# Biopyriboles and polysomatic series 

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#### Abstract

Many crystals may be regarded as made of layer modules that are chemically distinct and that in different crystals may occur in different proportions, giving rise to polysomatic series. The humites constitute a classic example, as do certain phyllosilicates and hydroxides. Amphiboles, in particular, may be factored by (010) cuts into layers that are alternately pyroxene and trioctahedral mica (or talc). The pyroxenes, amphiboles, and trioctahedral micas thus form several such series and we shall designate these minerals, collectively, as the biopyriboles, following Johannsen (1911). Each such series shows interesting relationships connecting the unit cells and space groups of successive members. The existence of these series also leads to useful chemographic relations in composition space.


## Introduction

"Biopyribole" and "pyribole" are collective nouns that were coined by Johannsen (1911) as a solution to certain difficulties that often plague the field geologist. There are good crystallochemical reasons, however, for retaining or reviving these terms, cutting diagonally as they do across currently accepted lines of mineral classification. The close kinship between micas, pyroxenes, and amphiboles is often obscured by their relegation to separate chapters and sometimes even to separate volumes of standard texts and reference works. That amphiboles, like the humites and the chlorites, may be regarded in a sense as "mineralogical mules" derived from the mating of distinct species, in this case mica (or talc) and pyroxene, is a pedagogically useful concept that appears to have escaped general notice.

More specifically, amphiboles may be regarded structurally as consisting of layers, joined on (010), that are essentially trioctahedral mica (we shall here regard talc as a trioctahedral mica) and pyroxene, in an alternating sequence along the $b$ axis (Thompson, 1970). This interlayering has interesting chemical and physical aspects, as we shall see below. Some of the close relationships between these mineral groups have been discussed elsewhere, notably by B. E. Warren and co-workers (1928; 1929; 1930a, b; 1931) and by Bragg (1930, 1937), but not in terms of amphibole as a composite of the other two.

## Modular crystallography

Although it is necessary for many purposes to consider the structure of a crystal on an atom-by-atom basis, it is also often instructive to consider larger units or modules in exploring certain aspects of crystals, particularly if the crystals are complicated ones with many atoms in a unit cell. The coordination polyhedra, for example, that have been used effectively by Pauling $(1928,1929)$ and others are in this sense simple modules that greatly simplify the visualization of many complex structures. Larger modules may include clusters, rings, chains, or layers of linked polyhedra such as those we now use, following Bragg (1937) and others, in the systematic structural classification of the silicates and related minerals.

Unit cells are also in a sense modules that contain all the essential features of a crystal, and that in the crystal as a whole are repeated periodically in three dimensions. We may also go further and regard rows of unit cells as beam modules or layers of unit cells as layer modules that, if isolated, constitute one- and two-dimensional crystals, respectively. Beam modules and layer modules, however, may also be selected so that they do not necessarily contain complete unit cells, but nevertheless contain many of the essential features of the crystal as a whole. For beam or layer modules that do not contain complete unit cells, the specification of the crystal as a whole is complete only when the rules for their assembly have
been given. For layer modules, as in the classical treatments of polytypism (Baumhauer, 1915; but see Verma and Krishna, 1966, for a recent comprehensive summary), these rules of assembly are contained in the stacking formulas. Stacking formulas may also be devised, if needed, to specify the assembly of beams into layers.

Beam modules possess translational periodicity in one dimension and may have their own symmetry elements. Each is thus characterized by one of the symmetry groups appropriate to three-dimensional crystals with one-dimensional lattices. Terminology for such groups is in an unsettled state (see Holser, 1961, Table I), but we shall follow Niggli (1959) and translate his "Balkengruppen" as "beam groups". There is an infinite number of such groups, but for modules that can be assembled into complete threedimensional crystals we need consider only 75. Layer modules with periodicity in two dimensions are characterized by use of the 80 symmetry groups appropriate to three-dimensional crystals with two-dimensional lattices. Again following Niggli (1959) we shall call these "layer groups". We shall use a modified Hermann-Mauguin notation somewhat like that of Holser (1958) for beam and layer groups. The onedimensional lattice of a beam group will be indicated by the symbol $L$, and the two-dimensional lattices of layer groups will be designated as either $P$ (primitive) or $C$ (centered). Taking the direction of the lattice translation as "special" in beam groups, and the direction normal to the lattice translations as "special" in layer groups, we shall place the symbols for
symmetry axes that are in the special direction, or for symmetry planes whose poles are in the special direction, to the left of the lattice symbol, and all others to the right of it. In some layer groups, it will be necessary to place the symbol " 1 " to the left of the lattice symbol in order to distinguish it from a space group.

## Nature of the interlayering

In Figure 1 are cross-sections of three simple beammodules that may be regarded, though idealized, as the basic building blocks of all biopyriboles. In the sketches, the oxygen sites and most metal sites are indicated by means of regular octahedra and tetrahedra that are, of course, idealizations of the coordination polyhedra found in real crystals. The K sites in mica and the similar A sites in amphiboles cannot ordinarily be idealized as simple regular polyhedra, hence are shown by open half-circles (half-circles because they are split by the bounding planes of the modules). The same is true of the $M_{2}$ sites of pyroxenes and the $M_{4}$ sites of amphiboles, hence these are shown by solid half- or quarter-circles. The beam axes in pyriboles are parallel to $c$, and in micas are parallel to the stacking vectors of Smith and Yoder (1956) of the (001) layers in which they occur. The beam groups for these idealized modules are $L 2 / c$ (pyroxene) and $L 2 / m$ (amphibole and mica).

In Figure 2 we see, schematically, the way these beams are arrayed in pyriboles and in micas. In mica polytypes, beams in successive ( 001 ) layers commonly do not have parallel stacking vectors.

The dotted vertical lines in Figure 1b show that an

Table 1. Site correspondence in simple biopyriboles

| Crystals | Sites |  |  |  |  |  |  |  |  |  |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Pyroxene ( $\mathrm{C} 2 / \mathrm{c}$ ) | - | $M_{2}$ | - | $M_{1}$ | - | Si | - | - | * | $\mathrm{O}_{3}$ | $\mathrm{O}_{2}$ | - | $o_{1}$ | - |
| Amphibole ( $\mathrm{C} 2 / \mathrm{m}$ ) | A | $M_{4}$ | $M_{3}$ | $\mathrm{M}_{2}$ | $M_{1}$ | $\mathrm{Si}_{2}$ | $\mathrm{Si}_{1}$ | ${ }^{0} 7$ | $0_{6}$ | $0_{5}$ | $\mathrm{O}_{4}$ | $\mathrm{O}_{3}$ | $\mathrm{O}_{2}$ | $o_{1}$ |
| 1M Mica ( $\mathrm{C} 2 / \mathrm{m}$ ) | K | - | $M_{1}$ | - | $\mathrm{M}_{2}$ | - | Si | $\mathrm{O}_{1}$ | $\mathrm{O}_{2}$ | * | - | (OH) | - | $o_{3}$ |
| Point Group of site | $2 / m$ | 2 | $2 / m$ | 2 | 2 | 1 | 1 | m | 1 | 1 | 1 | m | 1 | 1 |

Site-labeling for the pyroxene is after Burnham et at (7967); that for the amphibole is after Ghose and Hellner (1959), and that for the mica is after Donnay et al (1964). In an idealized model such as that of Fig. 1 or 3 , the amphibole $0_{5}$ and $0_{6}$ sites lie in the ( 010 ) auts, hence their assignment is arbitrary. The above assigntents ${ }_{0}{ }_{0} f_{5} 0_{5}$ and $0_{6}$ are consistent with their locations in most real C2/m amphiboles. With axes corpesponding to those of the pyroxene, the amphibole would be $I 2 / \mathrm{m}$ and the miea $\mathrm{A} / \mathrm{m}$.


Fig. 1. Idealized beam-modules for biopyriboles as seen in crosssection. A sites of amphibole and K sites of micas are shown by open half circles. $M_{4}$ sites of amphiboles and $M_{2}$ sites of pyroxenes are shown by solid half circles or quarter circles. All other $M$ sites are idealized as regular octahedra of equal size, and all Si sites are idealized as regular tetrahedra of equal size. The ratio of the tetrahedral edge to the octahedral edge is $\sqrt{3} / 2$.
amphibole beam may be cut in such a way that a central portion that is essentially a mica beam may be removed. Upon removal of this central mica portion, the two other portions may be reassembled to form a pyroxene beam as in Figure 1c. In a simple amphibole such as tremolite, the modules of Figure 2b are all oriented the same way, but in other amphibole structures some beams are inverted by half-rotations about their long axes. In any case, the cuts that factor the individual beams into pyroxene and mica parts are aligned in (010) so that an entire model may in turn be factored by $(010)$ cuts into layer modules that are alternately mica and pyroxene along $b$. For our idealized models, the ( 010 ) cuts would be placed at $1 / 8$ and $3 / 8$ on the amphibole $b$ axis. This interlayering is shown in more detail in Figure 3.

## Idealized clinoamphiboles

In tremolite, an example of a simple clinoamphibole, the repeating unit of a single beam module has the formula $\mathrm{Ca}_{2} \mathrm{Mg}_{5} \mathrm{Si}_{8} \mathrm{O}_{22}(\mathrm{OH})_{2}$ and all modules are in the same orientation. The mica portion of the tremolite beam module has the formula $\mathrm{Mg}_{3} \mathrm{Si}_{4} \mathrm{O}_{10}(\mathrm{OH})_{2}$ (talc), and the remainder may be used to construct a pyroxene beam module with the formula $\mathrm{Ca}_{2} \mathrm{Mg}_{2} \mathrm{Si}_{4} \mathrm{O}_{12}$ (diopside). These formulas are also those of the corresponding ( 010 ) layer modules. In this example, the pyroxene layer modules belong to the layer group $2 / n P$ and the mica modules to the layer group $2 / m P$.

If the mica layers are removed from a tremolite model, each pyroxene layer may be translated $b / 4$ plus $c / 2$ relative to each of its nearest neighbors to form an idealized diopside model. Referred to con-


Fig. 2. Arrangement of beam-modules in micas (a), amphiboles (b), and pyroxenes (c). Only one mica layer is shown. The amphibole and pyroxene arrays may also be regarded as simple cleavage fragments. Dashed lines show cleavage orientations.
ventional amphibole axes, this model would have space group $I 2 / c$, but referred to conventional pyroxene axes, the space group would be $C 2 / c$ (Fig. 4). Similarly, the mica layers may be assembled by translations of $a / 2$ plus $b / 4$ plus $c / 2$ to form an idealized model of a $1 M$ talc. The space group of the mica (talc) model would be $A 2 / m$ if referred to conventional pyroxene or amphibole axes, and would be $\mathrm{C} 2 / \mathrm{m}$ if referred to conventional mica axes. The registry of the cleavage layers relative to their neighbors in such a mica model is not quite that found in real micas, but may be adjusted by small translations of these layers relative to their successors along the mica $a$ axis. The conventional $b$ axes of the three models correspond in orientation, but the $c$ axes of the pyriboles correspond to the $a$ axes of the $1 M$ mica (Fig. 4 ), and the $I$ cell of the pyroxene corresponds to the $C$ cell of the amphibole and vice versa (see Jaffe et al., 1968).

Although the width of each module is only one-


Fig. 3. Idealized section $\perp c$ of a $C 2 / m$ ( $I 2 / m$, if referred to pyroxene axes) amphibole such as tremolite. Dashed vertical lines show (010) cuts that factor it into mica and pyroxene layer modules.


Fig. 4. Section $\perp b$ of a biopyribole unit cell, as in a $C 2 / m \mathrm{mica}$, a $C 2 / m$ amphibole, or a $C 2 / c$ pyroxene. If the amphibole were referred to a body-centered $I 2 / \mathrm{m}$ cell, its axes would correspond to those of a $C 2 / c$ pyroxene. Mica and pyroxene axes correspond but with $a$ and $c$ reversed. If mica axes were labeled as in pyroxene, it would be $A 2 / m$. The $2_{1}$ axes parallel to $b$ are located at points marked M (in mica), A (in amphibole), and P (in pyroxene), 2-axes $\| b$ are shown by the standard symbol, and are common to all three.
half of the conventional $b$ axis of the complete structure, it is possible, in this example at least, to choose unconventional primitive cells for both the mica and the pyroxene such that each $(010)$ layer module may be regarded as a mosaic of primitive unit cells. Although we have been concerned here primarily with larger units than the individual atoms, the factoring of an amphibole into pyroxene-like and mica-like (010) layers can be carried even to the minute level of the atomic sites. For the example just considered, these sites correspond essentially as shown in Table 1.

There are many interesting relationships among the symmetry operations (rotations, reflections, translations) of a given mica model and a given pyroxene model, and those of the amphibole model that may be assembled from layer modules taken from the other two. To minimize confusion, we shall refer all three models to the conventional pyroxene axes. The amphibole in this example will thus be referred to $I 2 / m$ and the mica to $A 2 / m$. Letting subscripts distinguish between amphibole (A), pyroxene ( P ) and mica (M), we then have, with regular polyhedra,

$$
\begin{equation*}
b_{\mathrm{A}}=b_{\mathrm{P}}+b_{\mathrm{M}} \tag{1}
\end{equation*}
$$

and also

$$
\begin{equation*}
b_{\mathrm{M}}=b_{\mathrm{P}}=b_{\mathrm{A}} / 2 \tag{2}
\end{equation*}
$$

Furthermore, if the modules are to be joined in (010)

$$
\begin{align*}
& a_{\mathrm{M}}=a_{\mathrm{P}}=a_{\mathrm{A}}  \tag{3}\\
& c_{\mathrm{M}}=c_{\mathrm{P}}=c_{\mathrm{A}} \tag{4}
\end{align*}
$$

The centering translations may at first appear unrelated, but if we take the pyroxene-centering translation $\left(a_{\mathrm{P}}+b_{\mathrm{P}}\right) / 2$ and the mica-centering translation $\left(b_{\mathrm{M}}+c_{\mathrm{M}}\right) / 2$ in succession, the net effect with (1), (3), and (4) is $\left(a_{\mathrm{P}}+b_{\mathrm{P}}+b_{\mathrm{M}}+c_{\mathrm{M}}\right) / 2$, which is equivalent to the amphibole-centering translation $\left(a_{\mathrm{A}}+b_{\mathrm{A}}+\right.$ $\left.c_{\mathrm{A}}\right) / 2$. It is thus clear that the $I$-centered cell of the tremolite is a necessary consequence of the union, as indicated, of the $A$-centered cell of the talc and the $C$ centered cell of the diopside. These considerations also cast in a new light the translations for assembly of mica from amphibole after removal of the pyroxene modules as discussed earlier. These translations, from (1), (3), and (4) above, are simply the centering translation of the removed pyroxene.

In this example, all of the symmetry elements of the (010) pyroxene and mica modules are preserved in the composite amphibole structure. When the mica modules are assembled as mica or the pyroxene modules as pyroxene, however, new symmetry elements arise in the planes where the modules are joined, as a consequence of their mode of assembly. This can not, of course, occur when they assemble as amphibole, because the modules meeting at each (010) interface are then both chemically and physically distinct.

The two-fold axes parallel to $b$ are common to both types of module and to all three crystals that can be made from them. The two-fold screw axes parallel to $b$, however, have different locations (Fig. 4) in each of the three crystals that can be assembled from the layer modules. This may appear anomalous but is not. Because these screw axes invoke translation along $b$, they can not be symmetry elements of the isolated (010) modules but are rather related to their mode of assembly. With this in mind, we can show that their locations do indeed make sense. The $2_{1}$ operations for a $1 M$ talc and for diopside, taken successively, are equivalent in net effect to the amphibole $2_{1}$ operation plus a 2 -rotation. We thus see that here again the amphibole structure is a necessary product of the mating, as indicated, of the corresponding pyroxene and mica. An alternative operation, in fact, by which the pyroxene modules in an amphibole may be reassembled after removal of the mica ones is to subject each, relative to its neighbor, to the $2_{1}$ operation of the removed mica and vice versa. A useful way in which to remember the various possible assembly rules is that they undo the effects arising from the introduction of the alien modules.

Had we selected a clinoamphibole such as edenite in which the $A$ sites are occupied by Na or K , rather than vacant as in tremolite, the mica modules would assemble to form a 1 M Na - or K-phlogopite rather than talc. Thus, in a general way, hornblende may be regarded as interlayered biotite and augite.

## Idealized orthoamphiboles

In an idealized protoamphibole (Pnmn) structure (Gibbs, 1969) the beams are not all in the same orientation but are inverted in alternate (010) stacks along $b$. With the idealized formula $\mathrm{Mg}_{7}\left(\mathrm{Si}_{8} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}$ (neglecting lithium) the pyroxene layers would have the composition $\mathrm{Mg}_{4} \mathrm{Si}_{4} \mathrm{O}_{12}$ and the mica layers would again have the composition of talc. With removal of the mica layers, the pyroxene layers, $2 P 2_{1} 2_{1}$, may be assembled to form a protoenstatite (Pbcn, see Smith, 1959) and the mica layers would again assemble to form a $1 M$ talc. The assembly rules and the symmetry relations among the three corresponding structures are different here from those of our earlier example, but may be deduced by similar reasoning.
In an idealized anthophyllite (Pnma) half of the beam modules are again inverted, as in a protoamphibole, but according to a different scheme in which alternate beams within any (010) stack are inverted relative to their neighbors along $a$. These stacks are then arranged along $b$ so that the third is a duplicate of the first, and so on. The pyroxene layers ( $I P 2 / a$ ) have the composition $\mathrm{Mg}_{4} \mathrm{Si}_{4} \mathrm{O}_{12}$ and may be assembled to form enstatite ( $P b c a$ ). The mica layers $\left(m P a 2_{1}\right)$ again have the composition of talc but assemble to form a 20 talc ( $A 2_{1} m a$, with amphibole axes, or $\mathrm{Ccm} 2_{1}$ with mica axes) rather than a $1 M$ talc. With small translations of the mica beams parallel to $a$ (mica) in the cleavage planes, however, the symmetry of the reassembled mica model may be increased to Ccmm (mica axes) which is to be expected for a 20 mica.

## Non-idealized amphiboles

In real amphiboles or in more sophisticated models the structural details are different from those in our idealized examples, and the symmetry of a beam or layer module may be lowered by distortion of the idealized structure or by site-ordering, or by both. The basic topology of the amphibole structure, however, is still there, and this has been the main key to the general argument that amphibole may be regarded as interlayered mica and pyroxene.

## Polysomes and polysomatic series

Crystals that may be regarded as made of interlayered chemically distinct modules taken from other crystals are not uncommon among minerals. The humites constitute a series of such crystalline phases, in that they may be regarded as containing two types of module that can occur in several different ratios. There are also numerous examples involving phyllosilicates and hydroxides. Several of these examples will be considered in more detail below because of some interesting structural and chemographic relationships they share with the biopyribole series.

The phenomenon is akin to layer polytypism in many ways and has been called "mixed-layer polytypism" by Kohn and Eckart (1965) and others. Warshaw and Roy (1961) suggest "heteropolytypism" as opposed to ordinary or "homopolytypism." Polytypism, however, is properly a special case of polymorphism, and involves chemically identical modules rather than chemically distinct ones as in the phenomena considered here. The term "morphotropic" has also been applied, specifically to the humites (Penfield and Howe, 1894), but this again has been used in other senses that are not strictly comparable. The term "homologous series" has been used in chemistry for series of molecules, such as the alcohols or paraffins, that contain variable amounts of some common structural element. The term was first applied to crystals showing phenomena like that described here by Magnéli (1953), and has since been used by other authors (see, for example, Roth and Wadsley, 1965) in a more generalized sense, employing beam modules and even discrete blocks. In any case, despite the resemblance in certain chemical formulas, the physical phenomenon in organic chemistry is quite different from that in crystals, hence it seems unwise to use the same term for both. We shall therefore use the term polysome or "many substance" for a crystal such as that of an amphibole that can be regarded as made of chemically distinct layer modules. Similarly, a series such as the humites (including olivine) or any of several among the biopyriboles can be called a polysomatic series, and the phenomenon polysomatism. Polytypism may thus occur in a onecomponent system in the physicochemical sense, whereas a polysomatic series requires at least two components for its complete description.

Writing mica schematically as (M), pyroxene as (P), and amphibole as (MP), we may ask whether other members of the various biopyribole series may occur. Models of them can be constructed easily, whether or not they occur as real crystals. (MMP) or
$\left(\mathrm{M}_{2} \mathrm{P}\right)$, for example, would have a formula of the type $\mathrm{R}_{5} \mathrm{Si}_{6} \mathrm{O}_{16}(\mathrm{OH})_{2}$ and would have cleavage angles intersecting more obliquely than those of amphibole. [Since the first version of this manuscript was prepared, natural occurrences of not only (MMP) but also (MPMMP) have been reported by Veblen and Burnham (1975, 1976).]

## Chemical aspects

A consequence of the polysomatic relationships among the biopyriboles is that nearly all amphibole end-member formulas are linear combinations of a known, or at least plausible, pyroxene end-member formula and a mica end-member formula, and that for every pair of end-member formulas of pyroxene and trioctahedral mica there is a known or at least plausible amphibole formula. Some of the formulas so obtained are not known as single-phase compositions, but in most such instances can be identified at least as end-member components that reflect known compositional variations. Some simple examples are given in Table 2. Parentheses in Table 2 indicate components of real crystals that are not known in pure form. In a general way we may write

$$
\text { Biotite }+ \text { Augite }=\text { Hornblende }
$$

The exceptions to these pedagogically useful results are in the amphibole formulas that have occupied $A$ sites without an accompanying substitution of Al for Si in the adjacent tetrahedral sites. Thus for richterite (soda-tremolite) we write

Mica Module + Pyroxene Module $=$ Richterite

$$
\begin{aligned}
{\left[\mathrm{NaMg}_{3} \mathrm{Si}_{4} \mathrm{O}_{10}(\mathrm{OH})_{2}\right]^{+1}+} & {\left[\mathrm{NaCaMg}{ }_{2} \mathrm{Si}_{4} \mathrm{O}_{12}\right]^{-1} } \\
& =\mathrm{Na}_{2} \mathrm{CaMg}_{5} \mathrm{Si}_{8} \mathrm{O}_{22}(\mathrm{OH})_{2}
\end{aligned}
$$

and find that we cannot write neutral formulas for the modules. The same is true for arfvedsonite, $\mathrm{Na}_{3} \mathrm{Fe}_{4} \mathrm{AlSi}_{8} \mathrm{O}_{22}(\mathrm{OH})_{2}$. Note that in such an amphibole there is a marked departure from the rule of local valence saturation, in that the $A$ site must be surrounded entirely by oxygens that are already bonded to two silicons. One may speculate that some of these amphiboles may, in certain occurrences, arise through an ion exchange of the type $\mathrm{Na}_{2}$ for $\square \mathrm{Ca}$, at some time after primary crystallization.

A further consequence of the collinearity of polysomatic series in composition space is that it often simplifies the chemographic analysis of complex natural mineral assemblages, and that it may help locate simple degenerate subsystems in otherwise complex ones that one may wish to investigate experimentally. Specifically, two distinct mica formulas and a pyroxene formula (or vice versa) will always define a plane in composition space that is likely to also include a series of amphiboles. In many such instances the equilibrium among the biopyriboles can be strictly ternary even in systems where many chemical elements are involved. Some interesting and largely selfexplanatory examples are shown in Figure 5. Pyroxene formulas must conform to the general type $\mathrm{R}_{2} \mathrm{O}_{3}$ and may be combined with formulas such as $\mathrm{H}_{2} \mathrm{SiO}_{3}$,

Table 2. Polysomatic series in the biopyriboles

|  | $\begin{array}{cl} \text { Talc } \\ \mathrm{Mg}_{3} \mathrm{Si}_{4}{ }^{\mathrm{O}}{ }_{10}(\mathrm{OH})_{2} \end{array}+$ | $\begin{aligned} & \text { Diopside } \\ & \mathrm{Ca}_{2} \mathrm{Mg}_{2} \mathrm{Si}_{4} \mathrm{O}_{12} \end{aligned}$ | - | $\begin{aligned} & \text { Tremolite } \\ & \mathrm{Ca}_{2} \mathrm{Mg}_{5} \mathrm{Si}_{8} \mathrm{O}_{22}(\mathrm{OH})_{2} \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: |
|  | Tale + | Enstatite | = | Anthophyllite |
|  | $\mathrm{Mg}_{3} \mathrm{Si}_{4} \mathrm{O}_{10}{ }^{(\mathrm{OH})_{2}}$ | $\mathrm{Mg}_{4} \mathrm{Si}_{4} \mathrm{O}_{12}$ |  | $\mathrm{Mg}_{7} \mathrm{Si}_{8} \mathrm{O}_{22}{ }^{(0 \mathrm{H})}{ }_{2}$ |
|  | Talc + | Jadeite | - | Glaucophane |
|  | $\mathrm{Mg}_{3} \mathrm{Si}_{4} \mathrm{O}_{10}{ }^{(\mathrm{OH})}{ }_{2}$ | $\mathrm{Na}_{2} \mathrm{Al}_{2} \mathrm{Si}_{4}{ }^{\mathrm{O}} 12$ |  | $\mathrm{Na}_{2} \mathrm{Mg}_{3} \mathrm{Al}_{2} \mathrm{Si}_{8} \mathrm{O}_{22}{ }^{\text {(OH) }} 2$ |
|  | $(\mathrm{Fe}-\mathrm{Talc}) *+$ | Acmite | $=$ | Riebeckite |
|  | $\mathrm{Fe}_{3} \mathrm{Si}_{4}{ }^{\mathrm{O}} 10{ }^{(\mathrm{OH})_{2}}$ | $\mathrm{Na}_{2} \mathrm{Fe}_{2} \mathrm{Si}_{4}{ }^{\mathrm{O}}{ }_{12}$ |  | $\mathrm{Na}_{2} \mathrm{Fe}_{5} \mathrm{Si}_{8} \mathrm{O}_{22}(\mathrm{OH})_{2}$ |
|  | Phlogopite + | Diopside | $=$ | (K-Edenite) * |
|  | $\mathrm{KMg}_{3} \mathrm{AlSi}_{3} \mathrm{O}_{10}(\mathrm{OH})_{2}$ | $\mathrm{Ca}_{2} \mathrm{Mg}_{2} \mathrm{Si}_{4} \mathrm{O}_{12}$ |  | $\mathrm{KCa}_{2} \mathrm{Mg}_{5} \mathrm{AlSi}_{7} \mathrm{O}_{22}(\mathrm{OH})_{2}$ |
|  | (Na-Phlogopite)* + | Diopside | $=$ | Edenite |
|  | $\mathrm{NaMg}_{3} \mathrm{AlSI}_{3}{ }_{10}{ }^{(\mathrm{OH})_{2}}$ | $\mathrm{Ca}_{2} \mathrm{Mg}_{2} \mathrm{Si}_{4} \mathrm{O}_{12}$ |  | $\mathrm{NaCa}_{2} \mathrm{Mg}_{5} \mathrm{AlSi}_{7} \mathrm{O}_{22}{ }^{(\mathrm{OH})}{ }_{2}$ |
|  | Talc + | "CATS" | - | Tschermakite |
|  | $\mathrm{Mg}_{3} \mathrm{SI}_{4} \mathrm{O}_{10}{ }^{(\mathrm{OH})}{ }_{2}$ | $\mathrm{Ca}_{2} \mathrm{Al}_{4} \mathrm{Si}_{2} \mathrm{O}_{12}$ |  | $\mathrm{Ca}_{2} \mathrm{Mg}_{3} \mathrm{Al}_{4} \mathrm{SH}_{6} \mathrm{O}_{22}(\mathrm{OH})_{2}$ |
|  | *Parentheses indicate end members not known to exist stably as phases. |  |  |  |



Fig. 5. (a) through (f) are composition planes defined by $\mathrm{CaMgSi}_{2} \mathrm{O}_{6}-(\mathrm{Di})$ and the mica end-members $\mathrm{Mg}_{3}\left(\mathrm{Si}_{4} \mathrm{O}_{10}\right)(\mathrm{OH})_{2}-(\mathrm{Tc})$, $\mathrm{KMg}_{3}\left(\mathrm{AlSi}_{3} \mathrm{O}_{10}\right)(\mathrm{OH})_{2}-(\mathrm{Ph}), \mathrm{NaMg}_{3}\left(\mathrm{AlSi}_{3} \mathrm{O}_{10}\right)(\mathrm{OH})_{2}-(\mathrm{Na}-\mathrm{Ph})$, and $\mathrm{CaMg}_{3}\left(\mathrm{Al}_{2} \mathrm{Si}_{2} \mathrm{O}_{10}\right)(\mathrm{OH})_{2}-(\mathrm{Cl})$. These include the stable amphibole end-members $\mathrm{Ca}_{2} \mathrm{Mg}_{5}\left(\mathrm{Si}_{8} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-(\mathrm{Tr}), \quad \mathrm{NaCa}_{2} \mathrm{Mg}_{5}\left(\mathrm{AlSi}_{7} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-\left(\mathrm{Ed}_{5}\right), \mathrm{Na}_{2} \mathrm{CaMg}_{5}\left(\mathrm{Si}_{8} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-(\mathrm{Ri})$, and $\mathrm{KNaCaMg} \mathrm{Na}_{5}\left(\mathrm{Si}_{8} \mathrm{O}_{22}\right)$ $(\mathrm{OH})_{2}-(\mathrm{K}-\mathrm{Ri})$, and the metastable ones $\mathrm{KCa}_{2} \mathrm{Mg}_{6}\left(\mathrm{AlSi}_{7} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-(\mathrm{K}-\mathrm{Ed})$ and $\mathrm{Ca}_{3} \mathrm{Mg}_{5}\left(\mathrm{Al}_{2} \mathrm{Si}_{6} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-(\mathrm{Ca}-\mathrm{Ed})$. Lightly stippled areas are inaccessible to biopyriboles. (g) shows phases coexisting with diopside in the system that may be defined by $\mathrm{Di}, \mathrm{Ph}, \mathrm{Na}-\mathrm{Ph}$, and Cl . Darker stippling shows fields of amphiboles and micas that coexist with diopside. Micas appear when the amphibole is metastable relative to the mica-pyroxene pair.
$\mathrm{H}_{2} \mathrm{KAlO}_{3}$, or $\mathrm{H}_{2} \mathrm{NaAlO}_{3}$ to obtain coordinates containing an amphibole formula, a mica formula, or both. A four-component space obtained in this way is shown in Figure 6. These diagrams suggest several possibilities for mica or amphibole compositions not now known but that might yet be found.

## The humite series

The structural relationships between olivine (Bragg and Brown, 1926) and the humite minerals were first clarified by Taylor and West (1928). Taylor and West described them as a polysomatic sequence in which layers of olivine, $\mathrm{Mg}_{2} \mathrm{SiO}_{4}$, alternate with layers of the composition $\mathrm{Mg}[\mathrm{F},(\mathrm{OH})]_{2}$. The silica-free layers, however, cannot be assembled to form known $\mathrm{MgF}_{2}$ or $\mathrm{Mg}(\mathrm{OH})_{2}$ structures and, as pointed out by Ribbe et al. (1968), do not actually have the anionic content ascribed to them by Taylor and West. We shall here avoid these difficulties by taking norbergite as the limit of the series (it appears to be) and describing the others as interlayering of olivine and norbergite modules.

Both olivine and norbergite are orthorhombic with
space group Pbnm. We shall therefore cut each into layer modules (see Ribbe et al., 1968, Fig. 1; also Gibbs and Ribbe, 1969, Fig. 1; or Papike and Cameron, 1976, Fig. $4 a$ and $e$ ) by cuts at the mirror planes in ( 001 ), obtaining modules that are in alternate "left", $\mathrm{O}_{\mathrm{L}}, \mathrm{N}_{\mathrm{L}}$, or "right", $\mathrm{O}_{\mathrm{R}}, \mathrm{N}_{\mathrm{R}}$, orientation for each. Our cuts in norbergite pass between mirrorrelated pairs of fluorine atoms (or hydroxyl groups) rather than through them, as do the cuts of Taylor and West (1968). Olivine is then $\mathrm{O}_{L} \mathrm{O}_{\mathrm{R}} \mathrm{O}_{\mathrm{L}} \mathrm{O}_{\mathrm{R}} \ldots$. . or simply ( $O$ ), and norbergite is then $\mathrm{N}_{\mathrm{L}} \mathrm{N}_{\mathrm{R}} \mathrm{N}_{\mathrm{L}} \mathrm{N}_{\mathrm{R}} \ldots \ldots$ or simply ( N ). Our modules correspond to those of Taylor and West according to the scheme
$\ldots \frac{|\mathrm{A}| \mathrm{B}|\mathrm{A}|}{\left|\mathrm{O}_{\mathrm{L}}\right| \mathrm{O}_{\mathrm{R}} \mid}-$


Chondrodite is then in our notation $\mathrm{O}_{\mathrm{L}} \mathrm{N}_{\mathrm{R}} \mathrm{O}_{\mathrm{L}}$ $\mathrm{N}_{\mathrm{R}} \ldots$. . . or simply ( ON ); humite is $\mathrm{O}_{\mathrm{L}} \mathrm{O}_{\mathrm{R}} \mathrm{N}_{\mathrm{L}} \mathrm{O}_{\mathrm{R}} \mathrm{O}_{\mathrm{L}} \mathrm{N}_{\mathrm{R}}$ $\ldots$... or simply ( $\mathrm{O}_{2} \mathrm{~N}$ ); and clinohumite is $\mathrm{O}_{\mathrm{L}} \mathrm{O}_{\mathrm{R}} \mathrm{O}_{\mathrm{L}}$ $\mathrm{N}_{\mathrm{R}} \mathrm{O}_{\mathrm{L}} \mathrm{O}_{\mathrm{R}} \mathrm{O}_{\mathrm{L}} \mathrm{N}_{\mathrm{R}} \ldots \ldots$, or simply $\left(\mathrm{O}_{3} \mathrm{~N}\right)$ (see Papike


Fig. 6. The composition space $\mathrm{CaSiO}_{3}-\mathrm{MgSiO}_{3}-\mathrm{FeSiO}_{3}-\mathrm{H}_{2} \mathrm{SiO}_{3}$ showing the pyroxene quadrilateral Diop., Hed ,, $\mathrm{MgSiO}_{3}$, $\mathrm{FeSiO}_{3}$; the amphibole quadrilateral Trem., Fe-Trem., Anth., Grun.; and Talc, Fe-Talc.
and Cameron, 1970, Fig. 4b, c, and d). If the number of symbols (counting subscripts as multipliers) in the simplified formula is odd the structure is orthorhombic, $P b n m$, and if even it is monoclinic, $P 2_{1} / b$, if referred to axes labelled as they are in olivine, or $P 2_{1} / c$, if the axes of the monoclinic forms are conventionally labeled.


Fig. 7. Brucite-like ( $\mathrm{B}_{\mathrm{U}}, \mathrm{B}_{\mathrm{D}}$ ) and talc-like $\left(\mathrm{T}_{\mathrm{U}}, \mathrm{T}_{\mathrm{D}}\right)$ layer modules || (001) in serpentines or septechlorites (a) and in chlorites (b).

## Some layer silicates and hydroxides

Serpentines, septechlorites (Nelson and Roy, 1954), and chlorites (Pauling, 1930b) can be described as polysomes made of brucite or brucite-like modules and talc or talc-like modules; and the kaolin or kandite minerals (Brown, 1955) can be similarly described as made of gibbsite and pyrophyllite modules. We may select modules for lizardite or for septechlorite as shown in Figure 7a, and for chlorite as shown in Figure 7b. Formulas for lizardite or septechlorite may thus be written $\mathrm{T}_{\mathrm{U}} \mathrm{B}_{\mathrm{D}} \mathrm{T}_{\mathrm{U}} \mathrm{B}_{\mathrm{D}} \ldots \ldots$, or simply (TB); and a formula for chlorite may be written $T_{U} T_{D} B_{U} B_{D} T_{U T} T_{D} B_{U} B_{D} \ldots$, or simply ( $T_{2} B_{2}$ ). In lizardite we then have

$$
\begin{array}{ccc}
\text { Brucite }+ \text { Talc } & = & \text { Lizardite } \\
\mathrm{Mg}_{3}(\mathrm{OH})_{6}+ & \mathrm{Mg}_{3} \mathrm{Si}_{4} \mathrm{O}_{10}(\mathrm{OH})_{2} & = \\
\mathrm{Mg}_{6} \mathrm{Si}_{4} \mathrm{O}_{10}(\mathrm{OH})_{8}
\end{array}
$$ for kaolinite we have

Gibbsite + Pyrophyllite $=$ Kaolinite
$\mathrm{Al}_{2}(\mathrm{OH})_{6}+\mathrm{Al}_{2} \mathrm{Si}_{4} \mathrm{O}_{10}(\mathrm{OH})_{2}=\mathrm{Al}_{4} \mathrm{Si}_{4} \mathrm{O}_{10}(\mathrm{OH})_{8}$ and for chlorite

$$
\text { "Brucite" }+ \text { "Talc" }=\text { Chlorite }
$$

$$
\mathrm{Mg}_{2} \mathrm{Al}(\mathrm{OH})_{6}{ }^{+1}+\mathrm{Mg}_{3} \mathrm{AlSi}_{3} \mathrm{O}_{10}(\mathrm{OH})_{2}{ }^{-1}=
$$

$$
\mathrm{Mg}_{5} \mathrm{Al}_{2} \mathrm{Si}_{3} \mathrm{O}_{10}(\mathrm{OH})_{2}
$$



Fig. 8. A portion of the composition plane $\mathrm{MgO}-\mathrm{H}_{2} \mathrm{O}-\mathrm{SiO}_{2}$. Solid circles are hydroxy end-members of known minerals. Open circles are other compositions of interest. Polysomatic series include the humites: forsterite-clinohumite-humite-chondrodite-norbergite; the phyllosilicate-hydroxide series: talc-lizardite-brucite; and the biopyriboles: talc-anthophyllite-enstatite. (MMP) and (MPMMP) are new biopyriboles recently described by Veblen and Burnham (1975, 1976). Joins are merely to indicate collinearities, not coexistence.
which is reminiscent of the amphiboles richterite and arfvedsonite, in that the valence balance is module-to-module rather than internal to each.

## Pyroxenoids

Koto et al. (1976) have shown that the pyroxenoids may also be regarded as a polysomatic series containing pyroxene-layer modules (see also Prewitt and Peacor, 1964; and Burnham, 1971). The pyroxene slabs are here of clinopyroxene type and are cut parallel to (011) pyroxene, rather than (010) as in the biopyriboles. These alternate with wollastonite layers (W) and the following forms are known: (W), wollastonite; (WP), rhodonite; ( $\mathrm{WP}_{2}$ ), pyroxmangite-pyroxferroite; $\left(\mathrm{WP}_{3}\right)$, "ferrosilite III"; and (P), pyroxene. Their polytype formulas thus parallel those of the humites, with pyroxene playing here the role of olivine in the humites.

Some synthetic silicates in the system $\mathrm{MgSiO}_{3}{ }^{-}$ $\mathrm{LiScSi}_{2} \mathrm{O}_{6}$ have been shown by Takéuchi et al. (1977) to belong to yet another polysomatic series containing pyroxene-layer modules. In these the clinopyroxene slabs are cut parallel to (101) pyroxene. It is of interest that both pyroxenes and micas may be cut in more than one way to provide bases for polysomatic series.

## Some chemographic relationships

By selecting our reference compositions as $\mathrm{SiO}_{2}$, MgO , and $\mathrm{Mg}(\mathrm{OH})_{2}$ and plotting on the basis of anion fraction, we obtain the intriguing array of Figure 8 . In it we see not only the hydroxy end-members of the humites and a series of simple biopyriboles, but also the series talc-lizardite-brucite. Forsterite, hu-


Fig. 9. The composition plane $\mathrm{SiO}_{2}-\mathrm{NaAlO}_{2}-\mathrm{HAlO}_{2}$ showing the possible dioctahedral amphiboles: $\mathrm{Na}_{2} \mathrm{Al}_{4}\left(\mathrm{Si}_{8} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-(X)$ and $\mathrm{Na}_{3} \mathrm{Al}_{4}\left(\mathrm{AlSi}_{7} \mathrm{O}_{22}\right)(\mathrm{OH})_{2}-(Y)$, relative to quartz, albite, jadeite, nepheline, paragonite, and pyrophyllite.
mite, and hydroxy-norbergite are essentially desilicated enstatite, anthophyllite, and talc, respectively, and several other collinearites such as periclase-nor-bergite-lizardite are shown. This is but one of many interesting chemographic relationships among polysomatic series. By adding components such as $\mathrm{KAlO}_{2}, \mathrm{NaAlO}_{2}$, or $\mathrm{CaAl}_{2} \mathrm{O}_{4}$ to those of Figure 8, we may obtain composition spaces that show many biopyribole compositions relative to the compositions of other rock-forming minerals.

## The possibility of dioctahedral amphiboles

Although none are yet known, there seems to be nothing in principle that would prevent the existence of amphiboles with mica modules of dioctahedral rather than trioctahedral type. Dioctahedral modules might contain mainly $\mathrm{Al}, \mathrm{Fe}^{3+}$, or $\mathrm{Cr}^{3+}$ in the octahedral sites. If Al-rich, the $c$ axis (in pyroxene notation) would probably be relatively short and could be most readily combined with pyroxene modules, such as those of $\mathrm{Na}-\mathrm{Al}$ or $\mathrm{Li}-\mathrm{Al}$ pyroxeneś, that have comparable dimensions.

In Figure 9 we see that two such end-member compositions might occur in the composition plane $\mathrm{SiO}_{2}-\mathrm{NaAlO}_{2}-\mathrm{HAlO}_{2}$. Point $X$ corresponds to the formula $\mathrm{Na}_{2} \mathrm{Al}_{4} \mathrm{Si}_{8} \mathrm{O}_{22}(\mathrm{OH})_{2}$, and point $Y$ to the formula $\mathrm{Na}_{3} \mathrm{Al}_{5} \mathrm{Si}_{7} \mathrm{O}_{22}(\mathrm{OH})_{2}$, the first with its $A$ site empty, and the second with an occupied $A$ site. No minerals of either composition are known, but analyses of "gastaldite" (Strüver, 1876, 1887; Milch, 1907), now regarded as a variety of glaucophane rich in alumina, may be of this type (see also Bocquet, 1971, for more recent references). On the other hand, these may be simply faulty analyses of normal glaucophanes.

The first of the two formulas lies in the triangle quartz-albite-paragonite or in quartz-jadeite-paragonite, and the second on the join jadeite-paragonite. If such amphiboles do exist, their fields of stability will be limited by these alternative assemblages. If they occur at all, they probably do so only under conditions that are not normally accessible geologically. High pressures and low temperatures would probably be required, at least for the first relative to quartz-albite-paragonite.

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