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RE-EXAMINATION OF Pt ALLOYS FROM LODE AND PLACER DEPOSITS, URALS

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ABSTRACT

"Cuproplatinum", originally reported from the Urals, is shown to be tulameenite, ideally Pt_2FeCu, with an *a* of 3.9018(7), a *c* of 3.5845(13) A, and $Z = 1$. Tulameenite is a secondary mineral formed during serpentinization of ultramafic massifs and is usually found as a rim on primary platinum alloys. It occurs both in lode deposits and within placers derived from such deposits.

Keywords: tulameenite, "cuproplatinum", Urals, Nizhni Tagil, platinum lodes, Omutnaya river, Tulameen river, platinum placers, Pt alloys, platinum-group minerals, ultramafic massifs, serpentinization.

SOMMAIRE

Notre caractérisation du "cuproplatine" de l'Oural, en URSS, montre qu'il s'agit de la tulameenite. Elle possède une composition idéale Pt_2FeCu, et les paramètres réticulaires *a* égal à 3.9018(7), et *c* égal à 3.5845(13) A, pour $Z = 1$. La tulameenite est un minéral secondaire formé lors de la serpentinisation des massifs ultramafiques; elle se présente généralement sous forme de bordure sur les grains d'alliages primaires de platine. On la trouve aussi bien dans les gisements *in situ* que dans les graviers alluvionnaires qui en sont dérivés.

(Traduit par la Rédaction)

Mots-clés: tulameenite, "cuproplatine", Oural, Nizhni Tagil, gisements de platine, rivière Omutnaya, rivière Tulameen, graviers platinifères, alliages de Pt, minéraux du groupe du platine, massifs ultramafiques, serpentinisation.

INTRODUCTION

The discovery of platinum placers in the Urals occurred in 1819 when a silvery metal from the alluvial gold diggings south of Ekaterinburg (Sverdlovsk) was submitted for analysis. By the end of 1825, platinum alloys had been found in the gravels of rivers that drain the foothills of 11 localities of ultramafic instrusive bodies over a distance of about 500 km. In 1890, the first *in situ* platinum deposit was found, in the dunite massif at Nizhni Tagil (about 115 km NNW of Sverdlovsk), located approximately in the middle of a homogeneous metallogenetic province that extends for more than 600 km along the axial part of the Ural mountains (Betekhtin 1961). Thus, the years 1819 and 1890 may be considered historical milestones in the study of the mineralogy and geology of platinum deposits. It is also interesting to record that, though the first discovery of primary platinum mineralization consisted of a relatively small chromite body, exploration of the platiniferous dunite massifs of the Ural Mountains resulted in the discovery of approximately 600 sites of primary Pt mineralization with high Pt contents, and almost always in association with chromite (Betekhtin 1961). In their monograph, Duparc & Tikonowitch (1920) collected all the chemical data known up to that time on the Urals deposits, especially those reported in the first paper of Wyssotzky[1] (1903), as well as results of their own analyses, performed in Geneva. Copper was reported by Duparc & Tikonowitch (1920) in some of the analyses of platinum from Nizhni Tagil, with 5.2 wt.% as the highest value. In a later book, Vysockiy (1923) first mentioned copper-bearing platinum. He wrote that, besides iron-bearing platinum, platinum free of iron, palladium-, rhodium-, and iridium-bearing platinum, there exist also copper-bearing varieties, with the greatest copper contents occurring in varieties rich in iron and nickel. He reported a high copper content of 8.2 wt.% from Nizhni Tagil.

"Cuproplatinum" was first discovered by A.G. Betekhtin and A.N. Zavaritsky in 1924 (Betekhtin 1935) in lode ores in the Nizhni Tagil deposit. Betekhtin (1935) described "cuproplatinum" in reflected light as follows: white, compared to native platinum, with a greenish tint, isotropic, and softer than native platinum. Betekhtin (1961), for the first

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[1] Also spelled Wyssotsky, Vysockiy, Vysockij, Vysotsky, Visotsky, Vysotski, Vuisotzki, Visotski.

time, however, pointed out that in some polished sections, "cuproplatinum" with a high copper content is anisotropic. The chemical composition was given as Pt(+Ir) 65–78%, Fe 13–17%, Cu 5–13%, Ni 0.5–1.5%, corresponding to chemical formulas over the range Pt_5Fe_4Cu – $Pt_3Fe_3Cu_2$. Betekhtin stressed that "cuproplatinum" replaces native platinum during serpentinization. No X-ray-diffraction studies were reported on "cuproplatinum".

Cabri *et al.* (1973) found an alloy of platinum, iron, and copper in the platinum-bearing placers of the Tulameen river area, British Columbia, which they named *tulameenite* with the approval of the Commission on New Minerals and Mineral Names, International Mineralogical Association. Tulameenite was described as tetragonal, with $a = b = 3.891$ and $c = 3.577$ A.

In the early 1970s, the second author began to restudy (in Moscow) Betekhtin's collection of polished sections containing platinum alloys from the Urals. During this investigation, the new mineral *rhodplumsite* ($Rh_3Pb_2S_2$) was found (Genkin *et al.* 1983), and new varieties of ruthenium- and rhodium-bearing pentlandite were described (Genkin *et al.* 1974). Re-examination of Betekhtin's "cuproplatinum" by electron microprobe and X-ray diffraction led the second author to conclude that this mineral is identical to the tulameenite of Cabri *et al.* (1973), but these results were not published. During a visit to Moscow by the first author in 1989, it was decided to collaborate in order to compare the platinum-group minerals from the Urals with those from the Tulameen area. Some of the results of this collaboration are reported here.

SAMPLES AND METHODS

The samples studied originated both from platinum placers (Nos. 53, 64) and from primary platinum lodes (Nos. 45, 54, 59, 62). The placer samples are polymineralic nuggets (up to 12 mm in diameter) consisting largely of platinum alloys encapsulated in chromite from the Omutnaya river, Sissertj (near Sverdlosk). The samples from primary lode deposits are from the Nizhni Tagil dunite massif (Betekhtin 1961) and consist of platinum alloys within chromite aggregates, all enclosed by serpentinite.

Electron-microprobe analyses were done at CANMET on a JEOL 733 microanalyzer system. Analyses of platinum-group minerals and platinum-group-element-bearing minerals were carried out by wavelength-dispersion spectrometry at 20 kV, with a beam current of 20 nA, using the X-ray lines and standards reported by Nixon *et al.* (1990).

RESULTS AND GENERAL DESCRIPTION OF SAMPLES

The platinum-group minerals (PGM) identified by electron-microprobe analyses are indicated by an "x" in Table 1. X-ray diffraction was only performed on tulameenite from sample #45.

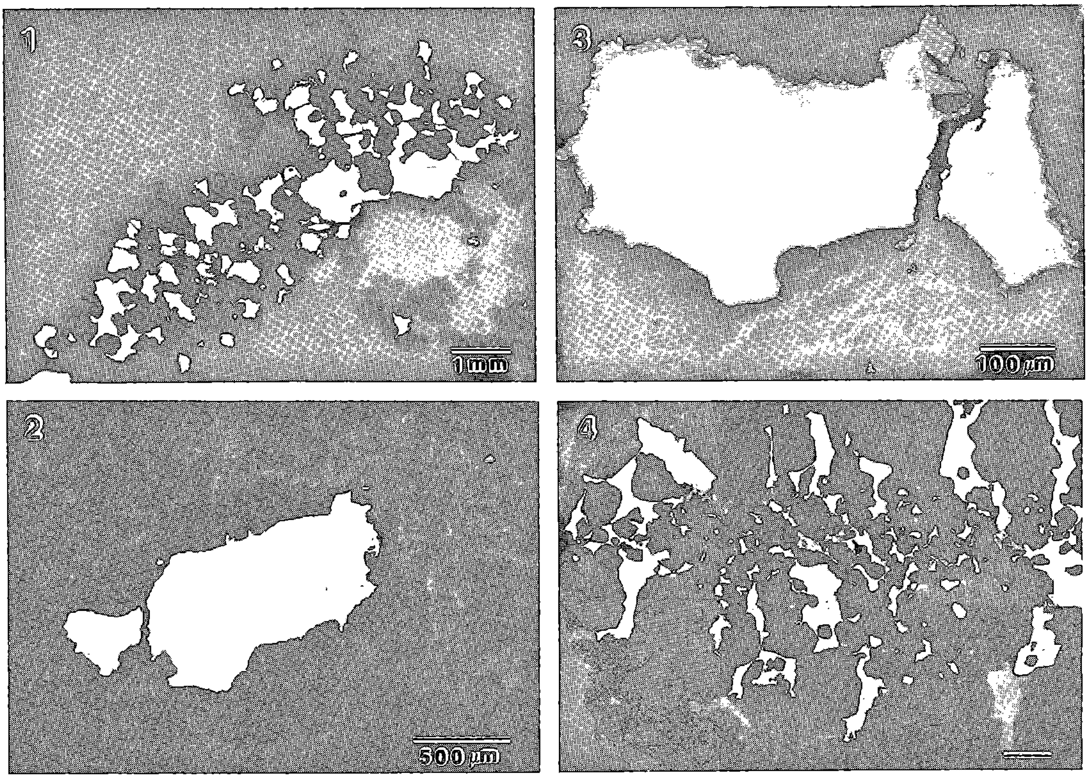
TABLE 1. PLATINUM-GROUP-ELEMENT-BEARING MINERALS FOUND IN BETEKHTIN'S SAMPLES

	lode samples				placer samples	
	#45	#54	#59	#62	#53	#64
Tulameenite	x	-	-	-	x	x
platinian palladian Cu	x	-	-	-	-	-
ruthenian platinian Cu	x	-	-	-	-	-
Osmium*	x	x	-	-	x	x
Iridium*	-	x	x	x	x	x
Tetraferroplatinum	-	x	x	x	-	-
Isoferroplatinum	-	x	x	x	x	x
Bowieite	-	-	-	-	x	x
Kashinite	-	-	-	-	x	x
Laurite	x	x	x	x	x	x
Rhodplumsite	-	-	-	-	x	-
UK Rh-Ir-Pt-sulfide	-	-	-	-	x	-

* using the nomenclature of Harris & Cabri (1991).

Lode samples

Sample #45 consists of tulameenite aggregates (as much as 3 mm by 10 mm) with heavily fractured cumulus chromite in a matrix of serpentine (Fig. 1). The size of the Pt-alloy masses and their association with chromite are similar to those of the placer nuggets described from Tulameen, British Columbia (*cf.* Fig. 11, Nixon *et al.* 1990). This should help allay concerns raised in the literature on the dissimilarity in size between concentrations of Pt-alloys in lodes and placer nuggets. Some of the tulameenite grains occur in serpentine, without chromite, and have a very irregular outline (Fig. 2). Tulameenite grains (analytical data in Table 2) may have a rim and contain microinclusions of platinian palladian copper ($Cu_{0.78}Pt_{0.19}Pd_{0.03}Fe_{0.01}Ni_{<0.01}$), which may be compared (Fig. 3) to similar occurrences at Tulameen (Nixon *et al.* 1990) and Thetford, Quebec (Corrivaux & Laflamme 1990). The native copper, in some cases, is a so-far-unreported ruthenian platinian variety [($Cu_{0.53}Ru_{0.22}Pt_{0.13}Os_{0.06}Pd_{0.02}Rh_{0.01}Fe_{0.01}Ni_{0.01}Ir_{<0.01}$), average of 3 analyses]. Tulameenite grains contain a few tabular and vein-like inclusions of osmium ($Os_{0.91}Ir_{0.05}Ru_{0.02}Pt_{0.01}Rh_{<0.01}$); one lath is actually included in chromite. Minor laurite, associated with a greyish anisotropic alteration product (Ru $\gg$ Os $>$ Mg $>$ Si $\gg$ Fe), also occurs in tulameenite Sample #54 consists of massive, relatively unfractured chromite with disseminated isoferroplatinum forming an



FIGS. 1-4. 1. SEM photomicrograph, back-scattered-electron image (BEI) of tulameenite with subhedral chromite within massive serpentinite. Sample #45. 2. Optical photomicrograph showing tulameenite grains with irregular ("corroded"?) margins in serpentine, without associated chromite. Sample #45. 3. SEM photomicrograph (BEI) of an area of tulameenite shown in Figure 2, illustrating typical rim and fine inclusions of platinian copper (grey) surrounded by fractured chromite. Sample #45. 4. A composite made from six optical photomicrographs showing the extensive distribution of isoferroplatinum and minor laurite within a chromite-rich matrix. Scale bar = 1 mm. Sample #54.

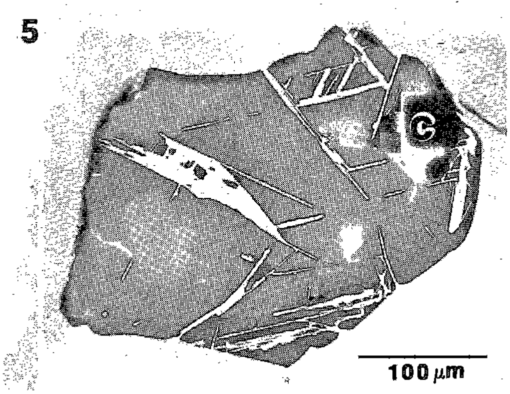
TABLE 2. RESULTS OF ELECTRON-MICROPROBE ANALYSES OF TULAMEENITE

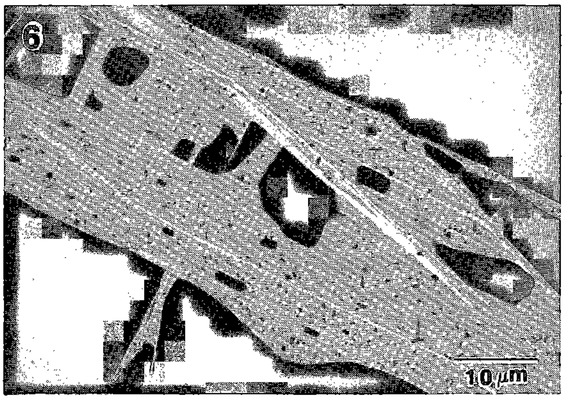
			weight per cent				
Sample	Pt	Ir	Rh	Fe	Cu	Ni	Totals
#45 Grain 1	76.4	0.28	n.d.	11.7	10.8	0.71	99.89
#45 Grain 2	76.7	0.29	n.d.	12.0	10.5	0.78	100.27
#45 Grain 3	75.9	0.32	n.d.	11.4	11.2	0.67	99.49
#45 Grain 4	76.3	0.30	n.d.	11.3	11.7	0.68	100.28
#45 Grain 5	76.1	0.28	n.d.	12.3	10.3	0.94	99.92
#45 Grain 6	76.2	0.31	n.d.	11.7	11.0	0.74	99.95
#53 12 anal.	75.9	1.8	0.12	10.7	12.2	0.15	100.87
#64 4 anal.	76.8	1.2	0.39	12.8	9.7	0.29	101.18
			atomic proportions				
#45 Grain 1	2.00	<0.01	-	1.07	0.87	0.06	2.00:2.00
#45 Grain 2	2.00	<0.01	-	1.09	0.84	0.07	2.00:2.00
#45 Grain 3	1.99	0.01	-	1.04	0.90	0.06	2.00:2.00
#45 Grain 4	1.98	0.01	-	1.02	0.93	0.06	1.99:2.01
#45 Grain 5	1.98	0.01	-	1.11	0.82	0.08	1.99:2.01
#45 Grain 6	1.98	0.01	-	1.06	0.88	0.06	1.99:2.00
#53 12 anal.	1.98	0.05	<0.01	0.98	0.98	0.01	2.03:1.97
#64 4 anal.	1.99	0.03	0.02	1.16	0.77	0.03	2.04:1.96

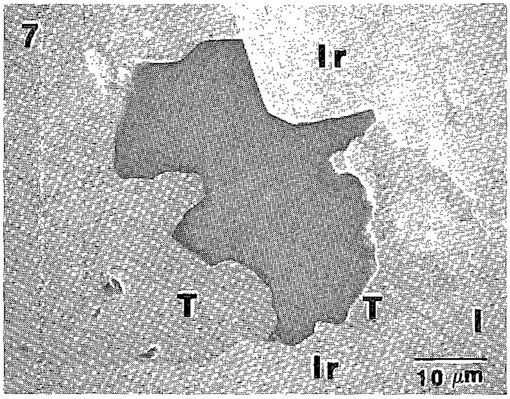
Os, Ru, Pd and Sb were sought, but not detected.

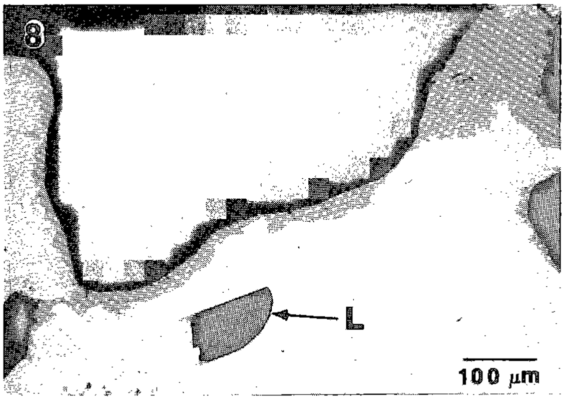
area of 7 × 22 mm (Fig. 4). The isoferroplatinum ranges in composition from $(Pt_{2.38}Ir_{0.24}Rh_{0.05}Pd_{0.01})_{\Sigma 2.68}(Fe_{1.13}Ni_{0.12}Cu_{0.07})_{\Sigma 1.32}$ to $(Pt_{2.47}Ir_{0.25}Rh_{0.04}Pd_{0.01})_{\Sigma 2.77}(Fe_{1.10}Ni_{0.07}Cu_{0.06})_{\Sigma 1.23}$ for six grains analyzed. The isoferroplatinum is rimmed by tetraferroplatinum and irregular patches of osmian platinian iridium (*e.g.*, $Ir_{0.67}Os_{0.17}Pt_{0.08}Rh_{0.04}Fe_{0.03}Ru_{0.01}$). Rarer grains of iridian osmium ($Os_{0.47}Ir_{0.37}Ru_{0.09}Fe_{0.04}Pt_{0.02}Rh_{0.01}$) and laurite also occur in the isoferroplatinum. Fresh and altered silicates are found to occur between cumulus grains of chromite.

Sample #59 contains similar PGM and has similar chromite–silicate textures to sample #54, except that the laurite exhibits oriented lath-like intergrowths of tetraferroplatinum in an isoferroplatinum matrix covering an area of 7 × 11 mm (Figs. 5,6). Most of the forsterite host is altered to serpentine, but one pristine inclusion of forsterite (60 × 90 μm), within Pt–Fe alloy, was identified by EDX analysis. The isoferroplatinum in the matrix $[(Pt_{2.30}Ir_{0.25}Rh_{0.04})_{\Sigma 2.59}(Fe_{1.15}Ni_{0.18}Cu_{0.08})_{\Sigma 1.41}]$ is similar in composition to that in the intergrowths $[(Pt_{2.30}Ir_{0.21}Rh_{0.05})_{\Sigma 2.56}(Fe_{1.17}Ni_{0.22}Cu_{0.05})_{\Sigma 1.44}]$. In the same way, tetraferroplatinum rims in the matrix have compositions (given in Table 3) that are close









FIGS. 5-8. 5. SEM photomicrograph (BEI) of a compositionally zoned laurite grain cross-cut by crystallographically oriented lamella-like PGE alloys and surrounded by a nearly continuous zone of tetraferroplatinum (grey) set in a matrix of isoferroplatinum. The laurite also contains or hosts a chromite inclusion (C). Sample #59. 6. SEM photomicrograph (BEI) of an area in Figure 5, showing details of the lamella-like PGE alloys cross-cutting the laurite. The enlargement shows details of oriented tetraferroplatinum lamellae (dark grey) in an isoferroplatinum matrix (grey) and oriented osmium laths (white), some of which are extremely thin (< 1 μm). Islands of laurite suggest areas that have not yet been replaced. Sample #59. 7. SEM photomicrograph (SEI) of a typical small irregularly shaped quartz inclusion closely associated with tulameenite (T), isoferroplatinum (I), and iridium (Ir). Sample #53. 8. SEM photomicrograph (BEI) illustrating typical replacement of isoferroplatinum (light grey) by an irregular rim of tulameenite (grey). Interestingly, a lath of osmium (white) does not seem to be replaced by the tulameenite. Of note, also, is a compositionally zoned grain of laurite (L). Black areas are chromite. Sample

TABLE 3. RESULTS OF ELECTRON-MICROPROBE ANALYSES OF TETRAFERROPLATINUM

	weight per cent							
Sample #54	Pt	Ir	Rh	Pd	Fe	Ni	Cu	Totals
Grain 1	72.8	7.4	0.67	0.18	15.6	3.4	1.1	101.15
Grain 2	73.4	6.4	0.63	0.17	15.4	3.2	1.2	100.4
Grain 3	73.6	6.2	0.66	0.17	15.4	3.3	1.4	100.73
Grain 4	73.5	5.5	0.58	0.16	15.5	3.7	1.7	100.64
Grain 5	73.7	6.0	0.66	0.22	15.9	3.5	1.2	101.18
Grain 6	72.2	6.2	0.88	0.16	15.7	3.4	1.5	100.04
Sample #59, 4 anal.	72.4	5.7	0.61	-	14.5	3.6	2.1	98.91
	atomic proportions							
Grain 1	0.96	0.10	0.02	<0.01	0.72	0.15	0.05	1.08:0.92
Grain 2	0.98	0.09	0.02	<0.01	0.72	0.14	0.05	1.09:0.91
Grain 3	0.98	0.08	0.02	<0.01	0.71	0.15	0.06	1.08:0.92
Grain 4	0.97	0.07	0.02	<0.01	0.71	0.16	0.07	1.06:0.94
Grain 5	0.97	0.08	0.02	<0.01	0.73	0.15	0.05	1.07:0.93
Grain 6	0.96	0.08	0.02	<0.01	0.73	0.15	0.06	1.06:0.94
Sample #59, 4 anal.	0.97	0.08	0.02	-	0.68	0.16	0.09	1.07:0.93

For sample #54 : in addition to the above elements, Os, Ru, and Sb were sought, but not detected; for sample #59: Pd, Os, Ru, and Sb were not sought.

to that of the tetraferroplatinum in the intergrowths $[(\mathrm{Pt}_{1.00}\mathrm{Ir}_{0.07}\mathrm{Rh}_{0.02})_{\Sigma 1.09}(\mathrm{Fe}_{0.69}\mathrm{Ni}_{0.15}\mathrm{Cu}_{0.08})_{\Sigma 0.92})]$. This is additional evidence supporting the proposal of Nixon *et al.* (1990) that primary Pt–Fe–Cu–Ni alloys have undergone subsolidus modification during cooling through the presumed presence of a miscibility gap within the Pt–Fe–Cu–Ni system, analogous to that implied to exist at low temperatures between pure PtFe and $\mathrm{Pt_3Fe}$ (Cabri & Feather 1975).

Sample #62 is similar to samples #54 and #59, and Pt–Fe alloys extend over an area of 10 × 23 mm.

Placer samples

Sample #53 (10 mm in diameter) is coated with a goethite-rich rim (as much as 400 μm in thickness) containing inclusions of quartz, magnetite and other minerals derived from the placers. The nugget consists largely of isoferroplatinum with rims of tulameenite, all encapsulated by chromite. The chromite grains are fractured on a fine scale (*cf.* Ramdohr 1980, p. 345), partly altered (especially on their rims) and veined by secondary silicates. In addition, some of the Pt–Fe alloy masses project veinlets into fractures in chromite, possibly by secondary mechanical reworking. Intergrown anhedral osmian platinian iridium. (*e.g.*, $\mathrm{Ir}_{0.49}\mathrm{Os}_{0.31}\mathrm{Pt}_{0.12}\mathrm{Ru}_{0.04}\mathrm{Rh}_{0.03}\mathrm{Fe}_{0.01}$) and laths of iridian osmium (*e.g.*, $\mathrm{Os}_{0.66}\mathrm{Ir}_{0.28}\mathrm{Ru}_{0.03}\mathrm{Pt}_{0.02}\mathrm{Rh}_{0.01}$) are common primary inclusions in the isoferroplatinum $(\mathrm{Pt}_{2.94}\mathrm{Ir}_{0.08}\mathrm{Rh}_{0.01})_{\Sigma 3.03}(\mathrm{Fe}_{0.92}\mathrm{Cu}_{0.05}\mathrm{Ni}_{<0.01})_{\Sigma 0.97}$. Iridium also occurs as fine blebs in the Pt–Fe alloy matrix. Laurite grains occur, showing marked compositional zoning as well as alteration along grain rims. The compositional zoning ranges from $(\mathrm{Ru}_{0.885}\mathrm{Os}_{0.045}\mathrm{Ir}_{0.02}\mathrm{Rh}_{0.01}\mathrm{Pd}_{0.01}\mathrm{Pt}_{0.01})_{\Sigma 0.97}(\mathrm{S}_{2.02}\mathrm{As}_{<0.01})_{\Sigma 2.03}$ to $(\mathrm{Ru}_{0.66}\mathrm{Os}_{0.24}\mathrm{Ir}_{0.05}\mathrm{Rh}_{0.02}\mathrm{Pd}_{0.01}\mathrm{Pt}_{<0.01})_{\Sigma 0.98}(\mathrm{S}_{2.02}\mathrm{As}_{<0.01})_{\Sigma 2.02}$ in a single grain. Minor quantities of rhodplumsite, bowieite, kashinite, and the unknown PGM also occur in the Pt–Fe alloy. Average of four analyses of one grain of the unknown PGM gave: Rh 35.1, Ir 28.9, Pt 11.5, Ru 0.15, S 24.2%; total 99.85 wt.% for a possible formula of $(\mathrm{Rh}_{1.83}\mathrm{Ir}_{0.81}\mathrm{Pt}_{0.32}\mathrm{Ru}_{<0.01})_{\Sigma 2.96}\mathrm{S}_{4.04}$. This composition is close to that of bowieite, yet appears to have a different stoichiometry. Desborough & Criddle (1984) reported another mineral close to bowieite with a metal-deficient composition. Silicate inclusions in Pt–Fe alloys include quartz (Fig. 7), diopside (?), and rarer intergrowths such as augite (?) plus serpentine (or chlorite).

Sample #64 (10 × 11 mm) has a similar goethite-rich rim and consists mainly of isoferroplatinum $(\mathrm{Pt}_{2.99}\mathrm{Ir}_{0.05}\mathrm{Rh}_{0.03})_{\Sigma 3.07}(\mathrm{Fe}_{0.88}\mathrm{Cu}_{0.04}\mathrm{Ni}_{0.01})_{\Sigma 0.93}$ with an irregular rim of tulameenite (Fig. 8) encapsulated in chromite. Laths of osmium are common, as are anhedral grains of iridium. Laurite is present and shows zoning as well as alteration rims (*cf.* sample #53); four small inclusions of bowieite–kashinite also were found. Small (<30 μm) silicate inclusions within Pt–Fe alloys are tentatively identified to be magnesio-hornblende, sodian alumino-silicic edenite, and augite. Some of these are composite inclusions such as augite plus muscovite (?) and quartz (<10μm) attached to hornblende.

Tulameenite

Results of quantitative microprobe analyses of tulameenite from three samples are given in Table 2. These analyses show that the mineral exhibits very consistent stoichiometry between total PGE and base metals (Fe,Cu,Ni). Grain 6, extracted from sample #45 for X-ray-diffraction (XRD) analyses, proved to be microcrystalline and not suitable for single-crystal XRD. Excellent 114.6 mm Debye–Scherrer patterns were obtained with Fe-filtered Co radiation, however, and gave a = 3.9018(7), c = 3.5845(13) Å (Z = 1); they have been submitted for inclusion in the JCPDS - International Centre for Diffraction Data (J.T. Szymański, pers. comm., 1990).

Tetraferroplatinum

Tetraferroplatinum was found in samples #54, #59, and #62, usually as an overgrowth or rim on isoferroplatinum in the former and both as a rim on isoferroplatinum and as oriented intergrowths in isoferroplatinum in the latter. Quantitative analyses reveal little variation and a slightly Pt-rich stoichiometry (Table 3).

DISCUSSION AND CONCLUSIONS

Betekhtin's "cuproplatinum" is identical to tulameenite, and quite likely Wyssotzky's copper-bearing platinum (for the most copper-rich examples) also is tulameenite. Betekhtin (1935) considered "cuproplatinum" to be an isotropic (= cubic) species distinct from native platinum. His later report (1961), stating that the most copper-rich grains of "cuproplatinum" are anisotropic, was not given appropriate notice by later authors (*e.g.*, Wright & Fleischer 1965, Cabri 1972, Ramdohr 1980). Such a statement, however, if made without confirmatory XRD data, is insufficient for consideration of a distinct mineral species. This is especially true for alloys, which may exhibit anisotropic effects from polishing. The similarity of Betekhtin's "cuproplatinum" to tulameenite was not raised by any representative during or after consideration of tulameenite as a new mineral species by members of the Commission on New Minerals and Mineral Names of the International Mineralogical Association in 1972. It is also apparent that "cuproplatinum" has not been recognized as a distinct mineral species, nor has it been widely used in the literature. In addition, "cuproplatinum" is not a suitable name for a tetragonal mineral because platinum itself is cubic.

We believe it appropriate to also discuss the paragenesis of tulameenite. Betekhtin (1961) proposed that tulameenite formed during serpentinization of the ultramafic massifs by solutions that travelled along the contacts of primary platinum alloy grains and chromite. He proposed that Fe and Ni in tulameenite were derived from the olivine. Copper was considered to have been derived by alteration of former sulfides (chalcopyrite, cubanite, pyrrhotite, pentlandite), which he observed only rarely as minute disseminations in unaltered dunite. These primary sulfides are never found in serpentinized dunite, but rare magnetite (from pyrrhotite) and millerite (from pentlandite) occur. Betekhtin also reported the occurrence of native copper (from Cu-bearing sulfides), as rare grains and as veinlets closely associated with thin veins of serpentine. Interestingly, Nixon *et al.* (1990) arrived at essentially the same conclusions for the paragenesis of tulameenite in their study of Pt mineralization in the Tulameen complex, British Columbia.

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