PALLADOARSENIDE Pd_2As – A PRODUCT OF MAYAKITE PdNiAs DESTRUCTION IN NORILSK SULFIDE ORES

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The paper describes mineral assemblages and genesis of mayakite and palladoarsenide in magmatic magnetite-pentlandite-chalcopyrite ores of the lower horizons of the Mayak Mine (Talnakh Deposit, Norilsk Ore Field). The mayakite studied here contained up to 1.5 wt.% Pt, whereas palladoarsenide contained up to 3 wt.% Cu and up to 2 wt.% Ni that substitute Pd. Microprobe analyses – 9 for mayakite and 4 for palladoarsenide – are presented in the paper. Palladoarsenide forms linear and branching metasomatic veinlets in mayakite and incomplete pseudomorphs after fine mayakite grains. Palladoarsenide is present at the locations where the ores are tectonized and have veinlets and metasomes of chlorite, carbonates, serpentine, anhydrite, makinawite, and magnetite. In these ore formations, ferroaugite is almost completed replaced by chlorite, carbonates, serpentine, and smectites. Possibly, palladoarsenide originated from the epigenetic processes of low-grade metamorphism (zeolite and prehnite-pumpellyite facies), which are widely abundant in the northwest of the East Siberian Plantform, where the Norilsk Ore Field is located.
8 figures, 2 tables, 27 references.
Key words: palladoarsenide, mayakite, norilsk sulfide ore, low-grade metamorphism.

Norilsk Ore Field

The Norilsk Co-Ni-Cu sulfide ore field contains 75% of the world resources of palladium and features a numerous amounts of palladium minerals. Those include stannides, plumbides, bismuthides, arsenides, antimonides, bismuthides-tellurides, cuprides-stannides, and other intermetallides (Genkin, 1968; Kulagov, 1968; Begizov *et al.*, 1974; Zolotukhin *et al.*, 1975; Genkin *et al.*, 1976; Begizov, 1977; Genesis..., 1981; Evstigneeva and Genkin, 1986; Mitenkov *et al.*, 1997; Naldrett, 2004; Spiridonov, 2010).

The Norilsk Ore Field is located in the northwestern corner of the ancient East Siberian Platform, in the zone of edge dislocations. The Norilsk deposit in the southwestern part of the ore field and the Talnakh deposit in the northeastern part of it belong to the plate cover of the platform. In this region, the Riphean-Vendian-Paleozoic sedimentary formations of the plate cover have the maximum thickness up to 14 km. The plate cover in the Norilsk region is dislocated, split by numerous faults, its sedimentary rocks and overlaying volcanic formations folded to form systems of flat brachysynclines and somewhat steeper anticlines. The cores of the brachysynclines are built by the multi kilometer strata of plateau basalts, traps formation, P_2-T_1 (Godlevskii, 1959). Some of the latest elements of the traps are gently dipping band-shaped ore-bearing gabbroid intrusions. The deposits are associated to the Norilsk, Upper Talnakh, and Taimyr intrusions of olivine gabbro-dolerites, gabbro-norite-dolerite, gabbro-anorthosites, gabbro-peridotites, troctolites, and gabbro-diorites. These rocks intruded along the zone of the steeply dipping Norilsk-Kharaelakh Fault with northeastern strike, and cross-cut the plate cover down to the bottom of the basalt strata. Magmatic sulfides are present as dissemination in ore-bearing intrusives, bodies, veins, and impregnations at the lower endocontact of these intrusives with underlying hornfelses; less abundant – in the main bulk of the intrusive and in the supra-intrusive zone. The isotope age of all constituents of the traps formation – volcanites, intrusive rocks, and magmatic Ag-Au-Pt-Pd-Co-Ni-Cu sulfide ores is 251 Ma (Naldrett, 2004; Spiridonov and Gritsenko, 2009). The isotope composition of lead in the ore-bearing intrusive rocks, magmatic sulfide ores, galena and Pd-Pt intermetallides of the Norilsk and Talnakh deposits is quite different. The isotope composition of lead of intrusive rocks, magmatic sulfide ores, and Pd-Pt intermetallides at Talnakh deposit is noticeably more radiogenic (Spiridonov *et al.*, 2010). This is evidence to the genetic relation of the ores to particular intrusions.

According to metaphoric expression of Mikhail Nikolaevich Godlevskii, the Norilsk sulfide ore is the "kingdom of sulfide solid solutions". Crystallization differentiation is the governing process in solidification of the Co-Ni-Cu-Fe sulfide melts. The early crystallization products are represented by the monosulfide solid solution (Mss) with hexagonal structure and composition close to pyrrhotite. Later crystallization products, enriched in copper to a various degree, Pd, Pt, Ag and Au, are represented by the intermediate solid solution (Iss) with cubic structure and composition varying from cubanite to chalcopyrite with variable nickel content. The lower and marginal zones of the magmatic sulfide deposits are composed of pyrrhotite group minerals — the products of solid phase transformations of Mss. These are gradually replaced by pyrrhotite, troilite, chalcopyrite, and cubanite in various combinations, and other minerals of the chalcopyrite group (talnakhite, mooihoekite) — the product of solid phase transformations of Iss. Both these varieties contain a significant amount of pentlandite as decomposition products of the Mss and Iss.

Minerals of platinum group elements in norilsk sulfide ores

Although the minerals of noble metals were extensively studied by M.N. Godlevskii, E.A. Kulagov, A.D. Genkin, A.A. Filimonova, T.L. Evstigneeva, V.D. Begizov, C.F. Sluzhenikin, G.A. Mitenkov, N.S. Rudashevskii, V.A. Kovalenker, E.V. Sereda, E.N. Sukhanova, I.N. Tushentsova, A.P. Glotov, and other researchers, we are only starting to understand the actual complexity of the noble metal mineral formation in the Norilsk sulfide ore deposits. The major part of Pd, Pt, Au, and Ag in these ores is represented by their own minerals, and minor quantities of these metals are contained in sulfides (e.g. palladium and silver in pentlandite, etc.). The minerals of platinum group elements (PGM) are similar in all the ore types, from pyrrhotite ores to chalcopyrite ores, talnakhite ores, and mooihoekite ores, only the quantitative ratios of PGM vary (Mitenkov *et al.*, 1997; Spiridonov, 2010).

Most geologists believe that PGM are products of magmatic crystallization. In 1960s, A.D. Genkin proposed that PGM could form at late-magmatic or post-magmatic conditions under the action of fluids (Genkin, 1968). Later on, it was proved (Kulagov, 1968; Genkin *et al.*, 1976; Evstigneeva and Genkin, 1986; Spiridonov *et al.*, 2003) that some of the PGM are metacrystals. The most recent observations show that the all of PGM in the norilsk ores originated from the replacing of the sulfide aggregates, silicates, and oxides in intrusive rocks and hornfelses (Spiridonov, 2005; 207; 2010). The exocontact zones of the sulfide aggregates, as well as the silicate matrix of the hornfelses spacially separated from the sulfides, contain metacrystals of atokite, tetraferroplatinum, rustenburgite, michnerite, sperrylite (formations up to 8 mm in size present in hornblende aggregates), electrum, mayakite, polarite, paolovite, and kotulskite. The size of the PGM metasomes is determined by the size of sulfide bodies: e.g. a few microns near the sulfide drops. Hence, the PGM genesis is likely penumatolytic. When they form, Pd, Pt, Au, Ag, Sn, Te, As, Sb, and Bi are brought by the fluids that originated at the stage of sulfide formation; in this process, Cu, Pb, Fe, and Ni are extracted from the replaced sulfide minerals. Through this mechanism, mayakite replaced pentlandite (pentlandite relics are common in mayakite metacrystals) (Spiridonov *et al.*, 2004). Some of the zones of the sulfide ores are dominated by stannides of Pd and Pt, whereas another zone may contain antimonides of Pd and Pt, and the next one — bismuthides and bismuthides-tellurides of Pd and Pt, and others — arsenides and arsenides-stannides of Pd and Pt. The PGM in Norilsk sulfide ores formed at extremely low sulfur fugacity ($f\ S_2$) under highly reducing conditions.

Early stage pneumatolytic PGM. These are the solid solutions with extensive replacement of Pd-Pt-Au and Sn-Sb-Bi-Pb-Te-As with characteristic decomposition patterns. Intergrowths of gold-containing tetraferroplatinum Pt_2Fe (Fe, Ni,Cu) with lamellae of gold-platinum-lead-bearing atokite Pd_3Sn as well as intergrowths of gold-platinum-lead-bearing atokite Pd_3Sn with lamellae of tetraferroplatinum are quite common. Also abundant are PGM metacrystals with the bulk composition (Pd,Pt)(Sn,Sb,Bi,Te,As) — equiatomic platinoid solid solutions. These minerals are composed of the products of decomposition of equiatomic platinoid solid solutions, which are usually represented by antimony-bearing paolovite (matrix) associated with a mass of geversite ($PtSb_2$) — insizvaite ($PtBi_2$) lamellae and niggliite (PtSn) microgrowths. The products of joint recrystallization of the above minerals are widely present.

Middle stage pneumatolytic PGM are mainly the products of transformation of the early

PGM, which typically occurs with addition of tellurium. Altaite PbTe and moncheite $Pt(Bi,Te)_2$ are products of this process. Moncheite plates are often overgrown by aggregates of zonal crystals of maslovite (PtBiTe), tellurium- and bismuth-bearing geversite, and tellurium- and antimony-bearing insizvaite. Alteration of early gold-bearing tetraferroplatinum and atokite with breakdown lamellae resulted in the formation of the aggregates of tetraferroplatinum (without breakdown patterns), zoned crystals of rustenburgite (Pt_3Sn), and atokite (Pd_3Sn) (also without breakdown patterns). These minerals are associated with zvyagintsevite (Pd_3Pb), stannopalladinite (Pd_5CuSn_2), and Au-Cu minerals. Low-tellurium parageneses are present: (1) thin-lamellar and graphic intergrowths of polarite (Pd_2PbBi), stannopalladinite, plumbopalladinite (Pd_3Pb_2), associated with mayakite and tetraferroplatinum; (2) plate aggregates of paolovite, paolovite with antimony-bearing paolovite, paolovite with stibiopalladinite (Pd_5Sb_2), all cemented by bismuth-bearing geversite and antimony-bearing insizvaite. Later minerals in this stage include taimyrite $(Pd,Pt)_9Cu_3Sn_4$ and tatyanaite $(Pt,Pd)_9Cu_3Sn_4$, which form replacement borders around large crystals of rustenburgite-atokite and pseudomorphs after their smaller crystals.

Late stage pneumatolytic PGM. The late pneumatilytic PGM are dominated by palladium minerals associated with minerals of Au-Ag, altaite, and hessite. The aggregates of these minerals grow over the earlier PGM and Au-Cu minerals, corroding them and forming separated nests. Commonly present are tellurium-bearing sobolevskite (PdBi), frudite ($PdBi_2$), non-zonal and zonal electrum and kustelite, bismuth-free geversite, antimony-free insizvaite, and cabriite (Pd_2CuSn).

Latest pneumatolytic PGM. The latest-formed mineral is sperrylite ($PtAs_2$), its crystal size varying from several microns to 11 cm. The sperrylite crystal boundaries cross-cut tetraferroplatinum, paolovite, insizvaite, geversite, rustenburgite, atokite, taimyrite, sobolevskite, frudite, cabriite, electrum and kustelite.

Mayakite PdNiAs in the Talnakh ore deposit, Mayak mine

Mayakite – monoclinic PdNiAs – was discovered and studied by T.L. Evstigneeva (Genkin *et al.*, 1976; Evstigneeva *et al.*, 2000) in the ore specimens from the eastern wing of the Komsomolskii mine, Talnakh ore deposit. Further on, mayakite from Talnakh deposit was described in several published studies (Begizov, 1977; Barkov *et al.*, 2000; Spiridonov, 2010).

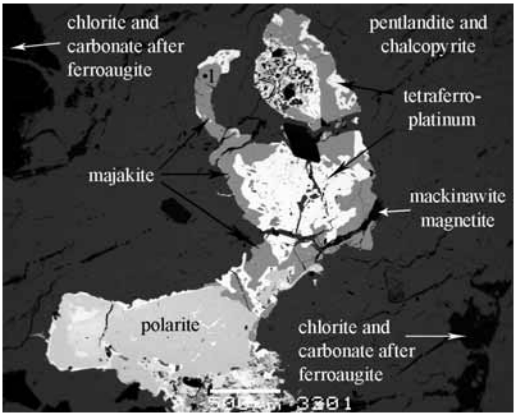
The Mayak mine locates in the southeastern part of the Talnakh Deposit. The lower part of the Upper Talnakh ore intrusion and underlying hornfelses contain lenses and veins of massive sulfide ores of pentlandite-chalcopyrite composition, sometimes with noticeable contents of magnetite (at the upper contact of the gently dipping veins) and ferroaugite. PGM form small metasomes in sulfide ores, which are distributed quite unevenly and are typically associated with either the upper selvage of gentle veins and bodies or lower contacts of xenoliths gabbro-dolerite and hornfelses in sulfide veins. PGM are more rare in silicate matrix at the exocontact zones of the sulfide ores. The size of the nests composed by noble metals ranges up to 7 mm, but typically less than 1 mm. The PGM are predominantly related to the middle stage and in some places contain significant amount of sperrylite.

Mayakite is present in magnetite-pentlandite-chalcopyrite matrix as separated metacrystals, oval-shaped metasomes, more often polymineral aggregates with tetraferroplatinum, polarite, palarstanide $Pd_3(As,Sn)$, plumbopalladinite, stannopalladinite (Fig. 1 – 4). In these aggregates, mayakite and other PGM are often overgrown by platinum, palladium-bearing tetraauricupride AuCu (Fig. 5), and zvyagintsevite (Fig. 6). Also, mayakite is present in intergrowths with stillwaterite Pd_8As_3 (Fig. 6), and the contacts of mayakite with polarite are often traced by frudite borders (Fig. 7).

The locations where the mayakite and palladoarsenide specimens were taken for microprobe analysis are marked by numbers in the figures; the numeration of analyses in the figures and tables is identical. The chemical composition of mayakite is quite constant with respect to main components (Table 1). The contents of impurity elements vary quite widely (wt.%): Pt 0 – 1.4, Fe 0.1 – 0.5, Au 0 – 0.2, Pb 0.01 – 0.4. The average composition of mayakite is given by (based on 9 analyses, wt.%): Pd 43.31; Pt 0.36, Au 0.09; Ni 24.39; Fe 0.14; Cu 0.09; As 31.38; Pb 0.10; Sn 0.05; Bi 0.05; Te 0.02; Total 99.98%; Calculation for 3 atoms correspondes the formula $Pd_{0.98}Ni_{1.00}Fe_{0.01}As_{1.01}$.

Palladoarsenide Pd_2As as a product of mayakite PdNiAs alteration in the sulfide ores of the Talnakh deposit, Mayak mine

Palladoarsenide – monoclinic Pd_2As – was discovered and studied by V.D. Begizov in the ores of the Talnakh deposit, which are developed by the Oktyabrskii mine (Begizov *et al.*, 1973; Begizov, 1977). The first find of palladoarsenide is represented by small veinlets and irregular-



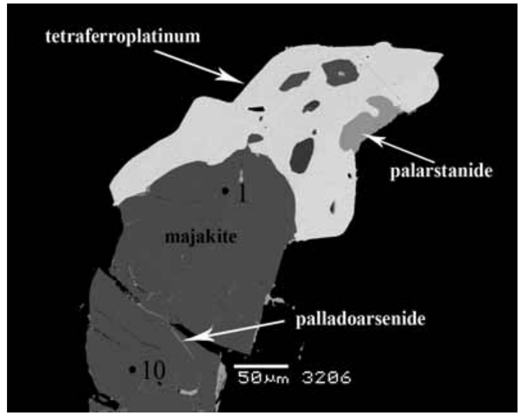
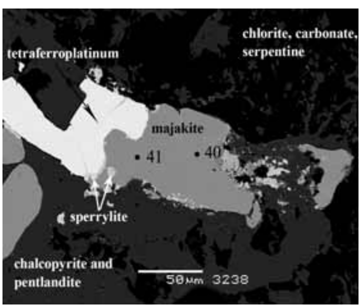


Fig. 1. Intergrowth of mayakite with tetraferroplatinum and polarite in the matrix of chalcopyrite and pentlandite. The ores contain veinlets of mackinawite, magnetite, chlorite, and carbonates. Ferroaugite is replaced by chlorite and carbonates. (BSE=image). From here on: specimen location: deep horizons of the Mayak Mine, Talnakh deposit. Numbers indicate the place and analysis for a particular mineral studied by electron microprobe.

Fig. 2. Intergrowth of mayakite, tetraferroplatinum, and palarstanide; thin streaks of palladoarsenide in mayakite (detail from Fig. 1) (BSE=image).



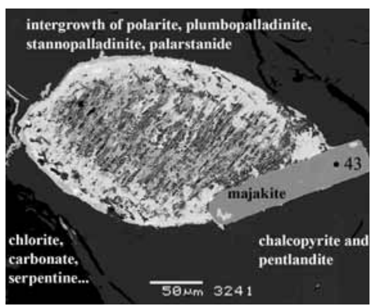
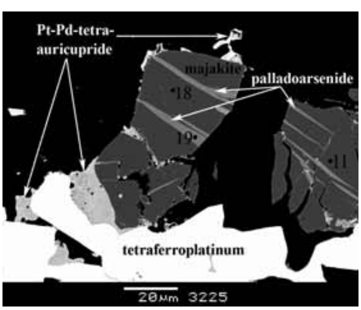


Fig. 3. Intergrowth of mayakite with tetraferroplatinum. Ferroaugite in sulfide matrix is replaced by aggregates of chlorite, carbonates, and serpentine (BSE=image).

Fig. 4. Intergrowth of a regular-shaped mayakite metacrystal with an oval aggregate of polarite plates, plumbopalladinite, stannopalladinite, and palarstanide in the matrix of chalcopyrite and pentlandite. Ferroaugite in the sulfide matrix is replaced by chlorite, carbonates, and serpentine (BSE= image).



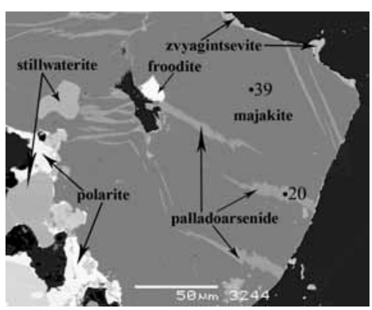
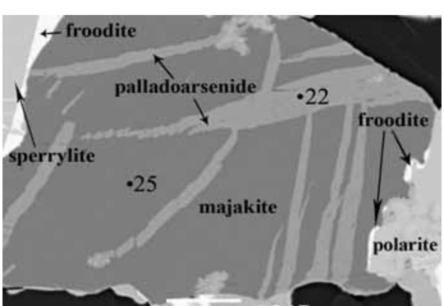


Fig. 5. Intergrowth of mayakite and tetraferroplatinum with a borber of platinum and palladium-bearing tetra-auricupride. Mayakite contains a net of metasomatic palladoarsenide veinlets. (BSE= image).

Fig. 6. Intergrowth of mayakite, stillwaterite, polarite, zvyagintsevite, and frudite. Mayakite contains a net of metasomatic palladoarsenide veinlets. (BSE= image).



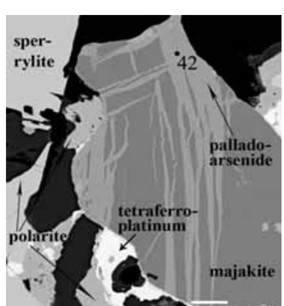


Fig. 7. Intergrowth of mayakite, polarite, and frudite. Mayakite contains a net of metasomatic palladoarsenide veinlets. (BSE=image). The scale bar — 10 microns.

Fig. 8. Intergrowth of mayakite, tetraferroplatinum, and polarite. Mayakite contains a net of metasomatic palladoarsenide veinlets. (BSE=image). The scale bar — 10 microns.

Table 1. **Chemical composition of mayakite (wt.%) from magnetite-pentlandite-chalcopyrite ores, Mayak mine, lower horizons**

Components	Analysis №								
	1	10	11	18	25	39	40	41	43
Pd	42.73	42.90	42.80	42.41	43.29	43.82	44.00	43.53	44.34
Pt	1.06	nd	0.80	nd	nd	1.40	nd	nd	nd
Au	nd	0.15	0.03	0.14	0.21	nd	0.03	0.22	nd
Ni	24.60	25.01	24.24	24.69	24.73	23.03	24.63	24.40	24.14
Fe	0.48	0.09	0.10	0.17	0.07	0.08	0.07	0.08	0.12
Cu	0.08	0.12	0.07	0.07	0.04	0.18	0.02	0.13	0.13
As	31.10	31.14	31.65	32.45	31.25	31.40	31.50	30.82	31.10
Bi	0.08	0.06	nd	0.07	0.09	0.07	nd	nd	0.11
Sn	0.05	0.08	0.13	nd	0.01	0.03	0.10	0.05	nd
Pb	nd	0.38	nd	0.01	nd	0.19	nd	0.30	0.06
Te	nd	0.02	0.07	nd	nd	nd	nd	0.09	0.01
Total	100.08	99.95	99.89	100.01	99.69	100.17	100.35	99.62	100.01
Atomic coefficient in formula calculated for 3 total atoms									
Pd	0.987	0.992	0.993	0.975	1.002	1.025	1.012	1.012	1.027
Pt	0.013		0.010			0.017			
Au		0.002		0.002	0.003			0.003	
Total	1.000	0.994	1.003	0.977	1.005	1.042	1.012	1.015	1.027
Ni	0.992	1.007	0.982	0.991	1.000	0.940	0.990	0.991	0.976
Fe	0.020	0.004	0.004	0.007	0.003	0.003	0.003	0.003	0.005
Cu	0.003	0.004	0.003	0.003	0.001	0.007	0.001	0.005	0.005
Total	1.015	1.015	0.989	1.001	1.004	0.950	0.994	0.999	0.986
As	0.983	0.984	1.004	1.021	0.990	1.004	0.992	0.980	0.985
Bi	0.001	0.001		0.001	0.001	0.001			0.001
Sn	0.001	0.002	0.003			0.001	0.002	0.001	
Pb		0.004				0.002		0.003	0.001
Te			0.001					0.002	
Total	0.985	0.991	1.008	1.022	0.991	1.008	0.994	0.986	0.987

Note: Electron microprobe Camebax, analyst I.M. Kulikova. Sb — not detected; nd — below detection limit.

shaped grains in chalcopyrite. It is not mentioned in later descriptions of the Norilsk Ore Field. As an accessory mineral, palladoarsenide was found in many deposits containing noble metals: chromites in the Stillwater layered basite-hyperbasite massif, USA (Volborth *et al.*, 1986), Bushveld, South Africa (Oberthür *et al.*, 2004), alpine type massifs of Oman (Auge, 1986; Ahmed, Arai, 2003), sulfide ores associated with Canadian komatiites (Chen *et al.*, 1993), Bulgarian porphyry-copper deposits (Auge *et al*.., 2005), and telethermal gold-palladium deposits (Cabral *et al.*, 2002).

We identified palladoarsenide in PGM aggregates with mayakite in the ores of deep horizons of the Mayak mine. Palladoarsenide forms straight and branching metasomatic veinlets in mayakite, incomplete pseudomorphs after small grains of mayakite (Fig. 5—8). The veinlets range up to 70 micron in length and 8—12 micron in thickness. In reflected light, palladoarsenide is almost undistinguishable from the surrounding mayakite. Therefore, its reliable identification is only possible by electron microscopy. This type of analysis provides new quality estimates of the chemical composition of the rare palladium arsenide (Table 2).

In the palladoarsenide, significant path of palladium (up to 10% of atoms) is substituted by copper, nickel, and iron, and, to a less extent, by platinum and gold. Copper impurities can comprise up to 2.8 wt.%, and nickel 2.2 wt.%. Note that there is almost no copper is present in the parent mineral, mayakite. In the studied palladoarsenide, only a negligible fraction of arsenic atoms is substituted for lead, bismuth, tin, and tellurium. The average composition of the palladoarsenide from the metasomatic veinlets in mayakite of the Talnakh deposit is as follows (based on 4 analyses, wt.%): Pd 68.67; Pt 1.03, Au 0.13; Cu 1.49; Ni 1.35; Fe 0.34; As 26.10; Pb 0.17; Sn 0.03; Te 0.03; Bi 0.02. This composition recalculated for 3 formula atoms corresponds the formula $(Pd_{1.84}Pt_{0.02}Cu_{0.07}Ni_{0.06}Fe_{0.02})_{2.01}As_{0.99}$. Probably, the mechanism of palladoarsenide formation is: $2PdNiAs \rightarrow Pd_2As + 2Ni\ \ell + As\ \ell$.

Table 2. **Chemical composition of palladoarsenide (wt.%) in metasomatic veinlets in mayakite from magnetite-pentlandite-chalcopyrite ores, Mayak mine, lower horizons**

Components	Analysis №			
	42	19	20	22
Pd	68.08	69.15	67.84	69.59
Pt	3.75	nd	0.35	nd
Au	0.17	nd	0.36	nd
Cu	0.44	2.21	2.77	0.54
Ni	0.81	1.17	1.23	2.19
Fe	0.66	0.21	0.18	0.30
As	25.85	26.36	25.86	26.32
Bi	nd	0.06	nd	nd
Pb	nd	nd	0.44	0.23
Sn	0.07	nd	nd	0.04
Te	0.09	nd	0.04	nd
Total	99.92	99.16	99.07	99.21
Atomic coefficient in formula calculated for 3 total atoms				
Pd	1.850	1.840	1.810	1.845
Pt	0.056		0.005	
Au	0.002		0.005	
Cu	0.020	0.100	0.125	0.025
Ni	0.040	0.055	0.060	0.105
Fe	0.034	0.010	0.010	0.015
Total	2.002	2.005	2.015	1.990
As	0.998	0.995	0.980	1.005
Pb			0.005	0.005

Note: Electron microprobe Camebax, analyst I.M. Kulikova. Sb — not detected; nd — below detection limit.

Palladoarsenide occurs in the zones where ores are sectioned by tectonic fractures and contain veinlets and metasomes of chlorite, carbonates, serpentine, anhydrite, mackinawite, and magnetite. In these ores, ferroaugite is almost entirely replaced by chlorite, carbonates, serpentine, and smectites (Fig. 1, 3, and 4). The formation of palladoarsenide is likely to be related to epigenetic processes of low-grade metamorphism in condition of zeolite and prehnite-pumpellyite facies, which are well represented in the northwest of the East Siberian Platform (Spiridonov *et al*., 2000; Spiridonov and Gritsenko, 2009). Mobility and addition of copper, which occurs on replacement of mayakite by palladoarsenide, is a characteristic process of low-grade metamorphism. Nickel and arsenic released due to destruction of mayakite might be involved in the formation of arsenide-carbonate veins, which are very common in the Talnakh region (Spiridonov and Gritsenko, 2009).

In the chromite ores of Stillwater (Volborth *et al*., 1986) and Oman (Auge, 1986; Ahmed, Arai, 2003), as well as in the sulfide ores of the Thompson komatiite belt (Chen *et al*., 1993), palladoarsenide is present in the serpentine zones, which correlates with our observations in Talnakh.

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