VARIATIONS IN THE COMPOSITION OF ORE-FORMING SULFIDES AND THE CONDITIONS OF FORMATION OF PYRITIC ORE BODIES

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In general form, the current ideas concerning the formation of pyritic ores are based on the recognition of their genetic relationship to foci of basaltoid early geosynclinal submarine vulcanism, of the prolonged, multistage history of their development, of the predominance of the hydrothermal-sedimentary and hydrothermal-metasomatic forms of ore deposition, and of the different degrees of their metamorphic transformation (from unaltered to strongly metamorphosed). These positions, which have been formulated most completely in the publications of V. I. Smirnov [15, 16], are based chiefly on numerous geologic-structural, lithologic-petrographic, regional-metallogenic, and other data, and also on studies of the mineral composition of these ores.

It is known that the mineral composition of the ores forming the deposits under consideration is quite uniform, being characterized by a constant sharp predominance of sulfides of iron, copper, and zinc (pyrite, chalcophyrite, sphalerite, etc.), which are stable over practically the whole range of endogenic mineralization conditions.

Thus the ore-forming sulfides as the main determining components of pyritic ore bodies are only a limited source of genetic information: at one level of the classic methods of study (optical microscopy, etc.) one can ascertain only their relative quantities, the morphology of the individual mineral grains and their aggregates, and some of their physical properties (hardness, thermoelectric power reflectivity, etc.), and diagnose (very tentatively) the rare minerals. The most important typomorphic features of the sulfides—their chemical composition—remains unknown because of the extreme difficulty of extracting, from the usually poly-mineralic ores with fine textures and complex structures, monomineralic fractions suitable for chemical analysis. Yet the contents of micro- and macroadmixtures in the sulfides, and the relationships of their main (constitutional) components, are the quantitative basis for distinguishing the generations of minerals, the characteristics of their zonality, the genesis of convergent structures, for determining certain physicochemical conditions of mineralization, etc.

All this explains the very great importance of the various forms of microanalysis, which yield fundamentally new information important for understanding the conditions of ore formation: the chemical composition of the individual mineral grains, and their areas only a few microns in size, observable under the microscope.

On the basis of previously developed particular methods of quantitative electron microprobe and laser-emission microanalyses [4,8,20], in combination with field and mineragraphic methods of study of the ores in a number of pyritic ore deposits of the Southern Urals, the Rudnyi Altai, the Northern Baikal, Japan, and other regions, the present author together with a group of students, apprentices, higher degree candidates, and colleagues in the Department of Mineral Resources of the Moscow University School of Geology has made a study of the composition of the ore-forming sulfides within individual ore bodies. It was found that the changes in composition of these minerals follow particular patterns in time and space, thus reflecting various phenomena of the complex picture of ore deposition and subsequent metamorphism.

Stages of Ore Formation (Generations of Pyrite). One of the main criteria of the stages of ore formation must be considered to be the presence of different generation of a mineral formed at different times. In the study of pyrite–the most common mineral in pyritic ores–it turned out that besides the usually observed differentiation of its grains according to morphology, paragenesis, and properties, its generations also clearly differ (within the scope of a single ore deposit) in composition. The most substantial is the variation in contents of microadmixtures—cobalt and nickel—of single metals, whose isomorphous presence in place of iron in the crystal lattice of pyrite (as well as chalcopyrite and pyrrhotite) is not subject to doubt. Experimental and theoretical studies by a number of authors [1,11] have shown that the concentrations of cobalt and nickel in sulfides are determined by their contents in the initial hydrothermal (or metamorphic) solutions, the chemistry of these solutions, the associations of sulfide minerals thus formed, and the physicochemical conditions of their crystallization (primarily the temperature). Thus the contents of these metals and especially their ratios may be adopted as indicators of the generation of pyrite.

An over-all consideration of the ores of pyritic-polymetallic deposits definitely reveals four generations of pyrite. The first generation (pyrite I) is usually a fine- and very fine-grained aggregate that is often typified by metacolloidal structures and framboids formed as a result of precipitation from solutions and subsequent crystallization. An allotriomorphic aggregate of medium-sized and large grains, usually with a zonal structure, corresponds to the second generation (pyrite II), which is understood to result from collective crystallization of the fine- and very fine-grained aggregate of pyrite I. A granoblastic aggregate of medium-sized and large grains, sometimes oriented, makes up the third generation (pyrite III), which is a product of metamorphic recrystallization of previously formed purite bodies. The fourth generation (pyrite IV) is represented by regular, euhedral, most often cubic crystals that as a rule are uncorroded, within a mass of other sulfides: chalcopyrite, sphalerite, pyrrhotite, and galena. The formation of this last generation is due to reprecipitation of part of the previously formed pyrite by hydrothermal, including metamorphic, solutions together with neogenic "late" sulfides. The amounts of these generations of pyrite are far from equal. As a rule, pyrite II and pyrite III predominate sharply. These, which are often hard to distinguish from each other, form the "basis" of the ore bodies, are present in all the types of ores, and are corroded and replaced by other sulfides. Pyrite IV is quite extensively developed in pyritic-polymetallic ores. It varies in form and size of its grains and in their association with other sulfides. Thus individual investigators have often distinguished a larger number of generations of this mineral than in the scheme outlined above.

Table 1 summarizes the characteristic compositions of the pyrite generations distinguished in four ore deposits in different regions of the USSR. It can be seen that the pyrite generations differ from each other within each ore deposit. The most sharply dif-

Table 1

Cobalt and Nickel Contents in Pyrite Generations of Pyritic and Pyritic-Polymetalic Ore Deposits

Ore deposit	Element	Generations of pyrite											
		I			II	III			IV				
			Framboids	Reniform			In sulfur-pyrites	Polymetallic	Corroded in sphalerite	In pyrrhotite	In chalcopyrite	In chalcopyrite	In sphalerite
Dzhusinskoe (Southern Urals)	Co Ni Co/Ni	12,5 5,5 2,3			14,1 4,2 3,4						5,7 5,2 1,2		
Shemonaikha (Rudnyi Altai)	Co Ni Co/Ni	7 2,5 2,8			8 2,5 3,2				2,5 5 0,5			5 6 0,8	5 3,5 1,4
Parandovskoe (Karelia)	Co Ni Co/Ni	13(59) 7(56) 1,9				10(74) 5,5(74) 1,8				15(83) 16(116) 0,9			
Kholodninskoe (Northern part of Baikal region)	Co Ni Co/Ni		8,1(12) 11,7(23) 0,7	5,6(26) 6,9(55) 0,8	5,9(39) 6,7(55) 0,9		7,7(45) 10(56) 0,8	7,2(35) 7(40) 1,0	8,9(44) 5,8(47) 1,5	17,8(99) 9,4(60) 1,9			

Note. Coefficients of variation in cobalt and nickel contents given in parentheses.

ferent is pyrite IV: in the majority of cases it is typified by a distinct diminution of the Co/Ni ratio relative to the pyrite of earlier generations. The exception is the Kholodninskoe ore deposit, where the picture is directly the opposite: this ratio increases sharply in the pyrite IV, which according to N. I. Bezmen's experimental data [1] may be explained by the hydrocarbonate (rather than chloride) composition of the late hydrothermal-metamorphic solutions from which the pyrite of this generation was precipitated.

On the example of the Parandovskoe and Kholodninskoe ore deposits (Table 1), one can quite clearly see a regular increase in the variation coefficients of the cobalt and nickel contents from the earlier generations of pyrite to the later. This testifies to the comparatively persistent, steady regime of formation of the initial pyrite masses with subsequent repeated regrouping of the material in its crystallization, recrystallization, and reprecipitation, which occurred during the stage-by-stage ore formation and metamorphism. This circumstance clarifies the ideas of F. Hegemann [21], G. A. Krutov [10], and B. Cambel and J. Jarkovsky [19] concerning the possibility of distinguishing "sedimentary" and "hydrothermal" pyrite accumulations, but now within a single ore deposit and a single ore body.

As was shown in a number of publications by the present writer and his colleagues [3, 8,14,24], the pyrites of different generations can also be differentiated by their S/Fe ratio and by certain physical properties. It must be emphasized that the typomorphic metals of these ores—copper, zinc, lead, and silver—correspond in their contents in the pyrite grains to the total amount of the surrounding typomorphic minerals (chalcopyrite, sphalerite, galena, or gray copper ore [tennantite], respectively. It seems that these metals are contained in the pyrite chiefly in the form of minute inclusions of the above-listed minerals.

Concealed Mineral-Geochemical Zonality as Indication of Mechanism of Formation of Ore Bodies. Study of the composition of individual grains of ore-forming sulfides (pyrite, chalcopyrite, sphalerite, pyrrhotite) and of sulfosalts (gray ore—tennantite) belonging to one (the predominant) generation within the contours of individual ore bodies has been the means of establishing concealed mineral-geochemical zonality. Numerous concrete examples of such zonality are cited in a number of publications by the present writer [5,6,7,9,17], and can thus be omitted from this paper, where the discussion can be limited to the phenomenon as a whole (Table 2). The specific characteristics of this zonality are the mineral by which it is determined, and the chemical element (or ratio of chemical elements) whose concentration is measured in the individual grains. At the present time, such a zonality has been established for a large number of pyritic and pyritic-polymetallic ore deposits, both by the changes in contents of microelements (Co, Ni, Cu, Zn, Ag) in pyrite and chalcopyrite and by the fluctuations of the contents of the elements determining the constitution of the minerals of variable composition (tennantite, pyrrhotite, chalcopyrite) or present in them as macroadmixtures (iron in sphalerite).

Two main structural types of this zonality have been established: repeating (concentric) and nonrepeating. The first type is characterized by either symmetrical or asymmetrical change in composition of the mineral from the central part of the ore body toward its periphery, with the formation of a concentric structural zonality. The second type is characterized by directional change in chemical composition of the mineral from one wall of the ore body to the other with the formation of a zonality of nonrepeating, "layered" structure.

In pure form, these types of zonality usually appear in comparatively small homogeneous ore bodies. In ore bodies of complex, usually heterogeneous composition, both nonrepeating and repeating types of zonality may appear. In connection with the latter, either both types of zonality may occur in the same mineral, but belonging to two generations, present in different and spatially separated types of ores of the body, or else these types of zonality may be combined in one type of ore, reflecting the regular changes in two different minerals.

The repeated (concentric) zonality may be both symmetrical and asymmetrical. Nonrepeating zonality in isolated cases (such as the "yellow" ores of the Shakanai ore deposit) is complicated by cyclicity. In the case of a symmetrical repeating zonality its contrast (that is, the gradient or sharpness of change in composition of the mineral is the same on both sides toward the contact of the ore body.

The hidden mineral-geochemical zonality of ore bodies in the majority of cases is primary—that is, was formed in the crystallization of the minerals that determine it. This is confirmed by the sharply weaker contrast of the identical zonality in the Proterozoic ore deposits as compared to the Paleozoic, in consequence of the more profound meta-

Table 2

Concealed Mineral-Geochemical Zonality of Pyritic Ore Bodies

I Character of ore bodies in which zonality is manifested	1) ore bodies of homogeneous composition 2) parts of ore bodies of homogeneous composition 3) zonal ore bodies (differentiated by types of ores)	
II Minerals and metals as indicators of zonality	1) pyrite (Co, Ni, Co/Ni) 2) chalcopyrite (Co, Ni, Co/Ni, Fe) 3) sphalerite (Fe) 4) pyrrhotite (Fe) 5) tennantite (As, Sb, As/Sb, Ag, Cu, Pb, Zn, Fe)	
III Structure of zonality in transverse sections through ore bodies	1) repeating (symmetrical, asymmetrical) 2) non-repeating (simple, complicated by cyclicity) 3) combined	
IV Contrast of zonality	1) contrasting 2) noncontrasting	
V Nature of zonality	1) deposition 2) deposition by stages	exhalational-sedimentary ore formation
	3) filtrational 4) precipitation	hydrothermal-metasomatic ore formation
	5) superimposed filtrational 6) veinlet-deposition	metamorphic ore formation
	7) diffusional redistribution	
Examples of zonality (ore deposits and character of zonality found in their ore bodies)	Gaiskoe (I_1, $II_{1,2}$, III_1, IV_1, $V_{3,5}$); Dzhusinskoe (I_1, II_1, III_1, IV_1, V_3); Osennee (I_1, II_1. III_2, IV_1, V_1); Priorskoe (I_3, II_1, III_2, IV_1, V_2); Oktyabr'skoe (I_1, II_3, III_1, IV_2, V_4); Guslyakovckoe (I_2, $II_{3,5}$, III_1, $IV_{1,2}$, V_4); Parandovskoe (I_1, $II_{1,2,3,4}$, $III_{1,2,3}$, $IV_{1,2}$, $V_{1,6}$); Kholodninskoe (I_2, II_1, $III_{1,2,3}$, $IV_{1,2}$, $V_{1,6}$); Sazare (I_1, II_1, III_1, IV_1, V_7); Shimokawa (I_1, II_1, III_2, IV_1, V_1); Shakanai (I_1, II_1, III_2, IV_1, V_1)	

morphic transformations, which are expressed in the recrystallization of sulfide masses (but retaining signs of their primary structures and textures), accompanied by "averaging" of the distribution of minor elements within them. Five types of primary zonality can be distinguished: depositional, stage-by-stage depositional, veinlet depositional, filtrational, and precipitational. Along with these, in the specific situation of intensive regional metamorphism under conditions of high pressure against a background of complete recrystallization and redeposition of the ores, one can discern the rise of a repeating concealed mineral-geochemical zonality resulting from differential migration of the material in the ore body by diffusion (as in the Sazare ore deposit, or the Sambagawa ore deposit that is localized in the metamoorphic belt of the outer zone in Japan).

Study of the concealed mineral-geochemical zonality in a large number of ore deposits has established the complex and varied nature of the precipitation of pyritic ores (in accordance with the five types distinguished above), has revealed volcanogenic-sedimentary (nonrepeating zonality, sometimes complicated by cyclicity), hydrothermal-metasomatic, and metamorphic (repeating zonality) ore bodies, and has demonstrated the variety of spatial orientations of the ore-localizing structures (symmetry of zonality) in different stages of ore formation.

Temperature of Ore Formation According to Distribution of Admixture Elements in Coexisting Sulfides. The distribution of admixture elements within an individual gtain of sulfide often turns out to be nonuniform. This usually happens when the grain of one sulfide is contained within a different sulfide: the concentration of the admixture element in the marginal part of a fairly large grain differs markedly from the concentration in its central part. Such a pattern, in particular, was established in the pyrites contained within the pyrrhotite ores of the Gornaya Abkhaziya area of the USSR [8]. It has now been shown [12] that the distribution of minor components is characterized by a strict and definite regularity, so that the distribution coefficients can be used to estimate the thermodynamic conditions determining the equilibrium of natural systems, particularly the temperature.

On the basis of N. I. Bezman's experimental data [2] on the distribution of cobalt and nickel in coexistent ore-forming sulfides (pyrite, pyrrhotite, and chalcopyrite), extrapolated into the region of low temperatures, and the use of electron-probe and laser microanalyzers by the present writer (together with Nat. E. Sergeeva, A. G. Tvalchrelidze, T. P. Kuznetsova, and N. V. Pozdnyakova), it has been possible to obtain the temperatures at which these minerals were formed in the ores of a number of ore deposits (Table 3).

Table 3

Temperatures of Formation of Pyritic Ores According to Distribution of Admixture Elements in Coexisting Sulfides

Ore deposit, ores	Admixture elements	Coexisting sulfides	Temperature, °C		Data by other methods
			Range	Average	
Shemonaikha: D copper pyrite—polymetallic C skarn	Co Co Co	 Py—Chp Py—Chp		190 330	V. V. Vortsepnev, homogenization of gas-liquid inclusions in garnets of epidosites, 320-350°
Osennee: D_2e copper-zinc thermal metamorphism	Co	Py—Chp Py—Chp		160 300	
Kholodninskoe: Pt pyrite-pyrrhotite of late hydrothermal-metamorphic stage	Co	Py—Po	250—400	315	By Arnold method 330—300° (av. 310°)
Parandovskoe: Pt pyrite-pyrrhotite of late hydrothermal-metamorphic stage	Co,Ni	Py—Po	420—430	420—430	By Arnold method (av. 360°)
Sazare: Pz pyrite-chalcopyrite with sphalerite	Co	Py—Chp		200	By Masao's estimate (analysis of metamorphic facies), 200—300°
Nikolaevskoe: D_2e pyrite-chalcopyrite	Co	Py—Chp	80—260	140	
Shakanai and Hanawa: Middle Miocene, "black" and "yellow" ores	Co	Py—Chp	70—140	100	1) Kajiwara[22], thermodynamic analysis of mineral parageneses, 80-250° 2) Thermobarometry (Tokunaga and Honma [25]), 70-150°
Kamyshinskoe: D_2e copper-zinc, copper	Co,Ni Co,Ni	Py—Chp Py—Chp		245 200	
Shimokawa: Mz pyrite-chalcopyrite-sphalerite with pyrrhotite	Co Co	Py—Po Py—Chp		320 325	
Gornaya Abkhaziya: J pyrrhotite-polymetallic with pyrite	Co	Py—Po	305—330		A. G. Tvalchrelidze, homogenization of primary inclusions in quartz syngenetic with pyrrhotite, 298—300°

The copper-pyrite—polymetallic ores of the Nikolaevskoe ore deposit in the Rudnyi Altai region were formed at temperatures in the range of 80-260°, with an average of 140°C. The closely similar exhalational-sedimentary "yellow" and "black" ores of the Hanawa and Shakanai ore deposits (of the Kuroko type, Japan), which are related genetically to acidic submarine volcanism on a continental crust, appear to have been formed in the temperature interval of 70-140°C (average about 100°C), which is consistent with the thermodynamic data of Kajiwara (80-250°C [22]) and the thermobarometric determinations of Tokunaga and Honma (70-150°C [25]).

The ores of the Kamyshinskoe ore deposit in the Rudnyi Altai) were formed in stages: hydrothermal solutions of the subsequent stages have been repeatedly superimposed on the volcanogenic-sedimentary sulfides, resulting in the formation first of copper-zinc ores at the temperature of 245°C and then of copper ores at the temperature of 205°C. Analagous in the mode of their formation are the Mesozoic superimposed copper ores of the Shimokawa ore deposit (in Japan), formed at temperatures of 320-325°C. The hydrothermal copper-pyrrhotite ores of the Gornaya Abkhaziya region were also formed at temperatures of 305-330°C.

The copper-pyritic—polymetallic ores of the Shemonaikha ore deposit, which were formed at the temperature of 190°C and are of volcanogenic-sedimentary origin, are complicated by a superimposed skarn-forming process (owing to the effect of the Zmeinogorsk granitoids), which took place at the higher temperature of 330°C. Two temperature values were likewise obtained also for the Osennee ore deposit in the Southern Urals—160°C for the volcanogenic-sedimentary ores and 300°C for the same ores in the zones of their thermal metamorphism by dikes of granitoids.

The pyrite-pyrrhotite ores of the Kholodninskoe deposit (in the Northern Baikal region), formed in the late hydrothermal-metamorphic stage [13], showed a temperature of formation in the 250-400°C range (average 315°C), which is about the same as the data obtained for these same ores by the Arnold method (300-330°C, average 310°C). The analogous bodies of the Parandovskoe ore deposit (in Karelia) correspond to the temperature interval of 420-430°C, which is somewhat above the temperature of their formation as obtained on the pyrite-pyrrhotite solvus by the Arnold method (360°C).

The copper—copper-pyrite Sazare ore deposit, which is localized in the Sambagawa metamorphic belt (Japan), together with its host rocks underwent intensive regional metamorphism under conditions of high pressures and low temperatures, which according to Masao's estimates [23] were 4-5 kbar and 200-300°C, respectively. The minerals in the ores were moreover entirely recrystallized, according to the present author's determinations, at temperatures of about 200°C.

On the whole, the temperatures cited above agree with the data obtained by other methods, and are consistent with the geologic conditions characterizing the formation of the separate ore deposits.

Conclusion. 1. Study of the composition of individual grains of the chief ore-forming sulfides in copper-pyritic ore deposits by the methods of electron-probe and laser-emission microanalysis has become a new source of important genetic information on the conditions of copper-pyrite ore formation. It was established that the variations in composition of these grains are characterized by definite and strict regularity in space and time.

2. Differentiation of pyrites by their contents of the admixture elements cobalt and nickel and their ratio, as well as by the ratio of sulfur to iron, in combination with their distinctive features of morphology and paragenesis, are important criteria for distinguishing generations of this mineral, confirming the duration and stages of ore formation and the evolution of the ore-forming solutions' composition with time.

3. The character of the hidden mineral-geochemical zonality detected by the change in composition of a sulfide of one generation within the contours of ore deposits reflects the variety of mechanisms of formation of the latter: volcanogenic-sedimentary, hydrothermal-metasomatic, and metamorphic.

4. The temperatures of mineralization, as determined by the distribution of microadmixtures of cobalt and nickel in the coexisting sulfides of a number of ore deposits, fall within a very wide range (from 70 to 430°C), reflecting the variety of conditions of volcanogenic-sedimentary, hydrothermal-metasomatic, and hydrothermal-metamorphic formation and thermal metamorphism of the pyritic ores.

5. The above-mentioned data on the variations in composition of the ore-forming sulfides in pyritic deposits support the prevailing idea of the polygenic and polychronic formation of these still largely puzzling geologic bodies.

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