

## Using Radioactive Elements and the Th/U Ratio in Study of the Geochemical Typification of Granitoids and Their Intrusive Character

L.P. Rikhvanov

Tomsk Polytechnic University, pr. Lenina 30, Tomsk, 634050, Russia

Received 20 March 2018; accepted 26 July 2018

**Abstract**—This paper considers the use of radioactive elements for the solution of geochemical problems related to granitoid magmatism. The metallogenic types of granites are recognized on the basis of the contents of radioactive (Th-bearing, rare-metal, U-bearing, gold ore, etc.) elements in them. Special attention is focused on the Th/U ratio, which is higher than 2.5 in primary igneous rocks, such as Li–F granites of the Ongon-Hayrhan-deposit (Mongolia). Granitoids with  $\text{Th/U} < 2$  (and, the more so,  $\text{Th/U} < 1$ ) cannot be considered igneous. They are either metasomatized or of metasomatic genesis. Petrochemical and geochemical data on these types of rocks should be used for petrological implications with great care. Radiogeochemical indices can and must be used as additional indicators of the genesis of rocks, when it is controversial because of the widely manifested convergence and metasomatism processes.

**Keywords:** uranium, thorium, Th/U ratio, granitoid magmatism, radiogeochemical typification, metasomatism

### INTRODUCTION

Natural radioactive elements (NRE) are abundant and thus have attracted attention as geologic indicators since their discovery.

At the early stages of the development of radiogeochemistry, the use of uranium, thorium, and radioactive isotope  $^{40}\text{K}$  was hampered by the lack of reliable measurement methods. By the beginning of the 1970s, many methods for determining natural radioactive elements (luminescence, photocolometry, neutron activation, delayed-neutron method, gamma spectrometry, etc.) appeared, which permitted a reliable high-sensitivity determination of these components in all natural objects both in laboratory and in the field, including remote (aerial and other), conditions.

This made it possible to obtain quickly a large amount of information on NRE geochemistry in different geologic processes and during magmatism of different types, especially granitoid one.

These data were first discussed at the First All-Union Radiogeochemical Conference in Novosibirsk in 1972, in which all great scholars in this field took part, such as A.I. Tugarinov, L.V. Komlev, V.I. Gerasimovskii, V.I. Baranov, A.A. Smyslov, F.P. Krendelev, Ya.N. Belevtsev, N.N. Amshinskii, etc.

Later, the role of NRE as indicators in geologic processes were discussed at the Second All-Union Conference in Du-

shanbe in 1975, at the Third All-Union Conference in Tomsk in 1992, and at the International Conferences “Radioactivity and Radioactive Elements in the Environment” in Tomsk in 1996, 2004, 2009, 2013, and 2016.

All these conferences were concerned with the geochemistry of uranium, thorium, and potassium during granitoid magmatism.

Similar problems were and are still studied in the USA, Europe, and other countries.

Numerous studies have shown that NRE occur in particular quantities in all world objects, including intrusive ones (Hamilton, 1989; Rikhvanov, 2004, 2017).

The geochemistry of NRE, first of all, U and Th, in natural objects of the Earth’s shells has been comprehensively studied (Smyslov, 1974; Vinogradov, 2013), especially in the lithosphere. Soviet and Russian geologists V.I. Vernadsky, A.P. Vinogradov, D.I. Shcherbakov, V.I. Baranov, V.I. Gerasimovskii, L.V. Tauson, L.V. Komlev, I.E. Starik, A.I. Tugarinov, Ya.I. Belevtsev, N.P. Ermolaev, A.A. Smyslov, E.B. Vysokoostrovskaya, E.V. Plyushchev, G.B. Naumov, V.P. Kovalev, V.A. Zlobin, etc. and non-Russian specialists J.A. Adams, C.V. Allegre, E.S. Larsen, C.L. Roders, T.G. Lovering, J.N. Rosholt, I.D. Davis, G.W. Vine, M. Pagel, etc. established the certain regularities of the behavior and accumulation of NRE during particular geologic processes, including magmatism (Smyslov, 1974; Hamilton, 1989).

For example, with increasing silica or alkali contents in the rocks, the contents of U and Th also increase, reaching a maximum in alkali granites (Smyslov, 1974; Hamilton, 1989). This regularity can be violated only in alkaline, alka-

✉ Corresponding author.

E-mail address: rikhvanov@tpu.ru (L.P. Rikhvanov)

line-ultrabasic, and ultrabasic rocks (Smyslov, 1974; Rikhvanov, 2002). For example, we have established that alkaline-ultrabasic rocks (ankaratrites) of the Erbin neck have the same NRE contents as alkali granitoids (U = 8.5 ppm, Th = 25.7 ppm, K = 1.6%), and the Altai rocks, known as lamprophyres, have abnormal contents of these elements (U = 6.5–7.6 ppm, Th = 55.3–59.0 ppm). In some cases, the contents of U and Th reach 16 and 136 ppm, respectively, with Th/U = 8.5, which undoubtedly indicates the extremely specific regimes of the rock formation (Vasyukova, 2017).

The NRE content tends to increase from early igneous complexes or phases to late ones, which reflects the general regularity of enrichment of residual melts with U and Th. The geochemical history of uranium during the differentiation of magmas is related to none of the rock-forming elements (Si, K, Na, etc.) (Tauson, 1961). A statistical relationship between NRE and petrochemical indicators of igneous rocks was established, which provided a radiogeochemical key to the determination of these rocks in the  $a-Q$  coordinates used in the petrochemical classification by A.N. Zavaritskii (Ermolaev and Sobornov, 1973).

The terms “carrier minerals” and “concentrating minerals” were introduced (Tauson, 1961), and major uranium species in rocks and minerals were identified (Smyslov, 1974).

It was established that the chemical properties (valence, ionic radii, solubility, etc.) of  $U^{4+}$  and  $Th^{4+}$  are similar to those of REE, Zr, Hf, and some other trace elements, while  $U^{6+}$  is strongly different from these ions.

In other words, U and Th have an identical geochemical history in magmatic and high-temperature processes, but in low-temperature fluid–water systems in exogenous conditions it is different because of the transition of  $U^{4+}$  into  $U^{6+}$ . The Th/U ratio is an indicator of this change. Unfortunately, insufficient attention is paid to this radiogeochemical parameter, which could often help to determine the genesis of igneous rocks (Allègre et al., 1986; Rikhvanov, 2002).

## MATERIALS AND METHODS

This paper is based on the author’s long-term personal observations of the radiogeochemical characteristics of rocks, metasomatites, and ores in different regions of Russia and the world and on numerous literature publications and reference books presenting Th and U as classical indicators of particular types of rocks. The data obtained by productive institutions (Sosnovgeologiya, Berezovgeologiya, Step’geologiya, etc.) were also used in aero-gamma spectrometric studies of the territories of Russia and Mongolia, for which maps of U, Th, and K were compiled. These maps give an idea of radiogeochemistry of the regions. The limits of detection of these elements by a modern KAS aerodevice are as follows: U—0.35 ppm, Th—0.5 ppm, and K—0.11%, which are lower than the Clarke contents of these elements in the Earth’s crust.

Sampling and preparation of the samples for a radiogeochemical study were performed in accordance with the available methodological recommendations based on numerous radiogeochemical and geochemical investigations by Russian geologists (N.I. Safronov, A.P. Solovov, A.A. Smyslov, etc.) and with regard to the techniques of geochemical analysis used in other countries, including the recommendations on and experience in the implementation of the MG-KhK-1000 program and International MRGK No. 259 Program.

Transparent or, in some cases, polished transparent and polished thin sections were prepared for almost each sample, which made it possible to obtain homogeneous pure geochemical samples without superimposed processes for a subsequent statistical processing.

Uranium and thorium, chosen as major elements for the typification of ore-magmatic systems, have unique nuclear-physical ( $\alpha$ ,  $\beta$ , and  $\gamma$  radioactivity) and chemical properties, which helped to elaborate and introduce a great number of both laboratory and field analytical methods. The metrological parameters of some of these methods are given in Table 1.

**Table 1.** Metrological parameters of some applied laboratory methods of rock analysis for uranium and thorium

Method	Sample mass, g	Detection limit, wt. %		Reproducibility, rel. %		Time of analysis	Sample preparation
		U	Th	U	Th		
Luminescence, TsAL-1	0.2	$3 \times 10^{-5}$	–	10	–	3 h	Chemical
Luminescence, TsAL-2	1.0	$5 \times 10^{-6}$	–	13	–	6 h	Chemical
Photocolorimetry	0.1	$2 \times 10^{-3}$	$1 \times 10^{-4}$	15	5–10	8–10 h	Chemical
Chemical, with weight analysis	1.0	$5 \times 10^{-3}$	–	5–10	–	1 h	Chemical
X-ray fluorescence spectrometry	1–3	$2 \times 10^{-4}$	$5 \times 10^{-4}$	5–10	10	2 min	Without chemical preparation
Gamma spectrometry of natural radioactivity	500	$2 \times 10^{-5}$	$4 \times 10^{-5}$	10–15	10–15	1 h	Without chemical preparation
Instrumental neutron activation gamma spectrometry	0.1	$1 \times 10^{-5}$	$1 \times 10^{-5}$	5–10	5–10	5 h	Without chemical preparation
Track fission radiography	0.01	$1 \times 10^{-8}$	$1 \times 10^{-7}$	15–50	15–50	24 h	Special
Delayed-neutron method	10	$2 \times 10^{-7}$	$4 \times 10^{-5}$	2–10	5–15	2.5 min	Without preparation

Comparison of different methods of analysis for radioelements showed that the delayed-neutron method (DNM), developed at Tomsk Polytechnic University, is the most highly sensitive and correct for analysis for uranium. The high accuracy of uranium determination by DNM is evidenced by data on the certification of the former USSR and GDR reference standards and modern IAEA (International Atomic Energy Agency) standards. We chose this method as basic for determining uranium in rocks, ores, and minerals (Vertman et al., 1977).

Instrumental neutron activation analysis is also a good method for multielement analysis. We widely use it because Tomsk Polytechnic University has a research nuclear reactor with good operating characteristics.

Field gamma-spectrometric analysis for radioactive elements attracted our attention because it is an express mobile high-sensitivity and high-accuracy method for the solution of applied radiogeochemical problems. Taking into account the basic requirements for gamma spectrometry (the measurement geometry, a possible shift of a radioactive equilibrium, and fluctuations in the instrument operation), one can achieve a satisfactory reproducibility of the results of measurement.

For example, the reproducibility of the results is 10–20 rel.% for uranium (determined from the content of radium) ( $U = 2\text{--}10$  ppm), 15–30% for thorium ( $Th = 3\text{--}20$  ppm), and 20% for potassium ( $K = 0.5\text{--}1.5\%$ ). The above metrological parameters permit using this method for the solution of many applied problems of radiogeochemistry.

Using gamma spectrometry, it is possible not only to apply a statistical approach in radiogeochemical studies, with unlimited measurements (usually, until three close values are obtained), but also to correct the field works, establish the tendencies and dynamics of changes in the parameters of rocks and ores directly during their geological study, and to make a more representative sampling for laboratory studies.

Numerous experimental determinations of the radiogeochemical parameters of geologic objects by different field gamma spectrometry methods (envelope method, averaged linear method, etc.) yielded similar results (differing by 15–20%). Moreover, the contents of accumulated natural radioactive elements measured by different instruments of the same type at the same site in different years differ by no more than 15–25%.

Thus, the field and laboratory methods for uranium and thorium determination yielded reliable data for the subsequent radiogeochemical typification of ore-magmatic systems and genetic constructions. An exception is X-ray fluorescence analysis, which yields highly erroneous contents of U and Th in the presence of interfering elements (Ba, Sr, Pb, Hg, Sb, Bi, and some others).

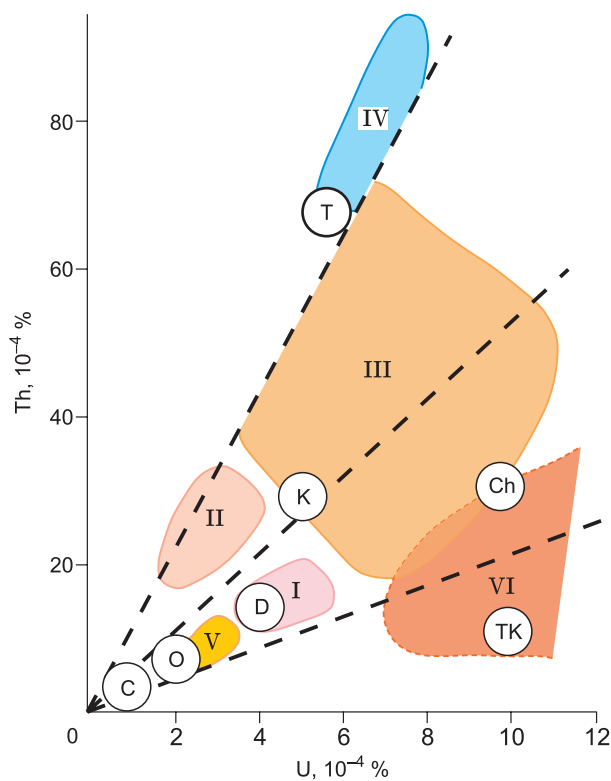
In recent decades, inductively coupled plasma mass spectrometry (ICP MS) has been widely applied in geochemical studies. This method permits accurate determination of a wide range of elements, including Th and U. We seldom used it in our research, but its results permitted us to assess the representativeness of particular geologic samples.

The average contents of radioelements in rocks, ores, and minerals were calculated from the results of their determination by at least two independent methods, which had a reproducibility of 10–30%.

## RESULTS

The problem of geochemical typification of granitoids has been discussed for decades (Tauson, 1961). Classifications of these rocks by the contents of accumulated uranium and thorium appeared as early as the 1950s. The first radiogeochemical classification of the USSR granitoids was proposed by L.V. Komlev in a closed ScD thesis in 1950. He recognized radiogeochemical groups of rocks, determined their ore potential types, and thus predicted areas promising for uranium deposits in Ukraine and Central Asia.

The above research work became widely known after publication by Smyslov (1974). In radiogeochemical classification diagrams (Rikhvanov, 2002), granitoids are well identified according to their metallogenic signatures (Fig. 1).



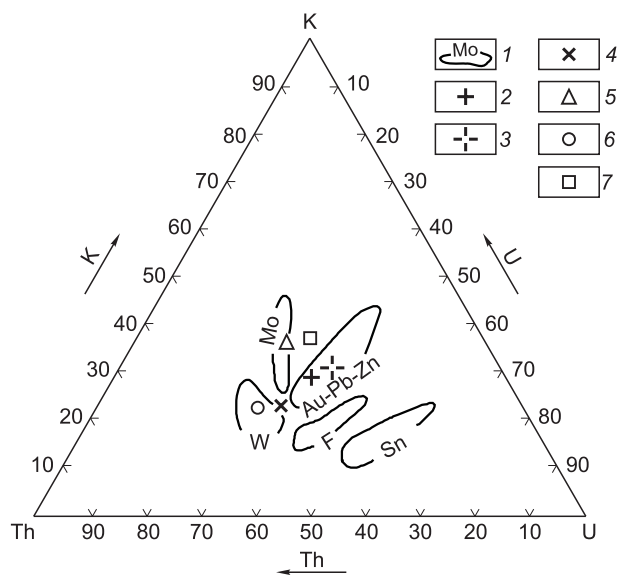
**Fig. 1.** Major radiogeochemical types of granites and the arrangement of some granitoids of the Altai–Sayan Folded Area in their composition fields (after (Smyslov, 1974), supplemented). I, normally radioactive granites,  $Th/U = 2.5\text{--}4.5$ ; II, enhanced radioactive granites,  $Th/U = 6\text{--}10$ ; III, highly radioactive rare-metal granites,  $Th/U > 5\text{--}10$ ; IV, highly radioactive Th-bearing granites,  $Th/U > 10$ ; V, weakly radioactive plagiogranites,  $Th/U < 2\text{--}5$ ; VI, highly radioactive, U-bearing granites,  $Th/U = 1\text{--}2$ . ASFA granites of: T, Tarak pluton; D, Dudet (Tigertysh) subtype (Andat, Sarala, etc.); O, Ol'gino subtype (Karlygan, Kolodzhul, etc.); Ch, Chebula complex; K, Kalguty pluton; TK, Talitsa–Karakol type; Ts, Tsentral'nenskii type.

Such diagrams make it possible to recognize different groups of granite plutons and complexes according to the radiogeochemical features of the rocks. For example, the Tsentral'nenskii, Dudet, and Ol'gino gold-bearing granite intrusions (Kuznetsk Alatau) and a complex of rare-metal granitoids of the Kalguty and other plutons (Gorny Altai) are arranged close to each other in the above classification diagram. The Tarak monazite-containing (group IV) and Talitsa–Karakol and Chebula U–rare-metal–REE granitoids (Altai and Kuznetsk Alatau) are localized separately from them, in the field of U-bearing granitoids.

European researchers assign high heat production granitoids to group VI in the above classification. They report that some types of uranium deposits are associated with such granitoids (Simpson and Plant, 1984).

A ternary U–Th–K diagram is widely used to interpret aerospectrometric data and separate granitoids of different metallogenic signatures according to radiogeochemical data. For example, the metallogenic typification by M.P. Nikolaenko and other researchers in Transbaikalia made it possible to recognize the fields of granitoids with definite types of regional endogenous deposits (Fig. 2).

By our request, the radiogeochemical data for some granitoid plutons on the eastern slope of Kuznetsk Alatau were processed by M.P. Nikolaenko. The diagram (Fig. 2) shows two groups of rocks, one of which falls in the field of Au-bearing Transbaikalian granitoids, and the other, in the field of Mo-bearing ones. Commercial molybdenum and gold deposits of the Altai–Sayan folded area (ASFA) (Kommunar, Sarala, Sorskoe, etc.) also lie in these fields. As follows from the above data, there are no Sn- and fluorite-containing granitoids on the eastern slope of Kuznetsk Alatau.



**Fig. 2.** Ternary U–Th–K diagram with fields of granitoids. 1, metallogenic type of Transbaikalian granitoids; 2–7, granitoid plutons from the eastern slope of Kuznetsk Alatau: 2, Ulen'–Tuim, 3, Solgon, 4, Askiz, 5, Saigachi, 6, Yuli, 7, Kotur.

As shown by numerous studies, the content of radioactive elements in granitoids is an indicator of not only the composition of the Earth's crust in which a magma chamber formed and evolved (Nozhkin, 1997) but also the crust evolution as a whole (Nozhkin, 1983; Nozhkin and Rikhvanov, 2014). The differentiation of the crustal matter was accompanied by the concentration of trace and radioactive elements (U, Th, and K). In the course of its evolution, the continental crust acquired mostly a dioritic composition, and its upper layer, a granodioritic one (Taylor and McLennan, 1985). Compared with the primitive mantle, the continental crust is 50–60 times richer in radioactive elements, and its upper granite-metamorphic layer is 150–160 times richer in them. Highly radioactive Na–K granitoids are the products of the highest degree of geochemical differentiation of the Earth's matter. They terminate the global stages of crustal growth and the formation and cratonization of continents.

Radioactive elements in igneous rocks, especially granitoids, are good indicators of some of their geochemical types, e.g., *A*-type, and their ore potential (Stussi, 1970; Hennesy, 1981; Rikhvanov, 2002). When *A*-type granitoids forming in intraplate settings are considered, it is implied that they are enriched in Th, U, Ta, REE, and other incompatible trace elements. Granitoids of the Arabian Shield or the Jos Plateau (Nigeria) (Solodov and Usova, 1986; Maurice and Charbonneau, 1987; Pollard, 1988) belong to the *A*-type. In the ASFA, the Chebula and some other complexes might be referred to *A*-type granitoids.

As shown by our (Rikhvanov, 2017) and other (Allegre et al., 1986; Ozima, 1987; Hamilton, 1989) studies, the Th/U ratio is an extremely important indicator. It varies over a very narrow range of values, starting from the Solar System as a whole (Th/U = 3.72) to its planets, the Moon (Th/U = 3.55), meteorites (Th/U = 2.5–8.6), and igneous rocks (2.5–5.5), with a predominance of values of 3.5–4.5. This suggests a general regularity of the distribution of Th and U, determined by the laws of the universe (Rikhvanov, 2004).

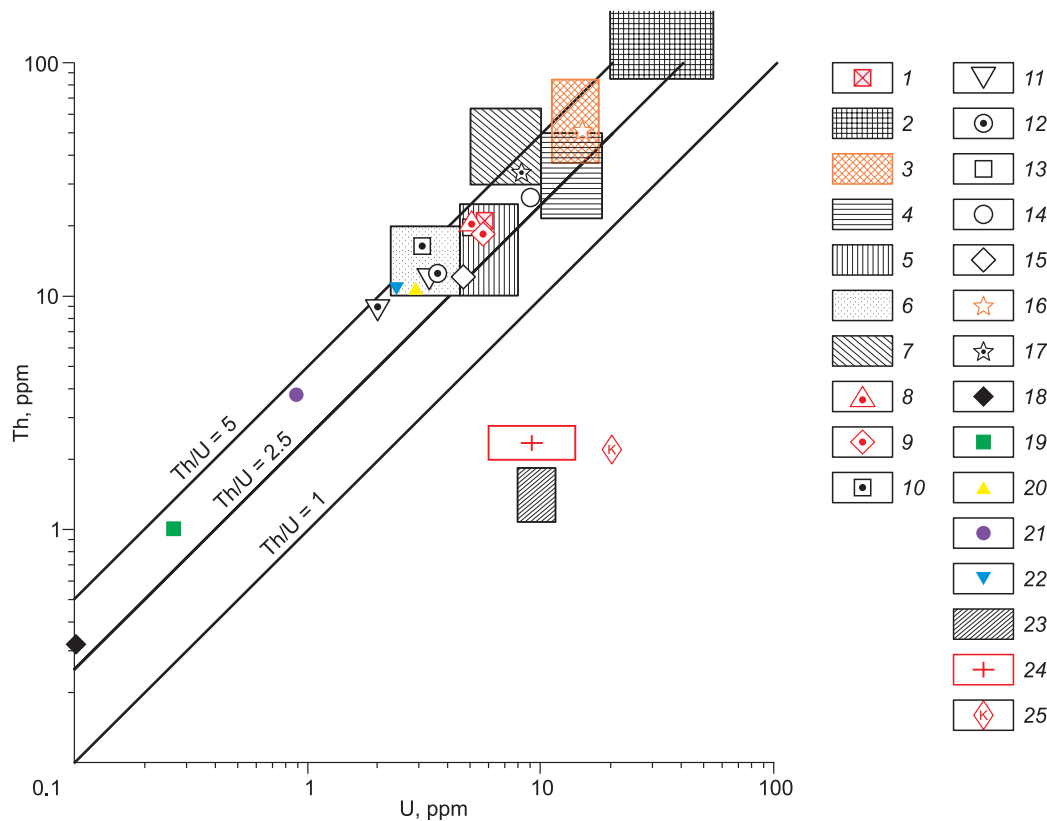
As noted by Vernadsky (1954), the constant Th/U ratio in the Earth's crust was noted by E. Rutherford in 1904 and by B. Boltwood in 1906. Vernadsky proposed to call this regularity the Rutherford–Boltwood law.

In other words, thorium is 3–5 times more abundant in nature than uranium. This proportion of Th and U is persistent in many rocks, especially igneous ones, except for chemogenic and biogenic rocks and products of metamorphism and metasomatism (Rikhvanov, 2002, 2004, 2017).

The Th/U ratio is a very important indicator of the magmatic nature of granites. In most of igneous rocks it is 2.5–7.0. In our opinion, this ratio is a criterion for the magmatic nature of rocks with convergence features.

There were attempts to treat the radiogeochemical parameters of rocks in terms of the geodynamic setting of their formation (Naumov et al., 2011), but this issue calls for a special study.

One of the debatable objects is the igneous rocks found in the area of the Ongon-Hayrhan deposit in Mongolia (Kova-



**Fig. 3.** Radiogeochemical typification of REE granitoids. 1, ongonites of the Ongon-Hayrhan deposit (Mongolia); 2–7, fields of metallogenic granitoids: 2, Ta–Nb granitoids of Nigeria (Rikhvanov, 2002); 3, Li granites of Corsica (Bonin, 1988); 4, Li- and Cs-containing and Ta- and Nb-containing Sn–W–Be granites (Stussi, 1970); 5, Sn–W–Be–U granites (Stussi, 1970); 6, W-bearing granites (Stussi, 1970); 7, U- and Th-bearing Cu–Mo–W granites (Stussi, 1970); 8, average world granite (Tweedie, 1979); 9, average granitoid formed during the late Paleozoic–Mesozoic activity in Central Siberia (Rikhvanov, 2002); 10, granitoids of Transbaikalia (from data by V.I. Medvedev et al.; radiogeochemical research materials of Sosnovgeologiya State Enterprise); 11, felsic volcanics of Transbaikalia (from data by V.I. Medvedev et al.; radiogeochemical research materials of Sosnovgeologiya State Enterprise); 12, ultrafelsic rhyolites of Gorny Altai (from data by Yu.A. Tikunov); 13, felsic glasses and rhyolites of Ethiopia (Rikhvanov, 2002); 14, rhyolites of Japan, JR-1 standard; 15, obsidian standard (USA); 16, NIMG granite standard (Africa); 17, porphyritic granites of Altai that host tungsten deposits; 18–22, estimated average contents of U and Th (Taylor and McLennan, 1985); 18, chondrites, 19, lower continental crust, 20, upper continental crust, 21, total continental crust, 22, pelagic clay, 23, Beauvoir topaz–rhyolitic granite (Beauvoir, France; sample from a depth of 400 m), 24, average Ta-containing spodumene granite of Altai; 25, average “kalgutite” from the Kalguty deposit (Altai).

lenko et al., 1971a). Similar rocks, e.g., the Kalguty dike complex in Altai and other areas, were also assigned to igneous rocks of this type (Annikova, 2003; Vladimirov et al., 2007).

Our study of the radiogeochemical characteristics of ongonites and kalgutites (Rikhvanov et al., 2017) showed that the dike rocks described as ongonites in Mongolia (they can be taken as a petrotype) are a complex with radiogeochemical signatures. The contents of U and Th are 5.7 and 20.6 ppm, respectively,  $Th/U = 3.6$ , and these rocks are similar in radiogeochemical parameters to the average world granites (Tweedie, 1979).

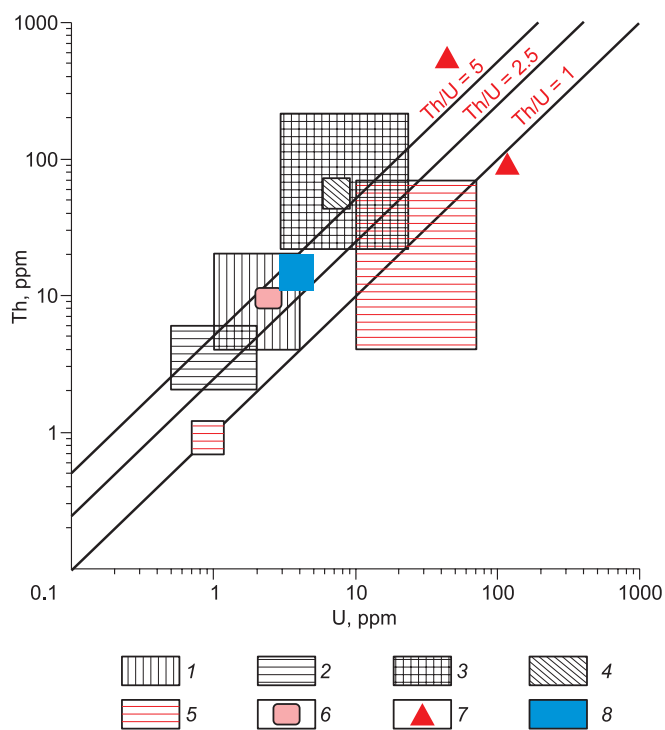
In these parameters the above complex is an analogue of classical intrusive rocks, as was stated earlier based on the results of comprehensive geological, mineralogical, and geochemical studies of ongonites in Mongolia (Kovalenko et al., 1971a).

In these parameters the studied petrotype of ongonites corresponds to W and Sn–W–Be–U granitoids (Stussi, 1970).

Dikes in Gorny Altai, identified as analogues of ongonites and called kalgutites (Annikova, 2003; Vladimirov et al., 2007), cannot be considered ongonites, because they have radically different radiogeochemical parameters ( $Th/U < 1$ ). This seems to be due to a high degree of their metasomatism, which is confirmed by a radiogeochemical diagram (Fig. 3).

A similar conclusion can be drawn for the Allakha pluton described as a new spodumene type of granites (Kudrin et al., 1994).

The structures, textures, and some geochemical features of the Allakha stock permit different concepts of its genesis. The conditions of occurrence of granites, their relationship with the host rocks, and the rims of fine-grained rocks simi-



**Fig. 4.** Radiogeochanical typification of granitoids of Mongolia. 1, gabbro-diorite–granite complex (Erdenedavaa, Khamar Davaa, Gargan, and Ilchir), 2, Lake (Hovd) and Turgen granitoid plutons, 3, Ulaan Del granitoids, 4, granitoids of the Turgen Gol terrane, near the Askhatin deposit, 5, Han-Bogdo granitoids, 6, Agardag, Altay, and Hovd granitoid plutons, 7, Haldzan-Buregtey granitoids, 8, subalkalic granitoids of the Lake terrane.

lar to chill zones at their endocontacts suggest the magmatic nature of the granites.

On the other hand, a number of features, such as the structural relationships among minerals (reflecting replacement and formation of porphyroblasts), gradual transition between different mineralogical and petrographic types of granites, etc., might indicate their metasomatic genesis. The geochemical spectrum of ores (the presence of Bi, Mo, Cu, W, Ag, and other trace elements) and the relationships among natural radioactive elements also suggest the metasomatic nature of these rocks.

For example, in the radiogeochanical diagram (Fig. 3), the field of the Allakha stock lies in the domain characteristic of metasomatized rocks with a  $\text{Th}/\text{U} < 1$ , which makes them radically different from, e.g., Li-containing granites of Corsica (Bonin, 1988) and Ta–Nb granites of Nigeria (Solodov and Usova, 1986; Pollard, 1988), whose  $\text{Th}/\text{U}$  values are typical of igneous rocks (Rikhvanov, 2002).

Note that some granitoids of Central Europe (e.g., the Beauvoir massif, Eibenstock) are of the same nature. They also fall in the field of hydrothermally altered rocks in the Th–U diagram (Fig. 3).

The ICP MS introduced into practice today is perfect for analysis for NRE. This gives a unique opportunity to check the representativeness of the samples used for the construc-

tion of various petrological and geodynamic models based on the  $\text{Th}/\text{U}$  ratio.

A radiogeochanical analysis of granitoids of Mongolia (Fig. 4), the so-called “country of granites”, shows different types of these rocks, which are usually of magmatic nature. But there are also geologic objects that have undergone a particular metasomatic transformation, and their current petrogeochemical characteristics differ from those of the primary igneous rocks (Rikhvanov et al., 2013).

Among such objects are the granite plutons of Mt. Haldzan-Buregtey, which host large reserves of Zr, Nb, Ta, REE, Th, and U, and the Han-Bogdo pluton. One of typical indicators of metasomatic transformation of these rocks is their great diversity. For example, eight mineral phases, including ones of metasomatic genesis, were recognized in the Haldzan-Buregtey granites (Kovalenko et al., 1995). Several mineral phases were also found in one of the largest alkali-granitoid massifs of Central Asia (Rikhvanov et al., 2013).

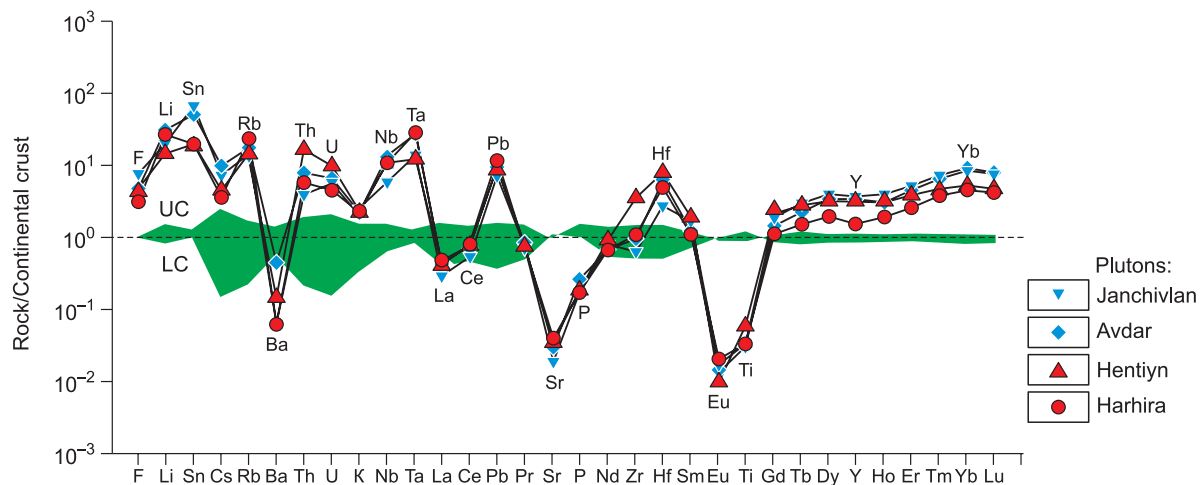
Of special interest in terms of radiogeochemistry is analysis of the so-called spidergrams, which are widely used today. Using spidergrams of radioactive elements (Fig. 5), it is possible to judge (with a certain probability) the correctness of attributing the studied pluton (complex) to an intrusive formation, and its petrochemical characteristics can be used for model constructions.

The presented spidergram (Fig. 5) clearly shows that three of the four plutons of Li–F–rare-metal granites in Mongolia (Avdar, Hentiyn, and Harhira) are Th-bearing, which is typical of intrusive rocks. As for complex plutons, such as Janchivlan, the points of Th and U in the spidergram indicate that the studied intrusive rocks are U-bearing and their initial petrochemical characteristics changed. It is hard to judge the types of these rocks from the diagram<sup>1</sup>, but it is known that the Janchivlan pluton includes greisen, microcline, and albitite bodies (Kovalenko et al., 1971b). It is not ruled out that in such complex plutons, fluid phases with uranium dominating over thorium are in equilibrium with magma at the final stages of its crystallization, which might influence the radiogeochemical parameters of rocks. But this issue calls for a special study.

In Siberian mountain–taiga regions, where detailed observations are not always possible, quartz–feldspathic metasomatites (microclinites, albitites, etc.) are often regarded and described as granitoids and pegmatoids with anomalous

<sup>1</sup> In 1969, together with outstanding Russian petrologist D.S. Korzhinskii, we examined ongonites, subvolcanic analogs of Li–F–rare-metal granites, discovered by V.I. Kovalenko. D.S. Korzhinskii had a good knowledge of Li–F granites of Mongolia. During the discussion of the examined exposures, he assumed a successive transformation of melt into fluid, i.e., fluid-magmatic rock genesis. This hypothesis was later confirmed by V.I. Kovalenko.

In Li–F granite plutons, there are sites of transition of rocks with typical magmatic structures into rocks subjected to autometasomatic processes. Based on the data obtained by L.P. Rikhvanov and the  $\text{Th}/\text{U}$  ratios, it is possible to study the successive transformation of melt into fluid during comprehensive investigation of well-exposed Li–F granite plutons in Mongolia (M.I. Kuz'min's note).



**Fig. 5.** Continental-crust-normalized trace-element spidergrams for Li–F–rare-metal granite plutons in the Early Mesozoic magmatism area in Mongolia. LC, UC, average compositions of the lower and upper continental crust, respectively.

radiogeochemical characteristics. Apparently, this anomaly is the only explanation of the low Th/U ratio in alkali granitoids of the Chebula complex and some Altai granitoids. Autometasomatism processes common in alkali granitoids significantly changed their radiogeochemical features. This is evident from the low Th/U ratio in these rocks (its weighted average is  $\sim 2$ ), which is significantly lower than its Clarke values in the Earth's crust and granites. For example, the average world granite (Tweedie, 1979) contains 5 ppm U and 21 ppm Th, with Th/U = 4.2.

In literature, however, the above fact is often ignored. For example, alkali granitoids of nine of the twelve studied plutons in East Sayan and Tuva (granite collections by V.N. Dovgal' and V.V. Minin) have Th/U < 2, and those of the Dugda, Duhu Nuur, Kodyros, Husun Gol, and Academic plutons are characterized by Th/U  $\leq 1$ , which unambiguously indicates their deep metasomatism.

There is a slight direct dependence of the Th/U ratio on the pluton size. In our opinion, this is due to a more intense autometasomatism of small bodies, which are often the apical parts of larger plutons that were not subjected to erosion and, probably, superposed metasomatism.

The determined contents of radioactive elements and Th/U ratios in the studied samples of igneous rocks show that up to 90% of the samples are initially igneous rocks and about 10% of the samples are not. Using the latter samples in petrological and geodynamic models can lead to erroneous conclusions.

## CONCLUSIONS

The common abundance of radioactive elements and their excellent nuclear-physical characteristics make it possible to determine these elements in all types of rocks with high metrological parameters.

The well-studied geochemical regularities of migration, scattering, and accumulation of radioelements and their species in rocks permit them to be used as indicators of crystallization and subsequent transformations.

By the example of granitoids it has been shown that the contents of radioactive elements reflect the geochemical and metallogenic features of rocks. Gold, molybdenum, and tungsten ores are localized in certain rocks. A-type granites have very specific radiogeochemical features.

The prototype of Li–F rocks in the Ongon-Hayrhan deposit has high contents of U and Th, 5.7 and 20.6 ppm, respectively, and Th/U = 3.6.

The Th/U ratio is a very important indicator. It can serve as a criterion for the decision whether the rocks under study are igneous or not. If Th/U < 1–2, then the rocks are surely not intrusive but metasomatic or metasomatized (Allakha pluton, Gorny Altai; kalgutites, etc.)

Radiogeochemical parameters, especially the Th/U ratio, can and should be used as additional indicators in study of igneous rocks whose genesis is controversial because of widely manifested convergence and metasomatism and at the stage of preliminary geochemical typification of granitoids.

## REFERENCES

- Allègre, C.V., Duprè, B., Lewin, E., 1986. Thorium/uranium ratio of the Earth. *Chem. Geol.* 56 (3–4), 219–227.
- Annikova, I.Yu., 2003. Rare-Metal Granites, Ongonites, and Elvans of the Kalguty Pluton, Southern Altai (Composition, Association with Mineralization, and Petrogenetic Model of Formation). PhD Thesis [in Russian]. OIGGM SO RAN, Novosibirsk.
- Bonin, B., 1988. Peralkaline granites in Corsica: some petrological and geochemical constraints. *Rend. Soc. Ital. Mineral. Petrol.* 43 (2), 281–305.
- Ermolaev, N.P., Sobornov, O.P., 1973. Mode of expression of radiogeochemical characteristics of rocks in a single coordinate system and its geochemical application. *Geokhimiya*, No. 6, 803–815.

- Hamilton, E.I., 1989. Terrestrial radiation—an overview. *Radiat. Phys. Chem.* 34 (2), 195–212.
- Hennesy, J., 1981. A classification of British Caledonian granites based on uranium and thorium contents. *Mineral Mag.* 44 (336), 449–454.
- Kovalenko, V.I., Kuz'min, M.I., Antipov, V.S., Petrov, L.L., 1971a. Topaz-containing quartz keratophyre (ongonite), a new variety of subvolcanic igneous vein rocks. *Dokl. Akad. Nauk SSSR* 199 (2), 430–433.
- Kovalenko, V.I., Kuz'min, M.I., Zonenshain, L.P., Nagibina, M.S., Pavlenko, A.S., Vladykin, N.S., Tseden, Ts., Gundsambuu, Ts., Goreglyad, A.G., 1971b. Rare-Metal Granitoids of Mongolia (Petrology, Distribution of Trace Elements, and Genesis) [in Russian]. Nauka, Moscow.
- Kovalenko, V.I., Tsaryeva, G.M., Goreglyad, A.V., Yarmolyuk, V.V., Troitsky, V.A., Hervig, R.L., Farmer, G.L., 1995. The peralkaline granite-related Khaldzan-Buregtey rare metal (Zr, Nb, REE) deposit, Western Mongolia. *Econ. Geol.* 90 (3), 530–547.
- Kudrin, V.S., Stavrov, O.D., Shuriga, T.N., 1994. A new spodumene type of Ta-bearing rare-metal granites. *Petrologiya* 2 (1), 88–95.
- Maurice, Y.T., Charbonneau, B.W., 1987. U and Th concentration processes in Canadian granitoids, their detection by airborne gamma ray spectrometry and their relationship to granophile mineralization. *Revista Brasileira de Geociências* 17 (4), 644–646.
- Naumov, V.B., Kovalenko, V.I., Dorofeeva, V.A., Girmis, A.V., Yarmolyuk, V.V., 2011. Average composition of magmatic melts in major geodynamic settings (from data of study of melt inclusions in minerals and of chilled glasses in rocks, in: Proceedings of the 11th International Seminar “Deep-Seated Magmatism, Its Sources and Plumes” [in Russian]. Irkutsk, pp. 130–167.
- Nozhkin, A.D., 1983. Geological and geochemical signs of maturity of Archean complexes and factors in the ore content of continental blocks of crust. *Geologiya i Geofizika (Soviet Geology and Geophysics)* 24 (8), 41–48 (35–40).
- Nozhkin, A.D., 1997. Petrogeochemical Typification of Precambrian Complexes in Siberia. ScD Thesis [in Russian]. Novosibirsk.
- Nozhkin, A.D., Rikhvanov, L.P., 2014. Radioactive elements in collisional and within-plate sodic–potassic granitoids: accumulation levels and metallogenic significance. *Geochem. Int.* 52 (9), 740–757.
- Ozima, M., 1987. *Global Evolution of the Earth*. Springer, Berlin, Heidelberg.
- Pollard, J.I., 1988. Petrogenesis of tin-bearing granites of the Emu-ford district, Herberton tinfield, Australia. *Austral. J. Earth Sci.* 35 (1), 39–57.
- Rikhvanov, L.P., 2002. Radiogeochemical Typification of Ore-Magmatic Systems [in Russian]. Izd. SO RAN, Filial “Geo”.
- Rikhvanov, L.P., 2004. Radioactive elements in geosphere shells, in: *Radioactivity and Radioactive Elements in the Human Habitat. Proceedings of the Second International Conference, Tomsk, 18–22 October 2004* [in Russian]. Tomsk, pp. 498–505.
- Rikhvanov, L.P., 2017. Radioactivity and radioactive elements as a factor of the geologic environment and its use in geosciences. *Razvedka i Okhrana Nedr*, No. 12, 55–61.
- Rikhvanov, L.P., Arbuzov, S.I., Galnemekh, O., Mashenkin, V.S., Batulzii, D., Gerel, O., Oyunbat, S., Garamjav, D., 2013. Radiogeochemical features of some granitoids of Mongolia and their rare-metal mineralization, in: *Radioactivity and Radioactive Elements in the Human Habitat. Proceedings of the Fourth International Conference, Tomsk, 4–8 June 2013* [in Russian]. TPU, Tomsk, pp. 451–456.
- Rikhvanov, L.P., Arbuzov, S.I., Batulzii, D., 2017. New data on the geochemistry of ongonites. *Geosfernye Issledovaniya*, No. 1, 50–59.
- Simpson, P.R., Plant, J.A., 1984. Role of high heat production granites in uranium province formation, in: De Vivo, B., Ippolito, F., Capaldi, G., Simpson P.R. (Eds.), *Uranium Geochemistry, Mineralogy, Geology, Exploration and Resources*. Springer, Dordrecht, pp. 167–178.
- Smyslov, A.A., 1974. Uranium and Thorium in the Earth's Crust [in Russian]. Nedra, Leningrad.
- Solodov, N.A., Usova, T.Yu., 1986. Ore Potential of Alkali Granites [in Russian]. VIEMS, Moscow.
- Stussi, J.-M., 1970. Rôles et significations du zircon et de la thorite dans la géochimie de l'uranium et du thorium des roches volcaniques associées au Culm des Vosges méridionales. *C. R. Acad. Sci. Paris* 271, 2255–2258.
- Tauson, L.V., 1961. Geochemistry of Trace Elements in Granitoids [in Russian]. Izd. AN SSSR, Moscow.
- Tauson, L.V., 1977. Geochemical Types and Ore Potential of Granitoids [in Russian]. Nauka, Moscow.
- Taylor, S.R., McLennan, S.M., 1985. *The Continental Crust: Its Composition and Evolution*. Blackwell, Oxford.
- Tweedie, J.R., 1979. Origin of uranium and other metal enrichments in the Helmsdale Granite, eastern Sutherland, Scotland. *Trans. Inst. Min. Metall., Sect. B* 88, 38–45.
- Vasyukova, E.A., 2017. Petrology and Fluid Regime of Formation of Lamprophyres of the Chuya Complex (Southeastern Altai–Northwestern Mongolia) [in Russian]. Izd. SO RAN, Novosibirsk.
- Vernadsky, V.I., 1954. *Selected Works. Geochemical Essays* [in Russian]. Izd. AN SSSR, Moscow, Vol. 1.
- Vertman, E.G., Stolbov, Yu.M., Meshcheryakov, R.P., 1977. The possibility of application of the delayed-neutron method for determination of uranium in geochemical studies. *Geokhimiya*, No. 9, 1337–1347.
- Vinogradov, A.P. (Ed.), 2013. *Major Geochemical Features of Uranium* [in Russian]. STT, Tomsk.
- Vladimirov, V.G., Annikova, I.Yu., Antipin, V.S., 2007. Ongonite–elvan magmatism in southern Siberia. *Litosfera*, No. 4, 21–40.

*Editorial responsibility:* M.I. Kuz'min