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MAGNETITE- AND ILMENITE-SERIES OF GRANITIC ROCKS AND THEIR POTENTIAL NIOBIUM AND TANTALUM MINERALIZATION WITHIN THE UKRAINIAN SHIELD

(Reviewed by the editorial board member O. Mytrohin)

The paper discusses differentiating between different types of granites that occur within the territory of the Ukrainian Shied. The classification in the article partly follows the principles formulated in the early taxonomies of granites, among which are the classifications proposed by Ishihara [11], Chappel and White [4], and a lower classification proposed by Tomurko and Shcherbakov [20]. The latter made an attempt to single out two contrasting granite varieties – apobasitic and apopelitic granites – that are commonly found within the Ukrainian Shield.

Grouping granites into different genetic types runs into difficulties due to heterogeneous origin of these formations, which results from merging parental mantle with crustal sources. Nevertheless, such classification might be used in making an accurate prediction about metallogenic specialization of different genetic types of granites. The redox state of granites formation is a major factor that can largely affect possible metal concentration in them. Based on this parameter granites can be classified into ilmenite and magnetite series of granites, which differ in geological environment of their formation [6, 11] and mineralization associated with them [1].

Within the territory of the Ukrainian Shield there can be distinguished entire provinces with distinct distribution of mostly ilmenite or magnetite series of granites that reveal different ways of ore mineralization associated with them. Columbite-tantalites are common ore minerals found within these series of granites, with niobium and tantalum being characterised by close lithofillic affinity to these formations. Within the Ukrainian Shield, columbite-tantalites were found in granites and associated rare-metal pegmatites, in some apogranitic metasomatites (greisens, quartzites, perthosites), and also in alkaline rocks of various origin. It is determined the basic features of columbite-tantalites, with Ta/Nb value, Fe/Mn ratio and presence of admixture elements to have close genetic relation to different granite series. The features of mineralization, defined to be typomorphic of ilmenite and magnetite series of granites, might be used to improve survey and exploration carried out in the Ukrainian Shield.

Introduction. A possibility to use accessory oxides (magnetite and ilmenite) was first outlined in the paper published by Ishihara in 1977 [11], in which they were determined to be mineralogical criteria for metallogenic specialisation of granitic rocks. This paper discusses distinguishing between magnetite and ilmenite series of granitic rocks that might be related, to some extent, to I- and S- types of granites according to genetic classification of Chappel and White [4], or apobasic and apopelitic varieties of granites according to lower mineralogical-geochemical classification of Tomurko and Shcherbakov [20] eleborated for different granitic complexes of the Ukrainian Shield. It is obvious that classifying granites into different genetic types, such as I (igneous), S (sedimentary), A (alkaline, anhydrous, anorogenic) ones, runs into difficulties Granitic magmas can be formed from different heterogeneous parental source (protolith), crustal or mantle one predominantly, or it can represent mixture melts of crustal and mantle origin [17]. At the same time such subdivision of granites into genetic series enables predicting the likely metallogenic specialization of granitic magmas. Among the major parameters which can essentially affect general metallogenic content of granitic complexes are the following: 1) type of granitic protolith (S, I, A – types); 2) a trend of evolution; 3) degree of fractionation; 4) redox state [1].

Background. The classifications mentioned above are interesting not only because they reveal features of metallogenic specialisation in the complexes of the Phanerozoic orogenic belts [1, 4], but they might be also used to interpret the likelihood of ore mineralization, that occurred in different felsic association and granitic complexes found in other geodynamic settings even in Precambrian structures, among which there is the Ukrainian Shield.

Within the Ukrainian Shield there might be outlined distinct regions which are characterised by the distribution of mostly ilmenite or magnetite series of granites. It is important to note, that magnetite series of granites may include some syngenetic ilmenite, while ilmenite series of granites are commonly characterized by the presence of ferric iron (Fe^{+2}) in the structure of iron arsenide (loellingite $FeAs_2$).

The presence of magnetite or ilmenite in granites can predominantly be indicative of redox state of the environment, in which these granite complexes form. According to the classification eleborated by Frost [6] there are three main conditions that can maintain magnetite in granitic rocks: 1) reduction resulting from the combustion of carbon on metasedimentary rocks melting; 2) magnetite consumption by reacting with the Fe-Mg silicates in reduced rocks; 3) magnetite consumption to make sodic pyroxenes and amphiboles in peralkaline rocks. It allows estimating potential ore mineralization that might be found in granites as well as it can help to define mineralogical-geochemical features of many ore elements which are sensitive to redox state, especially those of lithophilic affinity, with niobium and tantalum being most useful among them.

Both niobium and tantalum show close association with felsic rocks and tendency for accumulation in the latest phases of fractionated intrusive complexes. In the late

stages of granite evolution and superimposition of metasomatic alterations a wide range of tantalum-niobium minerals can be formed. Because of their close chemical affinity niobium and tantalum might be found in the same minerals, where these elements isomorphically replace each other. Occurring dessiminated in rockforming minerals, Nb^{5+} and Ta^{5+} can commonly be concentrated in some accessory phases during the stages of superimposed metasomatic alterations; the latter being characterized by activity of volatile elements. Columbite $(Fe,Mn)Nb_2O_6$, pyrochlore $(Na,Ca)_2Nb_2O_6(OH,F)$, ilmenorutile $(Ti,Nb,Fe)_3O_6$, fergusonite $(Y,Th)(Nb,Ta)O_4$. are among the minerals that can concentrate tantalum and niobium. These minerals can even concentrate up to 95 % of the total content of tantalum and niobium. Concentration of Ti^{4+} in the mineralogical environment being high, there can be observed processes of dissemination of Nb^{5+} and Ta^{5+} in titanium-bearing minerals. Alkaline associations are characterized by constant prevalence of niobium over tantalum.

Ta and Nb are also widely used to interpret different kinds of trace element in the environment indicative of granite rocks formation [16], to which discriminant diagrammes plotted in Nb-Y, Ta-Y, Rb-(Y+Ta) an Rb-(Y+Nb) coordinates are common. In spite of the fact, that these descriminators are commonly used for Phanerozoic granitic complexes and felsic associations, for which plate tectonics paradigm is widely accepted, these discriminative diagrams for Precambrian complexes are also mentioned in a range of recent publications.

Our primary aim in this paper is to discuss a possibility of distinguishing between different types of granites and other felsic rocks that occur within the Ukrainian Shied. The Ukrainian Shield is considered to be a part of Sarmatia segment of East European Craton that consists of both Archean and Proterozoic domains. Some researchers believe these domains to be fragments of a single craton [12, 18], for basic facts about plate tectonics in the Precambrian time remain limited [10]. Those who keep up a plate tectonics idea consider the Ukrainian Shield to be a Proterozoic collage of discrete terrains [5, 7]. Among the Archean terrains there single out the Azov one, which is situated in the East, the Dniester-Bug and Rosinsk-Tikich terrains, highly reworked in the Palaeoproterozoic, and the Middle Dnieper granite-greenstone terrain, never affected by Proterozoic processes. Palaeoproterozoic terrains are represented by the Kirovohrad domain (Ingul), which is located in the central part of the Ukrainian Shield and the Volyn domain, which comprises the westernmost part of the Shield (Figure 1).

Niobium-tantalum mineralization was found in different domains of the Ukrainian Shield. Among Precambrian rock series with associated mineralization there are distinguished rare-metal pegmatites rich in Li, Rb, Cs, Ta, Nb, Sn, Be, some apogranitic metasomatites (greisens, quartzites, perthosites), and also various alkaline rocks. Compositional affinity of Ta-Nb minerals to basic compositional features of rock series that host this mineralization is believed to be a common feature of Ta-Nb minerals found in different rock associations.

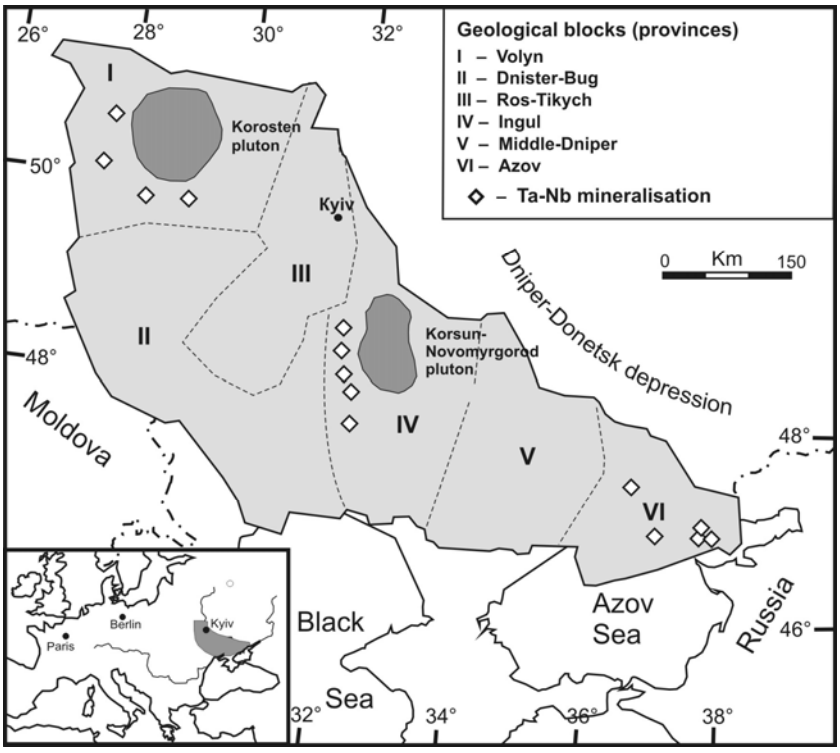

Figure 1. Schematic distribution of Ta-Nb mineralization within the Ukrainian Shield

Rare-metal pegmatites. Rare-metal pegmatites with Li, Rb, Cs and Ta-Nb mineralization were found in the Proterozoic (Volyn, Ingul) and the Archean (Azov) terrains. These rare-element pegmatites are classified as LCT type pegmatite [3]. Among the Precambrian rocks that host rare pegmatites with Ta-Nb mineralization there were distinguished metaluminous granite complexes (S-type), which are characterized by high potassic and alumina content, lack of fluorine, predominant ilmenite, as well as carbonaceous matter constancy. The age of RE pegmatites is unidentifiable because of the lack of accessory minerals commonly used as geochronometers (zircon, monacite). The age of granitic rocks that host rare-metal pegmatites is likely to be about 2.0 Ga for the Volyn and Ingul terrains and the same of 2.0 Ga (with some scientists scaling up to even the Archen age) for pegmatites of the Azov terrain.

Apogranitic metasomatites. The Nb-Ta-bearing apogranitic metasomatites of Proterozoic age were found in eastern Volyn and central Ingul terrains. They are characterized by the occurrence of large AMCG (anorthosite-

mangerite-charnockite-granite) complexes, such as Korosten (in Volyn) and Korsun-Novomyrhorod (in Ingul) ones, with which morion, topaz- and beryl-bearing chamber pegmatites are associated, and which are classified as NYF-type pegmatites [3]. The Volyn terrain also includes metaluminuos felsic associations of Osnitsk complex (I-type) approximately dated 2.0 Ga and Perga peralkaline associations (A-type) of 1750 Ga, the latter tracing Nb-Ta mineralization. Osnitsk complex, being part of Osnitsk-Mikashevichi Orogenic Belt bordering Sarmatia from Fennoscandia [2], is thought to have predated the formation of Korosten AMCG complex. Perga complex is supposed to have appeared after the formation of Korosten NYF-type pegmatites.

Alkaline rocks. There were found about 50 massifs and occurrences of alkaline rocks and carbonatites in the central (Ingul) and eastern (Azov) terrains of the Ukrainian Shield. The only alkaline formations evident in Kirovohrad terrain turned out to be alkaline rocks and kimberlites of the Proterozoic age (2.1-1.8 Ga). The Azov terrain prevalently includes occurrences of the Proterozoic and Phanerozoic (Devonian) ages. The two main alkaline associations found in the Azov terraine were defined to be alkaline-ultrabasic (carbonatitic) and gabbro-syenitic complexes. Alkaline rocks of the eastern terrain (Azov) and the western terrain (Ingul) reveal completely different geochemical properties. Those of the Azov terrain show features of alkaline-ultrabasic associations (high contents of incompatible rare elements such as Nb, REE, Zr, Y, Sr), whereas those in the Kirovohrad terrain are characterized by low contents of Nb and Zr, and REE. This fact is interpreted to have resulted from different geodynamic settings of their origin – extensional and collisional ones, respectively. Various mineral deposits of apatite, niobium, REE, yttrium and zirconium proved to be associated with the alkaline rocks and carbonatites of the Ukrainian Shield. Most Nb and Ta of Novopoltavka economic carbonatites (Chernihivka alkaline-ultrabasic massif of the Azov terrain) were found to be concentrated in fergusonite and hatchettolite.

Application. By using optical microscopy, common chemical analysis, spectral and XRF methods there was determined chemical and mineralogical composition of granitoids and other felsic varieties of rocks. Furthermore, microprobe analysis identified some features of rock forming, ore and accessory minerals, with probes to be JXA – 5, JCXA – 733 (Institute of Geochemistry, Mineralogy and Ore Formation, NAS of Ukraine), JCXA – 8200 (Scientific and Technical Centre, NAS of Ukraine), Cameca SX – 100 (Geological Institute, Slovakian Academy of Sciences).

The study of Ta-Nb mineralization distribution within the Ukrainian Shield shows that despite inherent geochemical affinity between tantalum and niobium, these elements behave differently throughout the processes of granite complex formation, which is especially indicative of Proterozoic granite complexes. It should be emphasized that granites themselves do not commonly show high concentrations of ore minerals. Their potential economic mineralization is mostly associated with the products of late to postmagmatic alteration stages of granite system. It is these stages during which volatile components become more active followed by hydrothermal stage, during which most of the ore elements are thought to be accumulated [19]. But at the same time different pegmatites and metasomatites (greizens, secondary quartzites, perthosites) show distinctly close spatial and genetic relation to certain Proterozoic complex found in the Ukrainian Shield, which may be classified into different ilmenite or magnetite series of granites.

Within the territory of the Ukrainian Shield there can be outlined two contrasting provinces – Volyn terrain in the eastern part and Ingul terrain in the central part of the Ukrainian Shield, within which one can spatially single out fields of rare-metal granites and pegmatites that reveal typical geochemical specialization on Li, Rb, Cs, Ta, Nb, Sn, Be. These areas are characterized by mostly Proterozoic granite complexes, which distribution can be based on the features of niobium and tantalum minerals formed in contrasting redox conditions [8, 9].

In the Volyn terrain, the westernmost part of the Ukrainian Shield, the major concentrations of Nb and Ta mineralization are confined to metasomatically altered alkaline granites of Perga (A-type) and Osnitsk (I-type) complexes. Felsic rocks of these complexes are characterized by high magnetite content and that's why they may be related to magnetite series granites. Regionally, both Perga and Osnitsk granite complexes of magnetite series are related to the Osnitsk-Mikashevichi Orogenic Belt, which marks geological border between Sarmatia and Fennoscandia, the northern margin of the Ukrainian Shield. Both terrains are interpreted to have had different geological histories before 2.0 Ga and to have merged in post 2.0 Ga period of geological history [2]. It is exactly the period during which Pegra and Osnitsk granite complexes of magnetite series formed. Both granite complexes show a wide distribution of superimposed processes of metasomatic alterations (greisenization, albitization and silicification) and associated mineralization, which is confined to these metasomatites. Magnetite series granites of Perga and Osnitsk complexes are characterized by high content of fluorine and wide distribution of magnetite, with Li micas (lithionite, zynvaldite), columbites $(Fe,Mn)(Nb,Ta)_2O_6$, pyrochlore $NaCaNb_2O_6F$, Ta-cassiterite (Ta crystals in cassiterite) commonly distinguished among ore mineral associations. It was identified Ta/Nb ratio ranging from 1/10 to 1/15 and values of Fe/Mn ratio reaching 10/1, as well as complete absence of admixture elements to be the typomorphic features of columbites found in these ores.

Ingul terrain, occupying the central part of the Ukrainian Shield, is also characterized by a wide distribution of rare-metal granites, which occurrences can be spatially related to the central Kirovohrad (Ingul) orogenic belt [13]. Most granite complexes that contain Ta-Nb mineralization are related to ilmenite series granite of S-type (Yaroshevka, Polohivka, Lipniazhka complexes) with geochemical specialization on Li, Rb and Cs. These granites are characterized by high content of potassium, oversaturation in alumina and practically complete absence of fluorine. Graphite and anthraxolite that are commonly found in these ilmenite series of granites can probably testify for predominant reduction conditions of their formation.

Polohivka, Mostove, Lipniazka, North-Stankuvatka and Nadiya rare-metal deposits were determined to contain tantalum-niobium mineralization with short range of Ta/Nb values and low values of iron (FeO/MnO = 2.80-6.56). Columbites-tantalites found here are marked by heterogeneous inner structure, with different mineralogical phases to be found in a single mineral grain. These mineralogical phases are characterized by high content (weight %) of Ta_2O_5 ranging from 9.80 to 71.0, and Nb_2O_5 varying from 10.6 to 70.1 [8, 9, 13, 14]. At the same time columbite-tantalites indicate to a high content of admixure elements (weight %) with TiO_2 reaching 5.88; WO_3 – up to 3.70; SnO_2 – up to 9.20; Sc_2O_3 – up to 5.40. Some associated ore minerals, namely ilmenorutile $(Ti,Nb,Fe)_3O_6$, tapiolite $Fe(Ta,Nb)_2O_6$, microlite $(Na,Ca)_2(Ta,Nb)_2O_6(O,OH,F)$, Ta-cassiterite, nigerite are also commonly found here.

Conclusions. The basic typomorphic features of columbite-tantalite minerals, specified by Ta/Nb values, Fe/Mn ratio and admixture elements are concluded to de-

pend on genetic granite types represented by magnetite and ilmenite series that are found in the Proterozoic complexes of the Ukrainian Shield. Magnetite series granites, which are considered to be formed in mostly oxidized environment (raised fO_2 values), are characterized by predominant niobium mineralization. At the same time ilmenite series granites, which are supposed to be formed in predominantly reducing environment, show rare-metal mineralization that is characterized by generally equal proportion of tantalum and niobium contents. This conclusion facilitates methods to optimize prospecting for potential rare metal mineralization associated with Precambrian structures within the Ukrainian Shield.

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МАГНЕТИТОВІ ТА ІЛЬМЕНІТОВІ СЕРІЇ ГРАНІТОЇДІВ ТА ОЦІНКА ЇХ ПОТЕНЦІЙНОЇ РУДОНОСНОСТІ НА НІОБІЙ І ТАНТАЛ В МЕЖАХ УКРАЇНСЬКОГО ЩИТА

У статті обговорюється можливість встановлення відмінностей між різновидами гранітів, які були сформовані в межах території Українського щита. Запропонована класифікація частково спирається на головні принципи, що були використані в деяких ранніх класифікаціях гранітів, серед яких є широко відома класифікація, яка була запропонована в роботах Ішихари [11] та Чеппела і Вайта [4], а також локальна (регіональна) класифікація, яка була запропонована в роботі Томурко та Щербакова [20]. У цій локальній класифікації була зроблена спроба навести відмінності між двома контрастними різновидами гранітів – апобазитовими та апопелітовими гранітами, які були встановлені в межах Українського Щита.

Класифікація гранітів на різні генетичні типи може стикатися з певними труднощами внаслідок гетерогенного генезису цих утворень – можливого змішування розплавів з їх батьківських мантійних та корових джерел. Але в той же час така класифікація може використовуватися для можливого прогнозу металогенічної спеціалізації різних генетичних типів гранітів. Серед параметрів, які можуть істотно вплинути на процеси накопичення металів в гранітах суттеву роль відіграють окислювально-відновні умови їх формування. На базі значень цього параметра граніти можуть бути підрозділені на два типи – граніти ільменітової та магнетитової серій, які характеризуються різними умовами їх формування [6, 11] і асоційованої мінералізації [1].

В межах території Українського Щита можливо виділити цілі провінції з відмінним розподілом переважно ільменітової або магнетитової серій гранітів. Ці граніти характеризуються різними стилям рудної мінералізації асоційованої з ними. Тантало-ніобати знаходяться серед типових рудних мінералів, які можуть бути встановлені в тісній асоціації з цими серіями гранітів, оскільки ніобій і тантал характеризуються наявністю тісної літофільної спорідненості з цими утвореннями. На Українському Щиті тантало-ніобати були знайдені в гранітах і асоційованих з ними рідкіснометальних пегматитах, деяких апогранітних метасоматитах (грейзени, кварцити, пертозити) а також в лужних породах різного генезису.

Було встановлено, що характерні ознаки тантало-ніобатів, серед яких значення Ta/Nb, відношення Fe/Mn і присутність елементів домішок, показують близьку спорідненість з різними генетичними серіями гранітів. Ці ознаки, які були встановлені як типоморфні для ільменіт і магнетитових серій гранітів, можуть бути використані для оптимізації проведення пошуково-розвідувальних робіт на Українському Щиті.

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МАГНЕТИТОВЫЕ И ИЛЬМЕНИТОВЫЕ СЕРИИ ГРАНИТОИДОВ И ОЦЕНКА ИХ ПОТЕНЦИАЛЬНОЙ РУДОНОСНОСТИ НА НИОБИЙ И ТАНТАЛ В ПРЕДЕЛАХ УКРАИНСКОГО ЩИТА

В статье обсуждается возможность установления различия между разновидностями гранитов, сформированных в пределах территории Украинского щита. Предложенная классификация частично базируется на основных принципах, использованных в некоторых ранних классификациях гранитов, среди которых широко известная классификация предложенная в работах Ишихары [11], а также Чеппела и Вайта [4], и локальная (региональная) классификация, предложенная в работе Томурко и Щербакова [20]. В данной локальной классификация была сделала попытка провести различие между двумя контрастными разновидностями гранитов – апобазитовыми и апопелитовыми гранитам, которые были установлены в пределах Украинского Щита.

Классификация гранитов на различные генетические типы может сталкиваться с определенными трудностями из-за гетерогенного генезиса этих образований – возможного смешивания расплавов с их родительских мантийных и коровых источников. Но в то же время такая классификация может использоваться для возможного прогноза металлогенической специализации различных генетических типов гранитов. Среди параметров, которые могут существенно повлиять на накопление металлов в гранитах важную роль имеют окислительно-восстановительные условия их формирования. На базе значений этого параметра граниты могут быть подразделены на два типа – граниты ильменитовой и магнетитовой серий, которые характеризуются различными условиями образования [6, 11] и ассоциированной минерализации [1].

В пределах территории Украинского Щита можно выделить целые провинции с отличным распределением существенно ильменитов или магнетитовых серий гранитов. Эти граниты показывают различные стили рудной минерализации ассоциированной с ними. Тантало-ниобаты находяться среди типичных рудных минералов, которые могут быть установлены в тесной ассоциации с этими сериями гранитов, поскольку ниобий и тантал характеризуются наличием тесного литофильного сродства с данными образованиями. На Украинском Щите тантало-ниобаты были найдены в гранитах и ассоциированных редкометальных пегматитах, некоторых апогранитных метасоматитах (грейзены, кварциты, пертозиты) а также в щелочных породах различного генезиса.

Было установлено, что характерные признаки тантало-ниобатов, среди которых значения Ta/Nb, отношение Fe/Mn и присутствие элементов примесей, показывают близкое сродство с различными генетическими сериями гранитов. Эти признаки, установленные как типоморфные для ильменитовых и магнетитовых серий гранитов, могут быть использованы для оптимизации проведения поисково-разведочных работ на Украинском щите.