Z. geol. Wiss. • Berlin 18 (1990) 5 • S. 447-457

Contribution to the Geochemical Study of the Nb-Ta Pegmatite of Muiane, P. R. of Mozambique

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With 6 Figures and 3 Tables

1. Geology of the region

The pegmatite of Muiane is located in the region of Alto Ligonha, which is an interesting geological province of Mozambique in the Mozambique belt. The Precambrian rocks of the region have the following sequence in decreasing chronological order (LONGYEAR 1955; FIGUEIREDO DE BARROS & MARTINS VICENTE 1963; OBRETENOV 1978; PAECH & SCHMIDT 1982; ELGUINE et al. 1983):

- Pegmatites
- Granitic intrusions
- Metasedimentary series
- Regional gneisses.

Their distribution is shown in Fig. 1.

The pegmatites mainly occur in the outer zones of the Metasedimentary series or in the border zones between the Regional gneisses and the rocks of the above mentioned series (PAECH & SCHMIDT 1982). The petrography of the metamorphic complex was studied in detail by SCHMIDT (1986). The pegmatites of the Zambezia province form two strings which reflect the two principal tectonic directions of the region.

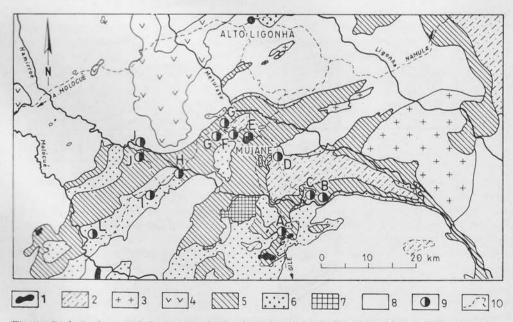


Fig. 1. Geological map of the region of Alto Ligonha (CORREIA NEVES & LOPES NUNES 1966)
1 - basic and ultrabasic rocks; 2 - equigranular granites; 3 - porphyritic granites; 4 - gneissic granites; 5 - schists;
6 - gneisses and metasedimentary rocks; 7 - quartzites; 8 - granitic gneisses; 9 - pegmatitic outerops; 10 - road

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The rare metal pegmatites are located parallel to each other in zones of NNE-SSW and NE-SW direction, corresponding to longitudinal veins, whereas the rare-earth pegmatites follow tectonic lines transversal to them thus having a NW-SE direction, corresponding to transversal veins (PAECH & SCHMIDT 1982).

2. Geological characterization of the ore deposit of Muiane

The main body of the ore deposit is a lenticular vein with NE-SW orientation at an angle of 25° to SE (LOPES NUNES 1972; OBRETENOV 1978; GOUK 1979), which is subjected to alteration processes, mainly kaolinization of feldspars and spodumene and cookeitization of spodumene and tourmaline (LONGYEAR 1955; FIGUEIREDO DE BARROS & MARTINS VICENTE 1963; v. KNORRING 1962; CORREIA NEVES et al. 1971; LOPES NUNES 1972; GOUK 1979). In the pegmatitic body xenoliths of neighbouring rocks occur, particularly in outer zones (Fig. 2), showing different degrees of assimilation. There is also a tectonic breccia which was formed later than the quartz nucleus and is composed

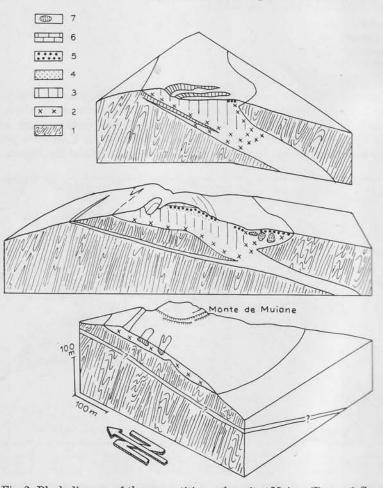


Fig. 2. Block-diagram of the pegmatitic ore deposit at Muiane (PAECH & SCHMIDT 1985) 1 — metamorphic schists; 2 — outer zone of the pegmatite; 3 — intern zone of the pegmatite; 4 — lithium zone of the pegmatites; 5 — breccia; 6 — quartz nucleus; 7 — xenolith

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of quartz, feldspars, micas and metamorphic schists (PAECH & SCHMIDT 1982). In the western and northern parts of the main body two dykes crop out which show some degree of mineral formation.

The pegmatite of Muiane shows a zoning into (Goux 1979):

quartz nucleus

- lithium zone
- inner zone
- outer zone.

This zonation is based on the distribution of the principal minerals, which are: quartz, microcline, albite, muscovite, biotite, cleavelandite, spodumene (cookeite), lepidolite, tourmaline and beryl.

In the various zones of the pegmatite the Ta mineralization is quite different:

Lithium zone: Nb-tantalite, microlite, and subordinate Mn-tantalite, Sb-tantalite, Ta-columbite, Sc-ixiolite, Bi-tantalite. Inner zone: Ta-columbite.

Outer zone: columbite (Ta-columbite).

The zonal structure is badly defined NE of the vein, where there is in fact no lithium zone and only the outer and inner zone are present (Fig. 3).

The pegmatite of Muiane is classified as a complex, zoned and differentiated pegmatite of sodolithic type.

3. Geochemistry of the pegmatite of Muiane

3.1. Trace elements in the pegmatites

Table 1 represents the mean contents and the CLARKE concentrations (CLARKE values after VINOGRADOV 1962 in RÖSLER & LANGE 1972) for each of the elements, as well as the values of the Nb/Ta ratio in the analysed pegmatites. From the CLARKE concentrations values we can conclude that the following elements have been enriched:

- in the main body:
 - lithium zone: W, Ta, Be, Sn, B, Nb, Ge (to a minor extent Co)

inner zone: W, Ta, B, Be, Nb, Ga, Co, Sn

outer zone: W, Sn, Ta, Nb, B, Be, Co, Ga

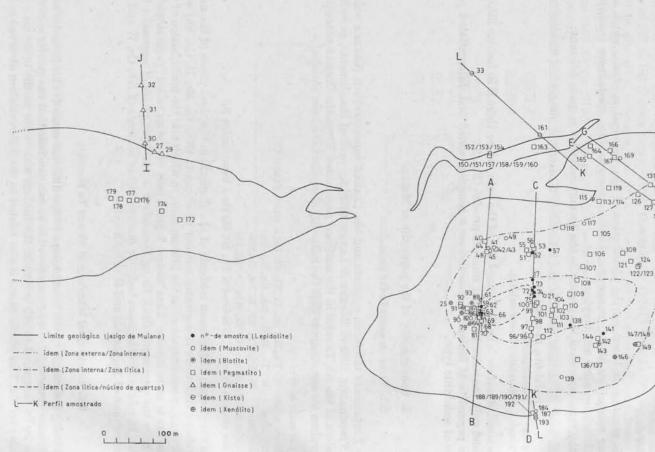
- in the northern dyke: W, Ta, Nb, Co, Be, Ga

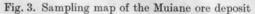
- in the western dyke: W, Nb, Co, Sn, Pb, Be, Ga (to a lesser extent B).

The comparison of the mean contents of the elements in the three zones of the main body shows that there is a greater number of elements (Nb, Ta, Be, B, Ga, Sn) with higher CLARKE concentrations in the lithium zone, in which also the kaolinization of the pegmatites is more intense. That means, a great part of the so called "pegmatophile" remain in the pegmatitic fluid, fixing themselves in the later phase of pegmatite formation. In relation to the main body we observe higher contents of Nb, Co, W and Pb and lower ones of Ta, Ba, Cu and B in both dykes.

3.2. Geochemical behaviour of Nb and Ta elements

Considering the values of the relationships in the different zones (Table 1): outer zone = 2.6, inner zone = 1.7, lithium zone = 0.5, we can conclude that the greater mineralization in Ta compared to Nb occurs in the lithium zone while in the outer zone Nb prevails over Ta. This phenomenon is also related to the differentiation observed in the





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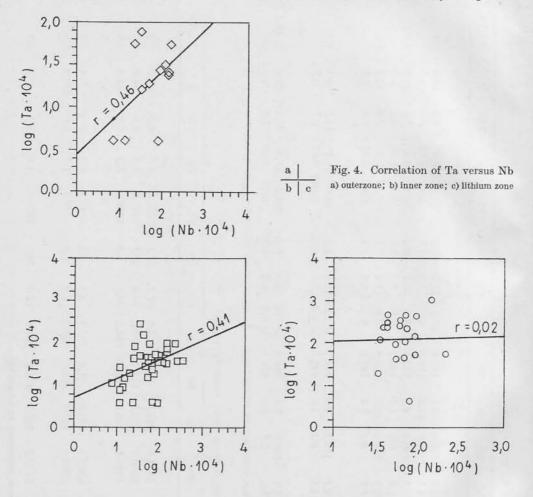
Table 1. Chemical composition in trace elements of the pegmatites

Section 20		Nb	Ta	Be	Ga	Mn	Sn	W	Ba	Co	Cu	Ti	Zr	Pb	В	Nb/Ta
		Main	body			2.57										
lithium zone	$ar{x}$ C	$\begin{array}{c} 64.6\\ 3.2 \end{array}$	$\begin{array}{c} 120.2\\ 48.1 \end{array}$	$\begin{array}{c} 49.0\\ 12.9\end{array}$	$58.9 \\ 3.1$	$788.0 \\ 0.8$	$\begin{array}{c} 28.1 \\ 11.2 \end{array}$	$\begin{array}{c} 165.6\\ 124.8\end{array}$	$93.3 \\ 0.1$	$\begin{array}{c} 22.9 \\ 1.3 \end{array}$	$5.9 \\ 0.1$	$51.3 \\ 0.01$	$\begin{array}{c} 9.0 \\ 0.05 \end{array}$	$5.3 \\ 0.3$	$55.0 \\ 4.6$	0.5
inner zone	$ar{x} \\ \mathrm{C}$	$51.3 \\ 2.6$	$30.2 \\ 12.1$	$11.2 \\ 2.9$	$41.7 \\ 2.2$	$\begin{array}{c} 190.3\\0.2\end{array}$	$4.3 \\ 1.7$	$\begin{array}{c} 154.9 \\ 119.2 \end{array}$	83.2 0.1	$\begin{array}{c} 38.9 \\ 2.2 \end{array}$	$9.5 \\ 0.2$	$\begin{array}{c} 51.3\\ 0.01 \end{array}$	$9.8 \\ 0.06$	$7.2 \\ 0.4$	$36.3 \\ 3.0$	1.7
outer zone	$ar{x} \\ \mathrm{C}$	$49.0 \\ 2.4$	$\begin{array}{c} 19.1 \\ 7.6 \end{array}$	$7.2 \\ 1.9$	$\begin{array}{c} 33.1 \\ 1.7 \end{array}$	$\begin{array}{c} 146.2\\ 0.1 \end{array}$	$\begin{array}{c} 19.8 \\ 7.9 \end{array}$	$\begin{array}{c} 186.2\\ 143.2 \end{array}$	$\begin{array}{c} 100.0\\ 0.2 \end{array}$	$\begin{array}{c} 33.9 \\ 1.9 \end{array}$	$8.5 \\ 0.2$	$\begin{array}{c} 288.4\\ 0.06\end{array}$	$\begin{array}{c} 12.6 \\ 0.07 \end{array}$	$9.2 \\ 0.6$	$\begin{array}{c} 27.5 \\ 2.3 \end{array}$	2.6
		North	n Dyke													
	$ar{m{x}}{ m C}$	$\begin{array}{c} 116.0\\ 5.8\end{array}$	$\begin{array}{c} 15.7 \\ 6.3 \end{array}$	$\begin{array}{c} 13.1\\ 3.4 \end{array}$	$\begin{array}{c} 32.4 \\ 1.7 \end{array}$	$\begin{array}{c}135.5\\0.1\end{array}$	$\begin{array}{c} 2.6 \\ 1.0 \end{array}$	$815.7 \\ 627.5$	$< 100 \\ < 0.2$	70.0 3.9	$3.5 \\ 0.07$		$\begin{array}{c} 11.7 \\ 0.07 \end{array}$	$\begin{array}{c} 11.3 \\ 0.7 \end{array}$	8.1 0.7	7.4
		West	Dyke													
	$ar{x} \\ \mathrm{C}$	$ 114.8 \\ 5.7 $	< 8 < 3.2	$7.6 \\ 2.0$	$33.9 \\ 1.8$	$\begin{array}{c} 166.7 \\ 0.2 \end{array}$	$7.6 \\ 3.0$	$616.6 \\ 474.3$	$< 100 \\ < 0.2$	$75.9 \\ 4.2$	$2.3 \\ 0.05$	$204.2 \\ 0.05$	11.0 0.06	$40.5 \\ 2.5$	$13.8 \\ 1.2$	> 14.3

Table 2. Chemical composition of the micas in trace elements (mean conter	Table 2. (Chemical	composition o	f the	micas in	trace	elements	(mean	contents	in	ppm)
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	Nb	Та	Ве	Ga	Mn	Sn	W	Ва	Co	Cu	Ti	Zr	\mathbf{Pb}	в	Nb/Ta
	Musco	ovites									1.1.1				
lithium zone	228.5	86.0	20.4	259.8	2511.6	265.3	53.4	110.0	7.3	19.1	391.2	4.0	4.9	36.1	2.7
inner zone	293.7	55.6	27.2	275.3	3360.7	255.0	121.7	101.9	10.9	11.8	478.7	4.0	3.3	84.8	5.3
outer zone	414.2	47.1	33.5	324.8	1229.1	506.5	60.6	134.5	10.9	6.0	1096.2	7.2	5.6	47.4	8.8
	Lepid	lolites													
lithium zone	95.7	407.7	17.4	167.0	3568.4	62.8	51.5	102.9	4.4	3.1	17.3	5.2	8.9	176.4	0.2
inner zone	78.0	269.1	19.6	232.0	3700.0	109.4	54.5	102.0	7.1	1.8	33.7	7.2	6.5	209.8	0.3
	Bioti	tes													
inner zone	671.6	80.1	13.1	111.1	3128.2	260.7	311.2	125.4	48.0	37.7	3636.6	27.4	11.6	28.8	8.4

Cr, Ni, V, Mo: Smaller than the sensibility limits of determination, respectively, 25, 2.5 and 1 ppm



pegmatite of Muiane, which is characterized by the predominance of the Ta minerals in the lithium zone and the Nb minerals in the outer zone.

The elements Nb and Ta, from the Vb group of the Periodical System, present very similar physical and chemical properties. The similarity of the ionic radii and the identity in the more stable valency state (+5) allow their association in nature and their mutual substitution in mineral structures. In a general way, both elements belong to the lithophile element group and are enriched in the residual solutions and in the later differentiates (ALEKSANDROV 1984; GRABEZHEV 1981).

In spite of this, there are certain differences between the chemistry of these elements. For example, Nb is a more reactive metal than Ta, presenting lower resistance to corrosion and, in general, reduces more easily than Ta (v) and hydrolises more slowly in hydrochloric acid solution (FAIRBROTHER 1967). Ta, with the atomic number 73, presents stronger metallic properties than Nb, with the atomic number 41. Consequently, alterations in the pH of the solutions initiate variations in the relative Nb and Ta concentrations and therefore, in the Nb/Ta ratio (ALEKSANDROV et al. 1985).

The increase of alkalinity in the final phase of pegmatite formation will then favour the Ta enrichment in the hydrothermal solution, leading to higher contents of this element in the lithium zone.

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Nb and Ta form coordination compounds with carbonates and fluorides, Nb presenting greater affinity for carbonates and Ta for fluorides (ALEKSANDROV & GARANIN 1980).

The greater enrichment of Ta in the lithium zone means that this element remains in the residual solution for a longer period of time, and the separation of both elements is attributed to the greater mobility and stability of the Ta-F complexes in relation to the Nb-F complexes (PARKER & FLEISCHER 1968).

In addition fluorine plays an important role in the Ta enrichment, with Ta tending to a higher concentration than Nb in the phase of pegmatitic crystallization (v. KNORRING & FADIPE 1981).

The progressive increase of Ta in relation to Nb and, consequently, lower values of Nb/Ta (2.6, 1.7 and 0.5 in the outer, inner and lithium zone, against the Nb/Ta CLARKE value of 8) is also linked to intense albitization zones (SKVORCOV et al. 1975; MÖLLER et al. 1983).

So the Nb/Ta ratio represents an indicator of the Nb and Ta mineralization. In the plots of Ta versus Nb (Fig. 4), the absence of correlation between the two elements again shows different tendencies in their fixation in the course of pegmatite formation.

The value of the correlation coefficient decreases from the outer to the lithium zone, in correspondence with the decrease of the Nb/Ta ratio from the outside to the inside of the pegmatic body.

A Stand	Nb	Та	Be	Ga	Mn	Sn	W	Ba	Co
Gneisses									
x	33.7	23.6	1.3	13.6	377.0	0.7	518.8	289.2	72.8
x C	1.7	9.4	0.3	0.7	0.4	0.3	399.1	0.4	4.0
Schists									
x	17.8	< 8	6.5	12.2	587.0	1.9	146.5	167.5	27.1
x C	0.9	< 3.2	1.7	0.6	0.6	0.8	112.7	0.3	1.5
Xenoliths									
x	60.6	33.4	19.9	32.1	485.6	30.8	156.2	236.9	31.8
C	3.0	13.4	5.2	1.7	0.5	12.3	120.2	0.4	1.8
	Cu	Ti	Zr	Pb	в	Cr	Ni	V	Mo
a .			T. all	Saler	lin de o	128	12.6413	ni olar	
Gneisses -	04.4	001-0	07.4		0 =	-0.4	00.0	#1 P	- 1
$\overline{\mathbf{x}}$ C	84.1 1.8	$\begin{array}{r} 2315.9\\ 0.5\end{array}$	$\begin{array}{c} 87.1\\ 0.5\end{array}$	$\begin{array}{c} 6.2 \\ 0.4 \end{array}$	8.7 0.7	$\begin{array}{c} 58.1 \\ 0.7 \end{array}$	$\begin{array}{c} 22.8 \\ 0.4 \end{array}$	$\begin{array}{c} 51.6\\ 0.6\end{array}$	< 1 < 0.9
Schists									
x	14.8	4289.7	68.0	5.2	0.8	139.8	27.0	74.7	1.5
x C	0.3	0.9	0.4	0.3	0.9	1.7	0.5	0.8	1.4
Xenoliths									
x	34.8	5639.1	112.7	10.1	52.7	87.4	14.0	67.9	2.1
x C	0.7	1.3		0.6	4.4	1.1	0.2	0.8	1.9

Table 3. Chemical composition of gneisses, schists and xenoliths of trace elements

 \overline{x} - mean content in ppm; C - CLARKE concentration

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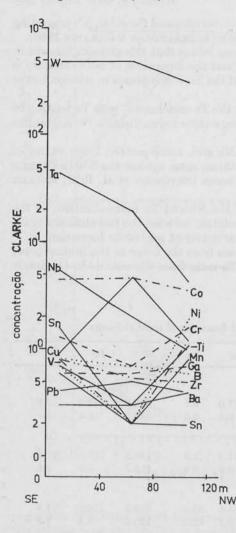


Fig. 5. Distribution of trace elements in the SE-NW-profile (Fig. 3, I-J) in gneisses of the footwall of the west dyke

3.3. Geochemical characterization of micas

Table 2 shows the mean contents of the trace elements in the micas of the main body as well as the values of the Nb/Ta ratio. By comparing the distribution of traces elements in muscovites and lepidolites of the inner and lithium zones we note that:

- The enrichment of the elements Nb, Ga, Be, Ti, Sn, Co, W and Cu is linked to muscovite formation, i.e., the contents of these elements are higher in muscovites than in lepidolites.

- The enrichment of the elements Ta, B, Mn, Pb and Zr is associated with lepidolite formation, i.e., the contents of these elements are higher in lepidolites than in muscovites.

Ba has more ore less the same values in both micas.

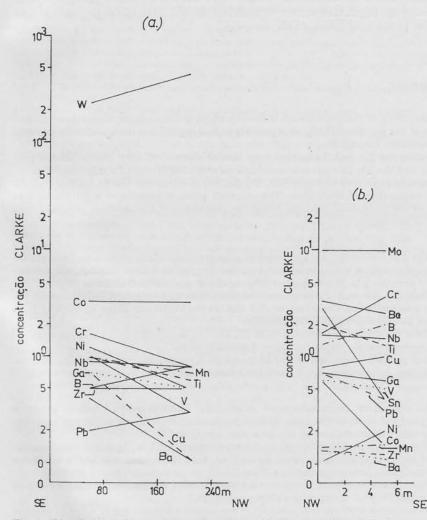
We also observe an increase of Be, Ga, Ti, Co, Mn and B from the lithium zone to the inner zone, with Ta, Pb and Cu behaving inversely. The elements Nb, Sn, W and Zr do not show an identical distribution in the two mica types. For example, in contrast to the trend in lepidolites, in muscovites Nb increases from the lithium to the inner zone. Nb, Ta and Be and Sn, which occur in high concentrations in the muscovite, recognized

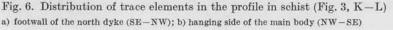
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in ore deposits containing mineralization of Nb and Ta minerals, of beryl and of cassiterite respectively, used as indicator elements (SHMAKIN et al. 1979).

The same way be considered in relation to lepidolites and biotites, which present the same characteristics, extending this rule to the micas in general. The Nb/Ta ratio has the lowest value in lepidolites, and in muscovites we observe a decrease from the outer to the lithium zone, corresponding to the Ta concentration in the minerals of the final phase of pegmatite formation.

Similarly, the biotites have more Nb than Ta (Nb/Ta = 8.4), i.e. an increase of the Ta in relation to the Nb in the later products of differentiation also corresponds to a decrease of biotites which do not occur in the lithium zone and are not frequent in the pegmatitic body. High quantities of Li in the original liquid are, probably, the reasons for the limited occurrence of biotites in rare metal pegmatites (ALEKSANDROV et al. 1985).





3.4. Trace element distribution in neighbouring rocks

Table 3 shows the trace element contents in the analysed rocks. These results show that in gneisses the elements W, Ta, Co, Cu and Nb have, on an average higher concentrations than the CLARKE ones. In schists, the mean contents of W, Be, Cr, Co and Mo exceed the CLARKE value.

The xenoliths of the pegmatite reveal, for the great majority of the elements, an enrichment compared to the schists. Ta, W, Co, Nb, Cu and Sn aureoles were identified in gneisses of the west dyke (Fig. 5); W, Co and less clearly Cr and Ni aureoles on the footwall and Mo, Be, Cr and less distinctly developed Nb, B and Ti aureoles at the hanging side of the main body (Fig. 6).

The primary aureoles are different at high and low exocontacts, showing the occurrence of metasomatic processes with different degrees of extension.

The neighbouring rocks (as well as the micas which were analysed in paragraph 3.3.) may thus represent an important source of material for the rare metal pegmatites and must be included in the evaluation of the reserves.

Summary

Through the examination of the geochemical behaviour of trace elements, in connection with the internal structure of the pegmatitic body, it is possible to find regularities in their distribution and relate them to pegmatite formation.

Although the elements Nb and Ta have a very similar chemistry, they show different geochemical features, and the Nb/Ta ratio is a useful indicator for the Nb and Ta mineralization.

The high contents of some rare elements (Nb, Ta, Be, Sn) in micas can also be used as an indicator for the corresponding mineralization. The observed primary aureoles of some elements in the exocontacts of the pegmatitic deposits, reveal the occurrence of metasomatic processes with different degrees of extension.

Zusammenfassung: Auf Grundlage der Untersuchung des geochemischen Verhaltens von Spurenelementen im Zusammenhang zur inneren Struktur der Pegmatite war es möglich, die Gesetzmäßigkeiten ihrer Verteilung in Beziehung zur Bildung der Pegmatite aufzuklären.

Obwohl die Elemente Nb und Ta sehr ähnliche chemische Eigenschaften haben, zeigen sie unterschiedliches geochemisches Verhalten, so daß das Nb/Ta-Verhältnis einen nützlichen Indikator für Nb- und Ta-Mineralisationen repräsentiert.

Die hohen Gehalte einiger Spurenelemente (Nb, Ta, Be, Sn) in den Glimmern können ebenfalls als Indikator für entsprechende Mineralisationen genutzt werden.

Die beobachteten primären Aureolen einiger Elemente im Exokontakt der Pegmatitlagerstätten offenbaren das Wirken metasomatischer Prozesse mit unterschiedlichem Grad der Ausdehnung.

Содержание: Исследование геохимического поведения элементов-примесей в связи с внутренной структурой пегматитов дало возможность выяснить закономерности элементного расспределения в зависимости от образования пегматитов.

Даже несмотря на то, что химические свойства элементов Nb и Ta очень сходны, эти элементы имеют различное геохимическое поведение, так что отношение Nb/Ta является полезным индикатором для минерализации Nb и Ta.

Высокие содержания некоторых элементов-примесей (Nb, Ta, Be, Sn) в слюдах также могут служить индикатором таких видов минерализации.

Рассматриваемые первичные ореолы некоторых элементов в экзоконтакте месторождений пегматитов отражают действие метасоматических процессов с различной степенью площадного развития.

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Manuscript received: 18th October 1988

Vortrag, gehalten anläßlich der Tagung "Beltrag der DDR zur geowissenschaftlichen Erforschung von Gebieten außerhalb von Europa" vom 4. bis 6. November in Köthen, veranstaltet von der Gesellschaft für Geologische Wissenschaften der DDR, Fachbereich Geologie, der Akademie der Wissenschaften der DDR, Zentralinstitut für Physik der Erde, Potsdam, und der Bergakademie Freiberg, Sektion Geowissenschaften

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