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Textural-structural characteristics and mineral types of ores of the Aghlig-Filfilli mineralization area of the Duruja zone

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Abstract. The black shale complex, which is organic carbon-bearing and characterized by syngenetic, epigenetic sulphidation and intense metamorphism in the Duruja structural-formation zone of the southern slope of the Greater Caucasus, is the focus of geologists' attention due to the mineralization areas, which are rich in significant non-ferrous, noble, radioactive metals, rare and rare earth elements. In order to determine the prospects of noble, non-ferrous, radioactive metals, rare and rare earth elements of the black shale of the Aghlig-Filfilli mineralization area of the Duruja zone, it is necessary to determine the mineralogical composition of the ores of the area, to study the textural-structural characteristics and to clarify whether the rocks of the zone are a source of complex ore raw materials. The object of the work is the mineragraphic and petrographic study of the numerous samples collected from the Aghlig-Filfilli mineralization area during the field expedition before the PhD admission period, during my post-graduate studies and currently in order to solve various important mineralogical and petrographic problems. In order to investigate the textural-structural characteristics and mineral types of the ores of the Aghlig-Filfilli mineralization area, thin sections and polished sections made from the samples were studied with the help of «Carl Zeiss» and «Polam-312» microscopes in transmitted and reflected rays. The composition, surface area and morphological characteristics of the monomineral samples were studied by the electron microscope «SEM». Accurate diagnosis of some minerals was carried out based on the results of their X-ray diffractometric analysis. According to the results of the research, the ores of the Aghlig-Filfilli mineralization area were formed in 3 stages: syngenetic, epigenetic and remineralization as a result of strong metamorphism of syngenetic formations. At the same time, the origin of the ores of the Aghlıg-Filfilli mineralization area was studied. According to the results of the research, it was determined that epigenetic mineralization in contact with Aralig and Zangi deep faults in the north and south of the Aghlig-Filfilli area is of magmatogene origin ($\delta^{34}S$=1.6-4.6), and syngenetic mineralization in the black shales towards the center is of biogenic origin ($\delta^{34}S$= -18 – -22.6).

Key words: mineral, ore, pyrite, texture, structure

Текстурно-структурна характеристика та мінеральні типи руд району зруденіння Аглиг-Філфіллі зони Дуруджа

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Анотація. Чорносланцевий комплекс, що містить органічний вуглець і характеризується сингенетичною, епігенетичною сульфідизацією та інтенсивним метаморфізмом у структурно-формаційній зоні Дуруджа на південному схилі Великого Кавказу, є центром уваги геологів завдяки зонам мінералізації, які багаті на важливі кольорові, благородні, радіоактивні метали, рідкісні та рідкоземельні елементи. Для визначення перспектив благородних, кольорових, радіоактивних металів, рідкісних і рідкоземельних елементів чорних сланців району зруденіння Аглиг-Філфіллі зони Дуруджа необхідно визначити мінералогічний склад рудних зон, вивчити текстурно-структурні характеристики та з'ясувати, чи є породи зони джерелом комплексної мінеральної сировини. Об'єктом роботи є мінералографічне та петрографічне дослідження численних зразків, зібраних із зони мінералізації Аглиг-Філфіллі під час польової експедиції до вступу в аспірантуру, під час мого навчання та зараз для вирішення різноманітних важливих мінералогічних та петрографічних питань. З метою дослідження текстурно-структурних характеристик і типів мінералів руд району зруденіння Аглiг-Філфіллі досліджено шліфи і поліровані шліфи зразків за допомогою мікроскопів «Carl Zeiss» і «Polam-312», в прохідних і відбитих променях. Склад, площу поверхні та морфологічні характеристики зразків мономінералів досліджували на електронному мікроскопі «СЕМ». Точну діагностику деяких мінералів здійснено за результатами їх рентгенівського дифрактометричного аналізу. За результатами досліджень встановлено, що руди району зруденіння Аглиг-Філфіллі формувалися в 3 етапи: сингенетичний, епігенетичний та ремінералізації в результаті сильного метаморфізму сингенетичних утворень. Одночасно вивчалося походження руд району зруденіння Аглиг-Філфіллі. За результатами досліджень встановлено, що епігенетичне зруденіння в контакті з глибинними розломами Аралiг і Зангі на півночі та півдні площі Аглиг-Філфіллі має магматогенне походження (δ34S=1,6-4,6), а сингенетичне зруденіння в чорних сланцях в напрямку до центру має біогенне походження (δ34S= -18 – -22,6).

Ключові слова: мінерал, руда, пірит, текстура, структура.

Introduction

The initial data (till 1917) related to the geology of the Azerbaijani part of the southeastern slope of the Great Caucasus was obtained as a result of the researches by V.H. Abikh, V.B. Simonovich, and later (1917-1956) K.N. Paffenhols, F.V. Drobyshev, V.V. Weber, A.N. Solovkin, A.Ch. Sultanov, V.Y. Khain. Z.Isayev, R.A.Yolchuyev, H.A.Aliyev, I.Sh.Mammadov, A.H.Baloglanov, Y.F.Podgorny conducted search and mapping work on the southern slope of the Greater Caucasus in 1956-1984, and identified promising ore areas such as Aghlig-Filfilli and Galajik in the Duruja field. In recent years (2020-2022), a group of researchers of the Institute of Geology and Geophysics of ANAS (Kerimov, Gadirova, Shikhova, 2014) studied the distribution characteristics of the noble, rare, radioactive metals and rare earth elements in the Aalenian sediments of the Duruja zone, and carried out the geochemical assessment of the rare earth elements' accumulating potential of these rocks.

The Duruja structural-formation zone, where we conducted our research, is located on the southern slope of the Greater Caucasus between the Zagatala-Govdag synclinorium (in the north) and the Vandam anticlinorium (in the south) (Shikhalibeyli, 1956). It is surrounded by the Gaynar deep fault from the north and the Zangi deep fault from the south. It is observed at a distance of more than 200 km, with a thickness of 0.3-4.5 km in the area between Lekitchay-Sulutchay. It consists of sediments of Lower and Upper Aalenian terrigenous-flyschoid formation (Mammadov, et al., 2001).

The Duruja anticlinorium is a horst-type fault-uplift structure, secondary faults are observed along the axis directions of its internal folds. This feature creates favourable conditions for the formation of ore accumulations in the zone. It is interesting that the numerous and different scale microfolds and landslide folds which were formed within the Duruja zone do not develop outside the zone (in structures in the north or south) and are not observed. The discovered ore fields, accumulations and points are mainly located in the pan-Caucasian and transversally hydrothermally-altered zones around faults related to structural faults. At the same time, it should be emphasized that intensive volcanism, which is in contact with the Duruja zone along the northern part of the Vandam zone, played an important role in the formation of their mineral composition.

The reason for the prospect of valuable, non-ferrous, radioactive, rare and rare earth elements of the Duruja structure-formation zone is not only the structural feature, but also because the zone differs sharply from the structures located in the north and south according to the lithological composition of the rocks. So, the Zagatala-Govdag structural-formation zone consists of Upper Jurassic and Lower Cretaceous terrigenous-carbonate rocks in the north, and the Vandam structural-formation zone consists of the Upper Cretaceous volcanogenic-sedimentary formation layer in the south. Both zones are resistant to tectonic stress and they are non-plastic. As a result of tectonic processes, the rocks of the Duruja zone, which consist of plastic clay schist and alternating clay schist and sandstones, crumble and change relatively easily. This situation creates conditions for the formation of secondary ore accumulations during subsequent processes.

Another factor affecting ore accumulation in the black schist formation of the Duruja zone, which is rich in sulphide and organic matter, is characterized by the sorption of $C_{organic}$ and sulfur in rocks, ore-washing, regenerating, oxidizing, barrier processes, activation of electrochemical processes during mineralization and other features. The geostructural position of the Duruja zone, the mineralogical composition of its rocks, the richness of $C_{organic}$, the occurrence of hydrothermally altered zones around faults in the Pan-Caucasus and transversal directions increase our interest in this zone, and these factors give reason to say that the rocks of the zone are promising for noble, non-ferrous, radioactive metals, rare and rare earth elements.

The Aghlig-Filfilli mineralization area, located in the Duruja zone of the southern slope of the Greater Caucasus, is more interesting in terms of prospects. All factors, which affect the abovementioned ore accumulation, are concentrated in this mineralization area.

This area is structurally located in the middle course of the Khalkhalchay River in the eastern part of the Shinchay-Filfillichay uplift block, belonging to the Aralig fault of the Duruja zone (Azadaliyev, 2006; Shikhalibeyli, 1956; Mammadov, et al., 2001). Mineralization developed here mainly along the tectonically complex contact between flyschoid and aspide schist layers. The black shales of the Aghlig-Filfilli ore field are characterized by unique structural and textural peculiarities, degree of metamorphism and high $C_{organic}$-bearing (Baloglanov and Podgornyi, 1982).

The Aghlig-Filfilli mineralization area has a very complex geological structure, as it is characterized by numerous uplifts and declines that are tectonically very intense. Here the intensive fracture and local deformation systems, including the Aralig fault, Gaynar and Zangi deep faults form their apophyses and extend in different directions (Mammadov, et al., 2000; Giorgobiani and Zakaraia, 2010). The Aralig depth fault forms the northeastern branch of the Zangi deep fault, has a pan-Caucasian extension and lies to the northeast at an angle of 40-50 degrees. The thickness of the changed zone along the fault is 40-50 meters. Alterations such as intense limonitization, pyritization, quartzization, carbonation, occasional chloritization and sericitization are observed in the wall rocks composed of flyschoids and black aspide schists (Chernyshov and Vologdina, 2014).

Fig.1. Appearance of altered flyschoids and black aspide schists around fault zones in the Aghlig-Filfilli mineralization area (macroscopic)

The fault zone is often accompanied by a large thickness of abraded clays. The alteration zone with a thickness of 3-4 m is observed in the wall rocks along the subduction (southern bedding) of the mineralization zone, where the rocks were subjected to intensive quartzization, baritization and carbonation (Borodayev and Mozgova, 1974).

The main difference of the Aghlig-Filfilli field from other ore fields located in the Duruja zone is that concretions consisting of pyrite, pyrite-clay schists, siderite, pyrite-perlite and clay schists are widespread here. The sizes of these concretions, which are mainly conchoidal, reach 100x50x20 cm (Fig. 2).

Fig. 2. Conchoidal concretions in the Aghlig-Filfilli mineralization area

The Aghlig and Filfilli mineralization zones are characterized by quartz-calcite-sulfide mineralization in crumbled, crushed clay shales. They are observed up to a distance of 1.5-2.0 km with a thickness of 5-8 m in several sections on the earth's surface. Sulphidation makes up 4-6% of the total rock mass in these zones (Smirnov, 1967; Baloglanov and Podgornyi, 1982).

The sulphide ores of the Duruja zone are characterized by impregnated, veinlet, veinlet-impregnated, bended, spotted, massive, brecciated and kernel-shaped textures. Rough-type ores, which are much more dominant due to the intensity of occurrence, are often represented by unevenly distributed sulfide impregnations in the wall rocks. Relatively distributed pyrite impregnations are also observed in the rock mass in mineralized areas. Sparse and dense types of impregnation are distinguished depending on the distribution intensity of impregnations in the rocks (Fig. 3a).

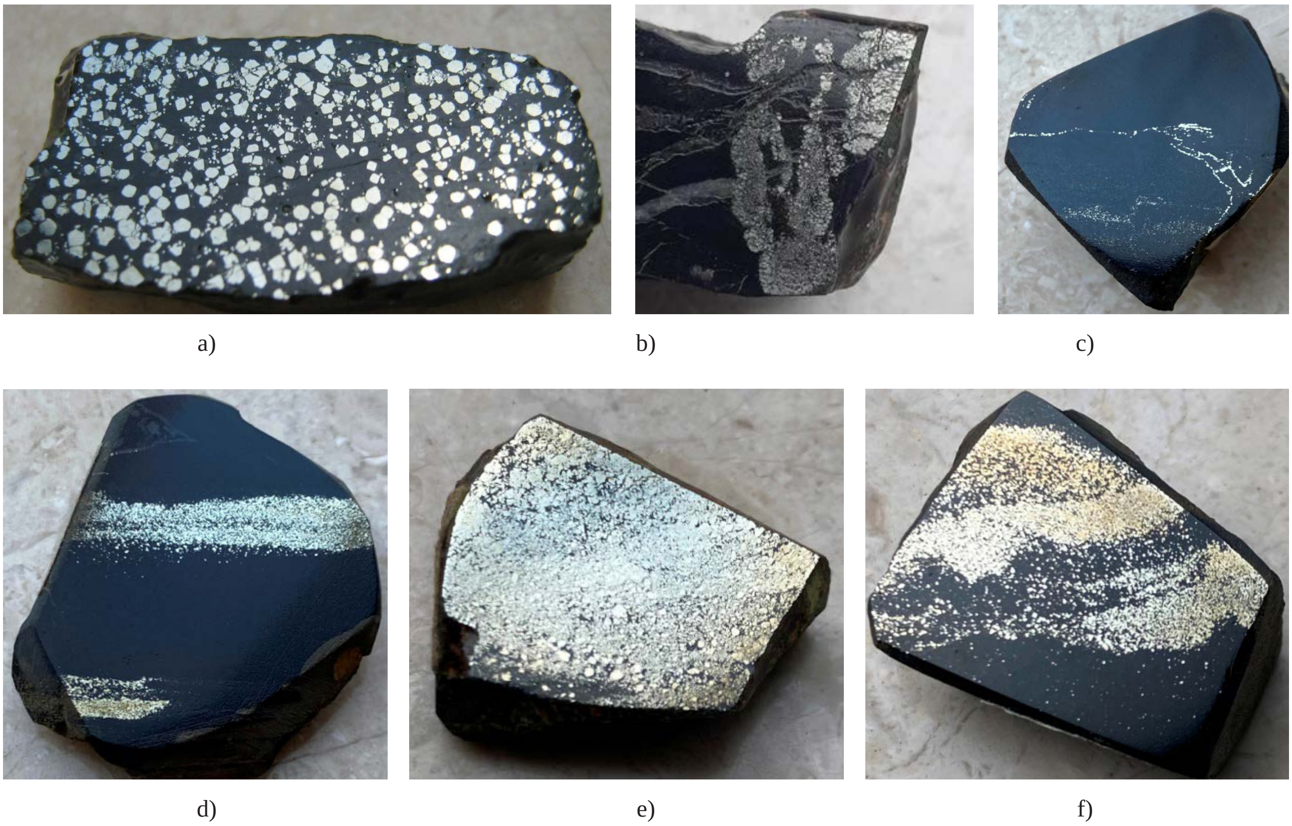


Fig. 3. Textures of ores in the Aghlig-Filfilli area (polished tuff):
a)impregnated; b) spotted; c) veinlet; d) layered; e) massive; f) kernel-shaped.

Ores with a *spotted* structure, which differ from impregnated ores according to the relatively large size and density of mineral aggregates, are also often observed (Fig. 3b).

Veinlet-type ores are observed in the distribution areas of epigenetic sulfides and are mainly represented by quartz and calcite accompanying these minerals. Veinlets of different directions, the thickness of which is usually several millimeters and sometimes reaches one centimeter, are often observed in a mutually intersecting, sometimes subparallel position (Fig. 3c).

The veinlets are usually oriented in different directions and are located in subparallel positions at separate intervals in *veinlet-impregnated*-type ores, which cut ore-hosting rocks and are always accompanied by sulphides. Sparsely and densely located occurrences are separated depending on the location character of the impregnation relative to each other in this type of texture, as in the case of impregnated ores.

Locally, the *banded* texture is manifested as alteration of sulphide bands and bands of non-ore minerals (Fig. 3d). Bands represented by ore minerals are often characterized by a lenticular shape. The densely located pyrite impregnations, which form the basis of sulfide bands and whose size is a few millimeters, are sometimes replaced by its massive aggregates in banded ores , the alternations of which are macroscopically visible. Occurrences in which the alternation of ore and non-ore bands is not clearly expressed are often observed.

Ores with a *massive* texture have a monotonous appearance and are characterized by the monotony of the pyrite aggregates that forms them. Such textured ores are often found in some parts of sulfide bands, which form a banded texture (Fig. 3e).

Formations with a *kernel-shaped* texture, which are related to the destruction of primary ores due to the influence of various factors and are mainly represented by secondary minerals, are observed in the mineralized zones (Fig. 3f).

The ores of our researched areas are characterized by a variety of structural types. The minerals that form the ores are represented by *hypidiomorphic-grained, allotriomorphic-grained, concentric-zonal, radial-radiant, globular, relict, druse, annular, limbate, cataclastic* and a number of other structured aggregates (Fig. 4).

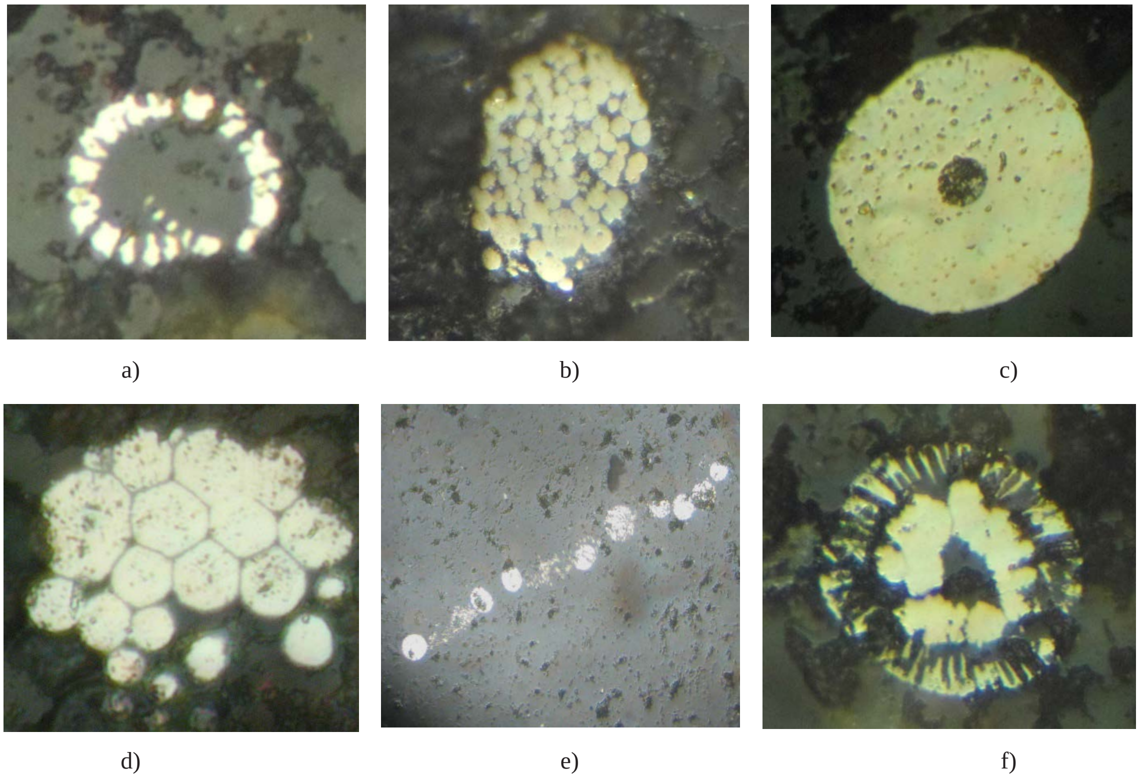


Fig.4. Structures of ores in the Aghlig-Filfilli area (microscopic (polished section)): a) annular polished section X 90; b) globular polished section X 120; c) spherical polished section X 90; d) framboidal polished section X 90; e) beaded polished section X 90; f) concentric-zonal polished section X 90

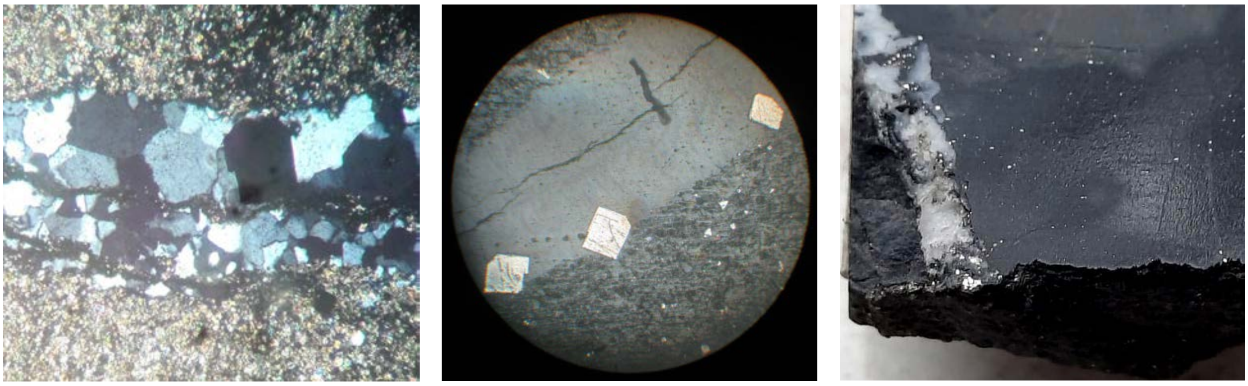
Occurring mainly as hypidiomorphic-grained formations and cubic crystals, pyrite also forms aggregates resembling globular, concentric-zonal, spherical, annular, feather and sheared, and sometimes honeycomb and cauliflower-like structures (Fig 4a,b,c,d,e,f). Sphalerite, chalcopyrite and galena aggregates are characterized by allotriomorphic-grained structure.

The most widespread mineral of ores that is characterized by a very simple mineral composition is *pyrite*, which is represented by three generations. Its early generation of syngenetic origin is observed as grains and aggregates of various shapes in ore-hosting rocks. It is often observed in the samples taken from fault zones that as the result of dynamometamorphism, pyrite is covered with numerous fractures.

Globular-grained pyrites and their aggregates-colonies are often observed in the rocks of the Aghlig-Filfilli area (Fig 4 b,c,d). These colonies differ according to the size and number of pyrite grains that make them up. Sometimes such aggregates of globular aggregates occur in cauliflower-like forms. Sometimes honeycomb-like occurrences of pyrite are observed in the ore-hosting rocks. Various ideas have been put forward in the geological literature about the formation of globular pyrite (Morgunova and Zhivaeva, 1987; Davidson, 1962). Some researchers emphasize the biogenic origin of such aggregates and attribute them to pyrite-generating microorganisms or metasomatic formations formed from dead microorganisms. The possibility of the formation of similar aggregates in an abiogenic way is also shown – they are formed from sulfide gels of hydrothermal origin (Towson, et al., 2015). It should be noted that the possibility of formation of globular pyrite aggregates of biogenic and abiogenic origin has been proven as a result of experimental studies.

Another generation of pyrite is observed in the ores as a result of strong metamorphism and recrystallization of its syngenetic formations (Sidorov and Tomson, 2000).

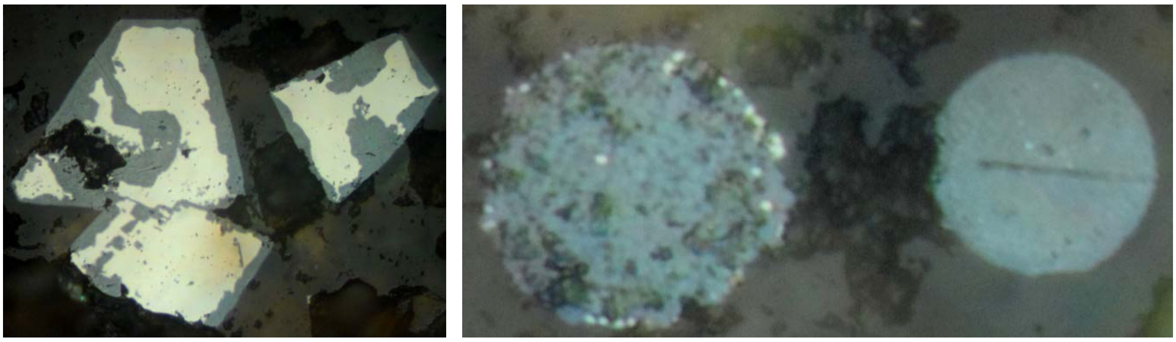
The third generation of the abovementioned mineral is of hydrothermal origin and occurs in the form of impregnation and veinlet-type formations (Fig. 5). Its cubic crystals and irregularly shaped aggregates are usually found in association with quartz.



a) b) c)

Fig. 5. Hydrothermal ore formation: a) thin section polished section X 90; b) polished section X 90; c) macro view.

Besides the well-chipped grains of pyrite, its dissected contoured formations are also observed. Pyrite grains and aggregates are corroded by chalcopyrite, sphalerite and galena (Towson, et al., 2015). Pyrite, which holds multiple impregnations of the wall rocks, is often cut by intersecting, sometimes subparallel veinlets of calcite Hypergene minerals of iron are often observed in the peripheral parts of pyrite grains in the form of rims surrounding them. Secondary iron minerals such as goethite, hydrogoethite, hydrohematite create pseudomorphoses on pyrite aggregates. If pyrite is not completely replaced by these hypergene minerals, its relics are preserved in these secondary minerals. Iron hydroxides, which often develop on the cubic grains of pyrite, preserve their appearance and occur in this form (Fig. 6).



a) b)

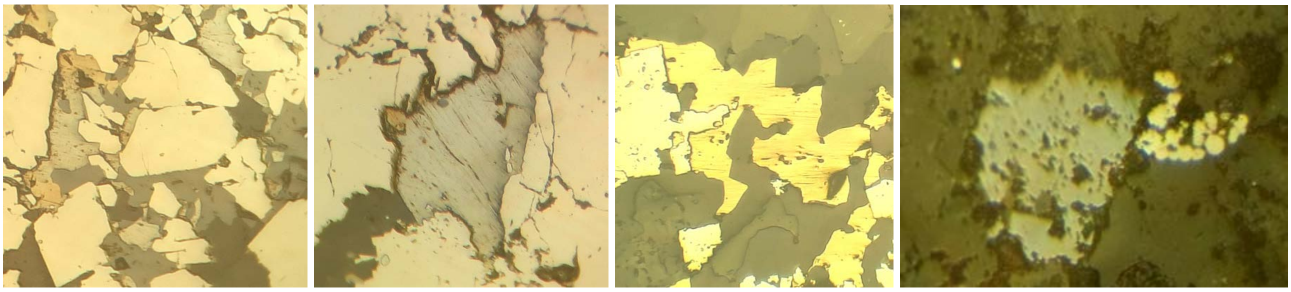
Fig. 6. Hypergenesis alteration of ores: a) crystalline pyrite polished section X 90; b) globular pyrite polished section X 90.

Sometimes framboidal aggregates of pyrite are also found in ores. If the globular aggregates of the mineral are observed individually and in the form of colonies in wall rocks, and often in certain layers, similar formations in ores are usually observed individually. The abovementioned aggregates of this generation of pyrite are assumed to be of abiogenic origin.

Although *sphalerite* dominates over other non-ferrous metal sulfides – galena and chalcopyrite, it is generally rare in ores. Sphalerite, which was found in some of the mineralization areas we studied, occurs as small irregularly shaped aggregates in a paragenetic association with galena and chalcopyrite. When etched by the vapours of chloroazotic acid under a microscope, it is revealed that its aggregates are characterized by an allotriomorphic structure (Mozgova, 1975). Sphalerite, the grains of which are measured in hundredths of a millimeter, is usually located among pyrite aggregates and corrodes them (Fig. 7a).

Irregular formations of *galena* are associated with sphalerite and chalcopyrite and corrode them by being located among pyrite aggregates. The aggregates of the mineral have an allotriomorphic grain structure, which grains are measured in tenths of millimeter (Fig. 7b**)**.

Chalcopyrite, which is far behind sphalerite and galena in terms of distribution, is observed in the interstices of pyrite grains in the form of small-sized fine-grained aggregates. Sometimes chalcopyrite, which occurs in small amounts in association with sphalerite and galena, forms veinlets that develop along fractures in aggregates of cataclized pyrite, which cement the disintegrated parts of the iron sulfide (Fig. 7c).



a) b) c) d)

Fig. 7. Polished sections x 90: a) pyrite-chalcopyrite; b) galena-pyrite; c) pyrite-galena-sphalerite-chalcopyrite; d) interaction of molybdenite-pyrite aggregates;

Arsenopyrite, which usually represented by rhombic idiomorphic grains and rod-like formations, is rarely found in association with pyrite in ores. Like pyrite, small impregnations of non-ore minerals are sometimes found in arsenopyrite.

According to the results of the geological explorations conducted by various geologists, as well as our research, a high concentration of molybdenum was determined in the mineralization areas of the Duruja zone (Yudovich and Ketris, 1991). For the first time, we discovered the rarely found scale-shaped formation of the specific mineral of the element – *molybdenite* in the area enriched with sulphide mineralization of this zone during the microscopic studies (Fig. 7d).

Marcasite, which is rarely observed in very small quantities in ores, occurs as irregularly shaped formations composed of small grains.

Pyrolusite, which is rare in ores, accompanies calcite and forms thin branch- and thorn-like aggregates – layered dendrites located along its veins.

Another manganese mineral, *psilomelan*, is represented by small-sized hidden crystalline aggregates and soot-like masses, which are rarely observed in clay shales. But, the presence of psilomelan in ores is related to the presence of xenoliths of rocks bearing this mineral.

The determination of iron oxides such as *magnetite* and *hematite* in ores, as well as titanium minerals such as *rutile* and *ilmenite* and small grains of other compounds is related to the observed presence of xenoliths of wall rocks containing these minerals in the sulfide mass.

Irregular shaped small grains of iron monosulfide – *pyrrhotite* are found very rarely in the composition of ores.

Conclusions

Investigating the potential of precious, non-ferrous metals, rare and rare earth elements accumulated in black shales is of great scientific and practical importance Sazonov, et al., 2011; Ganbarova, 2021; Sidorov and Tomson, 2000; Sazonov, et al., 2011; («Geochemistry, Mineralogy and Genetic Types of Rare Element Deposits», 1964). The characteristics of these rocks such as barrier, sorption, transport and reduction are important in the formation of anomalies of similar elements in black shales and in their role as a source in epigenetic processes.

The mineral composition and textural-structural characteristics of the ores were studied on the basis of mineralogical and mineragraphic studies of the samples taken during field expeditions from the Aghlig-Filfilli mineralization area, and the obtained results gave reason to say that there is mineralization related to various sources. Textures and structures specific to mineralization of different origins, which are mentioned in the article, confirm this. Structural aggregates of pyrite minerals such as «mineralized bacteria», «globulites», «spherulites», «framboids», «pyrite spheres» indicate their biogenic origin. It was determined during microscopic studies that this structure belongs to microorganisms or metasomatic formations formed on dead microorganisms (Fig.4-a, f). The study of pyrite-chalcopyrite-galena-sphalerite paragenetic mineral associations in the samples taken from hydrothermally altered around fault areas shows the occurrence of mineralization of epigenetic origin in the Aghlig-Filfilli area (Fig.7-a,b,c). At the same time, we observe the occurrence of the 3rd type of mineralization in the samples taken from remineralized areas related to metamorphism (Fig.3a).

So, according to the results of the research, it was determined that the ores of the Aghlig-Filfilli mineralization area were formed in 3 stages:

1. syngenetic;
2. epigenetic;
3. remineralization as a result of strong metamorphism of syngenetic formations.

For the first time, our study of the variation range of $\delta^{34}S$ in the sulphides of the Aghlig-Filfilli mineralization area is a clear proof of the accuracy of our research. So, globular pyrites have values in the range of $\delta^{34}S$= -18 – -22.6, and pyrite aggregates taken from hydrothermally altered around fault zones have values in the range of $\delta^{34}S$=+1.6 – +4.6.

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