# Rhonite in Molten Inclusions from the Olivine of Allivalite Nodules from Malyi Semyachik Volcano and Basalts of Klyuchevskoi Volcano, Kamchatka

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Abstract—A microprobe study of olivine found in allivalite nodules from lavas discharged by Malyi Semyachik Volcano and of olivine phenocrysts from basalts discharged by Klyuchevskoi Volcano revealed the presence of rhonite as a daughter mineral, for the first time ever. Rhonite was found in small  $(10-50 \, \mu m)$  grains that are xenomorphic in intergrowths with other minerals and have regular crystallographic outlines in contact with glass. We also found high-alumina clinopyroxene, chromium-free spinel, and hornblende. Residual glass is distinguished by its higher concentrations of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, alkalies, and by lower concentrations of FeO, MgO, and CaO. The chemical composition of the rhonite we studied is characterized by limited variations of the major components and fits the formula  $(Si,Al)_6(Ti,Al,Fe^{+3},Fe^{+2},Mn,Mg)_6(Ca,Na)_2O_{20}$  well. The data points in the composition of the rhonites we have studied lie in the overall field of much more variable compositions to be found in the field. Unlike the previous findings in alkaline and subalkaline rocks, the rhonite we describe in the present report was found in rocks of the tholeitic and calc-alkaline series.

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# **INTRODUCTION**

A microprobe study of olivine from allivalite nodules found in lavas of Malyi Semyachik Volcano and of olivine phenocrysts from basalts discharged by Klyuchevskoi Volcano revealed rhonite as a daughter mineral.

Rhonite is a silicate, a mineral of the enigmatite group, its name derives from the Rhoen massif in Germany. It is in the triclinic system and is characterized by the general formula  $X_2Y_6Z_6O_{20}$ , where X = Na, K, Ca; Y = Ti,  $^{VI}Al$ ,  $^{VI}Fe^{+3}$ ,  $Fe^{+2}$ , Mn, Mg; Z = Si,  $^{IV}Al$ , <sup>IV</sup>Fe<sup>+3</sup>. Rhonite is found as a rare earth in the form of subphenocrysts and microlites in alkali basalts, teschenites, limburgites, tephrites, and phonolites, as well as replacing amphibole xenocrysts and as an interstitial phase in rhonite xenoliths. The mineral associations that are typical of it contain titanaugite, kaersutite, diopside, forsterite, spinel, perovskite, magnesioferrite, and titanomagnetite [Mineral ..., 2001]. Rhonite was described as a daughter mineral in melt inclusions found in olivines of basanites in the North Minusinsk Basin and in southern Israel [Timina et al., 2006; Sharygin, 2002], in pyroxenes from lunar soil brought by the Luna 24 mission [Treiman, 2008], and in pyroxenes in the Haute Loire massif, France [Babkine et al., 1964]. The most complete reviews of rhonite can be found in [Johnston, Stout, 1985; Sharygin et al., 2007].

We discovered rhonite when studying melt microinclusions in olivines from olivine—anorthite (Ol—An) inclusions (allivalites) in a continuously differentiated flow (from basalt to andesite) discharged by Malvi Semvachik Volcano [Selvangin, 1979] and in basalts discharged by the Predskazannyi Vent on Klyuchevskoi Volcano [Khrenov et al., 1985]. The Ol-An inclusions consist of coarse-grained aggregates of olivine  $(Fo_{75-80})$  and anorthite  $(An_{90-95})$  with some stratification and transitions to eucrite. The grains of olivine are as large as 5 mm, while those of anorthite are up to 10 mm in size. The interstitial space frequently contains some amount of residual melt, which is usually crystallized to reach the stage of a fine-grained, porous, dolerite-like aggregate [Selyangin, 1979]. The rhonite-bearing olivines in the high-alumina basalts discharged by the Predskazannyi Vent have the composition  $Fo_{84-86}$ , are as large as 2.5 cm across, and are probably xenocrysts [Khrenov et al., 1985] or were produced by disintegration of lava-affine deep-seated cumulates.

The melt inclusions are ellipsoidal or irregular in shape, are from a few microns to 0.2–0.3 mm across, and usually have undergone partial crystallization. The daughter minerals include high-alumina clinopyroxene, spinel, amphibole, and rhonite. The glass in these inclusions is frequently penetrated by very thin

**Table 1.** Mean rhonite compositions

Sample #	LS16	LS20	LS26-28	KL119-3	KL119-5				
Number of analyses	umber of analyses 3		17	2	2				
$SiO_2$	25.51	25.45	25.78	29.06	26.35				
TiO <sub>2</sub>	5.17	4.82	6.05	3.80	3.19				
$Al_2O_3$	18.87	18.94	18.54	18.67	17.77				
$Fe_2O_3$	13.27	12.53	9.64	8.94	17.20				
FeO	13.29	14.02	14.88	17.09	9.53				
MnO	0.18	0.18	0.18	0.17	0.18				
MgO,	11.49	11.36	11.34	10.81	13.34				
CaO	12.56	12.49	12.41	11.82	11.96				
Na <sub>2</sub> O	0.65	0.45	0.59	0.99	0.75				
Total	101.00	100.23	99.42	101.33	100.26				
Formula-based amounts per 20 oxygen atoms									
Si	3.4096	3.4299	3.4908	3.8470	3.5157				
Ti	0.5198	0.4882	0.6159	0.3779	0.3196				
Al	2.9735	3.0091	2.9595	2.9128	2.7953				
Fe <sup>+3</sup>	1.3352	1.2711	0.9827	0.8902	1.7272				
Fe <sup>+2</sup>	1.4850	1.5797	1.6850	1.8918	1.0640				
Mn	0.0204	0.0204	0.0205	0.0183	0.0208				
Mg	2.2895	2.2813	2.2892	2.1327	2.6541				
Ca	1.7993	1.8041	1.8007	1.6765	1.7101				
Na	0.1676	0.1163	0.1556	0.2528	0.1932				
Mg#	0.61	0.59	0.58	0.53	0.71				
Total	14.0000	14.0000	14.0000	14.0000	14.0000				

Table 2. Theoretical rhonite formulas

Sample #	Formula
LS16	$(Si, ^{IV}Al)_6(Ti, ^{VI}Al, Fe^{+3}, Fe^{+2}, Mn, Mg)_{6.03}(Ca, Na)_{1.97}O_{20}$
LS20	$(Si, {}^{IV}Al)_6(Ti, {}^{VI}Al, Fe^{+3}, Fe^{+2}, Mn, Mg)_{6.08}(Ca, Na)_{1.92}O_{20}$
LS26-28	$(Si, ^{IV}Al)_6(Ti, ^{VI}Al, Fe^{+3}, Fe^{+2}, Mn, Mg)_{6.04}(Ca, Na)_{1.96}O_{20}$
KL119-3	$(Si, {}^{IV}Al)_6(Ti, {}^{VI}Al, Fe^{+3}, Fe^{+2}, Mn, Mg)_{6.07}(Ca, Na)_{1.93}O_{20}$
KL119-5	$(Si, {}^{IV}Al)_6(Ti, {}^{VI}Al, Fe^{+3}, Fe^{+2}, Mn, Mg)_{6.10}(Ca, Na)_{1.90}O_{20}$

dendrites of minerals and contains bubbles of a gas phase.

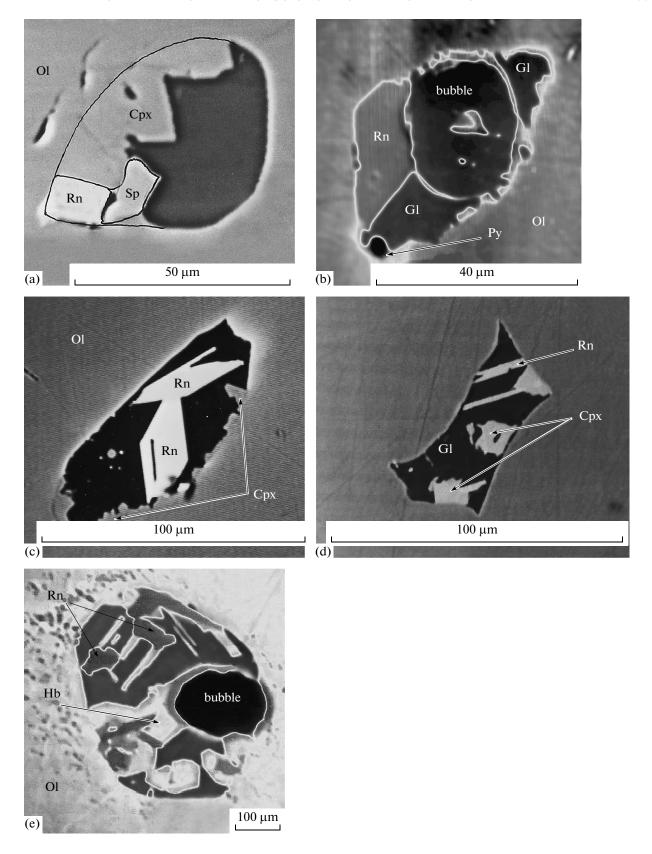
Rhonite is found as fine grains ( $10-50 \, \mu m$ ) that are xenomorphic in intergrowths with other minerals and that have regular crystallographic outlines at contacts with glass (Fig. 1). Although we have examined many melt inclusions, rhonite was found in few of these.

# **RESULTS**

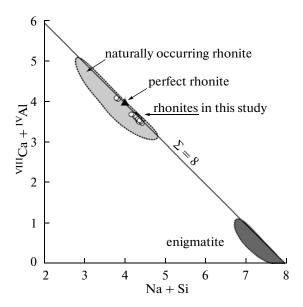
The chemical composition of the rhonite we have studied (Table 1) shows limited variations

(Si,Al)<sub>6</sub>(Ti,Al,Fe<sup>+3</sup>,Fe<sup>+2</sup>,Mn,Mg)<sub>6</sub>(Ca,Na)<sub>2</sub>O<sub>20</sub>, (Table 2). The data points lie in the general field of much more variable compositions of this mineral when it occurs naturally [Johnston and Stout, 1985] (Fig. 2).

Clinopyroxenes were encountered either in small crystals or in the rims at the olivine—melt boundaries. The pyroxenes typically have high alumina concentrations, which occasionally reach 16–17% Al<sub>2</sub>O<sub>3</sub>, low concentrations of silica (as low as 38%), and higher concentrations of titanium. Earlier studies by one of the present authors [Anan'ev, 1985; Anan'ev and Shnyrey, 1984] classified such high-alumina phases



**Fig. 1.** Microprobe photographs of rhonite-bearing inclusions: (a) sample LS16, (b) sample LS20, (c) sample LS26-28, (d) sample KL119-3, (e) sample KL119-5. Ol stands for olivine, Cpx for clinopyroxene, Hb for hornblende, Sp for spinel, Rn for rhonite, G1 for glass, and Py for pyrite. Images are in backscattered electrons. Images a, b, and c were subjected to additional treatment in order to identify the boundaries of mineral phases.



**Fig. 2.** A plot of Na + Si versus  $^{VIII}$ Ca +  $^{IV}$ Al for naturally occurring rhonite and enigmatite after [Johnston and Stout, 1985].

with very low concentrations of SiO<sub>2</sub> (38%) as garnets since they could not be studied optically. Stoichiometry does not distinguish between such pyroxenes and garnets and calculations show almost completely identical compositions of these phases with the garnet formula Si<sub>6</sub>(Ti,Al,Fe<sup>+3</sup>)<sub>4</sub>(Mg,Fe<sup>+2</sup>,Ca)<sub>6</sub>O<sub>24</sub>. However, such pyroxenes were also encountered in melt inclusions that did not contain rhonite, with the overall trend being continuous with Al<sub>2</sub>O<sub>2</sub> concentration. going from 3% to 17%, and having complementary variations in the concentration of SiO<sub>2</sub> that are not typical of garnet. Similar pyroxenes were also described elsewhere [Grib and Perepelov, 2008; Plechov et al., 2008; Sharvgin, 2002]. In addition, pyroxenes that were as high-alumina as the ones above (12% Al<sub>2</sub>O<sub>3</sub>) have been described as mineral impregnations in basalts from Auvergne, Eifel, and the Oslo area [Dobretsov et al., 1971], as well as in some alkaline and subalkaline rocks of Kamchatka [Volynets et al., 1990; Savel'ev and Filosofova, 2005].

Spinel is found as a high-alumina chrome-free variety with a comparatively low concentration of trivalent iron. Hornblende has a higher concentration of aluminum and forms both small dendrites that permeate the glass in the inclusions like felt and mature crystals.

The glass in these inclusions is a differentiate of the primary melt that has remained when the daughter minerals crystallized. It has higher concentrations of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and alkalies and lower concentrations of FeO, MgO, and CaO (Table 3).

It should be noted that nearly all previous finds of rhonite were in alkali rocks. The rhonite described in the present report was found in rocks of the tholeittic and calc-alkaline series that consist of high-alumina basalts [Khrenov et al., 1985], as well as of high-mag-

nesium basalts and andesites, whose continuous series was formed by fractionation of the allivalite—eucrite association and by mixing with dacite magma at the top of the magma column [Selyangin, 1979, 1987].

# **DISCUSSION OF RESULTS**

Numerous studies of rhonite-bearing mineral associations suggest a stable paragenesis of rhonite and high-alumina clinopyroxene [Timina et al., 2006; Sharygin, 2002; Kogarko et al., 2005; Magonthier and Velde, 1976 and elsewhere], while the other accompanying phases are characterized by more variable compositions and are not always present in these associations. Another stable feature to use to infer the presence of rhonite in a melt microinclusion is that the residual acidic glass should be high in alumina.

On homogenizing, the cryometry of rhonite-bearing melt inclusions in olivine extracted from alkaline rocks [Sharygin, 2002; Sharygin and Timina, 2008] in which gas bubbles contained liquid  $CO_2$  gave the result that the melt inclusions were captured in cores of olivine phenocrysts at  $T > 1300^{\circ}C$  and P > 3-5 kbars. The following sequence of phase generation was identified:  $\pm$  sulfide globule > Al-spinel (1320–1255°C > rhonite (1260–1180°C) > clinopyroxene (1240– 1130°C > apatite > ±amphibole, Fe-Ti-oxides (ilmenite or Ti-magnetite) (>1100°C) > glass (>1100°C). The glass in homogenized inclusions [Sharygin, 2002] is characterized by high concentrations of FeO (14.4–21), MgO (4.5–7.5), CaO (8.5–11),  $TiO_2$  (2.3–4.5), and  $P_2O_5$  (0.7–0.9) and by low concentrations of  $SiO_2$  (43.3–46.9),  $Al_2O_3$  (9–14), and  $Na_2O + K_2O$  (2.6–5.5 wt %). The residual glasses in the original (unheated) inclusions are substantially more acidic (in wt %):  $SiO_2$  (60.5–61),  $TiO_2$  (0.2–0.3),  $Al_2O_3$  (23.2–24.5), FeO (0.8–1.8), MgO (0.2–1.5), CaO (0.4–0.7), Na<sub>2</sub>O (5.2–6.6), K<sub>2</sub>O (4.9–5.8), P<sub>2</sub>O<sub>5</sub> (0.3-0.7), and Cl (0.3-0.4). With the exception of a smaller rise in alkalinity, this tendency of melt differentiation was also observed in rhonite-bearing inclusions from olivine in the rocks of normal alkalinity studied here.

Grapes et al. [2003] report that rhonite and accompanying minerals crystallized between 1190 and 1090°C and at pressures <0.5 kbars in basanites discharged by Mount Sidley, western Antarctica. Treiman [2008] conjectures that rhonite may have been produced by the reaction of titanium—oligoclase amphibole with pyroxene:

$$\begin{split} &Ca_2(\,M_4^{+2}\,Ti)(Al_2Si_6)O_{22}(OH)_2 + \,M^{+2}SiO_3\\ &= Ca_2\,M_5^{+2}\,Ti(Al_2Si_4)O_{20} + 3SiO_2 + H_2O. \end{split}$$

Appropriate calibration is supposed to allow that reaction to limit the bounds of possible fugacity of water and associated volatiles.

The early and long (1320–1130°C) liberation of as high-alumina phases as are spinel, rhonite, and clinopyroxene from a melt that has a low alumina con-

**Table 3.** Compositions of associated daughter minerals

Sample #	LS16		LS26-28		LS20	KL119-3		KL119-5		
Minerals	Срх	Sp	Gl	Срх	Срх	Gl	Срх	Срх	Hb	Gl
SiO <sub>2</sub>	45.42	0.00	63.64	44.37	43.30	64.44	39.49	39.44	42.35	56.85
$TiO_2$	1.76	0.44	0.93	1.77	1.84	0.20	2.07	2.57	0.75	0.62
$Al_2O_3$	10.40	58.13	22.17	13.19	13.52	22.82	15.34	16.91	17.82	23.95
$Cr_2O_3$	0.02	0.00	0.00	0.02	0.00	0.03	0.00	0.00	0.01	0.01
$Fe_2O_3$	2.20	8.86	0.00	2.00	2.82	0.00	6.29	5.54	8.23	0.00
FeO	7.40	19.97	2.07	7.34	6.08	1.88	5.22	5.43	0.00	2.06
MnO	0.19	0.19	0.04	0.22	0.16	0.04	0.14	0.19	0.27	0.09
MgO	11.30	14.35	0.54	9.77	9.62	0.40	7.67	7.42	15.93	0.67
CaO	20.66	0.00	3.72	21.60	22.50	4.23	22.42	22.28	10.14	4.98
Na <sub>2</sub> O	0.36	0.00	5.15	0.37	0.26	4.26	0.28	0.45	2.36	4.44
$K_2O$	0.00	0.00	1.54	0.09	0.00	1.20	0.00	0.00	0.20	1.97
$H_2O$	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2.13	0.00
Total	99.72	101.94	99.80	100.74	100.10	99.50	98.92	100.23	100.19	95.64
	Fo	rmula amo	unts (Cpx	per 6 oxyge	n atoms, H	lb per 23, S	p per 32, a	nd Gl per 8	3)	
Si	1.7021	0.0000	2.8221	1.6489	1.6200	2.8449	1.5137	1.4886	5.9427	2.6645
Ti	0.0499	0.0701	0.0309	0.0496	0.0517	0.0067	0.0595	0.0731	0.0796	0.0220
Al	0.4600	14.4532	1.1589	0.5775	0.5961	1.1876	0.6932	0.7521	2.9480	1.3230
Cr	0.0005	0.0000	0.0000	0.0006	0.0000	0.0012	0.0000	0.0000	0.0007	0.0005
$Fe^{+3}$	0.0620	1.4066	0.0000	0.0560	0.0795	0.0000	0.1815	0.1575	0.8691	0.0000
$Fe^{+2}$	0.2322	3.5231	0.0766	0.2281	0.1902	0.0693	0.1673	0.1713	0.0000	0.0809
Mn	0.0061	0.0337	0.0013	0.0068	0.0052	0.0015	0.0045	0.0060	0.0324	0.0037
Mg	0.6311	4.5134	0.0355	0.5412	0.5366	0.0266	0.4384	0.4177	3.3334	0.0465
Ca	0.8298	0.0000	0.1767	0.8602	0.9018	0.2001	0.9208	0.9008	1.5239	0.2501
Na	0.0264	0.0000	0.4427	0.0266	0.0189	0.3645	0.0211	0.0328	0.6418	0.4035
K	0.0000	0.0000	0.0870	0.0045	0.0000	0.0677	0.0000	0.0002	0.0362	0.1176
$H_2O$	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000
Mg#	0.73	0.56	0.32	0.70	0.74	0.28	0.72	0.71	1.00	0.36
Total	4.0000	24.0001	4.8317	4.0000	4.0000	4.7701	4.0000	4.0001	16.4078	4.9123

Note: Fe<sub>2</sub>O<sub>3</sub> in clinopyroxene, hornblende, and spinel was calculated in accordance with the stoichiometry of these minerals, Fe<sub>2</sub>O<sub>3</sub> in rhonite was calculated following [Johnston and Stout, 1985]. For hornblende we also calculated the content of water per one molecule H<sub>2</sub>O. The analyses were made at the Institute of Volcanology and Seismology using a Camebax microprobe. The accelerating voltage was 20 kV with a current of 20 nA through the sample. Analyst V.V. Anan'ev.

tent, such as that in a homogenized inclusion [Sharygin, 2002], added to its accumulation in the residual melt, seems paradoxical. It is to be supposed that this association of daughter minerals was preceded by crystallization of the host mineral (olivine) on the walls of vesicles and that this provided for a sufficient concentration of  $Al_2O_3$  by the time the rhonite-bearing association began to be liberated. This must have considerably expanded the inclusions during homogenization.

The textural features of rhonite suggest its being an "ephemeral" intermediate phase during crystallization of the spinel—pyroxene sequence [Sharygin and Timina, 2008], although we have failed to find any mention of disintegration structures of rhonite or

some reaction relationships of it with other phases. In our opinion, rhonite should be treated as a rare (rarely detected) accessory mineral.

A review of the accessible literature shows wide limits for the parameters of its stability: temperature (1260–1180°C), pressure (4–0.5 kbars), oxygen fugacity (ranging between that relevant to the iron—wuestite buffer in meteorites [Fuchs, 1971] and the fugacity controlled by the nickel—bunsenite equilibrium in the situation of amphibole replacement in xenoliths [Grapes et al., 2003]), concentrations (and apparently the composition) of volatiles, as well as the compositions of the magma melts from which rhonite was liberated (ranging from high- and normal-alkaline

(tholeiitic) and from ultramafic to acidic). Therefore, at the present state of our knowledge, we find no critical factor or a set of factors that control the generation of rhonite, although in statistical terms one could highlight such favorable factors as increased alkalinity of the melt and increased pressure.

Theoretically speaking, the evolution of a cooling magma captured by minerals in the form of microinclusions ought to be similar to the evolution of the bulk of magma outside the phenocrysts; this should in the general case (on a qualitative level) be facilitated by the presence of rhonite both in melt inclusions and in the bulk of olivine and olivine-free (olivine being liberated) alkaline rocks. One finds that boiling and crystallization occur in a similar manner for interstitial melts in fragments (nodules) of cumulates that are comagmatic to the volcanics that discharge them. The mineral/melt quantitative relationships in rhonitebearing inclusions (see Fig. 1) are similar to the degree of crystallinity in these interstices and the groundmass of basic to intermediate normal-alkaline rocks, which are, however, composed of ordinary single- and bipyroxene-plagioclase associations with titanomagnetite (±ilmenite) and acidic low-alumina glass. Whether the absence of free (not daughter) rhonite in these (even though as relicts) is a question of a more careful search or some essential difference between the conditions of evolution for a "conserved" and a free magma melt constitutes an intriguing petrogenetic problem that calls for broader and more detailed investigations using finer geochemical and experimental techniques.

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