

Microheterogeneity of the Koupol Deposit Fahlores as a Reflection of Changing of Physicochemical Parameters of the Ore-Forming Solution

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ABSTRACT: New data on a zonal structure of the Koupol deposit fahlores grains are given, and features of their chemism are shown. The fahlores chemical composition within the zones evolves from essentially arsenious (Fe-tennantite and Zn-tennantite), through mixed fahlores (Zn-tennantite-tetrahedrite arsenious and Zn-tennantite-tetrahedrite antimonous), to essential antimonous (Ag-bearing Zn-tetrahedrite). Varying chemical composition manifested as mineralogical-geochemical zonation of the fahlores grains is caused by changes of physicochemical conditions of the ore forming process during the time.

KEY WORDS: fahlore, mineralogical-geochemical zonation, physicochemical condition, Koupol deposit.

INTRODUCTION

Numerous publications devoted to fahlores (Kemkina, 2007; Spiridonov, 1985, 1984; Mozgova and Tsepin, 1983; Nekrasova and Sandomirskaya, 1979; Indoleev et al., 1974; Godovikov and Ilyasheva, 1972; Sakharova, 1966b; and others) describe this group of minerals as complex chemical compounds of a variable composition with a wide isomorphism both in cationic and anionic parts. Today, four mineral kinds, eight interspecific, and more than twenty intraspecific varieties of minerals of this group are

known (Mozgova and Tsepin, 1983). According to the above-mentioned researchers, variations of chemical composition of fahlores are caused by distinctions in physical and chemical conditions of the mineral-forming process, such as composition of the ore-forming solutions, their oxidation-reduction potential and acidity-alkalinity mode, and also chemical activity of sulfur and other elements. In this connection, a number of authors (Spiridonov et al., 1990; Gamyandin and Bortnikov, 1989; Moralev et al., 1988; Spiridonov, 1985; Mozgova and Tsepin, 1983; Rudashevskij et al., 1978; Andreeva and Loseva, 1976; Indoleev et al., 1974; Sakharova, 1966a) have made the attempts to connect the revealed by them features of the fahlores chemical composition with the certain conditions of mineral formation and to use these data as indicators of physical and chemical conditions of ore formation and evolutions of hydrothermal solutions responsible for a formation of ore deposits.

When studying ores of the Koupol gold-silver-bearing deposit, we have revealed cases of

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mineralogical-geochemical zonality of the fahlores grains that can also be used for finding out regularities in the evolution of chemical composition of the ore-forming solutions and for decoding of physical and chemical conditions of ores formation.

MATERIALS AND METHODS

The results we present here are based on the mineralogical-geochemical examination of more than 30 samples of ores of the Koupol gold-silver deposit. The polished samples of the ores have been studied on both the binocular reflected light microscope Zeiss AxioPlan 2 and the electron probe X-ray microanalyzer JXA-8100 (Japan). On the microanalyzer JXA-8100, researches were carried out in a mode of secondary electrons, allowing to receive the image of grains morphology and topography of their surface, as well as in a mode of reflected electrons, displaying contrast depending on average atomic number of element. The accelerating voltage is 20 kV, and the resolution of measurement is 137 eV at 5.9 keV. The precision of measurement depends on a matrix and analyzed element and changes within 0.01%–0.005%. The repeatability is 97%–98%. The electron probe X-ray microanalyzer JXA-8100 is equipped with the power dispersive analytical spectrometer Oxford Instrument INKA-sight (Great Britain). This spectrometer allows one to carry out quantitative analysis of the polished samples for definition of all elements heavier than Li in a point with locality from 7 μm for an easy matrix and to 1 μm for a matrix with large average atomic number and also to carry out quantitative analysis on all sample area.

MICROZONALITY AND CHEMICAL COMPOSITION FEATURES OF THE KOUPOPOL GOLD-SILVER-BEARING DEPOSIT FAHLORES

The Koupol deposit is located in the Anadyr area of Chukchi Autonomous Region (Northeast of Russia) on the northern flank of the Mechkerevskaya volcano-tectonic depression that is composed of Late Cretaceous volcanites of intermediate composition. These volcanites are intruded by small intrusions, subvolcanic bodies, and dykes of gabbro, diorites, dioritic porphyrites, andesites, basalts, dacites, and

rhyolite (Gluhov, 2008). The ore bodies are presented by small-sulphide quartz and adular-quartz veins and vein zones, which extend for about 3 800 m in a submeridional direction and have a width from 50 to 100 m.

Ores of the deposit belong to a gold-sulphosalt mineral type of gold-silver formations. They were formed in five stages: preproductive pyrite-adular-quartz, productive gold-pyrite-adular-quartz and gold-polysulfide-adular-quartz, post-ore antimonite-marcasite-quartz and hypergenic acanthite-jarosite (Vartanyan et al., 2005). According to the data of mineralogical researches, the fahlores minerals crystallized during both the gold-pyrite-adular-quartz and the gold-polysulfide-adular-quartz productive stages.

Microscopic studying of the Koupol deposit ores shows that the fahlores, along with other sulphosalts minerals (proustite, pyrargyrite, stephanite, and pearceite), are widely distributed and found in parageneses with native gold and argentiferous minerals. In this connection, their composition features and formation regularities acquire special interest. The fahlores form grains of various shape (from isometric and oval to extended and xenomorphic) and size (from thousand shares to 1–2 mm) hosted in quartz. Besides that, the fahlores make up close intergrowths with other ore minerals, such as pyrite, electrum, native silver, kuznetelite, stephanite, pyrargyrite, polybasite, and others. Individual grains of the fahlores contain microinclusions of galenite and naumannite. The fahlores are replaced by hypergenic minerals acanthite and covellite.

Intergrowths with chalcopyrite are most abundant. The fahlores of the late productive stage (gold-polysulfide-adular-quartz) are represented mainly by freibergite in the forms of simple crystalline aggregates. The fahlores of the early productive stage (gold-pyrite-adular-quartz), on the contrary, make up more complex crystalline aggregates observed as small inclusions, borders, and openwork edgings enveloping chalcopyrite (Fig. 1). In some cases, they make up aggregates of rhythmical alternation together with chalcopyrite. The chalcopyrite generally forms xenomorphic grains up to 0.4–0.6 mm in size with microscopic inclusions of fahlores and pyrite. The last one is presented by up to 0.08 mm spheroidal grains of sooty pyrite (which are often replaced by the fahlores) and to-a-variable-degree crystallized fine-

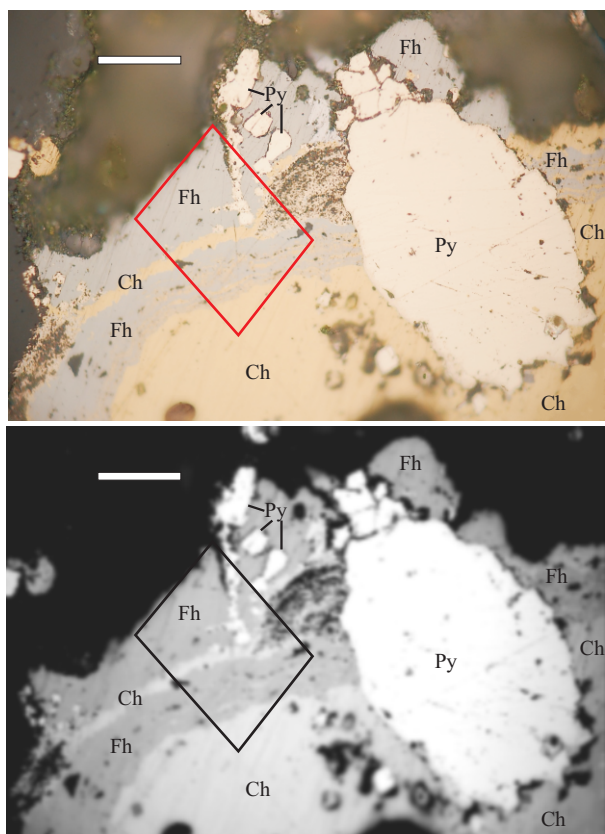


Figure 1. Intergrowth of chalcopyrite (Ch), fahlores (Fh) and pyrite (Py). Microphotographs in reflected light. The rectangle restricts the area shown in Fig. 3a. White marker is 40 μm .

grained pyrites, as well as by the pentagondodecahedral crystals of the later pyrite (Fig. 2).

Detailed study of interrelations of the fahlores of the early gold-pyrite-adular-quartz stage and the chalcopyrite has allowed us to reveal some interesting features. It has been observed under the binocular reflected light that the fahlores bordering the chalcopyrite form hardly perceptible alternating zones (strips), which at identical reflective ability have slightly different color shades. Studying these grains with the use of JXA-8100 microanalyzer in a mode of absorbed electrons (Figs. 3b–3h) has confirmed a zoned structure of the fahlores. In particular, the fahlores grains in Fig. 3b exhibit three and four consecutive zones of different thickness (from 0.005 to 0.015, less often to 0.03 mm) and color (gradual transition from dark grey to white). Boundaries between the zones are precise, sharp, and subparallel.

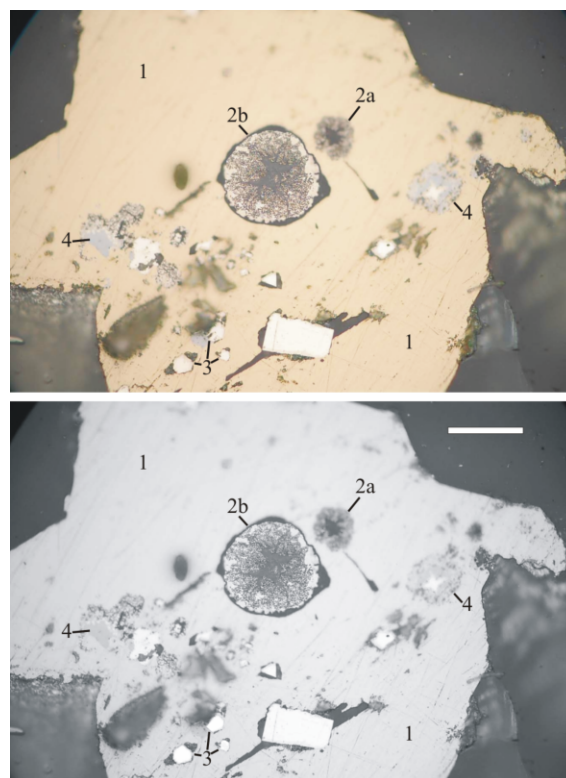


Figure 2. Inclusions of pyrite and fahlores in chalcopyrite. 1. Chalcopyrite; 2. spheroidal aggregates of pyrite: 2a. sooty-like, 2b. fine-grained; 3. pentagondodecahedral crystals of pyrite; 4. inclusions of fahlores. White marker is 100 μm .

An analysis of raster images (Figs. 3c–3h) of the same grains scanned in a mode of characteristic X-ray radiation (AgL_a , CuL_a , SbL_a , AsL_a , FeL_a , and ZnL_a) shows a heterogeneity of distribution of some chemical elements within the allocated zones that most likely is a consequence of distinction of their chemical composition. It is clearly visible on these images that the external (in relation to chalcopyrite) zones are more enriched in antimony and silver in comparison with the internal ones, which, on the contrary, are more abundant in arsenic. Concerning iron, zinc, and copper, the picture is not so expressive; nevertheless, it is possible to note that iron content is a little bit higher in the internal zones.

To find out the chemical composition of the specified zones, X-ray spectrum microanalysis has been executed (Table 1). The zones basic chemical elements were found to be Cu, Ag, Fe, Zn, Sb, As, and S. However, their contents in different zones vary over

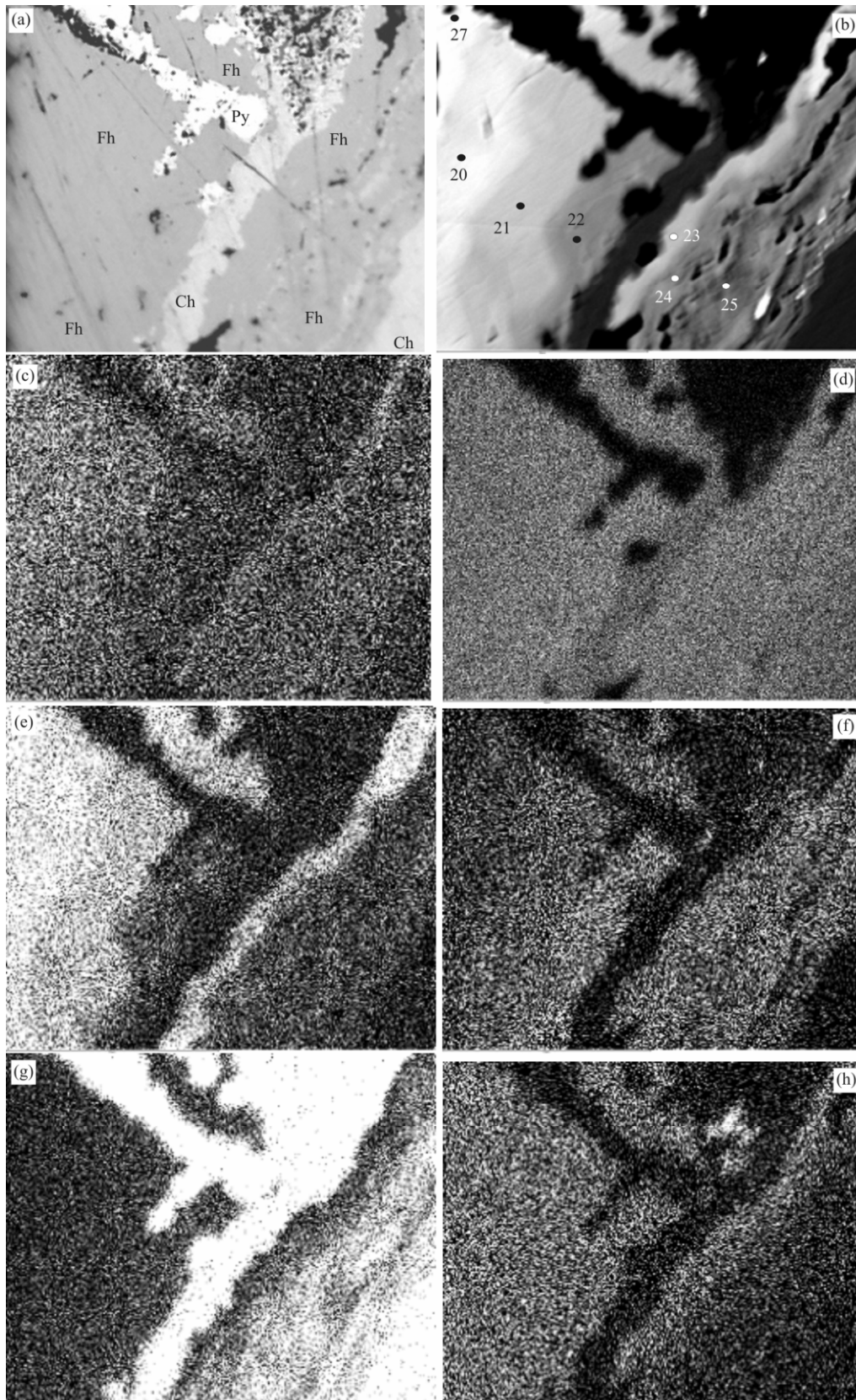


Figure 3. Microheterogeneity of fahlores. (a) Fragment of Fig. 1 within the restricted area; (b) microphotographs in the mode of absorbed electrons (points of observations are shown by figures); (c)–(h) raster images in the mode of characteristic X-ray radiation: AgL α (c), CuL α (d), SbL α (e), AsL α (f), FeL α (g) and ZnL α (h). Py. Pyrite; Ch. chalcopyrite; Fh. fahlores. Photo size is 100 μm \times 100 μm .

a wide range. For example, the amount of Cu varies from 33.59 to 42.59 (here and hereinafter in weight percent, wt.%); S from 23.55 to 28.82; Ag from 0.54 to 5.09; Fe from 1.25 to 5.49; Zn from 1.23 to 6.81; Sb from 3.32 to 23.24; and As from 2.97 to 17.67. At the same time, certain regularity in their distribution is observed. The internal zones are most enriched in arsenic (18.31 wt.% and 17.67 wt.%), iron (5.49 wt.% and 3.00 wt.%), sulfur (28.82 wt.% and 27.53 wt.%), and copper (41.53 wt.% and 42.59 wt.%), whereas the external zones have reducing amounts of these elements: arsenic up to 11.94 wt.% and 2.97 wt.%; iron

up to 3.27 wt.% and 1.25 wt.%; sulfurs up to 26.02 wt.% and 23.55 wt.%; and copper up to 40.13 wt.% and 33.59 wt.%. Distribution of such elements as antimony, zinc, and silver has an opposite tendency: they are the most abundant in the external zones (Sb 11.42 wt.% and 23.24 wt.%; Zn 5.51 wt.% and 6.81 wt.%; and Ag 2.07 wt.% and 5.09 wt.%) and the most scanty in the external (Sb 3.32 wt.% and 3.82 wt.%; Zn 1.23 wt.% and 5.78 wt.%; and Ag 0.54 wt.% and 0.72 wt.%). The fahlores intermediate zones are characterized by middle amounts of all the above-mentioned elements (Table 1).

Table 1 Chemical composition of fahlores zones (wt.%)

Points of observations	Zone	S	Fe	Cu	Zn	As	Ag	Sb	Σ
25*	Internal	28.82	5.49	41.53	1.23	18.31	0.54	3.32	99.24
24	↓	26.9	4.58	40.86	4.19	16.75	0.79	5.07	99.15
23	External	26.02	3.27	40.13	5.51	11.94	2.07	11.42	100.36
22	Internal	27.53	3.00	42.59	5.78	17.67	0.72	3.82	101.1
21	↓	25.85	2.11	39.44	5.9	11.18	1.89	12.56	98.94
20	↓	25.02	1.27	37.27	6.77	7.55	2.96	18.6	99.44
27	External	23.55	1.25	33.59	6.81	2.97	5.09	23.24	97.24

*. points of observations are shown on Fig. 3b.

Results of the X-ray spectrum microanalysis convincingly show that the zonality of the fahlores is caused by significant and often extremely contrasting fluctuations of arsenic and antimony contents and by their related changes of iron, silver, copper, sulfur, and zinc. There is a correlation between the varying contents of all elements within the zones under observation though. For example, greater amounts of silver and zinc correspond to a higher antimony content (Figs. 4a and 4b), and arsenic quantity increases as amounts of iron, copper, and sulfur increase (Figs. 5a and 5b). As it is known, the fahlores are characterized by wide isomorphism in both anionic part (semimetals group) and cationic part (metals group). The first type of isomorphism is caused by the internal crystallochemical reasons, and the second one is caused mostly by the action of external geochemical factors (for example composition of the ore-forming solutions). The main isomorphous monovalent metal of the fahlores is silver, which is mainly strongly attracted by

antimonous mineral kinds (Fig. 4a). We have established the maximum quantities of silver for tetrahedrite. Zinc and iron are the most common isomorphous bivalent metals in the Koupol deposit fahlores. The significant positive correlation of these metals with arsenic and antimony is observed. However, it should be noted that the zinc is more often associated with antimonous fahlores (Fig. 4b) and iron with arsenous one (Fig. 5a). The positive correlation is also established between copper and arsenic (Fig. 5b). Analysis of the data of the fahlores chemical composition shows that the cuprous fahlores are presented mainly by tennantite. Some fluctuations of the contents of above-mentioned metals are caused by microinclusions of sulfide minerals, such as pyrite, chalcopyrite, and arsenopyrite with which the fahlores form (as we above have specified) close intergrowths.

The chemical composition variations within the zones revealed in the fahlores are more evidently illustrated by crystallochemical formulas (Table 2),

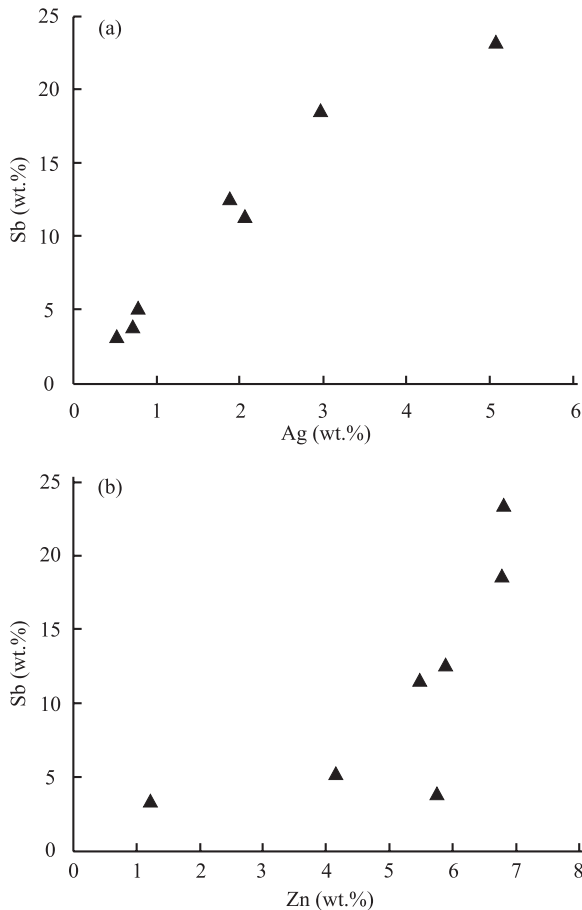


Figure 4. Correlation of contents Ag from Sb (a) and Zn from Sb (b).

which have been calculated according to the data of the X-ray spectrum microanalysis on the basis that the fahlore molecule contains 29 atoms in its formula. Table 2 shows that the zones revealed within fahlores grains are represented by different mineral kinds and their varieties. For example, the internal zones are composed of Fe-tennantite and Zn-tennantite. The intermediate zones are mixed in chemical composition and are composed of arsenious Zn-tennantite-tetrahedrite and antimonous Zn-tennantite-tetrahedrite. The external zones are represented by Ag-bearing Zn-tetrahedrite.

Thus, the composition of the fahlores within the zones under consideration changes from arsenious-rich to antimonous-rich through intermediate varieties, indicating the geochemical evolution of the ore-forming solution in time.

DISCUSSION

As mentioned above, the Koupol deposit fahlores

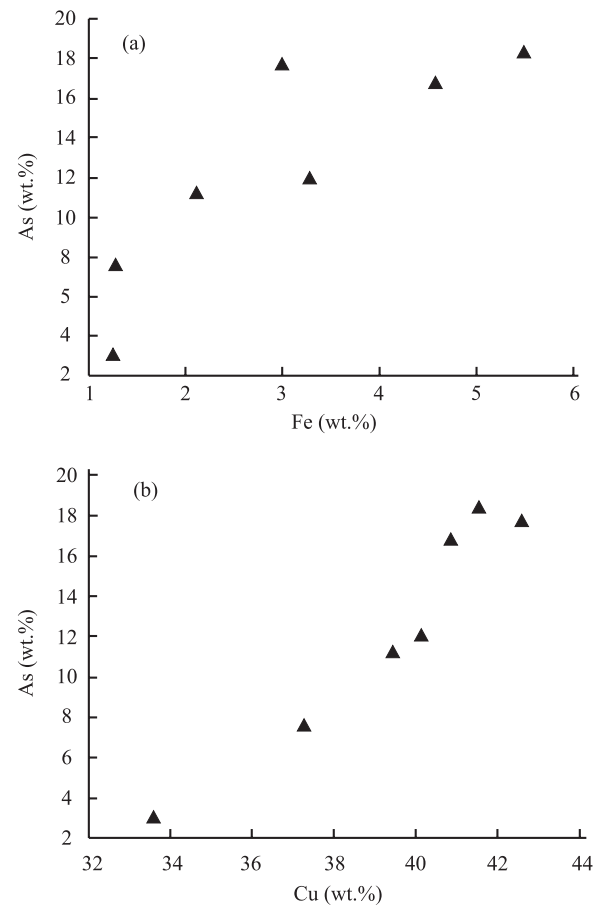


Figure 5. Correlation of contents Fe from As (a) and Cu from As (b).

form a single mineral association with sulfides, which are mainly presented by chalcopyrite and pyrite. The study of the fahlores-sulfides interrelations features (corrosive type of intergrowth borders, presence of fine inclusions of the fahlores, and replacement and encrustation of sulfides grains with the fahlores) testifies to the earlier crystallization of sulfides. In this connection, the study of these interrelations is very important for finding-out the succession and directivity of chemical reactions responsible for this mineral association formation. It is obvious that a subsequent crystallization of the fahlores occurred with the accompany of intensive chemical action of a hydrothermal solution on the earlier crystallized pyrite and chalcopyrite.

The change of sulfides deposition by the fahlores formation during the productive stage of ores crystallization is undoubtedly results of changed physical and chemical parameters of the ore-forming solution. One of these parameters is a temperature or, to be

Table 2 Crystallochemical formulas, kinds and varieties of fahlores

Points of observations	Crystallochemical formulas	Kinds and varieties
25	$[(\text{Cu}_{9.92}\text{Ag}_{0.07})_{9.99}(\text{Zn}_{0.28}\text{Fe}_{1.93})_{2.21}]_{12.20}(\text{Sb}_{0.30}\text{As}_{3.66})_{3.96}\text{S}_{12.84}$	Fe-tennantite
24	$[(\text{Cu}_{9.81}\text{Ag}_{0.11})_{9.92}(\text{Zn}_{0.97}\text{Fe}_{1.25})_{2.22}]_{12.14}(\text{Sb}_{0.64}\text{As}_{3.41})_{4.05}\text{S}_{12.81}$	Fe-tennantite
23	$[(\text{Cu}_{9.80}\text{Ag}_{0.20})_{10.00}(\text{Cu}^{+2}_{0.16}\text{Zn}_{1.31}\text{Fe}_{0.91})_{2.38}]_{12.38}(\text{Sb}_{1.47}\text{As}_{2.49})_{3.96}\text{S}_{12.66}$	Zn-tennantite-tetrahedrite
22	$[(\text{Cu}_{9.90}\text{Ag}_{0.10})_{10.00}(\text{Cu}^{+2}_{0.10}\text{Zn}_{1.32}\text{Fe}_{0.80})_{2.22}]_{12.22}(\text{Sb}_{0.47}\text{As}_{3.52})_{3.99}\text{S}_{12.79}$	Zn-tennantite
21	$[(\text{Cu}_{9.72}\text{Ag}_{0.28})_{10.00}(\text{Cu}^{+2}_{0.14}\text{Zn}_{1.44}\text{Fe}_{0.60})_{2.18}]_{12.18}(\text{Sb}_{1.64}\text{As}_{2.36})_{4.00}\text{S}_{12.82}$	Zn-tennantite-tetrahedrite
20	$[(\text{Cu}_{9.55}\text{Ag}_{0.45})_{10.00}(\text{Cu}^{+2}_{0.03}\text{Zn}_{1.69}\text{Fe}_{0.37})_{2.09}]_{12.09}(\text{Sb}_{2.50}\text{As}_{1.64})_{4.14}\text{S}_{12.77}$	Zn-tennantite-tetrahedrite
27	$[(\text{Cu}_{9.17}\text{Ag}_{0.83})_{10.00}(\text{Cu}^{+2}_{0.05}\text{Zn}_{1.74}\text{Fe}_{0.35})_{2.14}]_{12.14}(\text{Sb}_{3.33}\text{As}_{0.70})_{4.03}\text{S}_{12.83}$	Ag-bearing Zn-tetrahedrite

more exact, its reduction during the ore deposition. It has been established experimentally (Feiss, 1974) that stibium content in fahlores increased with reduction of the ore-forming solution temperature. For example, if a temperature of the ore-forming solution is 500 °C, the fahlores with high antimony and little arsenic (tetrahedrite) are formed (Sakharova, 1966a). A temperature of 400 °C and lower is sufficient for the tetrahedrite (fahlores with high antimony and little arsenic) to be formed. From this, it follows that tennantites have more high-temperature formations in comparison with tetrahedrites. Thus, the revealed by us zonality of the fahlores may be evidence for the reduction of temperature of the ore-forming solution during the ore deposition.

However, the temperature is not the only (or the main) factor that is responsible for a change of mineral formation conditions in time. A change of chemical composition of the ore-forming solution and its acidic-alkaline mode in particular is of no less importance. According to the data of experimental researches (Olshanskij and Ivanenko, 1958), solubility of iron, copper, lead, and several other metals sulfides increases with the change of the solution's acidity-alkalinity parameter from acidic to subacidic (pH>5). Other experimental data (Bobko and Lisetskaya, 1956) show that reduction of hydrothermal solutions acidity leads to the formation of arsenic and antimony complex anions and the residual solution enrichment with anions $(\text{AsS}_3)^{3-}$ and $(\text{SbS}_3)^{3-}$. These authors also note that arsenic anions become steady already in subacidic-neutral solutions (a favourable environment for tennantite formation), whereas antimony anions get

stability only in alkaline environments and break up in neutral-subacidic solutions, i.e., more alkaline environment is required for tetrahedrites to be formed. From this, it follows that the fahlores that crystallize under conditions of decreasing solution acidity are characterized by a different ratio of arsenic and antimony that is reflected in their mineral composition as a consecutive change of tennantite by tetrahedrite through intermediate varieties. Thus, we may conclude, on the strength of the above-cited data on mineralogical and geochemical zonality of the fahlores and the experimental data of the other authors, that the process of the Koupol deposit ores formation went on under conditions of distinct evolution of the ore-forming solution from the essentially acidic (sulfides crystallization) to alkaline (tetrahedrite crystallization) on a background of its temperature reduction.

It is worth noting that some researchers also use such a parameter as oxidation-reduction potential of the ore-forming solution to characterize physical and chemical conditions of ore formation (Spiridonov, 1985). For example, when the oxidizing potential of the ore-forming solution is high, high-cuprous fahlores are formed. If this potential is decreased, moderate-cuprous fahlores are crystallized. Moreover, the low oxidizing potential causes crystallization of small-cuprous fahlores. Following the given considerations, it is possible to conclude that the Koupol deposit fahlores were formed on a background of the oxidizing potential decrease during the ore deposition, because copper contents in the early fahlores makes up 42.59 wt.% and only 33.59 wt. % in the late ones.

Taking into account all the above mentioned de-

tails, the ore-forming mechanism for the early productive stage of the Koupol deposit can be presented as follows: at the initial stage, sulfide minerals have been generated first from high temperature and high acid at that time of primary ore-forming solution. The residual solution must have a reduction in concentrations of iron, copper, sulfur, zinc, and some other metals. The acidic-alkaline potential seems to be reduced as well. As the solution has obtained subacidic-neutral properties, a formation of arsenic sulfoanions began causing crystallization of tennantite; after reaching the alkaline properties, the generation of antimonies sulfoanions took place with further crystallization of tetrahedrite.

During the fahlores crystallization, complex anions $(AsS_3)^{3-}$ and $(SbS_3)^{3-}$ became bound with copper cations, which were partially taken from the residual solution, partially from the sulfides and mainly from chalcopyrite because the decrease of solution acidity increased the solubility of sulfides. On the dissolution of sulfides, the copper cations were completely inherited by the fahlores. As for iron cations, only a part of them have entered into the composition of minerals of this group (up to 5.49 wt.% maximum, see Table 1). The remained iron was redeposited in the form of sooty substance, which later underwent selective recrystallization that resulted in the formation of aggregates of fine-grained pyrite with metacolloidal structure. It is distinctly observed at the microscopic study of the Koupol deposit ores.

As already noted, gold and silver minerals are closely associated with the fahlores. Taking into account the revealed physical and chemical conditions of the fahlores formation, it is possible to make the assumption that gold and silver mineralization of the Koupol deposit occurred at final stages of the mineral formation process of productive stages, which were characterized by the low acidic-alkaline potential of the residual solutions.

CONCLUSIONS

1. It was revealed by the detailed microscopic researches with the use of X-ray microanalyzer that the fahlores of the Koupol deposit gold-pyrite-adular-quartz stage are represented by zonal aggregates of consecutive alternation of crystals of different chemical composition.

2. According to the data of the X-ray spectrum microanalysis, the zonality of the fahlores is a result of significant and often extremely contrast fluctuations of arsenic and antimony amounts and associated with them changes of quantities of iron, silver, copper, sulfur, and zinc.

3. Crystallization of the zonal fahlores of the Koupol deposit was caused by the evolution of chemical composition of the ore-forming solution from essentially acidic (crystallization of sulfides) to alkaline (tetrahedrite crystallization) on a background of its temperature reduction and decrease of the oxidation-reduction potential.

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