MINERALOGIA POLONICA
Vol. 9, No 1 — 1978

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THORIUM-BEARING URANINITE (BRÖGGERITE) FROM THE KARKONOSZE GRANITE

UKD 549.514.87:546.841:547.916:549.623.54:552:321.1(438-14:234.572)

Abstract. Automorphic thorium-bearing uraninite (bröggerite) has been found to occur commonly in association with biotite in the Karkonosze granites. It is coated with a metasomatic rim containing up to some wt. % U. The age of bröggerite, defined from the Pb content determined on electron microprobe, is identical with the K-Ar arge of the co-occurring biotite.

INTRODUCTION

A high uranium content in the granites of the Karkonosze massif (Jeliński 1965), averaging 12.7 ppm (1.9—62.5 ppm) in surface granites, suggests the possibility of occurrence of that element in the form of uranium minerals, as is the case in the uranium-bearing granites of the Franch Central Massif and Vendée (Coppens 1973). Since under the conditions of surface weathering uraninite is decomposed and uranium is either leached or changes its form of occurrence (disseminated in intergranular substance, disseminated in secondary minerals resulting from decomposition of rock--forming minerals, etc.), granite samples from Karkonosze IG-1 borehole located near Jakuszyce, west of Szklarska Poręba, attraced the authors attention. The granites from that borehole show below the weathering zone a still higher uranium content (19.8 ppm on the average, maximum content 90 ppm) that the surface granites. It has been assumed, therefore, that they may contain primary uranium minerals. In fact, microscopic studies in reflected light have revealed that the high-radioactivity centres localized using alpha-radioautography are microcrystals of a mineral with the habit and optical features of uraninite (Lis, Sylwestrzak 1977). As the presence of uraninite in the Karkonosze granites is of great metallogenic significance, this mineral was subjected to further investigations on electron microprobe.

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MODE OF OCCURRENCE OF URANINITE

Uraninite forms crystals exhibiting a hexagonal habit, with the edge length varying from a dozen or so to several dozen microns (Phots. 1—3). Uraninite microcrystals occur near biotite aggregates or on the edges of biotite plates and on its cleavage planes. Some microcrystals are coated with a thin (up to a dozen or so microns) rim of yellowish substance showing lower radioactivity. Some have also a very thin made up of pyrite which corrodes the surface of uraninite and penetrates inside. Small, roundish pyrite inclusions have also been noted in the outer parts of uraninite grains.

The distribution of uraninite microcrystals in the rock is most irregular. They are absent altogether in some thin sections while in others 20 microcrystals have been found in an area of 3 cm^2. The presence of uraninite microcrystals accounts for the high total content of uranium in the Karkonosze granites.

In reflected light, the uraninite is grey and isotropic, showing low reflectance.

The occurrence of uraninite in the vicinity of biotite provides evidence of the role of the latter mineral in the precipitation of uranium oxide. As a result of oxidation-reduction reaction between Fe^{2+} from biotite and U^{6-}, iron was presumably partly oxidized and uranium oxide precipitated. Somewhat later pyrite formed, with tetravalent uranium playing the role of reducer.

THE CHEMICAL COMPOSITION OF URANINITE

Analyses were carried out on a granite sample with a U content of 55 ppm, derived from Karkonosze IG-1 borehole from a depth of 39.2 m.

The following procedure was adopted: a dozen or so thin sections for investigations in transmitted light were prepared from the rock sample. Alpha-radioautography was used to localize uraninite microcrystals on the thin sections, and then fragments containing microcrystals were cut out. Each fragment was mounted on the microprobe stage and polished with diamond pastes.

Electron microprobe analysis of the chemical composition was made on theree uraninite microcrystals (Table 1). The instrument (an ARL SEMQ X-ray microanalyzer) was operated at an accelerating voltage of 20 kV and a probe current of 150 mA. Metalic U, ThO_2, Si, Pbs were used as standards and $U_{M\alpha}$, $Th_{M\alpha}$, $Pb_{M\alpha}$, $Si_{K\alpha}$ lines were utilized. Counting time was 100 sec. When calculating corrections, the amount of oxygen was assumed each time to be a complement to 100% of the sum of cations. This procedure was adopted because of poor reproducibility of determinations of light elements, as well as due to the lack of the proper standard, U_3O_8. Absorption correction was based on the generally accepted principles (Adler 1966) whereas the atomic number correction was calculated using Philibert's and Tixier's formula (1968).

As appears from electron microprobe analysis, the chemical composition of the microcrystals is very homogeneous. The uniform distribution

Table 1

The chemical composition of uraninite and coexisting minerals in wt. %

Sample No	U	Th	Pb	Si	Chemical formula
97/1	77.00	3.49	3.22±0.08	⩽0.03	$U_{2.74}Th_{0.13}Pb_{0.13}O_{8.63}$
98/1	77.64	3.69	3.18±0.08	⩽0.03	$U_{2.73}Th_{0.14}Pb_{0.13}O_{8.75}$
99/1	77.06	3.71	3.11±0.06	⩽0.03	$U_{2.73}Th_{0.14}Pb_{0.13}O_{8.52}$
*97/2**	4.46	0.24	⩽0.11	1.90	?
*97/5**	3.10	⩽0.14	⩽0.13	8.16	?

Analyst: H. Kucha.
⩽ — microprobe detectability limit.
* — as the compound failed to be identified, no corrections of the content of elements were made.

of the principal elements, i.e. U, Th and radiogenic Pb, has also been confirmed by electron microprobe studies (Phot. 3).

The proportions between the main components indicate that the mineral studied is mixture of oxides, UO_2 and UO_3, with an average formula close to U_3O_8. Uranium is partly replaced by thorium and radiogenic lead. A high thorium content (3.97—4.22 ThO_2), characteristic of pegmatite uraninites, defines the mineral studied as thorium-bearing uraninite — *bröggerite*.

SECONDARY ALTERATIONS ROUND MICROCRYSTALS

The bröggerite microcrystal embedded in a biotite plate near its edge (sample *97*) is coated with a thin layer (about 12 microns) of yellowish substance that formed as a result of destructive action of alpha-radiation emitted by radioactive elements (Phots. 1, 2, 3). Two electron microprobe analyses (Table 1 *97/2*, *97/5*) have revealed that the substance in question is enriched in U, Th and Pb but markedly deficient in silica. The presence of the rim is characteristically reflected in the microprobe distribution of elements (Phot. 3), specifically U and Si.

The formation of a reaction rim round microcrystals is due to that the bombardment with alpha-particles results in the breakdown of the structure of the surrounding mineral and oxidation of some elements, as well as increases the mobility of the components making up the rim. A part of these components migrate and metasomatic replacement can take place. These phenomena are most intense not in the immediate vicinity of the radioactive mineral but at a certain distance from it, which proves that the alpha-particle has a greater destructive effect when losing its energy. The mineralogical nature of the substance making up the rim has not been defined since this would require a complete analysis of its composition. It is feasible that it consists of hydrated compounds high in uranium.

The phenomena involving the breakdown of the structure and the formation of reaction rims near the microcrystals studied are consistent with those described by Hamilton (1958).

CONCLUSIONS

1. Uraninite appearing in the form of microcrystals in the Karkonosze granites is a thorium-bearing variety — *bröggerite*. It is worth noting that uraninite microcrystals found in the uranium-bearing granites of the French Central Massif belong, as a rule, to the same variety.
2. High uranium content in the Karkonosze granites is due to the presence of uraninite (bröggerite) microcrystals.
3. The presence of uraninite microcrystals implies that the Karkonosze granite massif is a metallogenic environment favourable to the formation of uranium deposits.
4. The age of uraninite (bröggerite) microcrystals determined by U-Th-Pb method on the basis of electron microprobe analysis (Lis, Sylwestrzak, Kucha — in press) is 299.8 MA (average from 3 analyses), being in good agreement with the K-Ar age of biotite containing these microcrystals — 301.8 MA. This age testifies to simultaneous crystallization of the two minerals.

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TORONOŚNY URANINIT (BRÖGGERYT) Z GRANITU KARKONOSZY

Streszczenie

Granity o wysokiej zawartości uranu cechują się zwykle obecnością mikrowrostków uraninitu (Coppens 1973). Niedawne badania autorów (Lis, Sylwestrzak 1977) wykazały, że mikrokryształy uraninitu występują także w granitach Karkonoszy. W próbkach granitu pobranych poniżej strefy wietrzenia w otworze Karkonosze IG-1 koło Jakuszyc stwierdzono obecność wrostków uraninitu o pokroju sześcianu. Ich wymiary wahają się w granicach od kilkunastu do kilkudziesięciu µm (fot. 1—3). Wrostki uraninitu występują w pobliżu skupień biotytu, na jego brzegach i płaszczyznach łupliwości. Są one niekiedy otoczone obwódką pirytu, który koroduje uraninit oraz cienką otoczką substancji o niskiej zdolności odbicia światła. Cechy optyczne uraninitu w świetle odbitym są typowe dla tego minerału. Rozmieszczenie mikrowrostków w skale jest nieregularne.

Badania za pomocą mikroanalizatora rentgenowskiego wykazały, że rozmieszczenie głównych pierwiastków w omawianym minerale jest jednorodne (fot. 3). Jego skład chemiczny jest zbliżony do U_3O_8, jednak wysoka zawartość Th pozwala określić badany minerał jako uraninit toronośny — *bröggeryt*.

Występującą w otoczeniu uraninitu substancję należy uznać za produkt metasomatycznego przeobrażenia w strefie reakcyjnej (fot. 3), w której struktura otaczającego minerału została zaburzona przez bombardowanie cząstkami α.

Przeprowadzone na podstawie wyników analizy w mikroobszarze oznaczenia wieku metodą U-Th-Pb wykazały (Lis, Sylwestrzak, Kucha — w druku), że uraninit powstał jednocześnie z zawierającym go biotytem.

OBJAŚNIENIA FOTOGRAFII

Fot. 1. Mikrofotografia bröggerytu (*br*) otoczonego strefą metasomatycznych przeobrażeń (*a*). Na kontakcie tej strefy z bröggerytem występuje piryt (*p*). Światło odbite. Pow. × 1000

Fot. 2. Mikrofotografia bröggerytu (*br*) otoczonego przez strefę zmienioną wskutek bombardowania cząstkami α (*a*). Na kontakcie bröggerytu i strefy zmienionej występuje piryt (*p*). Światło odbite. Pow. × 1000

Fot. 3. Ziarno bröggerytu otoczone przez strefę zmienioną wskutek bombardowania cząstkami α
a — EAI — elektronowy obraz absorpcyjny, *b* — obrazy rozmieszczenia U

Fot. 4. Ziarno bröggerytu otoczone przez strefę zmienioną wskutek bombardowania cząstkami α
a—*b* — obrazy rozmieszczenia Th i Pb
Na obrazie rozmieszczenia Pb widoczny wrostek galeny radiogenicznej. Pow. × 850

Fot. 5. Ziarno bröggerytu otoczone przez strefę zmienioną wskutek bombardowania cząstkami α
Obraz rozmieszczenia Si

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ТОРИЕНОСНЫЙ УРАНИНИТ (БРЕГГЕРИТ) ИЗ ГРАНИТОВ КАРКОНОШИ

Резюме

Граниты с высоким содержанием урана характеризуются присутствием микровключений уранинита (Коппенс, 1973). Недавние исследования (Лис, Сыльвестржак, 1977), показали, что микрокристаллы уранинита присутствуют тоже в гранитах из Карконоши. В образцах гранита, взятых ниже зоны выветривания в буровой скважине Карконоше ИГ-1 вблизи Якушиц, было обнаружено присутствие включений уранинита кубического облика. Их размеры колеблются от 10 до нескольких де-

сятков микронов (фото 1—3). Включения уранинита присутствуют вблизи агрегатов биотита — на его краях и поверхностях спайности. Иногда агрегаты окружены оболочкой из пирита, который корродирует уранинита, и тонкой оболочкой вещества с низкой способностью отражения света. Оптические свойства уранинита в отряжённом свете типичны для этого минерала. Расположение микровключений в породе очень нерегулярно.

Исследования при помощи рентгеновского микроанализатора показали, что расположение основных элементов в вышеупомянутом минерале является однородным (фото 3). Его химический состав сближается к U_3O_8, но высокое содержание Th разрешает определить исследуемый минерал как ториеносный уранинит — бреггерит.

Присутствующее вокруг уранинита вещество можно рассматривать как продукт метасоматического преобразования в зоне восстановления (фото 3), в которой структура окружающего минерала нарушилась из-за бомбардировки альфа-частицами.

Проведённые на основании результатов анализа в микрозоне определения по методу U-Th-Pb показали (Лис, Сыльвестржак, Куха — в печати, что уранинит образовался совместно с содержащим его биотитом.

ОБЪЯСНЕНИЯ К ФОТОГРАФИЯМ

Фото 1. Микрофотография бреггерита (*br*), окружённого зоной метасоматических преобразований (*a*). На стыках этой зоны с бреггеритом наблюдается присутствие пирита (*p*). Отражённый свет. Увеличение около × 1000

Фото 2. Микрофотография бреггерита (*br*), окружённого зоной измененной из-за бомбардировки α-частицами (*a*). На стыках бреггерита с измененной зоной наблюдается присутствие пирита (*p*). Отражённый свет. Увеличение около × 1000

Фото 3. Зерно бреггерита окруженное зоной измененной из-за бомбардировки α-частицами

a — электронный образ поглощения, *b* — образы расположения U

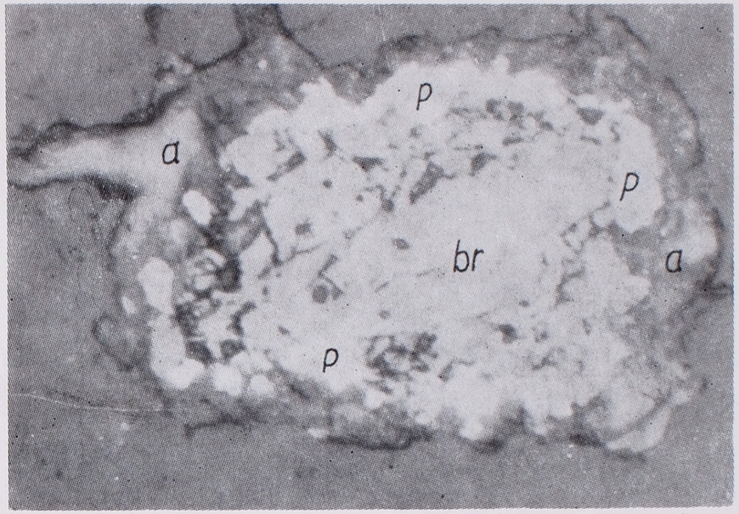
Фото 4. Зерно бреггерита окруженное зоной измененной из-за бомбардировки α-частицами

a—*b* — образы расположения Th и Pb.

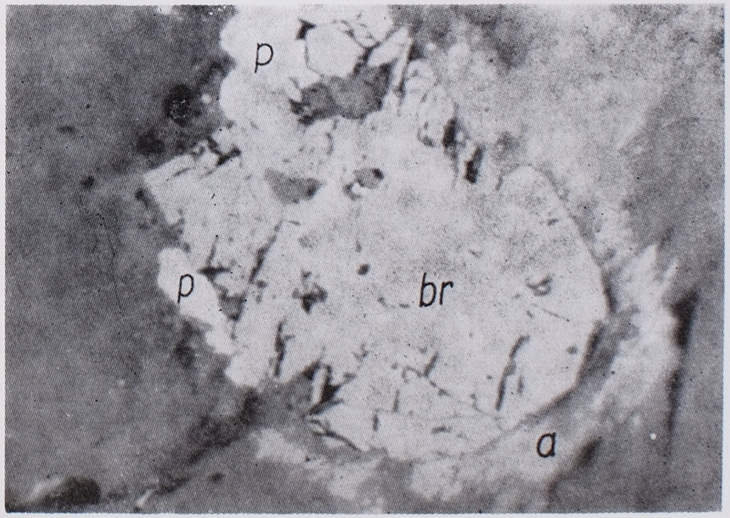
На образе расположения Pb видно включение радиогенного галенита. Увеличение × 850

Фото 5. Зерно бреггерита окруженное зоной измененной из-за бомбардировки α-частицами

Образы расположения Si. Увеличение × 850

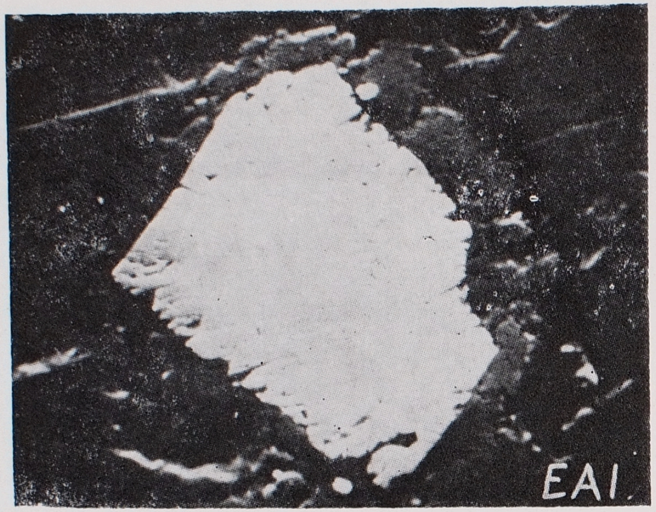


Phot. 1. Bröggerite (*br*) surrounded by zone of metasomatic alterations (*a*). On the contact with bröggerite occurs pyrite (*p*). Reflected light. Magn. × 1000

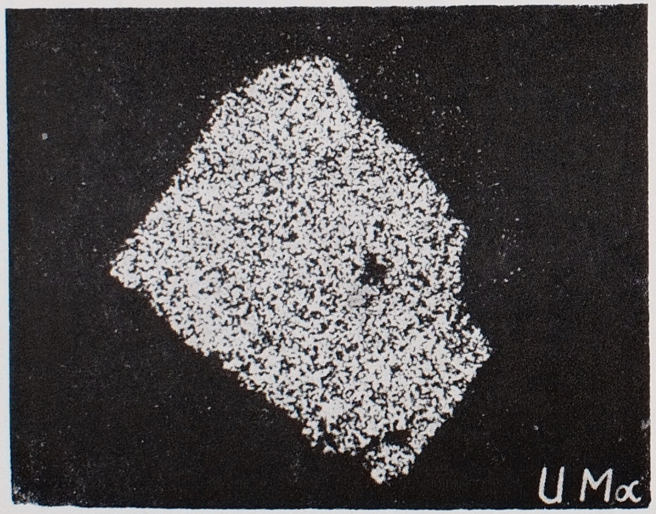


Phot. 2. Bröggerite (*br*) surrounded by zone altered due to bombardment α particles (*a*). On the contact of bröggerite and altered zone occurs pyrite (*p*). Reflected light. Magn. × 1000

a



b

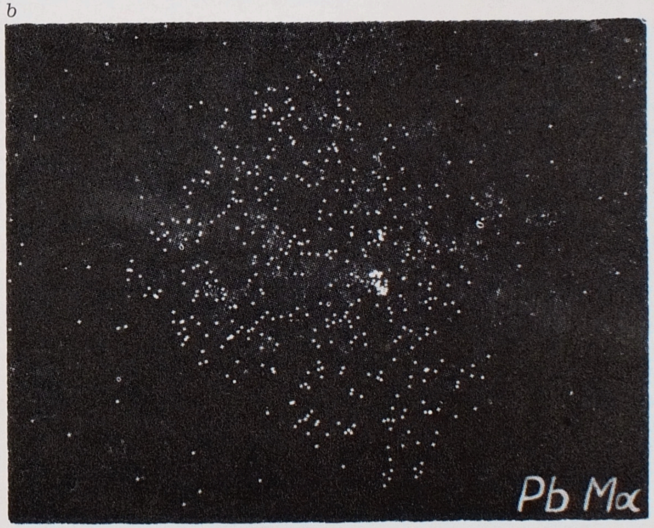


Phot. 3. Grain of bröggerite surrounded by altered zone due to bombardment α particles

a — EAI electron absorption image, b — images of U

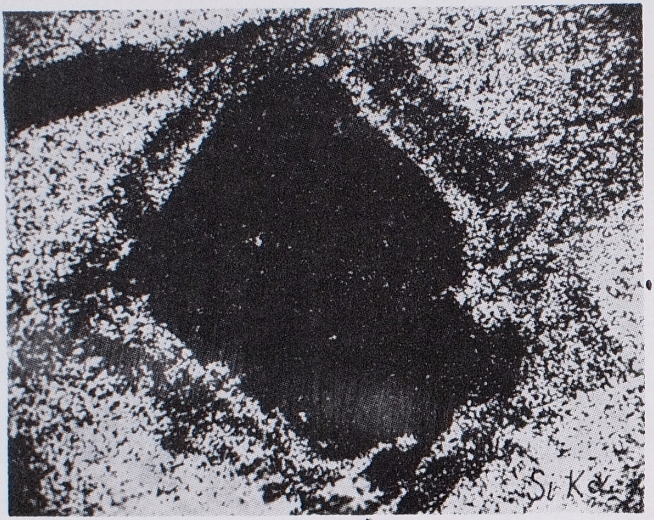
a

b



Phot. 4. Grain of bröggerite surrounded by altered zone due to bombardment α particles

a—b — images of Th, Pb distribution. On the image of Pb distribution an inclusion of radiogenic galena is visible. Magn. × 850



Phot. 5. Grain of bröggerite surrounded by altered zone due to bombardment α particles
Images of Si distribution. Magn. × 850