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Allanite-(Y) in Areas of Ongonite Magmatism in the Far East: Isomorphism and Petrogenetic Implications

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Abstract—The research history of allanite-(Y) has been considered. The composition and genesis of this mineral is discussed with allowance for new findings in Li–F granites of the Russian Far East. Compositional anomalies in allanite-(Y) reflect the metastable character of its structure. Types of zoning and trends in the chemical substitution of major components are described. Two crystal chemical modes of allanite-(Y) formation and the two-step isomorphism of its components are suggested. The main tendencies in the chemical evolution of allanite-(Ce) and allanite-(Y) in granitic rocks of the Far East are pointed out. Allanite-(Y) is formed in the anomalous geochemical setting that characterizes crystallization of fluid-saturated subalkaline granitic melt and is regarded as an index mineral of rare-metal ongonite magmatism completing the Late Cretaceous Pacific Orogeny.

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INTRODUCTION

Allanite was first described by T. Thomson in 1810 and named in honor of Thomas Allan (1733–1833), the Scottish mineralogist who discovered this mineral in eastern Greenland. Numerous synonyms, which were proposed as early as the 19th century in the process of allanite research (cerine, bucklandite, uralorthite, tautolite, pyrorthite, cerepidote, wasite), reflect allanite's extreme variability related to its metamict alteration and its isomorphism at four sites at once: $(Ca,Mn)(Y,REE)(Al,Fe^{3+},Cr,V,Mn^{3+})Al(Fe^{2+},Mg,Mn_2)[Si_2O_7[SiO_4]O(OH)$ (Semenov, 1958, 2001; Ercit, 2002; Gieré, 2004; Armbuster, 2006). For a long time, the names *allanite* and *orthite* denoted tabular crystalline and prismatic metamict hydrous varieties of the mineral, respectively. This division is now recognized as archaic, because metamictization, hydration, and the optic isotropy used for their identification are not strictly interrelated (Deer et al., 1962). In 1986, IMACNMNC made the choice in favor of *allanite* and Thomson's mineral was called allanite-(Ce) (Nickel and Mandarino, 1987). In 2006, allanite-(La) received the status of mineral species (Orlandi and Pasero, 2006).

RESEARCH HISTORY OF ALLANITE-(Y)

Allanite is distinguished by a wide isomorphism of REE. While REE patterns have a cerium maximum in the overwhelming majortity of cases, varieties sharply enriched in Y and HREE were periodically found over the two-hundred-year history of the research. It is noteworthy that name *orthite* was proposed in 1818 by J.J. Berzelius just for Y-allanite from Falun, Sweden. One of the first mentions of that finding belongs to Hermann (1848). He pointed out the chemical analysis of the sample from the famous Ytterby locality in Sweden, noted a high Y content in the orthite found together with gadolinite, and inferred replacement of La and Ce with Y. These data had been confirmed by Chernik (1908). He analyzed yttroorthite from Sweden, summarized the analytical data on orthite obtained in the 19th century, and called attention to other findings of anomalous high-Y orthites: Berlin's orthite in the Ytterby, Engström's wasite, Berzelius' pyrorthite found in Sweden, Kerntdt's bodenite and muromontite in Germany, and Mallet's orthite in the United States (Table 1, an.1). No information on the geological position and origin of these orthites is contained in publications by Hermann and Chernik; only their association with gadolinite from pegmatite and anomalous high water contents has been noted.

In light of recent tendencies in the procedure for recognizing new minerals, R. Hermann should be named the discoverer of allanite-(Y) as a compound, where one structural site is preferably occupied by a special chemical element. At the same time, Hermann (1848) called this mineral common orthite (gemeiner Orthit, in German) and did not separate it from Ce-bearing varieties. It should be remembered that the study of yttroorthite from Ytterby was conducted practically in parallel with the discovery and research of rare-earth elements, only 20 years after the discovery of isomorphism by E. Mitscherlich. Max von Laue still has not been awarded the Nobel Prize for his discovery of X-ray diffraction, and the number of mineral species scarcely exceeded 300. Therefore, Hermann and his colleagues—J.J. Berzelius, N. Engström,

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Table 1. Chemical composition of allanite-(Y), wt %

Component	Published data[1)]				Allanite from Far East[2)]					
	1	2	3	4	5	6	7	8	9	10
SiO_2	36.24	31.09	29.23	34.68	38.29	36.39	41.37	40.56	32.07	33.85
Al_2O_3	8.18	2.24	12.80	23.20	–	7.41	11.70	–	–	1.41
Fe_2O_3	–	–	2.60	2.54						
FeO[3)]	10.07	11.23	1.23	8.26	10.67	2.27	7.29	10.52	17.81	12.23
MgO	0.61	0.42	0.10	–	–	–	–	–	–	–
MnO	–	0.90	1.20	0.76	–	1.76	–	–	–	0.26
CaO	5.48	0.71	15.94	15.36	0.90	14.73	–	17.84	–	3.09
La_2O_3	–	3.54	3.24	–	–	–	–	–	–	0.07
Ce_2O_3	4.98	5.54	6.60	2.12	3.83	0.50	–	–	2.92	3.07
Nd_2O_3	–	–	–	–	2.92	–	–	–	5.60	3.96
Sm_2O_3	–	–	–	–	–	–	–	–	2.64	1.73
Gd_2O_3	–	–	–	–	2.84	1.34	–	–	3.97	3.65
Dy_2O_3	–	–	–	–	4.29	2.82	–	–	4.56	4.32
Er_2O_3	–	–	–	–	3.77	2.56	–	–	–	2.94
Yb_2O_3	–	–	–	–	3.79	2.13	–	–	–	2.14
Y_2O_3	29.81	37.14	13.30	8.70	29.75	28.09	31.15	31.08	29.30	27.19
ThO_2	–	–	–	0.22	–	–	8.53	–	1.14	0.30
H_2O	4.59	0.85	2.94	3.02						
Total[4)]	100.57	100.00	99.39	99.64	100.00	100.00	100.00	100.00	100.00	100.00
Formula coefficient[5)]										
Si	3.52	3.46	3.06	3.00	3.96	3.45	3.74	3.79	3.54	3.62
Al	0.94	0.29	1.58	2.32		0.83	1.25			0.18
Fe^{3+}	0.00	0.00	0.20	0.16	0.00	0.00	0.00	0.00	0.00	0.00
Fe^{2+}	0.82	1.05	0.11	0.59	0.92	0.18	1.22	0.82	1.65	1.09
Mg	0.09	0.07	0.02							
Mn^{2+}		0.08	0.11	0.06		0.14				0.02
ΣM	1.76	1.49	2.02	3.13	0.92	1.15	2.47	0.82	1.65	1.29
Ca	0.57	0.08	1.79	1.41	0.10	1.50		1.79		0.35
La		0.15	0.13							
Ce	0.18	0.23	0.25	0.07	0.15	0.02			0.12	0.12
Nd					0.11				0.22	0.15
Sm									0.10	0.06
Gd					0.10	0.04			0.15	0.13
Dy					0.14	0.09			0.16	0.15
Er					0.12	0.08				0.10
Yb					0.12	0.06				0.07
Y	1.54	2.20	0.74	0.40	1.64	1.42	1.50	1.54	1.72	1.55
Th				0.00			0.18		0.03	0.01
ΣA	2.29	2.66	2.91	1.88	2.48	3.21	1.68	3.33	2.50	2.69

[1)] (1) orthite from Ytterby, Sweden ($Ce_2O_3 = \Sigma Ce_2O_3 + La_2O_3$) (Hermann, 1848); (2) muromontite from Saxony, Germany ($La_2O_3 = \Sigma La_2O_3 + Nd_2O_3 + Pr_2O_3$, $Y_2O_3 = \Sigma Y_2O_3 + Er_2O_3$) (Chernik, 1908); (3) orthite from Falun, Sweden ($Y_2O_3 = \Sigma Y_2O_3 + Er_2O_3$, $Nd_2O_3 = \Sigma Nd_2O_3 + Pr_2O_3$) (Chernik, 1908); (4) yttroorthite from Sinyaya Pala vein, northern Karelia (Labuntsov, 1939); [2)] (5, 6) microcline–albite leucogranite with zinnwaldite, Badzhal district, Amur region; (7) microcline–albite leucogranite with zinnwaldite, Chaun district, Chukchi Peninsula; (8) granosyenite porphyry, Ichuveem dike swarm, Chukchi Peninsula; (9) greisenized biotite granite, Verkhneurmiyskiy pluton, Amur region; (10) average composition of allanite-(Y) from Far East (86 samples); dash and gap denote not detected and not determined, respectively; [3)] analyses 5–10: $FeO = \Sigma FeO + Fe_2O_3$; [4)] total also includes: (1) $\Sigma K_2O + Na_2O = 0.62$; (2) $K_2O = 0.17$, $Na_2O = 0.65$, $BeO = 5.52$; (3) $Na_2O = 0.08$, $K_2O = 0.12$; (4) $TiO_2 = 0.17$, $CO_2 = 0.61$; [5)] calculated on the basis of 8 cations with subsequent equilibration of Fe^{3+}/Fe^{2+} ratio on the basis of 12.5 oxygen ions (Ercit, 2002).

A.E. Kupfer, and N.I. Koksharov—distinguished only regional varieties of the mineral (uralorthite, muromontite, xanthorthite, bodenite, etc.) (Table 1, an. 1–3) and did not have an opportunity to estimate a degree of difference in the constitution of REE-epidote, Ce- and Y-orthites. V.I. Vernadsky was the first to make such a comparison and noted that *orthite* from pegmatite, which is enriched with the yttrium group of rare earth elements, "cannot be called orthite and requires a new name" (Vernadsky, 1929, p. 36).

The systematic study of Y-allanite was carried out by Labuntsov (1939), who was the first to find this mineral in Russia (in the Sinyaya Pala pegmatite vein in Karelia) and to describe it as a metamict mineral, which occurs as crystals up to 15 cm long and 5 cm thick incorporated into oligoclase. He determined the chemical composition and morphology of this mineral and defined it as a rare variety of yttroorthite. The Karelian allanite is anomalously rich in Al and Ca and relatively depleted in Y (Table 1, an. 4). Based on the association of yttroorthite with uraninite and on the occurrence of cyrtolite inclusions, Labuntsov considered yttroorthite to be the latest mineral formed at the pneumatolytic stage *D* (~600°C). Following Hermann, he called metamict varieties orthite and separated them from allanite as a mineral of crystalline appearance.

Lombaardite, which had been described by Nel and Strauss (1949) in quartz veins of Transvaal, was later discredited and identified as allanite with high Y (5.49 wt %) and Nd (5.81 wt %) contents (Neumann and Nilsen, 1962). Using XRF, Frondel (1964) managed to reveal allanite with elevated Y_2O_3 contents (7.5 and 7.7 wt %) in granitic pegmatites of Ontario Province, Canada. It was concluded that allanite with >5 wt % Y_2O_3 is a special variety of the mineral; an XRD difference between allanite-(Y) and allanite-(Ce) was established.

Semenov (1958, 2001) mentioned yttroorthite and regarded orthite as a complex rare earth mineral with a wide isomorphism and the ability to change a maximum of REE. Khvostova (1962) presented a review concerning the classification, composition, isomorphism, and abundance of orthite using the data on 190 samples from many localities around the world. A wide occurrence of Ce-orthite in granites, granitic pegmatites, and alkaline rocks was noted in contrast to rare yttroorthite and orthites with intermediate compositions.

Levinson (1966), citing Neumann and Nilsen (1962), considered allanite-(Y) a special mineral species. In recent summaries, despite insufficient data on unit-cell parameters of allanite-(Y), this mineral is unequivocally regarded as a mineral species (Semenov, 2001, Ercit, 2002; Armbruster et al., 2006; Krivovichev, 2008). To date, allanite-(Y) has been found in addition to the aforementioned localities in Italy (Lombardy, Piemont), Sweden (Värmland), Scotland (Skye Island), Japan (Fukushima), Norway (Nordland, Telemark), Spain (Cadalso de los Vidrios), and the United States (Arizona, Colorado, New York).

Thus, a special allanite species that differs from other minerals of this subgroup in extremely high Y content (up to 29.81 wt %) actually exists. It occurs extremely rarely and only in granitic pegmatites. Although allanite-(Y) has been declared to be a mineral species, its structure remains poorly studied and its origin and relationship to allanite-(Ce) is still ambiguous in many respects. Studies over the years have shown that the anomalous composition of this mineral is expressed in disturbed stoichiometric proportions of major components (Table 1, an. 1–4).[2] Information obtained by means of studying rare-metal granitoids in the Russian Far East clarifies some of these problems.

ALLANITE IN GRANITOIDS OF THE FAR EAST

It is known that allanite is not characteristic of the granitoid series in tin provinces (Marin, 2004). At the same time, allanite-(Ce) is a common accessory mineral in granites of the Pacific tin belt (Zagruzina, 1965; Isokh et al., 1967; Lyakhovich, 1979). Allanite-(Y) was identified as a subordinate accessory mineral in zinnwaldite granites of the Li–F geochemical type (Table 1, an. 5–7), which complete the Late Cretaceous granitic series of the Far East and control rare-metal mineralization at the largest deposits of the Amur region and Chukchi Peninsula, e.g., the Pravourmiyskoye (Brusnitsyn et al., 1993) and the Pyrkakai (Zagruzina, 1965; Alekseev, 2011) deposits. Allanite-(Y) was also found in the biotite granites, which make up batholiths of the Miao–Chang series in the Amur region (Izoch et al., 1967) and have been altered under the effect of younger Li–F garnites (Table 1, an. 9). Allanite-(Y) with anomalously high Ca content was established in association with allanite-(Ce) in low-silicic granosyenite, monzogranite, and granite porphyries of the dike fields in the Amur region and Chukchi Peninsula (Table 1, an. 8).

Allanite-(Y) from Li–F granite and granosyenite porphyry is dark red and translucent at the margins, with vitreous luster and uneven fracture. The mineral grains are xenomorphic in outlines, lenticular and duck-footed. They form peculiar chains and net venation along prototectonic and cleavage fractures in microcline and at boundaries of quartz and feldspar grains. Monocrystals are 10–50 μm in size; the reticulate segregations reach 150 μm in dimensions. Allanite is distinguished by extreme inhomogeneity and char-

[2] The related problems in formula calculation were discussed by Ercit (2002). A conclusion was drawn regarding the advantages of recalculation based on eight cations with the subsequent selection of the ratio Fe^{2+}/Fe^{3+} and equilibration of the composition for 12.5 O ions.

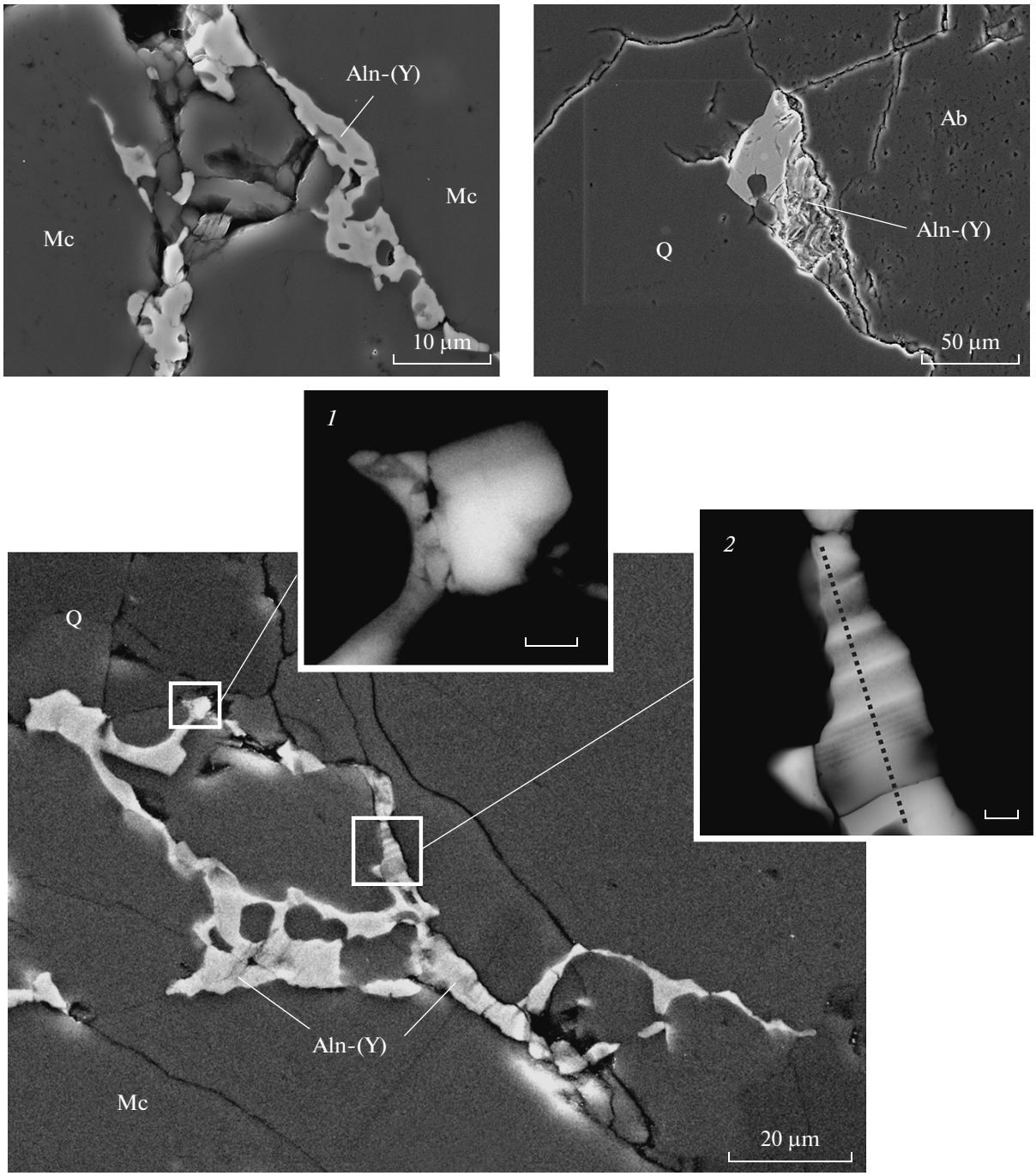

Fig. 1. Morphology and microheterogeneity of allanite-(Y) in zinnwaldite granite. Images in secondary electrons and BSE images in insets *1* and *2* (JSM-7001F, JSM-6460LV). Scale bars in insets corresponds to 1 μm. Dashed line of analytical profile (see Fig. 5) is shown in inset *2*. Aln-(Y), allanite-(Y); Mc, microcline; Ab, albite; Q, quartz.

acterized by fine oscillatory zoning. The finest brecciation is occasionally observed at the contact of grains with the adjacent matrix (Fig. 1). Allanite less frequently occurs as xenomorphic inclusions in accessory Y-bearing fluorite and tveitite-(Y) (Fig. 2). Another variety of allanite-(Y) has been identified in slightly greisenized biotite granite that hosts Li–F granite and probably is a pseudomorph after allanite-(Ce), a common accessory of the Far East granites. These pseudomorphs are 30–300 μm in size and vary in shape from irregular grains to euhedral and tabular crystals. A peculiar worm-shaped zoning is observed at the margins of large crystals (Fig. 3). Under optic microscope, allanite is nonuniformly reddish yellow, slightly pleochroic, and optically anisotropic. In physical properties, allanite-(Y) substantially differs from the allanite-

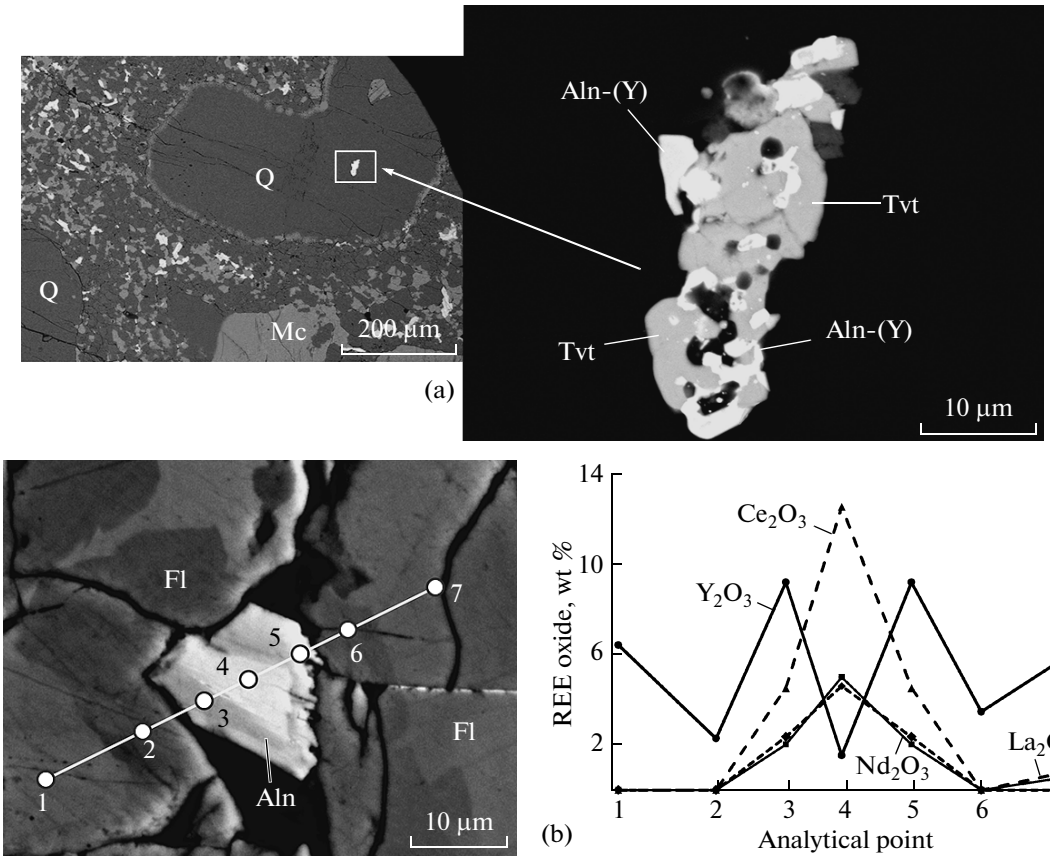

Fig. 2. Inclusions of allanite-(Y) in accessory fluorides. (a) quartz phenocryst with inclusion in rare-metal granite porphyry, on the left; inclusion of tveitite-(Y) (gray) and allanite-(Y) (white), on the right; (b) roughly zoned allanite with "cerium" core and "yttrium" rim as an inclusion in Y-bearing fluorite from zinnwaldite granite. The plot shows zonal REE distribution in allanite and host fluorite along line 1–7. Fl, fluorite; Tvt, tveitite-(Y); see Fig. 1 for other symbols. BSE images (JIB-4500, JSM-6460LV).

(Ce) that occurs in biotite granites of the Amur region (Table 2).

The structure and chemical composition of allanite was studied on JSM-6460LV, JSM-7001F, and JIB-4500 SEMs equipped with Oxford INCA Energy EDS at the National University of Mineral Resources. The observations were carried out in back-scattered and secondary electrons with a magnification of 85–43000. More than 320 analyses of allanite-(Ce) and 86 analyses of allanite-(Y) have been obtained (Table 1, an. 10).

Allanite-(Y) is variable in composition, which reveals special features in each petrographic selection. Allanite from zinnwaldite granite is the best studied. Y, Fe^{2+}, and Ca are its major components (Table 1, an. 5–7). HREE are the predominant admixtures; their concentrations are measured by a few weight percent (1.0–7.9 Gd, 1.9–7.6 Dy, 2.0–7.1 Er, 0.01–4.3 Yb); Nd and Ce are detected in lower concentrations: 1.4–9.1 and 0.2–6.7, respectively. Allanite hosted in feldspathic matrix is depleted in Al (Table 1, an. 5), but in varieties associated with fluorite and tveitite the Al contents increase to 2.6–4.3 wt %. Ca contents also increase to 14.4–17.7 wt % against the background of 0.2–2.4 wt %, as well as Mn contents to 1.4–3.0 wt %. The share of LREE is diminished (Table 1, an. 6). An important difference between allanite-(Y) and allanite-(Ce) is a low Mg content (<0.1 wt %). The Th content does not exceed 2.1 wt %, whereas allanite-(Ce) contains up to 6.2 wt % Th. A rare allanite-(Y) from Li–F granite on the Chukchi Peninsula contains 8.53 wt % ThO_2 (Table 1, an. 7). Allanite-(Y) in granitic rocks from the dike swarms of the Chukchi Peninsula and Amur region is distinguished by elevated contents of Ca (3.9–11.9 wt %), Al (4.3–10.7), and LREE; Gd and Dy are predominant among HREE (Table 1, an. 8). Allanite-(Y), which replaces predominant allanite-(Ce) in biotite granites of the Amur region, differs in having the highest Fe content (10.7–30.5 wt %), a deficiency in Ca, and elevated concentrations of LREE (Table 1, an. 9).

The total REE contents in allanites commonly vary from 15.4 to 27.4 wt % (Khvostova, 1962). In the Far East, allanite-(Y) is distinguished by much higher total REE content (20.8–47.2 wt %) in contrast to the moderate value (8.1–21.6 wt %) in allanite-(Ce). The REE patterns of allanite-(Y) have a positive slope with enrichment in HREE and a positive Dy anomaly

Table 2. Comparative characterization of allanite-(Y) and allanite-(Ce) from granites in the Far East

Typomorphic attribute	Allanite-(Y)	Allanite-(Ce)
Color of crystal	Reddish brown	Black, dark brown
Color in thin section	Reddish yellow, pale yellow	From orange-brown to reddish-brown
Luster	Vitreous	Resin
Grain size	Tens μm	Hundreds μm to 2 mm
Crystal perfection	Xenomorphic	Euhedral
Morphology	Flattened, duck-footed	Tabular (100), prismatic (001), (101)
Twins	No	Simple (100)
Birefringence	0.010–0.014 to 0.021	0.009–0.012 to 0.016
REE pattern	Yttrium	Cerium
Zoning	Mainly oscillatory	Mainly rough
Inclusions	No	Monazite, cerianite
Mode of occurrence	Along fractures and quartz–feldspar boundary	Intergrown with biotite
Mineral assemblage	Topaz, fluorite, ilmenite, monazite-(Ce), wolframoixiolite, cyrtolite, xenotime-(Y), samarskite-(Yb), aeschynite-(Y), strüverite, fergusonite, chernovite-(Y), tveitite-(Y)	Zircon, ilmenite, apatite, monazite-(Ce), rutile

(Figs. 4a, 4b, 4d). In the secondary allanite-(Y) that replaces allanite-(Ce), the Dy anomaly is negative (Fig. 4c). All studied allanites from the Far East, including Ce varieties, are distinguished by elevated Y and Nd concentrations; the latter reveals a constant positive anomaly of chondrite-normalized contents. Allanite from granosyenite porphyry dikes holds an intermediate position in terms of enrichment in HREE between allanites from zinnwaldite and biotite granites (Fig. 4d).

REE ISOMORPHISM AND CRYSTAL CHEMISTRY OF ALLANITE-(Y)

In allanite-(Ce),a deficiency of cations at site *A* is commonly compensated by a charge at site *M* (Peterson and MacFarlane, 1993; Catlos et al, 2000; Gieré, 2004). In our case, the situation is inverse: a deficiency in *M*-cations is compensated by an excess of cations at sites *A*; thereby REE apparently occupy a part of Ca vacancies at site *A1*. The distortion of structure expressed in anomalously high Si content (>3.5 apfu,

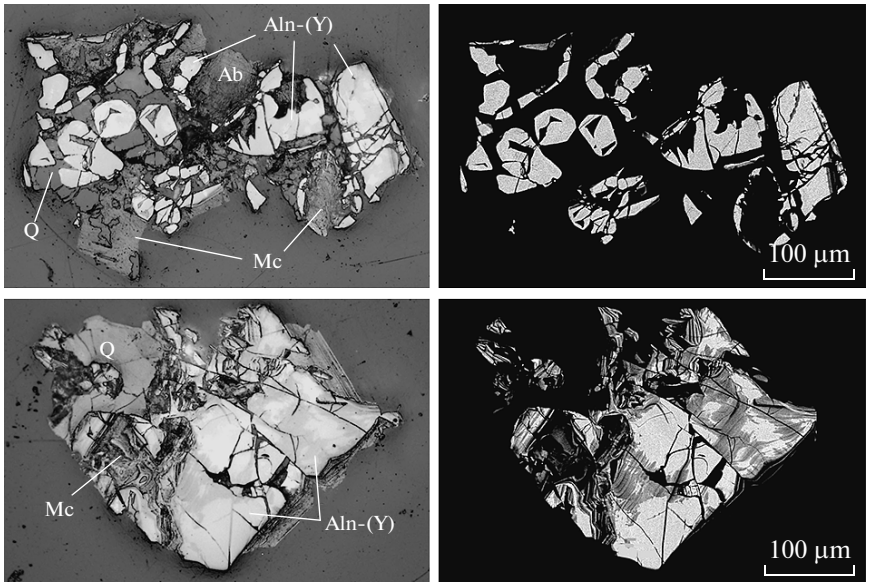

Fig. 3. Allanite-(Y) in biotite granite. Xenomorphic grains in quartz–feldspar aggregate (above) and a fragment of relatively large allanite-(Y) crystal with a spotty zoned core and rhythmically zoned rim (below). Images in reflected light, on the left (Olympus BX51) and BSE images, on the right (JSM-6460LV). See Fig. 1 for symbols.

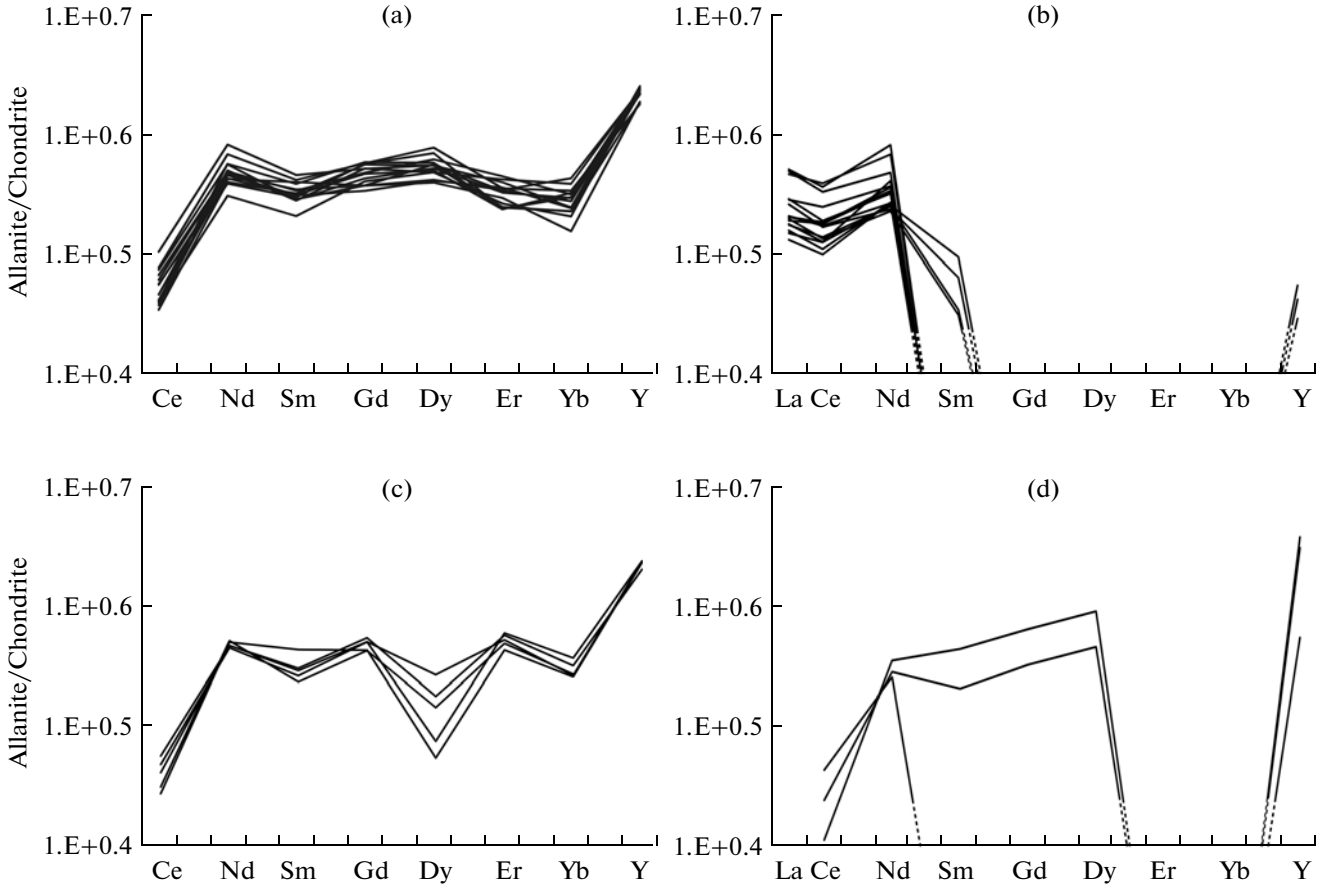

Fig. 4. Chondrite-normalized REE patterns of allanites from the Far East. (a) allanite-(Y) from minor zinnwaldite granite intrusions; (b) allanite-(Ce) from granitic batholiths; (c) allanite-(Y) from greisenized biotite granite; (d) allanite-(Y) from granite and granosyenite porphyries (dikes).

Table 1) does not allow studying isomorphism of allanite-(Y) in detail. Nevertheless, the compositional anomalies await interpretation. In this connection, the combination of *A1* and *A2* voids in the allanite structure with coordination numbers 7 and 8 is crucial. The polyhedrons of *A*-cations are face-sharing, and, according to the third Pauling's rule, this makes the structure of allanite instable. Although, REE^{3+} ions mainly occupy a more loose site *A2*, HREE can replace divalent ions at sites *A1* and *M2* owing to lantanoid compression (Gieré and Sorensen, 2004). Yb^{2+} and Dy^{2+} cations, which are able to replace Fe^{2+} and Ca^{2+} at site *A1* (Table 3), have the most advantageous properties. Origination of divalent REE is probably controlled by reductive crystallization conditions of the rare-metal melt and is a cause of the anomalous chemical composition of allanite-(Y) and the formation of the Dy anomaly (Fig. 4a). A deficiency in Yb, which has an advantage over Dy in the replacement of Ca^{2+} and Fe^{2+}, remains unexplained.

Table 3. Inferred REE isomorphism in allanite-(Y) from granites of the Far East

Parameter	Site/Coordination number					
	A1/7		*A2*/8		*M3*/6	
	replaced cation	isomorphic REE	replaced cation	isomorphic REE	replaced cation	isomorphic REE
Component	Ca^{2+}	Dy^{2+}, Yb^{2+}	Y^{3+}	Nd^{3+}, Gd^{3+}, Er^{3+}	Fe^{2+}	$Dy^{2+}Yb^{2+}$
Ionic radius	1.06	1.13, 1.08	1.019	1.109, 1.053, 1.004	0.78	1.07, 1.02
Charge/ionic radius	1.887	1.770, 1.852	2.944	2.705, 2.849, 2.988	2.564	1.869, 1.961

Ionic radii, after Shannon (1976).

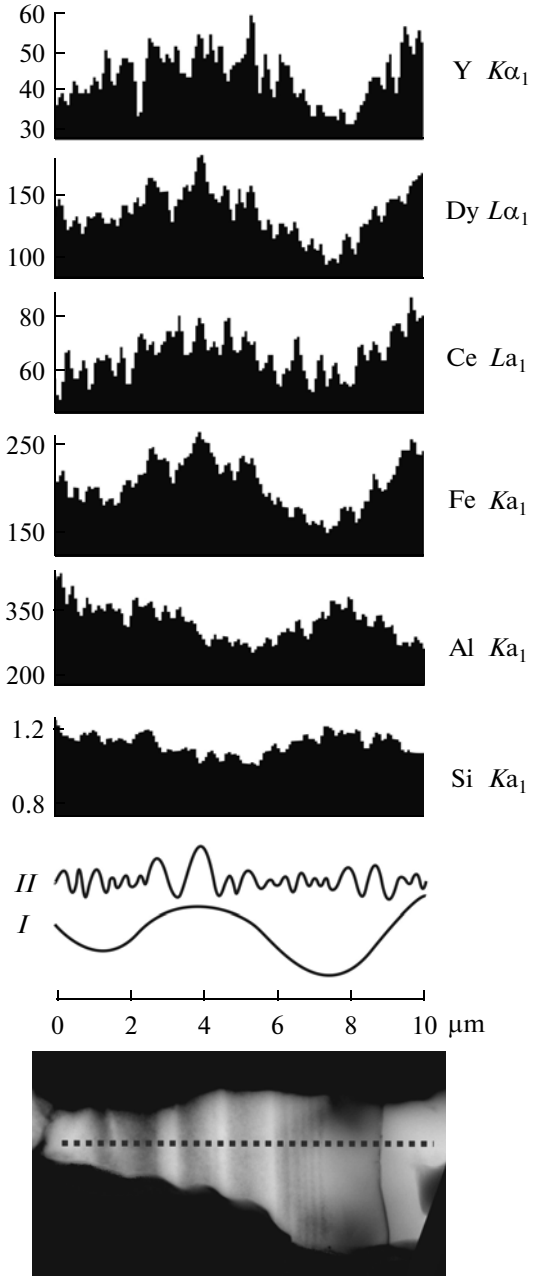

Fig. 5. Intensity variation of characteristic X-ray lines of components as a manifestation of the chemical zoning of allanite-(Y). I, II, types of intensity fluctuations (Fe$K\alpha_1$) that reflect rough (I) and fine oscillatory (II) zoning of allanite. The lower panel demonstrates a BSE image of allanite and a line of electronic scanning with a step of 0.1 μm; voltage is 25 kV.

In allanite-(Y) that replaces allanite-(Ce), on the contrary, a negative Dy anomaly is detected as a microgeochemical indicator of the alteration of allanite-(Ce) under conditions of HREE gain. This anomaly is a result of fractionation of Ca-bearing minerals containing isomorphic Dy^{2+} (tveitite, fluorite, fluocerite) at the magmatic stage. In addition, individual grains of allanite in greisenized biotite granite are enriched in LREE and Th as characteristic components of allanite-(Ce) (Table 1, an. 9). Taking into account their anatomy and association with magmatic allanite-(Ce), it can be suggested that metasomatic alteration of the latter consists of a loss of Ce, La, Nd, Ca, Al, and Fe^{3+} and a gain of Y, HREE, and Fe^{2+}.

The positive Nd anomaly characteristic of all allanites merits further investigation. In allanite-(Y), it can be explained by the crystal chemical affinity of Nd^{3+} to Y^{3+} (Table 3). It should be noted that positive anomalies are inherent to those REE, which are able to take a divalent state (Nd, Dy, Yb). The REE spectrum of allanite-(Y) probably reflects a special geochemical behavior of Nd^{2+}, Dy^{2+}, and Yb^{2+} under reductive crystallization conditions of subalkaline granitic melt.

Two kinds of zoning are inherent to the primary allanite: (1) the rough zoning of major element (Y, Fe, Al, Si, Ca) distribution and (2) the oscillatory zoning of trace elements (Gd, Dy, Er, Yb, Ce, Nd, Sm) (Fig. 5). Zoning (1) displays the domains enriched in Y and Fe or, conversely Si, Al, and Ca as a manifestation of the isomorphic relationships among these groups of elements. Zoning (2) shows harmonic oscillations of REE concentrations that substitute major components. The thickness of zones of this type is 100–500 μm (Figs. 1, 5).

The current crystal chemical concept of allanite does not allow us to use direct calculation of formulas on the basis of oxygen ions in assumption that all iron is ferrous (Finger et al., 1998). Calculation on the basis of eight cations with subsequent equilibration of Fe^{3+}/Fe^{2+} indicates silicon excess at site *A1*, as well as REE and Th excess at site *A2*. Unbalanced distribution of di- and trivalent cations at sites *M* is established. Similar features have been published earlier (Table 1). The range of site occupancies with allowance for 86 analyses is as follows: *T*(Si) 2.15–4.74; *A1*(Ca) 0.01–1.94; *A2*(Y, REE, Th) 1.14–2.89; *M1* (Al, F^{3+}, Ti) 0.01–2.48; *M2* (Al) 0.01–0.91; *M3* (Fe^{2+}, Mg, Mn) 0.09–2.19. Disproportion in occupancy of *A* and *M* sites in allanite structure, noted by Hoshino et al. (2005), attains anomalous values in allanite-(Y). Assuming that REE are confined only to voids *A*, their occupancies are estimated at 3.93 and 0.09 apfu, respectively. It should be emphasized that Y contribution is not higher than 2.03 apfu. We did not equilibrate cation fractions to the Si formula coefficient of 3.00, as has been recommended by Ercit (2002) and Armbruster et al. (2006). The empirical formula of allanite-(Y) in zinnwaldite granite is $Ca_{0.40}(Y_{1.50}REE_{0.84})_{2.34}Al_{0.12}Fe_{0.01}(Fe^{2+}_{0.65}Mn_{0.04})_{0.69}(Si_{3.85}O_{11})O(OH)$. In dikes, allanite-(Y) has the following composition: $Ca_{1.19}(Y_{1.50}REE_{0.50})_{2.00}Al_{0.70}(Fe^{2+}_{0.58}Mn_{0.02})_{0.60}(Si_{3.34}O_{11})O(OH)$.

The aforementioned crystal chemical anomalies are conventionally explained by a metamict transformation of allanite lattice (Campbell and Ethier, 1984; Armbruster et al. (2006), though in our case the Th

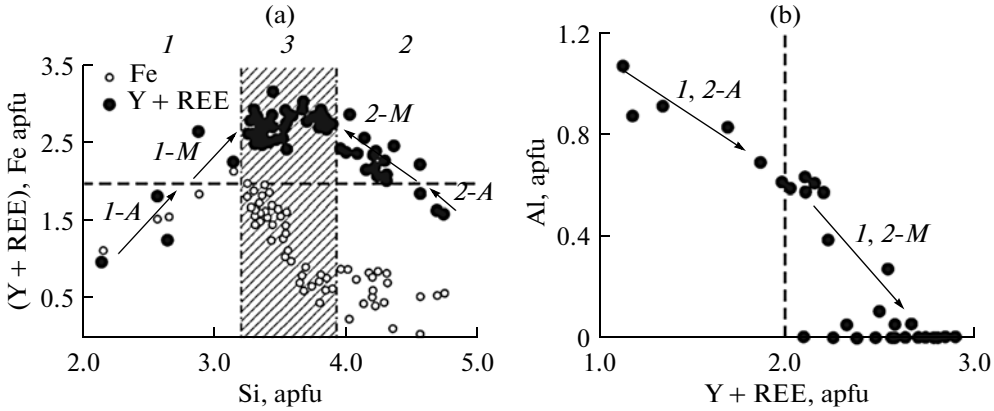

Fig. 6. Isomorphic trends of main components in allanite-(Y) from granitic rocks of the Far East. (a) REE (Y + Gd + Dy + Er + Yb) and Fe^{2+} cations versus Si; (b) Al versus REE. Fields in panel (a): *1*, stable structure of allanite-(Ce); *2*, instable and *3*, metastable structures of allanite-(Y). Arrows show isomorphic trends corresponding to transformation of stable and instable structure into metastable structure by two steps: *A-isomoprphism* (*1-A*, *2-A*) and *M-metamorphism* (*1-M*, *2-M*).

content is relatively low. The appearance of Th in the allanite structure is commonly related to variable valence of iron: $Th^{4+} + 2Fe^{2+} \leftrightarrow Ca^{2+} + 2Fe^{3+}$; $Th^{4+} + Fe^{2+} \leftrightarrow REE^{3+} + Fe^{3+}$. The leading role of ferrous iron in allanite-(Y), as well as positive Th–Fe and negative Th–REE correlations compel us to suggest Th isomorphism as $Th^{4+} + 2Fe^{2+}(Ca^{2+}) \leftrightarrow 2REE^{3+}$ or $Th^{4+} + 2Fe^{2+} \leftrightarrow REE^{3+} + Al^{3+}$. This implies that radiogenic disbalance of allanite-(Y) is conjugated with incorporation of HREE into its composition. In structurally anomalous individual grains with elevated Si content, the REE and Ca concentrations diminish, and this leads to capturing Th and subsequent metamictization of the structure.

At the same time, distortion of the allanite structure is directly related to its saturation with REE cations, which are especially large in allanite-(Y). The high-field-strength REE, being invaded at sites *A* and *M* (Table 3), substantially change the length of *A*–O and *M*–O bonds and *A*–O–*A* and *M*–O–*M* angles, forming the metastable structure of allanite-(Y). Variations in numbers of Y and REE atoms, as well as Fe relative to Si, determine two crystal chemical trends corresponding to the two means of allanite formation.

(1) **Allanite-(Ce) → allanite-(Y) transition** is characteristic of allanites with normal occupancy of *T* site (Si = 2.15–3.15 apfu) with positive correlation between Si cations and (Y + REE) plus Fe^{2+}. Allanite-(Y) is formed by the replacement of allanite-(Ce) under conditions of excess Ca, Al, and Fe^{3+} and a gradual transition from the stable structure of allanite-(Ce) to the metastable structure of allanite-(Y) (Fig. 6a, fields *1*, *3*) with an increasing role of REE and divalent ferrous cations: ${}^{M3}Fe^{3+} + {}^{A2}Ca \leftrightarrow {}^{M3}Fe^{2+} + {}^{A2}REE^{3+}$ (Dollase, 1971; Ercit, 2002; Gieré and Sorensen, 2004; Armbruster et al., 2006).

(2) **Anomalous allanite-(Y) → allanite-(Y).** This way is inherent to the primary allanite hosted in Li–F granite with an inverse relationship between numbers of Y + REE cations and Si. At the high saturation of leucogranitic melt with silica and deficiency in Ca, the crystallizing anomalous allanite-(Y) acquires instable structure oversaturated with Si tetrahedrons (3.15–3.50 apfu) and composed of discontinuous chains of *M*-octahedra containing distorted *A*-voids. The transition from the anomalous to the metastable structure of allanite-(Y) (Fig. 6a, fields *2, 3*) is realized by the replacement of Ca, Fe, and Al with REE accompanied by the formation of vacancies (Fig. 6b).

Irrespective of the mode of crystallization, the structure of allanite-(Y) is formed by two steps. At the first step, named *A-isomorphism*, an increase in the number of REE from 1.00 to 2.00 apfu at *A*-sites (Fig. 6a, *1-A* and *2-A* arrows) is accompanied by the expulsion of Al from *T* site, an increase in Fe^{2+} at sites *M3* and *M1* up to 2.00 apfu, the removal of Ca, Mn, Th, and Al (Fig. 6b). Thereby, vacancies $3Ca^{2+} \leftrightarrow 2REE^{3+} + {}^{A1}\square$ are formed (Peterson and MacFarlane, 1993), whereas *A2* voids and conjugated ${}^{M3}Fe^{2+}$ and ${}^{M1}Al^{3+}$ octahedra are distorted. The epidote structure becomes degraded in direction *1-A* and stabilizes in direction *2-A*.

At the second step of *M-isomorphism* (Fig. 6a, *1-M* and *2-M* arrows), the gain of REE above 1.50 apfu gives rise to further distortion of *M*-octahedra chains and *A*-voids. Ca–REE replacement is completed at *A1* site, and low-volume LREE cations (Dy^{2+}, Yb^{2+}, and probably, Er^{3+}, and Y^{3+}) appear at *M1* and *M3* sites owing to lattice defects: ${}^{M3}Fe^{2+} + {}^{M1}Al \leftrightarrow 2REE^{3+} + {}^{M2}\square + {}^{M3}\square$ (Table 3). Allanite is abruptly depleted in Ca and Al and its structure remains stable in the limited metastable region of relations between mineral-forming components (Fig. 6a, field *3*).

The behavior and role of LREE permanently contained in allanite-(Y) remains ambiguous. It cannot be ruled out that a cause of disturbed stoichiometry should be looked for in the crystal chemical competition of Y, Er, and Yb with Nd, Ce, and Sm. In rare cases of joint crystallization with fluorite and tveitite, allanite is distinguished by heterogeneity expressed in sharp fluctuations of REE concentrations, which

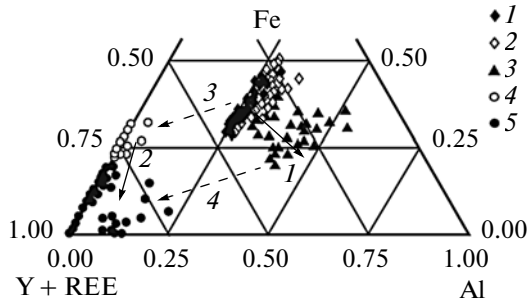

Fig. 7. Evolution of allanite composition in intrusive complexes of the Far East. Allanite-(Ce) from (*1*) biotite granite, (*2*) biotite granite porphyry, (*3*) granosyenite porphyry; allanite-(Y) from (*4*) biotite granite and (*5*) rare-metal zinnwaldite granite and granosyenite porphyry. Arrows in figure: evolution of compostion of (*1*) allanite-(Ce) and (*2*) allanite-(Y); allanite from (*3*) biotite granite and (*4*) rare-metal granosyenite porphyry and zinnwaldite granite.

reflect the isomorphism Y + HREE ↔ LREE with increasing Y/Ce ratio (Fig. 2).

The average number of Y + REE cations (2.48 apfu) indicates the complete occupancy of site *A2* and the partial occupancy (0.77 apfu) of site *A1* (taking into account Ca = 0.23 apfu). The excess of REE atoms participating in the inferred metamorphism at *M* sites is 0.71 apfu, while the corresponding excess of Si atoms in anionic framework of the structure reaches 3.6 – 3.0 = 0.6 apfu. Thus, the structure of allanite-(Y) is metastable in its nature and exists only within a narrow range of crystal chemical and probably physicochemical conditions. Since the XRD data on allanite-(Y) remain insufficient, further study of its crystal structure is needed.

ALLANITE AS A PETROGENETIC INDICATOR

A relatively low abundance of allanite-(Y) in nature is related to the instability of its crystalline lattice and has definable petrologic and geochemical causes. The mineral is formed in a reductive medium oversaturated with REE, water, and fluorine. In contrast to allanite-(Ce), which is contained in granitoids, syenites, gabbroic rocks, schists, gneisses, skarns, and fenites, etc. (Zdorik, 1989; Semenov, 2001; Gieré and Sorensen, 2004), as one of the major REE concentrators in the continental crust, allanite-(Y) has been found largely in granitic pegmatites. The assemblage of allanite-(Y) with gadolinite, fergusonite, yttrotantalite, xenotime, etc. points to rare-earth and rare-metal specialization of the parental magma.

According to the tradition founded by V.I. Vernadsky, granitic rocks are subdivided into orthite (titanite–orthite, orthite-bearing) and monazite (ilmenite–monazite, monazite-bearing) mineral types (Vernadsky, 1929; Vinogradov and Dubrovsky, 1967; Lyakhovich, 1979; Zdorik, 1989). These mineral types are related to different petrographic rock associations: the orthite type, to granitic association of metaluminous calc-alkaline granitoids, and the monazite type, to alaskite and subalkaline leucogranitic associations of peraluminous subalkaline granites (Lee and Barston, 1967; Marin, 1976, 2004; Zdorik, 1989; Broska, Petrík, and Williams, 2000; Gieré and Sorensen, 2004). Different mineragenic implications are imparted to the above mineral types. The orthite type is characteristic of granitoid series in the molybdenum provinces, whereas the monazite type is inherent to the tin provinces (Marin, 2004). Nevertheless, the intrusive rocks containing the studied allanite-(Y) are classified as tin granites. This contradiction is probably explained by the fact that granitic rocks with allanite-(Y) belong to the final members of the rare-metal intrusive series of the Far East, which are distinguished by elevated alkalinity and enrichment in REE (Gonevchuk, 2002). Allanite-(Y) is a product of specific postorogenic subalkaline peraluminous ongonitic magmatism. The absence or insignificant occurrence of such minerals-concentrators of HREE as garnet, titanite, and zircon in Li–F granites (Table 2) is an additional factor that facilitates crystallization of allanite-(Y).

In contrast to allanite-(Ce), which is a relatively early magmatic mineral (Meintzer and Mitchell, 1988; Peterson and MacFarlane, 1993; Hoshino et al., 2005; Spürgin et al., 2009) crystallized at 850–790°C (Broska, Petric, and Williams, 2000), allanite-(Y) is formed at the late stage of crystallization of the residual fluid-saturated subalkaline granitic melt. According to the crystal morphology of zircons, the crystallization temperature of parental granite does not exceed 650°C. In the Far East, allanite-(Y) bears many attributes of late magmatic origin. Among these are veinlet and nework modes of occurrence in aggregate of rock-forming minerals; microstructure of crushing (Fig. 1), and association with fluorite, tveitite, and chernovite. The chondrite-normalized REE patterns with positive slope (Figs. 4a, 4c, 4d) provide evidence for the contribution of fluorine-bearing aqueous fluids to the formation of allanite-(Y), as established for altered granitic rocks and pegmatites in Scotland, Virginia, and California (Exeley, 1980; Sawka et al., 1984; Meintzer and Mitchell, 1988). The oscillatory zoning of allanite expressed in fluctuation of REE concentrations (Fig. 5) also indicates that mineral crystallized from the melt, which changed its composition in the process of mineral fractionation with rhythmically variable partition coefficients owing to a periodic supply of fluorine to the system (Sawka, 1984). The above-described case of the zonal transition of allanite-(Ce) into allanite-(Y) within fluorite and the occurrence of the outer rim deficient in Y (Fig. 2) reflects the important role of F-bearing fluid in the formation of allanite-(Y). The worm-shaped structures observed at margins of allanite-(Y) pseudomorph after its cerium counterpart (Fig. 3) assume various explanations. Ercit (2002) referred such a structure to metamict breakdown of the mineral. At the same time,

it is similar to the primary structures, which arise during fast growth of spinel in undercooled basaltic melt (Roeder et al., 2001) or by crystallization of impact melt in meteorites (Davis et al., 1991) under conditions of abrupt disturbance of component partitioning in the crystal–medium system.

With allowance for the available data, it can be suggested that allanite was formed as a product of late magmatic crystallization of subalkaline granitic melt enriched in Y, Fe, F, and H_2O under relatively low-temperature reductive conditions. Like hydrothermal ixiolite in pegmatite with its disordered orthorhombic structure, which follows ordered magmatic columbite crystallized in the parental melt, stable allanite-(Ce) gives way to metastable allanite-(Y) at the transition from biotite granite to F-rich zinnwaldite granite. Formation of the late magmatic allanite-(Y) is accompanied by pseudomorphs of this mineral after accessory allanite-(Ce) in the biotite granite.

The finds of allanite in the rocks pertaining to the various intrusive complexes give an opportunity to trace the evolution of its composition during consecutive episodes of magmatic activity in the Far East. The most representative data have been obtained for the Late Cretaceous Miao Chang intrusive series, where allanite was identified in rocks of three consecutive intrusive complexes: (1) biotite granites of batholiths, (2) granitic dike swarms, and (3) lithium-fluorine zinnwaldite granite. Allanites make up yttrium and cerium series of generations. Allanite-(Ce) evolves from the main intrusive phase of biotite granite via a supplementary phase to rare-metal syenite porphyry dikes in line with the succession described by Hoshino (2005) for allanites from continental margins. From early to late generations, the Al content increases, whereas REE content remains at approximately the same level (Fig. 7, arrow *1*); the contribution of Y and Gd also increases. The evolution of allanite-(Y) is expressed in increasing REE contents primarily at the expense of Y, HREE, and Nd (Fig. 7, arrow *2*).

As can be seen, no gradual transition exists between allanite-(Ce) and allanite-(Y). The mineral assemblage of allanite-(Ce) and allanite-(Y) is forbidden. Allanite-(Ce) gives way to allanite-(Y) in time in both host and rare-metal granites. In the early biotite granite, this is expressed in the aforementioned metasomatic replacement of allanite-(Ce) with allanite-(Y) (Fig.7, arrow *3*). In the rare-metal intrusive series, the accessory allanite-(Ce) in dikes is followed by the accessory allanite-(Y) contained in Li–F zinnwaldite granite (Fig. 7, arrow *4*). A tendency to complicate the structure of accessory silicate minerals in subalkaline granite complexes with geologic time (Marin, 1985) is confirmed.

The data presented in this paper show that allanite-(Y) can be used as a petrogenetic guide for geological, geodynamic, and mineragenic typification of parental igneous rocks. This mineral is a product of the crystallization of extremely evolved fluid-saturated granitic melt or metasomatic substitution of allanite-(Ce) under relatively low-temperature (<650°C) reductive conditions. Allanite-(Y) is characteristic of subalkaline leucogranite complexes that complete postorogenic granitic series of the Far East continental margins. Allanite-(Y) is an indicator of rare-metal ongonitic magmatism of the Far East.

CONCLUSIONS

(1) The research history of allanite-(Y) shows that this mineral is not abundant because its formation requires special geochemical conditions. Allanite-(Y) is characterized by anomalous and steadily nonstoichiometric chemical composition. The enrichment in Y, HREE, and Fe^{2+} and depletion in Ca, Fe^{3+}, Al, and Th are typical, as well as a yttrium-type chondrite-normalized REE pattern with Dy and Nd anomalies.

(2) The structure of allanite-(Y) is metastable due to metamictization and the complex two-step isomorphism of HREE, Y, Ca, and Fe, which leads to formation of structural defects. Allanite-(Y) was formed in two ways: allanite-(Ce) → allanite-(Y) and anomalous allanite-(Y) → allanite-(Y).

(3) The evolution of allanite composition reflects the geochemical evolution of the granitic series of the Far East, from high-Al allanite-(Ce) in early biotite granite to ferroan allanite-(Y) in rare-metal zinnwaldite granite. Allanite-(Y) is formed in the anomalous geochemical setting inherent to crystallization of fluid-saturated subalkaline granitic melt and serves as a guide of rare-metal ongonitic magmatism completing the Late Cretaceous Pacific Orogeny.

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