Classification of Eudialyte-Group Minerals

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Abstract—According to the rules accepted by the Commission on New Minerals, Nomenclature and Classification, International Mineralogical Association, the definition of a new mineral species within the eudialyte group should be based on the general crystal chemical principle of the predominance of a new element at one or several key sites. To date, 25 minerals of the eudialyte group have been approved in line with this recommendation. Additionally, 50 samples (including potential new species of the eudialyte group) have been investigated using IR spectroscopy and X-ray diffraction. The new data require the modification and complementation of the current systematics of the eudialyte group. The eudialyte-type structures contain unique fragments that involve species-forming key sites. The crystal chemical classification that develops in this study is based on the hierarchical principle that takes into account the following features (listed in order of their lowering classification rank):

(i) simple or modular character of the crystal structure and value of c parameter (about 30 and 60 Å for 12- and 24-layer minerals, respectively);

(ii) symmetry determined by cation ordering in the octahedral ring, i.e., the possibility of dividing site $M(1)$ into $M(1a)$ and $M(1b)$;

(iii) the distribution of different components into five $M(2)$ square-based sites;

(iv) the water content (subdivision of the eudialyte-group minerals into low-and high-hydrous species with Na > ($H_2O + H_3O$) and Na < ($H_2O + H_3O$), respectively.

The 12-layer minerals are subdivided into eudialyte- and oneillite-type species. The further subdivision of these two types into subtypes is based on the predominance of octahedra, tetrahedra, or vacancies at sites $M(3)$ and $M(4)$ in the centers of nine-membered Si,O-rings and different combinations of these variants. Both *type* and *subtype* terms are used here in the crystal chemical sense. Among 24-layer minerals, the first subdivision is based on the degree of hydration; the second subdivision into subtypes takes into account combinations of predominant components (Zr + Zr, Ti + Ti, or Zr + Ti) in Z octahedra of different modules (floors). Various eudialyte-group minerals that belong to different crystal chemical taxons are briefly characterized.

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INTRODUCTION

Scientific interest in eudialyte, a worldwide abundant zirconosilicate, is due, in particular, to its pivotal role in the mineralogy and geochemistry of Zr. In the pioneering works on the crystal chemistry of eudialyte, Nikolai Vasil'evich Belov has foreseen that more than one mineral is hidden behind this name. He wrote that "… minerals of eudyalite group can be rock-forming in pegmatites." (Belov, 1976).

According to the recommendations of the Subcommittee on eudialyte of the Commission on New Minerals, Nomenclature, and Classification of the International Mineralogical Association (CNMNC IMA), the composition of the eudialyte-group minerals can be written with allowance for crystal chemical features, such as $N(1)_3$ $N(2)_3 N(3)_3 N(4)_3 N(5)_3 M(1)_6 M(2)_{3\,-\,6} M(3) M(4) Z_3$ $[Si_{24}O_{72}] O'_{4\,-\,6} X_2$ (Johnsen et al., 2003a), where $N(1-5) =$

Na, H_3O^+, K, Sr, REE, Y, Ba, Mn, and Ca; $M(1) =$ Ca, Mn, REE, Na, Sr, Fe; $M(2) =$ Fe, Mn, Na, Zr, Ta, Ti, K, Ba, and H_3O; $M(3)$ and $M(4) =$ Si, S, Nb, Ti, W, Na; Z = Zr, Ti, Nb; O' = O, OH, H_2O; $X(1)$ and $X(2) =$ Cl, F, H_2O, OH, CO_3, SO_4, AlO_4, and MnO_4. The literal notations indicate the set of elements and their site in the crystal structure (Fig. 1). As a rule, one letter corresponds to a group of closely spaced sites rather than to one site, i.e., to a microblock in the unit cell, which can be occupied on a competitive basis by components that differ in size, coordination number, and charge. Even with this reservation, the aforementioned formula does not involve all kinds of crystal chemical features of the eudialyte-group minerals, in particular possible additional sites of H_2O or cation ordering in an octahedral ring with doubling site $M(1)$ to form sites $M(2)$ and $M(1b)$.

The heteropolyhedral framework of eudialyte is made up of three- and nine-membered rings of silica tetrahedra and six-membered rings of $M(1)O_6$ octahe-

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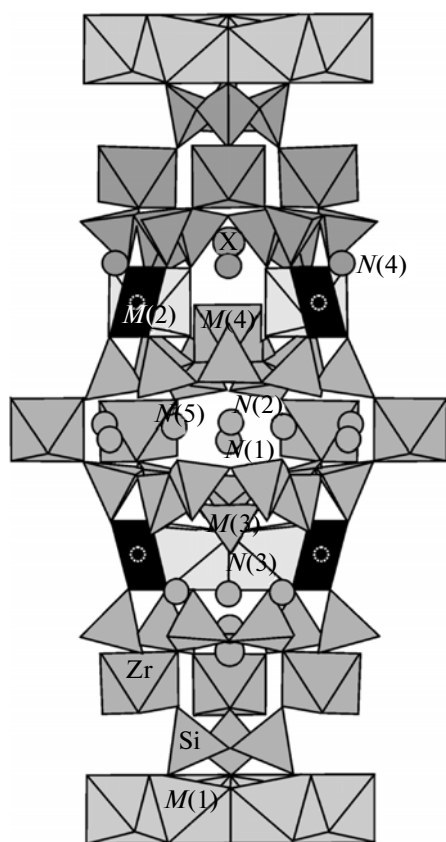[1] Corresponding author: R. K. Rastsvetaeva. E-mail: rast@ns.crys.ras.ru

Fig. 1. Polyhedral image of structure of eudialyte-group minerals. Letters denote cation sites.

dra joined by isolated ZrO_6 octahedra. The cations of alkali and alkali-earth metals, hydronium, and transition elements, additional large anions, and water molecules are disposed in the cavities of the framework. The variety of the eudialyte-group minerals with similar structural motifs is related to the key sites or microblocks, where local but crucial structural readjustments with change of valency and coordination number and the substitution of an atom with a group of atoms are possible.

Species with relatively low-ordered 12-layer structure described by a hexagonal unit cell with a ~14 Å and c ~30 Å are the most abundant in the eudialyte group. The ordering of cation occupancy, which can touch on almost all intraframework and framework sites (except for Si sites of Si,O-rings), gives rise to the formation of superstructures in some eudialyte-group minerals with multiple c period about 60 Å discovered by A.P. Khomyakov in the Lovozero and Khibiny plutons, Kola Peninsula, Russia. In the notations recommended by the Subcommittee, the composition of these 24-layer minerals is schematically described by the general formula $[N(1)N(1)*N(2)N(2)*N(3)N(3)*N(4)N(4)*N(5)N(5)$ $*N(6)N(7)]_3[M(1)M(1)*]_6[M(2)M(2)*]_3M(3)M(3)*$

$M(4)M(4)*Z_3Z_3^*[Si_{24}O_{72}][Si_{24}O_{72}]*\varphi_4\varphi_4^*X_2X_2^*$. Asterisked and nonasterisked symbols denote the homological sites of the first and second modules of the structure, respectively.

CRYSTAL CHEMICAL PRINCIPLES OF CLASSIFYING EUDIALYTE-GROUP MINERALS

According to the rules approved by CNMNC IMA (Johnsen et al., 2003a), the recognition of a new mineral in the eudialyte group is based on crystal chemistry and, for all minerals, the principle of a new element dominating at one or several key sites is universal. With allowance for these recommendations, 25 mineral species have been established so far in the eudialyte group. More than 50 varieties and potential new minerals have been studied with IR spectroscopy and X-ray diffraction.

The new data present a novel view on the classification of the eudialyte-group minerals. Rastsvetaeva (1992, 2006, 2007), Johnsen and Grice (1999), and Rastsvetaeva and Khomyakov (2003) summarized the crystal's chemical data on eudialyte at different stages of its investigation. However, until now, there has been no complete crystal chemical systematics that includes all minerals of the group.

First of all, it should be noted that there is no unsolved contradiction between the general rules of CNMNC IMA, which determines mineral species based on the predominant element at the structural site and hierarchical classification. The first attempt to coordinate both principles was made by Khomyakov (2007). According to his systematics, at the first level, the minerals are divided into two subgroups with 12-layer (a ~ 14.2, c ~ 30 Å) and 24-layer (a ~ 14.2, c ~ 60 Å) structural motifs. The next level of subdivision (subgroups of the second order) takes into account the attribution of the mineral in one of three space groups, i.e., (a) $R\bar{3}m$, (b) $R3m$, and (c) $R3$. Thus, the subgroups of the second order are divided into six structural types, i.e., 12a (subgroup of eudialyte), 12b (subgroup of kentbrooksite), 12c (subgroup of oneillite), 24a (subgroup of alluaivite), 26b (subgroup of rastsvetaevite), and 24c (subgroup of labyrinthite). This classification is based on general crystallographic characteristics (unit-cell dimensions and symmetry) without reference to the local crystal chemical features. In addition, most 24-layer minerals are centrosymmetrical and belong to $R\bar{3}m$ and $R\bar{3}$ space groups, which have not yet been found.

Previously, Bulakh and Petrov (2003, 2004) published formal hierarchical systematics of eudialytes based on the chemical composition without making allowance for fine crystal chemical features. The advantage of the formal hierarchical approach consists of the simple systematization of the data by chemical

composition (all components are listed in descending order of their content in atomic percents), which is convenient for processing large bodies of information and their inclusion in databases. At the same time, this classification does not reflect either the genetic or structural features of any sample.

Upon the development of a formal hierarchical systematization of the chemical compositions of eudialytes, Bulakh (2008, 2010) proposed mineral names, such as *eudialyte-(Ca,Fe)* or *eudialyte-(Ca,Mn)*, and reduced the entire range to a few mineral species with many chemical and structural varieties. At the same time, mineral formulas were revised taking into account the charge-compensation rule.

Yaroshevsky and Bagdasarov (2008) made other proposals concerning the systematics of the eudialyte-group minerals and the simplification of their nomenclature. These proposals are also based on the bulk chemical compositions of minerals without providing in-depth details on their role in the crystal structure. As a result, the authors introduced mineral names, such as *eudialyte-(X)*, where X = Na, Mg, Fe, NaFe, Mn, REE, NaREE, and Ca …, *niobo-eudialyte-(X)*, *titanium-eudialyte-(X)* but retained alluaivite, despite that it is also a Ti-dominant mineral, and *hydronio-boeudialyte*. It seems that this approach rules out not only the structural, but also genetic, implications of the mineral species. For example, in the eudialyte-group, Mn minerals can dominate at the $M(1)$, $M(2)$ site, or one of two sites N, and each of these minerals is related to special paragenetic assemblage.

The attempts to introduce efficient nomenclature, when a mineral name bears certain information on the mineral nature, would be welcomed. However, the current nomenclature of the eudialyte-group minerals based on the crystal chemical classification must take into account not only the chemical variations, but also the distribution of components by structural sites. The main problem in the systematics of these minerals is that the isomorphic chemical substitutions with regard to the structures of these minerals are not always understood in the classical sense (according to Goldschmidt), i.e., the replacement of one atom by another atom at the site with specified coordinates. In our opinion, isomorphism is the competition of two groups of atoms that cannot occur simultaneously in the same domain of the given unit cell, so that one block replaces another block that is close in size and shape, but has a distinct inner structure. In this case, the unit cell itself is retained. This generalized concept of isomorphism was suggested by Frank−Kamenetsky (1962, 1964) and was subsequently developed by Bulakh (1989, 1998), Bulakh, Krivovichev, and Zolotarev (1995), and Solodovnikov (2005) with the introduction of notions of block isomorphism and the assembly of a structure from fragments. As a rule, in these fragments (group of atoms and ions), a key element with the highest force characteristics or the greatest scattering ability is distinguished.

This paper, which does not focus on the development of nomenclature for eudialyte-group minerals, is concerned with the elaboration of convenient (as it is possible for the minerals with so complex and variable structures) and simple crystal chemical classifications.

CRYSTAL CHEMICAL CLASSIFICATION

The structural and crystal chemical classification of the eudialyte-group minerals proposed in this study (see also Rastsvetaeva and Chukanov, 2009) is based on the hierarchical principle, which takes into account the following features (in descending order of their rank):

(a) the modular character of the structure and a value of parameter c (~30 Å and ~60 Å for the 12- and 24-layer structures, respectively);

(b) the symmetry, which is determined by cation ordering in a six-membered ring composed of edge-shared octahedra accompanied by the breakdown of the octahedral Ca site $M(1)$ into $M(1a)$ and $M(1b)$ and the transformation of the Ca ring into a ring with alternation of Ca-octahedra and Mn- or Fe-centered octahedra, or Mn- and Na-octahedra;

(c) the distribution of various components by five key microdomains, including

(i) square $M(2)O_4$ and its derivatives with a coordination number of 5 or 6, i.e., $M(2)O_5$ and $M(2)O_6$ polyhedra centered by Fe, Mn, Na, Zr, and Ta atoms [below, the general symbol $M(2)$ is retained for all near-square subsites];

(ii) two microdomains close to the centers of two nine-membered rings $M(3)$ and $M(4)$ containing tetrahedra (SiO_4 and less frequent SO_4) or octahedra (NbO_6, WO_6, TiO_6);

(iii) the two most isomorphically capacious sites $N(3)$ and $N(4)$, along with their encirclement.

(d) the degree of hydration with subdivision into minerals with Na > (H_2O + H_3O) and Na < (H_2O + H_3O) (in apfu).

According to the aforementioned principles, the 12-layer members of the eudialyte group are subdivided into minerals with $M(1)$-disordered (eudialyte type, space group $R3m$ or $R\bar{3}m$) and $M(1)$-ordered (oneillite type, space group $R3$) structures (Tables 1, 2).

The second step of the subdivision of the eudialyte-type structures is based on centering nine-membered Si,O-rings by the additional Si-tetrahedra (T), $M(3)$- and $M(4)$-octahedra (M), and vacancies (□) or combination of these variants (Fig. 2). Thus, the eudialyte-type structures are subdivided into the structures of eudialyte (1.1 in Table 2), kentbrooksite (1.2 in Table 2), etc.

At the third step, the eudialyte-type structures are subdivided depending on the occupancy of the near-square site $M(2)$ = Fe (1.1.1) and $M(2) \neq$ Fe (1.1.2)

Table 1. Structural classification of eudialyte-group minerals

12-layer structures (Z = Zr)								24-layer structures			
1. Eudialyte-type structures					2. Oneillite-type structures***			3. Structures of low-hydrous members Na > H₂O + H₃O			4. Structures of high-hydrous members Na < H₂O + H₃O
1.1 variant $(T + T)$*	1.2 variant $(T + M)$**	1.3 variant $(M + M)$	1.4 variant $(\square + T)$	1.5 variant $(\square + \square)$	2.1 variant $(T + T)$	2.2 variant $(\square + T + M)$	2.3 variant $(\square + T)$	3.1 variant $Z_{Ti} + Z_{Ti}$	3.2 variant $Z_{Zr} + Z_{Ti}$	3.3 variant $Z_{Zr} + Z_{Ti}$	4.1 variant, hydrated eudialytes

Note: T is the tetrahedron in the center of nine-membered ring; M is the octahedron in the center of a nine-membered ring; $\square$ is the vacancy in the center of the nine-membered ring; Z_{Zr} is zirconium in the analog of the Z position; Z_{Ti} is titanium in the analog of Z position. * Subsequently, the eudialyte-type structures (variant $T + T$) are subdivided by a degree of hydration (aqualite and four varieties at the end of column in Table 2) and then, by cation at the $M(2)$ site. ** Subsequently, the eudialyte-type structures (variant $T + M$) are subdivided based on the degree of hydration (five samples at the end of the column in Table 2), then based on the major cation at the axial site $M(3)$, followed by site $M(2)$ and the major cation at site $N(4)$. ***The further subdivision of oneillite-type structures is performed in compliance with the cation couples $M(1a) + M(1b)$.

(Fig. 3), whereas minerals with the kentbrooksite structure are subdivided depending on the occupancy of axial octahedra M = Nb (1.2.1), W (1.2.2), and Mn (1.2.3). The minerals with kentbrooksite structure are further subdivided by the major cation at site $M(2)$, then by the major cation at site $N(4)$.

Oneillite-type structures, i.e., 12-layer structures with ordered $M(1)$-cations in octahedral ring and symmetry lowered down to $R3$, are also subdivided at the first step based on the centering of nine-membered Si,O-rings. The subsequent subdivision of these structures is conducted by different couples of $M(1a) + M(1b)$ cations.

At the first step, 24-layer minerals of the eudialyte group are subdivided into low-hydrous and high-hydrous members (Tables 1, 3), and subsequent subdivisions are based on the composition at sites of Z-octahedron at two different structural stages. Symbols Z_{Zr} and Z_{Ti} denote that site Z is occupied by Zr or Ti, respectively).

The minerals that belong to the selected cells of the crystal chemical systematics are briefly characterized below.

12-Layer Eudialyte-Type Structures

Most 12-layer minerals of the group have these structures, which are characterized by the six-membered ring consisting of equivalent Ca-dominant octahedra and by symmetry $R3m$ or $R\bar{3}m$. Depending on the dominant structural element (Si-tetrahedra T, octahedra M, or vacancy $\square$) near the centers of two nine-membered Si,O-rings in the structure of eudialyte, four variants of the structure are distinguished, i.e., eudialyte proper, kentbrooksite, ikranite, and mogovidite. Recently, the fifth variant ($M + M$) of the Nb- and Ti-rich sample (Rastsvetaeva et al., 2009b) has been added.

(1) Variant $(T + T)$ is observed in the structure of Si-rich minerals with additional Si-tetrahedra predominant near both centers of nine-membered rings. In turn, these structures are detailed by the occupancy of the $M(2)$ site at the center of the oxygen square or coordination polyhedra based thereon.

Eudialytes with the Fe-dominant site M(2). Only one mineral species is known satisfies this condition, and it is found in the crystal chemical subtype of eudialyte. This is *eudialyte proper*, i.e., $Na_{15}Ca_6Fe_3Zr_3Si(Si_{25}O_{73})(O,OH,H_2O)_3(Cl,OH)_2$ (Goly-
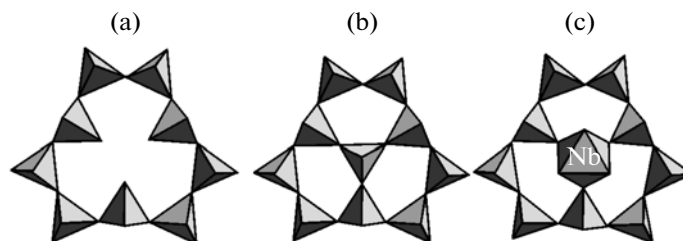

Fig. 2. Main structural fragments of the eudialyte-group minerals: (a) 9-membered Si,O-ring [Si_9O_{27}], (b) planar disk [$Si_{10}O_{27}(OH)$], and (c) heteropolyhedral planar disk [$Si_9O_{27}M(OH)_3$].

Table 2. Classification of 12-layer structures of eudialyte-group minerals and their members

1. Eudialyte-type structures					2. Oneillite-type structures		
1.1 variant $(T + T)$	1.2 variant $(T + M)$	1.3 variant $(M + M)$	1.4 variant $(\square + T)$	1.5 variant $(\square + \square)$	2.1 variant $(T + T)$	2.2 variant $(T + M)$	2.3 variant $(\square + T)$
Eudialyte proper	Kentbrooksite	*Zirconium–niobium silicate*	Mogovidite	Ikranite	Raslakite	Oneillite	*Low-Ca sample 2676*
Manganoeudialyte	Ferrokentbrooksite			Eudialyte Giuseppetti	Voronkovite		
Hyperzirconium sample-1	Carbokentbrooksite				*Low-Ca sample 2656*		
Low-Fe sample-2	Andrianovite				*Mn, Ca-ordered eudialyte*		
Low-Fe sample-5	Georgbarsanovite				*Mn,Ca-ordered eudialyte-2*		
Sample 1	Feklichevite				*Hyperzirconium sample - 2*		
Sample 7	Taseqite						
Sample 8	Zirsilite-(Ce)						
Sample 9	Golyshevite						
Sample 10	Khomyakovite						
Sample 12	Manganokhomyakovite						
Sample 14	Johnsenite-(Ce)						
Sample 15	*Feklichevite Khibiny*						
Low-Fe sample-4	*Barsanovite red-brown*						
Na-sample 3458	*Eucolite 368*						
K,Na-ordered eudialyte 3445	*Eucolite REE-Fe*						
High-Ti eudialyte	*Eucolite REE-Mn*						
Aqualite	*Sample 4*						
Hydrated sample-1	*Sample 11*						
Hydrated sample-2	*Sample 13*						
Hydrated sample-3	*Sample 16*						
Hydrated sample-6	*Sample 17*						
	Low-Fe sample-1						
	Potassium–hydronium sample						
	Hydrated sample-4						
	Hydrated sample-5						
	Low-Fe sample-3						

Note: Here and in Table 3, samples with the studied crystal chemistry and uncertain status as mineral species are given under provisional names and italicized.

shev, Simonov, and Belov, 1971), which contains basic elements in proportion close to stoichiometric Na : Ca : Fe : Si ~ 15 : 6 : 3 : 26. As a consequence, the site at the center of square is almost completely occupied by Fe^{2+} and tetrahedral complex anions composed of $10SiO_4$ tetrahedra are contained instead of two nine-membered Si,O-rings. Thus, the square- coordinated divalent iron and the tetrahedral cations at axial sites $M(3)$
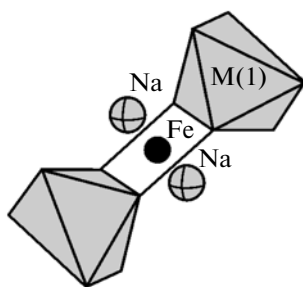
Fig. 3. Square and near-square $M(2)$ microdomains.

and $M(4)$ near centers of nine-membered Si,O-rings are characteristic of eudialyte proper.

Eudialytes with Mn-dominant site $M(2)$ are exemplified by high-Mn *manganoeudialyte*, i.e., $Na_{14}Ca_6Zr_3[Si_{26}O_{72}(OH)_2](Mn,Fe)_3(H_2O,Cl,O,OH)_6$ (Nomura et al., 2010). The Mn octahedron, which consists of four oxygen atoms of square and two water molecules on both sides is dominant in the near-square area. The mineral is distinguished by a high water content introduced as H_2O molecules and OH groups surrounding all near-square $M(2)$ polyhedra, as well as the apices of axial Si-tetrahedra and the sites of additional X anions.

Minerals that are rich in Si and depleted in Na, including *aqualite* $(H_3O)_9 Na_2(K,Ba,Sr)_2Ca_6Zr_3Si_{26}O_{66}(OH)_9Cl \cdot H_2O$, which was described by A. P. Khomyakov from the Inagli pluton, South Yakutia, is referred to this variant $(T + T)$ of the eudialyte-type structures. The mineral is highly hydrated (up to 10 wt % H_2O) and extremely depleted in Na and Fe oxides (3.5 wt % in

total). The mineral is highly saturated with hydronium group, which is predominant among the extra-framework cations and occurs at all large-cation sites (Rastsvetaeva and Khomyakov, 2002). Later, we found aqualite from hydrothermally altered pegmatite in the Kovdor alkaline ultramafic pluton.

(2) Variant $(T + M)$. Minerals with this structure are the most diverse. If site $M(4)$ on axis *3* is occupied by Si atoms, then in microdomain $M(3)$, Nb o,r less frequently, W octahedron is formed. The minerals with predominant Mn or Fe at the near-square site are distinguished within this variant.

Niobium minerals with the structure of kentbrooksite, which contains Mn at site $M(2)$, are *kentbrooksite* proper, i.e., $(Na,REE)_{15}(Ca,REE)_6Mn_3Zr_3Nb(Si_{25}O_{74})F_2 \cdot 2H_2O$ (Johnsen et al., 1998); its carbonate analog is *carbokentbrooksite*, i.e., $(Na,\square)_{12}(Na,Ce)_3Ca_6Mn_3$ $Fe_3Zr_3Nb[Si_{25}O_{73}](OH)_3(CO_3) \cdot H_2O$ (Khomyakov et al, 2003); K-rich mineral *andrianovite*, i.e., $Na_{12}(K,Sr,Ce)_3Ca_6Mn_3Zr_3NbSi[Si_3O_9]_2[Si_9O_{27}]_2(O,OH)_4(H_2O,CO_3,Cl)_2$ (Rastsvetaeva and Khomyakov, 2005); and REE mineral *zirsilite-(Ce)*, i.e., $(Na,\square)_{12}(Ce,Na)_3Ca_6Mn_3Fe_3Zr_3Nb[Si_{25}O_{73}](OH)_3(CO_3) \cdot H_2O$ (Khomyakov et al., 2003). All of the above-listed minerals are characterized by predominant polyhedra MnO_5 in microdomain $M(2)$ (unlike to eudialyte proper, in structure of which the coordination square $Fe^{2+}O_4$ is located in microdomain $M(2)$). Pentatopes MnO_5 are apex-shared with Nb octahedra to form clusters $[NbMn_3]$, which causes the acentricity of the structure and, in some cases, the piezoelectric effect.

Niobium minerals containing Fe at site $M(2)$ are *ferrokentbrooksite*, i.e., $Na_{15}Ca_6(Fe,Mn)_3Zr_3Nb[Si_{25}O_{73}]$

Table 3. Classification of 24-layer structures of eudialyte-group minerals

Low-hydrous (Na > H_2O + H_3O)			4. High-hydrous (Na < H_2O + H_3O)
3.1 alluaivite-type structure $Z_{Ti} + Z_{Ti}$	3.2 dualite-type structure $Z_{Zr} + Z_{Ti}$	3.3 rastsvetaevite-type structure $Z_{Zr} + Z_{Zr}$	4 variant $Z_{Zr} + Z_{Zr}$
Alluaivite *Alluaivite Khibiny*	Dualite	Rastsvetaevite Labyrynthite *K-depleted rastsvetaevite (sample 3765)* *Centrosymmetrical analog of labyrynthite* *Na,K-sample 1408* *Hypermanganese sample 3248* *Centrosymmetrical sample 3043_7* *Hydrated sample-I* *Hydrated sample-II*	*Hydrated rastsvetaevite (sample 3369)*

Note: See Table 2 for notes.

$(O,OH,H_2O)_3(Cl,F,OH)_2(CO_3) \cdot H_2O$ (Johnsen et al., 2003b); *feklichevite*, i.e., $Na_{11}Ca_9Fe_2Zr_3NbSi_{25}O_{73}$ $(OH,H_2O,O,Cl)_5$ (Rastsvetaeva, Ekimenkova, and Pekov, 1999); *georgbarsanovite*, i.e., $Na_{12}(Mn,$ $REE,Sr)_3Ca_6Fe_3^{2+}Zr_3NbSi_{25}O_{76}Cl_2 \cdot H_2O$ (Khomyakov et al., 2005); *taseqite*, i.e., $Na_{12}Sr_3Ca_6Fe_3Zr_3NbSi_{25}O_{73}(O,OH,H_2O)_3Cl_2$ (Ekimenkova, Rastsvetaeva, and Khomyakov, 2000a); and *golyshevite*, i.e., $(Na,Ca)_{10}Ca_9(Fe^{3+},Fe^{2+})_2Zr_3$ $NbSi_{25}O_{72}(CO_3)(OH)_3 \cdot H_2O$ (Chukanov et al., 2005). Feklichevite and golyshevite are examples of the eudialyte-group minerals with extremely high Ca contents (up to ~16 wt % CaO). Ca atoms occupy not only octahedral sites of six-membered rings, but also dominate at site $N(4)$. Thus, the minerals contain a layer composed of Ca-centered polyhedra, and the six-membered rings are a part of this layer. The structure of these minerals is similar to that of kentbrooksite, but the pentatope there in is dominated by Fe to form clusters $[NbFe_3]$, which emphasize acentric structures that result in a stable piezoelectric effect in georgbarsanovite. This structure differs from the structure of eudialyte proper, which is close to centrosymmetrical. The differences between the ten niobium minerals considered above also concern the occupancy of site $N(4)$. In kentbrooksite, ferrokentbrooksite, and carbokentbrooksite, this site is occupied by Na; Sr and REE are predominant in taseqite and zirsilite-(Ce), respectively; and, in georgbarsanovite, it is occupied by Mn, Sr, Ca, and some other cations with predominant Mn. In the structure of andrianovite, K, Sr, and Ce with predominant K occupy site $N(4)$.

W-bearing analogs of kentbrooksite are *khomyakovite*, i.e., $Na_{12}Sr_3Ca_6Fe_3Zr_3W[Si_{25}O_{73}](O,OH,H_2O)_3$ $(OH,Cl)_2$ (Johnsen et al., 1999); *manganokhomyakovite*, i.e., $Na_{12}Sr_3Ca_6Mn_3Zr_3W[Si_{25}O_{73}](O,OH,H_2O)_3$ $(OH,Cl)_2$ (Johnsen et al., 1999a); and *johnsenite-(Ce)*, i.e., $Na_{12}(Ce,La,Sr,Ca,\square)_3Ca_6Mn_3Zr_3W(Si_{25}O_{73})$ $(CO_3)(OH,Cl)_2$ (Grice and Gault, 2006). These minerals are rather rare and have been described from the Saint-Hilaire pluton in Canada; their structures contain polyhedral clusters $[WFe_3]$ or $[WMn_3]$, and site $N(4)$ is occupied by Sr or Ce (in johnsenite). Johnsenite also contains carbonate groups as species-forming components.

(3) Variant (M + M). This structure is characteristic of the recently studied potential new mineral $[Na,H_3O,Ca,K]_{15}Ca_6[\square,Mn,Fe]_3[Zr_{1.5}Nb_{1.5}][Si_{24}O_{68}]$ $(O,OH)_4][Ti_2](OH)_5Cl \cdot 4H_2O$ (Rastsvetaeva, Aksenov, and Chukanov, 2009) from the Kovdor phlogopite deposit, Kola Peninsula, where it is a secondary mineral that replaces a primary mineral from the eudialyte group. The low Zr and high Nb contents are typical of this mineral. Nb and Zr atoms occupy octahedral Z site of the framework, whereas Ti atoms occupy sites $M(3)$ and $M(4)$ on axis 3 in the center of nine-membered Si,O-rings (Fig. 4). The mineral can be classi-
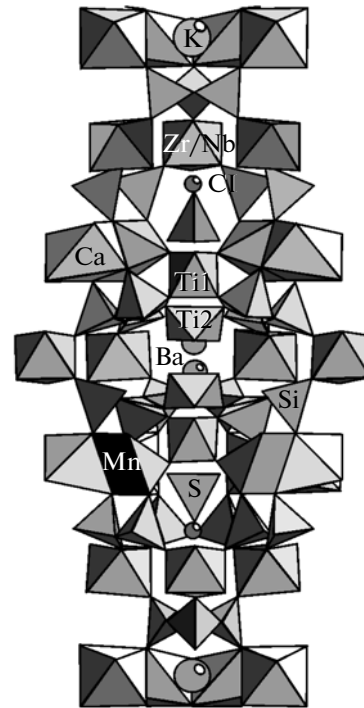

Fig. 4. Structure of Zr,Nb,Ti-eudialyte.

fied as either zirconosilicates and niobosilicates with the eudialyte-type structure.

(4) Variant ($\square$ + T) is exemplified in only one member, *mogovidite*, i.e., $Na_9(Ca,Na)_6Ca_6$ $(Fe^{3+},Fe^{2+})_2Zr_3[Si_{25}O_{72}](CO_3)(OH,H_2O)_4$ (Rosenberg et al., 2005), which is characterized by the highest Ca content among eudialytes. Ca occupies not only octahedral sites of the six-membered ring (as in most eudialyte-group minerals), but also the sites of extraframework cations, which dominate over Na at sites $N(3)$ and $N(4)$ to form a double Ca-layer.

(5) *Variant* ($\square$ + $\square$). Relatively rare samples depleted of metal cations are closest to eudialyte proper. To date, only 11 such minerals have been found and studied, and two of them (ikranite and aqualite) have been approved by CNMNC IMA as independent mineral species. Variant ($\square$ + $\square$) is observed in the structure of *ikranite*, i.e., $(Na,H_3O)_{15}(Ca,Mn,REE)_6Fe_3^{2+}Zr_3(\square,Zr)(\square,Si)Si_{24}$ $O_{66}(O,OH)_6Cl \cdot nH_2O$ ($n = 2-3$) (Rastsvetaeva and Chukanov, 2003). The mineral is characterized by predominant vacancies at both $M(3)$ and $M(4)$ sites in the center of the nine-membered ring. According to Mössbauer spectroscopy, 84.56% of iron is trivalent in ikranite, which is consistent with the chemical data. Another member of 12-layer ($\square$ + $\square$) minerals with the eudialyte-type structure is *Giuseppetti eudialyte*, i.e., $(Fe^{2+},Fe^{3+},Mn,Mg)_3Zr_3(Zr,Nb)_x(Ca,REE)_6Na_{12}[Si_9O_{27-y}$ $OH_y]_2[Si_3O_9]_2Cl_2$ (Giuseppetti, Mazzi, and Tadini, 1971), the structure of which is described by cen-
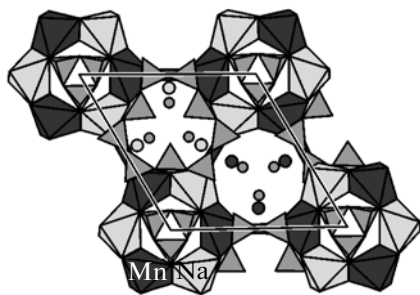
Fig. 5. Ordering of Na and Mn in octahedra of six-membered ring in structure of voronkovite.

trosymmetrical group $R3m$. Additional Zr atoms, along with Nb atoms, are located in the center of nine-membered rings, which make up 0.6 atoms of maximal two atoms per independent part of the unit cell. Making allowance for the predominant vacancy at two intraring sites $M(3)$ and $M(4)$, we placed the mineral near ikranite in the classification table.

12-Layer Oneillite-Type Structures

Most eudialytes contain either six Ca atoms in formula ($Z = 3$), which are necessary and sufficient to build octahedral ring, or slightly lower amount of Ca partly replaced with Mn, Na, and REE. The significant depletion of Ca in some minerals gives rise to the transformation of the six-membered ring. In Ca-poor minerals, $M(1)$ cations are ordered by alternating sites $M(1a)$ and $M(1b)$, so that the homogeneous six-membered ring is broken down into two triplets of octahedra, which are not equivalent in composition and size. As a result, the m plane disappears and symmetry decreases to $R3$.

(1) Variant (T + T) comprises two minerals species, raslakite and voronkovite. The ordered Ca and Fe are established in the $M(1)$ octahedra of the six-membered ring in the structure of *raslakite*, i.e., $Na_{15}Ca_3Fe_3(Na,Zr)_3Zr_3(Si,Nb)(Si_{25}O_{73})(OH,H_2O)_3(Cl, OH)$ (Ekimenkova, Rastsvetaeva, and Chukanov, 2000b). *Voronkovite* with the simplified formula $Na_{15}(Na,Ca,Ce)_3(Mn,Ca)_3Fe_3Zr_3Si_{26}O_{72}(OH,O)_4Cl$ H_2O described by Khomyakov from the Lovozero pluton, Kola Peninsula exhibits the transformation of six-membered rings. In the structure of voronkovite (Rastsvetaeva and Khomyakov, 2000), Ca does not dominate in any octahedra and is replaced by distinct cations, such as Mn and Na (the first and second parentheses of the formula). As a result of the occurrence of two octahedra with different volumes (distances Mn–O and Na–O are 2.22 and 2.45 Å, respectively), three isolated Mn-octahedra are combined with three Na-centered octahedra instead of the ring radical (Fig. 5) and by Na atoms in polyhedra $N(3)$ and $N(4)$ to form a layer. Another feature of voronkovite is the high Si

content with statistically disarranged additional Si tetrahedra.

(2) Variant ($T + M$) is exemplified in *oneillite*, i.e., $Na_{15}Ca_3Mn_3Zr_3Nb(Si_{25}O_{73})(O,OH,H_2O)_3(Cl,OH)_2$ (Johnsen, Grice, and Gault, 1999), from the Saint Hilaire pluton, Quebec, Canada, the structure of which is characterized by two types of disk-shaped platforms, i.e., a platform consisting of ten Si tetrahedra and a platform composed of nine Si tetrahedra and one M octaherdron. In the six-membered ring of oneillite, Ca octahedra (average Ca–O distance 2.43 Å) alternate with Mn-centered octahedra (average Mn–O distance 2.23 Å). The centers of nine-membered rings are occupied by the Nb octahedron and Si tetrahedron, respectively. Atoms of Fe are at the basis of the square in the pentatope.

Structures of Low-Hydrous 24-Layer Minerals

Structures with variable occupancies of Z octahedra by Zr or Ti atoms are distinguished in this group of minerals.

(*1*) The alluaivite structure (variant $Z_{Ti} + Z_{Ti}$). *Alluaivite* $(Ti_{2.3}Nb_{0.7})(Ca_{4.5}Mn_{1.5})(Na_{1.9}1Sr_{0.3}REE_{0.1})$ $[Si_3O_9]_2[Si_{10}O_{28}]_2[Cl_{0.8}(H_2O)_{1.6}K_{0.1}]$ described by Khomyakov from the Lovozero pluton, Kola Peninsula became the first titanium member of the eudialyte group. The absence of Fe, along with an excess of Na in the mineral, makes it possible to accommodate part of the Na in the interstice between two Ca-centered octahedra of two neighboring rings (square and near-square sites). The linear group of three Na-centered polyhedra (two septatopes and square-coordinated site between them), which was described for the first time by Rastsvetaeva et al. (1990), is this mineral's calling card. Another feature of this titanosilicate is the complete occupancy of the centers of nine-membered rings by Si atoms, which have identical orientations along axis *3* in each pair of rings. The presence of 12 additional Si tetrahedra for the entire cell leads to the complete transformation of the nine-membered rings $[Si_9O_{27}]^{-18}$ into ten-membered disk radicals $[Si_{10}O_{28}]^{-16}$.

(*2*) *The dualite structure (variant $Z_{Zr} + Z_{Ti}$).* A single member is *dualite*, i.e., $Na_{30}(Ca,Na,Ce,Sr)_{12}$ $(Na,Mn,Fe,Ti)_6Zr_3Ti_3MnSi_{51}O_{144}(OH,H_2O,Cl)_9$, which was described by Khomyakov from the Lovozero pluton. This Ti-rich zirconium mineral was named after the Latin *dualis*, which emphasizes its major feature, i.e., the Zr- and Ti-dominant octahedra alternate along axis *3* in the doubled cell (Rastsvetaeva, Khomyakov, and Chapuis, 1999) (Fig. 6).

(*3*) *The rastsvetaevite structure (variant $Z_{Zr} + Z_{Zr}$).* *Rastsvetaevite*, i.e., $Na_{27}K_8Ca_{12}Fe_3Zr_6Si_4[Si_3O_9]_4$ $[Si_9O_{27}]_4(O,OH,H_2O)_6Cl$, which was found by Khomyakov at Mt. Rasvumchorr, Khibiny pluton, Kola Peninsula, is characterized by anomalous enrichment in K. The main cause of doubled period c (Rastsvetaeva and Khomyakov, 2001a) is the occur-

rence of alluaivite- and eudialyte-shaped cation layers (modules), which in structure and composition. The main crystal chemical feature of the first layer is the K-dominant sites $N(6)$ and $N(7)$ (67% occupancy of each site) and the complete absence of Fe atoms, the site of which in the center of the oxygen square at the junction of rings [Ca_6O_{24}] is occupied by Na atoms (33% occupancy). All Fe atoms occur in the second layer. In addition to the planar group [FeO_4], the cation layer of this module contains [FeO_5] and [MnO_5] semioctahedra, which are characteristic of the kentbrookite structure and some other members of eudialyte group. The relative amounts of the alluaivite, eudialyte, and kentbrookite structures in rastsvetaevite are about 50, 40, and 10%, respectively. The swelling of the alluaivite layer in the structure of rastsvetaevite is caused by predominant K cations localized in zeolite cavities of this layer and explains the abundant hydrated analogs of rastsvetaevite in the Khibiny pluton and leaching of Na from this mineral when it is treated by water. These features are not characteristic of typical eudialyte.

Labyrynthite, i.e., $(Na,Sr,K)_{35}Ca_{12}Fe_3Zr_6$ [$Si_{52}O_{144}$]$(O,OH,H_2O)_9Cl_3$, which is described by Khomyakov from Mt. N'orkpakhk of the Khibiny pluton is also classified among minerals with rastsvetaevite structures. The mineral is characterized by lowered symmetry to $R3$ (Rastsvetaeva and Khomyakov, 2011b) compared to the space group $R3m$, which is typical of eudialytes; elevated Na; and lower Fe (2.7 wt % compared to a norm of 5–7 wt % FeO). The main feature of alluaivite fragment is general enrichment in Na and the occurrence of three Na-centered polyhedra, i.e., two septatopes and one site with square coordination localized between them. Another attribute of the alluaivite cation layer is the complete occupancy of nine-membered rings with additional Si atoms, which are similarly oriented along axis *3* in the opposite direction with respect to Si tetrahedra of eudialyte module. This is also the cause of the doubled period along this axis. The eudialyte module characterized by the square coordination of Fe is statistically supplemented (~25%) by the kentbrookite module, which contains pentatopes MnO_5; the axial niobium octahedron is replaced by the TiO_6 octahedron.

CONCLUSIONS

The proposed crystal chemical approach to the systematics of the eudialyte-group minerals makes it possible to characterize all minerals of this group in the same terms and to predict the discovery of new minerals and their varieties. This approach is helpful for reconstructing of the conditions of mineral formation, including local conditions, and has practical implications when the ion-exchange properties of the eudialyte-type minerals and their ability to concentrate rare elements are taken into account. The number of localities of the eudialyte-group minerals will undoubtedly
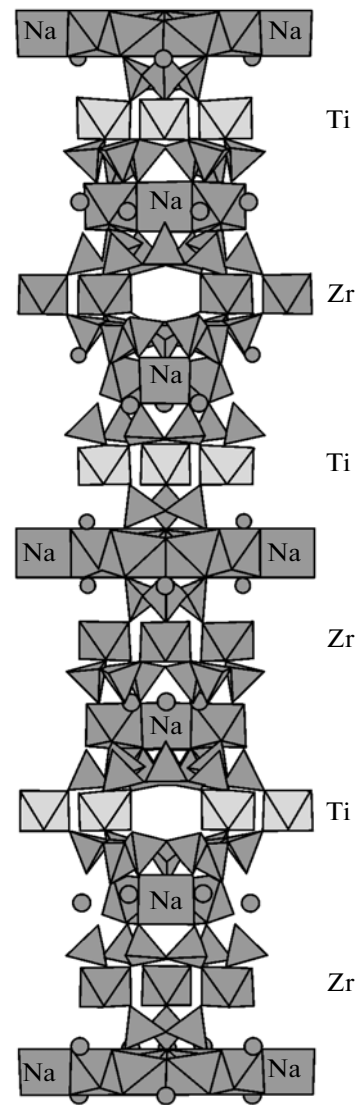

Fig. 6. Structure of dualite.

increase, and their taxonomy will expand and develop further as new facts are acquired. For example, we currently distinguish five variants of occupancy of sites $M(3)$ and $M(4)$ in the 12-layer eudialyte-type structures; so far, the sixth variant ($\square + M$) remains hypothetical. The same can be said of oneillite-type structures, where three hypothetical variants of site occupancy close to the centers of nine-membered tetrahedral rings are known.

Progress in physical analytical methods in recent decades facilitated a rapid increase in the number of mineral species; each year, CNMNC IMA approves more than 50 new mineral species. The introduction of rational principles into the systematics and nomenclature of eudialyte-group minerals could be useful for the development of the taxonomy of other groups of

minerals characterized by the complex mechanism of isomorphic substitutions at several structural sites.

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