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# **Iron-magnesium minerals from differentiated rocks of Lovozersky alkaline massif**

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**Abstract.** The compositions of the main rock-forming iron-magnesium minerals (clinopyroxenes, alkaline amphiboles, biotites) from the rocks of the Lovozersky alkaline massif have been studied by microprobe analysis. Clinopyroxenes of the studied rocks represent aegirine-diopside-hedenbergite solid solutions. They form trends on the triangular diagram Aeg-Di-Hd, testifying for the higher alkalinity of the clinopyroxenes from juvitic rocks and eudialite-lujavrites. Amphibole alkalinity increases in the sequence sodalitesyenite→lujavrite→eudialite-lujavrite→juvitic rocks. Sometimes biotite occurs in sodalite-syenites and corresponds to phlogopite-annite series. The compositions of the iron-magnesium minerals are connected to the exchange equilibria and depend on mineral forming *T-P* conditions formation. This fact allows the application of mineral geothermometers in order to estimate the forming conditions of the corresponding assemblages. Using Bt-Cpx and Amph-Cpx geothermometers we estimated the forming temperatures of clinopyroxene-bearing parageneses of the investigated sodalite-syenites. They vary in the range 990 to 595- 630°C showing different stages of crystallization with the separation of several generations of certain minerals (clinopyroxenes, in particular).

*Key words*: clinopyroxene, alkaline amphibole, biotite, mineral geothermometers

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#### **Наталия Ивановна Сук, Алексей Котельников, Андрей Ковальский. Железномагнезиевите минерали на диференцираните скали от Ловозерския алкален масив**

**Резюме.** С микросондов анализ е изследван съставът на железно-магнезиевите скалообразуващи минерали (клинопироксени, амфиболи, биотити) от скалите на Ловозерския алкален масив. Клинопироксените на изследваните скали принадлежат на изоморфната редица егирин-диопсидхеденбергит. На триъгълната диаграма Aeg-Di-Hd те формират тренд, сочещ че по-алкалните клинопироксени принадлежат на ювитовите скали и на евдиалитовите луяврити. Алкалността на амфиболите се увеличава в последователността содалитов сиенит→луяврит→евдиалитов луяврит→ювитови скали. Понякога в содалитовите сиенити се появява биотит с флогопит-анитов състав. Съставът на Fe-Mg минерали е свързан с равновесното изоморфно заместване и зависи от *Р-Т* условията на минералообразуване. Това ни позволява да приложим минерална геотермометрия и с нейна помощ да оценим условията на формиране на съответните парагенези. С Bt-Cpx и Amph-Cpx геотермометър ние оценяваме температурата на кристализация на клинопироксен-съдържащите парагенези в изследваните содалитови сиенити от 990 до 595-630°C. Тези резултати сочат наличието на различни стадии на кристализация с няколко генерации на отделни минерали (в частност клинопироксени)

### **Introduction**

Lovozersky alkaline massif on the Kola Peninsula is one of the largest layered intrusions accompanied by giant deposits of trace and radioactive elements, which are concentrated in the minerals loparite and eudialite. The following zones are distinguished in the massif: the complex of eudialite lujavrites in the upper part of the massif, consisting of interlayered leucocratic, mesocratic, and melanocratic varieties, eudialite-lujavrites, fojaites, and urtites; the underlying thick differentiated complex, composed of repeating three-membered units of fojaite-urtite-lujavrite; the complex of nepheline-, nepheline-hydrosodaliteand poikilitic syenites, which are confined mainly to the border zones of the massif and probably underlie the differentiated complex; the vein complex of alkaline rocks (Gerasimovsky et al. 1966; Vlasov et al. 1959; Bussen & Sakharov 1967). Sodalite-syenites, constituting both layers among differentiated rocks and independent bodies are also distinguished.

We investigated sodalite-syenites occurring as layers or lenses among fojaites, lujavrites and trakhitoid lujavrites. The samples of sodalite-syenites were selected from boreholes 447 and 904, which penetrated the rocks of the differentiated complex in the northern and southern parts of the massif respectively, and from borehole 164 penetrating the rocks of the upper member of the eudialite-lujavrite complex. Loparite-bearing rocks (lujavrites, juvites, fojaite-juvites, urtites, etc.) including ore horizons were studied too. This kind of samples was selected from the same boreholes, as well as from borehole 178, passing through the rocks of the eudialitelujavrite complex. The detailed locations of the samples used in this study are given in Table 1. Rock-forming minerals in the studied varieties of rocks are represented by nepheline, feldspar (albite, potassium feldspar), clinopyroxenes

(aegirine, aegirine-augite), alkaline amphibole, as well as sodalite and biotite in sodalitesyenites. The following accessory minerals are distinguished: eudialite, ramsaite, sphene, apatite, carbonates, iron sulfides (pyrite and pyrrhotite), manganoan ilmenite, etc. Loparite depending on the amount being in the rock can be both rock-forming (in ore horizons) and accessory mineral. Secondary natrolite and analcime are available too.

The mineral compositions were determined using a Camebax electron microprobe, equipped with a LINK AN10000 energydispersive X-ray microanalyzer and a CamScan MV2300 (VEGA TS 5130MM) digital scanning electron microscope with YAG detectors of secondary and back-scattered electrons and a Link INCA Energy energydispersive X-ray microanalyzer with a semiconductor Si(Li) detector. The results of X-ray spectral microanalysis were processed by

Table 1. *Location of the samples used in this study* 

	Sample	Borehole	Depth
Rock type	number	number	(m)
Sodalite syenites	$C-61$	447	717
Sodalite syenites	904-22	904	410
Sodalite syenites	$EV-6$	164	435
Lujavrites with	447-7	447	955
loparite			
Lujavrites with	$C-102$	447	1021
loparite			
Lujavrite with	202-5	202	63
eudialite			
Eudialite	185-11	185	165
lujavrite			
Lujavrite-juvite	$C-74$	447	855
with loparite			
Juvite with	904-26	904	465
loparite			
Loparite fojaite-	904-34	904	580.5
juvite			
<i>Fojaite-juvite</i>	904-49	904	815

means of the INCA Energy 200 program, and the obtained data were further recalculated using a program package developed at the Institute of Experimental Mineralogy, Russian Academy of Sciences.

#### **Analysis of iron-magnesium minerals**

The main rock-forming iron-magnesium minerals of the studied rocks are clinopyroxenes, alkaline amphiboles, and locally biotites. The compositions of the iron-magnesium minerals reflect the exchange equilibria and depend on forming *T-P* conditions. Thus, the study of their compositions is necessary in order to reconstruct the forming conditions of the corresponding assemblages and makes it possible to apply mineral geothermometers such as Bt-Cpx, Amph-Cpx, Amph-Bt. In this paper the compositions of the main rock-forming ironmagnesium minerals are considered.

### *Clinopyroxenes*

In the studied rocks they are represented by aegirine-diopside-hedenbergite solid solutions of variable composition. Examples of compositions of clinopyroxenes from different rocks are represented in Table 2. Several generations are distinguished in each sample. Aegirineaugites are elongated crystals, up to 2-3 mm in size. Prismatic aegirine crystals are smaller than aegirine-augites and may occur as inclusions in nepheline. Intergrowths of clinopyroxene crystals from different generations, showing no reaction relationships, have been found (Fig. 1a). Sometimes pyroxene of a late generation overgrows earlier aegirine-augite crystals, and the development of late pyroxene in fractures in aegirine-augite was observed. In such cases, both pyroxenes have rugged margins (Fig. 1b). Resorption of early pyroxene generations have been observed locally.



Fig. 1a-b. Various clinopyroxene (Cpx) generations from the sodalite syenites. (Bt) biotite. BSE image

Sample number	$C-61$		904-22		$EV-6$				
Cpx generation	Cpxl	Cpx2	Cpx3	Cpx1	Cpx2	Cpx1	Cpx2	Cpx3	Cpx4
SiO <sub>2</sub>	52.17	50.96	51.03	50.81	51.33	51.71	52.18	52.38	51.28
TiO <sub>2</sub>	1.35	1.56	0.52	1.65	0.89	1.48		0.34	0.48
$\text{Al}_2\text{O}_3$	1.57	1.60	1.17	1.05	0.92	1.74	0.64	1.07	1.16
MnO	0.72	1.20	1.75	1.26	1.17	0.94	0.87	1.38	1.33
FeO	7.30	13.00	19.14	17.77	21.82	9.80	14.35	16.41	19.54
MgO	13.35	9.33	5.81	6.64	4.18	11.94	9.78	7.74	5.91
CaO	21.83	18.88	15.57	13.31	10.35	19.53	18.11	15.99	14.18
Na <sub>2</sub> O	1.71	3.47	5.01	6.42	8.24	2.86	4.07	4.64	6.12
$K_2O$		$\overline{\phantom{0}}$	-	0.17			-	0.05	
ZrO <sub>2</sub>			٠	0.92	1.10				
Number of analyses	3	$\overline{4}$	8	4	5		$\mathfrak{D}$	3	6

Table 2. *Mean chemical compositions of clinopyroxenes from the rocks of Lovozersky massif. Means calculated from microprobe analytical results, normalized to 100 wt.%. (n.a) not analyzed* 

![](_page_3_Picture_175.jpeg)

![](_page_3_Picture_176.jpeg)

Compositions of clinopyroxenes of different generations are well illustrated by the triangular diagram Aeg-Di-Hd. The sodalitesyenite samples from boreholes 447 and 164, which are located in the northern part of the massif, not far from each other, form two distinct branches on the diagram (Fig. 2a) (Suk et al. 2007): (I) from  $Di_{70}Hd_{10-15}Aeg_{20-15}$  to Di<sub>30-35</sub>Hd<sub>25-30</sub>Aeg<sub>45-35</sub> (high-temperature branch) and (II) from  $Di_{25-30}Hd_{15-10}Aeg_{60}$  to almost pure aegirine (low-temperature branch).

Similar trends of compositions were obtained by Korobeynikov and Laayoky (1993) and Kogarko et al. (2006) for clinopyroxenes from different rocks of Lovozersky massif. Our data allow the estimation of the trends of clinopyroxene compositions in single sample that reveals crystallization sequence and characterizes the change of physico-chemical conditions during the formation process of a certain rock.

In some cases we managed to use Amph – Cpx and Bi – Cpx geothermometers (Perchuk 1970; Perchuk & Ryabchikov 1976) to determine the formation temperatures of clinopyroxenes, containing significant fractions of hedenbergite constituent. Temperatures were determined for touching mineral pairs. In addition, the compositions of amphibole and biotite inclusions in pyroxene were used. The temperatures of formation of several pyroxene generations were determined in a single sample. They vary in the range from 990 to 595-630°C, reflecting different crystallization stages. Such a compositional evolution of clinopyroxene probably corresponds to changes in the physico-chemical conditions of their formation (in particular, an increase in alkalinity).

Pyroxenes from the sodalite syenite samples collected from borehole 904, which is situated in the southern part of the massif, form a compositional trend (Fig. 2b), corresponding to branch II in Fig. 2a. As can be seen from

![](_page_4_Figure_4.jpeg)

Fig. 2a-d. Clinopyroxene compositions from different rocks of Lovozersky alkaline massif (mol.%). Temperatures are determined by clinopyroxene geothermometers. Mineral abbreviations: (Aeg) aegirine, (Di) diopside, (Hd) hedenbergite

the diagram, the composition of clinopyroxene evolves toward an increase in alkalinity. Crystallization temperatures of 740-750°C were obtained for the most magnesian clinopyroxenes and amphiboles from these rocks.

A trend of compositions of the studied loparite-bearing lujavrites, juvitic rocks (juvites, fojaites, urtites, etc.) from a differentiated complex, as well as eudialite lujavrites from the complex of eudialite lujavrites, corresponds to the low-temperature branch of the trend for clinopyroxenes of alkaline rocks and to the low-temperature trend for clinopyroxenes of sodalite syenites of Lovozersky massif (Fig. 2c, d). This fact seems to be connected with the increased alkalinity as well as with the enhanced contents of fluid components (including salt) during their formation. Due to the low fraction of the hedenbergite constituent in the composition of clinopyroxenens from lujavrites and juvitic rocks the mineral geothermometers with their participation cannot be used for these rocks. This is so, because the existing clinopyroxene-using geothermometers are made without considering the data on solid solutions of alkaline clinopyroxenes, containing significant amount of  $Fe<sup>3+</sup>$ . However, a comparison of the obtained trend of clinopyroxene compositions (Fig. 2c) with the trend of clinopyroxenes from sodalite syenites (Fig. 2a) of Lovozersky massif (Suk et al. 2007) makes it possible to assume that temperatures of clinopyroxene formation in lujavrites and juvitic rocks did not exceed 600- 650°C. Such comparison seems rightful, since the samples of these rocks and sodalite syenites are collected from the same drill holes and located in immediate depth proximity: sodalite sienites constitute layers or lenses among fojaites, lujavrites and trachytoid lujavrites of the differentiated complex of the massif that allows the assumption of similar *P-T* parameters for the evolution of these rocks.

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The diagrams in Fig. 3 illustrate correlations between the concentrations of some elements in the composition of clinopyroxenes from sodalite syenites (Suk et al. 2007). The content of Mg is inversely proportional to total Fe (Fig. 3a). The contents of Fe and Na are linearly dependent, but there are two trends (Fig. 3b) with different slopes: (I) corresponding to branch I and (II) corresponding to branch II in Fig. 2a. Similar patterns can be seen in the Na-Mg diagram (Fig. 3c). This is obviously related to a change from the aegirine-diopside-hedenbergite composition of clinopyroxenes to a substantially aegirinediopside composition, i.e., a change from the isomorphic substitution  $[Ca+(Mg,Fe^{2+})] \Leftrightarrow$  $(Na+Fe^{3+})$  to  $(Ca+Mg) \Leftrightarrow (Na+Fe^{3+})$  coupled with an abrupt increase in alkalinity.

The pyroxenes usually contain considerable amounts of Ti (up to 1.78-2.40 wt.%) TiO<sub>2</sub>) and sometimes Zr (up to  $1.62 \text{ wt.} \%$  $ZrO<sub>2</sub>$ ). The concentrations of these element are negatively correlated  $(r_{xy} = -0.83, n = 21)$  with the content of the aegirine constituent, which implies the isomorphic substitution  $[(Ti, Zr) +$  $Fe^{2+}$ ]  $\Leftrightarrow$  2Fe<sup>3+</sup> proposed by Semenov (1972). It is interesting that Ti is usually enriched in relatively higher temperature clinopyroxenes (branch I in Fig. 2a), whereas Zr is higher in medium temperature clinopyroxenes (branch II in Fig. 2a). To some extent it corresponds to the conditions of eudialite-bearing rocks formation, which has been discussed above.

#### *Alkaline amphiboles*

They usually form intergrowths with pyroxene crystals and occur as inclusions in them. Some examples of pyroxene-amphibole relationships are shown in Fig. 4. The compositions of some amphiboles studied are given in Table 3. The presence of fluorine in their composition is characteristic. Alkaline amphiboles represent a solid solution from richterite-edenite-riebeckite

![](_page_6_Figure_0.jpeg)

to riebeckite-arfvedsonite composition. Amphiboles from juvitic rocks differ from the amphiboles in lujavrites and sodalite syenites by their higher content of riebeckite constituent. Amphibole alkalinity increases in the sequence sodalite syenite–lujavrite–eudialite lujavrite– juvitic rocks (Fig. 5). Here amphibole compositions are consistent with the general direction of compositional variations in clinopyroxenes. An increase in the fraction of aegirine constituent in the clinopyroxene composition is accompanied by a decrease in the Ca fraction (increase in Na fraction) in the amphiboles. This also indicates that the general evolution of the massif included an increase in alkalinity. It should also be emphasized that the increase of

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![](_page_6_Figure_2.jpeg)

Fig. 3a-c. Correlations of selected element concentrations (*apfu*) in the compositions of clinopyroxenes

mole portion of Mn in amphiboles is synchronous with their alkalinity rise.

#### *Biotites*

Some samples of sodalite syenites contain biotite, which forms individual laths and intergrowths with aegirine-augite or occurs as inclusions in clinopyroxene (Fig. 6). Several generations of biotite with different  $X_{Mg}$  values were distinguished in some samples (Table 4). The composition of biotite corresponds to the phlogopite-annite series with  $X_{Mg}$  value from 0.495 to 0.675 (a more magnesian biotite with  $X_{\text{Mg}}$ =0.675 can be found locally among inclusions in pyroxenes). The presence of fluorine in the composition of biotite is also characteristic.

![](_page_7_Picture_0.jpeg)

Fig. 4a-b. Relationships of clinopyroxene (Cpx) and amphibole (Amph) in the sodalite syenites of the Lovozersky massif. (Sdl) sodalite; (Kfs) potassium feldspar; (Ne) nepheline; (Ap) apatite; (Spn) titanite (sphene). BSE image

![](_page_7_Figure_2.jpeg)

Fig. 5. Compositions of amphiboles from the rocks of Lovozerskii massif. Mineral abbreviations: (Arf) arfvedsonite, (Ed) edenite, (Gln) glaukophane, (Rbk) riebeckite, (Rich) richterite

![](_page_8_Picture_83.jpeg)

![](_page_8_Picture_84.jpeg)

![](_page_8_Picture_2.jpeg)

Fig. 6. Relationships between clinopyroxene (Cpx) and biotite (Bt) in the sodalite syenites of the Lovozersky massif. BSE image

#### **Discussion of the results**

The study of the compositions of iron-magnesian minerals from the rocks of Lovozersky alkaline massif makes it possible to estimate the forming temperatures of their assemblages and trace the evolution parameters of the rocks.

The data obtained allowed us to distinguish several crystallization stages of the rocks. The crystallization of the sodalite syenites (boreholes 447 and 164) (Fig. 2a), for example, included the stages of early clinopyroxene and amphibole (~990°C), later clinopyroxene with biotite  $(\sim 700^{\circ}C)$ , and late clinopyroxene with biotite and amphibole (595- 630°C). These stages of sodalite syenite formation are in good agreement with the aforementioned crystallization sequence of minerals in the poikilitic sodalite syenites proposed by Vlasov et al. (1959).

Table 4. *Mean chemical compositions of biotites from the sodalite syenites of Lovozersky massif. Means calculated from microprobe analytical results in wt.%* 

Sample number		$EV-6$		
<b>Biotite</b> generation	Bt 1	Bt <sub>2</sub>	Bt <sub>3</sub>	Bt
SiO <sub>2</sub> TiO <sub>2</sub>	41.88 1.45	40.60 1.72	39.84 2.08	41.07 2.04
$Al_2O_3$	10.73	11.10	11.34	11.11
MnO FeO	1.54 14.03	1.58 16.08	1.56 18.88	1.37 15.78
MgO Na <sub>2</sub> O	18.12 0.47	16.16 0.51	14.24	16.57 0.34
$K_2O$ F	9.77 2.77	10.25 1.71	10.06 1.61	9.74 2.36
Total	100.76	98.13	99.61	100.38
Number of analyses	4	$\mathfrak{D}$	1	6

On the basis of our experimental data (Kotelnikov et al. 2004, 2005), temperatures for the formation of the coexisting sodalite phases (chlorine- and sulphate-bearing) were estimated. They were 625-530°C. These values are in good agreement with the temperatures obtained from other mineral geothermometers and consistent with the general temperature trend of these rocks. Similar temperatures of 750-505°C were obtained for a sodalite-bearing assemblage from the Lovozersky massif by Ustinov et al. (2006), who used a sodalite geothermometer constructed on the basis of the investigation of oxygen isotope partitioning between the silicate and sulphate components of synthetic sulphate-bearing sodalites. The obtained data allowed us to calculate the average forming temperature of 600±50°C for the sodalite assemblages.

Moreover, the amount of excess silica in nepheline (Deer et al 1966) gives some information on forming temperatures of nepheline assemblages in the studied rocks. Excess silica content in the nepheline of lujavrites varies from 4.5 to 6 mol.%, that corresponds to temperatures from 600 to 760°C according to the data of Hamilton (Deer et al. 1966). In the juvites, urtite-juvites and fojavite-juvites it is less than 3 mol.%, that corresponds to temperatures lower than 500°C. This reflects highertemperature conditions of nepheline formation from lujavrites. In the nepheline from sodalite syenites excess silica content was up to 9 mol.%. This evidences for a rather high forming temperature of nepheline from sodalite syenites.

The compositions of coexisting alkaline feldspars yielded a temperature of  $\sim$ 450 $\degree$ C, which is not the temperature of feldspar formation, but was obtained for the compositions of coexisting albite and potassium feldspar produced by unmixing of their solid solution (Suk et al. 2007). The wide occurrence of natrolite in the samples indicates the influence of relatively low-temperature (<250°C) hydrothermal processes.

Our data reflect the continuous process of formation of mineral assemblages in these rocks, starting from a temperature of ~990°C and including the formation of several generations of certain minerals (e.g., clinopyroxenes at  $\sim$ 700 and 595-630 °C) (Suk et al. 2007) to temperatures of  $\sim$ 250 $\degree$ C reflecting postmagmatic hydrothermal processes.

Moreover, the temperatures of the initial stages of mineral formation, estimated by mineral geothermometry correspond to the experimental temperatures of complete melting of the rocks of the Lovozersky massif under a water pressure of 1 kbar: from 1010°C for urtites to 800°C for fojaites enriched in volatile components, including F and Cl (Kogarko & Romanchev 1977). The temperature of complete melting of the average composition of the differentiated complex of the Lovozersky massif was estimated as 1070°C under dry conditions and 910°C under a water pressure of 1 kbar (Kogarko & Romanchev 1977).

### **Conclusions**

(1) The compositions of the main rock-forming iron-magnesium minerals (clinopyroxenes, alkaline amphiboles, biotites) from the rocks of the Lovozersky alkaline massif were determined by microprobe analysis.

(2) Clinopyroxenes of the studied rocks represent aegirine-diopside-hedenbergite solid solutions of variable composition. Alkaline amphiboles represent a solid solution from richteriteedenite-riebeckite to riebeckite-arfvedsonite composition. Amphibole alkalinity increases in the sequence sodalite syenite-lujavrite-eudialite lujavrite-juvitic rocks. Biotite is locally observed in sodalite syenites and corresponds to the phlogopite-annite series.

(3) Using various mineral geothermometers, the temperature range of formation of the clinopyroxene-bearing assemblages of the sodalite syenites was estimated from ~990 to 595-630°C. These estimates reflect various stages of crystallization with separation of several generations of certain minerals (clinopyroxenes in particular). Clinopyroxene stages of crystallization include the stages of early clinopyroxene and amphibole (~990°C), later clinopyroxene with biotite  $({\sim}700^{\circ}C)$ , and late clinopyroxene with biotite and amphibole (595- 630°C). These data are in agreement with the crystallization sequence of the poikilitic sodalite syenites proposed by Vlasov et al. (1959).

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