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Vasyl Guliy, Natalia Bilyk

*Ivan Franko National University of Lviv,
4, Hrushevskoho St., Lviv, Ukraine, 79005,
vgul@ukr.net*

MAIN PECULIARITIES OF FLUID SYSTEMS UNDER APATITE FORMATION IN CARBONATE-BEARING ENDOGENOUS ASSOCIATIONS

Fluid systems of apatite from different carbonate-bearing endogenic association were studied by decrepitation analysis, high temperature chromatography analysis, and gas analysis of individual inclusions. Carbon and oxygen isotopic composition of different genetic types of the carbonate rocks have been used to determine main tendency of its formation and secondary alteration. The information about chemical composition of inclusions for the apatite and diopside from carbonatites, metamorphic rocks and scarns has been obtained. H₂O and CO₂ are main components of the inclusions from apatite from different associations. There is a great similarity between compositions of the inclusions in different kind of studied rocks.

Key words: apatite, carbonates, carbonatite, metamorphic apatite-carbonate rock, scarn, isotopic indicators, fluid, inclusion, mineral forming system, Precambrian.

Introduction. Apatite is widely spread in exogenous (for example, in phosphorites of different Phanerozoic sequences, or as authigenic mineral in sandstones of the Brest depression) and endogenous (for example, in kimberlites or Australian lamproites) formations, suggesting its stability in specious limits of depths, temperature and pressure.

Due to well extended isomorphic substitutions in different structural positions, apatite often put into practice as an indicator of conditions under mineral-forming processes to build of petrologic models and to construct a set of exploration and estimation criteria for industrial mineralization in different regions.

Peculiarities of apatite chemical composition in various formations of different origin, but with similar mineralogical compositions or such structural position within separate geological blocks, are very important also. Particularly, in carbonatites with simple mineralogical composition, where apatite likes endogenous carbonates plays important role, apatite-carbonate rocks within regional metamorphosed sequences, lime or magnesium scarns, main constituents of the rocks are apatite and carbonates with subordinated amounts of diopside, phlogopite etc.

At the same time, formal similarity of apatite-carbonate associations and often – analogous geological-structural position in geological sections led to uncertainty concerning geological processes, which are responsible for its formation. For example, in spite of investigations during almost century after determination as a separate group of carbonate rocks endogenous

in origin by W. Breger (1921) carbonatites have not yet precise terminological and classification definitely among various scientists. R. Mitchell [13] successfully shown present situation in this sphere by paraphrased sentence from H. H. Read – carbonatites and carbonatites and carbonatites... In his substantial review of peculiarities of the formations which are determined in various geological structures of the world as “carbonatites”, R. Mitchell suggested important role of CO₂, fluorine, water and other volatile components in formation of carbonatites, but he didn't give detail characteristic of relationships these components and specific differences of carbonatite compositions. In spite of skeptical attitude to anatectic “pseudocarbonatites” determined in different sectors of the Grenville Series [9, 10, 11] R. Mitchell prompted [13] its possible origin due to anatactic melts of the crust rocks, and suggested its feasible relationships to calc-silicate scarns or veins under significant role of fluids.

State of problem. Apatite is a permanent constituent of carbonate formations, which are endogenous in origin – carbonatites, apatite-bearing metacarbonate Precambrian rocks and Precambrian scarns. Commonly apatite in these rocks is accessory mineral, but sometimes its amounts are significantly high and such rocks can be determine as phosphorus or same additional industrial components ores. Attempts to determine perspectives of the carbonate rocks on different ores (rare metals, REE, apatite, phlogopite, etc.) with mineralogical and isotopic-geochemical criteria were unsuccessful. Taking into consideration important role of volatile components in fluids, we supposed its influence on ore specialization of investigated objects as well as on scale of mineralization.

To vagueness on origin of apatite-carbonate association in carbonatites as a whole noted above, we should add problems in its determination in connection to age of hosting massifs. Generalized model of formation the Phanerozoic carbonatites within the zone-concentric ultrabasic and alkaline complexes with carbonatites is popular [2], but for Precambrian linear carbonatites it is not so suitable because some rock components are not wide developed or even absent. As a result, there are substituted rock varieties (mainly silicate) not typical for the Phanerozoic complexes or determination of metamorphic carbonate rocks of basement as carbonatites. Such situation is typical for the Aldan Shield, where at the first stage of discovery of the Seligdar apatite deposit within metamorphic metasedimentary carbonate rocks of the Fedorovskaia Formation [1] shape of the main ore body have been determined as a pipe similar. In addition to this circumstance and due to formal similarity of apatite-carbonate associations of the rocks and some carbonatites idea about carbonatite origin of the Seligdar deposits was born [15]. The first isotopic data on carbon and oxygen in the Seligdar carbonates have been shown values typical for sedimentary carbonate rocks and the initial opinion about mantle origin of carbonate masses the author [15] transformed into a new variant of crust-mantle carbonatites. This idea is in accordance with models, which were proposed for the carbonatites of the Grenville Series in limits of primary carbonate metamorphic rocks transformation [9, 10, 11].

Little late, when exploration of apatite ores similar to the Seligdar deposit at the Aldan Shield was broaden, a number of new deposits composed by similar apatite-carbonate ores were founded, and lithological and stratigraphic controls of the mineralization have been established. New significant isotopic carbon and oxygen data for carbonates from the Seligdar deposits and new discovered objects were obtained [4]. Without any doubts, these data indicate sedimentary origin of studied apatite deposits at the Aldan Shield [4, 6].

Apatite-bearing primary sedimentary carbonate rocks of the Aldan Shield were affected in local manifestations by secondary alteration due to young episodes of tectonic-magmatic ac-

tivization similar to process in limits of the Grenville Series at the Canada Shield. Redistributions of the primary concentration of phosphorus were determined with appearance of new mineral phases including late carbonates. Betafite was founded in such locations and accepting of components from hosting rocks, particularly from titanite, which supplied Nb and Ta due to alterations [7]. Isotopic research of such formations has been shown later origin of carbonates if compare to carbonates from the main ore bodies and confirmed possibilities to determine origin and scale late formations described early [4].

Tendencies of isotopic carbonate and oxygen variations in different generations of carbonates similar to peculiarities of isotopic changes which were determined by D. Lentz for the Precambrian rocks at the Canadian Shield. He used these variations to create general model of transformation crust carbonate material to carbonatites in his opinion [9, 10, 11].

Given above information and analysis on the grounds of endogenous carbonates determination in addition to our previous data on specific composition of fluid phases in different genetic types of rocks [5] we involved to determine similarity and difference of apatite from carbonatites, metamorphic carbonate rocks and scarns. Very important moment from methodological position is to use the same lab instruments for our purposes as well as necessity to create geological and mineralogical basement for each investigated objects, which were studied, to determine scale of secondary transformations.

Initial geological materials and methods of investigation. We used for our purposes a set of samples from apatite-bearing rocks, which are regarded as models for different genetic kinds. Selected samples were grouped into three main types to characterize (1) carbonatites (apatite-carbonate ores of the Kovdor deposits [2] with some amount of silicates – diopside and phlogopite, and apatite-carbonate ores from the Gornoie Ozero carbonatite massif [2]), (2) apatite-carbonate metamorphic ores of the Seligdar type (Seligdar deposit, manifestations of the Chuga River at the Aldan Shield), and (3) Precambrian scarns of the Aldan Shield (Emeldzhak, Elkonka, Katalakh deposits), which were determined as products of magnesium metasomatism. In addition to these objects, we studied a number of deposits and manifestations from Finland (Silliniarvi), Ukraine (Nova Poltavka), Canada (Richardson Mine) etc. to get more information on mineralogical composition and peculiarities of formation of all studied genetic groups.

In spite of rich mineralogical composition of involved objects, for special lab analysis we used samples with simple apatite-carbonate or apatite-silicate associations (Fig. 1) to avoid of vagueness because of different phases with volatile components. Although there are a set of ideas on the Precambrian scarns formation, we did not check details of them and selected materials as apatite, calcite, diopside crystals within clear geological position in nests, veins and druses.

Analytical methods. After routine geological, petrographical and mineralogical observations, we prepared monomineral fractions of apatite, carbonates, diopside for future investigation of fluid inclusions, determination of fluid composition, and isotopic analysis. Firstly, we used heavy liquids to get initial lab samples, and then prepared final material after hand peaking with binocular microscope.

To get a general knowledge about fluid phase in analyzed apatite we involved decrepitation analysis with thermo-vacuum instrument DV-2. Heating of the samples (0.50–0.25 mm in diameter, similar weigh – 200 mg) put into quartz tubes were carried out from 100 up to 800 °C under vacuum. Detection of the effects conducted via pressure variations due to decrepitation of inclusions in mineral and generalized data are fixed at the final diagram.

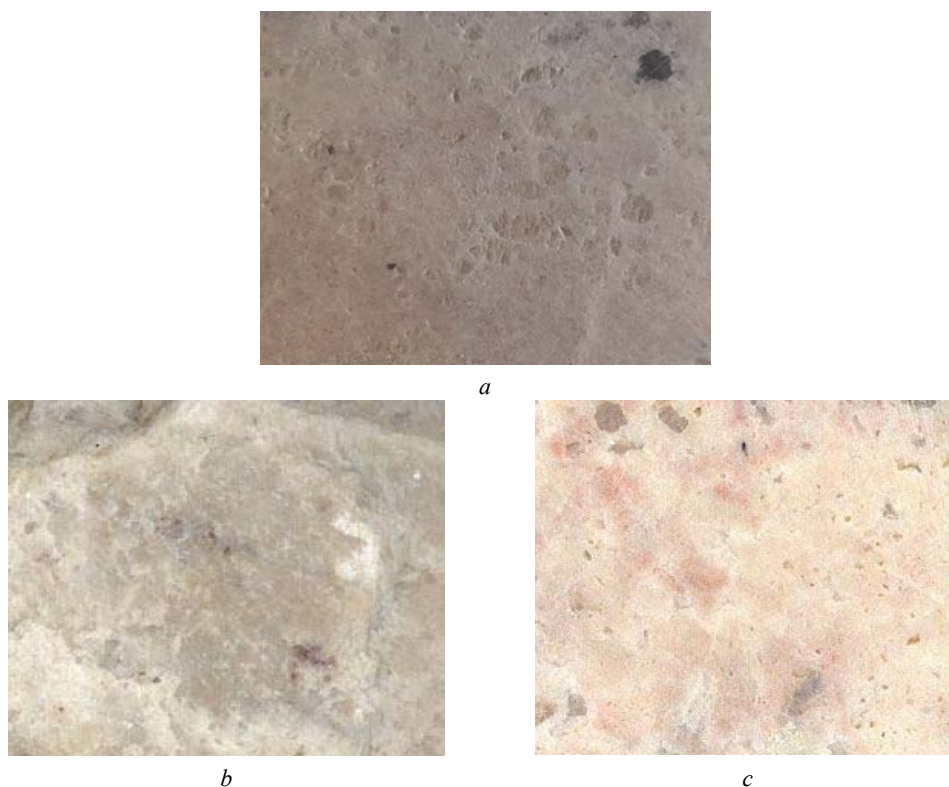


Fig. 1. Apatite-carbonate rocks of simple mineralogical compositions:

a – carbonatite Silliniarvi deposit (Finland); *b* – metamorphic rock of the Grenville Formation, Richardson Mine (Canada); *c* – metacarbonate rocks of Seligdar deposit (Aldan Shield).

Because there is a difficult in investigation of separate inclusions in apatite of different genesis due to small dimensions of the inclusions and bad quality of the material, we studied only separate samples to get general opinion about character of inclusions. Method of individual inclusions investigation has been used in order to characterize position of inclusions in mineral, its size, and future chemical analysis of inclusions.

Application of individual inclusion analysis has a number of limitations, and the most important is information on fluid phase only in limited points. To extract and analyze fluid inclusions in total volume of sample the method of high temperature gas chromatography developed by F. Letnikov [12] has been applied. Early expediency and precision of this method were under debates [12], and we should add some more arguments. Firstly, the method has used to analyze of rock samples, and such mixing materials could give some declination. We used during investigation only pure mineral samples, but we checked final results to avoid conversions joint components by correlation diagrams. Additional verification of the results we did with data of individual inclusion composition.

Taking into consideration possibilities to determine differences in origin of primary carbonate concentrations and development after secondary phases and often even late veins by isotopic composition of carbon and oxygen [4], we prepared a set of carbonate samples from carbonatites, metasedimentary carbonate apatite-bearing rocks and carbonates from different

scarns to supply additional information for diagnostic of carbonate transformations in various genetic groups of apatite-carbonate associations.

Oxygen and carbon isotopic compositions in the carbonate samples were determined in the Laboratory of Stable Isotopes (Institute of the Lithosphere of Marginal Seas, Moscow). Carbon dioxide was released from carbonates using the $PbCl_2$ method, proposed by Yu. Borshchevskii et al. (1974). Relative difference in oxygen and carbon isotope ratios in CO_2 (gas) was measured on the mass spectrometer Varian MAT-250. 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 $\delta^{13}C_{\text{samp}}$ (PDB) and $\delta^{18}O_{\text{samp}}$ (SMOW). The oxygen correction was taken into account for $\delta^{13}C_{\text{samp}}$ (PDB).

Obtained results and discussion. Data from decrepitation observations we used to build a set of diagrams for all intervals of analyzed temperatures from 100 up to 800 °C (Fig. 2). Almost for each observed samples peaks of the decrepitation activity were detected for both high (750–800 °C), and low (100–200 °C) temperature conditions.

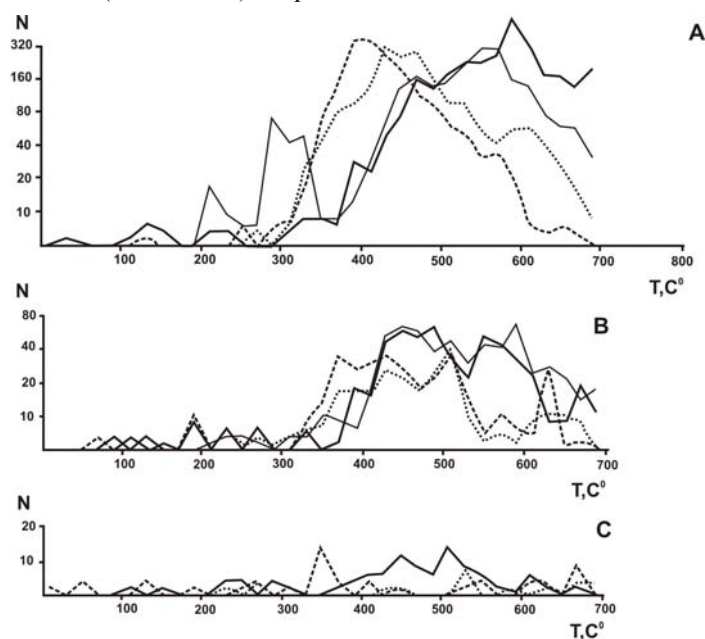


Fig. 2. Decrepitation curves of apatite from apatite-carbonate (A), apatite-silicate-carbonate (B), and apatite-silicate rocks (C).

There is a clear difference between apatite curves from carbonate and silicate variations of apatite ores after intensity of decrepitation activity and temperature levels (see Fig. 2). Apatite from silicate rocks is characterized by low approximately similar data of decrepitation activity along whole temperatures of analysis. At the same time, for apatite from carbonate rocks maximum of decrepitation activity lies in wide limits of temperature from 350 до 650 °C, but some variations are detected for every samples.

Changes of apatite decrepitation for the same rocks can be explained by influence of inclusion compositions, because under different temperature of mineral formation various in com-

positions inclusions have been led to different decrepitation activity. Results of thermic investigations of apatite show absence of influence of its physical properties on decrepitation activity. Therefore, composition of inclusions is a main factor of decrepitation activity of the mineral.

Size of gas inclusions in apatite is variable, and changes from 0.0n up to 0.00n mm (Table 1) and is not depends from composition of apatite-bearing rocks. At the same time, composition of gas phases is very changeable from absence up to 100 % of volume. Unusually high concentrations (sometimes up to 100 %) of nitrogen and rare gases were detected in apatite from diopside rocks. Similar high concentrations of these gases are fixed in diopside coexisting with apatite. In such cases, sizes of inclusions from apatite and diopside are similar.

Table 1

Results of quantitative analysis of gas phases compositions in individual inclusions in apatite from different type of the Precambrian carbonate-bearing rocks

Number of analyses	Diameter of bubble used in analysis, mm	Components, concentration volume %			
		H ₂ S, SO ₂ , SO ₃ , NH ₃ , HF, HCl, CO ₂	CO	H ₂	N ₂ + rare gases
Apatite-carbonate rocks					
Average (n = 17)	0.0307	54.3	15.0	11.1	32.6
Limits of data	0.0066–0.0786	19.0–100.0	7.5–28.0	5.5–24.0	11.0–81.0
Apatite-pyroxene rocks					
Average (n = 9)	0.0367	25.8	0.0	–	70.2
Limits of data	0.0150–0.0912	0.0–60.0	–	–	8.0–100.0
Scarns					
Average (n = 10)	0.0256	73.8	0.0	0.0	26.2
Limits of data	0.0090–0.0480	37.5–100.0	–	–	0.0–62.5

According to high temperature gas chromatography, H₂O is dominated component in apatite and diopside, and CO₂, CO, H₂, CH₄ were detected in less amounts (Table 2). Besides these components nitrogen as N₂ and rare gases were found also.

To analyze and compare inclusions compositions of apatite form different rocks we calculated coefficients of reducing and total gas indicator (see Table 2). Series of quantitative ratios between various phases (H₂/H₂O, CO/CO₂, etc.) have been used for discussion also.

Mixing fluid system with domination of sum of gases CO₂, CO, CH₄, H₂ under H₂O is a main characteristic of apatite in general. High contents of water in apatite from the Ukduska deposit are exceptions.

According to estimation of statistic parameters of distribution of components amount of water in inclusions is homogenous. Variations of values in general for apatite are less than its limits for separate genetic type. Maximum H₂O value (5.16 ml/g) is determined in apatite from the Ukduska deposit.

Limits of CO₂ values variations are more significant. For example, inclusions in apatite from the Seligdar deposit contain minimal value of CO₂ – 1.33 ml/g, and maximum – 13.6 ml/g. More high contents of CO₂ are common characteristic for apatite from apatite-carbonate association. But in some cases for apatite in association with silicates more significant CO₂ values has been determined (for example, at the Kovdor deposit up to 4.06 ml/g).

Table 2

Composition and characteristics of fluids in apatite
 after high temperature chromatographic analysis, ml/g

Location, deposits, rocks	Components						Coefficients*	
	H ₂ O	CO ₂	CO	CH ₄	H ₂	N ₂	C _r	Σ _{gas}
Kovdor, carbonatites	0.71	4.06	0.02	–	Traces	0.020	0.004	4.100
Gornoie Ozero, carbonatites	0.73	1.17	0.06	–	–	0.060	0.030	1.290
Oshurkovo, apatite-bearing diorites	0.86	0.44	0.94	–	–	–	0.720	1.380
Sliudianka, magnesian scarns	0.62	3.67	0.12	–	–	0.007	0.030	3.797
Katalakh, mag- nesian scarns	1.49	2.76	0.05	Traces	Traces	0.030	0.012	2.840
	1.87	1.30	Traces	–	–	0.040	0	1.340
Seligdar, apa- tite-carbonate rocks	1.68	13.60	0.11	–	Traces	–	0.007	13.710
	1.18	1.33	0.09	–	–	0.007	0.030	1.427
Chuga, apatite- carbonate rocks	0.42	0.19	–	–	–	–	0	0.190
Ukduska, apatite-diopside rocks	3.73	0.26	–	Traces	–	–	0	0.260
	0.33	0.22	Traces	–	Traces	–	0	0.220
	1.89	0.58	–	–	–	0.040	0	0.620
	5.18	0.38	0.20	–	0.12	0.060	0.006	0.640

* C_r = (H₂ + CH₄ + CO) : (CO₂ + H₂O); Σ_{gas} = CO₂ + CO + N₂.

Distributions of CO₂ values are relatively homogenous, although anomaly enrichment of CO₂ values is found in apatite from the Oshurkovo deposit.

Clear difference of inclusion compositions in apatite, if compare to diopside, is extremely low contents of hydrogen and CO, traces of methane and, accordingly, close to zero values of reducing coefficients (Fig. 3). H₂O and CO₂ are main oxidized phases in apatite (see Table 2). For H₂O (average 2.78, 1.43 and 1.68 ml/g) and CO₂ (average 1.02, 7.47 and 2.03 ml/g) are typical ratios close to 2:1. Nitrogen as a N₂ is a permanent constituent of the inclusions, but under low levels in limits 0.0n ml/g (see Table 2).

Obtained data on the isotopic composition of carbonate samples from the carbonatites, apatite-carbonate ores of the Seligdar and similar in composition and genesis deposits, and scarns are given in δ-values relative to the PDB standard for carbon and the SMOW standard for oxygen (Table 3). In the δ¹³C–δ¹⁸O coordinates the isotopic compositions of carbonates from layered apatite-carbonate rocks of all apatite deposits yield a scatter of δ¹⁸O values from +18.0 up to +23.4 ‰ without any depending from apatite resources.

The studied carbonates are characterized by positive δ¹³C values (up to +6.0 ‰, PDB) in sheeted apatite-carbonate bodies of the Seligdar deposit and its family. Carbonate from skarns of the Emeldzhak deposit is depleted in ¹³C and ¹⁸O (up to δ¹³C = –6.6 ‰, PDB, and δ¹⁸O = +14.0 ‰, SMOW). Similar, but more big in scale, depleted ¹³C and ¹⁸O values we determined for carbonates from the Precambrian linear carbonatites (see Table 3).

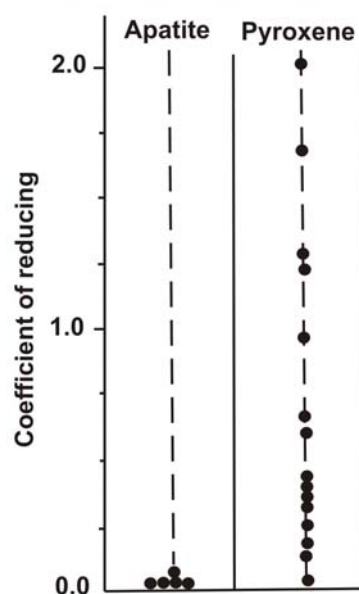


Fig. 3. Comparison of reducing coefficients for apatite and pyroxene.

Table 3

Isotope C and O composition of carbonates from different carbonate-bearing formations

Number of samples	Deposits	$\delta^{18}\text{O}$ ‰, SMOW	$\delta^{13}\text{C}$ ‰, PDB	Notes
8	Nova Poltavka, Dubravka	7.2–9.3	(-7.6)–(-4.7)	Linear Precambrian carbonatites
25	Seligdar, Niriandzha, Mustolaakh, Chukurdan	18.0–23.4	1.1–6.0	Layers of apatite-carbonate rock
6	Emeldzhak	13.5–14.0	(-3.8)–(-6.6)	Vein, nest and druse

Conclusions. Determination of decrepitation activity of apatite from carbonate and silicate mineral associations showed main difference in shape of generalize curves due to enrichment of the rocks by carbonate component. In spite of different genesis of the carbonate rocks forms of decrepitation activity (see Fig. 2, *A*) are similar in general for apatite from apatite-carbonate associations of the rocks. Increasing of diopside amount in apatite-carbonate-silicate varieties of the rocks led to decreasing of the decrepitation activity (see Fig. 2, *B*) up to four times, under keeping the same intervals for peaks. Apatite from mineral association poor in carbonates is characterized by the most low decrepitation activity (see Fig. 2, *C*). Last indicates important role of environment enrichment components, which are responsible for carbonate formation.

Chemical analyses of individual inclusions in apatite as well as total fluid phase determined by high temperature gas chromatography showed great similarity in quality composition of the fluids. Only quantity variations of the same components are important under forma-

tion of different mineralogical associations. There are not any specific “carbonatite” or scarns fluids, and only hosting geological environment is responsible for appearance of various mineralogical associations.

That is extremely significant for understanding diamond formation also [3, 14]. Early [6] we noted similarity of qualities composition of fluids in diamond, but recently idea about special new type of “diamond-forming” fluids was introduced [8].

Isotopic data indicate clear difference in nature of carbonate materials from whole three groups of the carbonate rocks. Carbonates from metamorphosed apatite-carbonate rocks are determined specific rich in heavy carbon and oxygen isotopes as signatures of a primary sedimentary protolith. Such data have been indicated apatite-carbonate rocks as a conservative system, which only locally was altered during late epochs of activation. CO₂-component is responsible for redistribution of primary carbonate materials, and due to isotopic investigation we can assumed about scale of mobilization by fluids of the primary formations. The extent to which fluids are involved in metamorphism (and metasomatism) is still in debate and constitutes one of the more important questions in understanding of regional metamorphism, metasomatism and formation of carbonatite melts.

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Василь Гулій, Наталія Білик

*Львівський національний університет імені Івана Франка,
вул. Грушевського, 4, Львів, Україна, 79005,
vgul@ukr.net*

ОСОБЛИВОСТІ ФЛЮЇДНИХ СИСТЕМ ПІД ЧАС ФОРМУВАННЯ АПАТИТУ В КАРБОНАТНИХ ЕНДОГЕННИХ АСОЦІАЦІЯХ

Наведено результати вивчення флюїдних систем в апатиті з різних карбонатних порід ендегенного походження на підставі даних декрепітаційного аналізу, високотемпературної газової хроматографії та газового аналізу окремих включень. Особливості хімічного складу апатиту з формацій різного генезису, однак подібного мінерального складу важливі з огляду на те, що мінерал є постійною складовою карбонатних утворень ендегенного походження – карбонатитів, докембрійських скарнів і апатитоносних метакарбонатних порід, а склад його головних компонентів та елементів-домішок часто успішно використовують як індикатор петрологічних процесів.

Формальна подібність апатит-карбонатних асоціацій, а часто й аналогічне геолого-структурне положення зумовлюють невизначеність щодо геологічних процесів, які відповідали за їхнє формування. Під час визначення подібних і відмінних рис апатиту з карбонатних порід різного генезису ми брали за основу дослідження флюїдної фази. Вивчали карбонатити (apatit-карбонатні руди Ковдорського родовища, які містять діюксид і флюгопіт, а також апатит-карбонатні руди карбонатитового масиву Горное Озеро), апатит-карбонатні метаморфічні руди типу Селігдар (Селігдарське родовище, прояви р. Чуга на Алданському щиті) та докембрійські скарни Алданського щита (родовища Емельджак, Ельконка, Каталах), які вважають продуктом магнезійного метасоматозу. Використано матеріали з низки родовищ і проявів Фінляндії (родовище Сіллінярві), України (родовище Ново-Полтавка) і Канади (рудник Річардсон).

Визначено подібність параметрів декрепітаційної активності апатиту з різногенетичних карбонатних порід. Виявлено відмінності в цій активності апатиту з карбонатних і силікатних різновидів апатитових руд. У разі збільшенні кількості діопсиду в апатит-карбонат-силікатних утвореннях декрепітаційна активність апатиту знижується до чотирьох разів. Найнижчу декрепітаційну активність має апатит з мінеральних асоціацій, збіднених карбонатами, а це засвідчує важливу роль процесу збагачення середовища компонентами, що відповідальні за утворення карбонатів.

За даними високотемпературної газової хроматографії, в апатиті й діопсиді, що з ним асоціює, переважають H_2O і CO_2 , у меншій кількості наявні CO , H_2 , CH_4 . Виявлено також N_2 і рідкісні гази. Результати хімічного аналізу окремих включень в апатиті та загальної флюїдної фази засвідчили значну подібність за якісним складом флюїдів.

Дані щодо ізотопного складу вуглецю й кисню використано для визначення головних тенденцій формування карбонатних мінеральних асоціацій та їхнього наступного перетворення. Отримані результати стосовно складу включень у мінералах з карбонатитів, метаморфічних порід і скарнів свідчать, що головними компонентами включень в апатиті є H_2O і CO_2 . Роль флюїдів такого складу у формуванні апатиту під час метаморфізму, утворення карбонатитових розплавів та метасоматозу подібна, тому нема відмінностей між апатитом з відповідних порід як продуктом цих процесів.

Ключові слова: апатит, карбонати, карбонатит, метаморфічна апатит-карбонатна порода, скарн, ізотопні індикатори, флюїд, включення, мінералоутворювальна система, докембрій.