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Article

The epidote–törnebohmite polysomatic series: structural relationships and complexity

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Abstract

The ET_n homologous row of the epidote–törnebohmite polysomatic series is considered, which includes minerals of the epidote ($n = 0$) and gatelite ($n = 1$) supergroups and radekškodaite group ($n = 2$). The crystal structures of members of the series are based upon the alternation of epidote (E) and törnebohmite (T) two-dimensional modules. The aristotype structure types of the members of the row crystallise in the $P2_1/m$ space group with the unit-cell parameters $a \approx 8.90$, $b \approx 5.65$, $c \approx (10.10 + 7.50n)$ Å, $\beta \approx 116.5°$ and $V \approx (455 + 338n)$ Å^3. The general formula for the members of the row can be expressed as $A_{2(n+1)}M_{n+3}[Si_2O_7][SiO_4]_{2n+1}X_{n+2}$ (X = O, OH and F). The structure model for the hypothetical ET_3 member of the series is constructed and the general formulae are derived for the calculation of the number of atoms per unit cell and the number and multiplicities of the atom sites for any value of n. The concept of K-sequence is introduced that is analogous to the concept of a Wyckoff sequence. The general formulae for the information-based complexity parameters are derived for the ET_n homologous row of the epidote–törnebohmite polysomatic series with different parities of n. The present absence of the members of the series with $n > 2$ reflects the entropic restrictions on the polysomatic series that confirm the principle of maximal simplicity for modular inorganic structures.

Keywords: polysomatism; polysomatic series; crystal structure; chemical composition; epidote supergroup; gatelite supergroup; radekškodaite group; törnebohmite; structural complexity; information-based complexity parameters

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Introduction

Polysomatism is an important concept in structural mineralogy and inorganic crystal chemistry that allows one to establish relationships among different structure types (Thompson, 1970, 1978), correlate their chemical composition, structure and properties (Veblen, 1991; Ferraris, 1997), and predict new structure types (Merlino and Pasero, 1997; Collins *et al.*, 2017; Ogawa and Walsh, 2025), etc. Though the term 'polysomatism' is due to Thompson (1978), the concept of modular construction of crystal structures and structural homology was recognised long before (see, e.g. Magnéli, 1953; Bragg and Claringbull, 1965). Ferraris *et al.* (2005) provided both conceptual and factual overviews of the concepts of modularity and polysomatism (note that the term 'polysome' is different from 'polymorph'; in contrast to polymorphs, different polysomes may have different chemical compositions). A number of recent works outline the usefulness of these approaches to the interpretation and description of both minerals and synthetic inorganic compounds (Cámara *et al.*, 2021; Malcherek *et al.*, 2021; Ciobanu *et al.*, 2022; Rao *et al.*, 2022; Yudintsev *et al.*, 2022; Conconi *et al.*, 2023; Hatert *et al.*, 2023; Aksenov *et al.*, 2023, 2024; Campo-Rodriguez *et al.*, 2024; Tsuchiya *et al.*, 2024).

This paper is devoted to the analysis of the epidote–törnebohmite polysomatic series. It was first proposed by Bonazzi *et al.* (2003) on the basis of their discovery of gatelite-(Ce), a mineral with a structure that can be described as an ordered alternation (ET) of epidote (E) and törnebohmite (T) two-dimensional modules. Further discoveries led Bonazzi *et al.* (2019) to the establishment of the gatelite supergroup that currently contains seven mineral species. In 2020, Kasatkin *et al.* discovered the natural ETT ($= ET_2$) polysome of the epidote–törnebohmite polysomatic series and defined the radekškodaite group that now contains three members: radekškodaite-(Ce), radekškodaite-(La) and zilbermintsite-(La) (Kasatkin *et al.*, 2020, 2024).

In addition to the interest from the crystal-chemical point of view, the minerals of the epidote–törnebohmite polysomatic series attract attention as a potential source of rare earth elements (REE) (Hentschel *et al.*, 2020). For instance, allanite is mined as one of the REE ore minerals in the Nechalacho deposit (Northwest Territories, Canada) (Haque *et al.*, 2014; see also: Xiao and Zhang, 2024). As the REE content of the epidote–törnebohmite polysomatic series increases from epidote to radekškodaite, further investigations are warranted to define the role of these minerals as REE concentrators in natural systems.

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This paper is part of a collection in tribute to the work of Edward Grew at 80

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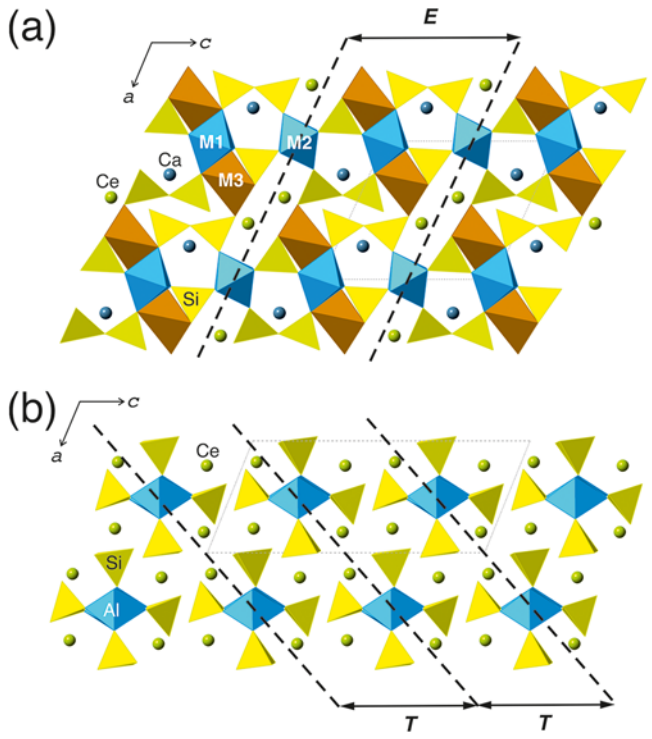


Figure 1. The crystal structures of allanite-(Ce) (a) and törnebohmite-(Ce) (b) projected along the *b* axes with the indication of the *E* and *T* modules by the dashed lines, respectively. Legend: $M^{3+}O_6$ octahedra = light-blue; $M^{2+}O_6$ octahedra = brown; SiO_4 tetrahedra = yellow; Ce and Ca (A) atoms are shown as green and blue balls, respectively. Drawn using the *CrystalMaker 8.7* program.

The aim of the present study is to derive the crystal-chemical formulae for the ET_n row of the series ($n \geq 0$) and analyse the evolution of information-based structural complexity parameters with the increasing *n* value. We shall demonstrate that for $n > 4$, the total information content of the crystal structure aristotypes is above 1000 bit per unit cell, which makes the corresponding polysomes unlikely to occur in nature as ordered crystals.

General crystallography of the epidote–törnebohmite series

Homologous rows

The crystal structures of allanite-(Ce) and törnebohmite-(Ce) as end-members of the series with indications of the *E* and *T* modules, respectively are in Fig. 1. The modular description of the representative members of the polysomatic series are in Table 1. It should be noted that the existing members of the series (Table 1) represent only one possible homologous row, namely, one that includes addition of *T* modules to the initial *E* module. This homologous row can be described by the general formula ET_n. Other homologous rows can be theoretically foreseen, such as EET_n, $EETET_n$ and $EETEET_n$. As only the ET_n members of the series have been observed in nature so far, only this homologous row is considered here.

Aristotype structures

Each member of the polysomatic series can be characterised by its aristotype structure, which is defined here as a structure type with maximal possible symmetry. For instance, the crystal structure of perbøeite-(Ce) is an aristotype for the *ET* member, whereas that of gatelite-(Ce) is not, as the latter is a 2×1×1 superstructure with regard to its aristotype; its *a* parameter is doubled (Table 1). By analogy, the experimentally determined crystal structure of törnebohmite-(Ce) does not correspond to its aristotype, owing to its 1×1×2 superstructure. Table 2 contains full information about the aristotype structure model of törnebohmite-(Ce). The aristotype structures of all members of the ET_n homologous row of the epidote–törnebohmite polysomatic series belong to the $P2_1/m$ space-group-type. Their chemical formulae and approximate unit-cell parameters are listed in Table 3 as dependent upon the *n* value (i.e., the number of törnebohmite modules in a polysome). Figs 2a and b shows crystal structures of perbøeite-(Ce) and radekškodaite-(Ce) as aristotype structures for the *ET* and ET_2 members of the polysomatic series, respectively.

K-sequences

The concept of *K*-sequence is a generalisation of the concept of Wyckoff sequence that reflects the diversity and numbers of Wyckoff sites occupied in a particular crystal structure (Hornfeck, 2022). Usually, the Wyckoff sequence contains the space-group number followed by a sequence of Wyckoff letters corresponding to the sites occupied by atoms in a particular crystal structure. In cases of more than one site of particular Wyckoff type, the number of such sites is indicated by a superscript. The following example is taken from Hornfeck (2022). The unit cell of the crystal structure of $RbAg_4I_5$ (space group $P4_132$ (no. 213)) contains 80 atoms distributed over six Wyckoff sites (crystallographic orbits) of four different types: two 24*e* (*x*, *y*, *z*); one 12*d* (1/8, *y*, *y* + ¼); two 8*c* (*x*, *x*, *x*); and one 4*a* (3/8, 3/8, 3/8). The total Wyckoff sequence is 213 e^2 *d* c^2 *a*. The *K*-sequence is a sequence of multiplicities of the occupied Wyckoff sites with the number of sites with the same multiplicities written in a subscript. For the crystal structure of $RbAg_4I_5$, the *K*-sequence is simply $24^2 12^1 8^2 4^1$. The *K*-sequence does not distinguish between different Wyckoff sites with the same multiplicity. For instance, the crystal structure of allanite-(Ce) (Hoshino *et al.*, 2005) contains one 2*a* (0, 0, 0), one 2*c* (0, 0, ½), thirteen 2*e* (*x*, ¼, *z*), and three 4*f* (*x*, *y*, *z*) sites. The Wyckoff sequence is 11 f^3 e^{13} *c* *a*, whereas the *K*-sequence is $4^3 2^{15}$.

The *K*-sequences in Table 4 are for the representative members of the epidote–törnebohmite polysomatic series listed in Table 1 classified according to the atom types (A, M, Si, O and X) as well as *K*-sequences for the total crystal structures. The detailed analysis of the dependencies of the *K*-sequences upon the *n* value for the known members of the ET_n homologous row allows one to derive general formulae, which are different for even and odd *n*. The corresponding formulae are $4^{(5n+7)/2} 2^{9n+14}$ and $4^{(5n+6)/2} 2^{9n+15}$ for even and odd *n* values, respectively. The number *v* of atoms in the unit cell is expressed by the same formula, irrespective of the parity of *n*: $v = 14(2n + 3)$. In contrast, the numbers *k* of occupied Wyckoff sites (crystallographic orbits) are also dependent on the parity of *n* and are equal to $(23n + 35)/2$ and $(23n + 36)/2$ for odd and even *n*, respectively. It should be emphasised, however, that the formulae obtained (see also Table 3) are for the aristotype structures of the respective polysomes. In reality, structure types may be different for the same aristotype, owing to the formation of superstructures, as is the case for gatelite-(Ce) that has the 2×1×1 superstructure relative to its aristotype (as a consequence, the space-group type changes from $P2_1/m$ to $P2_1/c$).

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Table 1. Representative members of the epidote–törnebohmite polysomatic series

Mineral name	Chemical formula	MC**	Space group	*a*, Å	*b*, Å	*c*, Å	β, °	*V*, Å^3	Ref.
Allanite-(Ce)	$(CaCe)(Al_2Fe^{2+})[Si_2O_7][SiO_4]O(OH)$	*E*	$P2_1/m$ (11)	8.905	5.761	10.123	114.78	471.5	1
Perbøeite-(Ce)	$(CaCe_3)(Al_3Fe^{2+})[Si_2O_7][SiO_4]_3O(OH)_2$	*ET*	$P2_1/m$ (11)	8.894	5.619	17.755	116.95	794.8	2
Gatelite-(Ce)	$(Ca,Ce)_4(Al,Mg,Fe)_4[Si_2O_7][SiO_4]_3(O,F,OH)_3$	*ET*	$P2_1/c$ (14)	17.770	5.651	17.458	116.18	1573.3	3
Radekškodaite-(Ce)	$(CaLa_5)(Fe^{2+}Al_4)[Si_2O_7][SiO_4]_5O(OH)_3$	ET_2	$P2_1/m$ (11)	8.960	5.727	25.113	116.63	1152.0	4
Törnebohmite-(Ce)	$Ce_2Al[SiO_4]_2(OH)$	*T*	$P2_1/c$ (14)	7.383	5.673	16.937	112.04	657.5	5
Törnebohmite-(Ce)*	$Ce_2Al[SiO_4]_2(OH)$	*T*	$P2_1/m$ (11)	7.383	5.673	8.964	112.04	328.8	5

*aristotype structure model; ** modular composition
References: (1) Hoshino *et al.* (2005); (2) Bonazzi *et al.* (2014); (3) Bonazzi *et al.* (2003); (4) Kasatkin *et al.* (2020); (5) Shen and Moore (1982).

Table 2. Aristotype structure model for törnebohmite-(Ce)

Atom	Site	*x*	*y*	*z*
Al	2*d*	½	0	½
Si1	2*e*	0.529	¼	0.419
Si2	2*e*	0.931	¼	0.338
O1	2*e*	0.382	¼	0.933
O2	2*e*	0.749	¼	0.969
O3	4*f*	0.508	0.012	0.720
O4	2*e*	0.041	¼	0.548
O5	2*e*	0.694	¼	0.553
O6	4*f*	0.999	0.476	0.789
OH	2*e*	0.324	¼	0.429
Ce1	2*e*	0.813	¾	0.456
Ce2	2*e*	0.219	¾	0.370

Table 3. Crystallographic parameters and formulae for the calculation of information-based complexity parameters for the aristotype structures of the ET_n row of the epidote–törnebohmite polysomatic series

n	odd	even
Chemical formula	$A_{2(n+1)}M_{n+3}[Si_2O_7][SiO_4]_{2n+1}X_{n+2}$ (X = O, OH, F)	
Space group	$P2_1/m$ (11)	
a [Å]	≈ 8.90	
b [Å]	≈ 5.65	
c [Å]	≈ (10.10 + 7.50*n*)	
β [°]	≈ 116.5	
V [Å^3]	≈ (455 + 338*n*)	
k [orbits]	(23*n* + 35)/2	(23*n* + 36)/2
v [atoms]	14(2*n* + 3)	
K-sequence	$4^{(5n+7)/2}2^{9n+14}$	$4^{(5n+6)/2}2^{9n+15}$
I_G [bits/atom]	$\log_2[7(2n+3)] - (5n+7)/[7(2n+3)]$	$\log_2[7(2n+3)] - (5n+6)/[7(2n+3)]$
$I_{G,total}$ [bits/cell]	$14(2n+3)\log_2[7(2n+3)] - 2(5n+7)$	$14(2n+3)\log_2[7(2n+3)] - 2(5n+6)$

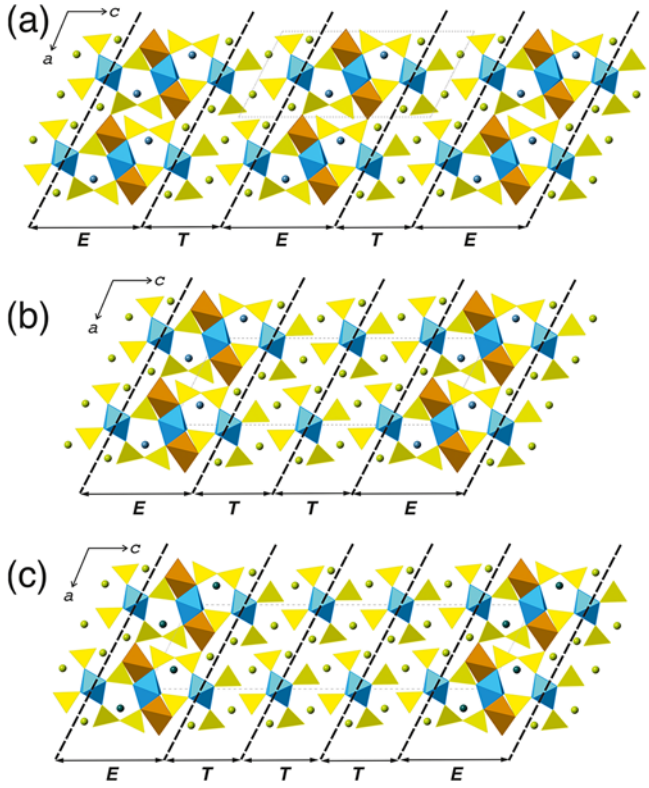


Figure 2. The crystal structures of: (a) perbøeite-(Ce); (b) radekškodaite-(Ce); and (c) hypothetical ET_3 members of the epidote–törnebohmite polysomatic series projected along the *b* axes with the indication of the *E* and *T* modules by the dashed lines. Legend as in Fig. 1.

On the basis of atom coordinates for the ET_n members of the series with $n = 0$, 1 and 2, the structure model for the hypothetical ET_3 polysome with $n = 3$ was constructed (see Table 5 for crystallographic parameters and Fig. 2c for the structure diagram).

Information-based analysis of structural complexity

The evolution of structural complexity with the changing *n* value in the ET_n homologous row of the series was investigated using information-based measures first proposed in Krivovichev (2012, 2013) and developed in Krivovichev *et al.* (2022).

For a crystal structure having *v* atoms per reduced unit cell split into *k* crystallographic orbits with multiplicities (m_1, m_2, …., m_k), the amounts of structural Shannon information per atom (I_G; in bits/atom) and per unit cell ($I_{G,total}$; in bits/cell) are calculated according to the following equations:

$$I_G = -\sum_{i=1}^{k} p_i \log_2 p_i \quad \text{(bits/atom)} \qquad (1)$$

$$I_{G,total} = v \times {}^{str}I_G = -v \times \sum_{i=1}^{k} p_i \log_2 p_i \quad \text{(bits/cell)} \qquad (2)$$

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Table 4. *K*-sequences and information-based complexity parameters* for some members of the epidote–törnebohmite polysomatic series

Mineral name	MC**	*n*	Space group	*K*-sequences					Complexity parameters			
				A	M	Si	O+X	total	*k* [orbits]	*v* [atoms]	I_G [bits/atom]	$I_{G,total}$ [bits/cell]
Allanite-(Ce)	*E*	0	$P2_1/m$	2^2	2^3	2^3	$4^3 2^7$	$4^3 2^{15}$	18	42	4.107	172.477
Perbøeite-(Ce)	*ET*	1	$P2_1/m$	2^4	$4^1 2^2$	2^5	$4^5 2^{12}$	$4^6 2^{23}$	29	70	4.786	335.050
Gatelite-(Ce)	*ET*	1	$P2_1/c$	4^4	4^4	4^5	4^{22}	4^{35}	35	140	5.129	718.100
Radekškodaite-(Ce)	ET_2	2	$P2_1/m$	2^6	$4^1 2^3$	2^7	$4^7 2^{17}$	$4^8 2^{33}$	41	98	5.288	518.242
Hypothetical	ET_3	3	$P2_1/m$	2^8	$4^2 2^2$	2^9	$4^9 2^{22}$	$4^{11} 2^{41}$	52	126	5.628	709.137
General (n = even)	ET_n	n	$P2_1/m$	$2^{2(n+1)}$	$4^{n/2} 2^3$	2^{2n+3}	$4^{2n+3} 2^{5n+7}$	$4^{(5n+6)/2} 2^{9n+15}$	(23*n*+36)/2	14(2*n*+3)	see Table 3	
General (n = odd)	ET_n	n	$P2_1/m$		$4^{(n+1)/2} 2^2$			$4^{(5n+7)/2} 2^{9n+14}$	(23*n*+35)/2			
Törnebohmite-(Ce)	*T*	-	$P2_1/c$	4^2	4^1	4^2	4^9	4^{14}	14	56	3.807	213.212
Törnebohmite-(Ce)***	*T*	-	$P2_1/m$	2^2	2^1	2^2	$4^2 2^5$	$4^2 2^{10}$	12	28	3.522	98.606

*without contributions from H atoms
**modular composition
***aristotype structure model

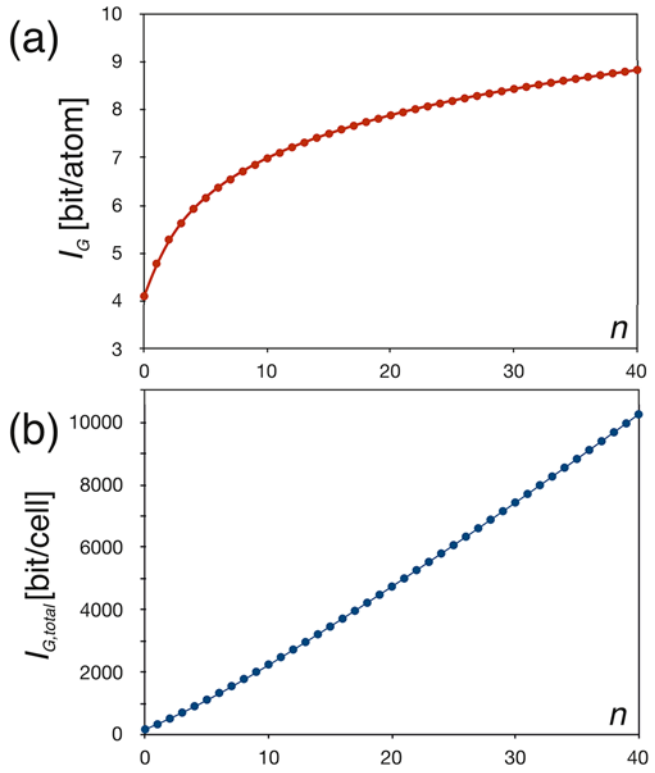


Figure 3. The dependence of the atomic (a) and total (b) information contents *versus n* value for the ET_n homologous row of the epidote–törnebohmite polysomatic series.

where p_i is the random choice probability for an atom from the *i*th crystallographic orbit, that is:

$$p_i = m_i / v \quad (3)$$

The structural complexity parameters for the representative members of the series listed in Table 1 are given in Table 4. The analytical expression of the *K*-sequences given in Table 3 (note their dependence on the parity of *n*) allows for the derivation of formulae for the calculation of structural complexity parameters using *n* as the only variable. The formulae are given in Table 3, whereas Fig. 3 shows the behaviour of both I_G and $I_{G,total}$ values with increasing *n*. The dependence is almost linear for the total amount of structural information amount exceeding the level of 1000 bits/cell for $n = 5$. At this value, the structure belongs to the very complex class (Krivovichev, 2013, 2014), though there are no principal obstacles for its formation either in nature or experiments. However, possible natural occurrences of such structures would be very rare, as even the radekškodaite-group members are considered as rare mineral species, presently known from one world locality only (Mochalin Log, South Urals, Russia, Kasatkin *et al.*, 2020, 2024).

Conclusions

As has been pointed out elsewhere (Krivovichev, 2013, 2014; Krivovichev *et al.*, 2022), modularity (or polysomatism) is one of the most important complexity-generating mechanisms in the realm of mineral and synthetic inorganic structures. In the complexity analysis of polysomatic series, it is important to look at the complexity of aristotype structures (maximal-symmetry realisations of polysomes). The example of the epidote–törnebohmite

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Table 5. Atom coordinates* for the aristotype $A_8M_6[Si_2O_7][SiO_4]_7X_5$ structure of the hypothetical ET_3 member of the epidote–törnebohmite polysomatic series

Atom	Site	*x*	*y*	*z*	Atom	Site	*x*	*y*	*z*
M1	2*a*	0	½	0	O6	4*f*	0.2384	0.4930	0.0100
M2	2*e*	0.3124	¾	0.0670	O7	2*e*	0.3740	¼	0.9665
M3	4*f*	0.0280	0.4995	0.1600	O8	2*e*	0.4848	¼	0.2090
M4	4*f*	0	½	0.3850	O9	2*e*	0.0559	¼	0.3530
A1	2*e*	0.7710	¼	0.0494	O10	2*e*	0.9676	¾	0.1910
A2	2*e*	0.2294	¾	0.7210	O11	2*e*	0.0944	¼	0.1320
A3	2*e*	0.5923	¼	0.3620	O12	4*f*	0.3041	0.4748	0.3420
A4	2*e*	0.4306	¾	0.1820	O13	4*f*	0.8140	0.5129	0.1090
A5	2*e*	0.6161	¼	0.1360	O14	4*f*	0.3271	0.4784	0.1140
A6	2*e*	0.2470	¾	0.2770	O15	4*f*	0.7842	0.5123	0.3340
A7	2*e*	0.7530	¼	0.4930	O16	4*f*	0.7138	0.5231	0.2000
A8	2*e*	0.4080	¾	0.4080	O17	2*e*	0.0541	¼	0.0480
Si1	2*e*	0.2014	¼	0.1020	O18	4*f*	0.2419	0.4883	0.2130
Si2	2*e*	0.3556	¼	0.2290	O19	2*e*	0.5174	¼	0.0560
Si3	2*e*	0.7027	¾	0.0890	O20	2*e*	0.5534	¾	0.1030
Si4	2*e*	0.8393	¾	0.2160	O21	2*e*	0.4520	¼	0.2830
Si5	2*e*	0.6719	¾	0.3150	O22	2*e*	0.5360	¾	0.3340
Si6	2*e*	0.1828	¼	0.3280	O23	2*e*	0.0600	¼	0.2750
Si7	2*e*	0.3412	¼	0.0120	O24	2*e*	0.9590	¾	0.2690
Si8	2*e*	0.3280	¼	0.4550	O25	4*f*	0.2158	0.0123	0.4364
Si9	2*e*	0.8172	¾	0.4419	O26	2*e*	0.4130	¼	0.5096
O1	2*e*	0.9256	¼	0.4070	O27	2*e*	0.4640	¼	0.4356
O2	2*e*	0.1068	¾	0.1380	O28	2*e*	0.9440	¾	0.4165
O3	2*e*	0.9504	¼	0.1830	O29	4*f*	0.6960	0.9748	0.4279
O4	2*e*	0.0639	¾	0.0420	O30	2*e*	0.9400	¾	0.4951
O5	2*e*	0.5870	¾	0.2600	O31	2*e*	0.0744	¾	0.3630

*$P2_1/m$, $a = 8.9702$, $b = 5.7044$, $c = 32.600$ Å, $\beta = 116.766°$.

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polysomatic series considered here demonstrates that aristotype structures of the end-members of the series (epidote and törnebohmite) are relatively simple (172.477 and 98.606 bits/cell, respectively). The simplest combination of the *E* and *T* modules is that of *E*:*T* = 1 realised in the crystal structures of gatelite-supergroup minerals with the aristotype structure model more complex than those of the end-members (335.050 bits/cell). The addition of *T* modules results in the increasing structural complexity; the structures with $n > 2$ are presently unknown as minerals or synthetic compounds. It is likely that the higher members of the series can be found at the contacts between grains of lower members. It is of interest that mineral diversity generally correlates with the increasing trend of complexity in the polysomatic series: the epidote supergroup contains at present at least 36 mineral species, whereas the gatelite supergroup has seven, and the radekškodaite group only three. This further confirms the principle of maximal simplicity for modular inorganic crystal structures proposed in Krivovichev (2021): structures with minimal information dominate over other structures with the same building principles. This principle has entropic origins and reflects the tendency of inorganic nature to form structures of high entropy.

Finally, it should be pointed out that temperature-driven phase transitions can be expected for those members of the series that form superstructures relative to their aristotypes (e.g. törnebohmite and gatelite-(Ce)) with the space-group change from $P2_1/c$ to $P2_1/m$ with twofold unit-cell volume contraction.

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Competing interests. The authors declare none.

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