



UDC 551.24:552.311(5-012)

EVOLUTION OF MINERAL FORMS OF RARE ELEMENT ACCUMULATION IN ORE-BEARING GRANITES AND METASOMATITES OF VERKHNEURMIYSK ORE CLUSTER (PRIAMUR REGION)

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It has been attempted to expand existing understanding of accessory mineralization evolution of rare metal-granite series at post-magmatic stage of their development and formation of associated hydrothermal deposits. Composition and distribution of rare elements of Verkhneurmiysk ore cluster have been examined from the position of mineralogy: the study focused on accessory and ore minerals Sn, W, Nb, Ta, Bi, Y, rare earth elements in rare metal Li-F granites and associated metasomatites. It has been discovered that accessory magmatic and hydrothermal mineral complexes share the same geochemical features, are formed under the leading role of abovementioned elements and consistently follow each other over time. It has been traced how mineral forms of accumulation of Sn, W, Nb, Ta, Y and rare earth elements evolve in the processes of magmatic crystallization and post-magmatic metasomatism in the time series: rare metal granites → zwitterites → tourmalinites → chloritites. Mineral rocks of each stage were noted to inherit mineralogical and geochemical distinctions from the rocks of the previous stage. A significant number of minerals, forming in the course of two-three stages, have been discovered, as well as omnipresent magmagenic-hydrothermal minerals. For a number of accessory minerals of rare metal granites post-magmatic generations have been identified. Special diversity among accessories of rare metal granites and zwitterites was observed in tungsten, tin and bismuth minerals.

Composition of Verkhneurmiysk ore cluster mineralization is in many aspects similar to the one of metasomatic accessory minerals of rare metal granites. Tungsten-tin deposits of Verkhneurmiysk ore cluster are polyformational and combine mineralogical features of cassiterite-quartz and cassiterite-silicate formations. Evolution of mineral forms of the key rare elements (Sn, W, Bi, Nb, Ta, rare earth elements) points to a genetic link between tungsten-tin mineralization of zwitter-tourmalinite formation and rare metal Li-F granites of the region. The evidence of that is a specific genetic category of accessory materials, formed as a result of pseudomorphism of protominerals, – transformational minerals or metasomatic accessory minerals. The list of metasomatic accessories includes exactly those minerals which are considered indicators of ore generation capacity of Far Eastern leucogranites: cassiterite, wolframite, scheelite, a number of sulphides.

Similar qualitative composition of magmatic and post-magmatic minerals demonstrates metallogenic specialization of parent granite magma. Predicted mineragenic significance of research in the field of mineral forms evolution of rare elements accumulation in ore-bearing granites and metasomatites is associated with possibilities to assess metallogenic specialization of parent granite magma. Understanding of accessory paragenesis evolution should serve as a base for exploration geo-technologies in the Far East metallogenic province.

Key words: evolution, accessory minerals, rare metal granites, zwitterites, tourmalinites, Verkhneurmiysk ore cluster, Priamur region

How to cite this article: Alekseev V.I., Sukhanova K.G., Gembitskaya I.M. Evolution of Mineral Forms of Rare Element Accumulation in Ore-Bearing Granites and Metasomatites of Verkhneurmiysk Ore Cluster (Priamur Region). *Zapiski Gornogo instituta*. 2017. Vol. 224, p. 149-155. DOI: 10.18454/PMI.2017.2.149

Introduction. Russian state-run program «Development of industry and improvement of its competitiveness» (2013) implies creating conditions for launch and complex development of rare and rare earth metal deposits. Judging by accounted reserves of most rare metals, Russian raw materials base holds a leading position in the world. However, due to certain geological and economic reasons (metal content in the ore, geographic and economic location of mining regions etc.), Russia yields to countries-producers of rare metal concentrate. One of the causes of this disadvantage is insufficient information about conditions of formation and localization of Russian deposits linked to rare metal granites.

Recent history of regional geologic research in Russia can be divided into three stages: 1) 1938-1964; 2) 1964-2005; 3) starting from 2005. The second stage, marked with meso-scale mapping of mining regions, led to the discovery of ore-bearing rare metal granites (RG), which allowed to collect rich mineralogical and petrographic materials and to reveal key features of RG petrogenesis [3-5, 11]. It has been identified that RGs are magmatic rocks containing industrial rare metal (Ta, Nb, W, Sn, Li, Be, REE) accessory mineralization (65-70 mineral species). It has been noticed that in granitoid series the number of mineral species and diversity of mineral

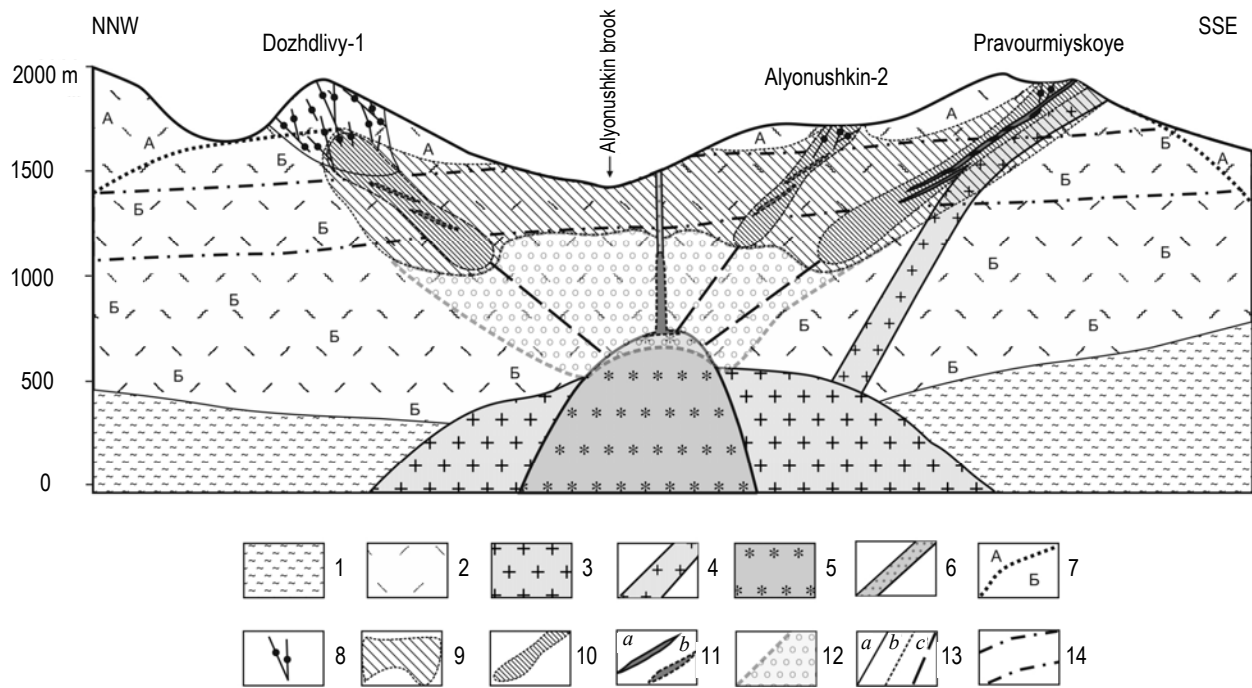


Fig. 1. Upper stage of Verkhneurmisk ore-magmatic system
(based on materials by Yu.B. Marin, G.T. Skublova, V.I. Alekseev et al., 1990, supplemented by V.I. Alekseev)

1 – crystalline schists; 2 – rhyolite ignimbrites; 3 – apical prominence of biotite leucogranites in Verkhneurmisk block; 4 – dyke of biotite granite-porphyrries; 5 – inferred deep intrusion of Li-F granites; 6 – dyke of siderophyllite-albite granites; 7 – boundary of regional propylites zone: A – albite-amphibole, Б – biotite; 8 – mineralized zones of zwitter and tourmalinites, unbroken; 9, 10 – stockworks with zwitter aureoles of siderophyllite-quartz facies: medium (9), intensive (10); 11 – zwitter ore zones of siderophyllite-topaz-quartz facies: proven (a) and inferred in the depth (b); 12 – bulk zwitterization; 13 – boundaries: geological (a), metasomatic (b), tectonic (c); 14 – boundaries of ore deposition zone

classes rise from granodiorite-granites to late subalkaline-alkaline rare metal granites, also significantly increases the role of oxides, fluorides, carbonates, phosphates and silicates of rare and rare earth elements. Complication of the structure of accessory minerals occurs in the same direction: from oxides with coordination geometry to layered and skeletal ones; from silicates with isolated Si-tetrahedra to skeletal structure. By the late members of granitoid series – rare metal granites – there is an increase in occurrence frequency and mineral content of volatile components, rare and rare earth metals, notably with progressively low bulk earth values [13].

One of the most popular concepts of hydrothermal ore genesis implies a magmatic source of fluids and mineral matter. The presence of genetic links between mineralization and granitoids as well as diversity of its manifestations have been highlighted by A.G. Betekhtin (1955), Yu.A. Bilibin (1955), I.G. Magakyan (1974), V.I. Smirnov (1982), L.N. Ovchinnikov (1988) and other researchers. On the example of Verkhneurmisk ore cluster in Mid-Priamur region the authors attempted to expand existing understanding of accessory mineralization evolution of rare metal-granite series at post-magmatic stage of their development and formation of associated hydrothermal deposits.

Geology of research object. Badzhal ore region, a part of Khingano-Okhotsk tin-bearing magmatic province in Priamur region [9], has been selected as a reference research region. Geology of the region and the largest Verkhneurmisk Cu-W-Sn ore cluster is described in the article [4]. In the eastern exo-contact of Verkhneurmisk granite batholite an RG areal has been detected, which controls numerous rare metal mineralizations and the largest Verkhneurmisk tungsten-tin deposit [1, 2, 7, 9]. An important feature of Verkhneurmisk ore-magmatic system (OMS) is a wide development of sequentially formed tin-ore topaz-siderophyllite zwitter, tourmalinite and chlorite metasomatites, composing metasomatic zones and stockworks, including ore bodies



of Pravourmiysk deposit, mineralizations Alyonushkin, Omot-Makit, Wolfram-Makit, Dozhdliiv and others (Fig.1). Placement and zoning of zwitter, tourmalinites and chlorites is controlled by Pravourmiysk rare metal complex [2]. It is a highly relevant issue to compare mineral composition of ore-bearing granites and wallrock metasomatites.

Research methodology and method. Scientific and methodological base of this research is the classification of rare metal granites by S.M.Beskin – Yu.B.Marin (1976-2013) [5]. The model of deep focal structures by I.N.Tomson and M.A.Favorskaya (1973; 1992) [8] has been used, supplemented with the concept of Far Eastern OMS by V.G.Gonevchuk [9]. The basic research principle was integration of mineralogical and petrologic information at different levels of matter composition: separate minerals, combined rocks and whole formations. Research methods included: geological mapping of key ore cluster, regional correlation of granitoid and metasomatic mineralizations, formation analysis. In order to solve petrologic tasks, a method of electronic petrography has been used based on combination of optical and electron-microscopic methods of mineral examination (SEM, EMPA, CL, SIMS). The key executive party was research-academic center «Fundamental research on minerals-indicators of petro- and ore-genesis» of Saint-Petersburg Mining University.

Evolution of mineral forms of rare element concentration in ore-bearing granites and metasomatites of Verkhneurmiysk ore cluster. Detailed examination of the largest deposits of the region allowed to trace the evolution of rare metal mineralization in the time series: RG → zwitter → tourmalinites → chlorites. On the one hand, accessory magmatic and hydrothermal mineral complexes of various stages share the same geochemical features and are formed under the leading role of W, Sn, Nb, Ta, Bi, Y and REE, on the other – they consistently follow each other over time. Mineralogic-geochemical evolution of rocks involves consistent alteration of mineral concentration forms of rare and ore elements (inherent and trace ones). Rocks of each stage have been noted to inherit mineralogical and geochemical distinctions from the rocks of the previous stage. A significant number of minerals, forming in the course of two-three stages, have been discovered, as well as omnipresent magmagenic-hydrothermal minerals: cassiterite, ferberite, scheelite, rutile, native bismuth, monazite.

RGs and post-magmatic zwitter induce intensive accumulation of lithophile elements: F, Li, Nb, Ta, W, Y, REE, Th, U. Moreover, minerals Li, Nb, Ta are unusual for late metasomatites, whereas F, W, Y, REE, Th, U form at all development stages of ore-magmatic systems. Typomorphic lithophile elements of geochemical ore processes, associated with RG penetration, are F, W, Y, REE, Th, U. Fluorine is a part of fluorite, topaz, phosphates. Tantalum-niobium, tungsten, yttrium-rare earth and thorium-uranium mineralizations are the most characteristic ones of RGs and zwitter, where, along with chemically specific phases (ferberite, monazite, xenotime, scheelite, allanite, chernovite, thorite, uraninite), there is an abundance of complex oxides with Nb, Ta, U, Th, Y, heavy REE, W, Sn (samarskite, fergusonite, bismuth pyrochlore, ishikawaite, euxenite). Many minerals Nb, W, Y, REE (scheelite, ferberite, cyrtolite, monazite) can also be found in recent hydrothermalites (see Table).

Minerals of chalcophile elements occur in the course of development of rare metal granites and form at all stages of post-magmatic hydrothermal process. There is «maggene-pneumatolytic» complex of minerals Sn, As, Bi, the content and diversity of which sharply increase at the transition from magmatic to zwitter stage and then go down; «hydrothermal» complex includes minerals Cu, Zn, Pb, In, Ag, the content and amount of which steadily go up in the late stages of hydrothermal process. The key chalcophile elements – Sn, As, Bi – form oxygen compounds (oxides, arsenates, wolframites), among them the basic ore minerals of zwitter and tourmalinites – cassiterite and arsenopyrite; they also form a part of tantalum-niobates. Starting from zwitter stage, the role of Sn, As, Bi sulphides steadily increases: from loellingite, arsenopyrite and bismuthinite to copper sulphostannates (stannite, stannoidite, mawsonite, kesterite etc.), sulphosalts of Ag, Cu, Bi (see Table). Acces-



sory chalcophiles – Cu, Zn, Pb, In, Ag – are unusual for granites. They occur as sulphides, arsenates, sulphosalts in zwitter and at later stages become ore elements. Typomorphic elements of chalcophile mineralization of tungsten-tin deposits of the region are Cu, In.

Evolution of mineral associations of rare elements in ore magmatic system with rare metal granites in Verkhneurmiysk ore cluster (Priamur region)

Rare element	Li-F granites and ongonites	Zwitter	Tourmalinites	Chloritites
Nb	<u>Wolframoixiolite</u> , <u>samaraskite</u> , aeschynite, ishikawaite, fergusonite, pyrochlore, bismuth pyrochlore, ilmenorutile, euxenite, columbite, (rutile, <u>ilmenite</u>)	Fergusonite, euxenite, pyrochlore, (ferberite, rutile)	(Rutile, ferberite)	–
Ta	<u>Wolframoixiolite</u> , ixiolite, ishikawaite, euxenite, columbite, (<u>samaraskite</u> , bismuth pyrochlore)	(Euxenite)	–	–
W	<u>Wolframoixiolite</u> , ferberite, scheelite, (<u>samaraskite</u> , aeschynite, rutile, ilmenite, ishikawaite, bismuth pyrochlore)	<u>Ferberite</u> , scheelite, (rutile, euxenite, cassiterite, pyrochlore, ilmenite)	Ferberite, (rutile)	Scheelite
Sn	Cassiterite, ixiolite, (<u>wolframoixiolite</u> , rutile, ishikawaite)	<u>Cassiterite</u> , stannite, sakuraiite, kesterite, (rutile)	<u>Cassiterite</u> , stannoidite, mawsonite, stannite, kesterite, (<u>chalcopyrite</u> , titanite)	Cassiterite
Bi	Bismuth pyrochlore, bismuth, (ishikawaite, <u>samaraskite</u> , aeschynite)	Bismuth, bismuthinite, mozgovaite, wittichenite, bismite, gananite, hedleyite, zavaritskite, bismuthoferrite	Bismuth, bismuthinite, wittichenite, (tennantite)	Bismuth, benjaminite
Y, REE	<u>Monazite</u> , cerianite, xenotime, <u>samaraskite</u> , ishikawaite, fergusonite, allanite-(Y), aeschynite, chernovite-(Y), britholite-(Y), euxenite, tveitite, (apatite, <u>fluorite</u> , <u>zircon</u>)	<u>Monazite</u> , xenotime, allanite-(Ce), fergusonite, euxenite, (<u>fluorite</u> , cyrtolite)	Monazite, (cyrtolite)	(Epidote)

Note. Key accessory and ore minerals are underlined. Minerals in the brackets contain the rare element as a significant (> 0.1 %) impurity.

New information on RG accessory mineralization of Verkhneurmiysk ore cluster shows that their composition contains a wide representation of late- and post-magmatic minerals (fergusonite, euxenite, native bismuth, pyrochlore etc.), observed even in zwitter. Post-magmatic generations have been identified for a number of accessory minerals of rare metal granites: ferberite, wolframoixiolite, fergusonite, scheelite, fluorite, monazite, rutile and others (Fig.2). An important role in the complex of hydrothermal accessories is played by sulphides: arsenopyrite, bismuthinite, pyrite, pyrrhotine, chalcopyrite, galenite, sphalerite [7, 9]. We have also detected metasomatic minerals which previously have not been reported in the Far East region: sakuraiite, cupropearceite, argentotennantite, mozgovaite, karibibite etc. [3]. Special diversity among RG accessories and zwitter was observed in tungsten (wolframite, wolframoixiolite, scheelite etc.), tin (cassiterite, ixiolite, stannite, kesterite, sakuraiite etc.) and bismuth minerals (bismuth pyrochlore, gananite, wittichenite etc.) [15] (see Table, Fig.2).

As follows from the Table, composition of ore mineralization in Verkhneurmiysk Cu-W-Sn ore cluster is in many aspects similar to the one of metasomatic accessory minerals of rare metal granites. However, only tungsten has identical mineral forms in granites and ores, being a part of wolframite, scheelite, wolframoixiolite (Fig.2). As an isomorphic trace element, tungsten is also included in a number of accessory and hydrothermal minerals: tantalum-niobates, ilmenite, rutile [3, 15]. Other ore elements – Nb, Sn, As, Bi, Pb, Zn – are mostly found in granites as isomorphic accessory components or as a part of less common accessory minerals; at late- and post-magmatic development stages of ore-magmatic systems they form ore minerals. E.g., niobium and tantalum compose

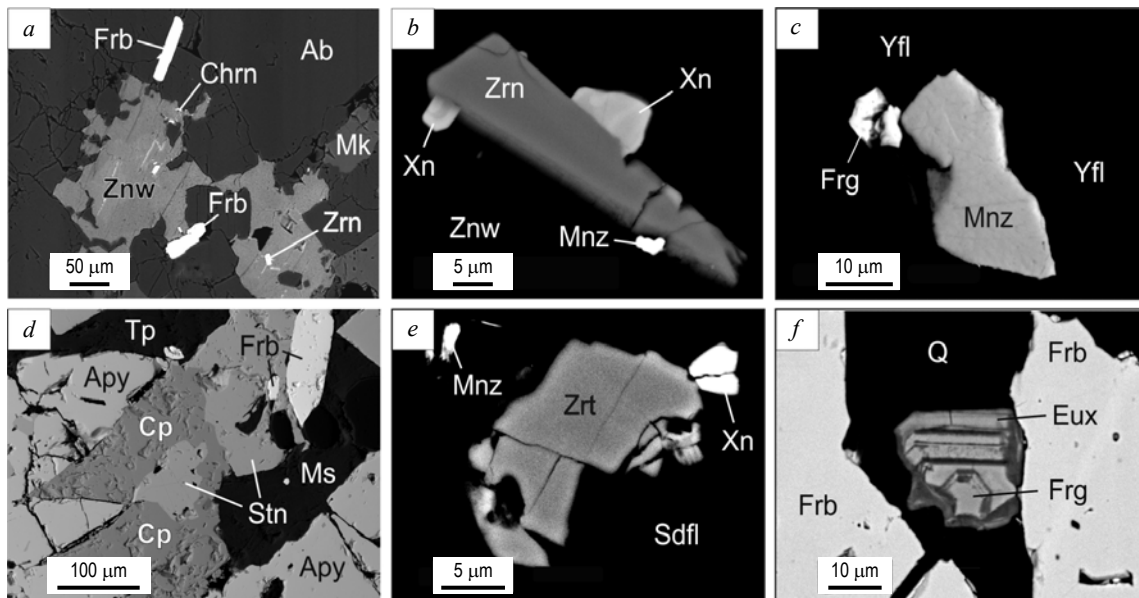


Fig.2. Inherent composition of accessory and ore mineralization of zinnwaldite rare metal granites (a-c) and topaz-quartz-siderophyllite zwitter (d-f) of Verkhneurmisk ore cluster.

Minerals: Ab – albite, Apy – arsenopyrite, Chrn – chernovite-(Y), Cp – chalcopyrite, Eux – euxenite-(Y), Frb – ferberite, Frg – fergusonite-(Y), Mk – microcline, Mnz – monazite-(Ce), Ms – muscovite, Q – quartz, Sdfl – siderophyllite, Stn – stannite, Tp – topaz, Xn – xenotime-(Y), Yfl – yttrifluorite; Znw – zinnwaldite, Zrn – zircon, Zrt – cyrtolite

accessory and ore ferberite, as well as a whole range of tantalum-niobates, including fergusonite (Fig.2). Magmagenic-hydrothermal evolution of mineral species centers around Nb, Sc de-enrichment of wolframite and increasing diversity of rare earth trace elements in fergusonite.

Rare earth elements form their own mineral phases in RGs and metasomatites: monazite-(Ce), xenotime-(Y), allanite-(Y), euxenite-(Y) etc. Accessory cassiterite turned out to be less common in leucogranites and rare metal granites than it could have been expected from published works [3, 15]. The reason for that is presence of other late-magmatic tin carriers in RGs: rutile (up to 22.92 % SnO₂), wolframoixiolite (up to 2.65 % SnO₂), struverite, titanite. Occasional cassiterite findings in rare metal granites were of late-magmatic origin. Bismuth concentrates in the form of bismuth pyrochlore, in the native state or as a trace element in ishikawaite, samarskite, aeschynite.

Discussion of results. Deep leucocratization of crust rocks under the influence of intratelluric fluids can cause formation of ore-magmatic systems with rare metal granites [6, 8, 9]. The largest Pravourmisk tin and tungsten deposit is spatially joined with an RG complex of the same name [2, 3, 15]. From the position of existing classifications [10], tungsten-tin deposits of Verkhneurmisk ore cluster are polyformational and combine mineralogical features of cassiterite-quartz and cassiterite-silicate formations. A single tungsten-tin zwitter-tourmalinite ore formation associated with RGs has been identified [1, 2, 7, 10]. A characteristic distinction of mineralogic-geochemical evolution of zwitter-tourmalinite deposits is inheritance of rare-element composition of RG accessories by post-magmatic minerals, in the course of which the ore-forming role passes from key lithophile components (W, Nb, Y, REE) to accessory chalcophile ones (Sn, Bi, As, Cu, Pb) [3].

Evolution of mineral forms of rare elements points to genetic links between zwitter-tourmalinite formations and RGs. This evolution can be explained in the following way. An important RG distinction is a constant presence of geochemically and crystallochemically related groups of accessory mineral species [5, 11, 16]. Such accessories usually form characteristic attachments, which contain information about conditions of their occurrence and growth [3, 12]. Unique distinctions of rare metal-granite magma facilitate simultaneous crystallization of related accessory minerals [11, 16]. Chemical micro-heterogeneity and evolution of the melt, accumulation of rare elements in residual pore solutions determine the sequence of crystallization and replacement of early mag-



matic accessory minerals (topaz, fluorite, ilmenite, zircon, monazite, xenotime, ferberite, wolframoixiolite, fergusonite, aeschynite, thorite, spodumene, amblygonite) by late-magmatic ones (ilmenorutile, cassiterite, bismuth pyrochlore, samarskite, allanite, ishikawaite, chernovite, scheelite, rooseveltite, britolite, asselbornite, philipsbornite, cerianite, thorianite, pyrofanite) [3]. Special genetic category of minerals forms as a result of crystallochemical adjustment (pseudomorphism) of magmatic protominerals to new conditions – transformational minerals [14]. Such late- and post-magmatic accessories, forming as a result of interference between crystallizing melt and the fluid or post-magmatic RG transformation, were referred to as «metasomatic» by V.V. Lyakhovich [12]. It should be noted that the list of metasomatites includes only those minerals that are considered indicators of ore forming capacity of Far Eastern leucogranites: cassiterite, wolframite, scheelite, arsenopyrite, loellingite, bismuthinite etc. [3, 8, 9].

Similar qualitative composition of magmatic and post-magmatic minerals reflects metallogenic specialization of parent granite magma [6, 7, 9]. Ore elements can concentrate in granites at the magmatic stage, forming an RG accessory-ore mineralization [13], dissipate as trace elements in rock-forming and accessory minerals or accumulate in residual melt-solutions and form post-magmatic ore mineralization [3, 6, 12, 13]. According to principles described in publications [6, 12, 13, 15], presence of accessory minerals containing ore elements (Sn, W, Bi, Nb, Ta, REE) and their evolution should be regarded as indicators of genetic link between W-Sn mineralization of zwitter-tourmalinite formation and RGs of Verkhneurmisk ore cluster.

Conclusions

1. From the position of existing classifications [10], tungsten-tin deposits of Verkhneurmisk ore cluster are polyformational and combine mineralogical features of cassiterite-quartz and cassiterite-silicate formations.

2. Evolution of mineral form concentration of rare and ore elements in Verkhneurmisk ore-magmatic system has been identified, which points to a genetic link between ore-metasomatic zwitter-tourmalinite formation and RGs.

3. Species composition, constitution and typomorphism of RG accessory minerals and associated hydrothermalites serve as indicators of forming conditions for granitoid series and potential ore-bearing capacity of specific intrusions. Predicted mineragenic significance of mineral forms evolution of rare elements accumulation in ore-bearing granites and metasomatites is associated with possibilities to assess metallogenic specialization of parental granite magma. Understanding of accessory paragenesis evolution should serve as a base for exploration geo-technologies (panning method, mineralogic-geochemical mapping etc.) in the Far East metallogenic province.

Acknowledgements. *Research has been carried out with financial support from Russian Foundation for Basic Research (project № 14-05-00364a) and Ministry of Education and Science of Russian Federation (basic and project parts of the state order in the field of science № 5.9248.2017/VU in 2017-2019).*

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The paper was accepted for publication on 11 November, 2016.