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GENESIS AND TYPOCHEMISM OF LAMPROPHYLLITE-BARYTOLAMPROPHYLLITE SERIES MINERALS FROM LUJAVRITE-MALIGNITE COMPLEX OF KHIBINY MASSIF

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The detail analysis of chemical composition and character of postmagmatic alteration of lamprophyllite-barytolamprophyllite series minerals from lujavrite-malignites of Khibiny massif was made by local roentgenospectral and electron-microscopic methods. It is determined that in lujavrites high-barium lamprophyllite is a typomorphic accessory mineral. In malignites two stages of lamprophyllite alteration are ascertained, which correspond to two stages of their formation: 1) at the stage of primary rocks (lujavrite or titanite trachytoid melteigite-urtites) transformation in result of K,Si-metasomatosis the recrystallization of primary Ba-lamprophyllite without change of chemical composition (in case of lujavrites) and enrichment of primary strontium lamprophyllite by barium and potassium (in case of melteigite-urtites) take place; 2) at the stage of low-temperature rocks transformation by action of solutions enriched by strontium and/or calcium the replacement of Ba-lamprophyllite by strontium analogue (in malignites genetically connected to lujavrites) and development of titanite after Ba,K-lamprophyllite (in malignites connected to ijolite-urtites) occur.

It is detected that the character of postmagmatic alteration of primary strontium lamprophyllite in «porphyraceous malignites» is also the evident of primary rocks (trachytoid ijolites) transformation during K,Si-metasomatosis. 4 figures, 1 table and 16 references.

Lamprophyllite, $(Sr, Ba, K)_2Na(Na, Fe, Mn)_2$ Ti₃(Si₄O₁₆)(O,OH,F)₂, is one of the most characteristic accessory minerals of Khibiny alkaline massif. It occurs in rocks of almost all this massif complexes: nepheline syenites, melteigiteurtites, apatite-nepheline rocks, ristschorrites, lujavrite-malignites. A whole number of works was devoted to lamprophyllite, however, the systematical study of accessory lamprophyllite from rocks of Khibiny massif, in particular, the study of genetic aspect of its mineralogy, was not practically undertaken till now. The minerals of lamprophyllite-barytolamprophyllite series from rocks of lujavrite-malignites complex have become the object of present study.

The crystal structure of lamprophyllite has a layered constitution, which allows free entering of large cations (Sr, Ba, K, Ca, Na) into inter-layer space and significant variations of their ratios (Rastsvetaeva *et al.*, 1995a, Rastsvetaeva, Dorfman, 1995b). This crystal structure peculiarity makes lamprophyllite perspective from the point of view of use it as one of the mineralogogeochemical indicators of changes of minerogenesis environment conditions.

General characteristic of research objects

Lujavrites and malignites in Khibiny massif are spatially connected to trachytoid melteigite-urtites of Central Arc. Their alternating bed bodies are traced in upper part of this complex along contact of Kukisvumchorr-Rasvumchorr apatit-nepheline deposit with overlapping ristschorrites up to the upper course of Kuniiok. The lujavrites from Khibiny are the late melanocratic phase of nepheline syenites; they consist of orthoclase, nepheline, and dark-coloured minerals, are characterized by clear trachytoid texture, being the result of orthoclase laths orientation. The question about malignites genesis remains disputable. According to opinion of one researches group, they are the late phase of melteigite-urtites or ristschorrites (N.A. Eliseev, T.N. Ivanova, S.I. Zak et al.). Others, I.A. Zotov, B.Ye. Borutzky, A.I. Serebritskii, consider them as a product of metasomatic alteration (orthoclazization) of melteigiteurtites or ijolites at their contact with lujavrites. khibinites, and lyavochorrites. In present research malignites are considered as rocks of metasomatic genesis, according to analysis of earlier works and features, determined by us. On their mineral composition they are divided into three types: 1) malignites (below denoted by us as «malignites-L») formed as a result of metasomatic alteration of lujavrites and confined to endocontact zone of lujavrites with trachytoid melteigite-urtites; they have mineral composition similar to that of lujavrites, but with poikilitic structure caused by large poikiloblasts of orthoclase and amphibole; 2) malignites (denoted as «malignites-U») confined to exocontact zone of lujavrites with titanite trachytoid melteigite-urtites and formed as a result of metasomatic alteration of the latter; they are also characterized by poikilitic structure, but according to their composition, are closer to melteigite-urtites; these rocks contain

Rocks	lujavrites				«malignites-L»				«malignites-U»			«porphyric malignites		
Phases		Ι	I-a			I, II-a		II-b	Ι	ІІ-а		I	П-а	
Nº	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Na ₂ O	10.32	9.05	9.24	9.09	9.65	9.53	9.77	11.95	10.50	9.15	8.67	9.53	9.35	9.07
K ₂ O	1.01	1.70	1.21	1.12	1.71	1.50	1.67	1.89	1.77	1.88	2.69	1.78	1.76	2.56
CaO	1.00	0.97	0.91	0.87	1.02	0.92	1.17	1.57	1.35	1.11	1.34	1.39	1.22	0.94
SrO	10.55	7.53	5.26	3.93	5.30	8.07	9.60	11.62	14.59	8.46	6.39	14.48	9.74	4.81
BaO	10.80	15.28	19.71	21.39	17.05	14.35	10.09	4.08	3.26	14.21	18.03	5.88	10.68	19.71
MgO	0.35	0.44	0.41	0.45	0.00	0.43	0.48	0.52	0.70	0.26	0.62	0.77	0.45	0.53
MnO	1.57	1.22	1.66	1.60	2.09	1.76	1.87	1.36	1.27	2:30	1.70	0.85	1.15	0.00
FeO	2.96	2.64	2.70	2.56	2.71	2.61	2.98	3.87	4.63	2.66	2.85	4.24	3.64	2.34
Al_2O_3	0.27	0.38	0.51	0.68	0.44	0.35	0.27	0.00	0.20	0.27	0.24	0.50	0.24	0.61
SiO_2	29.70	29.08	28.65	27.85	30.90	29.37	30.06	32.60	29.55	30.95	29.59	30.20	29.54	30.35
TiO_2	27.97	26.86	25.62	26.08	30.00	26.27	27.62	30.45	27.77	29.65	28.07	29.35	27.18	28.77
Nb_2O_5	0.77	0.11	0.00	0.20	0.80	0.10	0.69	0.90	0.39	0.00	0.15	1.72	0.25	0.17
Total	97.27	95.26	95.88	95.82	101.67	95.26	96.27	100.81	95.98	100.90	100.34	100.69	95.20	99.86
					Numbe	rs of ion	s on the	basis of	(Si + Al) =	- 4				
Na	2.67	2.38	2.45	2.46	2.38	2.48	2.49	2.84	2.73	2.27	2.25	2.40	2.43	2.66
К	0.17	0.29	0.21	0.20	0.28	0.26	0.28	0.30	0.30	0.31	0.46	0.29	0.30	0.42
Ca	0.14	0.14	0.13	0.13	0.14	0.13	0.17	0.21	0.19	0.15	0.19	0.19	0.18	0.13
Sr	0.82	0.59	0.42	0.32	0.39	0.63	0.73	0.83	1.14	0.63	0.50	1.09	0.76	0.36
Ba	0.56	0.81	1.06	1.17	0.85	0.76	0.52	0.20	0.17	0.71	0.95	0.30	0.56	0.99
Mg	0.07	0.09	0.09	0.09	-	0.09	0.09	0.10	0.14	0.05	0.12	0.15	0.09	0.10
Mn	0.18	0.14	0.19	0.19	0.23	0.20	0.21	0.14	0.14	0.25	0.19	0.09	0.13	-
Fe	0.33	0.30	0.31	0.30	0.29	0.29	0.33	0.40	0.52	0.28	0.32	0.46	0.41	0.25
Al	0.04	0.06	0.08	0.11	0.07	0.06	0.04	-	0.03	0.04	0.04	0.08	0.04	0.09
Si	3.96	3.94	3.92	3.89	3.93	3.94	3.96	4.00	3.97	3.96	3.96	3.92	3.96	3.91
Ti	2.80	2.74	2.63	2.74	2.87	2.65	2.74	2.81	2.80	2.85	2.83	2.87	2.74	2.78
Nb	0.05	0.01	0.01	-	0.05	0.01	0.04	0.05	0.02	-	0.01	0.10	0.02	0.01
Ba: <u>S</u> r	0.69	1.37	2.53	3.67	2.17	1.20	0.71	0.24	0.15	1.14	1.91	0.27	0.74	2.77

Table 1. Chemical composition of lamprophyllite-barytolamprophyllite series minerals from lujavrite-malignites complex rocks of Khibiny massif

Note:

Phase I — primary lamprophyllite of the rock; I-a — barytolamprophyllite (in lujavrites); II-a — lamprophyllite, formed during K,Si-metasomatosis; II-b — lamprophyllite, formed by influence of the late more low-temperature solutions. Analysts: an. 1-4, 6, 7, 9, 13 — V.V. Khangulov (Camebax SX-50, IGEM RAS); an. 5, 8, 10-12, 14 — N.V. Trubkin (electron microscope JSM-5300 with X-ray spectrometer Link ISIS, IGEM RAS). An. 9, 14 — average on two analyses; an. 2, 3, 5, 8 — on three; an. 12, 10, 11 — on five; an. 6, 13 — on seven; an. 1, 7 — on ten analyses

significantly more nepheline and dark-coloured minerals than «malignites-L», and less potash feldspar; titanium enrichment is also their characteristic feature; 3) «porphyric malignites», according to S.I. Zak terminology (Zak et al., 1972), which are traced in lower part of trachytoid melteigite-urtites series at the contact with underlying khibinites and among ristschorrites, and apparently, represent the small-grained trachytoid ijolites altered by metasomatic processes. They are characterized by porphyritic structure caused by the poikilocrystals of lamprophyllite or feldspar, which stand out among more small-grained matrix, composed by dark-coloured minerals and nepheline, and the trachytoid texture caused by orientation of aegirine-hedenbergite needle-shaped crystals and laths of metasomatic potash feldspar replaced nepheline. Outcrops of these rocks are noted at Mt. Poachvumchorr. Bedrocks of «malignites-L» and «malignites-U» are traced at Mt. Kukisvumchorr.

Morphology, typical assemblages and chemical compositions of lamprophyllite from lujavrite-malignites

In lujavrites lamprophyllite forms prismatic crystals of goldish-brown colour from some millimeters up to 1 cm in size and their growths disposed between high-barium (up to 3.0 -3.5 % BaO) orthoclase laths, among magnesioarfvedsonite and aegirine. Lamprophyllite is associated with eudialyte (proper eudialyte, according to (Johnsen *et al.*, 2003)), titanite, rarely rinkite and apatite. Here lamprophyllite is one of the latest mineral phases: it forms later than dark-coloured and most of accessory minerals, i. e. rinkite, apatite, which inclusions are observed in the crystals of lamprophyllite, titanite (sometimes replaced by lamprophyllite), and eudialyte.

Primary lamprophyllite from lujavrites, according to its chemical composition, is highbarium, although there are perceptible variations in content of both BaO, in most of cases from *10-11 to *15 %, and SrO, from *8 to 11 % (Table 1); the ratio (in *atoms per formula unit*) Ba/Sr=0.7-1.4. Continuous isomorphous series between Sr- and Ba-dominant specimens is traced (Fig. 1). Potassium and calcium are contained in lamprophyllite in subordinate amounts. The content of manganese is also insignificant.

At the late mineral formation stages barium lamprophyllite are replaced by varieties with yet higher content of barium right up to bary-tolamprophyllite (the content of BaO increases to 20-21%, the content of SrO is near 4-5%; Ba/Sr = 2.5-3.7, an. 3, 4 in Table 1), growing at the grains edges. Apparently, this phenomenon is caused by accumulation of barium in residual mineral forming environment because of its non-cogerency.

The appearance of high-barium lamprophyllite in lujavrites of Khibiny is their distinctive typomorphic peculiarity. The predominantly strontium lamprophyllite with significant content of manganese is spread in the analogous rocks from Lovozero massif (Kola Peninsula): lujavrites from differentiated complex of lujavrites-foyaites-urtites; eudialyte, lamprophyllite, and porphyric lujavrites (Bussen, Sakharov, 1972; Vlasov et al., 1959). Strontium lamprophyllite is observed also in leucocratic nepheline syenites of Kibiny: khibinites, foyaites etc. Barium lamprophyllite and barytolamprophyllite till now are noted practically only in pegmatites and late cross-cutting veinlets, which occur among apatite-nepheline rocks and nepheline syenites in Khibiny (Dudkin, 1959; Peng Tze-Chung, Chang Chien-Hung, 1965; Kapustin, 1973; Rastsvetaeva, Dorfman, 1995a) and Lovozero massifs, Kola Peninsula (Semenov, 1972; Kapustin, 1973; Bussen, Sakharov, 1972; Bussen et al, 1978), and also among albitized fenites at Turii cape (Kola Peninsula), metasomatites in Inagli and Murun massifs (South Yakutiya) (Rastsvetaeva et al., 1995b; Lazebnik et al., 1998), syenitized schists in Botogol'skii massif (East Sayan) (Kapustin, 1973). Moreover, high-barium lamprophyllite is described in a number of other massifs in the world (Zaitsev, Kogarko, 2002).

«Malignites-L» are characterized by irregular distribution of lamprophyllite. Here there are its large (up to 2-3 cm and larger) poikilocrystals (with inclusions of aegirine, nepheline, apatite, sometimes fersmanite), growing among



FIG. 1. The ratios of barium, strontium, and potassium (in apfu) in lamprophyllite from lujavrite-malignites complex rocks of Khibiny massif: lujavrites (1); «malignites-L» (2); «malignites-U» (3); «porphyric malignites» (4); and nepheline syenites of Khibiny massif (5); lujavrites of differentiated complex and eudialyte lujavrites of Lovozero massif (6)

poikilocrystals of high-barium orthoclase and Na-Ca-amphiboles of magnesioarfvedsonite richterite series. Rarely lamprophyllite forms small tabular crystals. In a number of cases lamprophyllite is corroded by high-barium orthoclase. The mineral assemblage, coexisting with lamprophyllite, is close to «lujavrite» one: it contains eudialyte, titanite, rarely rinkite, later Sr-apatite (6 – 10% SrO), taseqite, growing after proper eudialyte (Johnsen *et al.*, 2003), fersmanite, sometimes pectolite, which accumulations are noted into interstitions between other minerals grains.

The chemical composition of early lamprophyllite from «malignites-L» is in whole identical to barium lamprophyllite and barytolamprophyllite from lujavrites: BaO — from »10 to 17%, SrO — 5 – 10%; Ba/Sr = 0.7 – 2.2 (Table 1, an. 5-7). However, here barium lamprophyllite is, as a rule, changed. In the most of cases in its poikilocrystals there are, together with Badominant parts, the areas with gradually decreasing barium content (up to 4% BaO) and increasing content of strontium (on average »12 % SrO, Table 1, an. 8). These areas are traced parallel cleavage cracks of lamprophyllite poikilocrystals (Fig. 2). The ratio Ba/Sr in these zones is decreased up to 0.25, on chemical composition they corresponds to proper lamprophyllite, i. e. strontium lamprophyllite.

In the levels of **«malignites-U»** lamprophyllite, in contrast to above-mentioned rocks, is noted enough rarely. Its prismatic crystals are nearly 0.3 - 0.5, seldom 1 cm in length. They are located together with dark-coloured minerals (mainly aegirine, rarely magnesioarfvedsonite are noted) between poikilocrystals of potash

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FIG. 2. The development of strontium lamprophyllite after poikilocrystal of high-barium lamprophyllite in «malignites-L» (image in reflected electrons, scanning electron microscope JSM-5300, IGEM RAS). Ba-lamp — high-barium lamprophyllite; Sr-lamp — strontium lamprophyllite; Amph magnesioarfvedsonite



FIG. 3. The replacement of primary strontium lamprophyllite by barium-potassium lamprophyllite during K,Si-metasomatosis in «malignites-U» (image in reflected electrons, scanning electron microscope JSM-5300, IGEM RAS). Balamp — high-barium lamprophyllite; Sr-lamp — strontium lamprophyllite; Neph — nepheline; Aeg — aegirine

feldspar (barium-free, in contrast to «malignites-L») and Na-amphiboles of magnesioarfvedsonite — richterite series. Lamprophyllite is corroded by feldspar in different degree. It associates also with eudialyte group minerals (eudialyte with high-sodium and potassium-sodium chemical composition and taseqite, growing after it), djerfisherite, late titanite, Sr-apatite (the content of SrO up to 10%), pectolite, and pyrochlore. The latter is noted, as a rule, in cavities and crack of altered lamprophyllite.

In these rocks early lamprophyllite is significantly strontian, the content of BaO is not exceed on average 3% (Table 1, an. 9). Very often it is, in different degree, sometimes practically wholly, replaced by late barium-potassium lamprophyllite (Fig. 3). The content of SrO is near to 6-8%, BaO » 14-18%, Ba/Sr » 1.1-1.9, the content of K₂O in most high-barium

parts reaches 2.7% (Table 1, an. 11) (in primary lamprophyllite of above-mentioned rocks, this ratio fluctuates from 1.0 to 1.7%, and in late lamprophyllite it achieved 1.9%, Table 1). On the diagram, showing the ratios of barium, strontium, and potassium in *apfu* (contained in Table 1), the points of its chemical compositions are slightly displaced in «potassium» area (Fig. 1). At yet later stages Ba,K-lamprophyllite is, in its turn, replaced by titanite (Fig. 4).

In **«porphyric malignites»** lamprophyllite forms large poikilocrystals (up to 2–4 cm in size) among matrix, consisting of fine-needleshaped crystals of aegirine-hedenbergite and magnesioarfvedsonite and small grains of nepheline. Here lamprophyllite is corroded by potash feldspar and often replaced by micrograined aggregates of feldspar and aegirine. Except lamprophyllite, the typical accessory minerals here are eudialyte group minerals (high-sodium and potassium-sodium eudialyte is most widespread), forming large accumulations in these rocks, and also rinkite, lorenzenite, titanite, apatite, and others.

On its chemical composition lamprophyllite in «porphyric malignites» is strontian (Table 1, an. 12), however, also as in «malignites-U», it is often replaced in significant degree by lamprophyllite, enriched by barium and potassium. In late lamprophyllite the content of BaO achieves 19%, $K_2O - 2.6\%$ (Table 1, an. 14). On Fig. 1 the points, corresponding to chemical compositions of this lamprophyllite, lies in the area close to chemical composition of late Ba,Klamprophyllite from «malignites-U».

Genesis of lamprophyllite in the rocks of lujavrite-malignites complex

The analysis of the evolution of chemical composition of lamprophyllite-barytolamprophyllite series minerals from the studied complex rocks and the character of their alteration as a result of postmagmatic processes, compared with peculiarities of general evolution of accessory mineralization of lujavrite-malignites, allows in significant degree reconstructing the history of lujavrite-malignites formation. The analysis of alterations, detected for lamprophyllite from malignites of this complex, is high informative in genetic relation.

Barium lamprophyllite from «malignites-L». «Malignites-L» is seems to be mainly the product of lujavrites transformation as a result of K,Simetasomatosis, and barium lamprophyllite is here inherited from initial rock. This assumption is confirmed by following: 1) in malignites of these levels there are the mineral relics of primary assemblage, which is analogous to «lujavrite» one; 2) early postcrystallization changes, i. e. recrystallization of mineral grains, formation of poikilitic structure, corrosion of primary minerals by orthoclase; 3) gradual transition between lujavrites and «malignites-L», existing of «transitional» varieties with composition, corresponding to lujavrites, but combining trachytoid («lujavrite») and poikilitic («malignite») parts of rock. On this stage barium lamprophyllite is exposed to recrystallization and forms poikilocrystals.

The later transformation of lamprophyllite in «malignites-L» is a gradual replacement of barium lamprophyllite by significant strontian one, which, apparently, was influenced by late solutions, enriched by strontium and calcium. Formation of late «Ca-Sr-mineral assemblage», containing Sr-apatite, taseqite, pectolite, and fersmanite, in «malignites-L» can be explained by action of these solutions.

Lamprophyllite from «malignites-U». Most probably, early (strontian) lamprophyllite from «malignites-U» is also inherited from protorocks, in that case from trachytoid titanite melteigite-urtites, later undergone a K,Simetasomatosis transformation. The relics of these rocks in a number of cases are noted in «malignites-U». Moreover, the absence of sharp contacts between these rocks (they are connected to each other by gradually transitions), indirectly indicates the existence of their genetic connection with trachytoid melteigiteurtites. In trachytoid melteigite-urtites accessory lamprophyllite is not enough widespread. According to data of Arzamastsev et al. (1987), its amount is not exceeding 0.9% from total rock mass, which, probably, explains the rarity of lamprophyllite in «malignites-U».

The character of postmagmatic alteration of lamprophyllite, the barium and some calcium enrichment and corrosion by potash feldspar (identical to character of lamprophyllite alteration in massive melteigite-urtites, undergone an intensive transforming with formation of ristschorrites complex rocks during K,Si-metasomatosis), indicates the significant role of K,Si-metasomatosis in formation of «malignites-U». The similar scheme of lamprophyllite alteration was, in particular, described (Ageeva, 2001, 2002) for massive urtites, which is a main «matrix» for ristschorrites formation. The rising alkalinity of mineral formation environment, which causes the increase of activity of most basic components (in this case barium) and the decrease of activity of less basic ones (i. e. strontium), is a reason of barium activity increasing during K, Si-metasomatosis, according to O.A. Ageeva data.

In «malignites-U» as well as in «malignites-L» the more low-temperature lamprophyllite alteration takes place because of influence of late solutions. Here the character of lamprophyllite alteration is different: in contrast to «malignites-L», the development to titanite pseudomorphs after Ba,K-lamprophyllite is typical for these rocks. However, later transformation of «malignites-U», probably, was realized as a result of influence of the same solutions, enriched by calcium and strontium that solutions caused transformation of «malignites-L». The composition of late mineral assemblage, close to composition of «Ca,Sr-assemblage» of «malignites-L» indicates that. The former assemblage contains Sr-apatite, pektolite, pyrochlore, tasegite, and is practically distinguished from the later one only by wide prevalence of late titanite, forming pseudomorphs after lamprophyllite here.

Lamprophyllite from «porphyric malignites». As it is evident from above-mentioned data, the early strontium lamprophyllite in «porphyric malignites» is undergone by practically the same alterations that lamprophyllite in «malignites-U», these changes, apparently, caused by processes of K,Si-metasomatosis: lamprophyllite poikilocrystals are intensive corroded by late potash feldspar, the enrichment of strontium lamprophyllite by barium and potassium is observed. Small-grained trachytoid ijolites, spatially connected to «porphyric malignites», are, possibly, the primary rocks, which strontium lamprophyllite of «porphyric malignites» was inherited from.

Conclusion

The data, observed as a result of detail analysis of the chemical composition of lamprophyllite and the peculiarities of its postcrystallization alteration, allow determine following.

1. The enrichment by primary high-barium lamprophyllite is a typomorphic peculiarity of lujavrites from Khibiny. At present moment barium accessory lamprophyllite is noted neither in analogous rocks, lujavrites of Lovozero massif, no in other (leucocratic) nepheline syenites of large high-alkali massifs (Khibiny, Lovozero *etc*).

2. In «malignites-L» (genetically connected to lujavrites) and «malignites-U» (connected to melteigite-urtites) the character of alteration of lamprophyllite-barytolamprophyllite series minerals and minerals, associated with them, indicates two stages of their formation. The first stage is a transformation of initial rocks (lujavrites for «malignites-L» and trachytoid titanite ijolites for «malignites-U») during K,Simetasomatosis. In the case of «malignites-L» it



FIG. 4. Titanite pseudomorph after barium-potassium lamprophyllite in «malignites-U» (image in reflected electrons, scanning electron microscope JSM-5300, IGEM RAS). Ba,Klamp — barium-potassium lamprophyllite; Titn — titanite; Neph — nepheline; Aeg — aegirine-hedenbergite

is accompanied by recrystallization of primary barium lamprophyllite, forming poikilocrystals, without change of chemical composition; in the case of «malignites-U» there is enrichment of primary strontium lamprophyllite by barium and potassium. The second stage is the more low-temperature transformation, connected to influence of late solutions, enriched by strontium and calcium. In «malignites-L» barium lamprophyllite and barytolamprophyllite are replaced by proper lamprophyllite (strontian), in «malignites-U» titanite replaces them at this stage. The influence of these solutions causes calcium-strontium character of late mineralization in «malignites-L» and «malignites-U».

3. In «porphyric malignites» the alteration of early strontium lamprophyllite, inherited from their protorocks (small-grained trachytoid ijolites), caused by transformation of the latter during processes of K,Si-metasomatosis (as well as in «malignites-U»).

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