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FEATURES OF REE AND Sr DISTRIBUTION IN APATITE OF DIFFERENT GENESIS FROM THE ALDAN SHIELD (SIBERIAN PLATFORM)

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Results of REE and other isomorphic substituted elements distribution in apatite from carbonate, silicate rocks, and skarns from the Aldan Shield (Siberian Platform, Sakha (Yakutia) Republic) as well as peculiarities of its composition are given. Initial data were obtained by electron probe microanalyses (EPMA) in order to clarify the apatite forming process and determine its peculiarities during origin of the investigated rocks and ores.

Analyzing of the studied elements was made on the apatite sections, which were prepared due to cut of the samples perpendicularly to the L_6 axis of the mineral. Received concentration data authors used to create a set of component distribution maps for every studied element. Comparison of the concentration isopleths have been shown complicated pictures of the studied elements distribution besides relatively well prismatic crystal habit of the apatite.

Any clear differences in elements distribution within apatite crystals from the genetically various mineral associations have not been detected. Heterogeneous character of element distribution within the same apatite indicates on irregular components supply for different growth sectors of apatite.

Key words: apatite, REE, Sr, isomorphism, apatite-carbonate and apatite-silicate rocks, skarn, Precambrian, Aldan Shield.

Apatite is a unique mineral for investigations of sedimentary, metamorphic and magmatic processes (in which apatite is typical accessory phase or, more rarely, rock forming component, as in carbonatites, pegmatite, skarns, etc.) due to wide developed of isovalent ($Ca^{2+} \rightarrow Ba^{2+}$, Mn^{2+}, Sr^{2+}; $F^- \rightarrow Cl^-$, OH^-, etc.) and heterovalent ($2Ca^{2+} \rightarrow Na^+ + REE^{3+}$; $Ca^{2+} + P^{5+} \rightarrow REE^{3+} + Si^{4+}$; $2P^{5+} \rightarrow S^{6+} + Si^{4+}$; $Ca^{2+} + P^{5+} \rightarrow S^{6+} + Na^+$, etc.) [3, 10, 14, 15] isomorphic substitutions.

Many investigators have examined the possible role of elements-substituent in apatite from different formations and have suggested that the observed enrichments in some components are probably the result of formation in specific petrologic processes. Indicative examples in this sense are high concentration of strontium (more than 5 wt. % of SrO) in apatite from alkaline rocks from the Hibiny and Murun Massive (Russia), REE (more than 2 wt. % of REE sum) in apatite from carbonatites [12, 13], and manganese (up to 4 wt. % of MnO) in apatite from some pegmatites [2, 10, 14], etc.

These researches have been started firstly during of applying period of the “wet chemistry” classic method, and, therefore, obtained results often reflect of bulk apatite composition of

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sample even with its high purity and refining from mechanical inclusions. To do petrologic conclusions the scientists calculated obtained in such way data and received average volumes for each detected element in apatite which than compared to average contents of the same elements in apatite from the etalon petrologic formations. With improving of the analytical methods and progress in its precision a number of elements with low and very low levels of concentrations in apatite were detected. Application of X-ray fluorescence (XRF), instrumental neutron activation (INAM) and inductively coupled plasma mass spectrometry (ICP-MS) methods helped in characteristics of elements for coexisting with apatite minerals (oxides, silicates, carbonates, etc.) with low levels of the similar components [2, 7]. Due to these significant peculiarity new possibilities for determinations the same elements from different generation of the minerals and, therefore, obtaining general pictures elements distributions from the beginning up to final stages of mineral and rocks forming processes.

For estimations of similarity–difference between separate minerals and different generations of the same mineral in respect to common petrologic etalons, besides average amounts, investigators involved various component diagrams which are regarded as marks for typical petrologic processes. For example, chondrite-normalized REE distribution patterns for apatite as well as for coexisting phases have routine application for modern works [7, 8]. Sometimes, for illustrative purposes and comparisons geologists employed shale-normalized REE distribution patterns for apatite or associated minerals from distinguished sedimentary in origin rocks.

In any auspicious case, there is a big vagueness concerning determined elements in host apatite. Improving of local analytical methods and its wide development for apatite investigations, have been support success in determination of separate elements species in the mineral and determination of numbers minerals-inclusions which conclude of Fe, Mn, S, Si, etc. At the same time, using of the different precise methods, various natures of separate elements in apatite have been detected both as isomorphic traces in its structure and as small inclusions in host mineral. Future development of investigations of the endogenous rocks, which were early regarded as exotic formations, with increasing of their role as a source of critical metals for modern technique and high technology apatite became significant mineral as a bearing phase of important industrial elements or is permanent component in the mineral associations from ore of such elements.

Analysis of last investigations. Much less works have been conducted on metamorphic rocks and especially on determining the speciation of different elements in apatite from the Precambrian rocks and phosphorus, iron and REE ores. REE, Sr, etc. contents and their normalized patterns are widely used for recognition of the primary nature different rocks and for the assessment of their subsequent alterations. In contrast to the considerable elements mobility during hydrothermal and metasomatic alterations, apatite from metamorphic rocks is often demonstrated inert behaviour of its components. Due to this case there is a possibility to distinguish primary protolithic and subsequent processes signatures. At the same time, any detail investigation in this direction necessary to carry out with determination of location forms for each element admixture in apatite. The discovery of the Seligdar apatite deposit in the Precambrian rocks of the Aldan Shield (Siberian platform) [4] has stimulated scientific investigations of this unusual for metamorphic blocks apatite-bearing formations. Noted above peculiarities of elements distribution in the metamorphic apatite can be apply for understanding nature of its location as well as origin of apatite-bearing ores.

At the earlier stages of apatite ores exploring at the Aldan Shield, in addition to geological and structural data [4, 6], geochemical and isotopic evidences were appeared [3, 9, 13] on their origin due to primary sedimentary protolith rich initially in phosphorus concentrations. Based on formal similarities in mineralogical composition and apatite enrichment of the metamorphic rocks, little knew before, some investigators proposed ideas about possible metasomatic nature of the apatite rocks, their carbonate origin or connection to ultrabasic-alkaline with carbonatite formation, etc. Besides geochemical data from geologists [9, 13], which shown difference in REE and Sr enrichment of apatite from carbonatite, ultrabasic and alkaline rocks, some subsequent works [12] contain declarations on unique nature of the Seligdar apatite deposit.

In elaboration of our previous works on chemical composition of apatite from different formations of the Aldan Shield, its peculiarities from exactly metamorphic rocks, grade and variations of elements admixtures, and their often dual character of location in the crystals [11, 16], we collected direct data on spatial distribution of the significant elements within apatite from three famous genetic types of the apatite-bearing rocks. Taking into account suggestions on relationships of apatite enrichment and composition of phosphorus-bearing rocks in the deposits and peculiarities of elements distribution in host apatite we attempted to determine the picture of its distribution, differences for various genetic apatite types and origin of apatite in connection with composition variations of the mineral forming fluids.

Geological setting and main rocks types. Following the Seligdar deposit, other apatite prospects were a1so discovered in the Aldan Shield [6, 8] and supplied geologists with new evidences on metamorphic origin of apatite significant mineralization here. Since then hypotheses of its origin as exotic to the Aldan Shield (a collapse caldera above an ancient volcano, carbonatites, a two-stage graben, a stockwork over a mafic-ultramafic intrusion, an explosion pipe, a meteorite crater, metasomatites after missourites, a neck of peculiar carbonate-sulphide lavas, etc.) have given place [4] to better documented ideas about the primary fold structure and metamorphogenic nature of the deposit.

Examination of the apatite-bearing rocks has established the predominant development of two geological-technological kinds of ores [6]: apatite-silicate (apatite, diopside, amphibole, phlogopite, feldspar, and the less abundant of scapolite, quartz, sphene, magnetite and pyrite) and apatite-carbonate (apatite, dolomite, calcite, Mg-silicates, quartz, sulphates, martite, feldspar). These have respectively been termed Khani- and Seligdar-type ores, after the two deposits, where the features of their composition and geological shapes are most clearly defined.

The apatite-bearing rocks made up lensed-banded sequences with gneisses and schists, which have all undergone the same stages of deformation and are marked by a single structural style. The similar geological-structural position of the apatite-bearing rocks in the section has led to a hypothesis that lithological and stratigraphic factors are decisive in control of their distribution. This has determined the direction of exploratory work and assessment of similar deposits in the metamorphic rocks of the Shield [4, 6, 8].

Sampling and analytical methods. During field trips and lab researches we studied apatite-silicate and apatite-carbonate metamorphic rocks from different localities at the Aldan Shield as well as skarns (apatite-silicate-carbonate rocks) mineral associations to find difference in origin of apatite crystals.

Taking into account various sharply isotopic compositions of the C and O from carbonates different in origin [9], we used this feature to detected secondary hydrothermal and metasomatic alteration with replacing of primary carbonate materials and sampled simultaneously apatite crystals and associated carbonate materials in the Seligdar, Birikeen, Chukurdan deposits within the metamorphic rocks, and in the Emeldzhak, Kuranakh, Katalakh deposits in skarns. We observed shape and field relationships between apatite and carbonates, and with hosting and associated rocks. Special attention has been paid in determination of apatite and associated minerals distributions within poor and rich in apatite places of the hosting rocks. Beside field investigation of apatite we also collected a set of the carbonate samples from different localities named above.

Beside routine geological investigation we carried out petrographic studying of thin sections to determine of crystallization ordering for minerals of ores, gneisses, schists, marbles, etc., which are typical for the deposits of region, as well as to check heterogeneity of individual minerals of the rocks to find sectors which were not affected by late alteration.

Previously estimated amounts of different components in the apatite by XRF, INAM and ICP-MS methods [7, 11, 16] were used to detect species of their presence in crystals from various rocks. We selected homogenous prismatic crystals with relatively similar sizes up to 1cm in length. To do analysis of the elements with evidences of their structural position, perpendicular to apatite L_6 axis sections were made.

The quantitative electron probe microanalysis (EPMA) of oxygen and other elements in variably sections of the apatite preparations has been involved to measure the elements concentrations in individual grains using facilities of the Ocean Research Institute (University of Tokyo, Nakano, Tokyo, Japan). Chemical analysis was carried out with an automatic 4-channel EPMA (JXA-733, JEOL) connected to a computer performing a standard correction online by the ZAF method in which the true concentration is theoretically calculated by correcting the atomic number, absorption and X-ray fluorescence effects on the intensity of X-ray [1, 5]. About 60 points on apatite sections were analyzed by electron probe microanalyzer.

The oxygen and carbon isotopic compositions in the whole-rock carbonates were analyzed in the Laboratory of Stable Isotopes (Institute of the Lithosphere of Marginal Seas, Moscow). Carbon dioxide was released from carbonates using the $PbCl_2$. The relative difference in oxygen and carbon isotope ratios in CO_2 (gas) was measured on a Varian MAT-250 mass spectrometer. The laboratory CO_2 was used as standard during isotopic measurement.

The accuracy of $\delta^{13}C$ and $\delta^{18}O$ measurement in whole-rock samples was ±0.1 and ±0.2 ‰, respectively. The Craig correction was taken into consideration for the determination of $\delta^{13}C_{samp}$ (PDB) and $\delta^{18}O_{samp}$ (SMOW). The oxygen correction was taken into account for $\delta^{13}C_{samp}$ (PDB).

Chemical composition of apatite in the main apatite-bearing rocks of the Shield. List of elements determined early by chemical analysis of the bulk apatite samples [2, 7, 12] have been reduced after apatite investigations with the XRF, INAM and ICP-MS methods [2, 7]. Admixtures of Al_2O_3, Fe_2O_3, MnO, and MgO detected before are represented by phases of mechanical particles. At the same time, permanent presence of Si, S, Sr, and REE in microprobe analysis (see Table) led to opinion on structural substituents main components of apatite by these elements. Apatite from three genetic types of rocks is rich in fluorine, and there is a clear similarity in the F levels between them, but concentrations of Si and S are higher in apatite from apatite-carbonate rocks and skarns, under very low (about 0.06–0.03 %) concentration of these elements in apatite from apatite-silicate rocks. Average grade of Sr is low in all analyzed samples, and much less than previously reported [12, 13] for apatite from carbonatites and alkaline rocks.

Apatite from apatite-carbonate rocks and skarns is rich in La and Nd, under low concentrations of Sm and Gd (see Table). At the same time, apatite from apatite-silicate rocks is poor in all REE. So, there are not any similarity in average REE compositions in apatite even with formally the same mineralogical associations, and apatite from etalons endogenous formations.

Average chemical composition of three apatite samples after microprobe analysis, wt. %

Element	Sample 1 (*n* =15), from apatite-carbonate rock		Sample 2 (*n* = 25), from apatite-silicate rock		Sample 3 (*n* = 19), from skarn	
	*1**	*2***	*1*	*2*	*1*	*2*
P	17.52	0.17	18.50	0.17	17.71	0.21
Ca	38.34	0.21	39.25	0.34	38.53	0.20
F	2.94	0.06	3.03	0.11	3.10	0.08
Si	0.40	0.07	0.06	0.05	0.48	0.12
Al	0.002	0.003	0.002	0.003	0.003	0.003
Fe	0.003	0.007	0.060	0.008	0.040	0.120
Mg	0.007	0.006	0.004	0.005	0.005	0.003
Mn	0.01	0.01	0.01	0.01	0.01	0.01
Na	0.080	0.030	0.003	0.007	0.010	0.030
K	0.003	0.007	0.002	0.004	0.003	0.005
La	0.30	0.08	0.05	0.06	0.37	0.09
Nd	0.28	0.10	0.06	0.09	0.30	0.10
Sm	0.06	0.04	0.02	0.03	0.05	0.04
Gd	0.09	0.06	0.01	0.02	0.08	0.06
Sr	0.09	0.02	0.06	0.03	0.04	0.08
S	0.30	0.06	0.03	0.03	0.17	0.08

* – average value;
** – standard deviation.

Spatial distribution of the constituent elements of the apatite. Obtained data on chemical composition of the analyzed apatite have been involved to create a set of component distribution maps for every studied significant element (Fig. 1–6).

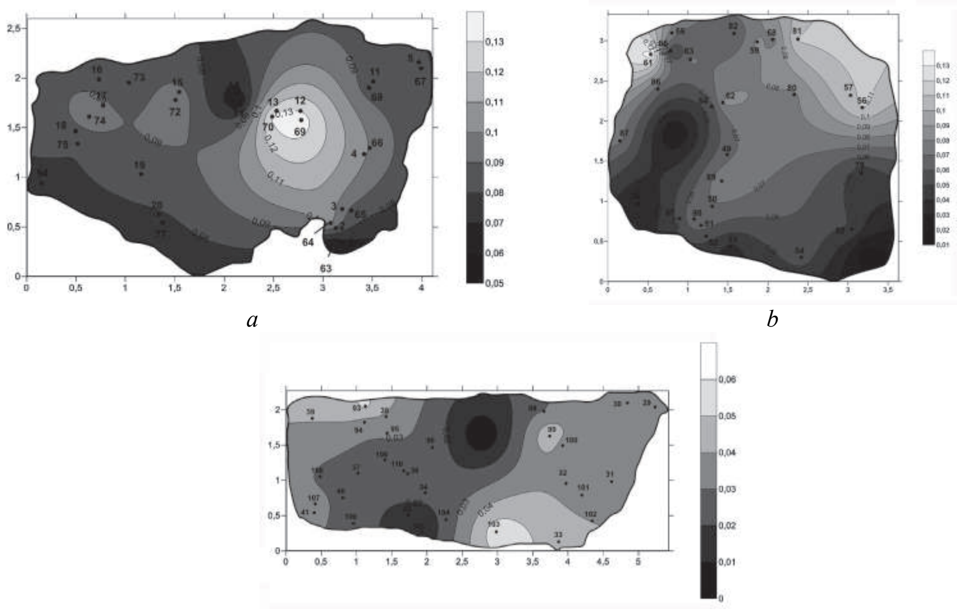
Pictures of Sr distributing in the separate apatite crystal are quite different for the mineral from apatite-carbonate and apatite-silicate rocks and skarns (see Fig. 1). There are series of irregular minimum and maximum concentrations in separate sectors of crystals.

At the same time there is a vague similarity in position of the Sr, Si and S concentration maximums for apatite from apatite-carbonate rocks (see Fig. 1, 2).

Some joint dim concentrations centre there is also in the pictures of Si and S distributions for apatite from skarns (see Fig. 2, 3).

Much more irregular pictures are characterized for REE distributions (see Fig. 4–6). First of all, we should note absence of any regulating in characteristics for separate REE. There are isolated concentration fields with various shapes without any correlations between La, Nd, Sm, Gd in apatite of the same genetic types of the apatite-bearing rocks. Analogous chaotic locations of the same elements are typical for all apatite crystals (see Fig. 4–6).

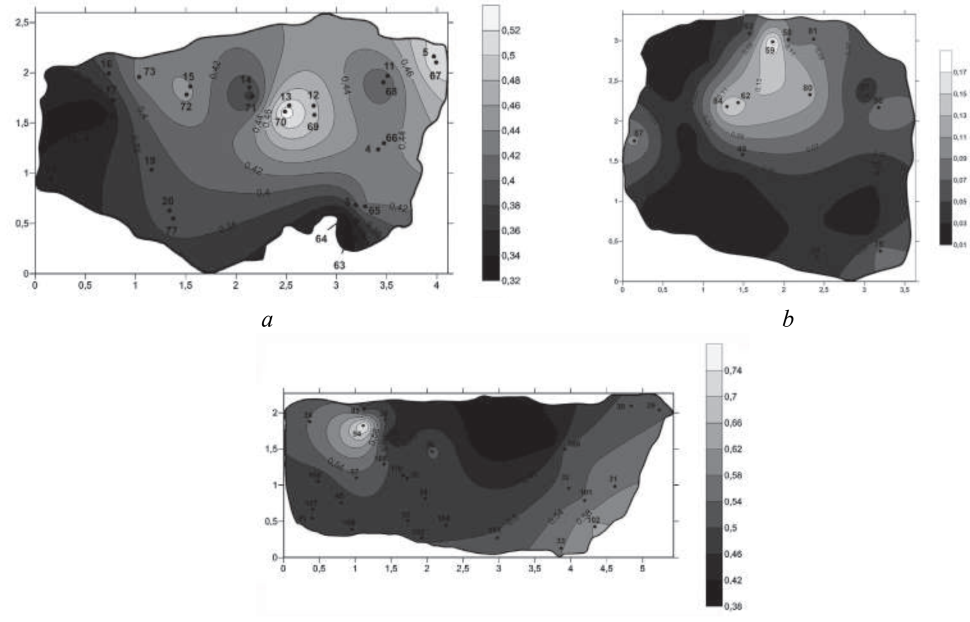
Isotope signatures in carbonates of the region. In addition to previously published results of isotopic investigations [8, 9], we studied isotopic composition of carbon and oxygen in carbonates of the various studied apatite-bearing rocks from the Aldan Shield in hope to estimate the differences for these so heterogeneous in geological sense formations as well as to avoid of products from the secondary processes during sampling. We suggested [6] high $\delta^{13}C$ value up to +5.5 ‰, PDB for carbonates from main ore bodies in the Aldan deposits. The isotopic compositions of carbonates from apatite-carbonate rocks of all apatite deposits of the Aldan Shield yield a scatter of $\delta^{13}C$ value from –0.5 up to +5.5 ‰ (PDB) and $\delta^{18}O$ value from +13.6 up to +22.2 ‰ (SMOW) [9].



a *b*

c

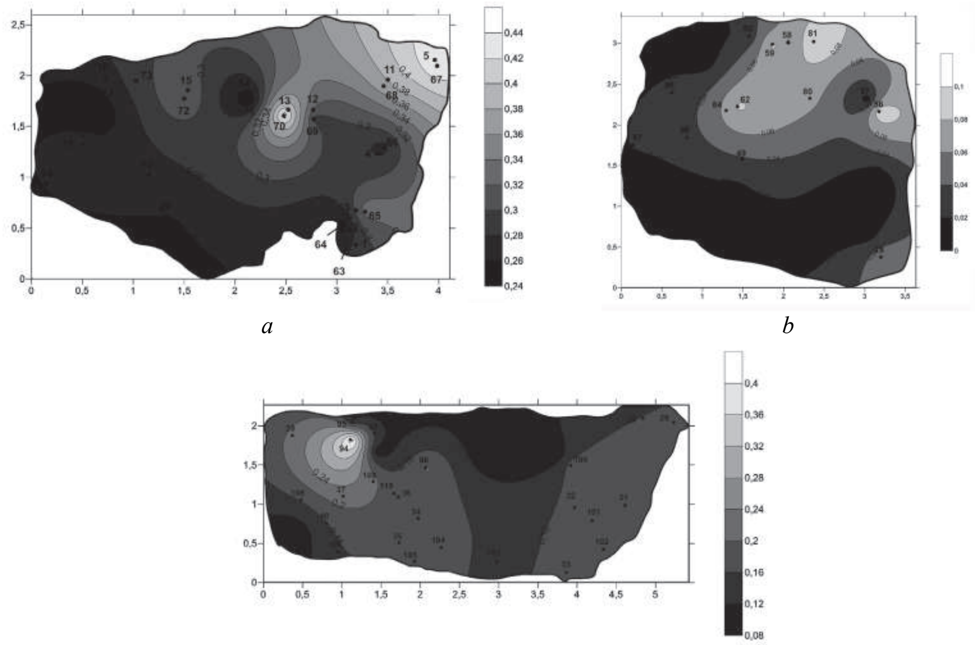
Fig. 1. Distribution of Sr in the separate apatite crystal from apatite-carbonate (*a*) and apatite-silicate (*b*) rocks and skarn (*c*).



a *b*

c

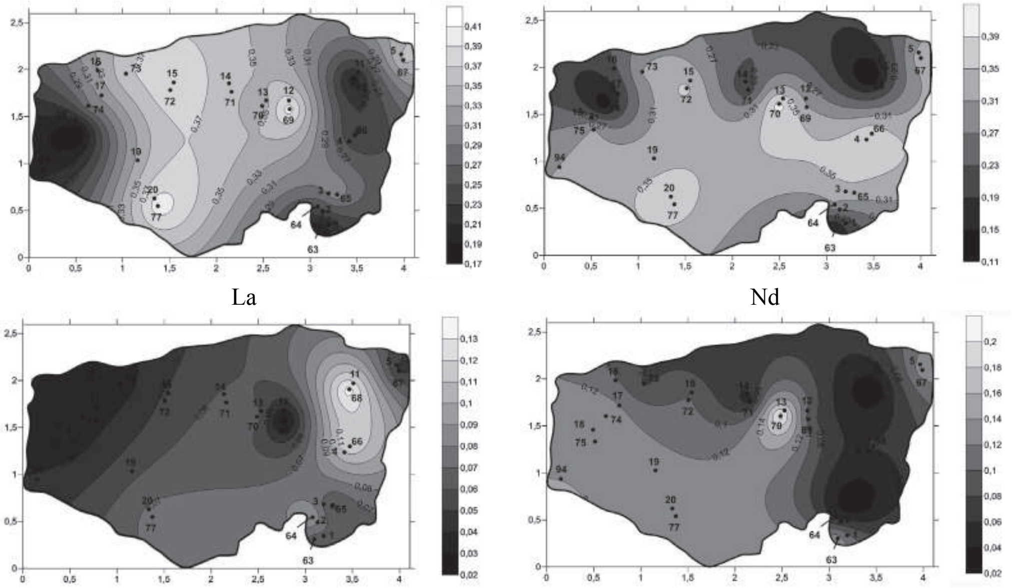
Fig. 2. Distribution of Si in the separate apatite crystal from apatite-carbonate (*a*) and apatite-silicate (*b*) rocks and skarn (*c*).



a *b*

c

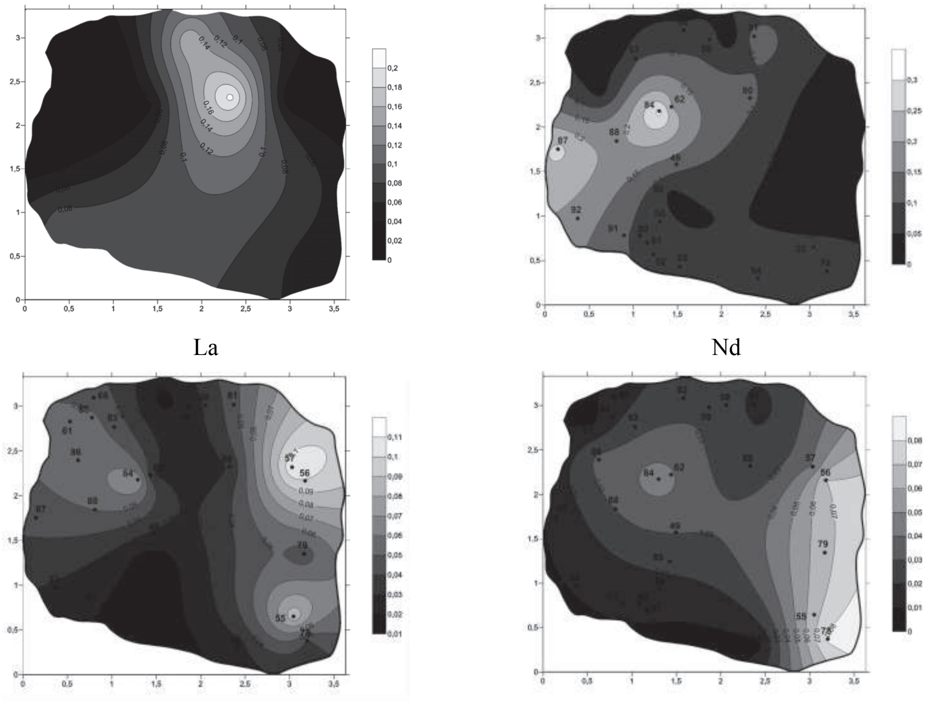
Fig. 3. Distribution of S in the separate apatite crystal from apatite-carbonate (*a*) and apatite-silicate (*b*) rocks and skarn (*c*).



La Nd

Sm Gd

Fig. 4. Distribution of REE in the apatite crystal from the apatite-carbonate rock.



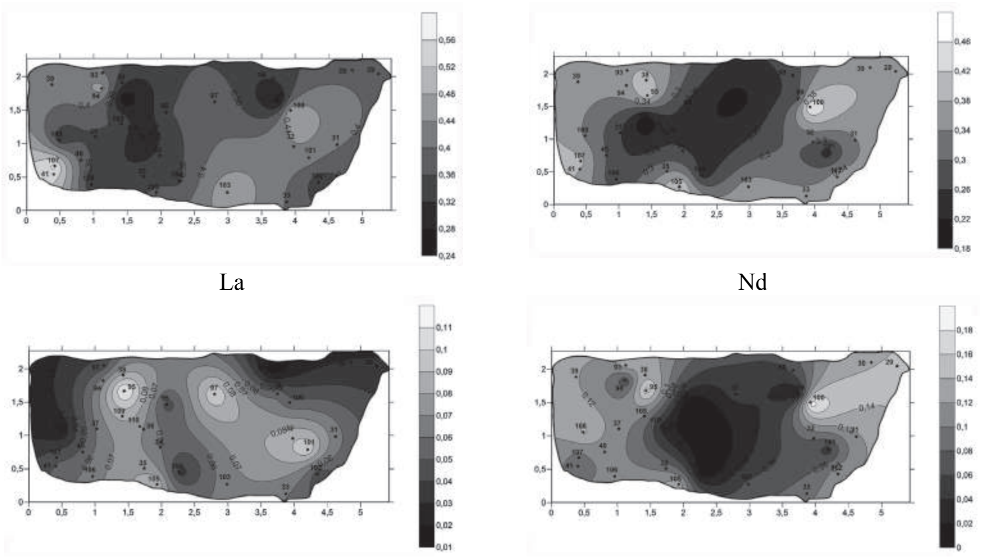
La

Nd

Sm

Gd

Fig. 5. Distribution of REE in the apatite crystal from the apatite-silicate rock.



La

Nd

Sm

Gd

Fig. 6. Distribution of REE in the apatite crystal from the skarn.

These values distinctly differ from data points of carbonates from apatite-free basement rocks (marble and calciphyres) and more young limestones of the Yudoma Formation. This difference is much sharper between isotopic data from carbonates from apatite ores and late veins and skarns, which are significantly enriched in light carbon isotope.

So, obtained data have been suggested on presence a number of element admixtures in apatite from all observed formations. Commonly they have dual nature and are represented by fine mineral inclusions or substituent elements which replaced in apatite structure some main components. Permanent presence of Sr, Si, S and REE as main replaced elements in apatite structure is clear evidence on enrichment by these components of initial solutions during apatite-forming processes without relation to its genetic nature. Quite different levels of studied elements in the investigated apatite from the Aldan Shield and some etalon endogenous object have been indicated obvious various natures of the apatite-bearing formations. From second side, with these data we obtained facts on absence of any similarity between carbonates from the apatite-bearing metamorphic rocks of the Aldan Shield and from carbonatites from famous provinces or similar endogenous formations.

The isotopic data have been shown inert behaviour of CO_2 during minerals formation and possibility to preserve of isotope marks on primary origin of carbonates from the rich in apatite metamorphic rocks of the Aldan Shield. We got evidences of inert role of CO_2 in its possible recrystallization from primary sedimentary protolith rich in phosphorus.

So, we can regard peculiarities of various elements distribution in apatite as a primary process. Comparison of the concentration isopleths have been shown complicated pictures of the studied elements distribution besides relatively well prismatic crystal habit of the apatite. Heterogeneous character of elements concentrations within separate crystals indicates on diverse and fluctuated elements supplying of different sectors of the crystals. It is evidence of more complicated model of apatite growth than proposed before due to layers-concentric rims surrounding initial centres of growth.

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ОСОБЛИВОСТІ РОЗПОДІЛУ РІДКІСНОЗЕМЕЛЬНИХ ЕЛЕМЕНТІВ ТА СТРОНЦІЮ В АПАТИТІ РІЗНОГО ГЕНЕЗИСУ З ПОРІД АЛДАНСЬКОГО ЩИТА (СИБІРСЬКА ПЛАТФОРМА)

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Відомо, що в апатиті з генетично різних геологічних утворень дуже поширені ізо- та гетеровалентні ізоморфні заміщення. Науковці часто використовують їх як індикатори петрологічних процесів формування апатитоносних порід. Такі дослідження започаткували ще в період використання класичного методу так званої мокрої хімії, тому отримані результати часто відображали валовий склад проб апатиту навіть за порівняно високого ступеня їхнього очищення від механічних домішок. Удосконалення локальних методів аналізу й широке впровадження їх під час досліджень апатиту сприяло визначенню форми наявності окремих елементів у мінералі та виявленню низки мінералів-включень, які є носіями, наприклад, заліза, манґану, сірки, кремнію тощо. Водночас виявлено різну природу цих елементів в апатиті: або як ізоморфна домішка в його структурі, або як дрібні включення в мінералі-господарі. З розвитком різноманітних досліджень порід, що їх раніше трактували як екзотичні, та зростанням ролі таких порід як джерела металів для сучасної техніки й високих технологій апатит стали вивчати прискіпливіше, оскільки він є і носієм низки важливих промислових елементів, і неодмінною складовою в мінеральних асоціаціях руд таких елементів.

У комплексах ультраосновних лужних порід з карбонатитами, у скарнах, лужних метасоматитах та інших утвореннях наявний апатит, у якому завжди є рідкісноземельні елементи і стронцій. Тому цей мінерал почали використовувати для з’ясування генезису й рудоносності таких, ще слабко вивчених порід. Раніше ми дослідили вміст і склад рідкісноземельних елементів та Sr в апатиті з декількох проб докембрійських порід дискусійного походження з використанням нейтронно-активаційного методу та методу масспектрометрії з індуктивно зв’язаною плазмою. Однак, попри високу точність методів, наші проби за природою могли бути неоднорідні, тому ми вирішили застосувати прилади для локальних досліджень: іонний та електронний мікроаналізатори, а також електронний мікроскоп з енергодисперсійною приставкою. Дослідження засвідчили наявність у мінералі рідкісноземельних елементів і Sr у формі як ізоморфних домішок у його структурі, так і мікровключень (~ 0,*n* мкм) – монациту й Ca-рідкісноземельного фосфату групи рабдофану (водних рідкісноземельних фосфатів).

Наведено результати подальшого вивчення апатиту, зокрема, щодо просторового розподілу окремих компонентів у різних секторах зерен мінералу. Описано результати розподілу рідкісноземельних та інших елементів, що наявні як ізоморфні домішки в апатиті з карбонатних і силікатних порід, а також скарнів Алданського щита (Сибірська платформа). Схарактеризовано особливості складу рідкісноземельних елементів та їхнього розподілу в межах кристалів апатиту різного генезису. Вихідні дані отримано в окремих точках кристалів під час дослідження їх електронним зондовим мікроаналізатором. Розподіл окремих елементів визначено на зрізах кристалів апатиту, які виготовили розрізуванням кристалів перпендикулярно до осі L_6. Отримані результати (концентрація елементів) вико-

ристано для побудови низки концентраційних карт розподілу окремих елементів у межах досліджуваних площин.

Порівняння концентраційних ізоплет засвідчило складну картину розподілу цих елементів, незважаючи на порівняно правильний призматичний габітус досліджуваних кристалів. Виявилось, що нема чітких і однозначних відмінностей у картині розподілу елементів у кристалах апатиту з мінеральних асоціацій різного генезису. Гетерогенний характер розподілу елементів у межах конкретного кристала свідчить про різнорідне живлення і флуктуаційне постачання окремих елементів у різні його сектори. Це є доказом складнішої моделі росту апатиту, ніж пропонували раніше – завдяки пошарово-концентричному наростанню навколо вихідного центра росту.

Ключові слова: апатит, рідкісноземельні елементи, стронцій, апатит-карбонатні й апатит-силікатні породи, скарни, докембрій, Алданський щит.