

Uranium oxidation states in allanite, fergusonite and monazite of pegmatites from Finland

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Abstract

Ion exchange was used to determine uranium oxidation states in Ce-allanites, fergusonite and monazite contained in Svecofennian granitic pegmatites from Finland. In allanites, uranium was partly oxidized; $U(IV)/U_{tot}$ range from 0.33 to 0.43. The minerals were metamictic. Fergusonite was found to be totally oxidized, giving 0.06 for the $U(IV)/U_{tot}$. The low ratio is assumed to be due mainly to the hydration of fergusonite. Of the samples analysed, the fergusonite and one allanite were from the same pegmatite, but they gave a different degree of oxidation for uranium. This finding may reflect the greater stability of allanite as compared to fergusonite. The monazite was indirectly found to be in reduced form ($U(IV)/U_{tot}$ 0.93). The method applied probably dissolved only the soluble oxidized form of uranium.

Key words: uranium, oxidation states, allanite, fergusonite, monazite, pegmatite minerals, Finland.

Introduction

Several minerals incorporate minor amounts of uranium even though uranium is not an essential component of their mineral structure. Allanite, fergusonite and monazite are relatively common minerals in granitic pegmatites which contain high amounts of uranium and thorium. In these minerals, uranium is assumed to be originally in reduced form (SMITH JR 1984), whereas oxides of Ta and Nb are usually reported to be oxidized to some degree (SMITH Jr. 1984, BURNS and FINCH 1999).

There are several factors that can oxidize uranium in a mineral. Oxidation may simply result from the redox potential change in the environment of the uranium atom. Another possibility is that oxidation may be caused by the decay process itself, e.g. the alpha recoil effect or energetic beta decay. Physico-chemical attacks, radiation damage or cationic substitutions can alter the redox conditions favouring, the higher oxidation state and more soluble form of uranium. The recoil atom may enter into a different lattice position and cause more radiation damage via the formation of an oxidized uranium ion. In addition, the decay product lead can oxidize reduced uranium.

Fluids enriched in chlorides, fluorides, phosphates or carbonates may significantly solubilize and transport uranium. The circulation of fluids in the vicinity of radioactive minerals is enhanced by the microfissures resulting both by radiation damage and from the increase in volume following hydration of the mineral. These microfissures may represent preferential channels for circulating fluids. Cleavage and faults, too, can hasten the breakdown of the crystal structure, thereby creating conditions enabling the oxidation of uranium. The extent of alteration was revealed by HECHT and CUNEY (2000): 75% of the uranium bound to monazite is leached out during monazite alteration. The alteration of monazite (HECHT and CUNEY 2000, POITRASSON et al. 2000, MATHIEU et al. 2001, READ et al. 2002) and of allanite (WOOD et al. 1997, SMITH et al. 2002) has recently been studied. Current research interest has also focused on the weathering processes of allanite (MEINTZER and MITCHELL 1988, HARLAVAN and EREL 2002). In contrast to monazite and allanite, fergusonite has seldom been studied (POITRASSON et al. 1998). In metamictic process, the crystal state of mineral becomes partly or wholly amorphous due to the radiation; chemical bonds are broken and structural disorders are formed in the mineral. Metamictization depends on the age, composition and structure of the mineral. Some annealing of the radiation damage can also occur at the same time. The metamictic process leads to changes in the physical properties of the minerals, and may also effect the oxidation states of uranium. The structural collapse and substitutions of the mineral that are brought about by radiation damage affect allanite and fergusonite; only weak corrosion, but no metamictization, has been observed in natural monazites. Research into radiation damage has focused mainly on monazite (MELDRUM et al. 1998) and allanite (JANECZEK and EBY 1993), and also - but less extensively - on zircon.

The mechanisms of complex alteration and radiation damage processes occurring in uranium-bearing minerals, as well as their time-scale are only partly understood so far. The effects of redox conditions and different ligands on alteration mechanisms are difficult to assess, owing to the complexity of the laboratory experiments involved. Although there has been discussion of uranium leaching and possible oxidation (POITRASSON et al. 2000), no studies have been reported. The extent of uranium oxidation in these uranium-bearing minerals is still unclear. The aim of this work was to provide information on the uranium redox state in natural allanite, fergusonite and monazite of geologically the same age.

Samples: their geology and chemical composition

The three allanites, one fergusonite and one monazite investigated in this study came from four different pegmatite occurrences in southern Finland (Table 1). The mineral grains studied were taken from larger grains. One allanite and fergusonite were from the same location, the Pyörönmaa pegmatite. All pegmatites belong to the REE-type according to the classification of ČERNÝ (1998). The Luumäki and Brändö pegmatites are allanite-monazite pegmatites, while the Pyörönmaa and Varala pegmatites are gadolinite pegmatites. Scarce U-Pb data for monazites, columbites and tapiolites from Finland indicated an age of 1.86-1.79 Ga (ALVIOLA et al. 2001). The geological environment and mineralogical studies of these pegmatites have been described by LOKKA (1935,1950), VORMA et al. (1966), SIIVOLA (1975), VAASJOKI (1977), LAHTI (1996), LAHTI and KINNUNEN (1992). Based on these papers a brief description of the geological setting of the minerals studied follows.

Brändö is situated in the archipelago of southwestern Finland. Red porphyritic granite is the prevailing rock, but there are also rocks resembling rapakivi granites. The allanite occurs in pegmatites as lustrous, jet-black lumps. The interface between the mineral and its host is generally distinct. The allanite is of the less thorium-bearing type. It is common with regard to its chemical composition, which has not changed very much.

The Pyörönmaa pegmatite lies in Kangasala, southwest Finland. The pegmatite is inside a late kinematic Svecofennian granite body. The allanite there occur as a minor constituent. The radioactive minerals are located in the intermediate feldspar-rich zone, where they form pockets. These ‘anomalies’ consist almost exclusively of black allanite, which in places is brown in colour due to alteration. Occasionally, fergusonite is a common mineral. Albite has crystallized before and after allanite, e.g., it is found as fracture fillings in allanite. The allanite crystals are mostly prismatic, but in places of indefinite habit, the crystals are massive or rounded grains of different size. Chemically they are Ce-allanites. XRD studies have proved the allanite to be in the metamict state.

Fergusonite has been found in association with allanite in Pyörönmaa pegmatite, but here the fergusonite is younger than allanite. Fergusonite occurs in three different phases: inside one morphologically single fergusonite crystal, there are three varieties, two being in the metamict state and the third in the crystalline state. These varieties are more advanced recrystallization products. The habit of the mineral is prismatic; the crystal faces are poorly developed. The mineral is transparent in thin splinters and the fracture is conchoidal. Some

crystals are brittle and inhomogenous due to hydration. The unheated material is X-ray amorphous.

The Varala pegmatite occurs about 14 km to the north-east of the Pyörönmaa pegmatite. The pegmatite itself is confined to even-grained granite that may be a satellite of the granite body where the Pyörönmaa pegmatite is situated. The XRD showed that the allanite is metamict. The occurrence of bastnäsite among the alteration products of allanite is in accordance with the high CO₂ content in the altered allanites. Allanite occurred both as prismatic crystals and as inclusions of indefinite habit. The allanite of Varala is among the most thorium-rich allanites.

The Luumäki pegmatite is situated at Kännätsalo, southeast Finland, in the Wiborg rapakivi massif. The pegmatite has similar mineral assemblages as the Pyörönmaa and Varala pegmatites. The dyke is composed of a quartz core and surrounded by two feldspar pegmatite zones. The coarse-grained inner feldspar pegmatite zone is enriched with rare earth minerals. The pegmatite is classified as a miarolitic pegmatite. Several complex processes, including hydrothermal alteration, have been in progress in the area. Dark brown monazite, mainly containing Ce and other lanthanoids, occurs near the quartz core, as large rounded crystals. The crystal faces are indistinctly developed. Biotite and hematite occur in strongly altered form.

Table 2 shows the chemical composition of the minerals studied here. The results given are averages and the standard deviations for the whole profile rather than one single value. This way of reporting indicates the variation for each element more clearly. The electron microprobe profiles for allanites show a homogeneous composition for the major elements; no internal zoning was observed. The maximum and minimum values of certain elements are shown in Figs. 1,2,4. The allanites contain approximately 11 wt% Ce₂O₃. The thorium oxide showed minor variation; this is often reported for allanites of pegmatites (JANECZEK and EBY 1993) (Fig. 1). The totals are also near the hundred level, which indicates that there have been no strong oxidizing alteration processes. The uranium content in allanites was under the detection limit, but the concentration was high enough to be determined by alpha spectrometry.

The microprobe results for fergusonite show a more heterogeneous composition. This is seen in the Fig. 2, where the ThO₂ and UO₂ profiles are presented (see also Fig. 3). The variation is in lumps rather than being caused by internal zoning. The totals were low, indicating possible mineral alteration, or rather hydration, process. Hydration means that there have been dislocations which have produced vacant places and substitutions with

OH-groups. This may have given rise to uranium oxidation. Owing to the high amount of oxidized uranium in fergusonite, it is more vulnerable to leaching and environmental changes, which may lead to erroneous dating results.

Although the monazite grain chosen seemed to be homogeneous at first, the microprobe analysis indicated an edge with lighter composition (marked with 5* in Table 2). This means there can be clear areas with different composition inside a grain. Inhomogeneity of uranium and thorium has been reported in allanites and monazite by JANECEK and EBY (1993). The cerium profile showed a homogeneous composition, but there was slight variation in the uranium and thorium profiles in both cases (Fig. 4). No internal zoning was observed.

Determination of uranium oxidation

The least altered part of the mineral crystals was chosen for uranium oxidation state determinations. Any oxidized surfaces were removed and the mineral grains were crushed into smaller pieces. These were checked by optical microscope, and crystals with the least impurities and heterogeneous phases were selected. Finally, the most suitable mineral grains were hand-picked for chemical analyses.

The uranium oxidation states were determined by a chemical method based on ion exchange. The sample was dissolved in an acid mixture, containing 4.5 M HCl and 0.03 M HF, under anoxic conditions for several hours. The oxygen was removed from the acid solution in advance by purging with N₂ gas. The dissolution of the sample was accelerated by ultrasound. The uranium oxidation states were separated in an ion exchange column under a protective gas flow. Strong anion exchange resin (Dowex 1x4, 50-100 mesh) was regenerated to the same acid mixture as the dissolution of the sample was done. The reduced form of uranium passes through the column while the oxidized form is retained in the column. This is eluted by a dilute of 0.1 M HCl. The separated oxidation states were spiked with ²³²U tracer. The fractions were evaporated and purified by ion exchange. The uranium content of the fractions was measured by alpha spectrometry (ERVANNE and SUKSI 1996). The total uranium content of the sample was determined to check that the dissolution of the sample was complete, since after dissolution there was a white residue. The sample was refluxed progressively in concentrated HCl, HNO₃ and HF for about several hours before the uranium content was determined by ion exchange and alpha spectroscopy.

Results and discussion

After the allanites and fergusonite were dissolved, an insoluble white residue remained. To confirm that all of the uranium was leached to the solution, the dissolution time was increased to several hours. No remarkable change in the uranium concentration was observed. In the case of monazite, however, a fourteen-fold increase in the amount of dissolved uranium was seen. When compared against the results of the microprobe analyses, the uranium content obtained by chemical dissolution were lower. This means that the acid mixture used for dissolution was not suitable for dissolving all of the uranium contained in the monazite samples. However, if we assumed that the major part of the dissolved uranium content is the easily leachable portion of uranium, i.e. oxidized uranium, we can indirectly calculate the distribution of the uranium oxidation state by subtracting this amount of oxidized uranium from the total amount of uranium determined with longer dissolution time. Thus, monazite occurs almost totally in the reduced form, $U(IV)/U_{tot}$ 0.93. The results for the uranium oxidation state are shown in Table 3. All of the allanites are oxidized to some extent. The $U(IV)/U_{tot}$ ranged from 0.33 to 0.43. The radioactivity content was of about the same magnitude in all of the allanites. Dated ages of the rare-element pegmatites fall in a narrow time range, about 1.86-1.79 Ga. The age is strong parameter for introducing a dose in the crystal. During the radioactive decay, the recoil effect may cause uranium oxidation, but the magnitude of this phenomenon cannot be so high as to explain the oxidation state distribution obtained here. If, on the basis of the age, we calculate the amount of ^{238}U which has decayed and assume that all of this would lead to uranium oxidation, we would arrive at about 0.7 for the $U(IV)/U_{tot}$ ratio. But this value is too low, since some of the ^{234}U has also decayed at the same time. On the basis of the age, we arrive at radiation doses which according to JANECEK and EBY (1993) are high enough to generate significant radiation damage. One factor affecting the oxidation of uranium can be the decay product of lead, Pb^{+4} , which can be a strong oxidant as it is unstable in the presence of U^{+4} . The reaction is so-called auto-oxidation (BURNS and FINCH 1999). In addition, we do not know the initial distribution of the uranium oxidation states. Is all of the uranium originally in the reduced form?

The fergusonite was found to be strongly oxidized. The oxidizing environment was also seen in the microprobe analyses. With regard to fergusonite, hydration is probably the reason for the lower uranium oxidation states. The increase in volume following hydration may introduce microfissures, which are channels for circulating fluids. However, strong fluid interaction would have been seen more clearly in the uranium isotope ratio.

Hydrothermal alteration is not probable because in that case the uranium would have leached out. In addition, the content of radioactive elements in the fergusonite was higher than in the allanites, which introduces a higher radiation dose to the mineral. The metamictization and deterioration of the crystal structure is more profound.

Conclusions

The uranium oxidation states obtained for allanites were about the same for the three allanites studied here ($U(IV)/U_{tot}$ 0.33-0.43). This may partly be due to the geologically similar age (Svecofennian) of the samples. The reason for the low $U(IV)/U_{tot}$ ratio is unclear and needs further investigation. The fergusonite was found to be in oxidized form ($U(IV)/U_{tot}$ 0.06). This is in accordance with the altered structure, as indicated by the microprobe profile, which was probably due to hydration. The monazite was indirectly found to be in reduced form ($U(IV)/U_{tot}$ 0.93), since the dissolution was incomplete. A more suitable dissolution method for monazite samples is required.

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Table 1. Minerals and their general formulas are reported by LOKKA (1935,1950), VORMA et al. (1966), SIIVOLA (1975), VAASJOKI (1977), LAHTI (1996), LAHTI and KINNUNEN (1992).

Sample	Mineral	General formula	Site
1	Allanite	$(\text{Ca,Ce})_2(\text{Fe,Al})_3\text{Si}_3(\text{O,OH,F})_{13}$	Brändö, Ahvenanmaa
2	Allanite	$(\text{Ce,Ca,Y})_2(\text{Al,Fe})_3(\text{SiO}_4)_3(\text{OH})$	Pyörönmaa, Kangasala
3	Allanite	$(\text{Ca,Ce,Mn})_2(\text{Al,Fe})_3\text{Si}_3(\text{O,OH,F})_{13}$	Varala, Kangasala
4	Fergusonite	$(\text{Y,Ce})(\text{Nb,Ta})\text{O}_4$	Pyörönmaa, Kangasala
5	Monazite	$(\text{Ce,Th,Nd,Lu})\text{PO}_4$	Luumäki, Kännätsalo

Table 2. Microprobe analyses. The chemical composition (wt%) of the minerals was determined by using a Cameca Camebax SX50 electron microprobe at the Geological Survey of Finland. The microprobe was operated at an accelerating voltage of 15 V, and the beam current was 20nA. The beam diameter was approximately 5µm. The value is an average of the profile and its standard deviation.

Analyses	1	2	3	4	5	5*
SiO ₂	30.64(0.18)	29.64(0.18)	29.65(0.23)	0.69(0.47)	-	-
Al ₂ O ₃	13.09(0.25)	14.80(0.13)	14.88(0.12)	n.d.	n.d.	n.d.
FeOtot	15.32(0.24)	14.10(0.16)	13.90(0.21)	n.d.	0.02(0.03)	0.01(0.02)
CaO	11.28(0.11)	5.52(0.09)	5.49(0.09)	1.72(0.57)	0.32(0.03)	0.27(0.10)
UO ₂	n.d.	n.d.	n.d.	2.08(0.72)	0.43(0.08)	0.04(0.06)
ThO ₂	1.57(0.16)	1.30(0.22)	1.23(0.12)	0.75(0.33)	13.59(0.17)	3.68(1.56)
PbO	0.10(0.09)	0.14(0.11)	0.11(0.09)	0.57(0.18)	0.73(0.20)	0.18(0.14)
Y ₂ O ₃	0.02(0.02)	1.94(0.14)	2.01(0.07)	25.38(1.07)	0.09(0.02)	0.07(0.02)
La ₂ O ₃	8.10(0.14)	3.47(0.27)	3.75(0.11)	0.03(0.05)	8.63(0.12)	9.91(0.29)
Ce ₂ O ₃	11.10(0.32)	10.71(0.39)	11.20(0.29)	0.15(0.12)	34.42(0.50)	38.81(0.52)
Pr ₂ O ₃	0.95(0.22)	1.93(0.33)	1.98(0.19)	0.09(0.10)	3.44(0.20)	3.84(0.07)
Nd ₂ O ₃	2.56(0.23)	3.98(0.25)	4.11(0.22)	0.63(0.21)	10.83(0.38)	12.34(0.57)
Gd ₂ O ₃	0.06(0.11)	1.10(0.32)	0.97(0.20)	3.29(0.28)	0.50(0.24)	0.53(0.22)
Dy ₂ O ₃	n.d.	n.d.	n.d.	4.10(0.21)	0.27(0.08)	0.16(0.14)
Sm ₂ O ₃	0.11(0.11)	1.39(0.19)	1.33(0.15)	0.68(0.19)	1.34(0.14)	1.65(0.22)
Ta ₂ O ₅	-	-	-	3.06(0.25)	-	-
Nb ₂ O ₅	-	-	-	41.65(1.96)	-	-
MnO	0.47(0.05)	5.18(0.17)	4.79(0.11)	0.12(0.09)	0.05(0.05)	0.05(0.03)
MgO	1.04(0.12)	n.d.	n.d.	n.d.	0.01(0.01)	0.01(0.01)
TiO	0.69(0.10)	0.01(0.01)	n.d.	n.d.	n.d.	0.04(0.05)
P ₂ O ₅	0.12(0.03)	0.03(0.03)	0.03(0.03)	-	24.10(0.21)	27.44(0.43)
F	0.27(0.11)	0.40(0.10)	0.32(0.10)	0.69(0.25)	0.71(0.13)	0.70(0.10)
Total	97.42(0.81)	95.68(0.58)	95.81(0.93)	86.68(1.98)	99.50(0.65)	99.73(0.42)
N	29	16	18	58	12	5

5* is the edge of the monazite grain. n.d. means not detected. N is number of analyses per profile.

Table 3. The oxidation state distribution for uranium and the corresponding isotopic ratios.

Sample	U(IV)/U _{tot} (%)	²³⁴ U/ ²³⁸ U(IV)	²³⁴ U/ ²³⁸ U(VI)
1	32.14±1.96	1.15±0.07	0.940±0.076
	32.72±1.52	1.01±0.05	0.992±0.035
average	32.50±1.20	1.06±0.04	0.983±0.032
2	33.20±1.83	0.982±0.062	0.946±0.059
	36.74±2.11	1.11±0.07	0.937±0.069
average	34.72±1.38	1.04±0.05	0.942±0.045
3	45.11±2.28	1.07±0.06	0.875±0.052
	40.35±2.38	1.09±0.07	0.962±0.067
average	42.83±1.65	1.08±0.05	0.907±0.041
4	4.64±1.21	1.11±0.36	0.994±0.177
	6.10±0.51	1.03±0.10	1.23±0.10
average	5.88±0.47	1.04±0.09	1.18±0.09
5	3.57±0.35	1.14±0.13	0.951±0.076
	7.29±0.44	0.878±0.077	0.929±0.077
average	5.01±0.27	0.946±0.066	0.940±0.054

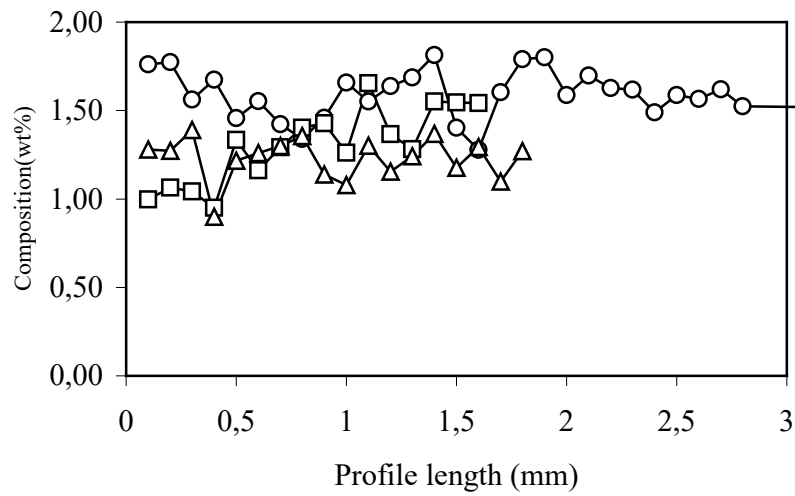


Fig. 1. ThO₂-profile of allanites: (o) sample 1, (□) sample 2 and (Δ) sample 3. The profile lengths are 1(2.8mm), 2(1.5mm) and 3(1.7mm).

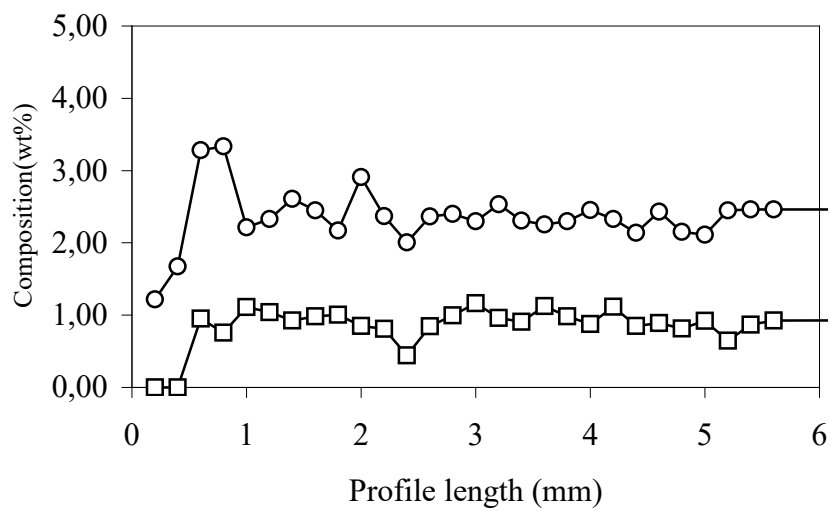


Fig. 2. ThO₂(□) and UO₂(o)-profiles of fergusonite. The profile length is 5.6mm.

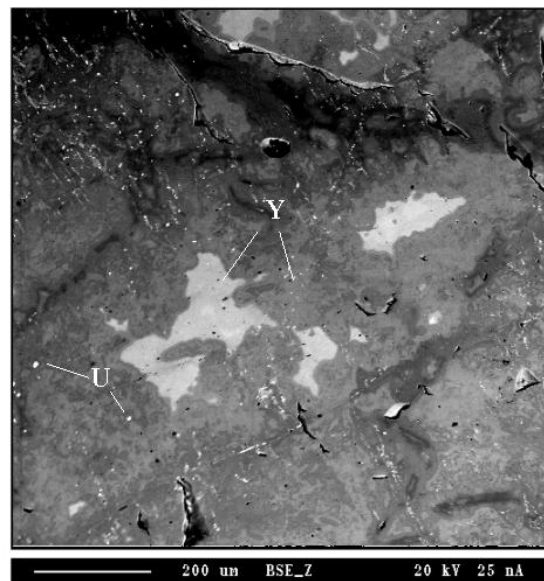


Fig. 3. Backscatter electron image of the heterogeneous fergusonite. The areas enriched in heavy elements appear lighter.

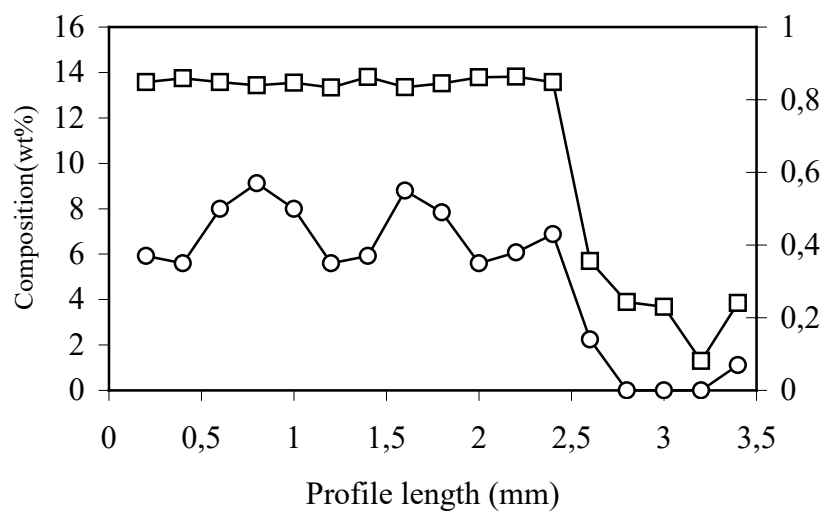


Fig. 4. ThO₂(□) and UO₂(○)-profiles of monazite. The profile length is 3.4mm. The area of different composition is shown on the right.