The cummingtonite problem.

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Summary. An examination of available chemical analyses shows that the anthophyllite and cummingtonite series of amphibole minerals are not isodimorphous. Whilst the anthophyllites are truly lime-free amphiboles, small amounts of calcium are essential to stabilize the monoclinic lattice of cummingtonites, which should be regarded as lime-poor amphiboles, analogous to the lime-poor pyroxenes. The existence of synthetic clino-anthophyllites is considered.

THE relationship between cummingtonite and anthophyllite has been a source of discussion since 1930, when Johansson made a comparative study of these minerals and tremolite. This work showed the similarity of the structures of the two monoclinic minerals, and the difference between these and the orthorhombic anthophyllite lattice.

An examination of the available chemical analyses (Layton, 1959) shows that although cummingtonite and anthophyllite can both be represented approximately by the formula $(Mg,Fe)_7Si_8O_{22}(OH)_2$, the most commonly occurring anthophyllites are magnesium-rich, whilst cummingtonites are usually iron-rich. For this reason, Winchell (1931) suggested that the magnesian composition is more stable as an orthorhombic structure, whilst the monoclinic form is more stable for iron-rich members. Sundius (1933) denied that the two minerals were isodimorphous and showed that anthophyllites contain 6–9 mol. % more alumina and ferric oxide than otherwise similar cummingtonites. This conclusion is also in general accord with Collins's (1942) results from Strathy. Winchell's further presentation of the case for isodimorphism in 1938 depends mainly on the accuracy of identification of supposedly high-iron anthophyllites. Rabbitt (1948) showed by inference that Winchell's examples were in fact probably cummingtonites and that the existence of high-iron anthophyllites is doubtful.

The rocks of the Orijarvi region were quoted by Eskola (1950) as examples of the occurrence of anthophyllite and cummingtonite of identical composition in the same rock. Eskola cited optical evidence only as the basis of his identification, despite Rabbitt's (1948) demonstration that this can lead to serious errors. Eskola also compared the relationship between cummingtonite and anthophyllite with the dimorphism between enstatite and clino-enstatite.

It seems to us that the following three points have a significant bearing on the problem: aluminium is present in quantity in some anthophyllites but is rare in cummingtonites; although some of the minerals from Strathy described by Collins (1942) as cummingtonites are relatively high in alumina, no X-ray data are given and it is probable that these are in fact anthophyllites. It cannot be maintained that a high aluminium content alone causes the change from monoclinic to orthorhombic symmetry, in view of the existence of high-aluminium monoclinic forms such as tschermakite. Manganese and calcium are usually present in significant quantities in cummingtonite, but are generally virtually absent in anthophyllites.

Many authors have noted the presence of calcium and manganese in cummingtonite, but it does not seem to have been suggested that they should be regarded as essential constituents. The present writers suggest that the monoclinic symmetry of cummingtonite is caused by small amounts of calcium and manganese occupying the positions of eightfold co-ordination, which are more fully occupied by calcium in the calciferous amphiboles such as tremolite. The ionic radius of manganese (0·83 Å. for eight co-ordination, Green, 1959) is larger than those of magnesium and ferrous iron to such an extent that it may conceivably occupy positions similar to calcium (1·03 Å.). A similar calcium–manganese diadochy seems to exist in some other minerals—for example, the garnets. It is clearly impossible to decide the location of the calcium and manganese by examination of the chemical analyses, but support for the above suggestions may be forthcoming from detailed X-ray analysis (it should be emphasized that the only certain distinction between the cummingtonite and anthophyllite groups requires the use of X-rays).

It is interesting to note that Eskola's (1950) cummingtonite from Muuruvesi contains 3·96 % of alkalis. The equivalent anthophyllite contains a negligible quantity. This is a further confirmation that the two minerals do not have identical compositions as claimed by Eskola and are not isodimorphous. The substitution of alkalis in the cummingtonite structure presents no difficulties on the present hypothesis.

The orthorhombic structure allows a closer packing of the silicon–oxygen chains than the monoclinic structure (Whittaker, 1960), and in magnesium-rich environments the small size of magnesium will undoubtedly favour the former symmetry. In addition, magnesium-rich environments are commonly relatively poor in calcium. Thus, a chain-chromite dunite quoted by Dunham *et al.* (1958) contains 30·8 % MgO and only 0·8 % CaO. This has been altered hydrothermally to a chain-chromite–anthophyllite rock containing 28·1 % MgO and 0·2 % CaO.

On average, the anthophyllite series may contain 0·5 % of lime and alkalis. Amounts greater than this in older analyses probably represent impurities. It seems quite possible, however, for very small amounts of calcium and sodium to enter the lattice without destroying the orthorhombic symmetry, and for the resultant strain to be taken up by localized distortion. It is also possible that these atoms may occupy the normally vacant 'A' position (Whittaker, 1960). Because of the small size of aluminium, a similar argument to that for magnesium applies to the aluminium-rich anthophyllites or gedrites.

The increase in ionic radius from magnesium to iron, coupled with the fact that more calcium and manganese are normally present in environments richer in iron, means that under these circumstances the difference in packing density between the orthorhombic and monoclinic structures is less marked and the presence of calcium or manganese ensures stability of the monoclinic form. Some confirmation of the suggested cummingtonite formulation is provided by Eskola (1950) who points out that cummingtonite, not anthophyllite, normally appears in paragenesis with hornblende. On the basis of the theory suggested here, anthophyllite would only form together with hornblende if the rock was deficient in calcium and manganese to such an extent that none was available for the formation of cummingtonite.

In an experimental investigation of the cummingtonite–anthophyllite series, F. R. Boyd (Geophysical Laboratory, 1954–55 Annual Report, p. 117) claimed to have synthesized both amphiboles from mixes free from manganese and lime. The synthesis was found to be easier in the more iron-rich batches and asbestiform anthophyllites were said to form in the range $Mg_{50}Fe_{50}$ to $Mg_{40}Fe_{60}$, although it is also stated that no amphibole richer in iron than $Mg_{50}Fe_{50}$ could be produced. In accordance with the above theory, the instability of iron rich compositions is to be expected in the absence of calcium. It is also important to note that the monoclinic forms were only obtained in the composition range $Mg_{85}Fe_{15}$ to $Mg_{70}Fe_{30}$ at temperatures above 800° C. and close to the apparent breakdown interval. We believe that these clino-amphiboles should be regarded as clino-anthophyllite and not cummingtonite, and that we see here a close parallel to the relationships between enstatite and clinoenstatite. The synthesis of iron-rich clino-amphibole similar in composition to naturally occurring grunerite is only to be expected in the presence of calcium or manganese.

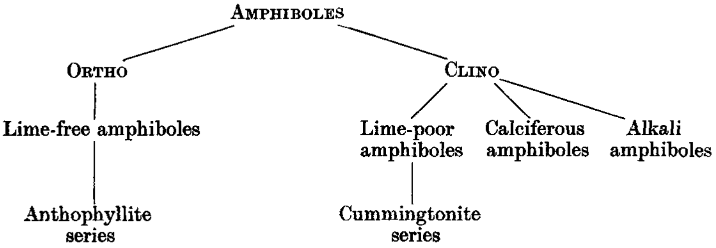
It is also possible to find naturally occurring examples of the persistence of monoclinic symmetry even in amphiboles rich in magnesium, if calcium and manganese are present. This is well shown by the analysis of a tirodite (Bilgrami, 1955).

It is of some interest to compare the cummingtonite–anthophyllite minerals with the pyroxene group. The calcic pyroxenes are monoclinic, as also are the high-temperature forms of the lime-free pyroxenes. There is an obvious analogy between the lime-poor pyroxene pigeonite and cummingtonite. So far as the writers are aware, there is no recorded instance of an ex-solution relationship between tremolite and anthophyllite due to instability of an original cummingtonite with falling temperature. It would seem, however, that this situation, analogous with the formation of diopside lamellae in orthopyroxene, is theoretically possible.

In conclusion it may be stated that whilst the formula

$$(Mg,Fe)_7Si_8O_{22}(OH)_2$$

may adequately represent the composition of the anthophyllite series, the suggestions given above, together with a detailed examination of available analyses (Layton, 1959), show that the formula for the cummingtonite series should be written as $(Ca,Mn)_x(Mg,Fe,Mn)_{7-x}Si_8O_{22}(OH)_2$ where x is less than one. The major divisions of the amphibole series are then seen to be as in the diagram below:



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