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LETTER

Platinum-group elements in ores from the Kalmakyr porphyry Cu–Au–Mo deposit, Uzbekistan: bulk geochemical and laser ablation ICP-MS data

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Abstract We studied primary ore samples from Kalmakyr, a giant Cu–Au–Mo porphyry deposit in eastern Uzbekistan. Disseminated and stockwork-type high-grade Cu–Au–Mo mineralization showed average concentrations of 55 ppb Pd, 5.5 ppb Pt, 0.95 ppb Rh, 0.49 ppb Ir, and 4.1 ppm Au (n=8). This type of mineralization is characterized by the presence of pyrite, chalcopyrite, molybdenite, and gold. A peak Pd content of 292 ppb was determined in a base-metal-rich quartz vein in granodiorite porphyry, which contains galena, sphalerite, chalcopyrite, tetrahedrite, and gold. Palladium correlates with Cu, Ag, Se, and S. Mineralogical and laser ablation ICP-MS study confirmed that Pd is homogeneously distributed in chalcopyrite, which contains up to 110 ppm Pd, and tetrahedrite, containing up to 20 ppm Pd. An assessment of the Pd and Pt budget at Kalmakyr showed the potential of approximately 17 t of Pd and 1.7 t of Pt.

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Introduction

Alkaline porphyry deposits represent significant gold resources owing to their large size. Anomalous platinum-group element (PGE) concentrations have been reported from a number of porphyry copper deposits worldwide, e.g., Copper Mountain and Galore Creek in the Cordillera of British Columbia; Allard Stock, La Plata Mountains and Copper King in USA; Majdanpek, Veliki Krivelj and Bor in Serbia; Elacite in Bulgaria (Tarkian et al. 2003); Skouries in Greece; Mamut in Malaysia; Ok Tedi and Panguna in Papua New Guinea; Santo Tomas II in Philippines (Tarkian and Koopmann 1995); Bozshakol in Kazachstan; Sora, Aksug and Zhireken in Russia; Erdenetiun-Obo in Mongolia and others (see summary paper of Economu-Eliopoulos 2005). Tarkian and Stribrny (1999) reported the results from a PGE study in sulfides and flotation concentrates from 33 porphyry copper deposits, where seven deposits revealed relatively high Pd contents (130–1,900 ppb) that were associated with high Au contents (1–28 ppm). They also identified the presence of platinum-group minerals (PGM) which occurred as inclusions in chalcopyrite.

Palladium is recovered as a by-product during the processing and refining of gold ores at a number of gold deposits, including the giant Cu–Au–Mo porphyry deposit of Kalmakyr in Uzbekistan. However, PGE data on porphyry systems from Uzbekistan are very limited (Turesebekov et al. 2005; Tarkian and Stribrny 1999).

In this paper, we present, for the first time, results from a geochemical and laser ablation ICP-MS study of PGE distribution in various types of ores from the world-class Kalmakyr Cu–Mo–Au porphyry deposit in Uzbekistan.

Samples and analytical techniques

Eleven representative ore and rock samples of about 1 kg each from the Kalmakyr deposit in Uzbekistan were studied for their PGE and other trace elements. The studied samples were collected from the 505 m level, i.e., the richest ore zone with 1–2% Cu, 0.2–0.5% Mo, 1–2 g/t Au, and the 520 m level with 0.3–0.4% Cu, 0.2–0.5% Mo, 0.5–0.6 g/t Au (V. A. Gornov, mine chief geologist, pers. com.) in the Kalmakyr open pit (Fig. 1). For sample description see Table 1. The samples were pulverized in the laboratories of the Czech Geological Survey (Prague). The PGEs were preconcentrated into Ni-button and analyzed by ICP-MS at the Faculty of Science, Charles University in Prague. Trace elements and sulfur were determined using ICP-MS following a lithium metaborate/tetraborate fusion and nitric acid digestion at Acme Labs Ltd. in Canada.

The mineral composition of all samples was characterized using optical microscopy and SEM. Concentrations of PGEs in minerals from sample K8 were analyzed by laser ablation ICP-MS technique. The sample was mounted in epoxy and polished for backscattered electron imaging and analysis using a solid state Nd:YAG laser (UP-213, New Wave Research) and a single collector sector field ICP-MS (Element 2, Thermo Finnigan) at the University of Bergen. The sample was ablated in He, the laser was fired at 10 Hz with 1.8 J/cm^2 fluence, producing laser pits of 80 µm in diameter. Signals were acquired for masses 34 (S), 99-101-102 (Ru), 105-106-108 (Pd), 188 (Os), 191-193 (Ir), 194-195 (Pt), and 197 (Au) using one measurement per mass peak and 10 ms dwell time. The signal of ^{34}S was used as internal standard to correct for differences in the ablation yield assuming stoichiometric composition of the minerals. The synthetic pyrrhotite sample PO725-B2 (Memorial University of Newfoundland) doped with PGEs and Au was used for calibration and quality control of analyses.

Geology and ore mineralogy of the Kalmakyr deposit

The Kalmakyr deposit belongs to the Almalyk ore field and is a part of the Chatkal–Kurama ore district in eastern Uzbekistan (Fig. 2). The Cu mineralization at Almalyk was discovered in 1926, exploration was undertaken from 1931 to 1941 and between 1947–1951, and open pit mining commenced at Kalmakyr in 1954. Additional exploration and delineation work between 1961 and 1980, and from 1986 to 1996 culminated in the estimation of reserves/resources of 2,000 Mt of 0.38% Cu, 0.6 g/t Au, 0.006% Mo at a 0.2% Cu cut-off, plus 1,700 Mt of lower grade of 0.15–0.19% Cu (Golovanov et al. 2005).

The mineralization is hosted mainly by plutonic rocks, which are related to the large Almalyk monzonitic pluton extending far beyond the deposit. The distribution of ore is controlled by the morphology of porphyry stocks and dykes and by linear fracture zones related to the Kalmakyr and Karabulak faults. As a result, the stockwork is represented by a cone surrounding the quartz monzonite porphyry and most intense fracturing and the high-grade ore are related to intersections of porphyry contacts with east–west and north-eastern faults. The total stockwork dimensions are 3,520× 1,430 m; the maximum vertical extent is 1,240 m (Shayakubov et al. 1999). The currently mined Kalmakyr ore contains 0.58% Cu, 0.05% Mo, 0.5 g/t Au, 3 g/t Ag, 1.5–2.0% S and admixtures of Se, Te, Re, Bi, and In. All Mo-rich ore bodies contoured by 0.008% Mo are located at the periphery of the Cu ore stockwork; the distribution of gold is controlled by the copper mineralization (Shayakubov et al. 1999).

Results and discussion

Geochemistry and mineralogy

The results of PGE, Au, and other selected trace and major elements in the studied ore and host rock samples (n=12) are summarized in Table 2. The maximum values in our sample set are 16.3 ppm Au, 292 ppb Pd, 13.1 ppb Pt, 4.1 ppb Rh and 1.27 ppb Ir, respectively (Table 2). The average ore of the 12 samples from Kalmakyr is 2.4 wt.% Cu, 0.18 wt.% Mo, 4.1 ppm Au, 55.2 ppb Pd, 5.5 ppb Pt, 0.95 ppb Rh, and 0.49 ppb Ir (Table 2). The average copper grade is the same as that of the sulfide flotation concentrate from the Skouries porphyry deposit, Greece (Tarkian and Stribrny 1999), and very close (2.7 wt.%) to the flotation concentrate from the Zhireken porphyry deposit, Russia (Berzina et al. 2005). The average Pd value (55.2 ppb) for our ore samples is in the range of the values from flotation concentrates from the Assarel porphyry deposit, Bulgaria (54 ppb), Grasberg deposit, Indonesia (58 ppb) and Sora porphyry deposit in Russia (50 ppb) (Economu-Eliopoulos 2005).

To assess the PGE and Au potential of porphyry system, it is important to analyze representative ore samples and to ensure that the ore samples are not part of the zones dominated by chalcocite and covellite, as they may exhibit high Au contents and negligible Pd–Pt values due to preferential mobilization during supergene enrichment and/or later hydrothermal overprint (Wood 2002). As we sampled representative ore only from a very limited and the richest part of the deposit, we recalculated the total reserves

Fig. 1 Location of sample collection at the Kalmakyr open pit (view towards south-east, 40°49′10″ S, 69°38°32″E)

of 2,000 Mt (avg. Cu=0.38%) to 316 Mt (avg. Cu=2.4%). Assuming ca. 316 Mt reserves and the average concentrations for the set of studied ore samples (Pd=55.2 ppb and Pt=5.5 ppb), the potential of the Kalmakyr deposit would be ca. 17 t Pd and 1.7 t Pt.

The chondrite-normalized PGE pattern for our average Cu–Au–Mo ore reflects Pd and Au enrichment (Fig. 3) and is very similar to that of the continental crust, which is, however, characterized by much lower PGE values (Fig. 3). Chondrite-normalized PGE–Au patterns of Au–Ag ores from the Kochbulach and Kyzylalma epithermal deposits in the Chatkal–Kurama ore district in Uzbekistan also show a strong Ir and Pd depletion and similar Ru, Rh, Pt, and Au enrichment when compared to the Kalmakyr ore (Fig. 3).

Although there is variation throughout each mineralized zone, the Pd/Pt and Pd/Cu ratios seem to be characteristic of each porphyry Cu–Au–Mo deposit, possibly reflecting the composition of parental magmas (Economu-Eliopoulos 2005). The average Pd/Pt ratio (11.8) and Pd/Cu ratio (23) in our Cu-Au-Mo ore samples are close to other alkaline porphyry copper deposits (e.g., 10 and 20 from Galore Creek in British Columbia, respectively, Economu-Eliopoulos 2005). The Au/Pd ratio in Cu–Au–Mo ore (192) from Kalmakyr is, however, notably higher than that

Table 1 Description of the studied samples from the Kalmakyr deposit

Sample	Type of ore/mineralization	Main ore minerals
Level 505 m (the richest ore zone 1–2 wt.% Cu, 0.2–0.5 wt.% Mo, 1–2 g/t Au)		
K1	Cu–Mo mineralization in stockworks in granodiorite porphyry	Pyrite, molybdenite, chalcopyrite, gold
K2	Cu mineralization disseminated in granodiorite porphyry	Chalcopyrite, pyrite, gold
K3	Cu–Mo mineralization disseminated in granodiorite porphyry	Pyrite, molybdenite, chalcopyrite, gold
K4	Cu mineralization in stockworks in granodiorite porphyry	Pyrite, chalcopyrite, gold
K5	Cu mineralization disseminated and in stockworks (mixed type) in granodiorite porphyry	Pyrite, magnetite, chalcopyrite, gold
K6	Granodiorite porphyry with low-grade mineralization	Minor pyrite and chalcopyrite
K7	Cu mineralization disseminated in silicified granodiorite	Chalcopyrite, pyrite, magnetite, gold
K8	Pb–Zn–Cu mineralization in quartz veins in granodiorite porphyry	Galena, sphalerite, chalcopyrite tetrahedrite, pyrite, gold
Level 520 m		
K9	Mo mineralization in stockworks in silicified granodiorite porphyry	Minor pyrite, molybdenite, chalcopyrite, galena, sphalerite, gold
K10	Diorite with low-grade mineralization	Minor pyrite, sphalerite
K11	Granodiorite porphyry with low-grade mineralization	Minor pyrite, sphalerite, galena, chalcopyrite

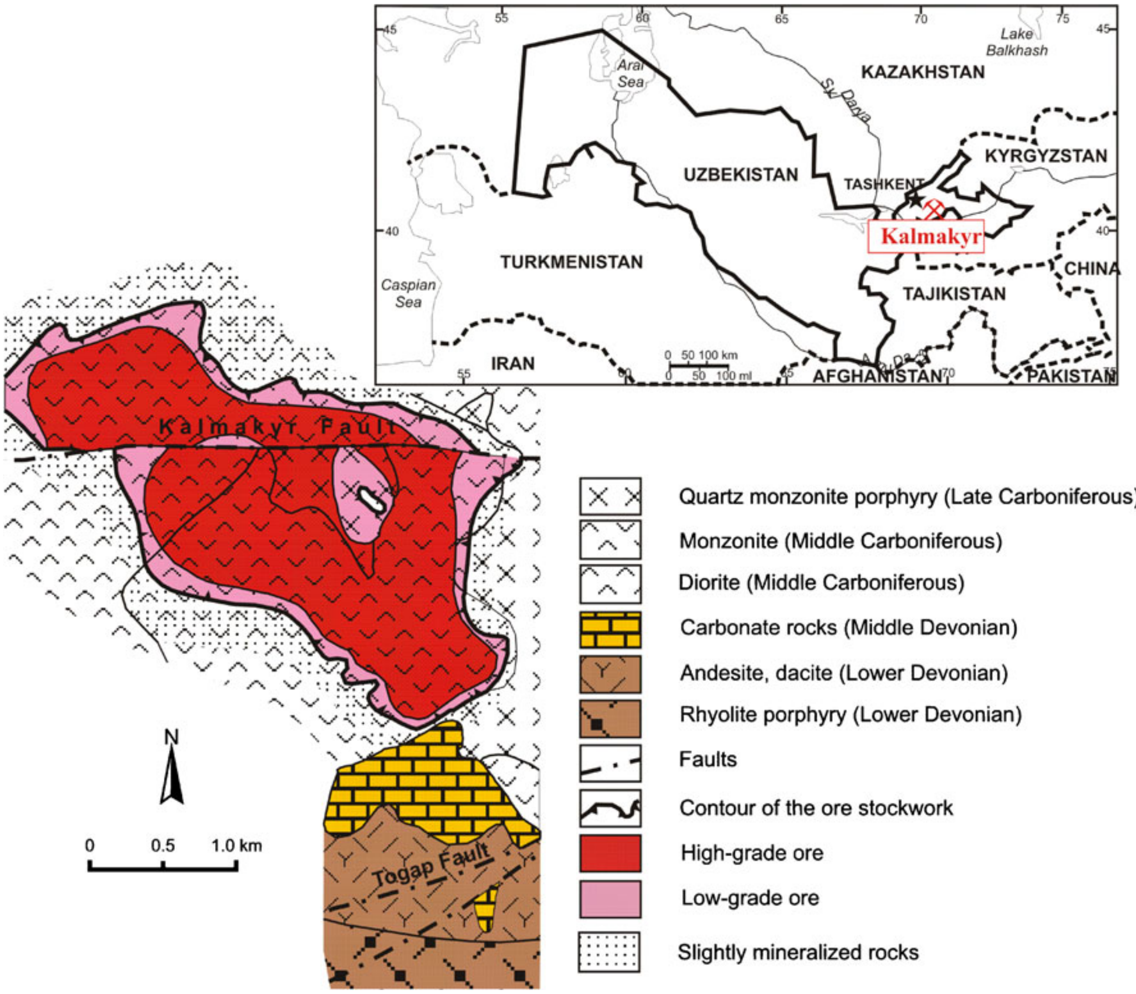

Fig. 2 Schematic geological map of the Kalmakyr deposit, adapted from Shayakubov et al. (1999)

of other world-class porphyry deposits where this ratio ranges from 0.1 to 89 (Economu-Eliopoulos 2005).

In the Kalmakyr deposit, Cu shows a significant relationship with Au and Pd, while Pd shows a strong link with Ag (Fig. 4) and also Se and S (Table 3). The correlation matrix (Table 3) for selected elements indicates that Pd is most likely associated with chalcopyrite and/or other Cu–Ag–Se–S phases, likely tetrahedrite or selenides or tellurides. No significant relationship between Pd, Pt, and Au was found. Similar values of the correlation matrix and a strong positive correlation (r+0.91) of Cu with Au and Pd have been reported from Cu–Mo porphyry deposits in Russia (Sora, Aksug, and Zhireken deposits) and Mongolia (Erdenetiun-Obo) by Sotnikov et al. (2001). Furthermore, PGM in porphyry systems have been identified as inclusions (2 to 20 µm) in chalcopyrite in the Elacite, Majdanpek, Mamut, Biga, and Skouries deposits (Tarkian and Stribrny 1999). PGM, usually of extremely small grain size, are represented by merenskyite ($Pd_{0.9}Pt_{0.1}Te_{1.8}Bi_{0.2}$) or solid solutions of merenskyite–moncheite ($Pt_{0.75}Pd_{0.25}Te_{1.5}Bi_{0.5}$); sperrylite ($PtAs_2$) and an unidentified Pd–Sb telluride in chalcopyrite was reported from the Mamut deposit (Tarkian and Stribrny 1999).

The primary Kalmakyr ore is characterized by a rather simple ore mineralogy represented mainly by chalcopyrite, molybdenite, pyrite, magnetite, galena, and sphalerite (see Table 1, Fig. 5). This is in agreement with data from Kovalenker et al. (2008). These authors described early sulfide–oxide mineral assemblages at Kalmakyr (pyrite, magnetite, hematite, rare chalcopyrite), followed by Au-bearing assemblages, which include chalcopyrite, bornite, molybdenite, pyrite, electrum, magnetite, hematite, minor base metal sulfides (galena and sphalerite), and rare sulfosalts and tellurides. Golovanov et al. (1988) described several mineral assemblages (from older to younger): quartz–magnetite, quartz–pyrite–molybdenite–chalcopyrite with gold, quartz–carbonate–polysulfide with gold, zeolite–anhydrite and barren calcite and barite veins and veinlets that complete the ore formation. Pyrite, magnetite, chalcopyrite, and gold are the principal ore minerals in the sample

Table 2 Geochemical composition of ore and rock samples from the Kalmakyr deposit

Element Sample	Mo wt.%	Cu wt.%	Pb ppm	Zn ppm	Ag ppm	Fe wt.%	S wt.%	Se ppm	Ir ppb	Ru ppb	Rh ppb	Pt ppb	Pd ppb	Au ppb	Pd/Cu	Pd/Pt	Au/Pd	Cu/Au
Ore samples																		
K1	0.5950	1.24	38	116	2.0	7.4	5.1	11	0.53	<2	0.70	3.1	4.3	1825	3.5	1.4	424	6.8
K2	0.0038	0.96	20	27	1.2	11.2	13	15	0.68	<2	1.05	5.2	5.3	969	5.5	1.0	183	9.9
K3	0.0661	0.49	11	27	0.7	1.7	1.5	3	0.25	<2	0.32	0.4	1.7	659	3.5	4.3	388	7.4
K4	0.0018	0.26	13	47	1.1	5.0	2.9	3	0.25	<2	0.16	1.7	17.3	986	66	10.2	57	2.7
K5	0.0007	2.97	14	59	3.5	9.5	6.7	27	0.32	<2	0.55	9.1	73.1	7355	25	8.0	101	4.0
K7	0.0003	7.10	28	23	10.3	10.8	10.9	52	0.34	<2	0.56	11.2	42.0	16337	5.9	3.8	389	4.3
K8[a]	0.0005	5.66	127900	71297	91.3	14.5	21.9	117	1.27	<2	4.11	4.5	291.7	3912	52	65	13	14.5
K9	0.7563	0.51	528	343	2.1	3.1	3.5	7	0.29	<2	0.16	8.9	6.3	695	12.4	0.7	110	7.3
Average ore, including K8 (n=8)	0.1781	2.40	16069	8992	14.0	7.9	8.2	29	0.49	<2	0.95	5.5	55.2	4092	23	11.8	74	5.9
Rock samples with low-grade mineralization																		
K6	0.0015	0.06	20	69	0.7	47.2	<0.5	3	0.16	<2	0.11	5.4	5.8	53	98	1.1	9.1	11.2
K10	0.0048	0.05	6	107	2.4	7.9	<0.5	<2	0.28	<2	0.18	13.1	11.4	66	252	0.9	5.8	6.9
K11	0.0030	0.02	151	214	<0.5	2.4	<0.5	<2	0.22	<2	0.09	1.8	<0.5	29	n.d.	n.d.	n.d.	6.9

[a] Anomalous Pb–Zn–Cu ore sample

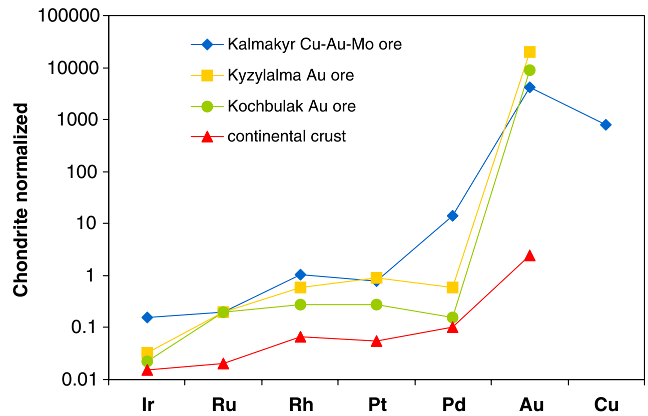

Fig. 3 Chondrite-normalized PGE-Au-Cu patterns for average ore from Kalmakyr and epithermal Au deposits (Kyzylalma and Kochbulach) in Uzbekistan (data from Vymazalová et al. 2009). Continental crust data are from GERM Reservoir Database http://www.earthref.org/, chondrite data from Anders and Grevesse (1989)

with highest gold content (K7—see Tables 1 and 2). The different geochemical composition of the sample K8 (max. Pd content—see Table 2) is reflected by a different ore mineralogy with dominantly galena, sphalerite, chalcopyrite, tetrahedrite, pyrite, gold, and electrum (Table 1).

Laser ablation study

Selected grains of chalcopyrite, pyrite, galena, and tetrahedrite from sample K8 with the highest Pd concentration

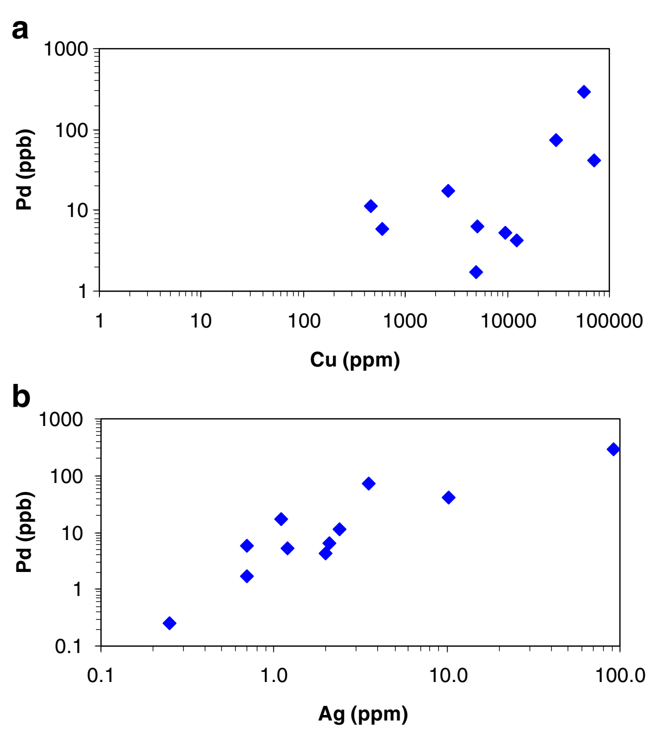

Fig. 4 Plot of Pd versus **a** Cu and **b** Ag

Table 3 Correlation matrix for selected major and trace elements from the Kalmakyr Cu–Au–Mo deposit (n=11)

	Au	Pd	Pt	Cu	Mo	Ag	S	Se
Au	1							
Pd	0.24	1						
Pt	0.46	0.02	1					
Cu	0.88	0.65	0.33	1				
Mo	−0.19	−0.22	0.03	−0.20	1			
Ag	0.16	0.98	−0.04	0.61	−0.17	1		
S	0.43	0.81	0.07	0.76	−0.15	0.81	1	
Se	0.48	0.95	0.11	0.84	−0.21	0.94	0.90	1

were analyzed for PGE and Au contents by laser ablation ICP-MS technique and the results are summarized in Table 4. Most of the analyses were below the limit of detection for the analytical parameters used. Accordingly, only data for Ru, Pb, and Au in pyrite, chalcopyrite, galena, and tetrahedrite are reported here. The highest Pd values were detected in chalcopyrite (110 ppm) and tetrahedrite (20.2 ppm). It should be noted that laser ablation signals for the analyzed elements, especially Pd (where detected), did not show short-time variations in intensity as would be expected for laser ablation of PGE inclusions. Such inclusions were also not visible on backscattered electron images of the sample. The observed steady and declining signal is rather typical of homogeneous element distribution in the analyzed minerals. Accordingly, it is suggested that the distribution of Pd in chalcopyrite and tetrahedrite is likely in the mineral crystal lattice, or alternatively, in sub-micron inclusions of PGE minerals that are homogeneously distributed throughout the host mineral grains.

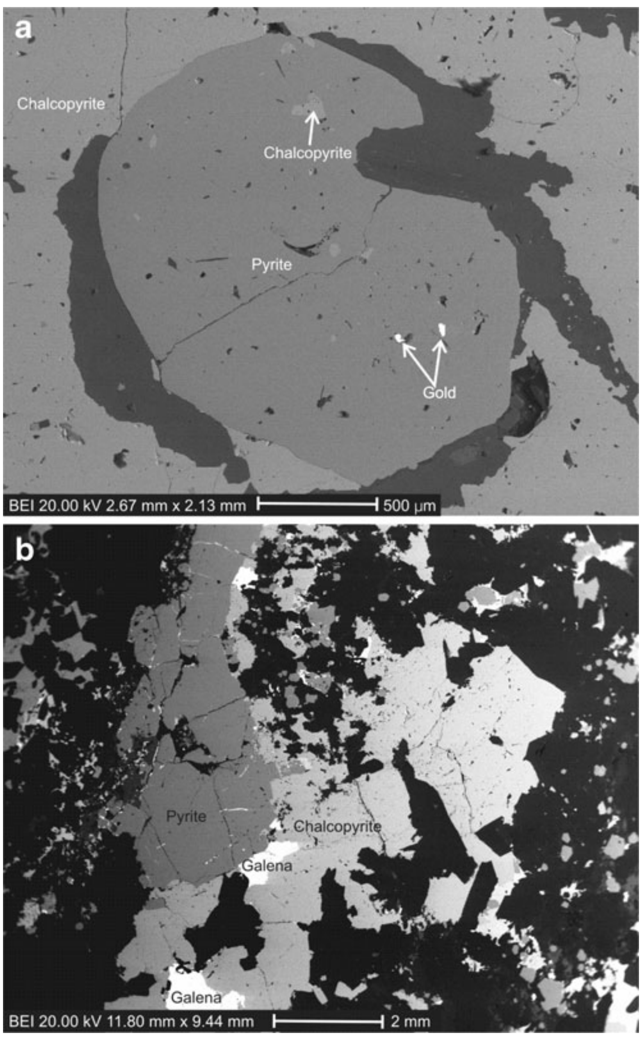

Fig. 5 Backscattered electron images of typical ore from the Kalmakyr deposit: **a** native gold in association with pyrite and chalcopyrite, **b** most abundant sulfides: chalcopyrite, pyrite, and galena

Table 4 Representative laser ablation ICP-MS analyses of PGE in sulfides from sample K8

Analysis #	Ru	d.l.	Pd	d.l.	Au	d.l.
Pyrite #1	0.44	0.12	b.d.l.		0.45	0.027
Pyrite #2	b.d.l.	0.14	b.d.l.		0.07	0.036
Pyrite #3	b.d.l.	0.11	b.d.l.		0.1	0.020
Pyrite #4	0.34	0.12	b.d.l.		0.28	0.024
Pyrite #5	0.55	0.12	b.d.l.		1.12	0.027
Pyrite #6	0.58	0.09	b.d.l.		0.32	0.022
Chalcopyrite #1	0.14	0.09	103.4	0.76	0.21	0.074
Chalcopyrite #2	0.1	0.09	110.4	0.74	b.d.l.	0.065
Chalcopyrite #3	b.d.l.	0.08	89.6	0.73	b.d.l.	0.054
Chalcopyrite #4	b.d.l.	0.07	87.3	0.67	b.d.l.	0.054
Galena #1	b.d.l.		0.23	0.17	0.14	0.014
Galena #2	b.d.l.		b.d.l.	0.16	0.24	0.013
Galena #3	b.d.l.		0.75	0.53	0.91	0.050
Galena #4	b.d.l.		0.95	0.69	0.98	0.052
Tetrahedrite #1	b.d.l.		19.3	0.74	b.d.l.	0.059
Tetrahedrite #2	b.d.l.		17.5	0.75	0.14	0.067
Tetrahedrite #3	b.d.l.		20.2	0.77	b.d.l.	0.075

Concentrations are in ppb

b.d.l. below detection limit

PGE transport

Generally, copper and precious metals are transported as chloride complexes by relatively hot (400–700°C) and saline and hypersaline (>70 wt.% $NaCl_{equiv.}$) hydrothermal fluids in porphyry systems. The main stage of mineralizations consists of bornite, chalcopyrite, pyrite and magnetite, as veinlets and disseminations associated with potassic alteration (Economu-Eliopoulos 2005).

At Kalmakyr, the early porphyry mineralization (pyrite, magnetite, hematite, and rare chalcopyrite—free of precious metals) was formed from high-temperature (597–525°C) and hypersaline (68.1–40.6 wt.% $NaCl_{equiv}$) fluids, while the later (precious metal-rich porphyry mineralization) formed from diluted fluids of much lower temperature (around 250°C) (Kovalenker et al. 2008). Under acid oxidizing conditions and temperatures up to 300°C, both Pt and Pd may be transported as chloride complexes (Gammons et al. 1992; Wood et al. 1992). Chloride transport is generally invoked for Cu and Au in the porphyry environment (e.g., Sillitoe 1993) and the association of Cu, Au, Pd, and Pt may be explained in part by a similar transport and deposition mechanism. Pd and Pt can also be transported as bisulfide complexes, but at levels one to three orders of magnitude lower than Au under the same conditions (Wood 2002). Recently, Heinrich et al. (2009) demonstrated that metal-rich magmatic vapors are sufficiently sulfur-rich to support the required Cu–S and Au–S complexes. Halter et al. (2002) demonstrated that magmatic sulfide melts can act as intermediate metal hosts, which preconcentrate Cu and precious metals during the evolution of the magmatic system before volatile saturation, and probably participate in an important step in the genesis of porphyry-type deposits. The laser ablation ICP-MS study in sulfides at Kalmakyr showed a close relationship of Pd and Cu (Ag, Se, S) without any important relationship with Au. We suggest that a strong Pd enrichment in base-metal-rich mineralization at Kalmakyr may be related to the presence of significant concentrations of other metal ions such as As, Bi, Sb, Se, and Te, possibly resulting in the formation of very insoluble compounds (submicron inclusions) with PGE.

Conclusions

The eight Cu–Au–Mo ore samples studied (Cu=2.4 wt.%, Mo=0.18 wt.%, Au=4.1 ppm) contain 55.2 ppb Pd, 0.95 ppb Rh, 0.49 ppb Ir, and 5.5 ppb Pt on average. This ore is characterized by rather high ratios of Au/Pd of 192, and low ratios of Pd/Pt of 4.2, Cu/Au of 4.7, and Pd/Cu of 11.1. The peak Pd concentration of 292 ppb is accompanied by significantly lower Pt (4.5 ppb), Rh (4.11 ppb) and Ir (1.27 ppb) values detected in a Pb–Zn–Cu-rich sample (Pb= 12.8 wt.%, Zn=7.1 wt.%, Cu=5.6 wt.%, Au=3.9 ppm) from a quartz vein in granodiorite porphyry. This sample shows very high values of Pd/Pt (65), Cu/Au (14.5), Pd/Cu (52), and rather low Au/Pd (13) ratios.

The chondrite-normalized PGE–Au pattern for the average ore from Kalmakyr shows a similar shape with that of the continental crust; however, at a higher level of PGE enrichment, possibly indicating a crustual source of these metals.

Pyrite, chalcopyrite, molybdenite, magnetite, and gold are typical of Cu–Au(Mo) ore while galena, sphalerite, chalcopyrite, tetrahedrite, pyrite, and gold dominate in the Pd-richest sample. Geochemical correlation study showed an important relationship between Cu and (Au, Pd). Palladium also showed a significant link with Ag, Se, and S. The laser ablation ICP-MS study of base metal sulfides confirmed that anomalous Pd concentrations are likely bound to the crystal lattice of chalcopyrite and tetrahedrite or, alternatively, are homogeneously distributed in submicron inclusions of PGE minerals.

We suggest that the primary Cu ore at Kalmakyr represents a significant source of PGE, and estimate a metal content of approximately 17 t Pd and 1.7 t Pt in the deposit.

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