

NEW DATA ON ALLANITE FROM VERKHOTURJE (FOR THE 180th ANNIVERSARY OF THE DISCOVERY OF ORTHITE IN RUSSIA)

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The history of the discovery of orthite in Russia (Verkhoturje, Central Ural Mountains) and the results of its investigation by A.Ya. Kupffer, G. Rose, R. Hermann, and N.I. Koksharov are briefly discussed in this article. Orthite was found as crystals in a pegmatite vein in biotite granites (Troitsky Stone). The main minerals are as follows: microcline, quartz, biotite, amphibole; minor and accessory species: allanite, magnetite, schorl, fluorapatite, zircon, epidote, calcite, chalcopryrite. New data on the composition and morphology of crystals are provided. The most developed forms are: (100), (001), (111), and (110); less developed forms are (101), (102), (103), (302), (304), (706), (221), and (112); occasionally — twins on (100) are found. Crystals are zonal and sectorial with respect to average atomic number and in the distribution of Ca, Fe, Al, REE, Th, and Ti. Orthite from Verkhoturje is allanite-(Ce); chemical composition (microprobe analysis, wt.%): SiO₂ 31.76–33.47; TiO₂ 0.10–0.23; ThO₂ 0.26–0.40; UO₂ 0.00–0.04; Al₂O₃ 16.85–20.05; Y₂O₃ 0.01–0.11; La₂O₃ 4.38–5.82; Ce₂O₃ 7.82–10.30; Pr₂O₃ 0.49–1.02; Nd₂O₃ 2.63–3.10; Sm₂O₃ 0.20–0.36; Gd₂O₃ 0.12–0.19; MgO 0.61–0.84; CaO 11.03–14.99; FeO 10.09–12.59; MnO 0.55–0.75; Na₂O 0.00–0.11; F 0.09–0.20; total 92.76–96.64. Comparison with the first analysis of R. Hermann (1848) (with La > Ce) is shown. The investigated mineral is poorly metamict, providing a reason for the underestimated totals of the analyses.

2 tables, 7 figures, 23 references.

Keywords: orthite, allanite, epidote group, pegmatites, Troitsky Stone, A. Kupffer, G. Rose, R. Hermann, N.I. Koksharov, A.E. Fersman.

Allanite a black aluminosilicate of rare earths, iron and calcium, was first described by T. Thompson and named in honor of the Scottish mineralogist Thomas Allan, who found this mineral in East Greenland (Thompson, 1810). The first find was soon followed by others: in 1818 Y. Berzelius found and described a similar mineral from Falun (Sweden) naming it "orthite" for its straight lines, and flat, radiant forms; in 1824 A. Levi discovered a black epidote near Arendal (Norway) and called it "bucklandite" in honor of the English geologist V. Buckland. There were also other numerous synonyms of allanite — orthite, reflecting properties of the mineral or its location (Armbruster *et al.*, 2006).

The discovery and investigation of orthite in Russia is connected with the names of famous Russian and European scientists and researchers — Adolf Kupffer, Gustav Rose, Rudolf Hermann, and Nikolai Koksharov.

A.Ya. Kupffer (Fig. 1), a native of Latvia, was awarded his PhD in Europe, followed by additional training in Paris (Pasetsky, 1984). In 1824 he was invited to join the faculty of Kazan University as a professor of physics, a post he held for four years. In 1828, because of his election as the academician of the Petersburg Academy of

Sciences, he moved to the capital. However, the summer before moving to St.-Petersburg, he undertook a journey across the Urals along the following route: Kazan' — Zlatoust — Yekaterinburg — Nizhnii Tagil — Kushva — Verkhoturje — Yekaterinburg — Kazan', accompanied by his colleague K. Klaus, a pharmacist and artist. Along the way, Kupffer carefully kept a diary in which he fixed the heights of ridges, their geological structure, meteoric conditions, the production and economic activity of factories and mines, and the living conditions and customs of urban and country people (Kupffer, 1833). In Verkhoturje (now a city in the Sverdlovsk oblast', Russia) Kupffer studied rocks on the bank of the Tura river around the so-called Troitsky (Trinity) Stone (Fig. 2), where he found orthite. He described the location as follows (Kupffer, 1833, p. 426): "Granites of Verkhoturje consist almost entirely of a white lamellar feldspar with a small amount of mica, and with still smaller amounts of quartz and garnet. These granites are most accessible on the left bank of the Tura River where they overhang as high cliffs. Rhomboidal prisms, with an angle of about 115°, of a black color, which melt easily in the flame of a blowpipe, and which were recognized by Mr. Berzelius as

orthite, are disseminated in this area through the right bank of the Tura river".

Kupffer provided some crystals to G. Rose, the mineralogy professor of the University of Berlin, for the further investigation of orthite from Verkhoturje. It should be noted that, before the discovery of orthite from Verkhoturje, many scientists (Thompson, Heindinger, Scherer, Breithaupt *et al.*) as well as Rose himself considered that allanite (orthite) belonged to the rhombic system due to the imperfection of the known crystals. Rose had investigated Kupffer's orthite after the completion of a famous expedition, led by Alexander von Humboldt, to the Urals and Siberia, in which he also participated (Rose, 1837). In one chapter, describing travel across the Central Ural Mountains, Rose noted a goniometrical investigation of orthite from Verkhoturje, and concluded that, as far as could be determined from, the angles of orthite that he could measure, the results were similar to those of epidote, and therefore, orthite is monoclinic. The forms found by G. Rose are *T*, *M* and *r* in the designations of Häuy for epidote (Rose, 1837), corresponding to *c* (001), *a* (100) and *r* (-101) today. Thus, Rose corrected an error of his predecessors. However, Rose did not manage to draw a full crystal of orthite: "*simple and twinned crystals occur, but, as far I can see, they are broken off at the ends*" (Rose, 1837; Koksharov, 1858). Without knowing the chemical composition of orthite, and solely on the basis of morphological features, Rose called orthite from Verkhoturje "black epidote", or bucklandite.

The chemistry of "black epidote" from Verkhoturje was established by Hans Rudolf Hermann (Fig. 3). In 1827 he left Dresden (Germany) for Moscow in order to organize a plant for production of artificial mineral waters. Subsequently, Moscow became a second home for Hermann (Trautshold, 1881; Liebman, 1956). He had



Fig. 1. Adolf Yakovlevich Kupffer (1799–1865) – the physicist, meteorologist, crystallographer, mineralogist, traveler. One of the founders of the meteorological and mineralogical services of Russia. Member of more than 20 Russian and foreign academies and societies (Pasetsky, 1984).

obtained no higher education, but, as a result of his intelligence and his enthusiasm for science, Hermann achieved amazing results. His skill in analytical chemistry of the REE, Zr, Th, U, Ta and Nb minerals is especially great. The father of modern crystallography, E.S. Fedorov, called Hermann one of founders of mineralogical science in Russia along with N.I. Koksharov and G. Rose.

In 1841 Hermann had received a sample of "chevkinite" from the Ilmen mountains for investigation. Following thorough evaluation, Hermann clearly proved its difference from chevkinite. He recognized the mineral as orthite and described it under the name uralorthite (Hermann, 1841). Thus, the Ilmen Mountains became the second location for orthite in Russia after Verkhoturje, and Hermann's chemical analysis was likely the first published for Russian orthite. Uralorthite probably originated in the No 22 pit according to M.P. Melnikov's numbering system (Popova *et al.*, 1980).

The find of "uralorthite" motivated Hermann to perform chemical analyses of other allanites,



Fig. 2. Verkhoturje. A view on the Holy Trinity Cathedral and the Troitsk Stone on the left bank of the Tura river – the locality of the initial find of orthite in Russia. Photo: S.A. Gavrilov, 2011.



Fig. 3. Rudolf Hermann (1805–1879) – scientist, mineralogist, one of the most authoritative Russian chemists-analysts of the middle of the 19th century, medicine doctor, a corresponding member of the Russian Academy of Sciences, the author of more than 180 scientific publications in Russian and German journals and transactions (Liebman, 1956).

Table 1. Composition of allanites from Verkhoturje (wt.%)

Compo- nents	Hermann, 1848*	Our data			
		1	2	3	4
SiO ₂	32.46	33.47	31.76	32.10	31.95
TiO ₂		0.10	0.16	0.19	0.23
ThO ₂		0.28	0.40	0.26	0.37
UO ₂		0.02	0.00	0.04	0.03
Al ₂ O ₃	18.09	20.05	17.10	17.46	16.85
Y ₂ O ₃	1.5	0.01	0.11	0.07	0.04
La ₂ O ₃	9.76	4.38	5.56	5.82	5.33
Ce ₂ O ₃	6.77	7.82	10.42	10.26	10.30
Pr ₂ O ₃		0.49	0.77	1.02	0.62
Nd ₂ O ₃		2.63	2.95	3.10	3.06
Sm ₂ O ₃		0.36	0.27	0.32	0.20
Gd ₂ O ₃		0.12	0.19	0.14	0.16
MgO	1.02	0.68	0.61	0.84	0.76
CaO	13.18	14.99	11.04	11.43	11.03
FeO	13.84**	10.09	11.32	12.59	11.06
MnO	traces	0.55	0.59	0.75	0.57
Na ₂ O		0.00	0.10	0.06	0.11
H ₂ O	3.40				
F		0.13	0.20	0.19	0.09
-O = F ₂		-0.05	-0.08	-0.08	-0.04
Total	100.02	96.17	93.55	96.64	92.76

Notes: * – CuO and Mn₂O₃ traces occur; ** – calc. on $\Sigma(\text{FeO} + \text{Fe}_2\text{O}_3)$.

An empty space means that component was not determined. 0.00 – component amounts are below detection limits. Analysis numbers correspond to those of the BSE image (Fig. 7); CAMECA SX 100 microprobe analyzer. analyst V.V. Hiller.

Empirical formula of allanite (calc. on cation sum = 8 apfu, when Si > 3.05 formula normalized for Si = 3 apfu;

Armbruster et al., 2006): Hermann – $(\text{Al}_{1.96}\text{Fe}_{1.06}\text{Mg}_{0.14}\text{La}_{0.11}\text{Ce}_{0.26}\text{Ti}_{0.01}\text{Si}_2\text{O}_7)\text{O}_{0.33}(\text{OH})_{2.06}$;

1 – $(\text{Ca}_{1.44}\text{Ce}_{0.26}\text{La}_{0.15}\text{Nd}_{0.08}\text{Pr}_{0.02}\text{Sm}_{0.01}\text{Th}_{0.01})_{1.97}(\text{Al}_{2.12}\text{Fe}_{0.76}\text{Mg}_{0.09}\text{Mn}_{0.04}\text{Ti}_{0.01})_{3.02}(\text{SiO}_4)(\text{Si}_2\text{O}_7)(\text{O}_{0.96}\text{F}_{0.04})\text{OH}_{0.70}$;

2 – $(\text{Ca}_{1.12}\text{Ce}_{0.37}\text{La}_{0.20}\text{Nd}_{0.10}\text{Pr}_{0.03}\text{Sm}_{0.01}\text{Th}_{0.01}\text{Gd}_{0.01}\text{Y}_{0.01}\text{Na}_{0.02})_{1.88}(\text{Al}_{1.90}\text{Fe}_{0.89}\text{Mg}_{0.09}\text{Mn}_{0.05}\text{Ti}_{0.01})_{2.94}(\text{SiO}_4)(\text{Si}_2\text{O}_7)(\text{O}_{0.94}\text{F}_{0.06})(\text{OH})_{0.35}$;

3 – $(\text{Ca}_{1.22}\text{Ce}_{0.35}\text{La}_{0.20}\text{Nd}_{0.10}\text{Pr}_{0.03}\text{Sm}_{0.01}\text{Th}_{0.01}\text{Na}_{0.01})_{1.93}(\text{Al}_{1.92}\text{Fe}_{0.96}\text{Mg}_{0.12}\text{Mn}_{0.05}\text{Ti}_{0.02})_{3.01}(\text{SiO}_4)(\text{Si}_2\text{O}_7)(\text{O}_{0.94}\text{F}_{0.06})(\text{OH})_{0.72}$;

4 – $(\text{Ca}_{1.15}\text{Ce}_{0.35}\text{La}_{0.15}\text{Nd}_{0.11}\text{Pr}_{0.02}\text{Sm}_{0.01}\text{Th}_{0.01}\text{Gd}_{0.01}\text{Na}_{0.02})_{1.87}(\text{Al}_{1.86}\text{Fe}_{0.87}\text{Mg}_{0.11}\text{Mn}_{0.05}\text{Ti}_{0.03})_{2.94}(\text{SiO}_4)(\text{Si}_2\text{O}_7)(\text{O}_{0.97}\text{F}_{0.03})(\text{OH})_{0.22}$

orthites, and "black epidotes" from Northern Europe and Verkhoturje (Hermann, 1848). There are now multiple accurate chemical composition data for orthite from Verkhoturje to compare with Hermann's (Table 1). Based on the analyses of the studied minerals and the goniometrical measurements of orthite crystals from Verkhoturje and "uralorthite" from the Ilmen mountains, performed by Rose, Hermann concluded that all the studied minerals are orthites, and members of the epidote group (Hermann, 1848). Somewhat later, N.I. Koksharov independently arrived at the same conclusion (Koksharov, 1848). Historically, the names "allanite" and "orthite" have been used to designate the tabular crystal and the prismatic metamict water-containing varieties, respectively. In 1986 the CNMMN IMA retained only the root name "allanite", and Thompson's mineral was called allanite-(Ce) (Nickel, Mandarino, 1987). Mineral species status was also assigned to allanite-(Y) and allanite-(La) (Armbruster et al., 2006). Thus, in compliance with the requirements of modern nomenclature, orthite from Verkhoturje is allanite.

During the preparation of this paper, we were unable to find any publications on the morphology of allanite from Verkhoturje. A visit by one of the authors (VAG) in 2013 to Verkhoturje did not solve the problem either, since allanite from the Troitsk Stone pegmatite veins was quite suitable for chemical analyses, but not for crystallographic measurements (Fig. 4).

Crystals of allanite from this locality, however, were found in collection of Alexander Evgenyevich Fersman. A.E. Fersman visited Verkhoturje twice as part of the Radium expedition founded by academician V.I. Vernadsky in 1911 for the study of the radioactive minerals and ores of Russia. In 1912 he visited the locality with V.I. Vernadsky, and in 1913 with the chemist-mineralogist B.A. Lindener. During the second trip, material with well-shaped allanite crystals was found, and the distribution of veins containing the mineral was also established. Fersman describes those places as follows: "In the Troitsk Stone the leucocratic veins with orthite occur, yet they are limited there, so it is not possible to extract any good material from them. But in a stone quarry in the countryside behind the Convent we succeeded in collecting excellent material. In a northwest angle of the quarry an especially rich pegmatite vein was discovered, which provided not only several chunks of rock containing this mineral, but also many regular crystals, with numerous faces and well-formed terminations" (Fersman, 1914, p. 159).



Fig. 4. Allanite crystal (1.8 cm) in a quartz-feldspar pegmatite. Verkhoturje, Troitsky (Trinity) Stone. Photo: V.A. Gubin.

Fig. 5. Allanite, a twin on {100} (1cm). Verkhoturje, a pit over the Convent. FMM, systematic collection, № 21851, collected by A.E. Fersman and B.A. Lindener, 1913. Photo: M.M. Moiseev.

Pegmatite veins containing allanite near Verkhoturje occur in a base biotite granite of the Hercynian age, without any stratification. Fersman considered these pegmatites the earliest, deep type I (allanite pegmatites subtype), corresponding to B- and C-geophases (Fersman, 1940). Pegmatites of the Troitsk Stone are mainly comprised of plagioclase; microcline, quartz, biotite and hornblende occur in lesser amounts. The list of pegmatite accessory minerals is also modest, and includes allanite, magnetite, chalcocopyrite, and schorl. Additionally we have collected samples of fluorapatite (with F = 3.43 wt.%, REE = 0.35 wt.%), zircon, epidote and calcite.

After his travels Alexander Evgenyevich donated the minerals he collected to a number of Russian museums and universities, but mainly to the Mineralogical Museum of the Russian Academy of Sciences. Mineralogical collections contributed by A.E. Fersman were organized systematically recently by V.D. Dusmatov based on locations and by numbers (Dusmatov, 2003). Among them, allanite-(Ce) samples from Verkhoturje also occur, collected by A.E. Fersman and B.A. Lindener in 1913 in the above mentioned stone quarry (№№ 702 – 707 from the deposits col-

lection and №№ 21846 – 21856 from the systematic collection).

Examination of the collection shows that allanite-(Ce) crystals (both single and twins) are fresh, black, and without character "coffee color" surface of change. Fragments of crystal faces are flat, but with little luster (Fig. 5); some crystals have well shaped terminations. Crystals cataloged in the systematic collection at № 21851 are of especially high quality. The size of separate crystals reaches 2.5 x 1 cm, although the average does not exceed 1 cm. Goniometrical measurements with a Fedorov stage, mounted under binocular microscope were taken (Fig. 6).

Except for flat faces, a considerable part of the surface of the crystals is covered with simultaneous growth induction surfaces of allanite with quartz, biotite and feldspar.

X-ray investigation shows this material is poorly metamict. The powder data of one of the crystal fragments (FMM № 21851) are shown in Table 2. In contrast to typical non-metamict allanite-(Ce) (Cech *et al.*, 1972), the powder data of allanite from Verkhoturje is characterized by a small set of lines, which are partially widened and blurred.

The chemical composition of allanite samples, collected in Verkhoturje by the authors, was

Fig. 6. Crystals and twins of allanite-(Ce) from Verkhoturje (FMM systematic collection, № 21851).

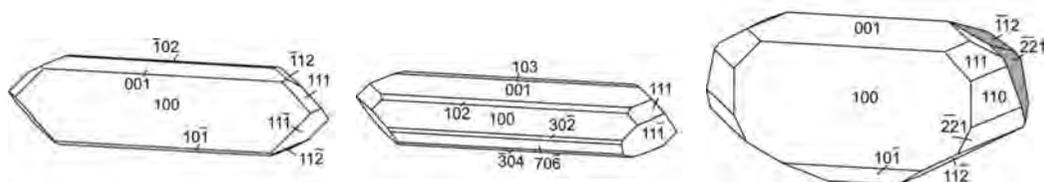


Table 2. X-Ray powder data for allanite-(Ce) from Verkhoturje, Mid. Ural, and Zambia

Verkhoturje (our data)		Luangwe, Zambia (Cech <i>et al.</i> , 1972)*		
<i>I</i>	<i>d/n</i> , Å	<i>I</i>	<i>d/n</i> , Å	<i>hkl</i>
		8	9.23	001
		11	8.11	100
		17	7.96	-101
		13	5.12	101
		10	5.02	-102
		5	4.89	011
10w	4.72	15	4.70	-110
		10	4.61	002
		1	4.06	200
		3	3.98	-202
		4	3.83	111
		8	3.79	-112
		9	3.60	012
90	3.523	45	3.53	-211
30	3.355	7	3.32	-210
		6	3.28	-212
		13	3.25	201
		2	3.17	-203
100	2.921	100	2.920	-113
30w	2.845	30	2.886	020
		16	2.829	211
		5	2.753	021
40w	2.707	65	2.714	013
		5	2.658	-303
50	2.619	40	2.627	-311
		20	2.561	202
		10	2.506	-204
10	2.462	8	2.447	022
10	2.415	14	2.413	-313
		13	2.337	-222
		8	2.301	-114
		5	2.291	-304
		2	2.233	-402
		7	2.210	122
		12	2.193	-123
		35	2.182	-401
20w	2.141	25	2.158	-403
		13	2.141	014
		19	2.135	-223
20w	2.117	11	2.105	023
		5	2.070	203
		3	2.056	322
		3	2.030	-205

Notes: w – wide line; operating conditions (allanite from Verkhoturje): photomethod, Guinier camera Huber 621 with quartz monochromator; CuK α , radiation; quartz used as internal standard, analyst V.Yu. Karpenko; * – data for *d/n* < 2.00 are omitted.

determined at the Institute of Geology and Geochemistry (Yekaterinburg) using a CAMECA SX 100 microprobe analyzer equipped with a wave dispersive spectrometer at 15 kV accelerating voltage, 100 nA probe current (Fig. 7, Table 2, analyst V.V. Hiller). The standards used were pure synthetic phosphates (*REE*)PO₄ (Y, La, Ce, Pr, Nd, Sm, Gd), Fe₂O₃ (Fe), TiO₂ (Ti), ThO₂ (Th), UO₂ (U), diopside (Si, Mg, Ca), rhodonite (Mn), jadeite (Na, Al), fluor-phlogopite (F on LPCO crystal analyzer). Maps were acquired for 260 minutes (each map) with an accelerating voltage of 15 kV and a beam current of 100 nA; scanning time for each map point was 100 ms. We analysed 15 points from which 4 typical points were chosen from different growth zones. The material we analyzed has selective-cerium composition of lanthanides with actual dominance of cerium. Thus, the mineral corresponds to allanite-(Ce). We note a rather large amount of magnesium (MgO ~ 0.9 wt.%), which can be explained by the increased Mg-content of the host granites. Weak metamictization of the mineral fixed by X-ray is probably related to the presence of trace amounts of Th. In general, modern microprobe analyses are close to that of R. Hermann (Table 1). However, in Hermann's analysis La prevails over Ce, and, according to the modern nomenclature of epidote-group minerals (Nickel, Mandarino, 1987; Armbruster *et al.*, 2006), it would be necessary to call this mineral allanite-(La). However, it may be premature to apply this label due to imperfections in the old analysis technique, as only one random section was investigated. In addition, at the time of Hermann's analysis (1848) some of the rare earth elements were not known yet, and some mixed data could lead to the corresponding result.

A wide variety of isomorphous substitutions are typical for allanites, as described by many authors (Hvostova, 1962; Kepizhinskas, Hlyostov, 1971; Gieré, Sorensen, 2004; Armbruster *et al.*, 2006; etc.). Along with the widespread scheme of heterovalent isomorphism Ca²⁺ + Al³⁺ ↔ REE³⁺ + Fe²⁺ it is necessary to specify others, as well. Some Mn, Mg and F indicates the presence of dolasseite-(Ce) and khristovite-(Ce) components and the implementation of the mechanisms Ca²⁺ + Al³⁺ ↔ REE³⁺ + (Mn, Mg)²⁺ and Al³⁺ + O²⁻ ↔ (Mn, Mg)²⁺ + F⁻ (Pecacor, Dunn, 1988; Pautov *et al.*, 1993). Th and U can enter according to the scheme (Th, U)⁴⁺ + 2M²⁺ ↔ Ca²⁺ + 2REE³⁺ (where M = Fe²⁺, Mn²⁺, Mg²⁺) (Gieré, Sorensen, 2004).

The complex growing zonality and sectoriality of an allanite crystal section observed in BSE mode does not always correlate with the

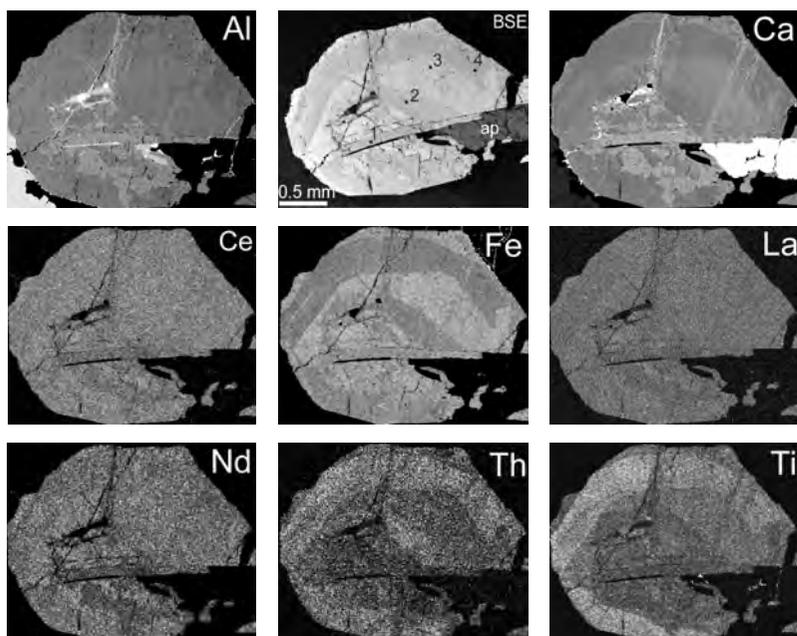


Fig. 7. Cross section of the allanite-(Ce) crystal. Back scattered electrons (BSE) and AlK α , FeK α , CaK α , TiK α , CeL α , LaL α , NdL α , ThM α X-ray radiation images. Analyst V.V. Hiller.

variability of amounts of a specific element, and most likely reflects the summary result of several schemes of isomorphous substitution (Fig. 7). In the central (lower) part of a random crystal section there are areas with an excessive amount of Ca and Al, which are close to epidote (Table 1, analysis 1). Spottiness possibly is connected with the crystal block growth there. Theoretically it is possible to find a rare-earth epidote in a full anatomic section along a crystal. Titanium and thorium both give a similar distribution picture, but their sectoriality is obviously different (Fig. 7). There is no expressed zonality for rare earth distribution in the studied sample, yet sectoriality is noticeable, especially for cerium and lanthanum. A lack of totals for analyses 2 and 4 most likely is connected with metamictization of the corresponding zones, probably caused by their enrichment with thorium; this is confirmed by the corresponding illustration (Fig. 7). A similar example can be seen for allanite-(Ce) from quartz veins of the Aldridge Formation, British Columbia, Canada (Campbell, Ethier, 1984).

The described heterogeneity of the studied crystal composition allows one to assume variations of composition of allanites both in the whole granite massif, and in the pegmatite veins.

The wide spread occurrence of allanite in the Verkhoturjsky region is has long been well known. In such a large mineralogical occurrence, considerable variations of forms and

composition of minerals in different rock phases and facies would be expected. This is confirmed by our additional research.

Verkhoturje is a region in which it is possible to study allanite typomorphism on the basis of mineralogical mapping. Allanites we have inspected from different collections, judging by induction surfaces, grew synchronously with surrounding magmatic minerals. Therefore, their typomorphism can be related to that of other minerals of the corresponding paragenesis.

The authors thank V.N. Onosova, T.B. Kochergina and Z.A. Spiridonova, library employees of Yekaterinburg, for their help in selection of historical literature, S.A. Gavrillov for a fine panoramic exposure of the Troitsk Stone, M.M. Moiseev for the twinned crystal photo, E.M. Spiridonov and L.A. Pautov for valuable advice and also Dr. M. Feinglos for English text editing.

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