A review of the data of the Mg-Fe-clinopyroxenes.

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IN recent years several occurrences of clinoenstatite and of clinohypersthene have been recorded, the identification depending on the oblique extinctions measured from a prominent cleavage. This is, of course, quite inconclusive, because many sections of hypersthene show extinction-directions oblique to the trace of a prominent cleavage, and much fuller data would be necessary before most of these occurrences could be accepted as monoclinic Mg-Fe-pyroxenes.

The terms clinoenstatite and clinohypersthene appeared for the first time in 1906 when both W. Wahl and A. Lacroix introduced them for the monoclinic Mg-Fe-pyroxenes which had been often observed in meteorites. Lacroix mentioned several meteorites in which these minerals were reported to have been present, while in his 'Minéralogie de la France' (1910) he noted the French occurrences. The only data given, apart from those on artificial products, are the constant lamellar structure parallel to (100) and the maximum extinction-angle of 22–29°, while the division between clinoenstatite and clinohypersthene was made at 10 % FeO to correspond with the division in the orthorhombic series. Michel (1913) found these minerals in many kinds of meteorites.

J. Ebelmen in 1851 was the first to produce $MgSiO_3$ artificially, but the monoclinic nature of his product was not recognized until 1881 when F. Fouqué and A. Michel-Lévy investigated the artificial production of minerals. In their book they give references to the other early workers who had produced this mineral without realizing that it was monoclinic. In 1906 A. Lacroix obtained the monoclinic form through inversion of the orthorhombic by heating the latter near its temperature of fusion. In this same year the Geophysical Laboratory at Washington began its thermal study of artificial melts with a paper on minerals of the composition MgSiO₃ by E. T. Allen, F. E. Wright, and J. K. Clement (1906). This paper and most of the subsequent ones give optical data on the various minerals produced, but a new form α -MgSiO₃ noted by E. T. Allen and W. P. White (1909) was shown later by N. L. Bowen and O. Andersen (1914) to be really forsterite. The papers of N. L. Bowen (1914) and O. Andersen (1915) give later data on the pure clinoenstatite produced by them. In more recent years, the study of systems containing CaO, MgO, FeO, and SiO₂ has greatly extended our knowledge of the Mg-Fe-clinopyroxene series and the relationships of the various phases are now well known (see N. L. Bowen and J. F. Schairer, 1932 and 1935; N. L. Bowen, J. F. Schairer, and E. Posnjak, 1933). The name 'ferro-silite' suggested by Washington has been adopted by N. L. Bowen (1935) for the FeSiO₃ molecule.

Very little structural work has been done on this series of minerals. In 1925 R. W. G. Wyckoff, H. E. Merwin, and H. S. Washington took powder photographs of several different pyroxenes and they state (p. 385) that clinoenstatite gives a diffraction-pattern which is, apparently, unlike that of any of the other pyroxenes examined. This statement, however, seems to refer to artificially produced clinoenstatite obtained by inverting enstatite from Espedalen at 1400° C. and the authors admit that this is unlikely to produce a pattern exactly similar to that of pure clinoenstatite. B. E. Warren and J. Biscoe (1931) took oscillation photographs of this mineral which showed the same pattern as those of diopside, but they did not state the nature of the actual material used. In 1935 N. L. Bowen and J. F. Schairer used powder photographs in identifying their artificial products. None of these papers give any reproductions of X-ray photographs of the minerals of this series.

In the case of terrestrial rocks, with the exception of the practically pure clinoferrosilite described by N. L. Bowen (1935) from the lithophysae of an obsidian from Naivasha, Kenya, the identification of all the reported occurrences examined rests on insufficient data. S. Tsuboi (1920, p. 83) reports clinohypersthene in a lava as having the same optical properties as the hypersthene which accompanies it, except that in the former the optic axial plane is transverse to the c-axis. In view of the fact that all known clinoenstatites and clinohypersthenes are optically positive and have the optic axial plane more nearly parallel than perpendicular to the c-axis, and that the maximum optic axial angle reported for them is equal to the minimum reported for the hypersthenes, it is obvious that much more data are required before this Japanese occurrence could be accepted as belonging to the series of minerals under discussion, especially as the characteristic polysynthetic twinning is absent in this case. J. Gotthard (1928) also reports an occurrence in a melaphyre of a clinohypersthene, similar to the hypersthene which accompanies it, except that it shows extinction-angles up to 10°. Clinoenstatite showing extinction-angles up to 30° was reported by A. Holmes and H. F. Harwood (1932) as cores of zoned enstatite in lapilli in a melilite-basalt from Uganda. Clinoenstatite is said by E. O. Teale (1928) to occur in a diamondiferous kimberlite pipe in Tanganyika, but no data are given, while P. A. Wagner (1925) describes polysynthetically twinned clinobronzite in a platinum-bearing norite on the grounds of a maximum extinction-angle of 3° 15′ in the symmetrical zone.

In describing the Johnstown meteorite, E. O. Hovey (1925) gives some data and an analysis of a sample of monoclinic pyroxenes which he calls clinohypersthene on the grounds of extinction-angles of about 8-15° and a lamellar structure, which he describes as 'polysynthetic twinning on the clinopinacoid (010)'. But this mineral is optically negative and its other properties agree with those of hypersthene, so the lamellar structure is probably that which is often seen in orthorhombic pyroxenes although as yet unexplained. The 'clinohypersthene' reported by H. Service (1934) has since been identified as hypersthene, the inclined extinctions and lamellar appearance having led to the mistake.¹ In his description of the geology of Dôgo in the Japan Sea, T. Tomita (1936) gives some data on a mineral which he suggests is a clinohypersthene of very low optic axial angle and for which he proposes the name 'gokaite'. The low 2V suggests the presence of some CaO, but the mineral is optically negative, while all the identified occurrences of this series and of the pigeonites are optically positive. Until further data are available it is impossible to include this 'gokaite' in the mineral series under discussion. M. S. Krishnan (1936) has suggested the presence of clinoenstatite in a meteorite along with enstatite on the basis of oblique extinction alone, while a reported occurrence in an igneous rock with more data (J. Verhoogen, 1937) is discussed below. In each of these cases the inclined extinctions are no proof of monoclinic symmetry, while a lamellar structure is quite common in orthorhombic pyroxenes.

In the case reported by J. Verhoogen (1937) from a lava the mineral in question is often zoned and the maximum extinction-angles rise from 7° in the cores to 16° or 18° on the outside. The data given for the hypersthenes are rather vague, but in the same section crystals with 2V 90° and 55° occur. The author thinks that these extinctions cannot be explained by the obliquity of the section if the mineral were ortho-

¹ Personal communication from Mr. Service.

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rhombic; but calculation shows that, for example, in a section inclined at 15° from the prism-zone and at 15° from normality to the optic axial plane the extinction-angle from the slow ray to the trace of c would be $5\frac{1}{2}^{\circ}$ where $2V_{\alpha} = 80^{\circ}$, and 16° where $2V_{\alpha} = 50^{\circ}$. Extinction-angles might well be measured on such a section, and the data are consistent with an orthorhombic pyroxene becoming markedly enriched in iron during crystallization, which explains the rise in refractive index outwards from the core, while the accompanying decrease of 2V over α accounts for the increase noticed in the extinction-angles on these oblique sections. The statement made that changes in pleochroism, colour, and birefringence are not related to change in extinction-angle is quite understandable, for, especially in sections showing low birefringence, which are those with the largest extinction-angles, the increase in birefringence would be hardly noticeable, while pleochroism is not connected with increase in iron.

The conclusion can, therefore, be drawn that no example of monoclinic Mg-Fe-pyroxene has been proved to occur in terrestrial rocks, except that of the iron end-member, clinoferrosilite, which itself may have been metastable and of which the occurrence is unusual. There is a possibility that members of this series may occur as fine intergrowths with other pyroxenes, but this has not yet been thoroughly studied.

In the supplementary volume of Hintze's 'Handbuch der Mineralogie' (1936) crystallographic data on clinoenstatite according to various authors are given; but as no reference is made there to the recent work of N. L. Bowen and J. F. Schairer (1935) and of N. L. Bowen (1935), the available optical data are indicated below. Contradictory statements have been made about the properties of members of this series, but it is now possible to discard some of these and to lay the basis of a table of optical properties which, however, requires much more data. The diagrams of A. N. Winchell (1935) and of T. Tomita (1934), of which the series under discussion forms a part, show the effects of an increase of lime. The optic axial plane for this series is perpendicular to (010), but beyond the value $2V = 0^{\circ}$, with increasing lime, it is parallel to (010). Up to the present, no division has been made between the MgSiO₃-FeSiO₃ series and the pigeonites, both of which are optically positive, and T. Barth (1931) uses the terms 'clinoenstatite' and 'pigeonite' indiscriminately, while G. T. Prior (1920) proposed to extend the former term to cover the pigeonites on the grounds that 'clinoenstatite' has essentially the same meaning as has 'enstatite-augite'. Where lime is appreciable he proposed the term 'calc-clinoenstatite'. But, in view

of our present knowledge, it is desirable to keep the terms 'clinoenstatite', 'elinohypersthene', and 'clinoferrosilite' quite separate from 'pigeonite' or 'enstatite-augite'.

Very few reliable analyses are available of these minerals, but the figure (fig. 1) shows that at present there is a gap between the pigeonites and the series of Mg-Fe-clinopyroxenes which have been prepared artificially and which are said to be common in meteorites, although



FIG. 1. Diagram showing relationship of the pigeonites to the Mg-Fe-clinopyroxenes.

Analysed pigeonites: 1, Mull (A. F. Hallimond, 1914); 2, from slag (N. L. Bowen, 1933); 3, Åland Islands, Finland (W. Wahl, 1906).

none from meteorites has been analysed. Much more reliable and complete data are required for minerals of this composition and this gap may prove to be non-existent, but at present it makes it unnecessary to draw a sharp dividing line between the pigeonites and the minerals of the clinoenstatite-clinoferrosilite series. A mineral which from its properties must lie quite close to this series is the 'pigeonite B' of E. P. Henderson and H. T. Davies (1936) from the Moore County meteorite. Its greater birefringence than the corresponding pure member of the series with the same value of γ shows that it probably contains a little CaO. A few occurrences of pigeonites with the optic axial plane perpendicular to (010) are recorded, but the available data are too scanty to be used in plotting them. T. Barth (1931, pp. 201-202) mentions one from the basalt of Katem quarry, Madras, with 2V 20°. A. Osann (1907, p. 708) describes one from a Tasmanian diabase, and P. D. Quensel (1912, p. 77) one from a Patagonian diabase, both of which have a low optic axial angle and an extinction-angle γ : c of 45°, which makes it impossible to plot them on the latest diagrams of A. N. Winchell (1935) or of T. Tomita (1934) recently improved by W. A. Deer and L. R. Wager (1938). W. Wahl (1907, pp. 6, 29, 64, 65, 68, and 90) gives two examples

of this type of orientation, from the diabase of Richmond, South Africa, and from the Juvinas meteorite, but when the old analyses of these are plotted the points lie on the wrong side of the line $2V = 0^{\circ}$, while a third analysis, from the meteorite of Novo-Urei, lies close to clinoenstatite, but the optic axial plane of the mineral is given as parallel to (010). In view of these discrepancies this point must be left undecided for the present.

N. L. Bowen and J. F. Schairer (1935, p. 199 and fig. 28) give the refractive indices, birefringence, extinction-angle, and optic axial angle of the artificial Mg-Fe-clinopyroxenes prepared by them, while N. L. Bowen (1935) gives data on the naturally occurring clinoferrosilite. In the former paper 2V is given as $20-25^{\circ}$ for the whole series, but in the earlier papers of E. T. Allen and W. P. White (1909, p. 30) and of N. L. Bowen (1914, p. 252) 2V for pure clinoenstatite is given as 53° . The whole series is optically positive with positive elongation and the optic axial plane is perpendicular to (010), while polysynthetic twinning on (100) is always present. The specific gravity of pure clinoenstatite is 3 ·19, but definite data on pleochroism and dispersion are lacking at present.

References.

- E. T. ALLEN and W. P. WHITE, Diopside and its relation to calcium and magnesium metasilicates. With optical study by F. E. Wright and E. S. Larsen. Amer. Journ. Sci., 1909, ser. 4, vol. 27, pp. 1–47.
- E. T. ALLEN, F. E. WRIGHT, and J. K. CLEMENT, Minerals of the composition MgSiO₃. Amer. Journ. Sci., 1906, ser. 4, vol. 22, pp. 385–438.
- O. ANDERSEN, The system anorthite-forstcrite-silica. Amer. Journ. Sci., 1915, ser. 4, vol. 39, pp. 407-454. [Min. Abstr. 2-20.]
- T. F. W. BARTH, Crystallization of pyroxenes from basalts. Amer. Min., 1931, vol. 16, pp. 195–208. [M.A. 5–218.]
- N. L. BOWEN, The ternary system diopside-forsterite-silica. Amer. Journ. Sci., 1914, ser. 4, vol. 38, pp. 207-264.
- ---- Crystals of iron-rich pyroxene from a slag. Journ. Washington Acad. Sci., 1933, vol. 23, pp. 83-87. [M.A. 5-454.]
- ----- "Ferrosilite" as a natural mineral. Amer. Journ. Sci., 1935, ser. 5, vol. 30, pp. 481-494. [M.A. 6-260.]
- N. L. BOWEN and O. ANDERSEN, The binary system MgO-SiO₂. Amer. Journ. Sci., 1914, ser. 4, vol. 37, pp. 487-500.
- N. L. BOWEN, J. F. SCHAIRER, and E. POSNJAK, The system CaO-FeO-SiO₂. Amer. Journ. Sci., 1933, ser. 5, vol. 26, pp. 193-284. [M.A. 5-454.]
- N. L. BOWEN and J. F. SCHAIRER, The system FcO-SiO₂. Amer. Journ. Sci., 1932, ser. 5, vol. 24, pp. 177-213. [M.A. 5-252.]
- --- The system MgO-FeO-SiO₂. Amer. Journ. Sci., 1935, ser. 5, vol. 29, pp. 151-217. [M.A. 6-352.]
- W. A. DEER and L. R. WAGER, Two new pyroxenes included in the system clinoenstatite, clinoferrosilite, diopside, and hedenbergite. Min. Mag., 1938, vol. 25, pp. 15-22.

- F. FOUQUÉ and A. MICHEL-LÉVY, Synthèse des minéraux et des roches. Paris, 1882; and Reproduction artificielle de divers types de météorites. Bull. Soc. Min. France, 1881, vol. 4, p. 279.
- J. GOTTHARD, O pyroxenech z vyvřelin okolí Lomnice nad Popelkou. Časopis Národního Musea, Praha, 1928, vol. 102, pp. 65–71. [M.A. 4-38.]
- A. F. HALLIMOND, Optically uniaxial augite from Mull. Min. Mag., 1914, vol. 17, pp. 97-99.
- E. P. HENDERSON and H. T. DAVIS, Moore County, North Carolina, meteorite—a new eucrite. Amer. Min., 1936, vol. 21, pp. 215–229. [M.A. 6-396.]
- A. HOLMES and H. F. HARWOOD, The petrology of the volcanic fields east and southeast of Ruwenzori, Uganda. Quart. Journ. Geol. Soc. London, 1932, vol. 88, pp. 370-439.
- E. O. HOVEY, A new meteoric stone from Johnstown, Weld County, Colorado. Amer. Mus. Novitates, 1925, no. 203. [M.A. 3-256.]
- M. S. KRISHNAN, The Tirupati and Bahjoi meteorites. Rec. Geol. Surv. India, 1936, vol. 71, pp. 144-149. [M.A. 6-394.]
- A. LACROIX, Le météorite de Saint-Christophe-la-Chartreuse. Bull. Soc. Hist. Nat. Ouest, Nantes, 1906, ser. 2, vol. 6, pp. 81–112.

er Klinoenstatit der Meteoriten. Centr. Min., 1913, pp. 161-163.

- A. OSANN, Ueber einen Enstatitaugit führenden Diabas von Tasmanien. Centr. Min., 1907, pp. 705-711.
- G. T. PRIOR, The classification of meteorites. Min. Mag., 1920, vol. 19, pp. 51-63.
- P. D. QUENSEL, Geologisch-petrographische Studien in der Patagonischen Cordillera. Bull. Geol. Inst. Upsala, 1912, vol. 11, p. 77.
- H. SERVICE, Note on the occurrence of clinohypersthene and enstatite-augite in the "norite" from Bluff, New Zealand. Trans. Roy. Soc. New Zealand, 1934, vol. 64, pp. 147-150. [M.A. 6-211.]
- E. O. TEALE, Tanganyika Territory; its geology and mineral resources. Mining Mag. London, 1928, vol. 39, p. 79. [M.A. 4-182.]
- T. TOMITA, Variations in optical properties, according to chemical composition, in the pyroxenes of the clinoenstatite-clinohypersthene-diopside-hedenbergite system. Journ. Shanghai Sci. Inst., Sect. 2, 1934, vol. 1, pp. 41-58. [M.A. 6-71.]
- ----- Geology of Dôgo, Oki islands, in the Japan Sea. Journ. Shanghai Soi. Inst., Sect. 2, 1936, vol. 2, pp. 37-146.
- S. TSUBOI, Volcano Öshima, Idzu. Journ. Coll. Sci. Tokyo, 1920, vol. 43, art. 6, pp. 1-146.
- J. VERHOOGEN, A monoclinic "hypersthene" from the Cascade lavas. Amer. Journ. Sci., 1937, ser. 5, vol. 33, pp. 63-69. [M.A. 7-24.]
- W. WAHL, Die Enstatitaugit. Helsingfors, 1906; and Tschermak's Min. Petr. Mitt., 1907, vol. 26, pp. 1-131.
- P. A. WAGNER, The platinum deposits in the western part of the Lydenburg district, Transvaal. South African Journ. Industries, 1925, vol. 8, pp. 90-113. [M.A. 2-440.]
- B. E. WARREN and J. BISCOE, The crystal structure of the monoclinic pyroxenes. Zeits. Krist., 1931, vol. 80, pp. 391-401. [M.A. 5-186.]
- A. N. WINCHELL, Further studies in the pyroxene group. Amer. Min., 1935, vol. 20, pp. 562-568. [M.A. 6-294.]
- R. W. G. WYCKOFF, H. E. MERWIN, and H. S. WASHINGTON, X-ray diffraction measurements upon the pyroxenes. Amer. Journ. Sci., 1925, ser. 5, vol. 10, pp. 383-397. [M.A. 3-186.]