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ON THE TRICLINIC MANGANIFEROUS PYROXENES

N. SUNDIUS, *Stockholm, Sweden.*

The following minerals are generally regarded as triclinic pyroxenes: rhodonite, bustamite, fowlerite, iron rhodonite, sobralite, and pyroxmangite. All these mineral species represent metasilicates of manganese with smaller or greater admixtures of $CaSiO_3$, $MnSiO_3$ and $FeSiO_3$. Fowlerite also contains ZnO, but it is uncertain if the whole amount of ZnO in reality enters in the constitution of the mineral and to what extent this oxide is caused by impurities. Besides the species named babingtonite also has been considered as a triclinic pyroxene, but the relations of this compound are still but little known. According to the available analyses it differs essentially from the first mentioned minerals through the presence of a considerable amount of sesquioxides, furthermore its manganese content is restricted. On these grounds babingtonite is not included in this work.

Among the manganese-rich triclinic pyroxenes rhodonite is often found developed as good crystals, and it has early attracted the attention of mineralogists. Goniometrically it has been thoroughly investigated, and several valuable works have been written on this subject, among which may be named those of G. Flink[1] and A. Hamberg.[2] As the mineral is triclinic, the choice of crystallographic axes is arbitrary and has varied with different authors, but most frequently the similarities with the monoclinic pyroxenes have been decisive for the orientation of the crystals. This orientation is to a great extent based on the cleavage relations. As in the monoclinic pyroxenes we find in the rhodonites two perfect cleavages, intersecting each other at about 87°30′ and in addition thereto two more inferior cleavages bisecting approximately the angles of the former. Furthermore, an inferior cleavage or parting according to (001) is often present, forming an angle of about 70° with the axis of the chief cleavages, the *c*-axis. Similar cleavage relations are found in all the other triclinic pyroxenes mentioned at the beginning of this

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[1] *Zeitschr. f. Kryst.*, 1886, Vol. **11**, p. 506.

[2] *Geol. Fören. Förhandl.*, Vol. **13**, p. 545, 1891.

paper, though the angle of the chief cleavages deviates somewhat in the bustamites.

On the triclinic pyroxenes a great number of analyses have been published, most of them relating to rhodonites and bustamites, the iron-rich species being more rare. On the whole the chemical relationships of the minerals must be considered rather well known. In spite of this it is a noteworthy fact that very little is definitely known about the general relations of the minerals. There are only a few reliable determinations of the optical properties and for some species they are quite lacking. Also the experimental work in the corresponding silicate systems has not been satisfactorily performed.

In the following an attempt is made at a more general treatment of the mineral group mentioned, especially as to the optical and chemical properties of the different members and as to the equilibrium relations in the system in which they fall. The very reason for this work was the discovery of a new locality of iron rich rhodonite, made several years ago by the writer at the mine of Tuna Hästberg in central Sweden.[3] The statement of the properties of this rhodonite made a closer investigation of the older known instances of related minerals in Sweden (Tunaberg and V. Silvberg) necessary and in connection with this a series of determinations on rhodonites and bustamites was started. When most of the necessary researches had been completed, the interesting paper by M. H. Hey on rhodonites and bustamites appeared.[4] In this paper a critical review of older determinations is given as also a statement on the true optical orientation. Concerning the first mentioned part of the research a reference at this place to the author named will be sufficient. Other results of a more general nature reported by Hey will be referred to in the discussion. Nearly simultaneously with the paper by Hey two publications appeared by H. Buttgenbach[5] and J. Melon[6] on the optical orientation of rhodonite. To these, too, further references will be made.

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In accordance with the custom adopted for the monoclinic pyroxenes the axis of the zone of the chief cleavages is regarded in the following discussion as the vertical c-axis and chosen as the central

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[3] *Geol. Fören. Förhandl., Stockholm,* Bd. **52,** p. 403, 1930.

[4] *Mineral. Mag.,* Vol. **XXII,** p. 193, 1929.

[5] *Acad. Royale de Belgique, Bull. Ser. V,* T. **XVI,** 1930, p. 35.

[6] *Ibid.,* p. 66.

point in the stereographic projections. This orientation is maintained for all the different minerals in order to get a better general survey, although, as will be seen in the following, a different orientation would correspond better to the conditions found in the bustamites. The measurements of the relative positions of the chief optical directions were made on a universal stage. For the measurements only sections have been used which permit the orientation of two vertical cleavages within the field of observation. In most cases the sections have been cut at angles greater than 60° to the vertical zone. In exceptional cases twinning lamellae parallel to (010) or sections parallel to a cleavage have been used. As the measurements comprise data of two sets of different types to be oriented, the crystallographical and the optical, and a later transformation on the stereographic net, the errors are rather large and the various measurements seldom agree exactly with each other. Even when the material used is excellent and the crystals are cut at very favorable angles to the cleavages and the bisectrices, differences from 2–3 degrees in the coordinates are not uncommon. In less favorable cases the differences may be considerably greater. On account of this a repetition of the measurements is necessary and when possible this has been made on sections of different grains. In this way the errors are diminished and generally they would seem not to exceed one or two degrees unless the material has been inferior (showing undulatory extinction or deformation of the cleavages).

The measurement of the axial angles, on the other hand, can be made with considerably greater exactness (errors about 0.5°), if both the axes are accessible.

Concerning the refraction, the only exact method of determination applicable to triclinic minerals of the present type is the use of polished plates on a total refractometer. The immersion method is less reliable, since the minerals split very readily parallel to the cleavages and all the chief optical directions are oblique to them. If it was impossible to get polished plates the values recorded through immersion have been checked by using sections perpendicular to the bisectrices and by measurements of the double refraction and of the axial angle.[7]

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[7] In all rhodonites and bustamites the bisectrix γ forms the smallest angle with the axis of the vertical cleavages and is best accessible in the powder. Hence only this bisectrix has been determined in many cases, when the immersion method has been employed. α and β were calculated from the double refraction and from the axial angle.

All the determinations on refraction were made with sodium light, except in those cases where the dispersion has been measured. For the researches made with the microscope white light was used. Errors originating from the strong dispersion of the axes or of the bisectrices, were reduced through screening in the objective and when necessary also in the condensor.

The determinations of the specific gravity made for this work refer to a temperature of 20°.

The material investigated by me comes for the most part from the collections of the mineralogical department of the State Museum in Stockholm. The specimens originating there are designated by the letters RM. From Professor A. Hamberg I received from his personal collection crystals from Harstigen, corresponding to the types analyzed by him. Furthermore I received from Professor A. Grönwall the original specimens from V. Silvberg, previously investigated by M. Weibull and belonging to the collections of the Mineralogical Inst. of the University of Lund. To all the gentlemen mentioned and to the director of the Mineralogical Department of the State Museum, Professor G. Aminoff, I wish to express my cordial acknowledgments.

Most of the new analyses necessary for the investigation have been performed by the chemist of the Geolog. Survey of Sweden, Dr. A. Bygdén. For his careful work and kind assistance I am very grateful. Also to Lic. Phil. G. Assarsson, assistant chemist at the Survey, I am much obliged for his assistance in making two analyses.

Owing to the readiness with which the minerals split along the cleavages the preparation of the slides is a somewhat complicated matter, especially if the specimens used are single crystals or single crystal fragments and the section is made at a large angle to the cleavages. To get usable slides in certain cases it has been necessary to develop a method of melting the fragments in glass (ordinary Na-glass or Pb-glass, melting point about 800–900°). Most of this work was done by the preparator of the Geol. Survey, G. Larsson.

GENERAL RELATIONS

In the triangular diagram in Fig. 1 all reliable analyses of natural triclinic Mn-rich pyroxenes known to the present writer from the literature, as well as those made for this work, have been plotted according to their content of $CaSiO_3$, $MnSiO_3$ and $FeSiO_3$, calculated as mol. per cent.[8] The natural minerals are never quite pure but hold some admixtures of MgO, often also of BaO and ZnO, the last-mentioned oxide especially being known from the Franklin Furnace specimens. Of these oxides BaO has been included with CaO. The amount of BaO is always very small and plays no part, the amount of MgO is greater, in most cases 0.5–1 and in certain cases 1.5–4.5 per cent by weight. This oxide has been included with FeO as being chemically most closely related to it. If MgO were omitted, the points on the diagram would move away from the $FeSiO_3$ corner but the displacements generally would be small and the general aspect of the diagram would not change.

The ZnO, contained in some analyses is not taken into consideration at all. This was done on the supposition that the presence of it is chiefly due to impurities of enclosed willemite grains, which cannot be avoided unless ultraviolet light is used. On the other hand, as is shown by the work of Hey and by the analysis No. 10 in this paper, the amount of the remaining ZnO, when the mineral powder is treated in this way, will be quite small and negligible.

A list of the analyses used for the diagram is given below. Of the older analyses all showing an excess or a deficit of SiO_2 greater than 3 per cent by weight, or holding considerable amounts of H_2O and CO_2 are excluded. The analyses of hedenbergite and wollastonite are taken from the mineralogical handbooks with the exception of two of the hedenbergites which correspond to the "green rhodonite" and "Herault" of the work of Wyckoff, Merwin and Washington on the pyroxene group.[9] Only hedenbergites with less than 5 per cent by weight of MgO have been considered.

1. Rhodonite, Vittinge, Finland, new, this paper No. 1, Table 1.
2. “ “, M. Saxén, *Fennia*, **45**, No. 11, p. 15, 1925.
3. “ Långban, *Hintze*, **II**, p. 1165, No. IX.
4. “ Viu at Turin, *Hintze*, **II**, p. 1165, No. V.
5. “ St. Marcel, Piemont, L. Colomba, *Atti R. Acc. Torrino*, **39**, 644, 1904.
6. “ Csucsom, *Hintze*, **II**, p. 1165, No. III.
7. “ Harstigen, Sweden, A. Hamberg, *Geol. Fören. Förh.*, **13**, p. 572, 1891.
8. “ “ “ “ “ “ “ “ “ “
9. “ “ “ “ “ “ “ “ “ “
10. “ “ “ “ “ “ “ “ “ “

[8] The small percentages of Fe_2O_3 present in the analyses have been calculated as FeO. In some cases all the iron is determined as Fe_2O_3, but this oxide as well as Al_2O_3 is probably always subordinate in the triclinic pyroxenes.

[9] *Am. J. Sc., Ser. 5*, Vol. **10**, 1925, p. 383.

11. Rhodonite, Val d'Err, Grisons, Switzerland, J. Jakob, *Min. Mitt.*, **1923,** Vol. 3, p. 236.
12. " Harstigen, Sweden, M. H. Hey, *Mineral. Mag.*, Vol. **XXII,** p. 193, 1929.
13. " Harstigen, Sweden, A. Hamberg, *Op. cit.*
14. " Långban, new, this paper, No. 4, Table 1.
15. " " " " " " 6, " ".
16. " Tetela, Mexico, *Hintze,* **II,** p. 1165, No. XXVIII.
17. " Långban, new, this paper, No. 5, Table 1.
18. Iron rhodonite, Gåsborn, Sweden, new, this paper, No. 11, Table 1.
19. " " Algier, *Hintze,* **II,** p. 1165, No. XXIX.
20. " " Tuna Hästberg, Sweden, new, this paper, No. 12, Table 1.
21. Bustamite, Tetela, Mexico, *Hintze,* **II,** p. 1165, No. XXVI.
22. " Radautal, J. Fromme, *T. M. P. M.*, **28,** 308, 1929.
23. " Långban, new, this paper, No. 9, Table 1.
24. " *Hintze,* **II,** p. 1165, No. VII.
25. " Långban, M. H. Hey, *op. cit.*
26. " " G. Lindström, *K. V. A. Övers.*, **1880,** 53.
27. " Franklin Furace, new, this paper, No. 10, Table 1.
28. " Långban, new, this paper, No. 7, Table 1.
29. " Franklin Furnace, M. H. Hey, *op. cit.*
30. " " " E. S. Larsen and E. V. Shannon, *Am. Mineral.*, 1922, Vol. **7,** p. 95.
31. Sobralite, V. Silvberg, M. Weibull, *K. V. A.*, *Övers.*, Stockholm, 1884, No. **9,** p. 29.
32. " " " new, this paper, No. 14, Table 1.
33. Sobralite+iron rhodonite, Tunaberg, J. Palmgren, *Bull. Inst.*, *Upsala,* Vol. **XIV,** p. 172, 1917.
34. " + " " " new, this paper, No. 16, Table 1.
35. Pyroxmangite, W. E. Ford and W. M. Bradley, *Am. J. Sc.*, Ser. 4, Vol. **36,** p. 169, 1913.
36. Mn-Hedenbergite, V. Silvberg, M. Weibull, *op. cit.*

On account of the distribution of the points in the diagram and with regard to the relations found in the slides of the pyroxenes studied, an attempt has been made to sketch the stability relations in the system composed of the three pure metasilicates. The relations found are rather simple. In the triangle we find two simple compounds, rhodonite and wollastonite and in addition hereto three composite compounds, hedenbergite, bustamite and sobralite. With this latter pyroxmangite is provisionally included, though the properties of this mineral are still to a great extent unknown. The pure $FeSiO_3$-pyroxene is not known among the natural minerals, nor is it obtained in experimental researches. The curves delimiting the stability fields are given with full lines in those parts where information has been gained from the paragenesis of two or three of the minerals in the same specimen. In most of these cases the minerals appear in a graphical or homoaxial intergrowth with each other. For the rest the course of the curves is drawn approximately on account of the distribution of the analyses.

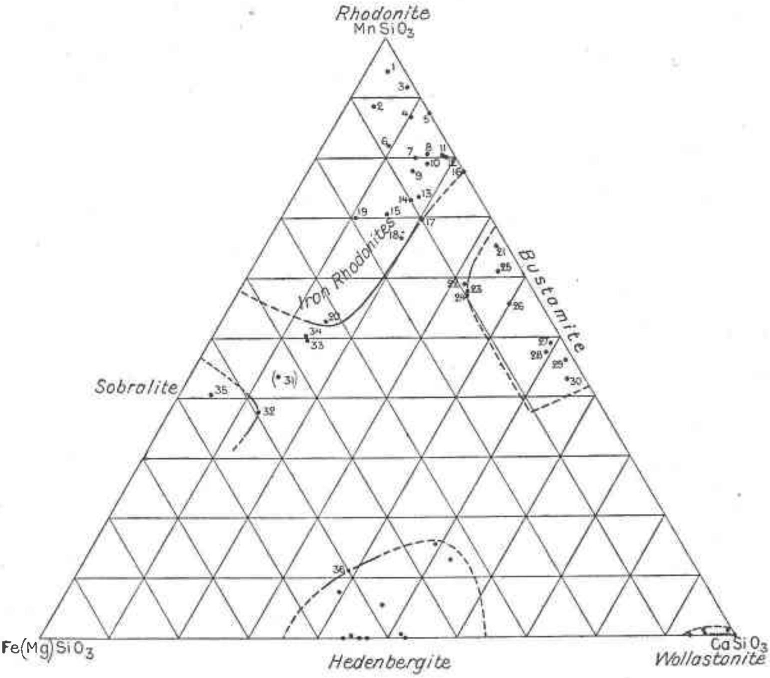

FIG. 1.

From the optical properties of the triclinic minerals described below, and from the determinations of their specific gravities, it appears that the relations between all the triclinic pyroxenes are non-isomorphic. The same holds good also for the triclinic pyroxenes and hedenbergite. With regard to the series bustamite—wollastonite, on the other hand, the relations found speak in favor of a true isomorphism, but in this series complications which will be discussed later enter and produce a discontinuity.

Rhodonite can take up Fe (Mg) SiO_3 to an extent of about 32–42 mol. per cent without losing its rhodonitic character. For those members of this series which are richer in $FeSiO_3$ than about 10 mol. per cent, the name iron rhodonite is considered to be appropriate by the present writer.[10] In no case are the iron rhodonites so far known poorer in $CaSiO_3$ than 10 mol. per cent, in most cases they contain 15–20 mol. per cent of this silicate. The highest Fe-rich iron rhodonite known is the Tuna Hästberg mineral described by the present writer in the paper just referred to (point 20 in the present diagram). It is homogeneous, but it may lie in the vicinity of the boundary of the stability field. This conclusion is drawn from the relations in the specimen from Tunaberg originally described by J. Palmgren and J. M. Sobral[11] and further examined by me in this paper. The analyses answering to this specimen are Nrs. 33 and 34. In the corresponding slides we do not find a homogeneous mineral but a mixture of three pyroxenes besides other minerals, removed from the analyzed powders by a careful separation. The

FIG. 2. Sobralite and iron rhodonite, homoaxially intergrown with each other. White = sobralite, grey = iron rhodonite. In the latter twinning lamellae parallel to (010). The partly clouded appearance of the sobralite is due to the presence of small, newly formed inclusions, probably of grünerite. The same mineral is also found as inclusions in the iron rhodonite, arranged along lines parallel to (001). Somewhat larger prisms of grünerite are seen at more points. Magn. 48×, crossed nicols.

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[10] Compare *Geol. Fören. Förh., Stockholm,* Bd. **52,** p. 403, 1931.

[11] *Bull. Geol. Inst., Upsala,* vol. **XIV,** 1917, p. 173; and vol. **XVIII,** 1921, p. 47.

pyroxenes present are iron rhodonite, sobralite and a diopsidic hedenbergite, all for the most part homoaxially intergrown with each other (Fig. 2). Of the pyroxenes the iron rhodonite is clearly present in greater quantity than the sobralite. The monoclinic pyroxene is present only in subordinate amounts. The mixture corresponding to the two points in the diagram, therefore, is not stable but has split up into three components. The place of the sobralite in the diagram is approximately stated by the analysis of the V. Silvberg-mineral (point 32) which optically is identical with the sobralite, from Tunaberg. Hence the boundary of the stability field of the iron rhodonites must be situated between the points 33–34 and the point 20 of the Tuna Hästberg iron rhodonite; and, as is shown later, the boundary of the sobralite-field is to be drawn through the sobralite-point. Thus there exists a distinct discontinuity area between sobralite and the iron rhodonites.

The unstable area between the sobralite and the iron rhodonites, on the one hand, and the monoclinic hedenbergites, on the other, must be considerably larger. According to the analysis quoted by Palmgren (referred to as No. 19 of Table 1 in this paper) $MnSiO_3$ in the diopside from Tunaberg amounts to about 6.2 mol. per cent. However, this pyroxene holds a considerable quantity of $MgSiO_3$ (20.7 mol. per cent) and belongs to a Mg-richer series than the diagram here presents. More relevant information is gained from the V. Silvberg minerals. In the specimens formerly examined by Weibull both iron rhodonite and sobralite are seen mixed with the monoclinic pyroxene named by him Mn-hedenbergite (No. 36 in the diagram), which holds 11.2 mol. per cent of $MnSiO_3$ but only 8.7 mol. per cent of $MgSiO_3$. Thus the area of discontinuity may run from the point represented by the Mn-hedenbergite to the points of the two minerals named. Of these no analysis is available of the iron rhodonite but optically it is about identical with the Tuna Hästberg rhodonite.

Rhodonite can dissolve about 20–22 mol. per cent of $CaSiO_3$, but from this limit it is not miscible with the bustamite. The discontinuity stretches from the limit named to 32–33 mol. per cent of $CaSiO_3$. The bustamites are known to contain a maximum $CaSiO_3$-content of 57 mol. per cent. The extent of the discontinuity area between the rhodonites and the bustamites is determined from a specimen received from the British Museum and originating in Långban (Nos. 5 and 9 of Table 1, Nos. 17 and 23 in the triangular diagram). In it rhodonite and bustamite are intergrown with each other in a granophyre-like manner (Fig. 3). Of the bustamite an analysis has been made previously by Hey, who kindly sent me the specimen. The two minerals, the bustamite and the rhodonite, have later been separated by me from each other and analyzed.

No case of paragenesis between bustamite and heden-gergite is known to the present writer. It is true that a chemical mixture corresponding to such a case would seem seldom to be realized in the natural rocks and mineral deposits. However, from the relations known it is evident that a discontinuity must exist. This can be concluded from the marked differences as to the optical and other properties of both the compounds. Furthermore, the quantities of iron in the bustamites are constantly very low (3.2 mol. per cent at most in the modern analyses; in one old analysis—*Hintze*, No. IV, p. 1165–, rejected for this work, FeO = 6.54 per cent by weight). In an Mg-free system the limit of the bustamite field

FIG. 3. Rhodonite (white) and bustamite (dark) graphically intergrown with each other. The small inclusions of a lamellar or tabular shape, occurring above the greater part of the bustamite, consist of diopside-perthite. The more irregular, partly ramified inclusions in the rhodonite in the upper part of the figure and to the right of the greater part of bustamite consist of calcite. Magn. 28×, crossed nicols.

would possibly lie at about 5 mol. per cent of $FeSiO_3$. The present point of the limit (10–11 mol. per cent) is influenced by the content of $MgSiO_3$, the effect of which is rather conspicuous on account of the low content of iron. The area of the hedenbergites, on the other hand, extends further in the direction of the bustamites, but even the most Mn-rich specimens known contain not more than 15 mol. per cent of $MnSiO_3$.

The relations of bustamite and wollastonite form a matter of great interest. When an orientation of the bustamites is chosen, which is in accordance with that of the rhodonites and the monoclinic pyroxenes, no similarities as to the crystallographical or optical properties are apparent in comparing bustamite with wollastonite. But if we choose the axis of the cleavage zone (the *c*-axis in the general orientation) as the *b*-axis, the similarities become evident. As will be shown in the discussion of the optical properties the bustamites alter their optical orientation markedly with increasing amount of CaO, and the change runs in the direction towards what is found in wollastonite. The cleavage angle of both the minerals is about 95°, but in all the other pyroxenes the angle is 91°40′–92°30′. Finally the optical properties and the specific gravity of wollastonite show a close agreement with the extrapolated values of the bustamites. Thus there are many reasons to suspect real isomorphic relations between these two compounds. Notwithstanding this, it is a remarkable fact that connecting mixtures between the two mineral species are quite lacking. The cause of this might be sought for in the absence of corresponding chemical mixtures in nature, but this argument seems not to be substantiated as the Mn-rich triclinic pyroxenes can be combined with free $CaCO_3$ and quartz or appear in carbonate layers. The real reason of the absence of intermediate forms in the series must rather be sought in the dimophism of the substance $CaSiO_3$ and the position of the inversion point in the system $CaSiO_3$–$MnSiO_3$. Of the two modifications known of $CaSiO_3$ only that which is stable at the lower temperature (below 1200°) can be regarded as isomorphous with the bustamites, the pseudo-wollastonite showing rather great dissimilarities compared with the latter. The forming of mix crystals, therefore, is restricted by the temperature limit of the inversion point, on the one hand, and by the eutectic point of bustamite-rhodonite on the other. The determination of the latter is not yet to be regarded as conclusive, but it points to about 1150°. Also if the real eutectic temperature is somewhat lower than this figure, and the position of the inversion point may be somewhat altered through the admixture of greater amounts of $MnSiO_3$, the range of temperature within which mix crystals of $CaSiO_3$ and $MnSiO_3$ are stable must be rather limited and be restricted to the vicinity of the discontinuity between rhodonite and bustamite (compare the schematic diagram in Fig. 4). The only possibility of getting mix crystals of greater Ca-content would be if the melting point of wollastonite could be realized, or if the crystallization temperature, through the influence of other substances present, could be lowered considerably below the liquidus curve and below the inversion point. The absence of corresponding crystal products shows that this is not realized in nature. The possibility of obtaining intermediate mixtures must therefore be restricted, and in spite of the

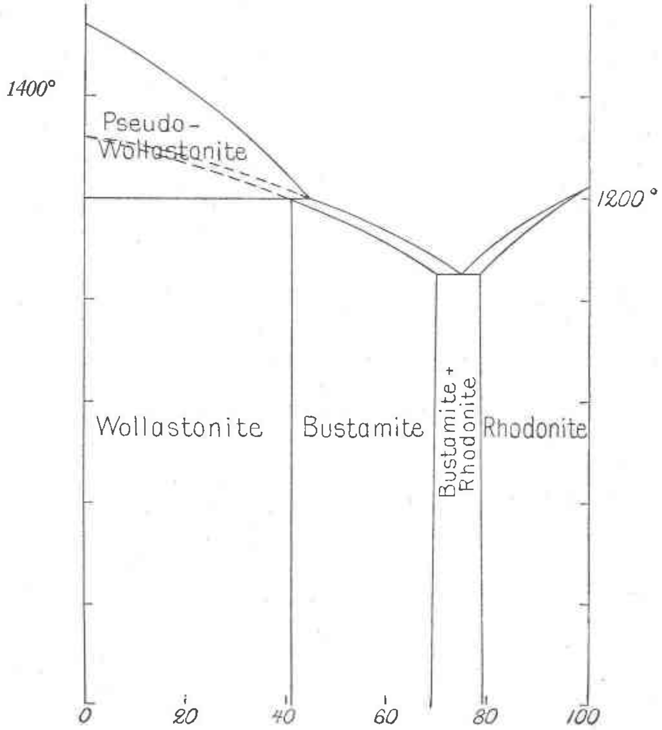

FIG. 4. Schematic diagram of the system $CaSiO_3$–$MnSiO_3$, chiefly constructed from the relations in the natural minerals.

fact that bustamite and wollastonite offer good isomorphic relations as to their properties, the corresponding system notwithstanding shows a marked discontinuity. In fact this discontinuity is greater than in any other of the binary systems present in the ternary diagram.

If these considerations are correct, the bustamites should be regarded as Mn-rich mix crystals of wollastonite rather than as independent minerals. The most convenient orientation of the bustamites should be the following: the cleavage face here designated as $(\bar{1}10)$ corresponds to (001) and the cleavage (110) to (100).

It should be remembered that the conception of a close relationship between bustamite and wollastonite is by no means new. As early as 1892 A. Hamberg[12] pointed out that the rhodonites (including bustamite) show as close relations to wollastonite as to the monoclinic pyroxenes. His opinion is based *inter alia* on the great capability of rhodonite to dissolve lime (in the bustamites). Among earlier orientations of the rhodonites those of v. Kokscharow and Des Cloizeaux are similar to that proposed here for the bustamite in choosing the axis of the vertical zone as the crystallographic *b*-axis. Recently B. Gossner and K. Brückl,[13] on the basis of researches on the atomic structure of rhodonite, have proposed an orientation differing only from that adopted by the present writer for the bustamite in using the opposite directions. However, no one of the authors named has realized the non-isomorphic relations between rhodonite and bustamite and the isomorphism between bustamite and wollastonite, nor has the chemical discontinuity between both the first named species been known previously.

The material investigated also gives some information concerning the equilibrium relations existing between rhodonite and diopside. In the specimens examined from Långban a small amount of diopside is present in most cases. This may be a Fe-poor species, because the diopsides analyzed from the Långban mines are of this type.[14] Furthermore the refraction is lower than that of rhodonite. In most cases the diopside appears as perthitically included small lath-shaped or more irregular patches, extended and ar-

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[12] *Op. cit.*, pp. 552–553.

[13] *Centralbl. f. Min., Geol. Pal.*, **1928**, Abt. A, p. 316.

[14] Two analyses of W. Hissinger and G. Rose show $MgO = 16.99–17.81$ per cent by weight, $Fe_2O_3 = 2.16–2.18$, "Mn-oxide" $= 1.45–1.59$. *S. G. U.* ser. Ca, No. 23, p. 35.

ranged parallel to (001) (Figs. 5–6). The dimensions of the inclusions decrease in size to those of minute lamellar interpositions or appear also as irregular streams arranged in the manner mentioned above. In the very MgO-rich rhodonite, investigated by me, small isolated but non crystallographically arranged grains of diopside are also present. Apparently the first-mentioned (perthite-like) inclusions are due to a separation of diopside from the rhodonite in the solid state, whereas the irregularly orientated grains have separated as free individuals at the same time as the rhodonite, or earlier. The amount of the perthitically enclosed diopside is small, perhaps 2–3 mol. per cent at most. However, the separation of the mineral is not complete, some parts of the rhodonites being clouded by the inclusions, others being clear and homogeneous. The greater part of the MgO of the analyses, therefore, belongs to a homogeneous solution of rhodonite and diopside. The highest Mg-content

FIG. 5. Rhodonite from Långban (Specimen 2), section about perpendicular to (001) and (110), and showing traces of the cleavage according to the latter face. In the rhodonite a few isolated and irregularly bounded grains of diopside are present. Furthermore numerous smaller, lamellar inclusions of the same mineral are visible arranged parallel to (001). To the left a fissure like line is seen along which the diopside–"perthite"–inclusions are abundant. Magn. 48×, crossed nicols.

rhodonite is No. 6 of Table 1 (the same as No. 15 of the triangular diagram), and it is this specimen which holds the isolated diopside grains in addition to the perthitic inclusions. For the analysis the powder was repeatedly treated with the solution of Clerici until a quite homogeneous fraction of the powder as far as spec. gravity is concerned was obtained. Most of the "free" diopside grains may have been avoided in this manner but not the perthitic inclusions. The resulting analysis, therefore, may be considered to mark the limit of the stability field of the rhodonites at the very crystallization temperature. Its $MgSiO_3$ content is 14.2 mol. per cent. On the other hand, rhodonite No. 5 of Table 1 (the same as No. 17 of the triangular diagram of Fig. 1) holds some sparse perthite inclusions of diopside. It may lie near the limit of the stability curve at a lower temperature. Its $MgSiO_3$-content is 6.2 mol. per cent. This is the only convincing case of a separation in the solid state found by the writer in the minerals investigated. It should be mentioned that no similar perthitic separation products are found in

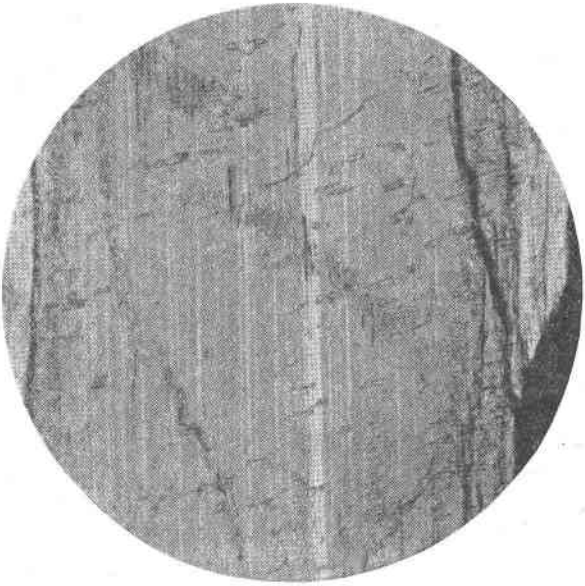

FIG. 6. The same rhodonite as in FIG. 5. Section about perpendicular to α. The white lines represent twinning lamellae parallel to (010). The rhodonite holds the same diopside-perthite-inclusions as in Fig. 4, and similar perthite-rich lines as in the latter. Magn. 48×, crossed nicols.

the bustamites. Also in the specimens answering to Nos. 5 and 9 of Table 1, in which the bustamite is graphically intergrown with rhodonite, the former is homogeneous though the rhodonite holds perthitic segregations of diopside. In both minerals the amount of MgO is similar (MgO = 5.9 and 6.2 mol. per cent). The solubility of the diopside, therefore, is greater in the bustamites than in the rhodonites.

This conclusion is valid only for the Fe-poor rhodonites. In the iron rhodonites the Mg-content is greater (up to 6 mol. per cent of $MgSiO_3$) without any separation of the Mg-component.

If we altered the diagram of Fig. 1 so as to form a tetrahedron and placed $MgSiO_3$ at the top, we should get a complete picture of the system of the sesquioxide-free natural pyroxenes. Thanks to the works of earlier authors, especially to those of R. B. Sosman[15] and B. Asklund,[16] we can imagine some of the chief features of the distribution of the minerals in it. Along the $FeSiO_3-CaSiO_3-MgSiO_3$-side we should get a continuous space area between hedenbergite, diopside and enstatite-clino-enstatite with the exception of a discontinuous area in the vicinity of the $MgSiO_3$-corner. The mentioned area of orthorhombic and monoclinic pyroxenes is in all probability separated from the areas of wollastonite-bustamite through a discontinuity area which extends to the corresponding gaps between bustamite-rhodonite and between iron rhodonite and sobralite. Whether any separate smaller mineral groups are situated in the interior of the tetrahedron, we do not know as yet. On account of the relatively low percentage of MgO in all the triclinic pyroxenes analyzed it does not seem probable that the stability areas of them will rise to any greater heights in the tetrahedron.

An artificial mineral perhaps related to the iron rhodonite or to the sobralitic group is the triclinic vogtite described by C. Hlavatsch[17] and A. F. Hallimond[18] from slags. However, the position of the mineral is not clear from the facts available. The analysis of Hallimond shows the following ratios of the chief oxides: $MnSiO_3 = 21.75$, $FeSiO_3 = 30.43$, $MgSiO_3 = 15.67$, $CaSiO_3 = 32.14$ mol. per cent.

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[15] *Journ. Wash. Acad. Science,* **1911,** 1, p. 86.

[16] *S. G. U., Ser. C,* No. **325,** p. 75. A diagram of the natural rock forming pyroxenes had already been compiled considerably earlier by H. E. Johansson (compare Asklund, p. 76).

[17] *Zeitschr. f. Krist.,* **42,** 1906–1907, p. 590.

[18] *Mineral. Mag.,* vol. **XVIII,** 1919, p. 369.

In the ternary diagram examined by me no series of unlimited solutions are represented, if we ignore the possible exception of the wollastonite-bustamite series. The type of equilibrium applicable in all other cases would be Roozeboom's No. V.

All the results outlined above refer to the natural minerals and to the relations in natural mineral mixtures. The results obtained in this way need not be similar to those obtained in artificial melts, as the crystallization temperatures in the natural systems will generally be lowered through the presence of other substances. In those cases where the distances of unmixing are small, as in the bustamite-rhodonite and iron rhodonite-sobralite systems, it might be suspected that the equilibrium type belonging to the melting temperature would be that of Roozeboom's No. III and that the discontinuity would not enter until a lower temperature is reached. This is possible, and then we should get equilibrium relations corresponding in principle to those of the alkali feldspars. A statement covering this question and of the lower temperature limit where possibly there exists unlimited mixing power, should be of great interest.

With the exception of the perthite-like inclusions of diopside in the rhodonites from Långban nothing is seen in the specimens studied which with any probability would indicate a splitting up of the components in the solid state. In the case of the intergrowth between bustamite and rhodonite, described from Långban, the minerals form a coarse, not homoaxial intergrowth which may best be called graphical. In the mineral mixtures from V. Silvberg, alluded to in the foregoing, the pyroxenes occur as separate grains. In the specimen from Tunaberg we find the following relations: the greater part of the iron rhodonite and all the sobralite are homoaxially intergrown. In addition we find a minor quantity of pure iron rhodonite. The diopside is for the most part homoaxially intergrown with the composite sobralite-iron rhodonite individuals, but rare grains of free diopside seem also to be present. These relations are best interpretated as due to a crystallization according to a system of limited solubility of the type indicated by the triangular diagram of Fig. 1.

Artificial rhodonite has been prepared by more workers and seems not to be difficult to obtain. According to F. Doerinckel[19]

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[19] *Metallurgie*, **1911**, p. 201, quoted by H. E. Boeke, *Grundlagen d. phys.-chem. Petrographie*, **1915**, p. 193.

it melts incongruently at 1215°. P. Lebedew[20] gives the melting-point at 1210°. F. M. Jaeger and H. S. Van Klooster found it at 1273°.[21] Researches on the melting-points and on some of the crystallization products obtained in the system $MnSiO_3-CaSiO_3$ have been made by A. S. Ginsberg[22] and S. Kallenberg.[23] Both of them found similar relations and according to them the original crystal products would form a continuous series of homogeneous compounds. A feebly marked minimum appears at 10–13 mol. per cent of $CaSiO_3$ at 1150°–1184°. Thus the corresponding type of equilibrium would be No. III. The melting point of rhodonite was found to be 1218° (Ginsberg) and 1180° (Kallenberg). The former figure is in close agreement with that of Doerinckel and Lebedew. The melting-point of $CaSiO_3$, according to the results of the Geophysical Laboratory, is 1540°.

These results seemingly agree with the conception that the bustamites are a Mn-rich series of wollastonite mix crystals. On the other hand, they do not show any traces of the complications introduced through the dimorphism of the $CaSiO_3$ component, nor does there appear to be any manifestation of discontinuity between rhodonites and bustamites. On the other hand, the researches on the crystalline products are not complete.

According to Kallenberg, pure $MnSiO_3$ forms homogeneous mix crystals with $FeSiO_3$ to an extent of about 50 mol. per cent of the latter.[24] Melts richer in iron could not be prepared on account of the reduction or oxidation of the iron. The results gained are in fair agreement with the relations found in the natural minerals, the critical mixtures beginning at 32–40 mol. per cent of Fe (Mg) SiO_3.

The artificially prepared rhodonite has been examined optically be several writers, though no exhaustive description of it has been given. Most reliable are the values of the spec. gravity and they vary between 3.63 and 3.716. The value given by Jaeger and Van Klooster (3.716) agrees fairly well with the figure calculated for the pure natural rhodonite (3.727). Ginsberg gives the refraction as 1.74. This would correspond to β in natural rhodonite (1.741), Kallenberg found $2V_\gamma$ rather small. The angle of natural rhodonite

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[20] *Zeit. f. Anorg. Ch.*, Vol. **70**, p. 313, 1911.

[21] *Proc. Akad., Amsterdam*, Vol. **XVIII**, p. 908, 1916.

[22] *Zeitschr. Anorg. Ch.*, **1908**, Vol. 59, p. 346.

[23] *Zentralbl. Min., Geol. Pal.*, **1914**, p. 388.

[24] *Zeitschr. Anorg. Ch.*, **1914**, p. 362.

is about 57.5°. Evidently the crystal products obtained by the authors named are identical with the natural rhodonite.

In an interesting paper published in 1927 A. N. Winchell discussed the relations of the triclinic pyroxenes.[25] The opinion is put forward that the rhodonites, bustamites, sobralite and pyroxmangite are all independent minerals or mineral groups. This reasoning is based on the valuable work of Wyckoff, Mervin and Washington[26] on the atomic structures of the different pyroxenes. Winchell's opinion is confirmed, on the whole, by the present investigation. The relations given in the triangular diagram of Fig. 1 agree well with the grouping of the pyroxenes made by the three writers mentioned on the basis of the atomic structures; but the provisional assembling of sobralite and pyroxmangite in one and the same group, made by me, is not in accordance with the respective X-ray patterns found. On this point it is to be remarked that if the X-ray examination of the sobralite was made on powder of the Tunaberg specimens, it must refer to a mixture of pyroxenic minerals of which two are triclinic (sobralite and iron rhodonite) and one is hedenbergitic diopside.[27] And the mode of distribution of the pyroxenes, as shown in the above, is of such a character that no one of them can be obtained in the pure state. The paper of the writers mentioned does not indicate whether this fact has been considered. Finally, the optical properties of the pyroxmangite are only partially known.

In the paper by M. H. Hey referred to above the series rhodonite-bustamite is considered as continuous. A similar opinion has also recently been expressed by N. H. Magnusson[28] in his paper on the Långban mines. However, no facts in favor of the opinion expressed are quoted by the last named writer. Hey bases his conclusion of the continuity in the series on three specimens examined by him and on determinations of earlier authors. However, when we consider the small distance of discontinuity in the series, it is apparent that the number of reliable determinations at the disposal of Hey was too small to allow of a true statement of the matter.

(*To be continued*)

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[25] *Am. Mineralogist*, **1927**, p. 10.

[26] *Am. Journ. Sc.*, Ser. 5, vol. **10**, 1925, p. 383.

[27] In addition thereto the specimens contain considerable amounts of other minerals.

[28] *S. G. U., Ser. Ca.*, No. **23**, 1931, p. 44.