Polysomatism and the classification of minerals

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ABSTRACT. — The concepts of polysomatism and polysomatic series take into account both chemistry and structure of minerals. Therefore they seem well worth considering for the purpose of emphasizing, for instance, parageneses and solid-state transformations of the minerals. The following new examples of polysomatic series are given.

Series of the inophite - The S_mX series is based on S (serpentine) and X [$Mg_6Si_2O_3(OH)_{14}\cdot H_2O$] modules. Known members are serpentine, carlosturanite and defective structures observed as lamellae intergrown within carlosturanite itself. The series accounts for minerals which can be defined as H_2O-rich and Si-poor serpentine. The stability of the members is discussed.

Series of the antigorite - The $S_m^+ S_{m'}^- T$ series is based on S (serpentine) and T (talc) modules (+ and − indicate the opposite polarities of the phyllosilicate layer). Polysomes of the series are serpentine itself and the antigorite minerals with different a parameters and Si/Mg ratios.

Series of the schafarzikite - Schafarzikite, versiliaite, apuanite and their defective structures are shown to be member of a polysomatic series Sc_mU_n based on Sc (schafarzikite) and U [$^{VI}Fe_4^{3+}{}^{IV}Fe_4^{3+}{}^{III}Sb_4^{3+}O_{16}S_2$] modules. Intergrowth of different polysomes explains the non stoichiometry revealed by some analyses.

Series of the 6 × 9 Å structures - The series A_mB_n is based on A (lawsonite) and B (AlSiO$_3$OH) modules. By coupling modules, also through translations and rotations, seven so called 6 × 9 Å structures are obtained: sursassite, macfallite, pumpellyite, julgoldite, ardennite (two types) and lawsonite.

Key words: polysomatism, classification of minerals, inophite, antigorite, schafarzikite.

RIASSUNTO. — I concetti di polisomatismo e di serie polisomatica tengono conto delle caratteristiche chimiche e strutturali dei minerali e vengono ritenuti utili per la classificazione di questi quando li si voglia raggruppare al fine di evidenziarne, per esempio, relazioni paragenetiche e trasformazioni allo stato solido. Si riportano i seguenti nuovi esempi di serie polisomatiche.

Serie dell'inofite - La serie S_mX si costruisce accoppiando moduli S (serpentino) e X [$Mg_6Si_2O_3(OH)_{14}\cdot H_2O$] e comprende il serpentino, la carlosturanite e strutture difettive osservate come lamelle nella carlosturanite stessa. La serie rende conto di minerali definibili come serpentini ricchi in H_2O e poveri in Si. Viene discussa la stabilità dei polisoma noti.

Serie dell'antigorite - La serie $S_m^+ S_{m'}^- T$ si costruisce accoppiando moduli S (serpentino) e T (talco) (+ e − indicano le polarità opposte che gli strati fillosilicatici possono assumere). Tale serie comprende il serpentino e l'antigorite pensata come un insieme di termini caratterizzati da peculiari valori del parametro a e del rapporto Si/Mg.

Serie della schafarzikite - Si mostra come schafarzikite, versiliaite, apuanite e le loro strutture difettive siano i membri di una serie polisomatica Sc_mU_n costituita da moduli Sc (schafarzikite) e moduli U [$^{VI}Fe_4^{3+}{}^{IV}Fe_4^{3+}{}^{III}Sb_4^{3+}O_{16}S_2$]. Le concrescite spiegano la non stechiometria trovata in alcune analisi.

Serie delle strutture 6 × 9 Å - Questa serie A_mB_n è basata sulla combinazione di moduli A (lawsonite) e B (AlSiO$_3$OH). Combinando tali moduli, anche tramite traslazioni e rotazioni, si mostra che a tale serie polisomatica appartengono sursassite, macfallite, pumpellyite, julgoldite, ardennite (due tipi) e lawsonite.

Parole chiave: polisomatismo, classificazione dei minerali, inofite, antigorite, schafarzikite.

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Introduction

The purpose of any classification is to establish a simple although comprehensive scheme, in order to collect, compare and distinguish individual objects and to predict unknown phenomena to some extent. Such a scheme must rely on few basic features of the objects to be classified. Whereas people usually agree on the aim of a classification, disagreement often arises just where basic features are concerned. In fact, individual aptitudes and immediate purpose largely determine the particular choice. For instance, different approaches such as X-ray diffraction or optical microscopy or field experience often lead to quite different classification of minerals. This is understandable and, maybe, fruitful as stated by LIEBAU (1985), who maintains that « the best classification which can be chosen is the one that is best able to serve the particular purpose under consideration ».

Both very broad and very specific classification schemes have been proposed for minerals. Some authors (e.g., STRUNZ, 1938; ZOLTAI, 1960; POVARENNYKH, 1972; LIMA DE FARIA, 1983; LIEBAU, 1985) preferred to cover very large topics and proposed quite broad classification schemes. Although the importance of these efforts can hardly be overemphasized, these comprehensive schemes may become partially inadequate, or cumbersome, when small arrays are to be dealt with. In the latter case a decreasing generality could be hopefully balanced by a deeper insight into the relevant phenomena. In fact, very specific classification schemes have been proposed for well defined classes of compounds [e.g., MEIER (1968) for zeolites; GOTTARDI and GALLI (1985) for natural zeolites; SMITH and RINALDI (1962) for framework silicates based on four- and eight-membered rings].

Most of the quoted approaches assume the chemical nature of the minerals just as a boundary condition, useful to define which objects are to be considered. In other words, structural crystallography may dominate over crystal chemistry, as chemistry does not directly enter within the set of basic rules. As a result, almost geometric schemes may be produced (LIMA DE FARIA, 1983).

The concepts of polysomatism and polysomatic series (THOMPSON, 1978) simultaneously take into account both structure and chemistry. Through these concepts, features such as solid-state transformations and paragenetic assemblages can quite straightforwardly be included within the classification scheme. Polysomatism, therefore, seems to us well worth considering when we are interested not only in the classification, but also in the behaviour of minerals, even though it suffers from lack of generality, from its rather specialized nature and limitations in the number of classified objects.

A few examples of polysomatic series will be shown in this paper, just to stress how the approach works. The list is by no means intended to be exhaustive, as the examples only refer to our recent work.

Polysomes and polysomatic series

The basic ideas that underlie the polysomatic theory may be found scattered throughout a literature dating back to when people started recognizing the occurrence of common structural and chemical modules that, differently combined, produce a whole family of closely related structures (e.g., MAGNÉLI, 1953; WARSHAW and ROY, 1961; KOHN and ECKART, 1965; ROTH and WADSLEY, 1965). THOMPSON (1970 and 1978), eventually, put forward a comprehensive formulation of the theory, exploited its implications in terms of compositional space, chemographic relationships and possible paragenetic assemblages, and discussed several examples of polysomatic series.

The basic building units of a polysomatic series are one-dimensional beam modules, or two-dimensional layer modules, whose structure and chemistry are usually idealized. By the combination of two, or perhaps more, modules, in different ratios and/or with different combination rules, several structures are produced which define the members of the polysomatic series. These series are collinear in composition, namely the idealized compositions of the members linearly range between the two end-member compositions. Polysomes (from the Greek = several bodies)

are akin to polytypes, in that some kind of mixed-layer polytypism can be found within them. A further improvement was attained on introducing exchange vectors (THOMPSON, 1981) which afford an efficient, and easily mastered, approach to isomorphic substitutions making allowance for compositions other than the end members. Typical examples of polysomatic series are biopyriboles (THOMPSON, 1978, 1981; VEBLEN, 1981), humites (THOMPSON, 1978), serpentine and chlorite (THOMPSON, 1978), pyroxenoids (KOTO et al., 1976).

The inophite polysomatic series

Crystal-chemistry

Inophite is the name of a polysomatic series which includes the serpentine-like mineral carlosturanite $Mg_{21}Si_{12}O_{28}(OH)_{34} \cdot H_2O$ and the related structures which occur

re. In fact, the structure of carlosturanite (fig. 1) just consists of the same octahedral sheet which occurs in $1:1$ trioctahedral silicates, with modifications in the tetrahedral sheet: rows of silicon vacancies in the tetrahedral centres, compensated by four hydrogen atoms, split the tetrahedral sheet into parallel hydrogen bonded strips. The silicate strip consists of triple chains which are generated by connection of four-repeat single-crankshaft chains. Depending on the abundancy and distribution of tetrahedral vacancies, many structures are possible, with different chain-multiplicity and chemistry (in particular, they have inversely correlated silicon and water contents).

These structures form a polysomatic series, as all of them are made of two chemically distinct modules, S and X in fig. 1. The S module is a (100) section of the flat-layer serpentine structure with $Mg_3Si_2O_5(OH)_4$ composition. The X module corresponds to
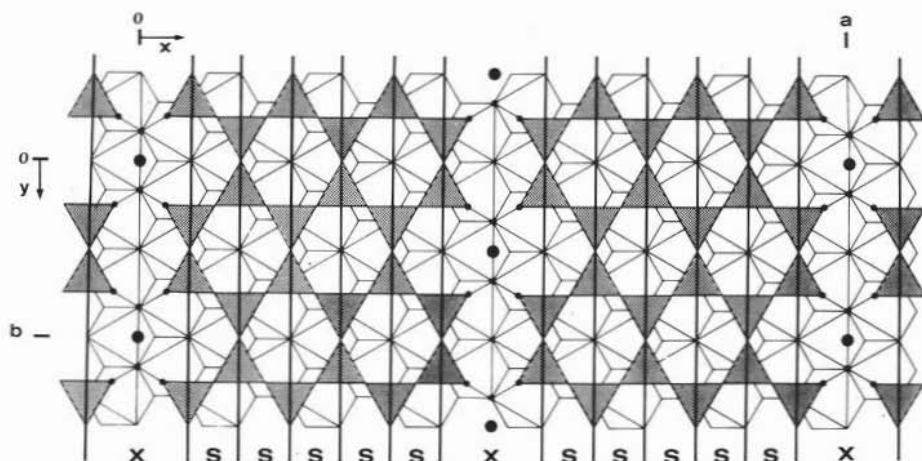

Fig. 1. — Structure of carlosturanite sliced into S (serpentine) and X (see text) modules. [After MELLINI et al. (1985)].

as fault lamellae within carlosturanite itself (COMPAGNONI et al., 1985; MELLINI et al., 1985). This has been the first polysomatic series to be voted and accepted as such by the International Mineralogical Association (IMA).

Carlosturanite is an asbestiform silicate, strongly reminding chrysotile in aspect and properties. The similarity of properties is due to the close structural and chemical relationships existing between carlosturanite and the idealized flat-layer serpentine structu-

an hypothetical hydro-silicate which can be derived from serpentine by substitution of $[(OH)_6 \cdot H_2O]^{6-}$ for $[Si_2O_7]^{6-}$ and has $Mg_6Si_2O_3(OH)_{14} \cdot H_2O$ composition. The complete series would be X, SX, ..., S_5X (carlosturanite itself), ..., S (serpentine); namely the different polysomes are structurally and chemically intermediate between the hypothetical hydro-sorosilicate X and the layer silicate serpentine S. The ideal chemical formula of the general S_mX polysome is simply $[Mg_3Si_2O_5(OH)_4]_m \cdot Mg_6Si_2O_3(OH)_{14} \cdot$

H_2O. As regards the unit cells, the b and c parameters are common to serpentine, whereas a is $[(m + 2) / 2]\, a_s$ ($a_s = a$ serpentine) and the cell is primitive when m is even. Viceversa, $a = (m + 2)\, a_s$ and the cell is C-centered when m is odd; for carlosturanite $(m = 5)$ $a = 36.70$, $b = 9.41$, $c = 7.291$ Å, $\beta = 101.1°$.

Several faults actually affect the a periodicity of carlosturanite and can be interpreted as chain multiplicity faults. Usually they are isolated lamellae, continuous within (010) and scattered throughout the carlosturanite matrix. Only other three periodicities besides the 18 Å one ($a/2$ of carlosturanite) have been observed in the lattice images. They are the 16 Å- and the 21 Å-periodicities, and the 24 Å-half-periodicity of the S_4X, S_6X and S_7X polysomes, respectively. Among them, S_6X and S_7X are common, whereas S_4X is rare.

be assessed. Carlosturanite is likely to be a real phase indeed, namely a stable assemblage, within the $MgO\text{-}SiO_2\text{-}H_2O$ system, as indicated by its abundance in the holotype locality and by its widespread occurrence. In particular, textural analysis at the optical and electron microscopy scale, indicated diopside + chrysotile + carlosturanite as a stable paragenetic assemblage. On this basis, COMPAGNONI et al. (1985) suggested $P_{H_2O} \approx 2$ kbar and $T \approx 250\text{-}300°$ C as likely conditions for the formation of carlosturanite in the light of the available data on the serpentinite metamorphism (TROMMSDORFF, 1982). Eventually, carlosturanite is replaced by brucite and chrysotile, according to the reaction:

$$Mg_{21}Si_{12}O_{28}(OH)_{34} \cdot H_2O \rightarrow$$
$$\text{carlosturanite}$$

$$6Mg_3Si_2O_5(OH)_4 + 3Mg(OH)_2 + 3H_2O.$$
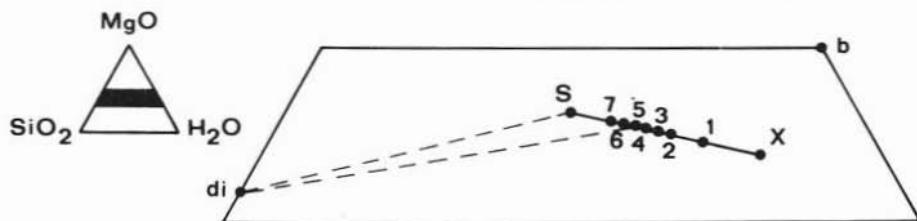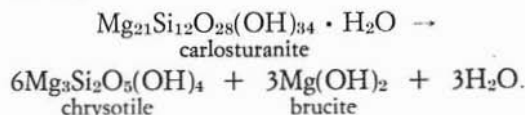$$\text{chrysotile} \qquad\qquad \text{brucite}$$

Fig. 2. — Chemical relationships among X, SX, ..., S polysomes of the inophite polysomatic series within the Mg-SiO₂-H₂O system (di = diopside, b = brucite).

Phase relationships

The whole compositional range of the inophite polysomatic series defines a tie-line which joins the serpentine composition to the X hydro-sorosilicate, within the $MgO\text{-}SiO_2\text{-}H_2O$ system (fig. 2). Most probably, carlosturanite is an important phase for metamorphosed maphics and ultramaphics. In fact, after the first description of the mineral, new findings were reported, from Taberg, Sweden (MELLINI and ZUSSMAN, 1986) and from several localities within the Lanzo Massif, Western Alps, serpentinites (unpublished data). On the other hand, no structure other than S_5X (carlosturanite) has yet been found as discrete crystals, but only as faults. On the basis of these observations the stability, or instability, of the different polysomes can

Polysomes other than S_5X are probably metastable, formed as growth defects, under $P\text{-}T$ conditions not far from the carlosturanite stability field. Quite interesting, the compositions of the observed faults just cluster around the carlosturanite composition (fig. 2), and two of them are less hydrated structures. A partially similar behaviour has been reported for other polysomatic series too, such as biopyriboles. In this series, no discrete crystal other than jimthompsonite or chesterite was found, whereas many different periodic or aperiodic structures were observed as metastable phases by HRTEM (VEBLEN and BUSECK, 1979).

The antigorite polysomatic series

Chemical analyses show (WHITTAKER and WICKS, 1970; WICKS, 1979; WICKS and

PLANT, 1979) that antigorite has a Si content definitely higher than that required by the ideal formula $Mg_3Si_2O_5(OH)_4$. Antigorite is based on the fundamental cell of serpentine except for the a parameter which, as for inophite, is a variable « multiple » of $a_s \approx 5.3$ Å. KUNZE (1961 and previous papers there quoted), on the basis of an experimental two-dimensional electron density map, proposed a structural model for antigorite with $a = 43.3$ Å, the most frequent value of this parameter. The model is characterized by a [100] sinusoidal modulation of the serpentine layer in such a way that tetrahedra belonging to adjacent half waves point in opposite directions, as shown in the highly schematic fig. 3. Whilst in crysotile the rolling of the layer compresses the octahedral sheet, in antigorite the folding (not shown in fig. 3) is opposite and compression is higher for the already smaller tetrahedral sheet. This sheet can therefore accomodate an extra [010] row of disilicate groups in each wave, in such a way that one out of two inversion sites has an octahedral row sandwiched between tetra-

inversion lines, SPINNLER (1985) describes antigorite in terms of a polysomatic series based on three modules. To avoid too strict a dependence upon a poorly known structure model, we propose a two-module polysomatic series to describe antigorite. Our modules are S, with the serpentine composition as for inophite, and T with the talc composition $Mg_3Si_4O_{10}(OH)_2$ to take into account the extra tetrahedra. These modules are (100) slabs one octahedron thick, i.e. $\sqrt{3}/2$ the octahedral edge. Noting with S^+ and S^- the modules with an opposite polarity of the tetrahedral sheet, the whole series can be written $S_m^+ S_{m'}^- \cdot T$ or $[Mg_3Si_2O_5(OH)_4]_{m+m'}$ $[Mg_3Si_4O_{10}(OH)_2]$. According to HRTEM images (YADA, 1979) and KUNZE's (1961) model, the condition $m = m'$ is not necessary; i.e., the two half waves can be of different length. In each wave the number of [010] octahedral rows per cell is $m + m' + 1$ and the Si/Mg ratio is given by $[2(m+m'+2)]/[3(m+m'+1)]$, i.e. $Si/Mg > 2/3$. By comparison, the m-th inophite member has a Si/Mg ratio given by $[2(m+1)]/[3(m+2)]$, i.e. $Si/Mg < 2/3$; in both cases when the
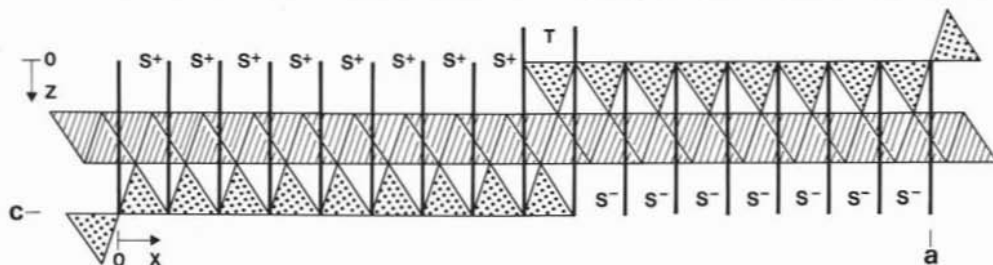

Fig. 3. — Schematic representation of the antigorite structure ($a = 43.3$ Å member) sliced into S (serpentine) and T (talc) modules.

hedra as in talc (T module in fig. 3). Details of the structure, particularly at the wave inversions, are still controversial (UEHARA and SHIROZU, 1985), but the number and the position of the extra tetrahedra accounting for the excess Si seem beyond dispute. According to this model, the variability of the a parameter can be connected with different lengths of the structural wave and requires a variable Si excess which does not contrast the available chemical data (WHITTAKER and WICKS, 1970).

In order to take into account the conclusions of KUNZE (1961) concerning the connections of the tetrahedra at the wave

number m of S modules tends to be infinite the ideal serpentine composition is obtained with $Si/Mg = 2/3$.

In the sample studied by KUNZE (1961) there are 16 octahedral rows across a and the average width of each row is $43.3/16 = 2.71 = 3.14 \sqrt{3}/2$ Å, which corresponds to the value expected for an ideal brucite layer (a_b = edge of the octahedron = 3.14 Å). According to the recent study by UEHARA and SHIROZU (1985) the different values measured for the a parameter of antigorite are multiples of $2.7 = a_b\sqrt{3}/2$ Å, more than of $a_s/2 = 2.6$ Å. Members with $m + m'$ odd and even thow P and C monocli-

nic cell, respectively; the value of the a parameter turns out to be $(m+m'+1)\,2.7$ Å and $2\,(m+m'+1)\,2.7$ Å in the two cases, respectively. Experimental a values not integer multiple of 2.7 Å are interpreted as average values from a mixture.

The schafarzikite polysomatic series

Schafarzikite, versiliaite and apuanite define a group of closely related minerals. Whereas schafarzikite is strictly an iron and antimony oxide, versiliaite and apuanite are more correctly defined as iron and antimony oxysulfides. Selected data for these minerals are given in table 1.

Schafarzikite was considered to be the

TABLE 1

Crystal-chemical data for minerals of the schafarzikite polysomatic series

Species	Ideal formula	a(Å)	b(Å)	c(Å)	Space group
Schafarzikite	$^{VI}Fe_4^{2+}\,^{III}Sb_8^{3+}O_{16}$	8.568	8.568	5.957	$P4_2/mbc$
Versiliaite	$^{VI}Fe_4^{2+}\,^{VI}Fe_4^{3+}\,^{IV}Fe_4^{3+}\,^{III}Sb_{12}^{3+}O_{32}S_2$	8.499	8.326	11.935	Pban
Apuanite	$^{VI}Fe_4^{2+}\,^{VI}Fe_8^{3+}\,^{IV}Fe_4^{3+}\,^{III}Sb_{16}^{3+}O_{48}S_4$	8.367	8.367	17.959	$P4_2/mbc$

parent structure of the whole group (MELLINI et al., 1979). Its crystal structure (fig. 4 a) is based upon chains of corner-sharing Sb^{3+}-O pyramids which are connected with chains of edge-sharing $Fe^{2+}O_6$ octahedra (ZEMANN, 1951; FISCHER and PERTLIK, 1975). The crystal structure of versiliaite is formally derived from schafarzikite (fig. 4 b), by substituting every fourth Sb^{3+} cation in the pyramidal chain by a Fe^{3+} cation, and
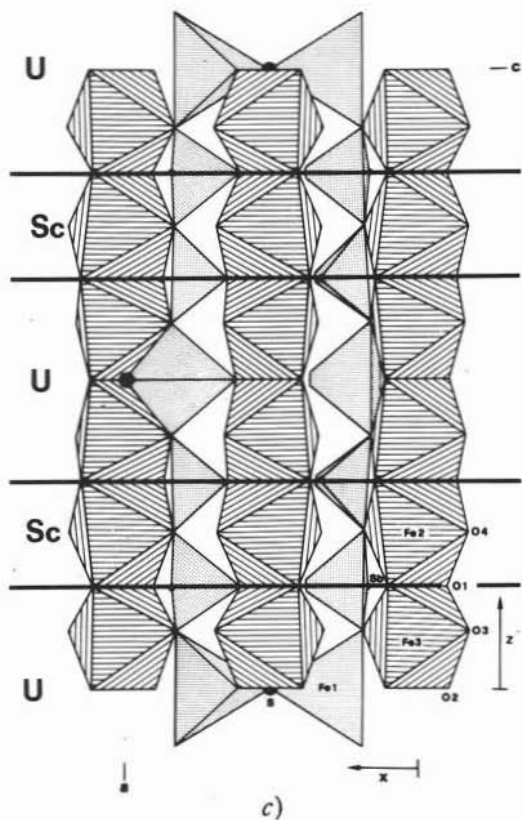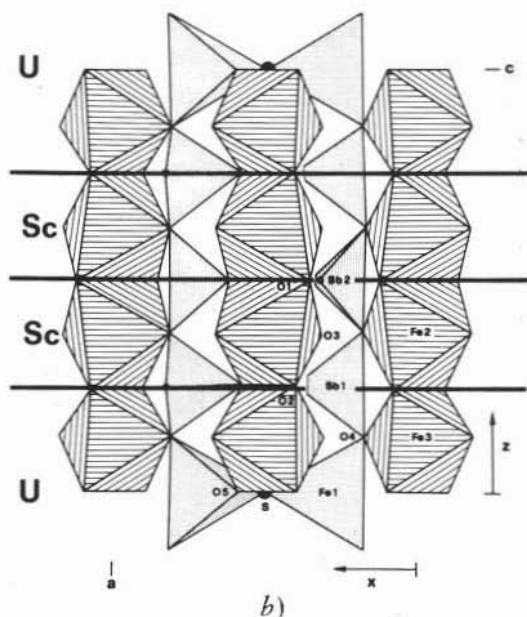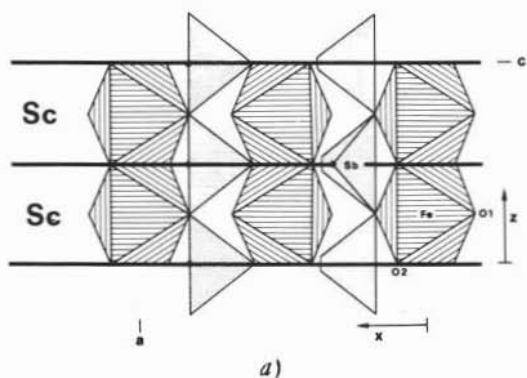

a)

b)

c)

Fig. 4. — Schafarzikite (a), versiliaite (b) and apuanite (c) structures shown as members of a polysomatic series generated by Sc (schafarzikite) and U (see text) modules.

introducing a sulfide anion between pairs of Fe^{3+} cations belonging to two adjacent chains (MELLINI and MERLINO, 1979). The excess negative charges introduced with the sulfide anions are balanced by oxydation of two octahedral Fe^{2+} to Fe^{3+} per each sulfide anion.

Similarly, the crystal structure of apunite (fig. 4 c) is formally derived assuming the same substitution every third Sb^{3+}. A major structural difference exists between these two derivative structures. Whereas the pyramidal chains are connected by sulfur bridges to build up double-chain ribbons in versiliaite, they form infinite sheets in the case of apunite. A scheme for the different connections is given in fig. 5.
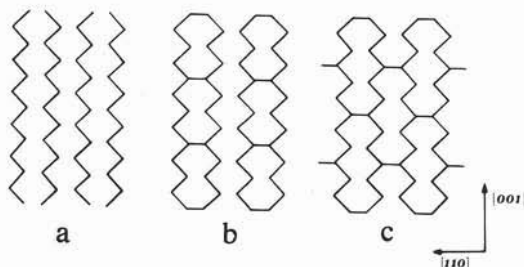

Fig. 5. — Connection of pyramidal chains through sulfur bridges in the schafarzikite polysomatic series: (a) single chain in schafarzikite; (b) double ribbons in versiliaite; (c) infinite layer in apunite.

Although MELLINI and MERLINO (1979) actually recognized the modular structure of these minerals, and described versiliaite as built up by two layers with $^{VI}Fe_4^{2+}$ $^{III}Sb_8^{3+}$ O_{16} and $^{VI}Fe_4^{3+}$ $^{IV}Fe_4^{3+}$ $^{III}Sb_4^{3+}$ $O_{16}S_2$ compositions respectively, the polysomatic nature of the whole group was not completely realized at that time. The systematic description of the group was instead performed mainly in terms of stuffed and substitutional derivative structures.

Adopting now the point of view of polysomatism, two fundamental layer modules can be recognized within the different structures, and they are suited to generate the whole family. These modules are sketched in figs. 4 and consist of (001) structural slabs. The first module, Sc hereafter, is just a (001) slab of the unmodified schafarzikite structure. The slab is one octahedron thick along [001], namely it has $c/2$ thickness. As shown below, the Sc module is to be

preferred over the two octahedra thick S layer which was chosen by MELLINI and MERLINO (1979), as this latter unit would fail to reproduce tetragonal structures such as apunite. The composition of the Sc module is obviously $^{VI}Fe_2^{2+}$ $^{III}Sb_4^{3+}$ O_8, namely schafarzikite itself is the first end-member of the polysomatic series. Its symmetry is described by the layer group 2Pba (THOMPSON, 1978), where 2 means a diad

TABLE 2
Crystal-chemical data for minerals of the 6×9 Å structures polysomatic series

Species	Ideal formula	a(Å)	b(Å)	c(Å)	β(°)	Space group
Sursassite	$Mn_2^{2+}(Al_3(OH)_3(SiO_4)(Si_2O_7))$	8.70	5.79	9.78	108.87	P2/m
Macfallite	$Ca_2(Mn_3^{3+}(OH)_3(SiO_4)(Si_2O_7))$	10.23	6.09	8.97	110.75	P2$_1$/m
Pumpellyite	$Ca_2(Al_3(OH)(SiO_4)(Si_2O_7))$	8.81	5.94	19.14	97.60	A2/m
Julgoldite	$Ca_2(Fe_3^{3+}(OH)_3(SiO_4)(Si_2O_7))$	8.922	6.081	19.432	97.60	A2/m
Ardennite	$Mn_4^{2+}(Al_6(OH)_6(AsO_4)(SiO_4)_2(Si_3O_{10}))$	8.71	5.81	18.52	--	Pnmm
Lawsonite	$Ca(Al_2(OH)_2(Si_2O_7)).H_2O$	5.80	8.83	13.20	--	Cmcm

axis normal to the layer module, and a and b are glide planes whose poles lie within the layer module. The second module, U, is two octahedra thick and contains all the modifications that produce the derivative structures: sulfide ion insertion, substitution of $^{IV}Fe^{3+}$ for $^{III}Sb^{3+}$ and of $^{VI}Fe^{3+}$ for $^{VI}Fe^{2+}$. It is just the same U layer already described for versiliaite, and has 2Pi layer group symmetry. As a consequence, the
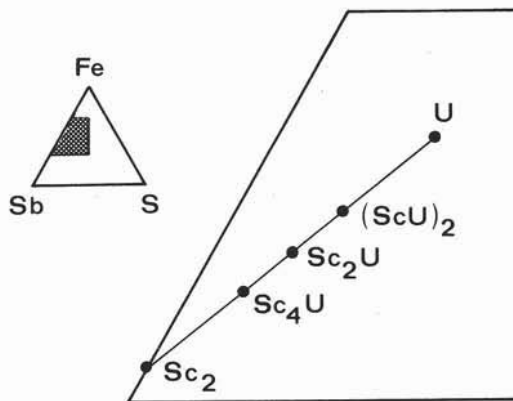

Fig. 6. — Chemical relationships among U, (ScU)₂ (apunite), Sc₂U (versiliaite), Sc₄U and Sc₂ (schafarzikite) polysomes of the schafarzikite polysomatic series within the Fe-Sb-S system.
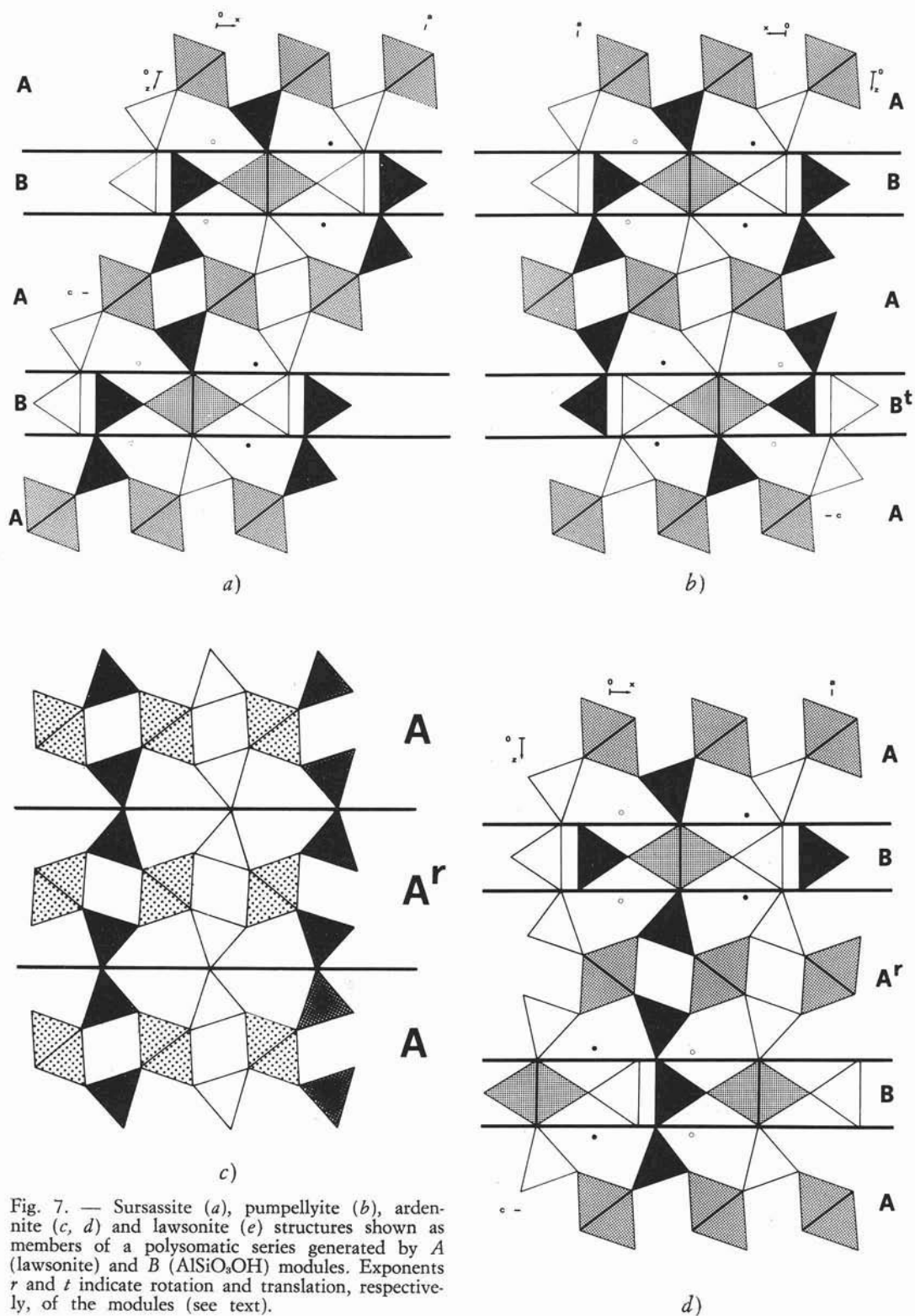
Fig. 7. — Sursassite (*a*), pumpellyite (*b*), arden-
nite (*c, d*) and lawsonite (*e*) structures shown as
members of a polysomatic series generated by *A*
(lawsonite) and *B* (AlSiO₃OH) modules. Exponents
r and *t* indicate rotation and translation, respective-
ly, of the modules (see text).

second end-member is the so called « 6 Å unknown orthorhombic structure » (MELLINI and MERLINO, 1979), namely it corresponds to the structure with the highest predictable sulfur content. With these choices, the different structures become

Schafarzikite Sc_2
Versiliaite Sc_2U
Apuanite $ScUScU = (ScU)_2$
Unknown 6 Å orthorhombic structure U
Unknown 18 Å orthorhombic structure (MELLINI et al., 1981) Sc_4U

Fig. 7 e)

The ideal unit cell content of the general Sc_mU_n member can be expressed as $(^{\text{IV}}Fe_2^{2+}\ ^{\text{III}}Sb_4^{3+}\ O_8)_m + (^{\text{IV}}Fe_4^{3+}\ ^{\text{VI}}Fe_4^{3+}\ ^{\text{III}}Sb_4^{3+}\ O_{16}\ S_2)_n$ and the c periodicity is $\approx (3\,m + 6\,n)$ Å. Orthorhombic Pbam structures occur when the polysomatic formula contains an even number of Sc layers between subsequent U layers. Otherwise, tetragonal $P4_2/mbc$ structures are obtained.

The chemical relationships among the different ideal polysomes are depicted in fig. 6. On considering the actual chemical data,

they plot along the tie-line, but are shifted from the expected positions. For instance, the chemical analyses for apuanite are found between the Sc_2U and the $(ScU)_2$ points. Similarly, versiliaite plots between Sc_2U, that is versiliaite itself, and Sc_2, that is schafarzikite. Namely, the crystal has bulk chemical properties intermediate between two adjacent ideal compositions. The explanation for this non-stoichiometric behaviour was given by MELLINI et al. (1981), who found intermixed occurrence of different structures.

The 6 × 9 Å structures

This group includes a number of structures, so called because of the approximate values of their axial translations (MOORE et al., 1985). Members of the group are lawsonite, sursassite (and the isostructural macfallite), pumpellyite (and the isostructural julgoldite), ardennite, orientite, ruizite, santafeite, bermanite. According to MOORE et al. (1985), they can be described in terms of common $_\infty^2[M_2\Phi_2(TO_4)_2]$ sheets occurring in all of them. Different intersheet material is sandwiched between sheets and gives rise to different tetrahedral polymerization. For istance, Si_2O_7 groups occur in lawsonite, SiO_4 and Si_2O_7 groups in sursassite and pumpellyite, SiO_4 and Si_3O_{10} groups in ardennite and orientite, Si_4O_{10} groups in ruizite. A few of these structures (table 2) will now be considered and their polysomatic nature will be stressed. Furthermore, the examples will show that either slightly different or highly different structures can be produced starting from equal amounts of the same, alternating fundamental layer-modules, just by different choice of stacking vectors.

Let us refer to sursassite as the starting member for the whole description. Its crystal structure (MELLINI et al., 1984) consists of edge-sharing octahedral chains, cross-linked by corner-sharing SiO_4 and Si_2O_7 tetrahedral groups (fig. 7 a). Following MOORE et al. (1985), the fundamental 6 × 9 Å sheet, named A in fig. 7 a, can be easily recognized. Two adjacent A modules sandwich the B module of fig. 7 a, having $M^{3+}SiO_3OH$ composition. For the
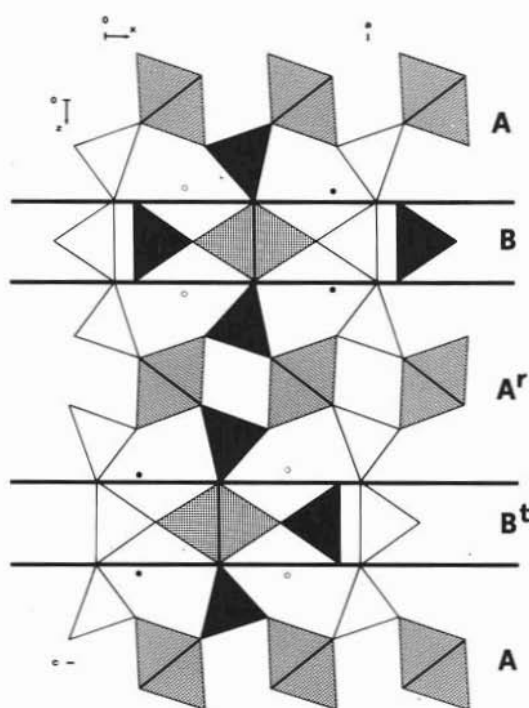
sake of generality, we extend now the A module to embed also the out-of-the-wall large cations, such as calcium or divalent manganese, and water molecules (X). Its chemical composition becomes $X_2M_2(OH)_2(SiO_4)_2$. Therefore, sursassite and isostructural macfallite can be easily described as consisting of alternating A and B modules (6.6 and 2.7 Å thick, respectively), AB being their polysomatic formula.

Pumpellyite and jugoldite are polytypic variants of the sursassite structural type. The most important difference is that, in the first case, similar tetrahedral groups face each other on the two sides of the octahedral chains (fig. 7 b), and, in the second case, different groups face each other (fig. 7 a). The difference is due to the different stacking vectors of the same fundamental modules. In particular, whereas each A layer of sursassite directly overlaps a corresponding A layer in pumpellyite, no direct overlap occurs for the B layers. In fact, every second B layer of pumpellyite is shifted by $1/2\ (\vec{a}+\vec{b})$ with respect to sursassite. The polysomatic formulae can be expressed as AB for sursassite, and as $ABAB^t$ for pumpellyite, with t indicating the $1/2\ (\vec{a}+\vec{b})$ vector. Similarly, more complex polytypes might be analyzed; for instance, the $\ldots t_s\,t_p \ldots$ 36 Å polytype reported by MELLINI et al. (1984) would become $ABABAB^tAB^t$.

The polysomatic analysis can be extended to other structures which do not otherwise reveal their close chemical and structural relationships with sursassite and pumpellyite. For instance, ardennite, $Mn_4^{2+}[Al_6(OH)_6(AsO_4)(SiO_4)_2(Si_3O_{10})]$, exhibits major structural modifications, as SiO_4 and Si_3O_{10} groups are now present. However, an appropriate slicing of its structure immediately reveals that ardennite too consists of alternating A and B modules, in equal amounts, and is a polymorphic modification of sursassite and pumpellyite (fig. 7 c). Whereas adjacent A layers are identical in sursassite and pumpellyite, they are correlated by a [100] twofold axis in ardennite. By specifying with the symbols A and A^r this relationship, the polysomatic formula for ardennite comes out to be ABA^rB. A further possibility is the introduction of B^t modules as those occurring in pumpellyite. Actually, they are possible, and the polysome ABA^rB^t

(fig. 7 d) just corresponds to a polysomatic variant for ardennite which was first assumed by MELLINI and MERLINO (1982) as possible structure for orientite, and then actually found to occur as fault structure in orientite crystals by MELLINI et al. (1986).

The end-members of this A_mB_n polysomatic series would have A and B compositions, respectively. As regards the A end-member, it actually exists and corresponds to lawsonite, $Ca[Al_2(OH)_2(Si_2O_7)]\cdot H_2O$ (fig. 7 e). Its polysomatic formula is AA^r, and, quite interesting, such a structure is also present as fault structure within the sursassite crystals (MELLINI et al., 1984). Possible B structures, which would have $AlSiO_3OH$ composition, do not seem to be known, instead. Obviously, other A_mB_n polysomes might exist and their chemical composition and d_{001} spacing would be obtained by the appropriate linear combination of A and B modules.

Conclusions

The fundamental question must now be faced of whether and where the approach is suitable.

A first, important application is connected with structural crystallography and topology of modular structures. For instance, in the case of ardennite and sursassite-pumpellyite, structures which would appear quite different when analyzed in terms of the tetrahedral polymerizations, immediately appear to be polymorphs and built up by the same fundamental modules.

A second field of application is due to those cases where the full understanding of the chemical data is quite important. This possibility comes out from the fact that polysomes just embed within their definition the required chemical information. An example is the non-stoichiometric behaviour within the schafarzikite group.

Bearing in mind the inophite polysomatic series as a third example, a third important reason can be found in the study of solid state transformations, growth defects, sub-solidus phenomena, parageneses and mineral equilibria, namely in mineral reactivity. In fact mineral reactivity strongly depends on

chemistry as well as on structure, and both these features are well taken into account by the use of the polysomatism theory.

As regards the identification of basic modules, it seems to us that an effort is required to identify the most significant ones, from the point of view of their physical soundness and possible crystalchemical use. This means that, based upon an appropriate choice, more and more structures may by found to be based upon these modules, just as these fundamental building units correspond to the most efficient ways of linking together individual atoms and represent energetically favourable pathways towards crystal growth.

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