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PRELIMINARY REPORT ON REE DISTRIBUTION IN TIN-BEARING SCHISTS OF THE STARA KAMIENICA SCHIST BELT (THE SUDETY MTS., POLAND)*

Abstract. Ten samples from the Przecznica area (Kamienica Schist Belt, Lower Silesia) were analysed with the INAA method. The samples represent cassiterite-mineralized and barren schists. The former are very low in Na, Rb and low in Cs, Cr, Sc and Th but enriched in Fe, Co, Zn, As, Sb and Se. Their REE contents are also low with LREE>HREE, relatively wide sample/chondrite pattern and $(La/Lu)_{cn}$ ratio from 2.5—7.3. The barren schists show much higher contents of Na, Pb, Cr, Th and Sc, and reveal parallel, narrow sample/chondrite pattern with LREE>HREE and $(La/Lu)_{cn}$ ratio from 6.6 to 16.5. Both sample sets show more flat sample/NASC patterns with LREE=HREE, roughly. Some samples reveal poorly marked Sm or Eu positive anomalies.

Chondrite/samples plots fit to that of the greenschist metapelite facies and to the NASC standard. The authors suggest that examined schists were formed during isochemical regional metamorphism. Redistribution of elements took place during hydrothermal solution activity which introduced Sn and sulphide-associated elements, and, simultaneously, removed Rb, Na, Cs, Sc, Th, Fe (to some extend) and REE.

Key-words: REE, trace elements, tin-bearing schists, hydrothermal mineralization.

INTRODUCTION

The Gierczyn-Przecznica tin-bearing schists constitute a part of the Stara Kamienica Schist Belt and extend over a distance of about 12 kilometers from Nove Mesto p/Smrkem in Bohemia, in the west to Przecznica in the east (Fig. 1). The schists form a part of the northern metamorphic cover of the Karkonosze Granite and are regarded as nember of the supracrustal series of inferred Algonkian age, precursors of which were presumably clayey shales (Kozłowski 1974). The rock-forming minerals were examined by Makała (1994).

The schists from Gierczyn-Przecznica area form relatively uniform rock complex. However, the northern part known from the drill cores shows distinct diversity in the mineralized (stratigraphically middle) part. The differences are caused by variable

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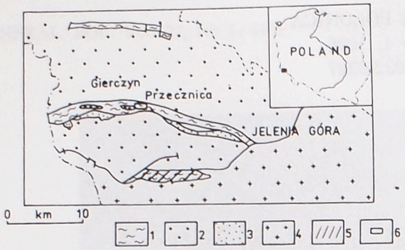

Fig. 1. Geological sketch of the northern metamorphic cover of the Karkonosze Granite (after Sawicki 1966, simplified)
1 — mica schists, 2 — gneisses, 3 — leucogranites, 4 — Karkonosze Granite, 5 — hornfelses, 6 — boreholes

proportions or absence of rock-forming minerals: biotite, muscovite, chlorite, chloritoid, quartz and garnets (almandine, Kozłowski et al. 1988). Petrographic composition controls the rock structure.

The tin mineralization shows unusual, stratiform geometry, perfectly concordant with the schist foliation. The ore zones are thin, flat lenses or pseudolayers showing diffused boundaries defined by chemical analyses, as cassiterite is usually macroscopically invisible. Ore mineralization includes also accompanying sulphides (pyrrhotite, chalcopyrite, pyrite, arsenopyrite and others). Small mines had been in operation from XVI to XVIII centuries and during the World War II (see Jaskólski 1948). Unsuccessful exploration had been undertaken in 1950—60-ties and again in 1980-ties.

The unique mineralization of the Stara Kamienica Schist Belt has been a matter of discussion since decades. Summary of numerous genetic concepts together with comprehensive references can be found e.g. in: Jaskólski and Mochnacka (1958), Szałamacha and Szałamacha (1974), Kowalski et al. (1978), Speczik and Wiszniewska (1984), Wiszniewska (1984), Siemiątkowski (1991), Cook and Dudek (1994). The present authors support the hydrothermal, generally post-metamorphic hypothesis.

Despite abundant literature, very little is still known about geochemistry of the ore and host rocks including the distribution of REE. The authors aim to apply the REE and other trace elements distribution patterns to the genetic interpretation of the Gierczyn-Przecznica mineralization.

MATERIALS AND METHODS

Samples were collected from drill cores in the Przecznica area. Most of them originate from the tin-bearing zone and its immediate vicinity encountered in the P2/7 exploratory well at depth 40.0—40.6 meters. Tin content in this zone reaches 0.49 wt.% (after industrial data). Single sample with traces of cassiterite comes from the P6/29 well and barren sample was taken from the P2/8 hole. Localization of samples is shown in Fig. 2.

Polished and thin sections were examined under NIKON and OPTIPHOT microscopes. Basing on the results of these observations 10 samples were selected for chemical analyses carried out using the instrumental neutron activation method (INAA). From this population 5 samples represent tin-bearing schists and 5 other are barren ones.

The INAA technique has been used for determination of trace elements including rare earths. The samples were analysed in several independent series under constant conditions. Both the reference materials and the single-element standard samples produced from standard solutions were used. The following reference materials were applied: SL-1, SL-3, SD-M-2/TM, SOIL-7 (supplied by the International Atomic Energy Agency, Vienna), GnA (obtained from the Central Geological Institute in Berlin — former GDR) and Rus-1, Rus-2 (distributed by the Permanent Standardisation Committee of the former COMECON).

Samples and standards weighting ca 100 mg each were wrapped in small (15 × 15 millimeters) envelopes made of a 99.99% aluminium foil, packed into a tight aluminium container and irradiated in thermal neutron flux in the 'Eva' nuclear reactor at the Institute of Atomic Energy in Świerk, Warsaw. Gamma-spectra of radionuclides induced in samples during irradiation were counted with the high purity germanium (HPGe) semiconductor detector of energy resolution 1.7 keV for the 1330 keV line of ^{60}Co. The spectra were then processed with a SPAAC software package (see Janczyszyn et al., in press). Due to the significant distance from our laboratory to the nuclear reactor and respective time of sample transportation after irradiation only radionuclides with half-lifes longer then about 15 h could be measured. Parameters of irradiation and counting are given in Table 1.

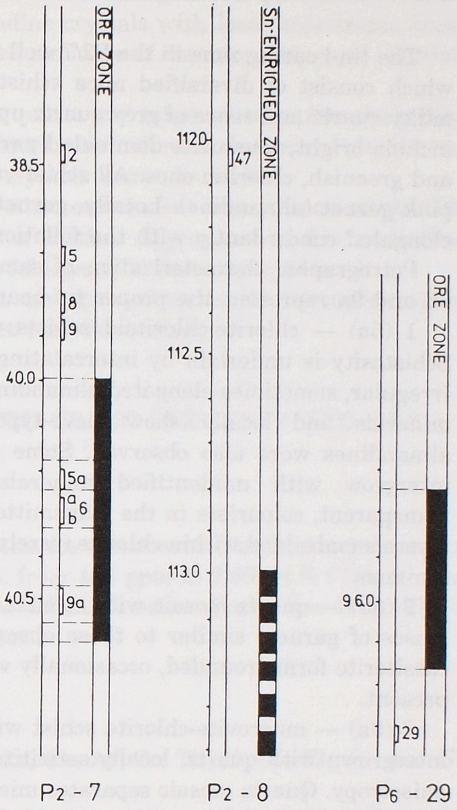

Fig. 2. Distribution of studied samples in the drill-cores from P2/7, P2/8 and P6/29 boreholes and their relationship to the ore zones (boundaries of ore zones after industrial data)

TABLE 1

Conditions of sample irradiation and counting

Neutron flux density	$2 \cdot 10^{13}$ n/cm^2 s	
Irradiation time [h]	8	
	Counting I	Counting II
Decay time	7 days	8—13 weeks
Counting time [s]	500	2 000

PETROGRAPHY OF TIN-BEARING SCHISTS

The tin-bearing zone in the P2/7 well includes the fragment of schists sequence which consist of diversified mica schists intercalated with layers and nests of milky quartz and zones of grey quartz up to several centimeters thick. The schists include bright, muscovite-dominated varieties intercalated with dark, biotite-rich and greenish, chloritic ones. All schist varieties may contain variable amounts of pink garnet (almandine). Locally, garnets form accumulations either nest-like or elongated concordantly with the foliation.

Petrographic characterization of samples is given below. — Samples 5a, 6a, 6b and 9a represent the proper tin-bearing zone (Fig. 2):

1 (5a) — chlorite-chloritoid schists with quartz, almandine and cassiterite. Schistosity is underlain by intercalating mica/chloritoid and quartz microlayers. Irregular, sometimes elongated almandine crystals penetrate into the surrounding minerals and locally show sieve-type intergrowths with quartz. S-shaped almandines were also observed. Some garnets reveal sericitization and locally intergrow with unidentified minerals of low birefringence. Cassiterite is transparent, colourless in the transmitted light and forms isometric or elongated crystals embedded within chlorite (rarely within garnets). Sulphides are relatively common.

2 (6b) — quartz mosaic with streaks of chlorite, relics of biotite and irregular lenses of garnets similar to those observed in samples 5a and 6a. Transparent cassiterite forms rounded, occasionally very fine isometric crystals. Sulphides are present.

3 (9a) — muscovite-chlorite schist with relics of biotite and isometric garnets intergrown with quartz, locally sericitized and intergrown with a mineral of low anisotropy. Quartz mosaic separates microlayers of micas. Transparent cassiterite forms fine, rounded crystals embedded within chlorite, quartz and muscovite. Transitions to semi-transparent SnO_2 are common. Disseminated cassiterite crystals form streaks. Small amounts of apatite and relatively common rutile and ilmenite were noted.

4 (6a) — strongly folded quartz-garnet-chlorite schist with rare chloritoid crystals and biotite flakes. The latter contain small crystals of a mineral which resembles zircon surrounded by pleochroic rims. Similar rims are observed in chlorites. Transparent cassiterite forms disseminated crystals or grape-like aggregates. Garnets are sericitized and show anisotropic areas. Pyrite, chalcopyrite, pyrrhotite and Ti-minerals were identified.

5 (29) — single sample from the P6/29 well composed of quartz with few chlorite intercalations. Rare cassiterite and more common sulphides were observed.

Samples 2, 5, 8, 9 and 47 represent barren schists from outside the tin-bearing zone (Fig. 2):

6 (5) — muscovite schists with biotite, quartz and local chlorite flakes. Occasionally, larger biotite flakes are present with fine zircon and/or monazite grains surrounded by pleochroic rims. Common ilmenite grains were observed.

7 (9) — muscovite schist with chlorite, biotite and garnets. Quartz microlayers are intercalated with mica ones. Almandine crystals with local anisotropic areas are subjected to local sericitization. Small amounts of ilmenite were seen together with tourmaline and apatite.

8 (8) — biotite-muscovite schist with quartz and large, idiomorphic almandine crystals (up to 3.5 millimeters in diameter) which show inclusions of quartz and micas. Single tourmaline and ilmenite crystals were also noted.

9 (2) — chlorite-biotite schists with distinct enrichment in postkinematic biotite. Chlorite originates from chloritization of biotite. Aggregates of fine-crystalline muscovite form pseudomorphoses after feldspars(?). Ilmenite is present.

10 (47) — chlorite schist with garnets and spots of muscovite. Strongly cracked garnets are chloritized. Feldspar veinlet cuts the sample.

DISTRIBUTION OF Sn, SELECTED TRACE ELEMENTS AND REE

Tin

The mineralized schists in which cassiterite was observed under the microscope show highly variable tin concentrations: from 406 ppm to 2.69 wt.% (Tab. 2). The barren schists reveal Sn contents below INAA detection limits (i.e. from 390 to 460 ppm, depending on the matrix effect).

Trace elements

In the studied samples Fe, Cr, Co, Zn, Sb, As, Sc, Th, U, Se, Rb, Cs and Na were determined (Tab. 2).

The two sets of schists — mineralized and barren — differ significantly in the content of trace elements. Particularly well-marked are differences in Na, Rb and Cs (Fig. 3). Less pronounced but visible are differences in Sc, Th and Cr contents (Fig. 4).

The mineralized schists are very low in Na and Rb (below detection limit), and low in Cs, Cr, Sc and Th. In contrast, enrichment is observed in Fe, Co, Zn, As, Sb and Se (Tab. 2) i.e. in elements which participate in sulphide stage of mineralization. However, the exception is sample 3 (9a) in which Sc, Th and Cr are more abundant that in the remaining mineralized schists.

Barren schists reveal much higher contents of Na, Rb, Cs, Cr, Th and Sc (Fig. 3, 4). Uranium concentration are low in both types of schists and do not show regularities.

Rare earth elements

The whole-rock REE contents are listed in Table 3 and distribution patterns normalized to chondrites and to the NASC (after Haskin et al. 1968) are shown in Fig. 5 and 6, respectively.

TABLE 2

Whole-rock anaalyses for selected elements of mica schists from Przecznica determined with INAA (Fe in wt.%, all other values in ppm) (samples 1—5 are tin-bearing schists, samples 6—10 are barren ones)

Sample[a] and lithology	Sn	Na	Rb	Cs	Th	U	Sc	Fe [wt.%]	Cr	Zn	Co	As	Sb	Se
1 (5a) chlorite-chloritoid schists	1 491	<147	<120	<0.4	2.4	2.1	2.94	10.1	14.9	297	40	27.1	<0.4	<3.9
2 (6b) chlorite-quartz schist	26 968	<160	<74	<0.4	2.0	1.5	4.4	13.2	12.1	242	73.8	5.3	1.4	9.2
3 (9a) muscovite-chlorite schist	1 326	<180	<40	0.9	9.2	2.5	8.9	14.4	47.9	778	17.2	69.1	0.8	<3.2
4 (6a) quartz-chlorite schists	8 756	<140	<79	0.8	3.0	<1	5.8	13.4	21.4	171	55.3	28	0.9	8.2
5 (29) chlorite-quartz schist	406	<270	<45	1.3	5.4	1.4	4.1	11.5	27.9	61.1	70.1	24.9	2.2	8.9
6 (5) muscovite schist	<430	1 570	272	9.8	16.3	2.8	18.9	6.5	79.5	69.3	18.2	<6.9	<0.7	<5
7 (9) biotite-muscovite schist	<450	1 700	230	9.1	17.7	2.1	19.4	5.9	89.8	43.9	15.1	<7.8	<0.7	<4.4
8 (8) biotite-muscovite schist	<390	1 140	171	7.5	12.8	2.4	7.5	5.1	57.1	45.9	16.1	<7.3	<0.8	<3.9
9 (2) chlorite-biotite schist	<390	1 330	258	10.2	15.5	2.1	16.8	5.2	82.3	42.7	16.1	<7.3	<0.8	<4
10 (47) chlorite schist		821	81	2.69	13.0	2.9	9.3	18.9	73	135	38	1.6	0.46	

a — numbers in brackets correspond to samples numbers in Fig. 2.

The mineralized schists have diversified sample/chondrite patterns with relatively wide range of values and LREE>HREE. The $(La/Lu)_{cn}$ ratio changes from 2.5 to 7.3. Poorly marked, positive Sm anomaly can be seen in samples 1 (5a) and 2 (6b) (Fig. 5). The sample/NASC patterns are more flat and ranges of values are more diversified. Roughly, LREE=HREE, however, sample 4 reveals LREE>HREE pattern (Fig. 6). Single sample 1 (5a) is particularly low in REE (Nd, Tb, Yb and Lu show contents below detection limit) (Tab. 3).

All the barren schists show roughly parallel, narrow sample/chondrite patterns (Fig. 5) with LREE>HREE and $(La/Lu)_{cn}$ ratio from 6.6 to 16.5 (Tab. 3). In single sample 7 (9) poorly marked, positive Sm anomaly appears. The sample/NASC patterns are flat but values are more diversified in comparison with the chondrite normalization. Samples 7 (9) and 10 (47) reveal poorly marked, positive Sm-Eu anomalies.

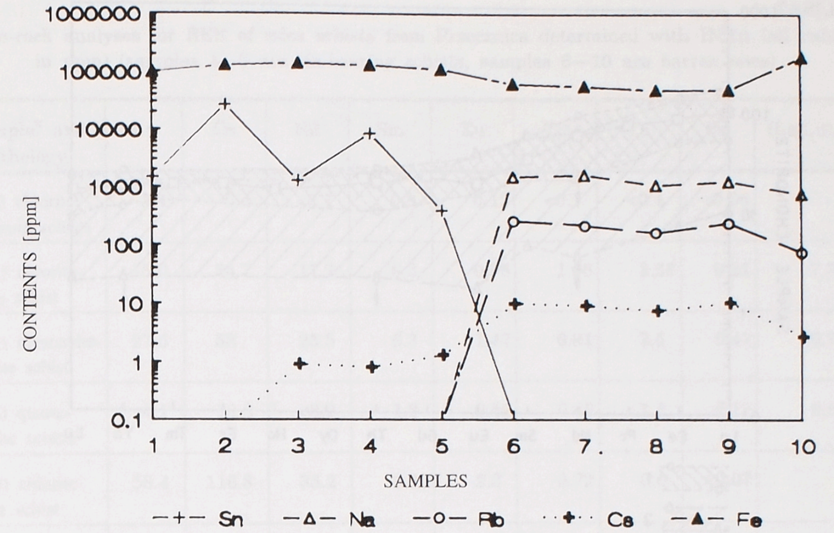

Fig. 3. Distribution of selected elements in the schists from the Przecznica area (numbers of samples correspond to the numbers in Table 2)

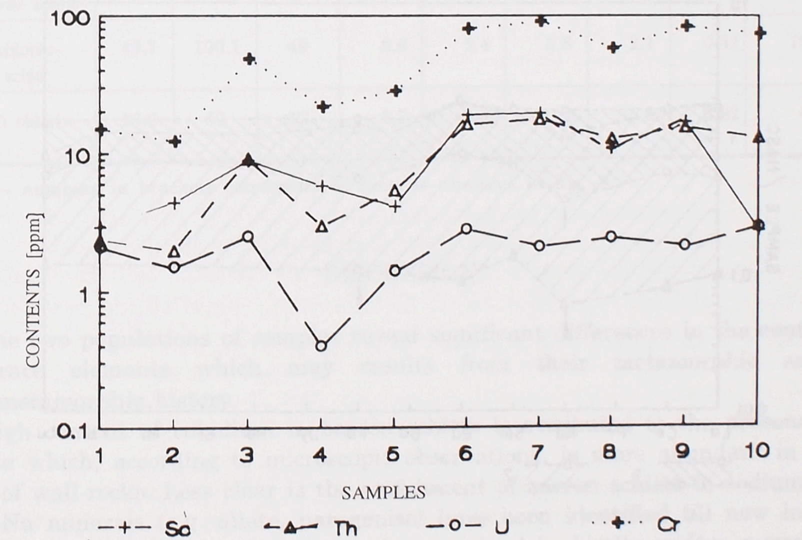

Fig. 4. Distribution of selected elements in schists from the Przecznica area (numbers of samples correspond to the numbers in Table 2)

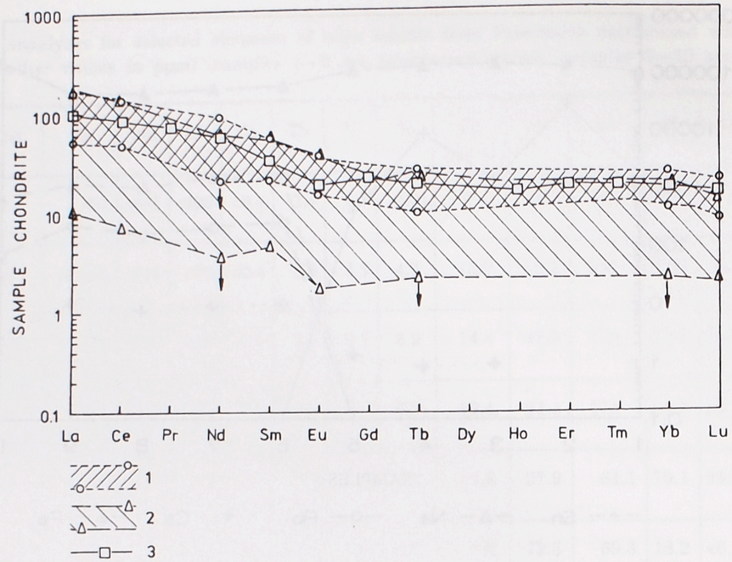

Fig. 5. Chondrite-normalized REE distribution patterns of schists from the Przecznica area
1 — barren schists, 2 — mineralized schists, 3 — NASC
(normalization after Haskin et al. 1968)

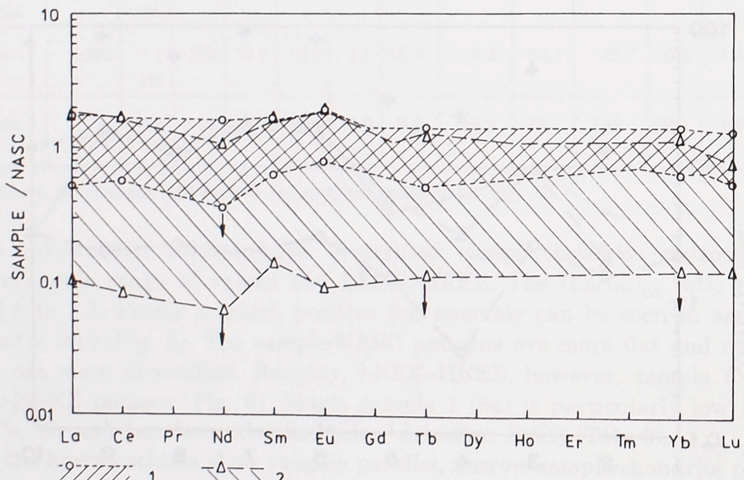

Fig. 6. NASC-normalized REE distribution patterns of schists from the Przecznica area
1 — barren schists, 2 — mineralized schists
(normalization after Haskin et al. 1968)

TABLE 3

Whole-rock analyses for REE of mica schists from Przecznica determined with INAA (all values in ppm) (samples 1—5 are tin-bearing schists, samples 6—10 are barren ones)

Sample[a] and lithology	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	$(La/Lu)_{cn}$
1 (5a) chlorite-chloritoid schists	3.4	6.4	<2.1	0.8	0.12	<0.1	<0.4	<0.06	
2 (6b) chlorite-quartz schist	15.0	34.7	17.7	6.3	0.98	1.08	2.58	0.21	7.3
3 (9a) muscovite-chlorite schist	27.5	58	25.5	6.3	1.47	0.91	3.5	0.41	6.9
4 (6a) quartz-chlorite schists	4.1	10.6	<8.2	1.8	0.59	0.46	1.6	0.17	2.5
5 (29) chlorite-quartz schist	58.4	116.8	35.2	8.9	2.3	0.72	0.6	<0.07	
6 (5) muscovite schist	53.1	113.4	51.8	9.7	2.13	1.2	2.0	<5.2	
7 (9) biotite-muscovite schist	56.8	122.4	53.5	9.6	1.76	1.2	4.4	0.62	15.0
8 (8) biotite-muscovite schist	43.6	91.1	42.1	7.5	1.5	0.7	3.2	0.5	9.4
9 (2) chlorite-biotite schist	49.7	100.1	49	8.6	2.4	0.8	2.1	0.31	16.5
10 (47) chlorite schist	16.9	42	<12	3.7	1.00	0.45	2.39	0.27	6.6

[a] — numbers in brackets correspond to samples numbers in Fig. 2.

DISCUSSION

The two populations of samples reveal significant differences in the contents of trace elements which may results from their metamorphic and/or post-metamorphic history.

High content of rubidium in barren schists is attributed to the presence of biotite which, according to microscopic observations, is more abundant in this type of wall-rocks. Less clear is the enrichment of barren schists in sodium. No high-Na minerals (e.g. albite, paragonite) have been identified till now in the Przecznica schists. This element can be contained in biotite and/or muscovite, eventually also in garnets. Depletion in Rb, Na (and Cs) is inferred to be caused by chloritization of biotite which is the principal hydrothermal alteration process observed in the Kamienica schists. Alternatively, this difference may originate from primary, low biotite content in the rock.

Scandium and chromium are also more abundant in barren schists. The first element is presumably hosted in biotite and garnets, the latter — rather occurs in biotite. Similarly to Rb and Na, depletion in Sc and Cr can be the result of chloritization of biotite and, eventually also the replacement (e.g. sericitization) of garnets — the process also commonly observed in the Kamienica schists.

Concentrations of uranium and thorium are low and are genetically related to the accessory minerals from which zirconium and/or monazite (Cook and Dudek 1994) were observed in the Kamienica schists. U and Th own phases have not been noticed despite the fact that U anomalies are known from the vicinity of the Kamienica Schist Belt. Thorium contents in barren schists are slightly higher than in mineralized ones whereas uranium remains fairly stable in both types of schists. Hence, Th/U ratio generally increases from mineralized to barren schists (Tab. 2).

The enrichment in Fe, Co, Zn, As, Sb and Se of the tin-bearing schists is clearly related to the presence of sulphides and seems to reflect the activity of hydrothermal solutions responsible for the formation of sulphide stage of mineralization.

The REE contents are presumably connected with micas, as quartz is rather low in these elements and as other REE-controlling minerals (e.g. feldspars and accessory phases) are rare or absent in the studied samples. The REE patterns show remarkable differences between mineralized and barren schists. Generally, the plots for barren schists are parallel and fall into the narrow range of values despite their distance both from the mineralized zones and between the sampled boreholes. Moreover, their chondrite/sample plots fit perfectly to that of greenschist metapelitic facies (Grauch 1989) and to the NASC standard (Fig. 5) which, to some extent, may represent the 'average' composition of shales. In contrast, mineralized schists generally show depletion in REE and more diversified composition.

No significant anomalies were observed for Eu and other REE. It is interesting because available data for biotites from metamorphic rocks (Grauch 1989) and biotites and chlorites from hydrothermally active granitic environments (see Alderton et al. 1980; Morteani et al. 1986,) usually reveal strong, negative Eu anomalies.

CONCLUSIONS

At this stage of studies data presented above are preliminary and do not permit the far-going, particularly genetic conclusions. However, apparent differences in abundance of selected elements (including REE) between barren and mineralized schists suggest the influence of processes which are here attributed to the activity of hydrothermal solutions.

If, hypothetically, barren schists represent the 'initial' concentrations of elements inherited from their sedimentary precursors, as suggested by their similarity to the NASC-derived REE pattern, it is suggested that schists from Przecznica were formed during an isochemical regional metamorphism episode. Furthermore, it can be proposed that redistribution of elements has occurred during the invasion of hydrothermal solutions. These solutions were responsible for introduction of Sn as well as sulphide-affiliated elements: Fe, Co, Zn, As, Sb and Se, and, simultaneously, for removal of Rb, Na, Cr, Sc, Th, Fe (to some extent) and REE.

Therefore, the REE patterns for mineralized schists reflect the overlapping of the two processes: regional metamorphism which affected whole schists sequence and metasomatosis, the latter being limited to the local, mineralized, tin-bearing zones. Hydrothermal metasomatic processes led to the depletion in REE, particularly in LREE, as suggested by somewhat higher $(La/Lu)_{cn}$ ratios for barren schists. However, more diversified patterns for the mineralized schists imply variable intensity of solutions action. It is supported by the pattern for poorly mineralized sample 5 (29) which appears to be intermediate between 'typical' barren and „typical" mineralized schist.

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PIERWIASTKI ZIEM RZADKICH W ŁUPKACH CYNONOŚNYCH PASMA STAREJ KAMIENICY (SUDETY, POLSKA)

Streszczenie

Wykonano badania dziesięciu próbek z otworów wiertniczych w Przecznicy, w najbardziej na wschód wysuniętej części strefy cynonośnej pasma łupkowego Starej Kamienicy. Pięć z nich to próbki okruszcowane kasyterytem, pięć — łupki płonne. Lokalizację prób przedstawiają rysunki 1 i 2. Wyróżniono tu łupki: chlorytowo-chlorytoidowe, chlorytowo-kwarcowe, muskowitowo-chlorytowe, kwarcowo-chlorytowe (okruszcowane kasyterytem) oraz muskowitowe, biotytowo-muskowitowe, chlorytowo-biotytowe i chlorytowe (nie zawierające kasyterytu).

Metodą neutronowej analizy aktywacyjnej oznaczono: Sn, Na, Rb, Cs, Th, U, Sc, Fe, Cr, Zn, Co, As i Sb oraz pierwiastki ziem rzadkich (Tab. 2 i 3). Łupki płonne od okruszcowanych różnią się rozmieszczeniem pierwiastków ziem rzadkich i wybranych pierwiastków śladowych. Łupki okruszcowane odznaczają się bardzo niską zawartością Na i Rb oraz niską koncentracją Cs, Cr, Sc i Th, przy równoczesnym wzbogaceniu w Fe, Co, Zn, As, Sb i Se (Tab. 2, Rys. 3, 4), tj. w pierwiastki uczestniczące w cynowym i siarczkowym stadium mineralizacji. Łupki płonne wykazują znacznie wyższe zawartości Na, Rb, Cs, Cr, Th i Sc (Rys. 3, 4).

Tabele 2 i 3 przedstawiają wykresy zawartości pierwiastków ziem rzadkich normalizowane do chondrytów i do NASC. Łupki okruszcowane wykazują znaczne zróżnicowanie zawartości oraz LREE>HREE, a także $(La/Lu)_{cn}$, który zmienia się od 2,5, do 7,3. Wykresy normalizowane do NASC ujawniają bardziej płaski przebieg z LREE=HREE. Jedna z próbek wykazuje szczególnie niskie koncentracje REE (Nd, Tb, Yb i Lu poniżej granic wykrywalności.). Dwie próbki wykazują słabe, pozytywne anomalie Sm. Łupki płonne wykazują prawie równoległe, zbliżone do siebie wykresy, z LREE>HREE i $(La/Lu)_{cn}$ wahającym się od 6,6 do 16,5. Wykresy normalizowane do NASC są płaskie, lecz zawartości w poszczególnych próbkach wykazują większe zróżnicowanie.

Na obecnym etapie badań przedstawione wyniki nie upoważniają do wysnucia daleko idących wniosków.

Skład pierwiastkow ziem rzadkich w łupkach płonnych jest zbliżony do średniego składu wzorca NASC. Pozwala to przypuszczać, że metamorfizm regionalny na badanym obszarze miał charakter izochemiczny i nie spowodował istotnych przemieszczeń REE. Przemieszczenia takie miały· natomiast miejsce w łupkach okruszcowanych i następowały pod wpływem roztworów hydrotermalnych. Doprowadzały one cynę i pierwiastki wchodzące w skład mineralizacji siarczkowej. Roztwory te powodowały równocześnie zubożenie skały w pierwiastki ziem rzadkich oraz w Na, Rb, Cs, Th i inne.