not many years ago mineralogists commonly considered that minerals were substances of simple and definite composition such that any correct mineral analysis could be represented by a simple formula. Innumerable formulas have been proposed for various minerals on the basis of this theory and many names have been created for such substances. Gradually we are learning that minerals vary a great deal in composition. When these variations in composition are continuous from that corresponding with some simple formula to that which must be represented by a different simple formula, it is still customary to consider that each simple formula represents a mineral, whereas, in fact, it represents only one limit in a continuous series of variation. It seems desirable, if not perhaps necessary, to revise our ideas so that our mineral names may correspond in meaning with the actual fact that minerals vary in composition through considerable ranges. It is doubtless true that the limits of variation need names in many cases, especially when these limits correspond with simple formulas. But it would probably lead to a decided improvement in the accuracy of our conceptions if we could discard the notion that these limits (or end-members of variation) are properly called minerals, and reserve that name for the entire series or system of variations. As a concrete illustration, every mineralogist will say that the composition of the common mineral called diopside is given by the formula, CaMg  $Si_2O_6$ ; nevertheless, no sample of this common mineral has yet been found in nature which corresponds exactly with this formula, and the variation from it amounts to very considerable percentages in some cases. Furthermore, these variations may continue without interruption till the formula is no longer even approximately correct. Then it is customary to assume that the substance has some other simple formula (although again it may differ considerably from that formula) and a new mineral name is applied to it. Accordingly, one mineral passes into another mineral by continuous variation, according to our present nomenclature. This condition is a consequence of our idea that minerals must be simple compounds of definite formulas. Would it not be much better to abandon the false idea that minerals are compounds which can be expressed in simple formulas, and accept the idea which expresses the

facts, namely, that nearly all minerals vary in composition through considerable ranges? If that were done, many names now given to minerals would be assigned to end-members, or, if preferred, to a definite (but necessarily arbitrary) portion of the total range of variation. The idea that the composition of any mineral can be expressed in a simple formula is responsible for the fact that in not a few cases no name has yet been given to the entire range of possible variations, although that is the thing which is most appropriately considered to be a mineral. Accordingly, some well known minerals have no name, although the end-members have names. As an illustration, the system magnesite-siderite-rhodochrosite may be mentioned.

Diopside is an end-member of another variation system which has been called pyroxene, in the narrow sense of that term. Diopside varies continuously from  $CaMgSi_2O_6$  to  $CaFeSi_2O_6$  (called hedenbergite), and also to  $MgMgSi_2O_6$  (called clinoenstatite), as well as nearly to  $FeFeSi_2O_6$  (called clinohypersthene). It also seems to vary continuously to  $NaAlSi_2O_6$  (called jadeite) and to  $NaFe-Si_2O_6$  (called acmite). Finally, it varies by including alumina or ferric iron, but only in limited amount, and apparently by forming mix-crystals with  $AlAlAl_2O_6$  or  $FeFeFe_2O_6$  or both. In this case the substance (not the end-member) is called augite.

Not long ago it seemed probable that the maximum tenor of iron in pyroxene<sup>1</sup> could be expressed by the formula MgFeSi<sub>2</sub>O<sub>6</sub>, and consequently this formula was assigned as that of the end-member called clinohypersthene. However, Bowen<sup>2</sup> has shown that a much higher tenor of iron is possible in pyroxene in slags, one sample containing 70 per cent FeSiO<sub>3</sub> (disregarding magnetite and sphalerite present as inclusions, it is about 80 per cent FeSiO<sub>3</sub>).

Bowen,<sup>3</sup> Schairer, Posnjak, and Schaller<sup>3a</sup> have also shown that the optical properties formerly attributed to hedenbergite (based on one determination) are incorrect.

For some purposes diagrams based on molecular proportions seem to be more useful than graphs based on proportions by weight.

Incorporating these changes, the relations between variations in composition and in optical properties in the clinoenstatite-diopside

<sup>&</sup>lt;sup>1</sup> Winchell, A. N., The New Mineralogy: Am. Mineral., vol. 18, p. 81, 1933.

<sup>&</sup>lt;sup>2</sup> Jour. Wash. Acad. Sci., vol. 23, p. 83, 1933.

<sup>&</sup>lt;sup>3</sup> Am. Jour. Sci., vol. 26, p. 260, 1933.

<sup>&</sup>lt;sup>3a</sup> Am. Mineral., vol. 14, p. 319, 1929.

series are shown in the first figure, which is based on the properties of pure artificial crystals as determined by Bowen.<sup>4</sup> It is well known that these mix-crystals are not stable at low temperature, separating into crystals of diopside and enstatite.



FIG. 1. Variations in composition and optic properties in the clinoenstatitediopside series. Data of N. L. Bowen: Am. Jour. Sci., vol. 38, p. 245, 1914.

The second figure shows the relations between variations in composition and optic properties in the diopside-hedenbergite series, including the corrected data for hedenbergite, as determined on artificial crystals,<sup>5</sup> and also on natural crystals.<sup>6</sup>

<sup>4</sup> Am. Jour. Sci., vol. 38, p. 245, 1914.

<sup>5</sup> For diopside: *Am. Jour. Sci.*, vol. **38**, p. 245, 1914. For hedenbergite: *Am. Jour. Sci.*, vol. **26**, p. 260, 1933.

<sup>6</sup> For diopside-hedenbergite series: Am. Jour. Sci., vol. 6, p. 511, 1923; vol. 26,



FIG. 2. Variations in composition and optic properties in the diopside-hedenbergite series.

The third figure shows the relations between variations in composition and in optical properties in the hedenbergite-clinohypersthene series, as determined on artificial crystals<sup>7</sup> as far as 56 mol. per cent FeSiO<sub>3</sub>, with extrapolation guided by measures on a highiron pyroxene found in a slag.<sup>8</sup> In this case also the mix-crystals are nct stable at low temperatures.

The fourth figure shows the relations between variations in composition and in optic properties in the MgSiO<sub>3</sub>-FeSiO<sub>3</sub> series of artificial monoclinic pyroxenes, as very recently determined by N. L. Bowen and J. F. Schairer.<sup>9</sup>

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p. 260, 1933; Am. Mineral., vol. 14, p. 319, 1929. Hedenbergite from Heroult, Calif., has  $N_g=1.752$ ,  $N_m=1.735$ ,  $N_p=1.726$  according to N. L. Bowen-personal communication, Oct. 23, 1934.

<sup>&</sup>lt;sup>7</sup> Am. Jour. Sci., vol. 26, p. 260, 1933.

<sup>&</sup>lt;sup>8</sup> Jour. Wash. Acad. Sci., vol. 23, p. 83, 1933.

<sup>&</sup>lt;sup>9</sup> Am. Jour. Sci., vol. 29, p. 151, 1935.





The fifth figure<sup>'0</sup> shows the relations between variations in composition and in optical properties in the entire clinoenstenitediopside-hedenbergite system. Under ordinary magmatic conditions mix-crystals containing more than about 50 per cent FeSiO<sub>3</sub> are quite unstable, and so this part of the diagram has no significance for rock-forming minerals; the upper left part of the diagram relates only to minerals of volcanic rocks, since the slow cooling of plutonic rocks leads to unmixing of these crystals. Hedenbergite is found chiefly in igneous contact zones and ore deposits, a mode of

<sup>10</sup> T. Tomita (*Jour. Shanghai Sci. Inst.*, **1**, **2**, pp. 41–58, 1934) has recently published a similar diagram, but presents a part of the ternary system: CaSiO<sub>3</sub>-MgSiO<sub>3</sub>-FeSiO<sub>3</sub> as part of a triangular diagram.



FIG. 4. Variations in composition and optic properties in the  $MgSiO_3$ -FeSiO<sub>3</sub> series of monoclinic pyroxenes. N. L. Bowen and J. F. Schairer: *Am. Jour. Sci.*, vol. **29**, p. 151, 1935.



FIG. 5. Variations in composition and optic properties in the clinoenstenitediopside-hedenbergite system. Based partly on data of N. L. Bowen: Am. Jour. Sci., vol. 38, p. 245, 1914, and vol. 26, p. 260, 1933.

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occurrence which suggests crystallization at temperatures lower than those of most magmas. Pyroxenes belonging to this system and not close to the diopside-hedenbergite series in composition are rare,<sup>11</sup> but enough of them have been analyzed and optically measured to make it possible to sketch the curves showing the optic properties. But the correct position of these curves is known much less accurately than the correct position of the curves in the four (boundary) binary series given in the other diagrams. In spite of this fact, it may be stated with some confidence that monoclinic pyroxenes showing considerable variations in optical properties from those expressed in this diagram will be found to differ materially from this system in their composition, for example, by the presence of other pyroxene molecules, or of alumina, in crystal solution. On the other hand, monoclinic pyroxenes, which do not differ materially in optical characters from the properties shown in the diagram, will not differ materially in composition from the system represented. Also, it is already known that alumina has little effect on the optic angle and indices, but a considerable effect (causing an increase of about  $3^{\circ}$  to  $5^{\circ}$ ) on the extinction angle.

<sup>11</sup> Examples follow: 1. Pigeonite, Onega Sea; W. Wahl: *T. M. P. M.*, vol. **26**, p. 27, 1907, with 51% Fe mol. and 43% Ca mol.  $2V=23^{\circ}-40^{\circ}$ ,  $N_{g}=1.740-1.742$ ,  $N_{m}=1.719$ ,  $N_{p}=1.716-1.718$ ,  $Z/c=45^{\circ}$ . 2. Pyroxene, Marquesas; T. F. W. Barth: *Am. Jour. Sci.*, vol. **21**, p. 377, 1931, with 29% Fe mol. and 41% Ca mol.  $2V=46^{\circ}$ ,  $N_{g}=1.728$ ,  $N_{m}=1.701$ ,  $N_{p}=1.695$ ,  $Z/c=39^{\circ}$ . 3. Pigeonite, Aland Island, W. Wahl: *T. M. P. M.*, vol. **26**, p. 18, 1907, with 39.5% Fe mol. and 17.5% Ca mol.  $2V=16^{\circ}-$ 20°,  $N_{g}=1.711$ ,  $N_{m}=1.691$ ,  $N_{p}=1.690$ ,  $Z/c=44\frac{1}{2}^{\circ}$ . 4. Augite, Ob. Veltlin; H. Küchler: *Chem. Erde*, vol. 1, pp. 58, 69, 70 (No. X), 1914, with 25.9% Ca mol. and 36.5% Fe mol.,  $N_{m}=1.7249$ ,  $Z/c=43^{\circ}$ . 5. Pigeonite, Mull; A. F. Hallimond: *Mineral. Mag.*, vol. **17**, p. 97, 1914, with 57% Fe mol. and 9% Ca mol.  $2V=0^{\circ}$ ,  $N_{g}=1.744$ ,  $N_{m}=N_{p}=1.714$ ,  $Z/c=40^{\circ}$ . 6. Pyroxene from slag, N. L. Bowen: *Jour. Wash. Acad. Sci.*, vol. **23**, p. 83, 1933, with 92% Fe mol. and 8% Ca mol.  $2V=20^{\circ}-$ 25°,  $N_{g}=1.785$ ,  $N_{m}=?$ ,  $N_{p}=1.745$ ,  $Z/c=36^{\circ}$ .

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