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Chemical composition and age of the accessory uraninite from granitoids of the crystalline basement of the Southern Yamal

Vera Vital'evna KHILLER*
Yuriy Viktorovich EROKHIN**
Kirill Svyatoslavich IVANOV***

The Zavaritsky Institute of Geology and Geochemistry of the Ural Branch of RAS, Ekaterinburg, Russia

The relevance of the work is due to the need to improve the method of chemical dating in application to high-uranium accessory minerals that cannot be date by isotopic methods of research.
The purpose of the work: the study of the accessory uraninite chemical composition from granitoids of the crystalline basement of Southern Yamal and the determination of its age.
Methodology of the research. Quantitative analysis of the chemical composition of zircon is performed with the X-ray spectral electron probe microanalyzer CAMECA SX 100 (electron beam diameter is from 1 µm, BSE, SE, Cat modes, determination of elements from beryllium to uranium). Spectra were obtained using inclined wave spectrometers; intensity measurement was carried out according to analytical lines: Th *Ma*, U *Mb*, Pb *Ma*, Y *La*, Si *Ka*, Ce *La*, La *La*, Nd *La*. Age calculation was carried out according to the known methods of foreign authors in addition to authors own developments.
Results. The chemical composition of uraninite is quite stable and quite contaminated, the main impurities are thorium (ThO_2 up to 5 wt.%), lead (PbO up to 3 wt.%) and yttrium (Y_2O_3 up to 3.7 wt.%). Based on the impurity content in Verkhnerechensky uraninite, its unit cell parameters were calculated as 5.475 ±5.476 Å. A substantial lead content and high crystallinity of the substance allows the use of this mineral as a mineral-geochronometer. The calculated weighted average age of uraninite is 258.7 ± 3.4 Ma (MSWD = 0.19); and isochron age is 259 ± 8 Ma (MSWD = 0.34).
Conclusions. Microprobe compositions of uraninite were obtained, and the Late Permian age of granitoids was determined by chemical dating. The given chemical composition data show that Verkhnerechensky uraninite differs sharply from most of its analogues from other rocks, but it correlates quite well with accessory uraninites from S-type granites. The mineral is very close to synthetic uraninite and is not metamict; it also does not contain significant amounts of more oxidized uranium (UO_3 и U_3O_8).

Keywords: uraninite, composition, chemical dating, granitoids, crystalline basement, Southern Yamal, Arctic.

Introduction

Recent studies of geology of the Arctic have acquired special importance because of connection with the predicted oil and gas potential of this huge and still poorly studied territory, as well as its upcoming, probably, division between neighboring countries in the Arctic region. The most important criterion is, as we know, the results of the study of the basement of the Arctic sedimentary basins. The Yamal Peninsula is the main gas province of our country, and one of the few places where the crystalline basement is still available for direct study, but with great difficulty. It is also important that oil and gas influxes at many fields of the Yamal Peninsula were found from the complexes of the Paleozoic folded basement. The crystalline basement of the oil and gas provinces remains one of the few (more or less promising) insufficiently studied objects; the granitoids of basements are the most promising for the search for oil and gas [1, etc.].

Uraninite is a common accessory mineral of granitoids, but it is normally not studied due to its very small size and great dispersion in the rock. At the same time, uraninite has long been used (quite successfully) as a mineral-geochronometer, and much earlier than other radioactive minerals (the first dating of the mineral was carried out in 1911 by A. Holmes). Unfortunately, many publications are relevant to vein or ore deposits of uranium [2, 3, etc.], but one can find some works on chemical microprobe dating of accessory uraninites in granitoids [4, 5]. We have decided to conduct a similar study for accessory uraninites from granites from the crystalline basement of the Verkhnerechenskaya oil and gas exploration area (Yamal Peninsula).

Geological position of the object of research

Granitoids in the crystalline (pre-Jurassic) basement of Southern Yamal were discovered only in one place within the Verkhnerechenskaya oil and gas exploration area. The area itself (the studied well No. 1 located there), was near the famous Novoportovskoye oil and gas field, about 50 km to the south-west and about 225 km to the north-east from the city of Salekhard (the administrative center of the Yamal-Nenets Autonomous Okrug).

Verkhnerechenskaya well No. 1 (Fig. 1) penetrated the crystalline basement (namely fresh granites) in the depth interval of 1748–2034 m. No deeper drilling was carried out; intrusive rocks higher in the borehole are overlapped by middle and upper-Jurassic deposits of sedimentary cover. Granites throughout the section of the borehole are represented by homogeneous light gray fine-grained differences of biotite-quartz-feldspar composition. It was found that they belong to monzole-granites, which were formed along the sedimentary substrate (S-type granites), most likely, in late horogenic conditions. The crystallization time of

✉hilvervit@mail.ru
https://orcid.org/0000-0001-84914958
**erokhin-yu@yandex.ru
https://orcid.org/0000-0002-0577-5898
***ivanovks@igg.uran.ru
https://orcid.org/0000-0002-8292-4658

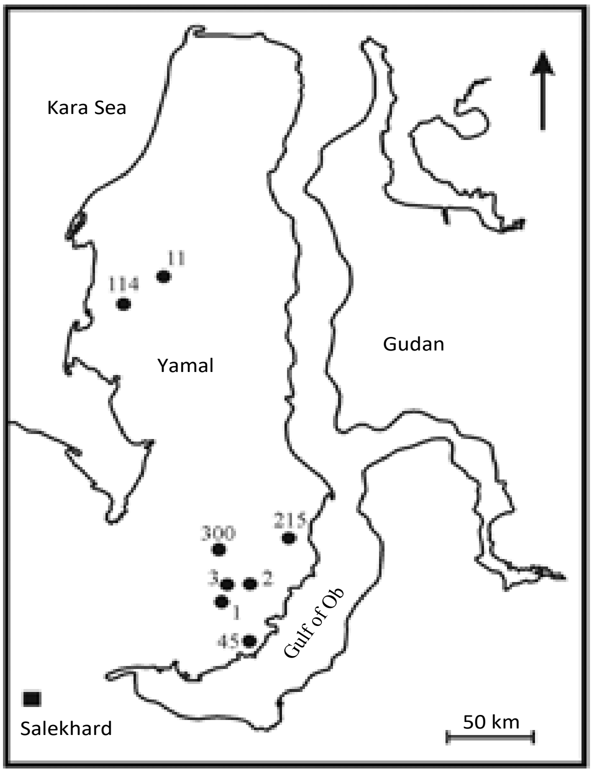

Figure 1. Location map of the wells penetrated the Paleozoic on the Yamal Peninsula; well numbers. 215 – Novoportovskaya; 300 – Zapadno-Yarotinskaya; 1–3 – Verkhnerechenskaya; 11 – Vostochno-Bovanenkovskaya; 45 – Syunai-Salinskaya; 114 – Bovanenkovskaya.
Рисунок 1. Схема расположения скважин, вскрывших палеозой, на полуострове Ямал; номера скважин: 215 – Новопортовская; 300 – Западно-Яротинская; 1–3 – Верхнереченская; 11 – Восточно-Бованенковская; 45 – Сюнай-Салинская; 114 – Бованенковская.

subalkaline leucogranites in the basement of the Verkhnerechenskaya area was estimated by chemical Th–U–Pb dating from accessory uraninite and monazite – 259.2 ± 3.6 Ma [6], and isotopic U–Pb dating from zircon – 254.0 ± 3.0 Ma [7]. Despite the detailed study of the material composition of monzoleogranites [8, 9], accessory uraninite was practically not described separately, even though it was an integral part of the bimineral isochron and rock dating was carried out on it.

Methods of research

The quantitative analysis of the chemical composition of uraninite was carried out using the CAMECA SX 100 electron probe microanalyzer (The Zavaritsky Institute of Geology and Geochemistry of the Ural Branch of the Russian Academy of Sciences, Ekaterinburg). Polished sections were made from granite samples, and then they were sprayed with a thin layer of carbon. Measurement conditions: accelerating voltage is 15 kV, current strength is 80 nA, electron beam diameter is 2 μm. Pressure in the chamber of samples is 2×10^{-4} Pa. Spectra were obtained using inclined wave spectrometers; intensity measurement was carried out according to analytical lines: Th Ma, U Mb, Pb Ma, Y La, Si Ka, Ce La, La La, Nd La. Measuring time of peak intensity for Th, U and Pb – 40 s, for Y and Si – 20 s, for the remaining elements – 10 s; against the background – half as much. Standard samples: ThO_2, UO_2, $Pb_2P_2O_7$, zircon, synthetic phosphates of REE. Detection limits of Th, U, and Pb were about 930, 650, and 430 g/t. Theoretical and practical substantiation of the chemical dating method using X-ray microprobe analysis is given in numerous publications on this subject [10, 11 etc.], including those for uraninite [4, 12, etc.]. The main condition of this method is: during evolution, the mineral did not lose radiogenic lead (i. e. the Th–U–Pb system was closed), the lead in the mineral was formed due to the decay of thorium and uranium.

Research results and their discussion

Uraninite, together with monazite-(Ce) forms abundant accessory impregnation in the granitoid, although less often than orthophosphate. It usually gravitates to places of accumulation of dark-colored minerals (mica and chlorite that replaces it), where it often forms small inclusions in the matrix of large grains of monazite and xenotime (Fig. 2, a) and composes well-formed square-section individuals (Fig. 2, b), up to 30 μm in diameter. In the BSE image mode, it differs well from other accessory minerals, having the brightest white color. Interestingly, uraninite in the form of inclusions was not observed in zircon [7, 13]; apparently, it crystallized itself later.

The chemical composition of uraninite is quite stable, although it varies slightly (Table). The main impurities are thorium, lead and yttrium. The following variation is observed for thorium – from 1.9 to 5 wt.% ThO_2; there is practically no variation for lead – 2.8–3.2 wt.% PbO. Large fluctuations in concentration are also observed for yttrium (Y_2O_3 from 0.4 to 3.7 wt.%). All these elements have an inverse correlation with uranium, which means that they are isomorphic components in the structure of the mineral. The sums of analyzes of uraninite are slightly underestimated, which is most likely due to the presence of other elements or higher uranium in the mineral. It is interesting that in terms of its composition, Verkhnerechensky uraninite differs sharply from the majority of its analogues [14, 15, etc.], but it is quite well correlated with accessory uraninites from S-type granites [5, etc.]. It is possible that our accessory uraninites, as part of the magmatic mineral association, have their own specific typochemistry characteristic only for such granitoids.

a b

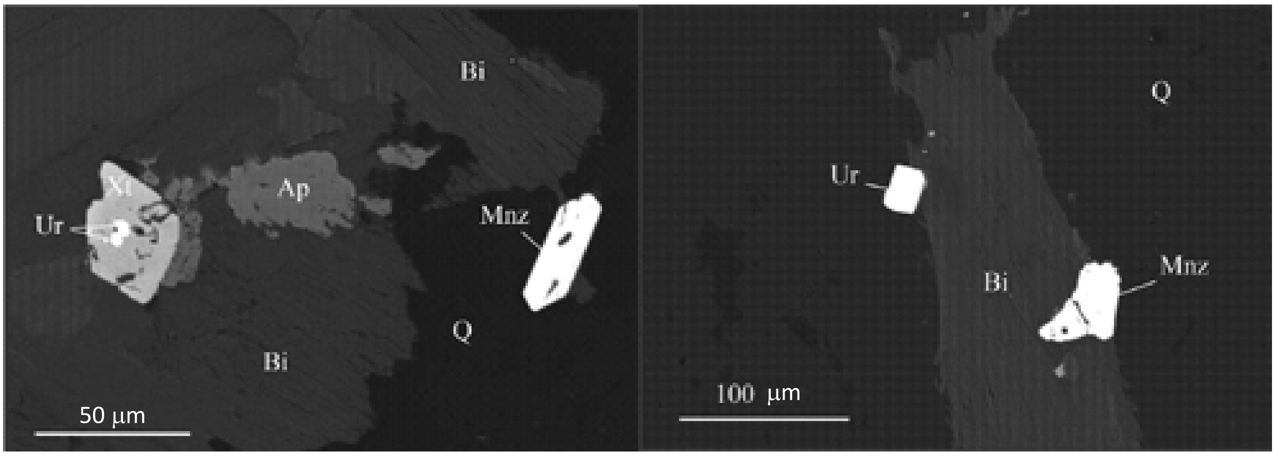

Figure 2. Grains of uraninite Ur, monazite Mnz, xenotime Xt and apatite Ap in the aggregate of quartz Q and biotite Bi from granitoids of the Verkhnerechenskaya area. CAMECA SX 100, BSE-image.
Рисунок 2. Зерна уранинита Ur, монацита Mnz, ксенотима Xt и апатита Ap в агрегате кварца Q и биотита Bi из гранитоидов Верхнереченской площади. CAMECA SX 100, BSE-изображение.

In 1959 years V. A. Leonova established the relationship between the unit cell parameters and the impurities in the composition of uraninite [16]. Using a graph, she estimated the change in the lattice parameters of the mineral upon inclusion of thorium, lead and rare earths, in at.%. With an average thorium content in Verkhnerechensky uraninite at 0.92 at.% Th, the unit cell parameter is estimated as a_o = 5.475–5.476 Å. For the synthetic reference uraninite, this parameter is defined as 5.4682 Å, and for natural uraninites it varies greatly from 5.33 to 5.49 Å. It is known that the unit cell parameter decreases with increasing oxidation state of uranium [17]. That is, Verkhnerechensky uraninite in terms of unit cell parameters is very close to the synthetic standard and is not metamict, and also contains an extremely small amount of more oxidized uranium (UO_3 and U_3O_8).

Chemical composition (in wt.%) of uraninite from granites of Southern Yamal.
Химический состав (в мас. %) уранинита из гранитов Южного Ямала.

Oxides	Analyses numbers										
	1	2	3	4	5	6	7	8	9	10	11
ThO_2	2.22	4.94	2.22	3.18	3.30	2.92	2.09	2.05	2.60	3.72	3.26
UO_2	88.79	83.18	88.08	87.59	89.85	86.56	87.24	88.45	85.03	88.41	88.98
SiO_2	0.09	0.32	0.13	0.68	0.15	0.26	0.10	0.10	0.03	0.54	0.29
Ce_2O_3	0.40	0.15	0.21	–	–	0.33	0.16	0.25	0.41	0.09	0.19
La_2O_3	–	–	0.01	–	0.18	0.01	–	–	–	–	–
Nd_2O_3	0.25	0.17	0.29	–	0.09	0.22	0.31	0.27	0.46	0.15	–
Y_2O_3	2.09	2.53	2.18	0.73	0.40	2.28	2.18	1.97	3.67	1.18	1.24
PbO	3.13	2.98	3.16	3.14	3.26	3.17	3.03	3.13	3.04	3.19	3.11
Total	96.97	94.27	96.28	95.31	97.23	95.73	95.12	96.21	95.24	97.28	97.07
Age, Ma	258	259	262	260	264	266	254	258	260	262	255

Oxides	Analyses numbers									
	12	13	14	15	16	17	18	19	20	21
ThO_2	2.35	3.31	2.17	3.64	1.88	1.93	2.21	2.99	2.70	2.16
UO_2	84.09	88.83	88.01	85.19	88.51	88.65	86.08	80.06	88.19	88.91
SiO_2	0.38	0.33	0.19	0.22	0.09	0.06	0.14	0.44	0.06	0.02
Ce_2O_3	0.05	0.04	0.02	0.24	0.25	0.04	–	0.15	0.13	0.05
La_2O_3	–	–	0.17	0.18	0.02	0.02	0.06	–	–	0.02
Nd_2O_3	0.20	–	0.15	–	0.33	0.17	–	0.21	0.29	–
Y_2O_3	2.21	1.28	1.95	1.88	1.98	2.01	2.04	3.21	2.50	1.98
PbO	2.93	3.18	3.14	3.08	3.13	3.14	2.99	2.81	3.12	3.13
Total	92.91	96.97	95.80	94.42	96.18	96.03	93.53	89.88	97.00	96.28
Age, Ma	254	260	261	263	258	259	254	256	258	257

Note: The Zavaritsky Institute of Geology and Geochemistry of the Ural Branch of RAS, microanalyzer CAMECA SX 100, analyst is V. V. Khiller.

The significant lead content obtained by microprobe analysis and the high crystallinity of substance make it possible to use this accessory uraninite as a mineral-geochronometer. The values of selective U–Th–Pb ages of uraninite lie in the range of 254–266 Ma with an error of 6–8 Ma and together give a weighted average age of 258.7 ± 3.4 Ma with MSWD = 0.19 (Fig. 3), which generally corresponds to the bimineral isochron previously given by us – 259.2 ± 3.6 Ma [6].

When constructing the PbO–UO_2^* dependence by the set of analysis points, accessory uraninites fall on one straight line (or isochron according to [14]), which indicates their one-time formation. Calculation of age by the angle of inclination of the isochron is 259 ± 8 Ma, MSWD = 0.34, probability = 0.997 (Fig. 4). In this case, the line crosses the origin of coordinates, which indicates the absence of inclusion or subtraction of lead during the evolution of uranium oxide.

Conclusions

Thus, we have studied in detail accessory uraninite from monzoleucogranites of the crystalline basement of Southern Yamal. Microprobe data were obtained on the composition of the mineral, and the Late Permian age of granitoids was determined by chemical dating. The given chemical composition data show that Verkhnerechensky uraninite differs sharply from most of its

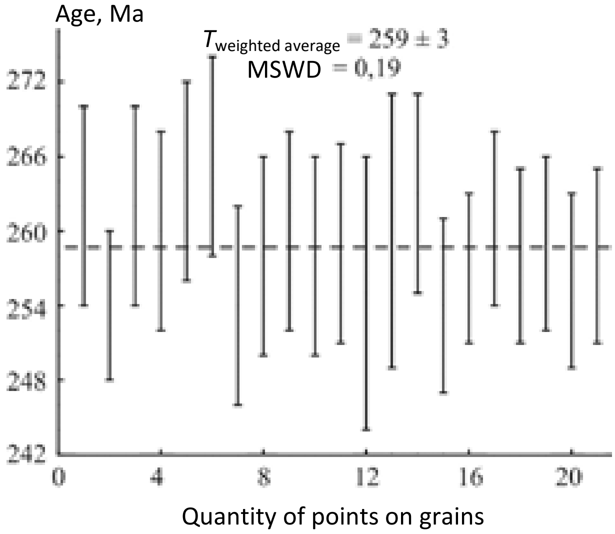

Figure 3. Variations in the values of Th–U–Pb ages and their weighted average for 21 selective determination in uraninite from Verkhnerechensky granites.
Рисунок 3. Вариации значений Th–U–Pb-возрастов и их средневзвешенная величина для 21 точечного определения в уранините из Верхнереченских гранитов.

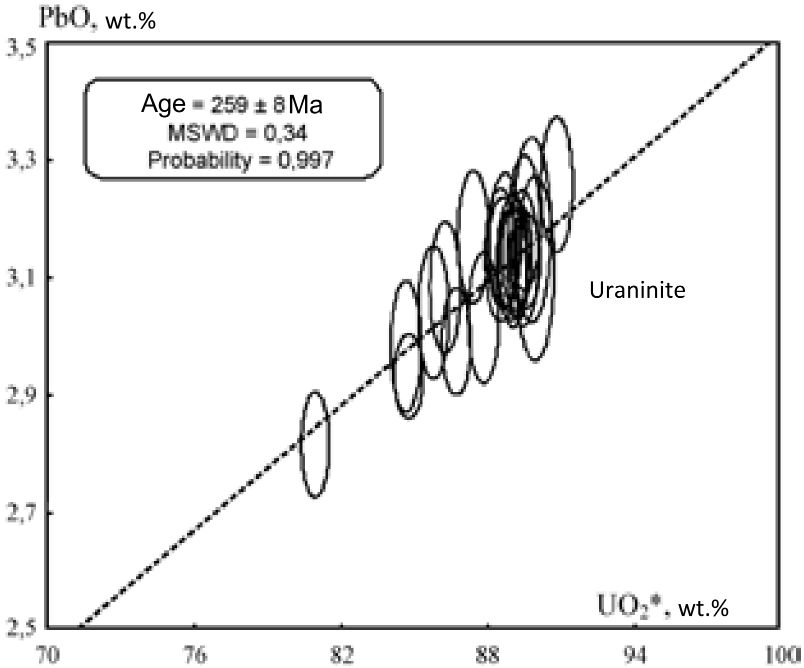

Figure 4. Isochron UO_2^*–PbO based on the results of uraninite analyses. UO_2^* = (UO_2+ThO_2^{eq}), where ThO_2^{eq} – the thorium content converted to the equivalent uranium content, capable of producing the same amount of Pb during the life of the system with the equality of U–Pb and Th–Pb-values of age.
Рисунок 4. Изохрона UO_2^*–PbO по результатам анализов уранинита. UO_2^* = (UO_2 + $ThO_2^{экв}$), где $ThO_2^{экв}$ – содержание тория, пересчитанное в эквивалентное содержание урана, способное произвести то же количество Pb за время жизни системы при равенстве U–Pb и Th–Pb-значений возраста.

analogues from other rocks, but it correlates quite well with accessory uraninites from S-type granites. Based on the impurity content in Verkhnerechensky uraninite, its unit cell parameters were calculated as 5.475–5.476 Å. It follows that the mineral is very close to synthetic uraninite and is not metamict, and also contains an extremely small amount of more oxidized uranium (UO_3 and U_3O_8).

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Химический состав и возраст акцессорного уранинита из гранитоидов кристаллического фундамента Южного Ямала

Вера Витальевна ХИЛЛЕР*
Юрий Викторович ЕРОХИН**
Кирилл Святославич ИВАНОВ***

Институт геологии и геохимии им. акад. А. Н. Заварицкого УрО РАН, Россия, Екатеринбург

Актуальность работы обусловлена необходимостью совершенствования метода химического датирования в применении к высокоурановым акцессорным минералам, которые невозможно датировать изотопными методами исследования.
Цель работы: исследование химического состава акцессорного уранинита из гранитоидов кристаллического фундамента Южного Ямала и определение его возраста.
Методология исследования: количественный анализ химического состава циркона выполнен на рентгеноспектральном электронно-зондовом микроанализаторе CAMECA SX 100 (диаметр пучка электронов от 1 мкм, режимы BSE, SE, Cat, определение элементов от бериллия до урана). Спектры получены на наклонных волновых спектрометрах, измерение интенсивности проводились по аналитическим линиям: Th *Ma*, U *Mb*, Pb *Ma*, Y *La*, Si *Ka*, Ce *La*, La *La*, Nd *La*. Расчет возраста проводился по известным методикам зарубежных авторов в дополнение к собственным наработкам.
Результаты. Химический состав уранинита вполне устойчивый и достаточно загрязненный, основными примесями являются торий (ThO_2 до 5 мас.%), свинец (PbO до 3 мас.%) и иттрий (Y_2O_3 до 3,7 мас.%). По содержанию примесей в верхнереченском уранините были рассчитаны его параметры элементарной ячейки 5,475–5,476 Å. Большое содержание свинца и высокая кристалличность вещества позволяют использовать данный минерал в качестве минерала-геохронометра. Рассчитанный средневзвешенный возраст уранинита 258,7 ± 3,4 млн лет (СКВО = 0,19), а изохронный – 259 ± 8 млн лет (СКВО = 0,34).
Выводы. Получены микрозондовые составы уранинита и методом химического датирования установлен позднепермский возраст гранитоидов. Приведенные данные по химическому составу показывают, что верхнереченский уранинит резко отличается от большинства своих аналогов из других пород, но вполне хорошо коррелируется с акцессорными уранинитами из гранитов S-типа. Минерал очень близок к синтетическому ураниниту и не является метамиктным, а также не содержит значимые количества более окисленного урана (UO_3 и U_3O_8).

Ключевые слова: уранинит, состав, химическое датирование, гранитоиды, кристаллический фундамент, Южный Ямал, Арктика.

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*hilvervit@mail.ru
https://orcid.org/0000-0001-84914958
**erokhin-yu@yandex.ru
https://orcid.org/0000-0002-0577-5898
***ivanovks@igg.uran.ru
https://orcid.org/0000-0002-8292-4658