Chemical composition and age of monazite-(Ce) in granitoids of the crystalline basement from the South Yamal

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Abstract

The relevance of the work is due to the need to improve the method of chemical dating as applied to high-thorium accessory minerals, which are difficult to date by isotope research methods.

Purpose of the work: study of the chemical composition of accessory monazite from granitoids of the crystalline basement of the South Yamal and determination of its age.

Research methodology: quantitative analysis of the chemical composition of monazite was carried out using X-ray spectral electron probe microanalyzer CAMECA SX 100 (electron beam diameter from 1 μm , BSE, SE, Cat modes, determination of elements from beryllium to uranium). The spectra were obtained with the help of inclined wave spectrometers, the intensity was measured using analytical lines: Th *Ma*, U *Mb*, Pb *Ma*, Y *La*, Si *Ka*, Ca *Ka*, P *Ka*, Ce *La*, La *La*, Pr *Lb*, Nd *La*, Sm *Lb*, Dy *La*, Gd *Lb*. The age calculation was carried out according to the well-known methods of foreign authors in addition to some developments of the author.

Results. The chemical composition of monazite makes it possible to classify it as a cerium variety, the content of radiogenic components varies greatly (in wt.%): ThO₂ – 5.37–16.31, UO₂ – 0.40–0.81, PbO – 0.08–0.19. There are significant concentrations of SiO₂ (up to 3.5 wt.%), Y₂O₃ (up to 1.8 wt.%) and CaO (up to 1.2 wt.%). It turns out that monazite implements hattonite (Th⁴⁺(U⁴⁺)+Si⁴⁺ → REE³⁺+P⁵⁺) and cheralite (Th⁴⁺(U⁴⁺)+Ca²⁺(Sr²⁺,Ba²⁺,Pb²⁺) → 2REE³⁺) isomorphism types. The decent content of lead and high crystallinity of the substance makes it possible to use this mineral as a geochronometer mineral.

Conclusions. New data on the chemical composition of monazite have been obtained, and the late Permian age of granitoids has been determined by microprobe dating. The values of the point U–Th–Pb ages of monazite together give a weighted average age of 256 ± 10 Ma (MSWD = 0.15) and an isochron of 254 ± 19 Ma (MSWD = 0.28), which almost ideally coincides with the results of isotopic U–Pb zircon dating from the same rock, 254 ± 3 Ma.

Keywords: monazite, chemical composition, chemical dating, granitoids, crystalline basement, South Yamal, Arctic.

Introduction

Research on geology of the Arctic has recently become particularly important in connection with the oil and gas potential of this vast and still very poorly studied territory, as well as its upcoming division between neighbouring countries in the Arctic region. The most important criterion is the results of the study of the crystalline basement of sedimentary basins in the Arctic.

The Yamal Peninsula is the main gas province of Russia, and one of the few places where the crystalline basement (albeit with great difficulty) is still accessible for direct study. Here the basement is located at relatively shallow depths of 1.5–1.7 km and drilling is not as expensive as in other parts of the West Siberian megabasin. It is also important that the inflows of oil and gas in many fields of Yamal were obtained from complexes of the Paleozoic folded basement. Crystalline basements of oil and gas provinces remain one of the only promising provinces but insufficiently studied. It is the granitoid of the basements that are most promising for oil and gas exploration [1].

Monazite is a widespread accessory mineral of granitoids but it is almost not studied mainly due to the well-functioning system for determining the age of accessory zircons using isotope mass spectrometers, and zircon itself is quite easy to

isolate from granites. At the same time, monazite, in comparison with uraninite (the first dating of which was carried out in 1911 by A. Holmes), has recently began to be used as a mineral-geochronometer. At first, monazite was dated by various isotopic methods [2, 3], and with the development of microprobe technologies in 1991, works on chemical microprobe dating of accessory monazites in granites began to appear [4, 5]. A similar study was carried out for accessory monazite from granites of the crystalline basement of the Verkhnerechenskaya oil prospecting area (Yamal Peninsula).

Geological position of the research object

Granitoids in the crystalline basement of the South Yamal were discovered just in one place within the Verkhnerechenskaya oil and gas exploration area. The area itself (well no. 1 located there) is located next to the well-known Novoportovskoye oil and gas field, about 50 km from the latter to the southwest and about 225 km northeast of the city of Salekhard (the administrative center of the Yamal-Nenets Autonomous Okrug).

Verkhnerechenskaya well no. 1 (Fig. 1) exposed the crystalline basement, namely fresh granites, in the depth interval 1748–2034 m. No deeper drilling was carried out; intrusive

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Figure 1. Layout of wells that exposed the crystalline basement on the Yamal Peninsula; well numbers. 215 – Novoportovskaya; 300 – Zapadno-Yarotinskaya; 1–3 – Verkhneretchenskaya; 11 – Vostochno-Bovanenkovskaya; 45 – Syunai-Salinskaya; 114 – Bovanenkovskaya.

Рисунок 1. Схема расположения скважин, вскрывших кристаллический фундамент на полуострове Ямал; номера скважин: 215 – Новопортковская, 300 – Западно-Яротинская, 1–3 – Верхнереченские, 11 – Восточно-Бованенковская, 45 – Сюнай-Салинская, 114 – Бованенковская.

rocks are overlain by the Middle-Upper Jurassic sedimentary cover higher along the well. Granites throughout the section of the well are represented by homogeneous light gray fine-grained varieties of biotite-quartz-feldspar composition. It was found that they belong to monzoleucogranites, which formed on a sedimentary substrate (S-type granites), most likely, in a late orogenic environment. The crystallization time of subalkaline leucogranites in the basement of the Verkhneretchenskaya area was estimated by the method of chemical Th–U–Pb dating using accessory uraninite and monazite, 259.2 ± 3.6 Ma [6] and isotopic U–Pb dating using zircon, 254.0 ± 3.0 million years [7, 8]. Despite a detailed study of the material composition of monzoleucogranites [9, 10], separately accessory monazite was almost not described, even though it was a component of the biminerall isochron and the rock dating was carried out.

Methods of research

A quantitative analysis of the chemical composition of monazite was carried out using the CAMECA SX 100 electron

probe microanalyzer (Institute of Geology and Geochemistry of the Ural Branch of the Russian Academy of Sciences, Ekaterinburg). Polished sections were made from granite samples, then they were sprayed with a thin layer of carbon. Measurement conditions: accelerating voltage is 15 kV, is 250 nA, electron beam diameter is 2 μ m. The pressure in the sample chamber is 2×10^{-4} Pa. The spectra were obtained with the help of inclined wave spectrometers, the intensity was measured using analytical lines: Th *Ma*, U *Mb*, Pb *Ma*, Y *La*, Si *Ka*, Ca *Ka*, P *Ka*, Ce *La*, La *La*, Pr *Lb*, Nd *La*, Sm *Lb*, Dy *La*, Gd *Lb*. Standard samples used for calibration: thorium and uranium oxides, diopside, synthetic REE phosphates, $Pb_2P_2O_7$. When performing quantitative analysis, the time for measuring the intensity at the peak (thorium – 60 s, lead – 120 s, uranium – 30 s, silicon, yttrium, and calcium – 20 s, and 10 s for the remaining elements) was twice as long as the background measurement time. The following detection limits were achieved for monazite: Th – 130, U – 60, and Pb – 53 ppm. The oxygen content was determined on the assumption of the stoichiometry of the composition. The theoretical and practical substantiation of the method of chemical dating using X-ray spectral microprobe analysis is given in numerous publications on this topic [11, 12], including the group of authors [13, 14]. The main condition of this method: in the course of evolution, the mineral did not lose radiogenic lead (i. e., the Th–U–Pb system was closed), all the lead in the mineral was formed due to the decay of thorium and uranium.

Research results and discussion

Monazite-(Ce) forms an abundant accessory inclusion in granitoid and is much more common than zircon and uraninite [15]. Usually it tends to accumulate dark-colored minerals (mica and chlorite replacing it), where it composes well-formed individuals of hexagonal cross-section, up to 100 microns in elongation. In the BSE image mode, monazite differs well from other accessory minerals, having the brightest white color and distinct zoning, expressed by different contents of thorium (Fig. 2). Judging by zoning, the crystals underwent three growth phases with a gradual increase in the thorium content. Interestingly, monazite in the form of inclusions was also noted in zircon [7, 8]; it crystallized simultaneously with zirconium silicate.

The chemical composition of monazite varies considerably (see table). According to the data of all the above 45 analyzes, we can say that all grains of monazite in the rock are represented by the cerium variety (since cerium significantly predominates over other light rare earths) with a high content of thorium. Other impurities include the presence of significant concentrations of silica (SiO_2 up to 3.5 wt.%), yttrium (Y_2O_3 up to 1.8 wt.%) and calcium (CaO up to 1.2 wt.%). The content of radiogenic components varies quite significantly (in wt.%): ThO_2 – 5.37–16.31, UO_2 – 0.40–0.81, PbO – 0.08–0.19.

It is known that for thorium and uranium impurities in monazite, a hattonite ($Th^{4+}(U^{4+}) + Si^{4+} \rightarrow REE^{3+} + P^{5+}$) and/or cheralite ($Th^{4+}(U^{4+}) + Ca^{2+}(Sr^{2+}, Ba^{2+}, Pb^{2+}) \rightarrow 2REE^{3+}$) type of isomorphism. From the available analyzes of the chemical composition of monazite, it can be argued that both types of isomorphism are realized in the phosphate matrix. In this case, recrystallization of monazite, diffusion losses of radiogenic lead can lead to a violation of the isolation of its U–Th–Pb system.

Figure 2. Zonal monazite grains in monzoleucogranite of the Verkhnerenchenskaya area. CAMECA SX 100, BSE-image.
Рисунок 2. Зональные зерна монацита в монцолейкограните Верхнереченской площади. CAMECA SX 100, BSE-изображение.

Figure 3. Variations in the values of Th-U-Pb-ages and their weighted average for 45 point determinations in monazite from the Verkhnerenchensk granites.

Рисунок 3. Вариации значений Th-U-Pb-возрастов и их средневзвешенная величина для 45 точечных определений в монаците из верхнереченских гранитов.

Figure 4. Isochron of ThO_2^* -PbO based on results of monazite analyzes. Here $\text{ThO}_2^* = (\text{ThO}_2 + \text{UO}_2^{\text{экв}})$, where $\text{UO}_2^{\text{экв}}$ – the uranium content converted to the equivalent thorium content, capable of producing the same amount of Pb during the lifetime of the system with the same U-Pb and Th-Pb age values.

Рисунок 4. Изохрона ThO_2^* -PbO по результатам анализов монацита. Здесь $\text{ThO}_2^* = (\text{ThO}_2 + \text{UO}_2^{\text{экв}})$, где $\text{UO}_2^{\text{экв}}$ – содержание урана, пересчитанное в эквивалентное содержание тория, способное произвести то же количество Pb за время жизни системы при равенстве U-Pb и Th-Pb-значений возраста.

Chemical composition of monazite from granites of the South Yamal, wt. %.
Химический состав монацита из гранитов Южного Ямала, мас. %.

Ox- ides	Analysis number																						
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23
P ₂ O ₅	27.30	26.61	27.90	27.23	26.16	25.89	28.88	29.25	29.89	28.60	27.98	29.38	28.18	24.39	26.87	28.00	25.63	28.33	26.17	27.93	27.85	26.39	29.88
ThO ₂	11.20	13.38	9.62	9.21	11.85	12.70	7.24	8.81	6.93	8.50	8.52	6.76	7.87	16.31	8.68	7.87	15.13	7.29	10.91	7.61	8.24	11.61	5.37
UO ₂	0.66	0.76	0.64	0.49	0.58	0.60	0.76	0.54	0.46	0.44	0.42	0.81	0.44	0.74	0.53	0.47	0.67	0.40	0.60	0.45	0.49	0.55	0.64
SiO ₂	2.27	2.69	1.87	1.91	2.46	2.61	1.08	0.92	0.57	1.37	1.68	0.94	1.56	3.50	2.02	1.59	3.13	1.41	2.41	1.72	1.83	2.45	0.92
Ce ₂ O ₃	26.84	25.81	27.46	27.09	25.80	25.79	26.86	24.73	26.72	27.66	28.05	26.82	28.70	23.85	27.46	27.75	25.36	29.13	26.36	29.22	29.13	25.86	27.92
La ₂ O ₃	12.50	11.93	13.05	12.94	12.03	12.50	12.74	10.30	11.20	12.79	13.83	12.80	13.90	11.14	13.22	12.97	11.63	14.31	13.05	14.51	14.64	12.41	13.64
Nd ₂ O ₃	11.17	10.64	11.15	11.63	11.38	11.01	11.13	12.00	12.57	11.58	11.17	11.37	11.36	10.93	10.88	11.57	10.87	11.44	11.00	10.40	10.41	11.16	11.26
Pr ₂ O ₃	2.87	2.88	2.92	2.89	2.94	2.78	2.93	2.96	3.26	3.17	3.08	2.95	3.08	2.76	2.88	2.96	2.77	2.88	2.86	2.90	2.97	2.86	2.98
Sm ₂ O ₃	1.72	1.64	1.60	1.73	1.82	1.55	1.85	2.56	2.46	1.73	1.53	2.11	1.53	1.69	1.58	1.75	1.68	1.58	1.46	1.41	1.36	1.64	1.69
Gd ₂ O ₃	1.20	1.15	1.13	1.24	1.29	1.01	1.45	1.95	1.71	1.20	0.96	1.52	1.08	1.15	1.14	1.30	1.16	1.05	0.95	0.90	0.92	1.18	1.50
Dy ₂ O ₃	0.31	0.32	0.41	0.32	0.32	0.33	0.57	0.66	0.50	0.34	0.23	0.55	0.26	0.27	0.29	0.41	0.31	0.26	0.28	0.27	0.20	0.33	0.57
Y ₂ O ₃	1.28	1.33	1.36	1.32	1.33	1.07	2.31	2.60	1.80	1.13	1.02	2.57	1.06	1.16	1.16	1.45	1.22	1.11	1.03	0.91	0.94	1.31	2.24
PbO	0.15	0.17	0.12	0.11	0.14	0.16	0.10	0.13	0.10	0.10	0.10	0.10	0.10	0.19	0.11	0.10	0.18	0.10	0.14	0.09	0.11	0.16	0.08
CaO	0.42	0.46	0.47	0.44	0.46	0.51	0.85	1.27	1.16	0.71	0.48	0.95	0.37	0.42	0.38	0.43	0.45	0.37	0.47	0.43	0.46	1.26	0.65
Total	99.90	99.77	99.70	98.56	98.55	98.51	98.74	98.69	99.32	99.32	99.06	99.65	99.47	98.49	97.22	98.61	100.21	99.65	97.68	98.74	99.55	99.14	99.34
Oxides	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	45
P ₂ O ₅	27.29	27.22	26.59	26.61	27.10	26.17	26.41	25.92	25.37	27.87	28.17	27.50	25.32	24.90	28.49	29.43	28.65	28.48	28.47	28.88	27.40	25.64	25.64
ThO ₂	12.84	9.41	8.81	9.82	8.78	9.43	11.73	11.69	12.47	7.61	7.43	9.48	14.11	11.27	4.74	6.07	5.35	8.01	7.65	7.75	8.18	13.09	13.09
UO ₂	0.59	0.44	0.45	0.44	0.55	0.64	0.59	0.63	0.64	0.43	0.42	0.62	0.74	0.58	0.32	1.70	0.34	0.40	0.47	0.65	0.39	0.69	0.69
SiO ₂	2.69	1.84	1.90	2.09	1.67	2.02	2.33	2.40	2.64	1.44	1.38	1.83	2.98	2.61	0.69	0.36	0.72	0.85	1.20	3.36	1.65	2.70	2.70
Ce ₂ O ₃	27.22	28.17	27.44	27.00	27.89	26.06	27.37	26.59	25.93	28.65	28.44	27.62	26.08	25.64	29.53	26.08	29.11	28.35	26.39	26.99	28.55	25.61	25.61
La ₂ O ₃	12.28	13.79	13.43	13.12	13.62	12.05	12.86	12.65	12.59	14.21	14.19	13.39	12.09	12.67	15.16	11.64	14.42	13.31	11.46	11.79	14.19	11.64	11.64
Nd ₂ O ₃	11.12	11.26	11.58	11.61	11.36	11.88	11.13	10.95	11.11	11.34	11.33	11.04	10.88	10.95	11.86	11.68	11.71	11.62	12.37	10.41	11.33	11.26	11.26
Pr ₂ O ₃	2.87	3.05	3.17	3.05	2.99	3.09	3.05	2.85	2.73	3.11	3.07	3.02	2.72	2.90	3.05	2.93	3.29	3.11	3.05	2.65	3.15	2.86	2.86
Sm ₂ O ₃	1.62	1.64	1.55	1.71	1.64	1.93	1.59	1.59	1.61	1.54	1.58	1.77	1.75	1.45	1.41	2.10	1.68	1.61	2.24	1.85	1.61	1.64	1.64
Gd ₂ O ₃	1.16	1.00	1.17	1.21	1.02	1.43	1.22	1.04	1.20	1.00	1.21	1.17	1.21	1.12	0.83	1.68	1.15	0.89	1.68	1.27	1.17	1.21	1.21
Dy ₂ O ₃	0.29	0.32	0.31	0.31	0.35	0.40	0.31	0.29	0.24	0.32	0.35	0.34	0.30	0.20	0.10	0.53	0.17	0.19	0.52	0.29	0.25	0.32	0.32
Y ₂ O ₃	1.05	1.02	1.06	1.11	1.18	1.47	1.20	1.23	1.25	1.29	1.30	1.40	1.28	1.01	0.72	2.23	0.91	0.92	1.95	1.23	0.96	1.30	1.30
PbO	0.17	0.13	0.12	0.13	0.12	0.14	0.13	0.14	0.15	0.10	0.11	0.13	0.18	0.14	0.06	0.12	0.06	0.10	0.09	0.10	0.11	0.17	0.17
CaO	0.37	0.36	0.33	0.36	0.42	0.47	0.43	0.40	0.42	0.47	0.48	0.51	0.41	0.45	0.79	1.36	0.76	1.10	1.04	1.21	0.37	0.43	0.43
Total	101.57	99.66	97.92	98.57	98.70	97.19	100.35	98.39	98.36	99.39	99.45	99.80	100.04	95.91	97.73	97.92	98.32	98.93	98.58	98.43	99.31	98.56	98.56

Note: Institute of Geology and Geochemistry of the Ural Branch of the Russian Academy of Sciences, CAMECA SX 100 microanalyzer, analyst V. V. Khiller.

In the well-known work [16], the parameter $\beta = (\text{Si} + \text{Ca})/(\text{Th} + \text{U} + \text{Pb})$, which characterizes the degree of charge compensation of Th^{4+} (U^{4+}) impurities, was considered as an indicator of isolation of the Th–U–Pb monazite system: it is considered to be closed when it is close to the unit. For the Verkhnerchensk monazite, the parameter $\beta = 0.97\text{--}1.03$, which indicates the isolation of the system and, therefore, the possibility of correct estimation of the age of the mineral.

The isolated system, sensitive lead content and a significant dispersion of thorium (and uranium, to a lesser extent) contents make it possible to use this accessory monazite as a geochronometer mineral. The values of the point U–Th–Pb ages of monazite lie in a wide range of 230–286 Ma and together give a weighted average age of 256 ± 10 Ma with MSWD = 0.15 (Fig. 3).

When plotting the $\text{ThO}_2^*\text{--PbO}$ dependence from a set of analysis points, accessory monazites fall on one straight line (or isochron according to [16]), which indicates their one-time formation. Calculating the age from the angle of isochron

inclination gives the dating of 254 ± 19 Ma, MSWD = 0.28, probability = 1.000 (Fig. 4). The content of non-radiogenic lead in monazite, calculated from the intersection of the Th*–Pb isochrone with the PbO-axis, is less than 0.005 wt.%, which is comparable to the limit of its detection in the mineral and therefore did not affect the age calculation. In general, the results obtained for monazite are very close to the U–Pb isotope dating for zircon from the same rock, 254 ± 3 Ma [8, 17].

Conclusions

Thus, we have studied in detail accessory monazite from monzoleucogranites of the crystalline basement of the South Yamal. New data on the chemical composition of the mineral have been obtained, and the Late Permian age of granitoids has been determined by microprobe dating. The values of the point U–Th–Pb ages of monazite together give a weighted average age of 256 ± 10 Ma (MSWD = 0.15) and an isochron of 254 ± 19 Ma (MSWD = 0.28), which almost ideally coincides with the results of isotopic U–Pb zircon dating from the same rock, 254 ± 3 Ma.

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Химический состав и возраст монацита-(Ce) в гранитоидах кристаллического фундамента Южного Ямала

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Аннотация

Актуальность работы обусловлена необходимостью совершенствования метода химического датирования в применении к высокотермическим акцессорным минералам, которые сложно датировать изотопными методами исследования.

Цель работы: исследование химического состава акцессорного монацита из гранитоидов кристаллического фундамента Южного Ямала и определение его возраста.

Методология исследования: количественный анализ химического состава монацита 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 проводилось по аналитическим линиям: Th Ma, U Mb, Pb Ma, Y La, Si Ka, Ca Ka, P Ka, Ce La, La La, Pr Lb, Nd La, Sm Lb, Dy La, Gd Lb. Расчет возраста проводился по известным методикам зарубежных авторов в дополнение к собственным работам.

Результаты. Химический состав монацита позволяет относить его к цериевой разновидности, содержание радиогенных компонентов сильно варьирует (в мас.%): ThO₂ – 5,37–16,31, UO₂ – 0,40–0,81, PbO – 0,08–0,19. Из примесей отмечаются существенные концентрации SiO₂ (до 3,5 мас.%), Y₂O₃ (до 1,8 мас.%) и CaO (до 1,2 мас.%). Получается, что в монаците реализуются хаттонитовый (Th⁴⁺(U⁴⁺)+Si⁴⁺ → REE³⁺+P⁵⁺) и чералитовый (Th⁴⁺(U⁴⁺)+Ca²⁺(Sr²⁺,Ba²⁺,Pb²⁺) → 2REE³⁺) типы изоморфизма. Приличное содержание свинца и высокая кристалличность вещества позволяют использовать данный минерал в качестве минерала-геохронометра.

Выводы. Получены новые данные по химическому составу монацита и методом микрозондового датирования установлен позднепермский возраст гранитоидов. Значения точечных U–Th–Pb-возрастов монацита в совокупности дают средневзвешенный возраст 256 ± 10 млн лет (СКВО = 0,15) и изохрону в 254 ± 19 млн лет (СКВО = 0,28), что практически идеально сходится с результатами изотопного U–Pb-датирования циркона из этой же породы, 254 ± 3 млн лет.

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

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