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THE APPLICATION OF ELECTRON MICROPROBE TO DATING OF U—Th—Pb URANINITE FROM THE KARKONOSZE GRANITES (LOWER SILESIA)

Abstract. Datings of uraninite microinclusions were carried out with the classical U—Th—Pb chemical method. Electron microprobe was used to determine the content of elements. The results obtained for three samples show insignificant scatter, and the average age of 299.8 million years is in accord with the age of biotite (301.8 mln years) in which the uraninite inclusions have been found. This shows that electron microprobe analysis is useful for geochronological studies.

INTRODUCTION

As a result of the development of mass spectrometry, the chemical U—Th—Pb dating of uranium and thorium minerals (Holmes 1911), resting on the assumption that all the lead present in a mineral is of radiogenic origin, was replaced by isotopic methods. However, the introduction of electron microprobe to these studies opened new prospects for the chemical dating of minerals. This applies primarily to uraninite and thorianite microinclusions present in some igneous and metamorphic rocks, as the small size of automorphic grains of these minerals can guarantee that lead contained in them is of radiogenic origin only.

The first datings of uraninite microinclusions with the aid of electron microprobe were carried out on uraninite from the Saint-Sylvestre uranium-bearing granites in the Central Massif (Barbier, Ranchin 1969) and the Mortagne granites in Vendée (Renard 1970).

URANINITE IN THE KARKONOSZE GRANITES

The Karkonosze granitoid massif forms the central part of a structure referred to as the Karkonosze-Izera block. The massif is built of coarse- and medium-grained porphyraceous biotite granites and equigranular biotite granites. From the petrochemical point of view, they are assigned to granodiorites and monzonitic granites (Borkowska 1966).

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Compared with the other Lower Silesian granitoid rocks, the Karkonosze granites show an exceptionally high uranium content (Jeliński 1965), averaging 12.7 ppm. The highest uranium content in surface samples is 62.5 ppm, running up to 90 ppm in core samples taken below the weathering zone.

The anomalously high uranium content in the Karkonosze granites suggests that uranium occurs here in the form of own minerals, as it does in some granitoid rocks of the Central Massif and Vendée (Coppens 1973).

Since uraninite is subject to oxidation in the weathering zone, and the released uranium is either leached or passes into other forms of occurrence, uraninite inclusions were sought in the granite samples from a borehole located to the west of Szklarska Poręba. The high-radioactive centres detected in thin sections by means of radio-autography of alpha particles proved, in fact, to be uraninite inclusions (Lis, Sylwestrzak 1977). Uraninite microcrystals have a cubic habit, sometimes slightly deformed. They vary from a dozen or so to several dozen μm in size, exceptionally attaining a size of 200 μm. All the observed inclusions occur in the vicinity of biotite concentrations or at the edges of biotite flakes. In reflected light uraninite is grey, isotropic. Some crystals are coated with a thin rim made up of pyrite which corrodes uraninite, penetrating inside the grains. The mode of occurrence and the habit typical of pegmatite uraninites show that the uraninite in question formed during the crystallization of the rock. Its occurrence in the marginal parts of biotite and in their vicinity indicates that this mineral played a significant role in the precipitation of uranium oxide. It is feasible that uranium oxide precipitated from the uranium-rich residual solutions as a result of the reaction between bivalent iron and hexavalent uranium.

CHEMICAL COMPOSITION AND AGE OF U—Th—Pb URANINITE

Chemical composition was determined for three uraninite microcrystals from a granite sample taken from the Karkonosze IG 1 borehole at a depth of 39.2 m. Uranium content in the sample was 55 ppm.

Electron microprobe analysis was carried out by one of the authors (Kucha) on an ARL SEMQ X-ray microanalyser, using metallic U, ThO_2, PbS and Si as standards and the following spectral lines: UM_α, ThM_α, PbM_α, SiK_α. The instrument settings were: accelerating voltage 20 kV, probe current 150 *n*A, counting time 100 sec. When calculating corrections, the amount of oxygen was assumed each time to be the complement to 100% of the sum of cations. This procedure was adopted because of the poor reproducibility of the determinations of light elements and the lack of a proper U_3O_8 standard. The generally accepted procedures were adopted to calculate corrections for the absorption of radiation (Adler 1966) and the atomic number difference (Philibert, Tixier 1968).

Since the accuracy of chemical datings is mainly affected by errors of determination of lead content, the determinations of these elements were made with the highest possible precision. The results listed in Table 1 show that the mineral in question is a mixture of UO_2 and UO_3 oxides with a molecular formula close to U_3O_8. In this mineral uranium is partly replaced by lead and thorium. Due to the high thorium content (over 3%), the mineral was defined as thorium-bearing uraninite — bröggerite (Kucha, Lis, Sylwestrzak 1978).

From the electron microprobe analysis it appears that U, Th and Pb are uniformly distributed on the surface of the grain sections studied. This substantiates the hypothesis that lead is of radiogenic origin.

To calculate the age of uraninite from its chemical formula, the following relation was used (Holmes, Lawson 1927):

$$\text{Age} = \frac{\text{Pb}}{\text{U}+0.38\ \text{Th}}\ 7400\ \text{MA}$$

where U, Th and Pb are expressed in weight % (Table 1).

Table 1

Chemical composition and age of U—Th—Pb uraninite

Sample No	U, %	Th, %	Pb, %	Age 10^6 years
97,1	77.0	3 49	3.22±0.08	304.2
98,1	76.64	3.69	3.18±0.08	301.5
99,1	77.06	3.71	3.11±0.06	293.7
				average 299.8

The isotopic age of biotite from the Karkonosze granites has been determined both by K—Ar and Rb—Sr datings. The former (Depciuch, Lis 1971) established the average age of 301.8 million years (307, 306, 305, 304, 304, 303, 303, 303, 302, 299, 296, 296, 295 mln years), while Rb—Sr dating (Przewłocki *et al.* 1962) indicated an age of 292 million years.

CONCLUSIONS

The U—Th—Pb dating of uraninite with the use of electron microprobe is in excellent agreement with the K—Ar and Rb—Sr datings of biotite in which uraninite occurs in the form of inclusions. This shows that the method of chemical dating combined with electron microprobe analysis is useful for the dating of uraninite micro-inclusions. At the same time, some genetic conclusions emerge:

1. The similarity of the age of biotite and uraninite testifies to the simultaneous crystallization of these minerals.
2. From the moment of their formation, both uraninite and biotite constituted closed systems in the geochronological sense. This further substantiates the hypothesis that the Karkonosze granite is an igneous granite that formed in one geologic episode.

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ZASTOSOWANIE MIKROANALIZATORA RENTGENOWSKIEGO DO DATOWANIA U—Th—Pb URANINITU Z GRANITÓW KARKONOSZY (DOLNY ŚLĄSK)

Streszczenie

Przeprowadzono datowania mikrowrostków uraninitu—bröggerytu tkwiących w biotycie granitów Karkonoszy. Datowania wykonano klasyczną metodą U—Th—Pb. Oznaczenia U, Th i Pb wykonano w mikroobszarze przy zastosowaniu mikroanalizatora rentgenowskiego. Uzyskane wartości wieku cechują się nieznacznym rozrzutem, a średni wiek 299,8 milionów lat jest całkowicie zgodny z oznaczonym metodą K/Ar wiekiem biotytu (301,8).

Zbieżność wieku biotytu i uraninitu świadczy o równoczesnym powstawaniu tych minerałów. Świadczy to również, że uraninit i biotyt stanowiły od chwili swego powstania układy zamknięte w sensie geochronologicznym. Potwierdza to pogląd, że granit Karkonoszy jest granitem magmowym powstałym w jednym epizodzie geologicznym.

Uzyskane wyniki świadczą o przydatności zastosowania mikroanalizy rentgenowskiej do badań geochronologicznych.

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ПРИМЕНЕНИЕ РЕНТГЕНОВСКОГО МИКРОАНАЛИЗАТОРА ДЛЯ U—Th—Pb ДАТИРОВАНИЯ УРАНИНИТА ИЗ ГРАНИТОВ КАРКОНОШЕЙ (НИЖНЯЯ СИЛЕЗИЯ)

Резюме

Проводилась датировка микровключений уранинита—брёггерита в биотитах гранита Карконошей. Датирование проведено классическим U—Th—Pb методом. Определение содержания U, Th и Pb проводилось на микрозонде. Полученные величины возраста характеризуются незначительным разбросом, а средний возраст 299,8 миллионов лет полностью совпадает с определенным K/Ar возрастом биотита ($301,8 \cdot 10^6$).

Совпадение возрастов биотита и уранинита свидетельствует об одновременном образовании этих минералов. Это свидетельствует также о том, что уранинит и биотит от момента своего образования в геохронологическом смысле представляли собой закрытые системы. Это подтверждает взгляд, что гранит Карконошей представляет собой магматический гранит, образованный в одном геологическом эпизоде.

Полученные результаты свидетельствуют о пригодности применения микрозонда в геохронологических исследованиях.