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Geochemistry and the form of «invisible» gold in pyrite from metasomatites of the Khangalas deposit, North-East of Russia

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Abstract. *The Khangalas orogenic gold deposit is located in the central part of the Yana–Kolyma gold belt, Northeastern Russia. Gold occurs in the native form in quartz veins, and in the so-called invisible form in pyrite and arsenopyrite of quartz-carbonate-sericite metasomatites. Pyrite and arsenopyrite are the most common ore minerals of the deposit. For the vein-veinlet and veinlet-disseminated types of mineralization of the Khangalas deposit, four generations of pyrite and two generations of arsenopyrite were identified. Despite the widespread occurrence of disseminated pyrite-arsenopyrite mineralization of metasomatites, its mineralogical-geochemical features, isotope-geochemical characteristics and mechanisms of location remain insufficiently studied, and its origin is debatable. The article presents the first results obtained in the studies of the geochemistry of the most common and industrially significant gold-bearing pyrite (Py3) from metasomatites. The elementary composition and morphology of crystals were studied with a JEOL JSM-6480LV electron scanning microscope equipped with an Energy 350 Oxford energy dispersion spectrometer. Trace elements in pyrites were determined using a New Wave Research UP-213 laser ablation system (USA), coupled with an Agilent 7700x quadrupole mass spectrometer (Agilent Technologies, USA). The total amount of impurities in pyrite-3 is 0.48–2.12 %, with an average of 1.11 %. The content of Au in a Py3 gross sample determined by the atomic absorption method is up to 39.2 ppm, silver up to 17.38 ppm. Typomorphic trace elements according to LA-ICP-MS analysis are As (4530–18790 ppm), Ni (8.2–1298 ppm), Co (0.23–505 ppm), Cu (0.5–19 ppm), Zn (3.5–6.4 ppm), Pb (0.5–860 ppm), Sb (0.3–407 ppm), Ag (0.008–1.01 ppm) and Au (0.1–15.9 ppm). Au is closely correlated with As (r = 0.9). Of ~ 100 grains of pyrite-3 examined, ~ 20 % contain microinclusions of galena and sphalerite in defects and crystal growth zones; tetrahedrite and freibergite are recorded in single samples. A microinclusion of native gold Au^0 was found only in one sample; it has a size of about 15 μm, fineness of 827 ‰. On the Au – As (mol %) diagram, the data points for Py3 samples from the Khangalas deposit fall below the solubility limit of Au in a solid. This indicates the form of “invisible” gold found in Py3 mainly as structurally bound Au^+. The presence of gold-bearing pyrite in metasomatites is of great practical importance and makes it possible to significantly expand the raw material potential of the Khangalas ore cluster.*

Key words: LA-ICP-MS trace element analysis, auriferous pyrite, form of «invisible» gold, Khangalas deposit, Yana–Kolyma gold belt, NE Russia.

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Intoduction

The Yana–Kolyma gold belt (YKGB) is the largest in northeast Russia [1–5]. The disseminated mineralization is widely manifested here both within the gold deposits [6–11] and in the regional sulfidation zones [12–14]. In particular, high gold contents in pyrite and arsenopyrite from metasomatic rocks were established at the Khangalas deposit located in the Olchan-Nera metallogenic zone of the central part of the Yana–Kolyma gold belt [15] (Fig. 1). It was discovered in 1947. When studying and developing the deposit until the 1990s, attention was paid to the vein type of mineralization [16, 17], in the 2000s, to mineralized crush zones with vein-veinlet mineralization [18, 19]. A detailed description of the geological structure of the Khangalas deposit is published in [5].

Pyrite-3 is the most widespread mineral of metasomatic rocks of the Khangalas deposit. Its geochemical characteristics and gold content are poorly studied, which makes it difficult to understand the origin of mineralization and assess the economic potential of the deposit. The article presents the first results of studying trace elements and the forms of gold in pyrites from metasomatites using modern analytical methods.

Materials and methods

Rock samples for mineralogical-geochemical and isotope-geochemical studies were collected from natural outcrops, walls and dumps of surface and underground mine workings. Mineralogy and textural-structural features of ores were studied with a ZEUS Axio optical microscope. The elementary composition and morphology of crystals were determined using a JEOL JSM-6480LV scanning electron microscope equipped with an Energy 350 Oxford energy dispersion spectrometer (20 kV, 1 nA, beam diameter 1 μm) (analyst Popov A.V., DPMGI SB RAS). Au and Ag grades of more than 2 ppm in a mass of ground pyrite were determined by an Agilent 4200 MP-AES atomic emission spectrometer with a microwave-saturated plasma. Au and Ag grades less than 2 ppm were determined by atomic absorption spectrometry with electrothermal atomization on an iCE 3500 spectrometer manufactured by ThermoScientific (analysts Sannikova A.E., Naryshkina E.L., Mikhailov E.I., IGABM

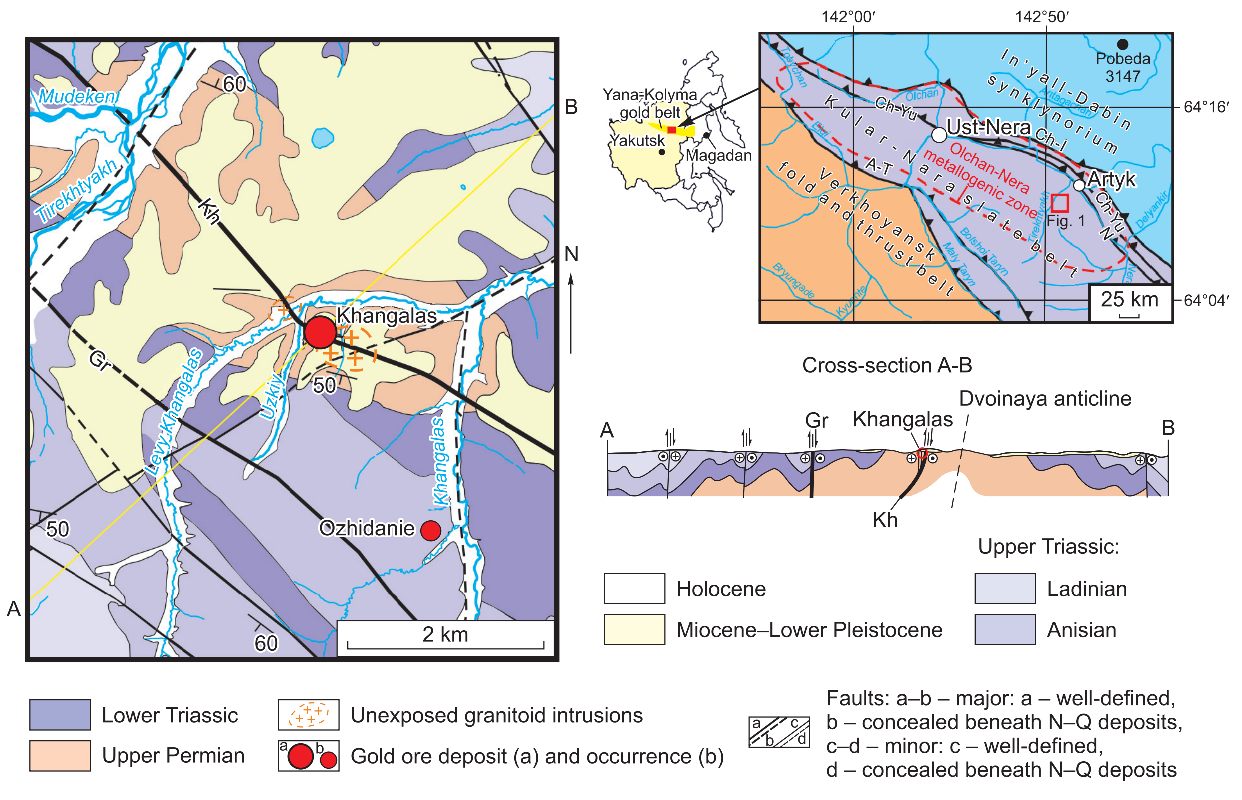

Fig. 1. Geological structure diagram and section of the Khangalas deposit (after Fridovsky et al. [5]). The inset map shows position of the Khangalas ore cluster. Faults: Ch-I – Charky-Indigirka, Ch-Yu – Chai-Yureya, N – Nera, A-T – Adycha-Taryn, Kh – Khangalas, Gr – Granitny.

SB RAS). The detection limits of the elements are from 0.0001 μg/ml and above. Trace elements were identified for 9 pyrite-3 grains from the Khangalas deposit by a New Wave Research UP-213 laser ablation system (USA) coupled with an Agilent 7700x quadrupole mass spectrometer (Agilent Technologies, USA) (analyst Artemyev D.A., Institute of Mineralogy, UrB RAS, Miass) [20–22].

Results and discussion

Several pyrite generations are shown for metasomatites of the Khangalas orogenic gold deposit (Fig. 2). Syngenetic framboidal pyrite (Py1) was formed after sedimentation, during diagenetic processes due to bacterial sulfate reduction. Framboids are represented by spherical intergrowths of pyrite microcrystals ranging in size from 10 to 100 microns. Pyrite microcrystals sometimes have a zonal structure and form dispersed or layered impregnations in a carbon-silica matrix. Epigenetic metamorphogenic pyrite (Py2) is characterized by disseminated areal distribution. Two morphological types of Py2 are distinguished: fine-grained pyrite ranging in size from 5 to 50 μm and coarse-grained from fractions of a millimeters up to 1–3 mm. Py2 is characterized by the cubic form of crystals and the cataclastic and corrosive microtexture. Py3 is developed in metasomatic rocks and associated with early arsenopyrite (Apy1). Those sulfides have a hypidiomorphic metagranular structure and form intergrowths with sharp boundaries. They have a disseminated and microlenticular texture. Py3 is characterized by complicated cubic or pentagonal dodecahedric forms. The size of the crystals attaineds 2–3 mm.

The total amount of trace elements is 0.48–2.12 %, with an average of 1.11 %. Table shows the most characteristic trace elements in pyrite-3 from metasomatized rocks of Khangalas deposit. As is the main impurity element of pyrite-3. Its content ranges from 0.45 to 1.88 %. The gold grade in a gross sample is 0.76–39.2 ppm as established by the atomic absorption spectrometry, and that in individual pyrite-3 grains (Fig. 2, *f*–*n*) estimated by LA-ICP-MS trace element analysis is 0.1–15.9 ppm. A close correlation is observed for Au with As (r = 0.9) and Cu (r = 0.92). In the ground gross sample the Ag content is 1.15–17.38 ppm as determined by the atomic absorption analysis, and according to LA-ICP-MS trace element analysis, in individual pyrite-3 grains (Fig. 2, *f*–*n*) silver is contained in a small amount (C_{Ag} = 0.008–1.01 ppm).

Silver weakly correlates with Au (r = 0.19). 20 % of ~ 100 Py3 grains examined with an optical microscope contain microinclusions of the polysulfide association minerals (galena, rarely sphalerite and chalcopyrite) (Fig. 2, *d*). Tetrahedrite and freibergite were recorded in single samples. A microinclusion of gold was found in only one sample of all

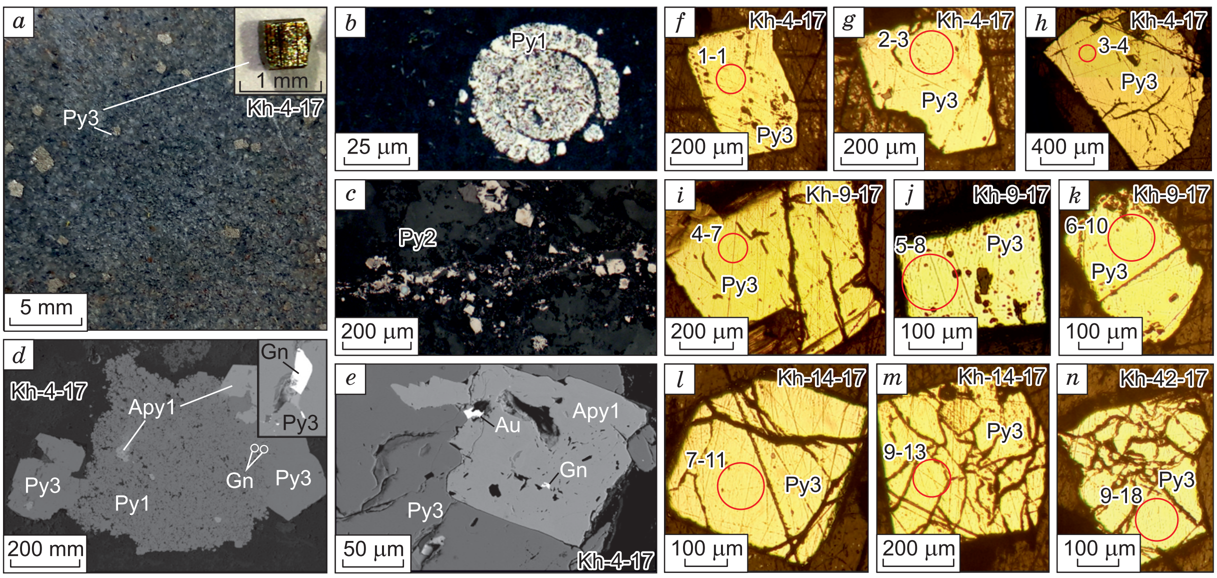

Fig. 2. Pyrite generations from metasomatites of the Khangalas deposit (*a*–*e*) and spot positions of LA-ICP-MS trace element analysis in Py3 (*f*–*n*): *a* – cubic crystals of Py3; *b* – syngenetic framboidal pyrite (Py1); *c* – fine-grained Py2; *d* – Py3 with galena inclusions in intergrowth with Apy1; *e* – Py3 in intergrowth with Apy1 with inclusions of gold and galena. Sample numbers and spot positions of LA-ICP-MS trace elements analysis correspond to numbers in Table.

Data of LA-ICP-MS trace element analysis of pyrite-3 from metasomatic rocks of the Khangalas gold deposit (all values in ppm, bdl – below detection limit)

Sample	Spot position	As	Au	Ag	Co	Ni	Cu	Zn	Pb	Sb	Ti	V	Cr	Mn	Mo	Te
Kh-4-17	1-1	5240	0.955	0.92	1.13	14.4	3.96	3.51	66	10.19	2470	7.16	10.3	0.85	0.079	0.056
	2-3	7630	0.502	0.0076	0.233	8.2	0.54	3.62	0.479	0.25	0.7	0.028	0.38	0.57	0.21	0.21
	3-4	4710	0.236	0.196	17.5	13.4	1.58	3.8	3.71	2	8.6	0.116	0.39	0.82	0.64	0.13
Kh-9-17	4-7	4530	0.507	0.85	1.15	74.9	3	4.2	860	407	0.78	0.058	0.52	0.41	0.059	0.083
	5-8	18790	8.83	0.055	7.05	39	19	4.7	3.35	6.19	36.1	0.35	0.98	0.4	0.047	bdl
	6-10	18550	15.85	0.8	21.3	56.9	18	5.3	26.7	40.1	2390	8.42	7.9	1.53	0.71	0.06
Kh-14-17	7-11	11060	2.5	1.01	505	690	7.3	6.4	23.3	8.42	77	0.235	0.58	7.55	1.04	0.31
	8-13	7340	0.143	0.062	41.6	1298	1.4	4.65	2.75	1.05	79	0.105	1.09	0.74	0.29	0.21
Kh-42-17	9-18	8640	1.028	0.128	43.5	64.9	1.29	4.22	3.34	1	208	0.274	0.77	0.73	bdl	bdl

examined ~ 100 grains, it has a size of about 15 μm, fineness of 827 ‰ (Fig. 2, *f*). It is assumed that native gold was formed synchronously with the lode gold of polysulfide association. Microinclusions in pyrite-3 are confined to defects and crystal growth zones. The Cu grade is low (0.5–19 ppm). The Ni grade ranges from 8.2 to 1298 ppm, with low (8.2–74.9 ppm) values prevailing. Co is also characterized by small grades (C_{Co} = 0.23–43.5 ppm), in only one sample does C_{Co} reache 505 ppm. Co has a moderate correlation with Ni (r = 0.42), and is closely associated with Zn (r = 0.8). Zinc is a constant impurity (v = 20 %) in pyrite-3; its content varies from 3.5 to 6.4 ppm, an average of 4.5 ppm. The amounts of Pb and Sb vary over a wide range from 0.5 to 860 ppm and from 0.3 to 407 ppm, respectively. There is a very close correlation between them (r = 0.996) and their negative correlation with Au is noted. This may indicate late processes superimposed on gold ore mineralization, and indirectly shows the polychronism and polygenicity of the Khangalas deposit. The titanium content varies widely from 0.7 to 2470 ppm, while in most samples the values are small (up to 208 ppm). Ti closely correlates with Cr (r = 0.99) and V (r = 0.96), the contents of which vary between 0.38–10.3 and 0.028–8.42 ppm, respectively. The sharply increased amounts of Ti (2470, 2390 ppm), Cr (7.9, 10.3 ppm) and V (7.16, 8.42 ppm) can be associated with microinclusions of Ti-, Cr- and V-containing minerals entrained from the host rocks during the growth of pyrite-3 grains. Mn varies from 0.57 to 7.55 ppm, Mo from 0.047 to 1.04 ppm, Te from 0.056 to 0.31 ppm. Figure 3 shows scatterplots of As vs. Au, Ag vs. Au, Zn vs. Au, Pb vs. Au, Cu vs. Au, Sb vs. Au, Te vs. Au, Co vs. Au, Ti vs. Au, As vs. Ni, Au vs. Ni, Zn vs. Ni.

Close relationship (r > 0.5) between Au and As in pyrite and arsenic pyrite from various types of gold deposits is noted by many authors [3, 23–26, etc.]. Elevated As contents are typical for pyrite with excess iron (S/Fe = 1.9–1.98). In the mineral structure, As isomorphically replaces S ($Fe_{1.00}(S_{1.98}As_{0.02})_{2.00}$), in some cases forming arsenic pyrite (As > 1.7 %), which is typical for reducing conditions [24, 27, 28]. Reich M. et al. [24] showed for epithermal and Carlin-type deposits an increase in the solubility of Au in the pyrite structure with increasing As content: $C_{Au} = 0.02 \cdot C_{As} + 4 \times 10^{-5}$. Based on the data of EMPA, LA-ICP-MS, SIMS and l-PIXE analyses, Deditius A.P. and co-workers [29], studied the mechanism of Au and As incorporation and the solubility of gold in pyrite from various types of deposits. In addition to the Carlin and epithermal types, pyrites of Cu-porphyry, Cu-Au, orogenic (OGD), volcanogenic-massive sulfide (VHMS), iron-oxide copper-gold (IOCG), and Witwatersrand Au and coal deposits were studied. The authors [29] found that on the Au–As diagram, the analytical results for pyrite from the gold deposits form a wedge-shaped zone and most points fall below the solubility limit of a solid defined by [24] (Fig. 4). They indicate that Au^{1+} is the dominant Au form in the arsenic pyrite of the studied deposits. Analytical data show that the solubility limit of Au in arsenic pyrite is ~ 0.02 for epithermal and Carlin-type deposits [24], and from OGDs ~ 0.004 [29] (Fig. 4). It appears that the solubility limit of Au depends on the crystallochemical properties of pyrite rather than on the geochemical environment of its formation [29]. Our results are in good agreement with the views of Reich M. et al. [24] and Deditius A.P. et al. [29]. Figure 4 shows that in the Au – As coordinates (mol. %), all at the studied

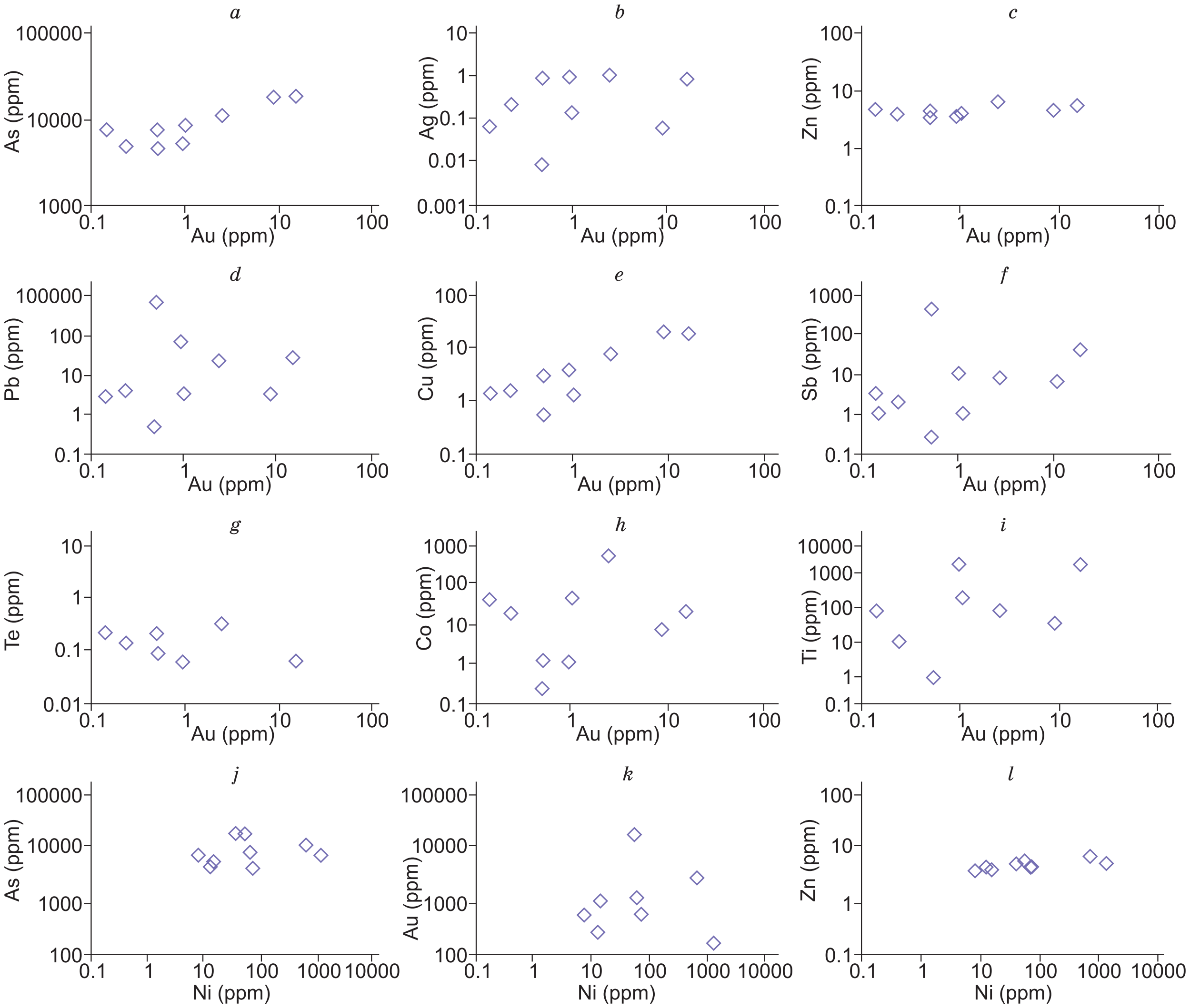

Fig. 3. Scatterplots of As vs. Au (*a*), Ag vs. Au (*b*), Zn vs. Au (*c*), Pb vs. Au (*d*), Cu vs. Au (*e*), Sb vs. Au (*f*), Te vs. Au (*g*), Co vs. Au (*h*), Ti vs. Au (*i*), As vs. Ni (*j*), Au vs. Ni (*k*), Zn vs. Ni (*l*).

samples of pyrites-3 from the Khangalas deposit fall into the field of structurally bound Au^{+} gold (Au/As < 0.02 according to [8] and < 0.004 – according to [29]). Arsenic at the Khangalas deposit controls Au behavior in the same way as observed for the orogenic and Carlin-type deposits (Figs. 3, 4) [24, 29]. These results are confirmed by rather low Au contents in the analyzed pyrites-3 – in most samples, Au does not exceed 2.5 ppm (see Table). This corresponds well with the studies of Tauson et al. [25], who established that the content of the Au structural form in all the samples of pyrite from deposits of different genetic types in Russia (large orogenic Natalka and Degdekan gold-quartz deposits, volcanogenic-plutogenic Dukat Au-Ag deposit, volcanogenic Dalnee and Oroch Au-Ag deposits, giant Sukhoi Log deposit with a controversial genesis, epithermal Pokrovskoye Au-Ag deposit, Amurskiye Dayki deposit with an unconventional type of mineralization, Zun-Kholbinskoe deposit with a controversial genesis) and Uzbekistan (epithermal Kochbulak and Kyzylalmasay Au-Ag deposits) does not exceed ~ 5 ppm. In only two samples from the metasomatites of the Khangalas deposit does the Au content reach more than 5 ppm (8.83 and 15.85 ppm). This may indirectly confirm the results of microscopic studies on the presence of a certain amount of the native form of gold in pyrites-3. Our results are also consistent with the viewpoints from [29] that pyrite in orogenic deposits usually contains less than 100 ppm Au. Higher contents are mainly associated with the presence of nano- and micro-

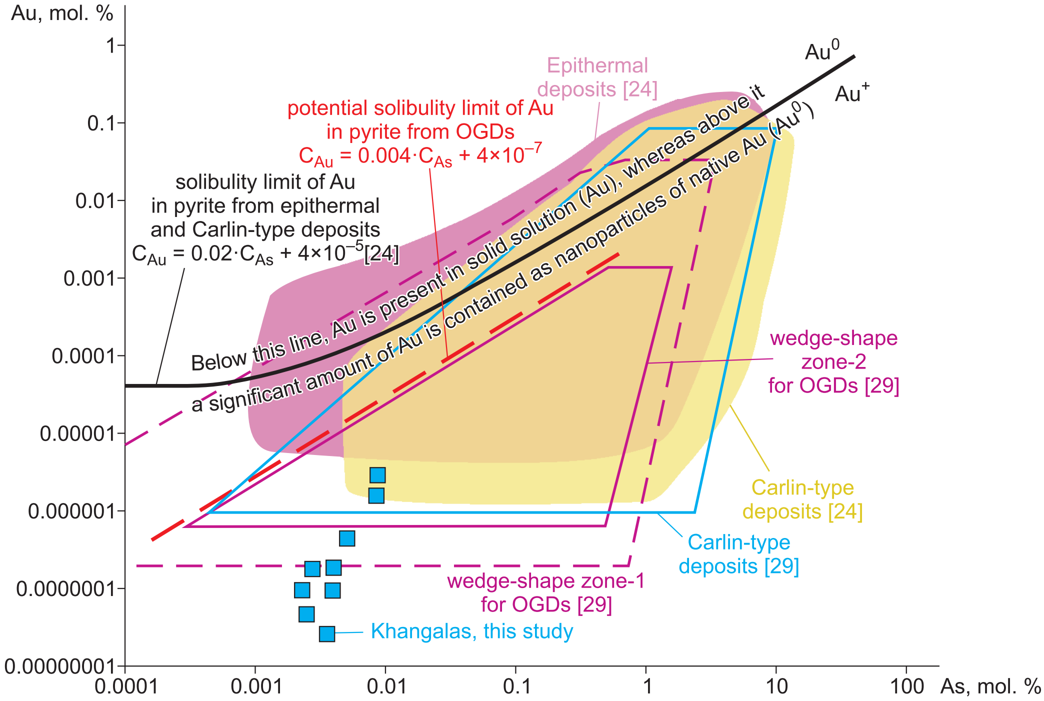

Fig. 4. Plot of log Au vs. log As (in mol. %) [24] for pyrite-3 from metasomatites of the Khangalas deposit.

particles [30]. The presence of native surface-bound (nano- or micro-) Au^0 in sulfides from metasomatites is also observed in deposits of various genetic types [24–26, 31].

Conclusions

In the metasomatites of the Khangalas deposit three pyrite generations were detected. The geochemistry of gold-bearing pyrite-3, as the most common and economically important, was studied. The presence of gold-bearing pyrite in metasomatites is of great practical importance and makes it possible to significantly expand the raw material potential of the Khangalas ore cluster. The total amount of impurities in Py3 is 0.48–2.12 %, with an average of 1.11 %. Arsenic is the main impurity; its grade ranges from 0.45 to 1.88 %. The grades of Au (0.1–15.9 ppm), Ag (0.008–1.01 ppm), Ni (8.2–1298 ppm), Co (0.23–505 ppm), Cu (0.5–19 ppm), Zn (3.5–6.4 ppm), and Pb (0.5–860 ppm) and Sb (0.3–407 ppm) are estimated. The grade of Au in Py3 gross sample determined by the atomic absorption method is up to 39.2 ppm, silver up to 17.38 ppm. The concentration of Au in pyrite-3 is directly proportional to arsenic grade (r = 0.9). In the examined ~ 100 grains of pyrite-3, microinclusions of galena and sphalerite are found; in single samples tetrahedrite and freibergite are noted. Microinclusion of Au^0 gold was found only in one sample; it has a size of about 15 μm, fineness of 827 ‰. It is assumed that it was formed synchronously with the vein gold-polysulfide association. Microinclusions are confined to defects and growth zones of pyrite-3 crystals. On the Au – As (mol %) diagram, the data points for Py3 samples from the Khangalas deposit fall below the solubility limit of Au in a solid. This indicates the form of «invisible» gold found in Py3 pyrite from the Khangalas orogenic gold deposit mainly as structurally bound Au^+.

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