



CHAROITE. EXPERIMENTAL STUDIES

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Abstract: The article provides an overview of experimental studies of charoite and charoite-containing rock formation hypotheses. The authors conducted experiments to clarify charoite and host rocks interaction and study charoite transformation processes at high temperatures. A series of experiments was aimed at improving the standard charoite samples. The experiments show the formation of polymineral reaction zones due to the contact interaction between charoite and microcline-arfvedsonite lamprophyre. By studying the newly formed phases, the authors reveal the distribution of elements by phases and establish their compositions. It is shown that thermal decomposition of charoite leads to the formation of wollastonite, which amount increases with the temperature increase from 800 to 1000 °C, and heating above 1200 °C leads to the formation of pseudowollastonite. The physicochemical simulation of charoite decomposition under the specified temperatures and pressure shows the following paragenesis: quartz, wollastonite, alkaline pyroxene (aegerine), microcline, rhodonite, and sphene.

The experiments prove the formation of charoite at low temperatures and the lack of silicate melt in the systems studied. The calculated values are consistent with the results of experiments conducted to study the charoite and host rocks interaction, which allows identifying phases potentially co-existing with wollastonite.

Special studies using by the coloring technique were conducted to improve the decorative properties of charoite. The color close to natural high-grade charoite coloration was achieved by keeping the rock samples in the active bright purple dye 4KT solution for 72 hours at a temperature of 70 to 90 °C.

Key words: charoite; Murun alkaline massif; experiment; fluid systems; physicochemical simulation; treatment

Recommended by E.V. Sklyarov

For citation: Marchuk M.V., Medvedev V.Ya., Ivanova L.A., Sokolova T.S., Danilov B.S., Gladkochub D.P. 2016. Charoite. Experimental studies. *Geodynamics & Tectonophysics* 7 (1), 105–118. doi:10.5800/GT-2016-7-1-0199.

ЧАРОИТ. ЭКСПЕРИМЕНТАЛЬНЫЕ ИССЛЕДОВАНИЯ

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Аннотация: Приведен обзор экспериментальных исследований чароита в России и в мире. Рассмотрены гипотезы формирования чароитовых пород. Для выяснения характера взаимодействия чароитового субстрата с вмещающими породами и исследования процессов преобразования чароита при повышенных температурах проведен ряд экспериментов. Кроме того, изучены возможности повышения сортности некондиционных

чароититов. При экспериментальном исследовании контактового взаимодействия чароитового субстрата с лампрофиром микроклин-арфведсонитового состава установлено образование реакционных полиминеральных зон. Исследование новообразованных фаз позволило изучить распределение элементов по фазам и получить их составы. Показано, что при термическом разложении чароита происходит его преобразование в волластонит, количество которого увеличивается с ростом температуры от 800 до 1000 °С, при нагревании выше 1200 °С образуется псевдволластонит. Согласно проведенному физико-химическому моделированию разложения чароита, из различных составов минерала при заданных температурах и давлении образуется следующий парагенезис: кварц, волластонит, щелочной пироксен (эгирин), микроклин, родонит и сфен.

Проведенные эксперименты подчеркивают низкотемпературный характер чароитообразования и показывают отсутствие силикатного расплава в исследованных системах. Результаты расчетов согласуются с проведенными экспериментами по взаимодействию чароита с вмещающими породами и его отжигу и позволяют определить возможные сосуществующие с волластонитом фазы.

Исследования по улучшению декоративно-художественных свойств чароититов методом окрашивания показали, что для получения окраски чароититов, близкой к окраске высокосортных природных образцов, требуется выдержка образцов в растворе активного ярко-фиолетового красителя 4КТ при длительности 72 ч и температуре 70–90 °С.

Ключевые слова: чароит; Мурунский щелочной массив; эксперимент; флюидные системы; физико-химическое моделирование; облагораживание

1. INTRODUCTION

Charoite is a unique mineral called “the lilac miracle of Siberia” [Rogova et al., 2013]. It is considered “the main mineralogical discovery of the second half of the 20th century” [Solyanik et al., 2008]. This rare gemstone has become widely known and recognized in the global market of ornamental stones due to its aesthetic and processing properties, such as unique lilac to purple coloration, structural features and high strength. The only currently known charoite deposit is Sirenevyi Kamen’ (Lilac Stone) located in the Murun alkaline massifs in the northwestern part of the Aldan Shield at the border of the Irkutsk region and Yakutia (Russian Federation) (Fig. 1). Reportedly, charoite has been recently discovered in the marine sediment of Kalpakam, the southeast coast of India [Deepthi et al., 2015].

The Sirenevyi Kamen’ deposit includes both charoite and charoite-containing rocks, as well as eluvial boulders of charoite [Bondarenko, 2009]. The charoite composition is $(K, Sr, Ba, Mn)_{15-16}(Ca, Na)_{32}[(Si_{70}(O, OH)_{180})](OH, F)_4 \cdot nH_2O$, and its structure is highly complex [Rozhdestvenskaya et al., 2010]. The main charoite chemical components are K, Na, Ca, Si, and H_2O ; the secondary components are Ba, Sr and Mn; the presence of traceable amounts of Fe, Mg, Al, Ti, Zr and Th is noted. Charoite is an alkaline calcium silicate with tubular Si-O-radicals. Its crystalline structure is quite specific, like that of other minerals of this group (kanasite, tinaksite, frankamenite, mizerite etc.) discovered in the Murun charoite mineralization field. Over 50 minerals are closely associated with charoite, and the latter does not occur in nature in its “pure” form [Konev et al., 1996; Solyanik, 2004; Vorobjev, 2008].

The geological structure of the Murun massif and a wide variety of rare and unique formations are factors contributing to the inconsistency of scientific views on the charoite mineralization origin. Today opinions differ on the conditions and mechanisms of the formation of charoite rocks [Bulakh, 1984; Biryukov, Berdnikov, 1993; Evdokimov, 1995; Konev et al., 1996; Mitchell, 1996; Vladykin, 2000, 2009; Reguir, 2002; Vorobjev, 2008]. It is assumed that the Murun massif was formed in the unstable tectonic conditions [Konev et al., 1996], as evidenced by numerous dykes of varied composition and strong foliation of igneous rocks. At the same time, there was a lack of tectonic control during the formation of the charoite field, and the charoite bodies intruded and dissected other rocks. However, conformable bedding of charoite veins and veinlets is also common. Probably, the intrusion of fluidized melt may be accompanied by shock decompression of the melt [Medvedev et al., 2014] and the formation of structures typical for torgolite disintegration. Some researchers support the hypothesis of the metasomatic origin of charoite in the exocontact halo of the Malomurun alkaline massif under the influence of fluids enriched with Sr, Ba, Mn, F, CO_2 , and alkalis [Biryukov, Berdnikov, 1993; Vorobjev, 2008; Bondarenko, 2009]. The metasomatic origin is viewed as a result of the interaction between the fluidized melt and the host rocks of different compositions, or the interaction between hydrothermal solutions and the host rocks, or the interaction between alkaline intrusions and carbonate host rocks. There is a hypothesis about the initial fluidized melts [Konev et al., 1996], which assumes that charoite is similar to pegmatites in the morphology of the veins, mineralogical diversity, large- and giant-grain struc-

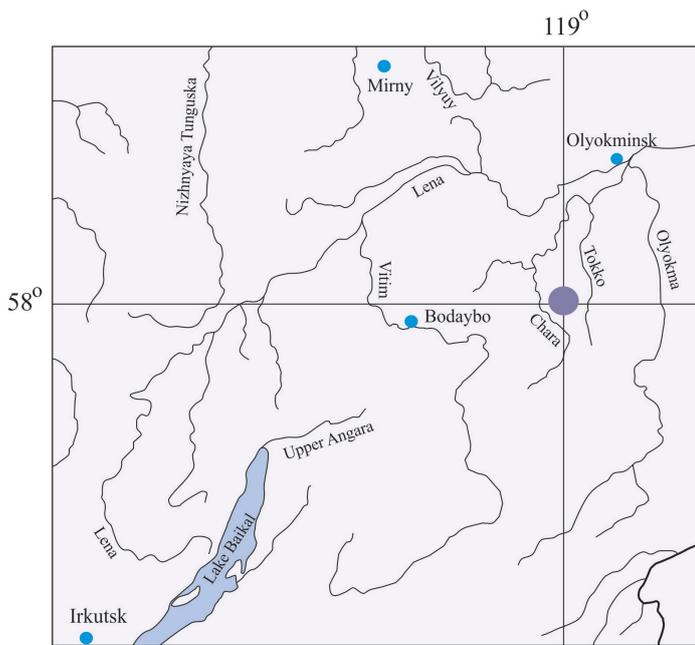


Fig. 1. The Sirenevi Kamen' deposit location map (marked by the lilac circle).

Рис. 1. Схема района расположения месторождения «Сиреневый камень» (отмечено сиреневым кружком).

ture, and relationships with the host rocks. According to [Vladykin, 2000] charoite is formed by the fractionation of the alkali melt. In [Vorobjev, 2008], the genesis of charoite is also considered as a combination of magmatic and metasomatic processes. Some researchers propose charoite and its paragenetic minerals can be classified as late-stage hydrothermal minerals. It is assumed that charoite, being formed in potassium-rich environments, is an equivalent of high-sodium silicates of late-stage mineral associations [Bulakh, 1984; Borisov, 1985; Evdokimov et al., 1985].

So, the challenge of the charoite formation involves a number of genetic problems, such as initial magma composition determining, magma crystallization and differentiation mechanisms, features of certain rocks types genesis in Murun alkaline complex and others. The wide complex of geochemical, mineralogical and other methods is employed in studies aimed at solving these problems, and an important component of such studies is thermodynamic simulation using experimental methods.

2. EXPERIMENTAL STUDIES REVIEW

Systematic experimental studies of charoite have been limited recently both in Russia and abroad due to a number of reasons, including the fact that the genesis of charoite mineralization remains unclear, and the

charoite rocks chemical composition and structure are unique. The concretion and intergrowth of phenocryst minerals (including quartz, microcline, aegerine, tinaksite, frankamenite, arfvedsonite, apophyllite etc.) are common in charoite, and experiments with a pure charoite material are thus challenging.

Termobarogeochemical studies conducted in the 1980s suggested the charoite rock crystallization from the residual melt fluid at temperatures ranging from 400 to 750 °C [Lazebnik et al., 1977]. The study of inclusions in charoite rocks was conducted by A.A. Borovikov at the Institute of Geology and Mineralogy, SB RAS, and revealed the charoite crystallization from the melt fluid at a temperature of about 800 °C. However, these results require confirmation by studies engaging the most typical charoite morphological varieties in the system.

The temperature of the dehydration reaction of charoite was established by the thermogravimetric analyses reported in [Lazebnik et al., 1977; Matesanz et al., 2008; Földvári, 2011] – thermograms clearly show two endothermal effects in the temperature ranges from 210 to 480 °C and 940 to 960 °C, when charoite loses water (the water content may amount to 5 wt. %) [Földvári, 2011]. The first temperature range is typical of the samples with high water contents, the second is the same for all the samples. According to [Lazebnik et al., 1977], these effects are due to the fact that the charoite samples contain water of three types: molecular water with strong intermolecular bonds, molecular water with weak intermolecular bonds, and loosely bound hydroxyl groups. Molecular water with weak intermolecular bonds is removed from charoite when it is heated to 300 °C, and the remaining water is removed in the temperature range from 900 to 1000 °C during the structural destruction of the mineral [Konev et al., 1996].

According to [Janeczek, 1991], the charoite structure was destroyed at a temperature of 970 °C, and only a crystalline phase – wollastonite – was diagnosed by the high-temperature X-ray method at 1000 °C. The formation of charoite was simulated in the experiments described in [Zaraisky, 2007] – the rock samples from the Malomurun massif (quartzite sandstone, dolomite, quartzite-richterite, and aegerine-K-feldspar fenite) were treated by alkaline solutions of potassium at T=550 °C and P=1 kbar. The experiments show that charoite does not form in dolomites, quartzite-richterite rocks and fenites. A mineral similar to charoite was obtained in the paragenesis with aegirine and potassium feldspar due to the experiment with the host quartzite sandstone in the alkaline solution containing K, Na, Ca, Ba, Sr, Fe, Al, Fe, Mn, F, and CO₂.

Charoite stability conditions depending on the chemical composition of the mineral-forming environment were studied by D.S. Glyuck [Vorobjev, 2008] in expe-

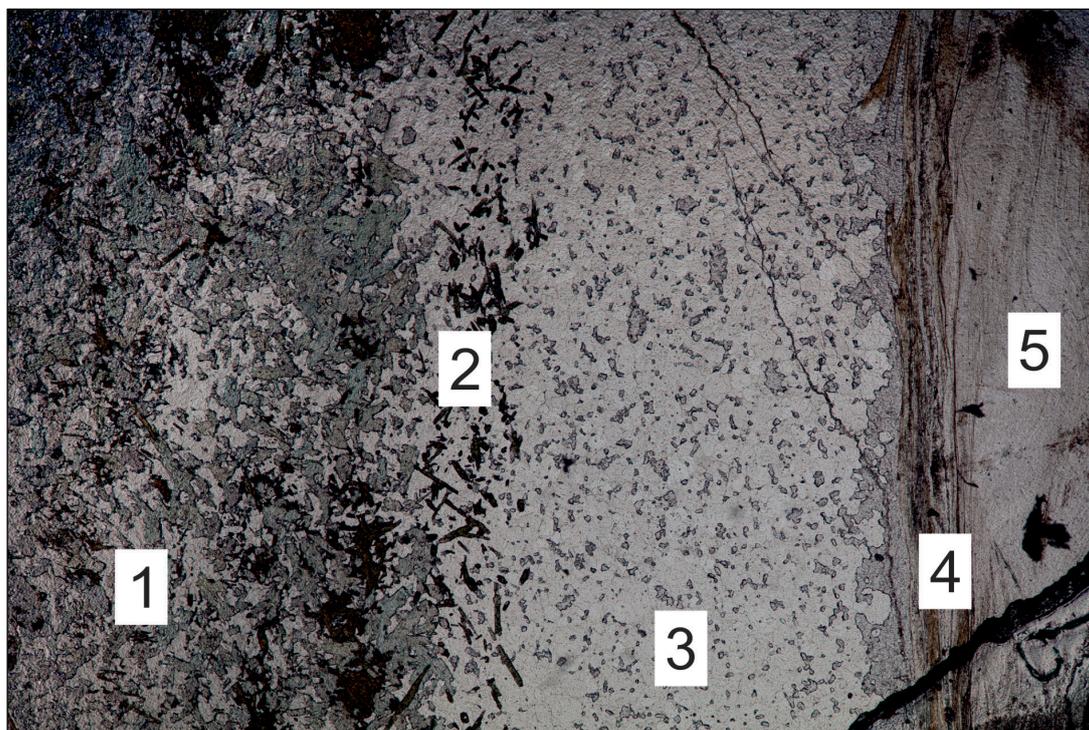


Fig. 2. The charoite melt and lamprophyre interaction contact zone. The sample was mined from the Staryi site.

Thin section; parallel nicols; the viewing field size is 7.2×4.8 mm. 1 – lamprophyre (arfvedsonite + microcline); 2 – aegerine; 3 – microcline; 4 – charoite interbedded with apophyllite; 5 – charoite.

Рис. 2. Контактная зона взаимодействия чароитового расплава с лампрофиром в образце с участка «Старый».

Шлиф, николи параллельны, поле зрения 7.2×4.8 мм. 1 – лампрофир (арфведсонит + микроклин); 2 – эгирин; 3 – микроклин; 4 – чароит с прослойками апофиллита; 5 – чароит.

periments conducted in specific isothermal-isobaric conditions ($T=550\text{ °C}$; $P=1\text{ kbar}$). In all the experiments, silicate melt was formed. However, neither the alkali concentration nor the composition of the mixture used in the experiments were published, and a description of the silicate melt composition was not provided.

A number of studies [Konev et al., 1996; Vorobiev, 2008] considered the successive transformation of host rocks xenoliths in charoitites. It was assumed this interaction has an effect on charoite rocks. It was noted at a temperature of 500 °C and a pressure of 1 kbar, a reaction rim was formed only in high alkalinity conditions. Such a transformation is evidenced by a sample from the Staryi (Old) site of the Sirenevyy Kamen' deposit. The sample shows charoite and lamprophyre contact and the reaction zone of the rocks. The contact zone looks as follows: lamprophyre (arfvedsonite + microcline) – aegerine – microcline – charoite interbedded with apophyllite – charoite (Fig. 2).

Such interactions raise a number of questions: what was the range of temperatures and pressures when the process was carried out, what was the fluid regime, and whether there was a charoite melt or melts of other compositions in this case.

3. EXPERIMENTAL STUDY OF THE CHAROITE STABILITY IN VARIOUS T–P CONDITIONS AND THE CHAROITE AND HOST ROCKS INTERACTION

Our study is focused on the high temperature charoite transformation, and the charoite and host rocks interaction. Our experiments are conducted using samples of charoite (with microcline and quartz impurities) and potassium K-feldspar-amphibole lamprophyre from the Staryi site of the Sirenevyy Kamen' deposit. Average chemical compositions of the initial samples are specified in Table 1.

Charoite transformation at high temperatures is studied in the experiments by heating the samples to temperatures from 800 to 1200 °C (at intervals of 50 °C) at the atmospheric pressure. The charoite samples (length up to 15 mm, and thickness from 2 to 5 mm) are put into microcrucibles made of aluminum oxide (or zirconium oxide in high-temperature experiments above 1200 °C), placed in the high-temperature furnace to be heated to the specified temperature, and kept for one hour in the specified conditions. Afterwards, the samples are tempered in air, for three minutes from 1200 to 400 °C, and for seven minutes from 400 to 100 °C.

Table 1. Average compositions of original samples

Таблица 1. Средний состав исходных образцов

Charoite substance		Lamprophyre		Charoite substance		Lamprophyre	
wt. %		wt. %		ppm		ppm	
SiO ₂	73.22	60.51		V	40	506.7	
TiO ₂	0.02	0.52		Cr	7	219	
Al ₂ O ₃	0.54	6.32		Ni	7	158.8	
Fe _{total}	0.17	4.78		Cu	160	690.6	
MnO	0.13	0.18		Zn	53	94.3	
MgO	1.84	11.22		As	21	105.5	
CaO	16.10	3.88		Y	60	15.7	
SrO	0.47	0.13		Zr	55	6.9	
BaO	2.02	0.04		La	231	34.7	
Na ₂ O	3.90	1.91		Ce	394	41.1	
K ₂ O	8.14	8.57		Nd	307	6.6	
P ₂ O ₅	0.02	1.25		W	8	2.2	
F	1.10	0.67		Pb	110	574.3	
S	0.05	0.05					
Cl	0.004	0.008					

On exposure at the samples for one hour at 800 °C, a charoite with reduced interplanar spacings is identified. When the sample is heated to 850–900 °C, two phases – charoite and wollastonite – are observed, and the charoite content is gradually decreasing from 90 to 80 %. At a temperature of 950 °C, the content of charoite drops abruptly to trace values. This experiment confirms the destruction of the charoite structure at temperatures above 900 °C and supports the research results published in [Janeczek, 1991]. Above 1000 °C, both the triclinic and monoclinic modifications of wollastonite are observed. When the samples are heated to 1100 °C, wollastonite and its high-temperature modification called pseudo-wollastonite are formed. When the charoite sample is subject to heating above 1200 °C for one hour, charoite is completely transformed into pseudo-wollastonite (Fig. 3). The charoite structure is similar to tinaksite or kanasite, while its silicon-oxygen framework corresponds to calcium silicates, and the charoite structure transformation into the lattice of triclinic wollastonite can be thus explained [Janeczek, 1991]. In our experiments, no charoite melt was formed.

Charoite and host rocks interaction. We studied the charoite and microcline-arfvedsonite lamprophyre interaction as a model of the natural transformation of host rock xenoliths in the charoite substance [Vorobjev, 2008].

In the experiments, a three-gram charoite sample (with microcline and quartz impurities) is crushed down to powder. The powder is put into a platinum capsule (9–10 mm in diameter; volume of 7–9 cm³). A lamprophyre sample (0.3–0.5 g) is added into the same capsule. The fluid composition in the capsule changes from pure H₂O to 85 % (NaHCO₃ + KOH) or (NaOH +

KOH) solution. The K/Na ratio in the system is 1.375 as calculated from the average ratio of these elements in charoite from the Staryi site. In the experiments at temperatures in the range from 500 to 800 °C and pressures from 0.5 to 1 kbar, the alkali content varies from 0 to 5 % in the (KOH + NaOH, KOH + NaHCO₃) solution. The initial charoite substance is bleached, and a recrystallized charoite (with traces of calcite) is observed on the lamprophyre surface. When the solution concentration reaches 10 %, no interaction takes place at the charoite powder and lamprophyre contact at 500 °C; however, charoite is partially transformed into wollastonite. When the solution alkalinity is raised to 50 % (KOH + NaOH), lamprophyre is transformed, and

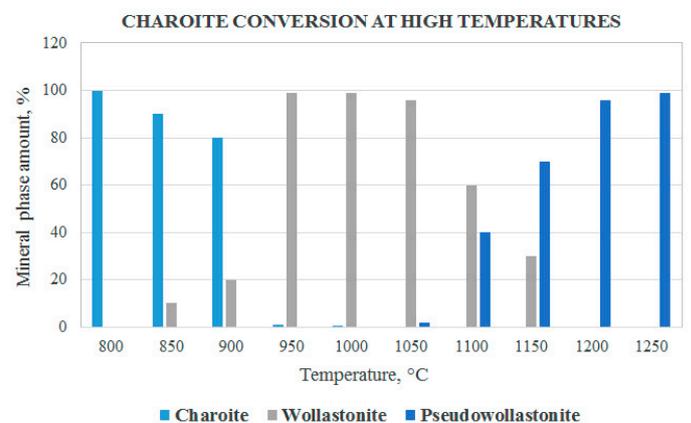


Fig. 3. The sample composition changing as a function of temperature.

Рис. 3. Изменение состава образца в зависимости от температуры.

sodium-containing phlogopite is formed, while charoite remains stable, even though its recrystallization takes place at the contact with lamprophyre. Increasing the (NaOH + KOH) alkalinity to 85 % results in the partial assimilation of lamprophyre to form fluorphlogopite, while charoite remains stable. This series of high-alkalinity experiments show a dense opaque (translucent in some cases) reaction zone formed at the lamprophyre and charoite substance contact. The zone contains microcline, calcite, and dolomite with traces of amphibole, and its widths amounts to 1.6 mm.

With the temperature increase up to 800 °C, the pressure of 1 kbar, and 10 % (KOH + NaHCO₃) solution, charoite is transformed in areas located at a distance from the contact with lamprophyre, and the recrystallization zone composed of wollastonite, mica phase and microcline is formed. At the charoite and lamprophyre contact, we observed the reaction zone containing wollastonite in the form of transparent columnar crystals (0.40–1.20 mm × 0.15 mm), microcline and recrystallized charoite with calcite traces, amphibole and aevilite, Ca₃(SiO₃·OH)₂·2H₂O. The reaction metasomatic column (Fig. 4, a, b) is quite similar to the natural one (see Fig. 2). We identified compositions of the newly formed phases and analyzed the distribution of elements. Various barium- and strontium-containing phases occur in some areas (Table 2).

4. THERMODYNAMIC SIMULATION

Analyses of the experiment results failed to determine the phase composition of the fine-grained reaction rim, and we attempt to employ the physicochemical simulation in order to establish a possible mineral association formed during the high-temperature decomposition of charoite. In order to identify the mineral phases formed under charoite annealing and coexisting with wollastonite, we use the Selector software [Chudnenko, 2010]. A model of charoite decomposition reaction is constructed for the temperature range from 700 to 900 °C at the pressure of 1 kbar. The 18-component model system (K, Na, Ca, Fe, Mg, Al, Si, Ti, Mn, Sr, Ba, P, F, Cl, S, C, O, and H) corresponding to the charoite chemical composition is used as a basis for calculations covering 116 potentially possible phases, including solids (minerals), gas and an aqueous solution. We used the thermodynamics databases from [Holland, Powell, 2011] and [Yokokawa, 1998] in calculations for minerals and simple substances. The gas phase data are from [Reid et al., 1977]. The aqueous phase data are from SUPCRT98 electronic database. The published chemical analysis data for charoite (Table 3) are used to construct the model. In the simulation, an excessive amount of H₂O (55.51 mol) is added to the charoite composition, which ensures the

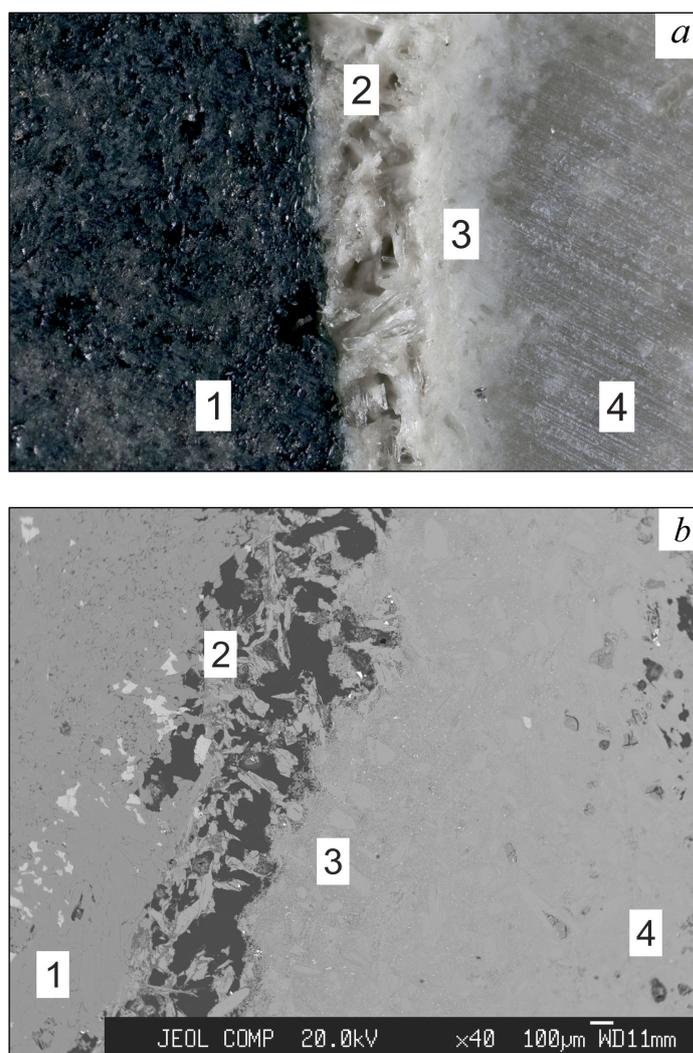


Fig. 4. The reaction rim resulting from the charoite substance and lamprophyre interaction in experimental conditions (T=500°C; P=1 kbar; 10 % concentration (NaHCO₃+KOH)).

a – the viewing field size is 7.5×5.1 mm; b – the image is taken in the backscattered electrons (zoom 40×). 1 – lamprophyre (arfvedsonite + microcline); 2 – wollastonite; 3 – wollastonite with microcline and charoite; 4 – charoite.

Рис. 4. Реакционная кайма, образовавшаяся при взаимодействии чароитового субстрата и лампрофира в экспериментах при T=500 °C, P=1 кбар, 10%-концентрации (NaHCO₃+KOH).

a – поле зрения 7.5×5.1 мм; b – снимок сделан в обратно рассеянных электронах, увеличение ×40. 1 – лампрофир (арфведсонит + микроклин); 2 – волластонит; 3 – волластонит с микроклином и чароитом; 4 – чароит.

permanent presence of the liquid phase, and a small amount of CO₂ (0.01 mol) is added to provide the conditions for potential formation of carbonates.

Our calculations show (Table 4) that at the given temperature and pressure, the following paragenesis is

Table 2. Composition of newly-formed phases (wt. %)

Таблица 2. Состав новообразованных фаз (вес. %)

Wallastonite		Microcline		Amphibole		Apatite	
SiO ₂	52.719	Na ₂ O	0.952	Na ₂ O	5.703	F	7.769
CaO	47.281	MgO	1.335	MgO	18.074	CaO	42.591
		Al ₂ O ₃	14.421	Al ₂ O ₃	0.000	SrO	11.243
		SiO ₂	61.754	SiO ₂	57.408	P ₂ O ₅	38.397
		K ₂ O	13.026	K ₂ O	4.914		
		CaO	1.073	CaO	3.261		
		TiO ₂	0.064	TiO ₂	0.334		
		MnO	0.681	MnO	0.079		
		FeO	5.292	FeO	10.227		

formed from charoite of various compositions: quartz, wollastonite, alkaline pyroxene (aegerine), microcline, rhodonite, and sphene. Besides, manganese oxide, barium carbonate and hydroxide, BaTiO₃, as well as BaSiO₃ and SrSiO₃ may be present in small amounts.

According to the simulation results, in the entire temperature range from 700 to 900 °C, the main stable products of the charoite decomposition reaction are quartz and wollastonite. In case of the charoite decomposition at 900 °C, the content of wollastonite can reach 40 mol. % (Fig. 5, a). Quantities of other mineral phases are insignificant and variable depending on the initial composition of charoite: pyroxene – up to 4.5 mol. %; microcline – up to 1 mol. %; rhodonite – up to 0.17 mol. %; sphene – up to 0.02 mol. %; and manganese oxide, barium carbonate and hydroxide, BaTiO₃, BaSiO₃ and SrSiO₃ – 3.5 mol. % in total. Figure 5, b,

shows the ratio of minerals (mol. %) formed from the charoite substance used in the experiments aimed to studying the charoite and host rock xenoliths interaction [Marchuk *et al.*, 2014]. It should be noted that the presence of the diopside component in pyroxene is typical of the given composition (column 4 in Table 3) as the content of MgO is higher than that in the composition of pure charoite (columns 1 to 3 in Table 3). Sphene and BaSiO₃ are also stable phases under the simulation conditions for this composition.

In the study of the charoite substance and lamprophyre xenoliths interaction, the experiments show that, in addition to wollastonite, traces of microcline and alkali phases of different compositions are contained in the reaction zone. Thus, the calculation results are consistent with the results obtained in the experiments aimed at studying the charoite and host rocks interac-

Table 3. Chemical composition of charoite (wt. %)

Таблица 3. Химический состав чаройта (вес. %)

Component	1	2	3	4
SiO ₂	58.50	57.31	58.94	73.22
TiO ₂	0.02	0.02	0.14	0.02
Al ₂ O ₃	0.03	0.01	–	0.54
Fe ₂ O ₃	0.14	–	–	–
FeO	–	0.02	0.01	0.17
MgO	–	–	–	1.84
MnO	0.10	0.35	0.23	0.13
CaO	21.90	21.01	21.12	16.10
SrO	–	1.43	0.64	0.47
BaO	2.53	0.65	3.22	2.02
Na ₂ O	2.26	2.29	2.62	3.90
K ₂ O	10.01	8.35	9.48	8.00
P ₂ O ₅	0.03	–	–	0.02
F	0.33	0.53	0.49	0.11
Cl ₂	0.02	–	–	–

Note. 1 – composition of charoite according to [Matesanz *et al.*, 2008]; 2 – composition of charoite according to [Rozhdestvenskaya *et al.*, 2010]; 3 – average composition of charoite according to five analyses in [Wang *et al.*, 2014]; 4 – composition of the charoite substance in Table 1.

Примечание. 1, 2 – состав чаройта из работ [Matesanz *et al.*, 2008] и [Rozhdestvenskaya *et al.*, 2010] соответственно, 3 – средний состав чаройта по пяти анализам из работы [Wang *et al.*, 2014], 4 – состав чаройтового субстрата из табл. 1.

Table 4. Products of high-temperature decomposition of charoite (mol. quantity)

Таблица 4. Продукты высокотемпературного разложения чаройта (мол. кол-во)

Mineral phases	Temperature, °C											
	700	800	900	700	800	900	700	800	900	700	800	900
	1			2			3			4		
Quartz (SiO ₂)	0.4942	0.5476	0.5550	0.4837	0.5343	0.5408	0.5122	0.564	0.5715	0.6517	0.7072	0.7224
Wollastonite (CaSiO ₃)	0.3905	0.3905	0.3905	0.3746	0.3746	0.3746	0.3766	0.3766	0.3766	0.2267	0.2267	0.2267
Aegirine (NaFeSi ₂ O ₆)	0.0017	0.0017	0.0017	0.0003	0.0003	0.0003	0.0001	0.0001	0.0001	0.0022	0.0022	0.0022
Diopside (CaMgSi ₂ O ₆)	-	-	-	-	-	-	-	-	-	0.0428	0.0428	0.0428
Microcline (KAlSi ₃ O ₈)	0.0006	0.0006	0.0006	0.0002	0.0002	0.0002	-	-	-	0.0096	0.0096	0.0095
Rhodonite (MnSiO ₃)	-	0.0014	0.0014	-	0.0049	0.0049	-	0.0032	0.0032	0.0017	0.0017	0.0017
Sphene (CaTiSiO ₅)	-	-	-	-	-	-	-	-	-	0.0002	0.0002	0.0002
Bixbite (Mn ₂ O ₃)	0.0007	-	-	0.0025	-	-	0.0016	-	-	-	-	-
BaCO ₃	0.0008	-	-	0.0040	-	-	0.0008	-	-	-	-	-
Ba(OH) ₂	0.0154	0.0162	0.0162	-	0.0040	0.0040	0.0184	0.0193	0.0193	-	-	-
BaTiO ₃	0.0002	0.0002	0.0002	0.0002	0.0002	0.0002	0.0017	0.0017	0.0017	-	-	-
BaSiO ₃	-	-	-	-	-	-	-	-	-	0.0123	0.0123	0.0123
SrSiO ₃	-	-	-	0.0138	0.0138	0.0138	0.0062	0.0062	0.0062	0.0042	0.0042	0.0042

Note. Columns 1, 2, 3, and 4 according to Table 3.

Примечание. Колонки 1, 2, 3, 4 – аналогичны Табл. 3.

tion and charoite annealing, which suggests that there are phases coexisting with wollastonite in the reaction rim during the charoite decomposition.

5. CHAROITE TREATMENT

The specific natural lilac to purple charoite coloration is due to impurity ions Mn²⁺ и Mn³⁺ in the mineral structure [Nikolskaya et al., 1976]. In this respect, Mn³⁺ has the main role. Besides, low concentrations of electron-hole centers put a slight effect on the color. The mineral is fluorescent in ultra-violet and green spectra in the photographic and X-ray excitation [Yarovoy, 1983]. According to [Nikolskaya et al., 1976], the charoite sample can be completely discolored by annealing in the reducing conditions at a temperature of 700 °C for three hours. The coloration can be restored by gamma irradiation, but become less intensive. After annealing in oxidizing conditions, coloration does not change. Subsequent gamma irradiation does not lead to any significant change in coloration. The methods of artificial mineral color changing in different redox environments [Medvedev, 1983] have provided a unique opportunity to track changes in the charoite chromophore and fluorescent characteristics [Yarovoy et al., 1987] in a variety redox conditions.

Charoite with high contents of impurity minerals looks less colourful, while its monomineral samples are recognized as highly decorative in terms of colors and patterns. Color shades depend on impurity minerals: honeylike (tinaksite), yellow (tokkoite), brown (titanite), black (aegirine), greenish (pectolite and microcline), and whitish (calcite and apofillite). Colouring with chemical agents is one of the techniques proposed to improve the colour-related characteristics of charoite [Ivicheva et al., 1998; Aleksandrova, Ivanova, 2015]. In this technique, charoite is impregnated with a selected colorant, and this process is accompanied by chemical reactions in the pore space of the rock sample. Options of using inorganic chromophores and synthetic dyes have been also studied. The technique ensures the color stability that is confirmed by corrosion tests and sufficient depths of the coloured matter, from the surface staining (for massive variations) to complete penetration of the dye (in samples with fiber and puckered structures).

Methods aimed at strengthening of charoitites are based on the use of high- and low-concentration dispersions and inorganic adhesives [Ivicheva et al., 1998]. Two types of inorganic adhesives can be used – soluble 'liquid' glass and silica sol obtained by the hydrolysis of tetraethoxysilane. Strengthening of porous samples is ensured by gradual (and, in some cases, repeated)

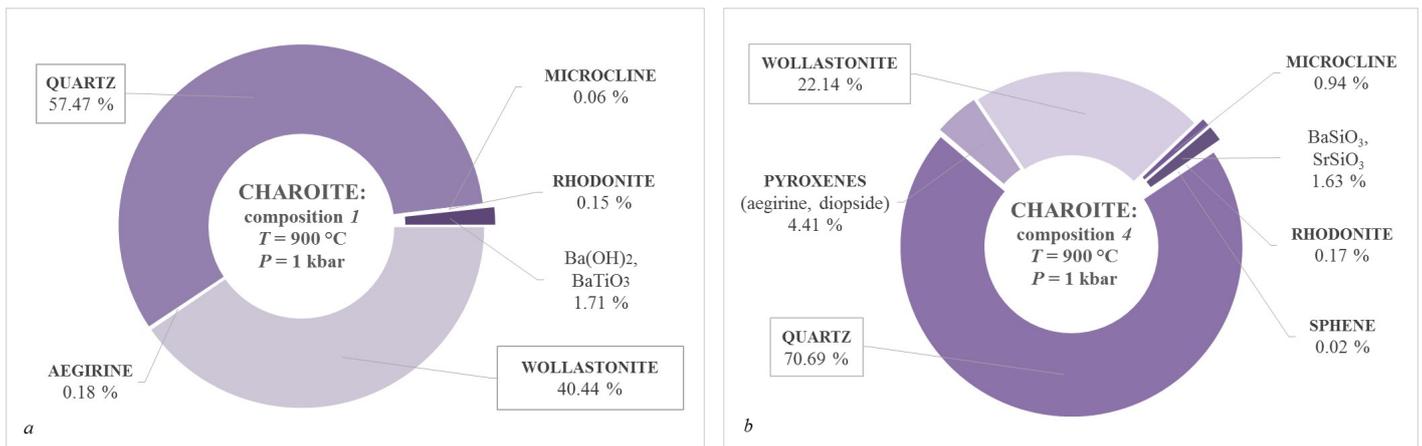


Fig. 5. Charoite high-temperature decomposition products (mol. %). The figure shows results of the physicochemical simulation by the Selector software. The simulation conditions are given in the figure. Chemical compositions of charoite 1 (a) and 4 (b) are given in Table 1.

Рис. 5. Продукты высокотемпературного разложения чаройта (мол. %) при физико-химическом моделировании с помощью ПК «Селектор». Условия моделирования приведены на рисунке. Химический состав чаройта 1 (a) и 4 (b) приведен в Табл. 1.

soaking of the sample rock with organosilicon, which is followed by heating, and modifying and hardening agents may also be used.

6. IMPROVING DECORATIVE PROPERTIES OF SUBSTANDARD CHAROITE SAMPLES

Coloring of charoitites samples is a complex multi-stage process, including sample preparation for coloring, coloring itself, fixing the result and testing the color stability against effects of water, alkaline solutions, sunlight and mechanical processing. The process cycle is 480–960 hours. The experiments are carried out in a temperature range from 20 to 100 °C. Charoitite samples preparation includes cleaning and surface degreasing by a soap-alcohol solution or acetone to provide for deeper penetration of the dye into the sample and to ensure the coloration uniformity.

Charoitites with different tones, structures, textures and mineral composition have been studied. Charoite contents in the samples varies from 20 to 70 %. Results of our experiments shows an optimal dye for charoite-containing rocks is synthetic – active bright purple dye 4KT, C₂₀H₁₄N₃O₁₅S₄Na₃Cu. By the type of the chromophore system, this dye is classified in the group of azo-dyes characterized by the presence in the molecule of -N=N- azogroup, connecting aromatic and heterocyclic compounds residues with each other and with the residues of compounds having active CH₂-group. Preliminary experimental studies have shown unacceptability of acetic acid as a reaction accelerator, as the coloring becomes unstable to any impact. In the experiments

with the sample treatment in the dye solution for 12–24 hours, only the near-surface layer (maximum 10 mm thick) is colored. To achieve optimal coloration (Fig. 6), the samples are kept in the dye solution for 72 hours, the temperature is raised to 70–90 °C, and NaCl is added to the 3–7 % aqueous solution of active bright purple dye 4KT as electrolyte in order to accelerate the diffusion.

7. RESULTS AND CONCLUSIONS

The charoite system experimental study overview emphasizes the need to develop more comprehensive and detailed experimental techniques for studying the conditions of the fluid-melt formation associated with the formation of charoite rocks.

It is established that in case of thermal decomposition, charoite is transformed into wollastonite which amount gradually increases with the temperature increase to 900 °C and, once the temperature is raised to 950 °C, grows dramatically to 95 % of the original sample weight. From the temperature of 1000 °C, both triclinic and monoclinic wollastonite modifications are observed. Further temperature increasing leads to the pseudo-wollastonite formation. Polymineral reaction zones of complex compositions are formed in case of the interaction between charoite and K-feldspar-amphibole lamprophyre, as revealed in the experiments using the samples from the Murun massif. Possibly, the difference in compositions of the natural and experimental interaction zones is due to discrepancy between the temperature parameters in the experimental and

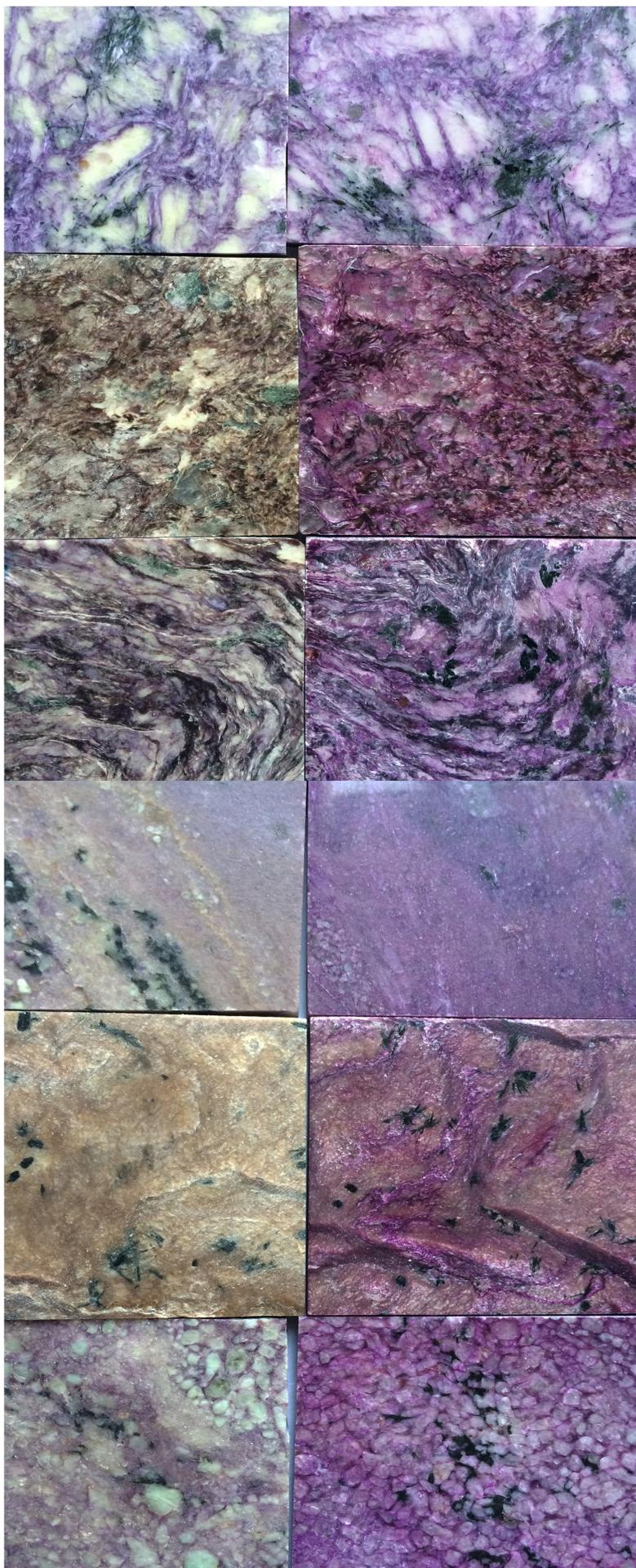


Fig. 6. Charoite samples: initial view (top row); the same samples after coloring (bottom row).

Рис. 6. Общий вид исходных образцов чароитов (верхний ряд), общий вид образцов после окрашивания (нижний ряд).

natural conditions and differences in the fluid composition, wherein the K/Na ratio may change at lower temperatures (about 500 °C). According to [Bondarenko, 2009], variations in the charoite compositions are fully similar to features of the alkaline trend of the Murun alkaline complex. For the early-stage charoite (metasomatic stage, high temperatures), K-component of the mineral-forming environment is considerable. The chemical activity of Na is enhanced with the decreasing charoite formation temperature. In nature, wollastonite is commonly associated with pectolite and charoite. Therefore, we can consider wollastonite is a typical rock associated with charoite, which crystallization conditions are similar. There is an opinion about the charoite formation by substituting wollastonite [Lazebnik et al., 1977]. The mineralogical and petrographic studies of charoitites show the possibility for the assimilation of lamprophyre xenoliths in the charoite substance with a specific distribution of pyroxene and K-feldspar [Vorobjev, 2008]. Based on the analysis of the charoitites and host rocks interaction in the natural and experimental conditions, it becomes possible to clarify the T–P conditions for the charoite formation. In particular, the experiments emphasize the low-temperature character of charoite formation and show a lack of silicate melt in the systems studied. The results obtained by the physicochemical simulation of the charoite decomposition reaction give grounds for the following conclusions: wollastonite and quartz together make up more than 97 mol. % of the total mass of

charoite; alkali mainly occur in the solution and, in small amounts, are present in aegirine and microcline; after the charoite decomposition is complete, manganese, strontium and barium form their own phases, which quantities are insignificant and would be even less if the model would take into account more complex schemes of isomorphism in minerals.

Charoite coloring by an active dye can be applied as a modern treatment method for the off-grade samples coloration and improving the colorimetry parameters of the material being processed, and can enhance the operational capabilities of the deposit. In our experiments, most of the samples acquired richer and attractive lilac and purple coloring, close to the natural charoite raw material.

8. ACKNOWLEDGEMENTS

The study was conducted using facilities of Centre for Geodynamics and Geochronology at the Institute of the Earth's Crust, SB RAS, Irkutsk, Russia by analysts Z.F. Ushchapovskaya and T.Yu. Cherkashina. The electronic probe X-ray analyses were performed in Vinogradov Institute of Geochemistry SB RAS by analyst L.A. Pavlova

The study was supported by the Russian Foundation for Basic Research (Grant Baikal No. 14-45-04108) and the Government of the Irkutsk Region.

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