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### **PECULIARITIES OF X-RAY LUMINESCENCE OF APATITE FROM CARBONATITES AND POSSIBILITIES OF THEIR USE FOR SEARCH PURPOSES**

X-ray luminescence of apatite from different genetic types of apatite-bearing rocks has been studied to elucidate its geochemical characteristics and the possibility of using it as an indicator mineral for ore formation conditions. Apatite of different genesis from the rocks of the Ukrainian, Aldan and Baltic shields, the Baikal region, the Sette-Daban ridge and the Maimecha-Kotui province of Yakutia has been researched. The luminescence intensity of characteristic isomorphic trace elements of apatite –  $TR^{3+}$  ( $Gd^{3+}$ ,  $Ce^{3+}$ ,  $Eu^{2+}$ ,  $Dy^{3+}$ ,  $Sm^{3+}$ ,  $Nd^{3+}$ ) and  $Mn^{2+}$  has been studied. The obtained results can be used to determine the genetic type of apatite-bearing rocks, the type of mineralization, the conditions of mineral formation and for research purposes.

*Key words:* apatite, carbonatites, X-ray luminescence, isomorphic trace elements, rare earth elements.

**Introduction.** Apatite is formed under various geological conditions and occurs in different types of igneous, sedimentary and metasomatic rocks. Igneous apatite ores supply up to 20 % of world phosphate production [11]. Apatite is an important accessory mineral of many igneous and metamorphic rocks. Its presence and distribution are often used to model geological processes. Apatite is the main industrial mineral in deposits of rare earth elements and phosphate raw materials, which are associated with carbonatites and associated alkaline rocks. The wide *PT*-range of magma crystallization conditions during apatite formation causes variations in its chemical composition [20, 21, 23–26]. Features of the structure of the mineral determine the possibility of concentration in it of elements that occur in the Earth's crust in small quantities (especially rare earths). The structure of apatite controls or affects the patterns of distribution of these elements depending on the physicochemical parameters of the igneous, metamorphic or metasomatic system. Therefore, apatite is considered a sensitive indicator of mineral formation conditions [9].

**Formulation of the problem.** Apatite as a characteristic accessory or rock-forming mineral in rocks of different genesis has been studied somewhat less compared to other rock-forming and ore minerals. This is largely due to the complexity of laboratory and analytical studies (separation of pure monofractions, determination of the trace elements content, etc.) [9]. Based on the features of apatite X-ray luminescence, we tried to determine its belonging to rocks of a certain genetic type and the possibility of using them for search purposes.

**Analysis of recent research and publications.** There are many publications devoted to the study of the luminescent properties of apatite. In [15], A. Portnov and B. Gorobets described the features of photoluminescence of apatite from rocks of different genetic type and the possibility of using luminescence data as indicator features of the mineral. In [7], V. Guliy, L. Kopylova and G. Kuznetsov presented the results of the study of apatite X-ray luminescence spectra from different rocks of the Aldan, Baltic and Ukrainian shields, the Baikal region, the Sette-Daban ridge and the Maimecha-Kotui province of Yakutia. The researchers concluded that the use of X-ray luminescent data does not allow determining the genetic type of primary apatite-bearing rocks. In addition, the authors concluded about the unified geochemical specialization for rare earth elements of individual rock massifs. Therefore, according to the X-ray luminescence of apatite, it is possible to determine its formation belonging to carbonatite-bearing complexes with a high probability. A. Tarashchan, G. Kuznetsov, and O. Povarennykh [18] investigated the luminescence spectra of rare-earth ions in apatite crystals (natural and TR-ion-activated by the solid-phase diffusion method) to determine the structural position of trace element centres. Work in this direction was continued by B. Gorobets and A. Rohozhyn [5]. In [9], O. Dubyna and co-authors substantiated the correlation between the geochemical parameters of apatite and its belonging to rocks of a certain genetic type, with which complex (apatite-ilmenite, apatite-rare earth, etc.) or only apatite mineralization associates. Scientists have found that the nature of the distribution of trace elements in apatite depends on the petro- and geochemical characteristics of the host rocks, the peculiarities of their differentiation and fractionation of magmatic melts from which they have been crystallized. In [8], O. Dubyna and colleagues focused on determining the content of rare earth and other elements in apatite from rocks of the Chernihivskiy carbonatite massif (Ukrainian Shield). The authors of [22] proposed to use the relationship between the composition of apatite, its structure, X-ray luminescence and the type and intensity of changes in porphyry rocks as a potentially fast and effective method for the study of porphyry mineralization.

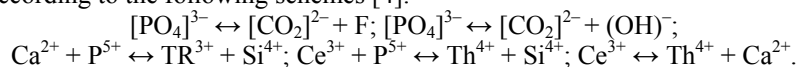
The **purpose** of the article is to analyze the X-ray luminescent characteristics of apatite from rocks of different genetic types and to evaluate the possibility of using the ratio of apatite luminescence intensities from carbonatites of different deposits for search purposes.

**Research methods.** X-ray luminescence spectra were studied in the spectral range of 200–800 nm. Excitation of the spectra was performed using an X-ray tube БСВ-21 with a cobalt anticathode at a voltage of 30 kV and a current of 15 mA. Measurements were performed at room temperature using an automated installation based on a monochromator МДР-23. A photoelectric multiplier ФЕП-106 was used as a radiation receiver. The registration of experimental data at the output of the ФЕП is provided by the use of a computer program, which set the required number of photons at each point of a given spectral interval and the step of scanning the spectrum, followed by processing the measurement results.

**Research results.** We studied apatite of different genesis from the rocks of the Ukrainian, Aldan and Baltic shields, the Baikal region, the Sette-Daban ridge and the Maimecha-Kotui province in Yakutia. Apatite-bearing rocks are represented by the following varieties: metamorphosed (primary-sedimentary) pyroxene rocks of the Precambrian [6]; marbles and calciphyres formed on Precambrian carbonate rocks [19]; apatite-carbonate deposits of the Aldan Shield, formed due to repeated recrystallization and metasomatic alteration of the primary sedimentary substrate or ancient crusts of weathering [3, 13]; Precambrian magnesium me-

tasomatites (skarns); granite pegmatites of Precambrian age [12]; gabbro [10]; pyroxenites [17]; ultrabasic complexes with carbonatites [16].

The structure of apatite was determined in 1930, and since then it has been repeatedly confirmed by many authors [1]. It is characterized by three types of polyhedra:  $Ca_I$  and  $Ca_{II}$  (in accordance with the two cationic positions in the structure) and  $[PO_4]^{3-}$  [11]. Coordination of  $Ca_I$  is 9O (more precisely, 6O + 3O), coordination of  $Ca_{II}$  – 6O + 1F (or 6O + Cl). The ratio of  $Ca_I : Ca_{II}$  is 2: 3 [14]. The presence of three crystallochemical positions in the structure of apatite – two cationic and one anionic – contributes to a broad heterovalent substitution in the mineral according to the following schemes [4]:



The distribution of isomorphic trace elements between crystallochemical positions will depend on the type of cation, its amount, as well as anionic substitutions [11]. TR and Sr atoms often replace  $Ca_{II}$  positions instead of  $Ca_I$  [2].

What is the reason for using the method of X-ray luminescence? Firstly, it is an express method, secondly, it requires a minimum amount of starting material, and thirdly, it is due to certain crystallochemical characteristics of apatite. The luminescence of natural apatite is due to the entry into its structure of ions of rare earth elements, which are in only one crystallochemical position of calcium [18]. There are intense bands  $TR^{3+}$  ( $Gd^{3+}$ ,  $Ce^{3+}$ ,  $Eu^{2+}$ ,  $Dy^{3+}$ ,  $Sm^{3+}$ ,  $Nd^{3+}$ ), as well as  $Mn^{2+}$  in the luminescence spectra of apatite. The luminescent properties of apatite have been studied in considerable detail [5, 7, etc.].

Table 1 shows the value of the intensity of luminescence centres in apatite from the rocks of different genesis.

Table 1

Intensity of luminescence centres in apatite

Apatite-bearing rocks	Luminescence centres						
	$Gd^{3+}$	$Ce^{3+}$	$Eu^{2+}$	$Dy^{3+}$	$Mn^{2+}$	$Sm^{3+}$	$Nd^{3+}$
Pyroxene rocks (31)*	$\frac{0-45^{**}}{10}$	$\frac{0-620}{86}$	$\frac{0-495}{141}$	$\frac{0-56}{16}$	$\frac{0-2800}{642}$	$\frac{0-940}{267}$	$\frac{0-80}{34}$
Calciphyres (10)	$\frac{0-40}{58}$	$\frac{0-300}{214}$	$\frac{0-850}{10}$	$\frac{0-22}{10}$	$\frac{55-2500}{438}$	$\frac{0-360}{99}$	$\frac{0-37}{17}$
Apatite-carbonate rocks (49)	$\frac{0-60}{3}$	$\frac{0-575}{120}$	$\frac{15-790}{120}$	$\frac{0-310}{15}$	$\frac{0-1620}{149}$	$\frac{0-2600}{193}$	$\frac{0-110}{30}$
Magnesium metasomatites (skarns) (7)	$\frac{0-8}{2}$	$\frac{0-178}{77}$	$\frac{37-210}{85}$	$\frac{2-10}{6}$	$\frac{18-635}{146}$	$\frac{0-60}{29}$	$\frac{0-10}{6}$
Pegmatites (28)	$\frac{0-17}{4}$	$\frac{0-100}{12}$	$\frac{42-329}{106}$	$\frac{4-46}{19}$	$\frac{120-8500}{2708}$	$\frac{0-1000}{130}$	$\frac{6-70}{21}$
Gabbro (11)	$\frac{0-87}{25}$	$\frac{0-1420}{253}$	$\frac{53-750}{225}$	$\frac{0-130}{32}$	$\frac{400-2300}{1170}$	$\frac{0-1820}{390}$	$\frac{0-110}{31}$
Pyroxenites (8)	$\frac{0-17}{5}$	$\frac{0-930}{321}$	$\frac{74-400}{229}$	$\frac{1-43}{18}$	$\frac{67-3840}{1248}$	$\frac{0-1500}{328}$	$\frac{0-80}{27}$
Carbonatites (107)	$\frac{0-11}{7}$	$\frac{0-351}{275}$	$\frac{0-700}{21}$	$\frac{3-44}{8}$	$\frac{30-1080}{737}$	$\frac{0-650}{48}$	$\frac{3-160}{11}$

\* Number of analyzes.

\*\* In the numerator – the limits of fluctuations, in the denominator – the average value.

Analysis of data from Table 1 testified as follows. Wide variations of the mean intensity were found for some luminescence centres:  $Mn^{2+}$  – from 146 in magnesium metasomatites to 2708 in pegmatites,  $Nd^{3+}$  – from 6 in magnesium metasomatites to 34 for pyroxene rocks,  $Ce^{3+}$  – from 12 in pegmatites to 275 for ultrabasic alkaline complexes with carbonatites. These data can serve as a preliminary criterion for the separation of apatite from different genetic types of rocks. More reliable information can be obtained through the use of factor analysis, which will be based on significant statistical material [7]. The factor diagram (Fig. 1), which is based on X-ray luminescence analysis, shows the figurative points of intensity of luminescence centres in apatite of different genetic types of rocks and the corresponding fields.

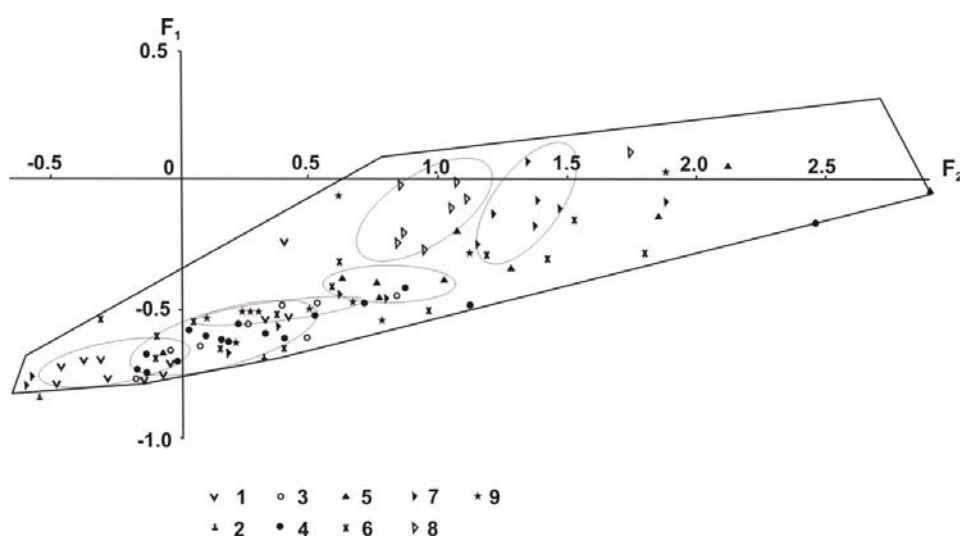


Fig. 1. Location of figurative points of luminescence centres intensity in apatite from ijolite-carbonatite complexes [7].

Apatite-containing rocks: 1 – ijolites; 2 – turjites; 3 – forsterites; 4 – apatite-forsterite-magnetite rocks; 5 – calcite-forsterite-magnetite rocks; 6 – calcite carbonatites; 7 – calcite-forsterite-magnetite rocks with tetraferriphlogopite; 8 – calcite carbonatites with tetraferriphlogopite; 9 – dolomite-magnetite rocks and dolomite carbonatites.

Figure 1 shows a clear trend: regardless of the genetic type of the analyzed rocks and their location (Kola Peninsula, Sette-Daban ridge in Yakutia), the fields of figurative points overlap, and all points fall into one large field bounded by the first and second factors. Therefore, according to X-ray luminescence, the studied apatite grains belong to carbonatite-bearing complexes. A smaller division of separate groups of figurative points showed that figurative points of apatite from earlier rocks (field of ijolites) are concentrated in the left part of the field, and the latest (field of calcite-forsterite-magnetite rocks with tetraferriphlogopite) – in the right one. Therefore, the intensity of the luminescence centres of rare earth elements in the apatite of each complex depends on the relative age of the mineral in consistently formed groups of rocks [7].

X-ray luminescence of apatite from carbonatite rocks was studied separately. Carbonatites and apatite-carbonate-magnetite rocks (ores) from the Kovdor deposit (Murmansk region) and carbonatites from the Yraas deposit (Maimecha-Kotui province, Yakutia) have been analyzed

(Table 2). It turned out that there are no significant differences in the values of the intensity of the centres of luminescence in apatite. Slightly lower luminescence intensity of  $\text{Eu}^{2+}$  and  $\text{Sm}^{3+}$  was recorded in apatite-forsterite carbonatites, and slightly lower intensity of  $\text{Eu}^{2+}$  luminescence – in apatite-carbonate-magnetite rocks (ores).

Table 2

Intensity of luminescence centres in apatite from carbonatites

Rocks	Luminescence centres						
	$\text{Gd}^{3+}$	$\text{Ce}^{3+}$	$\text{Eu}^{2+}$	$\text{Dy}^{3+}$	$\text{Mn}^{2+}$	$\text{Sm}^{3+}$	$\text{Nd}^{3+}$
Carbonatites, Kovdor	$\frac{0-25}{8}$	$\frac{0-750}{284}$	$\frac{0-168}{5}$	$\frac{3-28}{7}$	$\frac{172-1237}{682}$	$\frac{0-145}{38}$	$\frac{3-22}{9}$
Apatite-forsterite carbonatites, Kovdor	$\frac{4-9}{6}$	$\frac{86-270}{149}$	0	$\frac{3-8}{5}$	$\frac{594-1187}{843}$	0	$\frac{0-7}{4}$
Apatite-carbonate- magnetite rocks (ores), Kovdor	$\frac{4-25}{9}$	$\frac{86-925}{402}$	0	$\frac{2-17}{8}$	$\frac{128-1281}{832}$	$\frac{0-174}{57}$	$\frac{2-18}{10}$
Carbonatites, Yraas	$\frac{0-13}{4}$	$\frac{0-535}{185}$	$\frac{0-270}{17}$	$\frac{0-418}{34}$	$\frac{0-2001}{418}$	$\frac{0-234}{48}$	$\frac{0-43}{11}$

According to Tables 1 and 2, diagrams in the coordinates  $[\text{Ce}^{3+}/(\text{Ce}^{3+}+\text{Eu}^{2+})]$ – $[\text{Sm}^{3+}/(\text{Sm}^{3+}+\text{Mn}^{2+})]$  have been constructed (Fig. 2). In diagram 2, A, constructed according to Table 1, it is seen that each genetic type of apatite corresponds to a certain set of elements, and the concentrations of  $\text{Sm}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Eu}^{2+}$  and  $\text{Ce}^{3+}$  differ. The ratios of the intensity of X-ray luminescence centres in the mineral of apatite-carbonate rocks and pegmatites differ significantly. Instead, close geochemical formation conditions can be predicted for gabbro, magnesium metasomatites and carbonatites, as well as for calciphyres and pyroxenites. Therefore, using the values of the intensities of apatite X-ray luminescence, it is possible to predict the genetic type of the deposit. In diagram 2, B, constructed according to Table 2, we can see the narrow limits of fluctuations in the ratios of the intensities of apatite from carbonatites located in different areas. This suggests that they were formed under the same or similar geochemical conditions.

**Conclusions.** The peculiarities of the structure of apatite determine the concentration of many chemical elements in it, in particular rare earth elements, and affect their distribution in the mineral depending on the physicochemical parameters of the mineral formation environment. Therefore, apatite is considered a sensitive indicator of its formation conditions. The content of rare earth elements and X-ray luminescence of apatite are influenced by such factors as the chemical composition of the primary melt from which the rock has been crystallized, the degree of its differentiation, crystallization mechanism, saturation of the system with phosphorus, sequence and time of mineral crystallization, physicochemical parameters of magmatic system etc.

Studies of the X-ray luminescence of apatite from rocks of different genetic types indicate a certain difference in the intensities of the centres of luminescence in apatite of different genesis. This may serve as a preliminary criterion for the separation of apatite. Based on the data of factor analysis, we can assume that the intensity of the luminescence centres of rare earth elements in the apatite of each rock complex depends on the relative age of the mineral in the groups of rocks that have been successively formed.

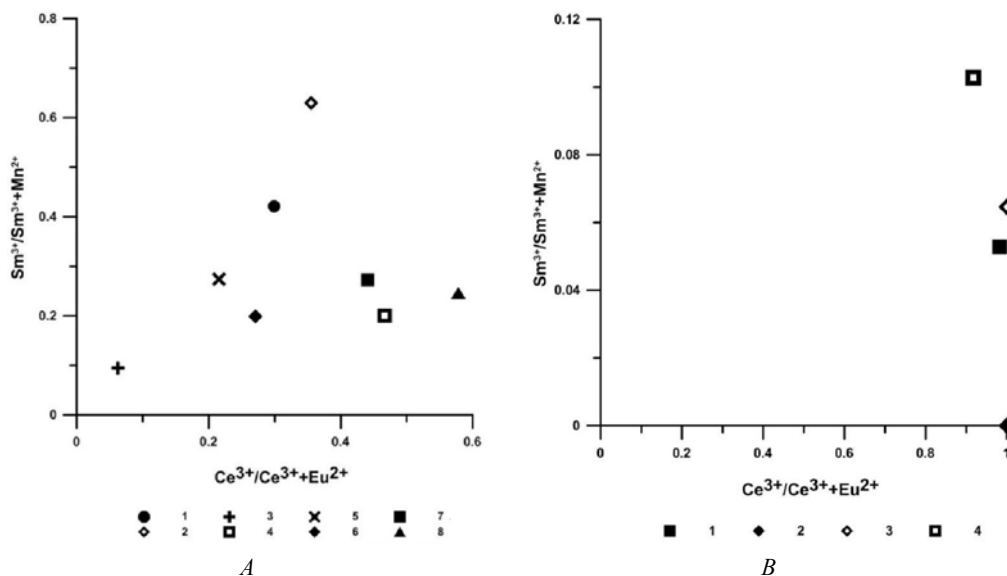


Fig. 2. Diagrams of the ratio of the X-ray luminescence centres intensities in apatite from different genetic type rocks (A) and carbonatites (B) in the coordinates of  $[Ce^{3+}/(Ce^{3+}+Eu^{2+})]-[Sm^{3+}/(Sm^{3+}+Mn^{2+})]$ .

A: 1 – pyroxene rocks; 2 – apatite-carbonate rocks; 3 – pegmatites; 4 – gabbro; 5 – calciphyres; 6 – pyroxenites; 7 – magnesium metasomatites (skarns); 8 – carbonatites; B: 1–3 – Kovdor deposit: 1 – carbonatites, 2 – apatite-forsterite carbonatites, 3 – apatite-carbonate-magnetite rocks (ores); 4 – carbonatites of the Yraas deposit.

Analysis of apatite from carbonatite rocks of different deposits showed minimal fluctuations in the ratios of X-ray luminescence intensity ( $Ce^{3+}$ ,  $Sm^{3+}$ ,  $Mn^{2+}$ ,  $Eu^{2+}$ ). This may indicate a close geochemical situation and, as a consequence, the only source of matter during the formation of carbonatites.

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### **ОСОБЛИВОСТІ РЕНТГЕНОЛЮМІНЕСЦЕНЦІЇ АПАТИТУ З КАРБОНАТИВ ТА МОЖЛИВОСТІ ЇХНЬОГО ВИКОРИСТАННЯ З РОЗШУКОВОЮ МЕТОЮ**

Досліджено особливості рентгенолюмінесценції апатиту з апатитонесних порід різного генетичного типу для з'ясування можливості застосування його як мінералу-індикатора умов рудоутворення. Вивчали апатит із карбонатитів, різноманітних апатит-карбонатних та ультраосновних порід, скарнів, пегматитів та інших утворень Українського, Алданського та Балтійського щитів, Прибайкалля, хребта Сетте-Дабан та Маймеча-Котуйської провінції Якутії. Для визначення інтенсивності люмінесценції характерних ізоморфних домішок апатиту ( $Gd^{3+}$ ,  $Ce^{3+}$ ,  $Eu^{2+}$ ,  $Dy^{3+}$ ,  $Sm^{3+}$ ,  $Nd^{3+}$ ,  $Mn^{2+}$ ) використано рентгенолюмінесцентний аналіз. Зроблено висновок, що значення інтенсивності люмінесценції  $TR^{3+}$  і  $Mn^{2+}$  в апатиті можна використовувати для визначення генетичного типу апатитонесних порід, типу мінералізації, умов мінералоутворення та з розшуковою метою.

*Ключові слова:* апатит, карбонатити, рентгенолюмінесценція, ізоморфні елементи-домішки, рідкісноземельні елементи.