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Rhenium-bearing molybdenite in the argillization zone of the Mikheevskoye molybdenum-copper-porphyry deposit (Southern Urals)

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Abstract

The purpose of the research. The work was carried out in order to determine the nature of rhenium content and typomorphic features of molybdenite from loose argillizite ores and to compare the obtained data with data on rocky ores of the Mikheevskoye deposit.

The relevance of the research. Molybdenite from porphyry copper deposits is one of the potential sources of rhenium for the domestic industry, which, taking into account the steadily growing interest of technologically advanced countries in superalloys (including those based on rhenium), determines the relevance of ongoing research.

Research methodology. The object of research is rhenium-bearing molybdenite isolated from loose argillizite metasomatites of the upper part of the deposit. The elemental composition of molybdenite was studied on a Cameca SX100 electron probe microanalyzer and compared with molybdenite previously isolated from rock ores.

Results and conclusions. The material composition of argillizites developed within the Mikheevskoye deposit has been refined, and the typomorphic parameters of molybdenite isolated from argillization zones have been established. Molybdenite grains are predominantly scaly and lamellar in shape, characterized by relatively stable concentrations of molybdenum and sulfur with significant variations in rhenium, the average value of which was 0.43%. The distribution of rhenium inside molybdenite grains usually has a point-spotted or concentric-zonal character. The results of the research showed the similarity of molybdenite from the zones of argillizites and rock ores both in the nature of the distribution of rhenium inside the grains and in the statistical parameters of the distribution of Re, Mo and S, which may indicate a slight influence of the temperature regime of the formation of ore-metasomatic associations on the main characteristics and the quality of the molybdenite deposit. Comparable characteristics have molybdenite of many other porphyry copper deposits in the world. A review of the factors influencing the distribution of rhenium in molybdenite of porphyry copper deposits has been made.

Keywords: rhenium, molybdenite, porphyry copper, argillizites, Mikheevskoe deposit.

Introduction

Rhenium is one of the most sought-after rare elements in modern industry, used in the aerospace industry, oil refining, precision instrumentation, medicine, electronics, etc. In the structure of world rhenium production (according to MetalResearch data as of 01/01/2015), Chile (53%), the USA (16%) and Poland (16%) occupy the leading positions with an annual production volume of about 70–80 tons; Russia's share here is almost imperceptible – less than 1%. In world practice, the main part of rhenium is extracted along the way from copper-molybdenum concentrates, and this metal practically does not form its own deposits. The exception is high-temperature fumaroles rich in rhenium of the Kudryavy volcano (Iturup Island), which were discovered as early as the 1990s; however, their industrial development is hindered by significant technological problems. Thus, in our country,

one of the main sources of rhenium (in addition to hydrogenous polyelement deposits of formation oxidation zones) at the moment are porphyry copper deposits. These include the Mikheevskoye deposit, the largest object of the porphyry copper family in the Urals.

The Mikheevskoye deposit is located in the Southern Urals within the Varna volcanogenic zone, its structure includes Late Devonian volcanogenic and volcanogenic-sedimentary formations of basaltic andesite and basaltic composition, with interlayers of terrigenous rocks and small bodies of serpentinites, as well as the granitoids of the Ulyanovsk (D_3) and Mikheevskoe (D_3-C_1) complexes [1]. The network of faults developed in the area of the deposit creates a linear-block structure with the development of north-south striking linear folds in the blocks, complicated by shear deformations.

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The age of mineralization by the Re–Os method is determined as Early Carboniferous (Tournaisian) [2]. The mineralization is confined to a band of dikes of quartz-diorite and diorite composition (mainly porphyritic), which extends between two diorite stocks of the Mikheevsky complex. The ore bodies of the deposit are stockwork-like zones of vein-disseminated copper-sulfide mineralization, which have a rather complex shape and are identified only according to sampling data. According to data from open sources, the identified resources of the deposit amount to 373.5 million tons of ore with an average grade of copper – 0.38%, gold – 0.1 ppm.

Results and their application

Within the deposit, metasomatites are represented by the following main types (according to [3] with additions by the authors):

1. Actinolite-epidote with magnetite, less often garnet (andradite);
2. Biotite-muscovite-K-feldspar with bornite and chalcopryrite;
3. Chlorite-epidote-sericite (propylites) with pyrite or pyrite and chalcopryrite;
4. Quartz-sericite with chalcopryrite and molybdenite;
5. Quartz-illite/hydromica-kaolinite (argillizite) with pyrite, chalcopryrite, bornite and molybdenite.

Argillizitic metasomatites were observed in the active quarry of the deposit in the form of steeply dipping zones with an apparent thickness of up to 40–50 meters or a series of closely spaced narrower zones. At the same time, weak manifestations of argillization superimposed on other types of metasomatites were often noted. In the upper part of the section, argillizites have an uneven areal distribution, which made it possible to distinguish them into a special technological type of ores – loose sulfide ores. The thickness of these formations varies from 1.6 to 46 meters, with an average of 14 meters; the boundary of the bark developed along them is not distinct. Zones of argillizite metasomatism are often associated with quartz-diorite porphyry dikes and their exocontacts. The argillizites typical for the deposit are whitish, light gray, greenish or yellowish formations, completely preserving the structural and textural features of the original rocks. Among the “loose” argillizites, there are separate thin quartz vein zones and areas of quartzization. Some mineralogical characteristics of these formations are given by the authors in [4]. According to the first results of ICP-MS analyzes, the geochemical specialization of argillizites of the Mikheevskoye deposit is determined by slight increases (by 20–40%) in the contents of copper, titanium, chromium, vanadium, strontium, and molybdenum with a more noticeable decrease (up to 50%) in the contents of arsenic, rubidium, and lead, tungsten, barium, cobalt, selenium and bismuth relative to the rocks of the substrate; the majority of elements in the process of argillization remain indifferent. Due to the small volume of statistical samples, no zonality or systematic distribution of elements in these formations has yet been established. Analysis of the distribution of elements in four samples of argillizites taken from two such zones within the horizons exposed by the quarry (southern edge, horizon +235 m and western edge, horizon +240 m), showed insignificant variations in the values of the main elements, with the exception of a number of chalcophile elements (copper, selenium, silver, gold, zinc, bismuth).

The main and often the only source of rhenium in porphyry copper deposits is molybdenite, a mineral that is chemically stable, close to the stoichiometric formula MoS_2 , and has minor impurities of Fe, Re, Se, Sn, W, Ti, and Bi. Molybdenite within the primary ores here has a discrete distribution and is visually often observed in association with quartz, pyrite and chalcopryrite, forming quartz-molybdenite, quartz-chalcopryrite-pyrite-molybdenite, quartz-carbonate-molybdenite veinlets of insignificant thickness (the first cm); molybdenite here has monomineral segregations (aggregates) up to 2–3 cm in diameter, usually represented by small-thin flakes, as well as “greases”.

Molybdenite within the established zones of argillization is isolated as rare small (fractions of millimeters) inclusions of lamellar, packet-like, scaly grains (fig. 1), as well as in intergrowths with pyrite and fine-grained quartz.

The study of the elemental composition was carried out on a Cameca SX100 electron probe microanalyzer at an accelerating voltage of 15 kV and a current of 20–60 nA (analyst D. A. Zamyatin, IGG, UB Russian Academy of Sciences, CUC “Geoanalyst”). When performing calibrations, standard samples were used: metallic rhenium, molybdenum, osmium, selenium, pyrite, sphalerite, wollastonite. The Mo content was measured using the L_β line; to determine S, the procedure for taking into account the superposition of the $\text{Mo}L_\alpha$ line on SK_α was used. The errors in determining the elements Mo and S are below 2.57 and 1.02 wt. %, respectively. At a number of points, the intensity of the $\text{Mo}L_\beta$ line was measured simultaneously on two spectrometers with PET crystal analyzers to reduce the error to 1 wt. %. The $\text{Re}M_\alpha$ (TAP crystal) and $\text{Re}L_\alpha$ (LLIF and LIF crystal) analytical lines were used to determine the Re concentration in molybdenite, similarly to [5]. The error and limit of detection of the Re content for the M_α line is below 0.09 wt. %, for the L_α line, 0.21 wt. %. The Re concentrations in molybdenites are determined from the M_α line; at a number of points, the concentration was checked and coincidence within the error with the measurement along the line L_α was shown. Inside one grain with wide variations in the Re concentration within the limits of 0.22–0.93 wt. % (sample Mikh-11-1), maps of the intensity distribution of the characteristic X-ray lines $\text{Re}L_\alpha$, $\text{Re}M_\alpha$, $\text{Mo}L_\beta$ and SK_α were obtained (fig. 2). Card registration time – 3 hours.

In total, 8 grains of molybdenite were analyzed, 74 representative measurements of the composition of molybdenite were carried out with a sum of more than 96%. The results of microprobe analysis showed that the distribution of sulfur and molybdenum in the composition of the grains is relatively uniform – the average values for the sample were 38.3 and 59.7 wt.%, respectively, with the distribution variation coefficients of both elements $V = 0.02$. Rhenium in the studied molybdenite is distributed very unevenly – with an average value of 0.43%, the content of this element varies within 0.1 – 1.72%, the coefficient of variation is $V = 0.79$, which is comparable with the values obtained from the results of the study of primary molybdenite ores [6].

In addition, regularities of rhenium distribution in molybdenite grains in primary ores ($n = 81$) and studied argillizites ($n = 74$) were compared. It is found that the distribution of Re is lognormal in both samples; comparison of independent samples according to the non-parametric Mann–Whitney test allows us to accept the hypothesis of the homogeneity of the

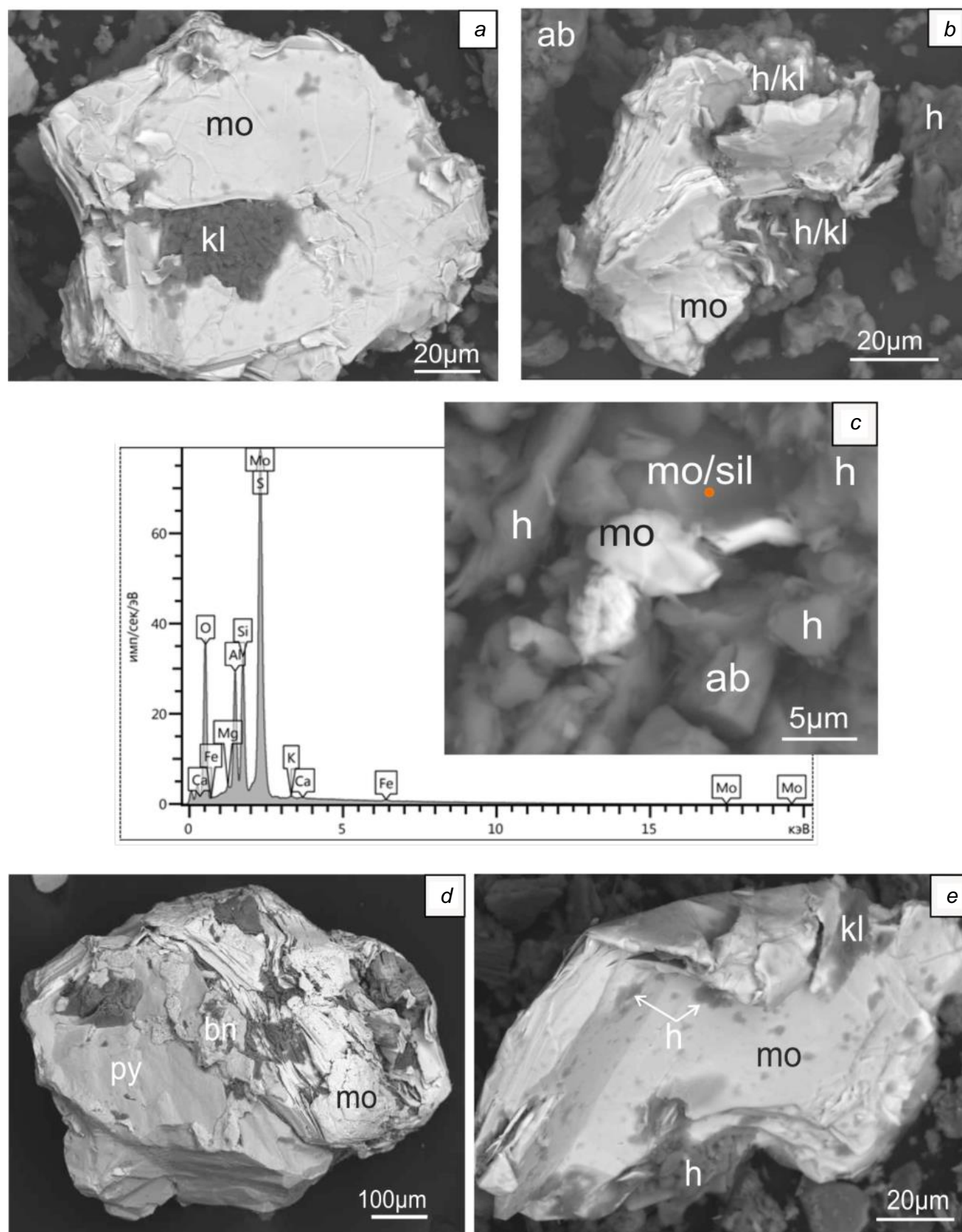


Figure 1. BSE images of molybdenite grains from loose argillizites of the Mikheevskoe deposit: a–c – fine and thin grains of molybdenite in association with hydromica, kaolinite, and albite (sample Mikh-13/18); d, e – larger segregations of molybdenite (sample Mikh-11-1), including intergrowths with pyrite and bornite (d); designation of minerals: ab – albite, bn – bornite, h – hydromica/illite, kl – kaolinite, mo – molybdenite, py – pyrite, sil – silicate film

Рисунок 1. Результаты СЭМ зерен молибденита, выделенного из аргиллизитов Михеевского месторождения: а–в – мелкие зерна от 50 до 150 мкм (проба Мих-13/18); г, д – более крупные до 1,0 мм и более (проба Мих-11-1)

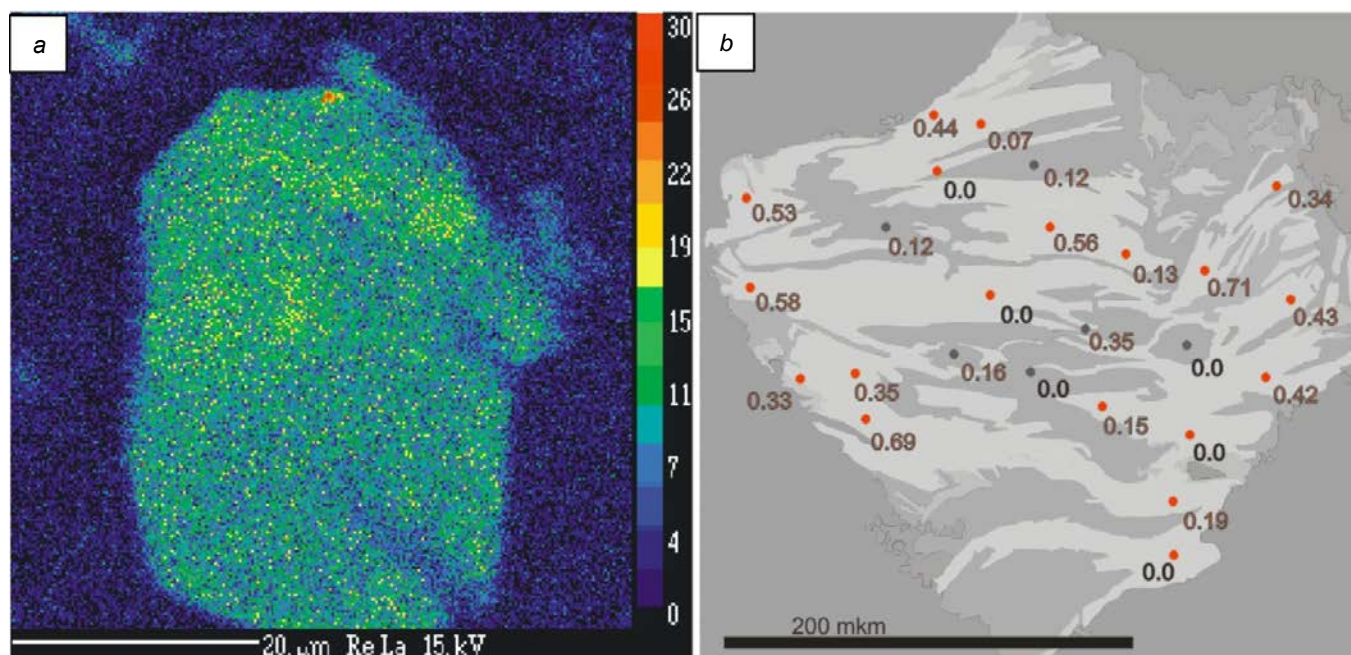


Figure 2. SEM data, microprobe scanning, and quantitative determination of rhenium: *a* – a map of the intensity distribution of the ReL_{α} line in a tabular molybdenite grain (sample Mikh-11-1); *b* – BSE-photo of a deformed scaly grain of molybdenite with points for determining the content of rhenium, wt.% (sample Mikh-14-1)

Рисунок 2. Данные СЭМ, микрозондового сканирования и количественного определения Re: *a* – деформированное чешуйчатое зерно молибденита в лучах BSE с точками определения содержаний, вес.% (проба Мих-14-1); *b* – карта распределения интенсивности линии ReL_{α} в зерне молибденита таблитчатой формы (проба Мих-11-1)

means ($U = 2174.5$; $p \leq 0.05$), i. e., about the absence of statistically significant differences in the distribution of rhenium in molybdenite of primary rock ores and metasomatites of argillization zones. Consequently, in the process of low-temperature metasomatism, the redistribution (input/removal) of rhenium in molybdenite practically does not occur.

As in primary ores, the distribution of rhenium in the molybdenite grains of the argillization zones has a point-spotted or concentric-zonal distribution with high variations of this component within the grains (fig. 2).

According to modern concepts [7, 8], this distribution can be due to several factors: 1) the ratio of 2H and 3R molybdenite polytypes formed under different conditions, 2) the presence of microdefects in the crystal lattice of this mineral, and 3) the possible presence of rhenite as a microimpurity. The question of the influence of polytypes on the ability to concentrate rhenium is currently open. There are a number of studies both in favor of the greater concentration possibilities of both 2H and 3R polytypes, but the factor of conditions for the formation of the mineral phase is more likely to be of greater importance [9, 10]. The uneven distribution of rhenium in the molybdenite of porphyry copper deposits in the world is also well known. So, at the Murat-dere deposit (Turkey), a high degree of heterogeneity in the distribution of rhenium in various generations of molybdenite was recorded – from 0.03 to 4% within the grain, which causes some zoning of the distribution, generally parallel to layering [11]. According to [12], high Re concentrations may be due to the isovalent substitution of Re for Mo in molybdenite and do not depend on the polytype; the possible presence of tellurides is also indicated here as an indication of the presence of rhenite in the ore system. It was also shown in [13] that the main factors affecting the distribution of rhenium in molybdenite of porphyry copper objects

are the composition of magmatics, the concentration of rhenium in the ore-forming fluid, and variations in the physico-chemical conditions of crystallization. Some regularities in the distribution of rhenium concentrations in molybdenite from a number of porphyry copper deposits in the Urals, related to the composition of the intrusions involved in the formation of these objects, the age of the deposits, and the geotectonic setting of their formation were also revealed [14]. The average content of Re in molybdenite of the argillization zone of the Mikheevskoye deposit is comparable with the concentrations of this element in molybdenites of porphyry copper deposits around the world (690 ppm) [11], reaching contents of ≥ 3000 ppm [15]. High concentrations of rhenium in molybdenite may also indicate a predominantly mantle source [16].

Conclusions

The results of the research allow us to make a preliminary conclusion that the molybdenite isolated from the argillizite of the deposit corresponds in its main characteristics to molybdenite from the rock ores of the deposit. It is characterized by slight variations in sulfur and molybdenum with wider variations of rhenium, the distribution of rhenium inside the grains is predominantly concentric-zonal and point-spotted and is comparable in statistical characteristics to molybdenite from rock ores.

Despite the rich diversity of low-temperature mineral phases in the argillizites of the Mikheevskoye deposit [3, 4], the expected changes in the composition and structural-geochemical features of rhenium-bearing molybdenite are not recorded here. The results of the work can be used in solving technological problems for the extraction of rhenium.

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Рениеносный молибденит зоны аргиллизации Михеевского молибден-медно-порфирикового месторождения (Южный Урал)

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Аннотация

Цель исследования. Работа выполнялась с целью определения характера рениеносности и типоморфных особенностей молибденита из рыхлых аргиллизитовых руд и сопоставления полученных данных с данными по скальным рудам Михеевского месторождения.

Актуальность. Молибденит медно-порфириковых месторождений – один из потенциальных источников рения для отечественной промышленности, что с учетом стабильно растущего интереса технологически развитых стран к суперсплавам (в том числе и на основе рения) определяет актуальность проводимых исследований.

Методы исследований. Объект исследований – рениеносный молибденит, выделенный из рыхлых аргиллизитовых метасоматитов верхней части месторождения. Проведены исследование элементного состава молибденита на электронно-зондовом микроанализаторе Cameca SX100 и сопоставление его с молибденитом, выделенным ранее из скальных руд.

Результаты и выводы. Уточнен вещественный состав аргиллизитов, развитых в пределах Михеевского месторождения, установлены типоморфные параметры молибденита, выделенного из зон аргиллизации. Зерна молибденита имеют преимущественно чешуйчатую и пластинчатую форму, характеризуются относительно стабильными концентрациями молибдена и серы при значительных вариациях рения, среднее значение которого составило 0,43 %. Распределение рения внутри зерен молибденита обычно носит точечно-пятнистый либо концентрически-зональный характер. Результаты исследований показали сходство молибденита из зон аргиллизитов и скальных руд как по характеру распределения рения внутри зерен, так и по статистическим параметрам распределения Re, Mo и S, что может свидетельствовать о незначительном влиянии температурного режима формирования рудно-метасоматических ассоциаций на основные характеристики и качество молибденита месторождения. Сопоставимые характеристики имеет молибденит многих других медно-порфириковых месторождений мира. Сделан обзор факторов, влияющих на распределение рения в молибдените медно-порфириковых месторождений.

Ключевые слова: рений, молибденит, медно-порфириковый, аргиллизиты, Михеевское месторождение.

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