THE GENETICS (FORMATION) OF RHYOLITE OCCURRING IN THE RUDABÁNYA MTS. (NE HUNGARY) ON THE BASIS OF REE AND OTHER TRACE ELEMENTS

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ABSTRACT

The authors performed a genetical investigation on the rhyolite occurring in the Rudabánya Mts., previously studied from a mineralogical-petrographical aspect using data on rare earth element composition as well as other trace elements. Measurements were performed using neutron activation analysis completed by — in case of some elements — quantitative spectroscopical analysis. Based on the result of the measurements the rocks were assigned to four groups by the help of cluster analysis. On the basis of the relations of incompatible and less incompatible elements they could observe that the rhyolite was formed partly by a partial melting of the crust and partly by differentiation processes within the magma chamber. The considerable negative anomaly of Eu encountered is explained by fractionation of the plagioclase, i.e., this fact would equally support that the rock was formed from differentiated remnants of the melted rocks. The triangular diagram of Th-Ta-Hf/3 (which can be effectively used for acidic rocks as well) indicates that the formation of the rhyolite can be connected to the volcanism of a plate margin being consumed (subduction). The authors could observe in one sample an essential enrichment in U, Y and lanthanids caused by, according to their investigation, a considerably high fluor-apatite content.

INTRODUCTION

The regional distribution, mineralogical composition of the rhyolite occurring in the Rudabánya Mts. as well as its relation with the embedding rocks were studied in details by SZAKMÁNY et al. (1989). This rock can be traced on the surface as well as in several boreholes at a relatively small area in several spots. Its mineralogical composition includes dominantly resorbed — originally idiomorphic — quartz grains and albitized sanidine due to subsequent Na-metasomatism, as well as some plagioclase of albite-oligoclase composition. The mafic constituents are baueritized and/or chloritized biotite. As accessories we can find some zircon, apatite, magnetite and ilmenite in the rock. As secondary minerals we can find calcite, chlorite, sericite, clay minerals and haematite formed from the primary constituents. The original matrix of the rock must have been mainly vitreous, that subsequently crystallized and silicified.

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The rhyolite bodies are situated in slate dated by Radiolaria stratigraphy to Middle Jurassic period (GRILL and KOZUR 1986). In course of the analysis of the contact between the rhyolite and the slate it was proved that the rhyolite intruded the loose hardly consolidated sediments in the form of half-quenched lava, and it was — to some extent — mixed with the former.

In the present paper the results of a detailed genetical analyses of the rhyolite on the basis of REE and other trace elements distribution are discussed.

DETERMINATION OF THE REE AND SOME OTHER TRACE ELEMENT CONTENT

Rare earth elements (La, Ce, Nd, Sm, Eu, Tb, Tm, Yb, Lu) as well as the majority of other trace elements (U, Th, Hf, Ta, Sc, Rb, Cs) were measured by neutron activation analysis. The measurements were performed in the Nuclear Technical Institute of the Budapest Politechnical University by J. BÉRCZI. The irradiation of the samples took 2 hours and a GeLi detector was used for detection. Quantitative evaluation of the results was performed using Ru standard.

For the determination of the trace elements Zr, Nb, Y and Be a quantitative spectroscopical method was used. The measurements were performed in the Spectroscopical Laboratory of the Department of Petrology and Geochemistry of the Eötvös Loránd University by Mrs. NAGY. Experimental conditions for the measurements were: graphite electrodes, 1:1 mixture of the powder of the sample and powdered graphite, P alfa interior standard, direct arch current (8 A) for 1.5 minutes (till complete evaporation), detection by PGS-2 lattice spectrograph, 2nd order of spectra.

DISTRIBUTION OF REE AND OTHER TRACE ELEMENTS IN THE RHYOLITE

The REE and trace element composition of the rock samples analysed are presented in Table 1. REE data, normalized for chondrite are presented (in a grouping) on *Fig. 1.* using the most generally accepted norm values for chondrite proposed by HASKIN *et al.* (1968). The 17 samples analysed can be assigned into four groups and the average values of these four groups are presented on *Fig. 1.* Within the individual groups the measured values are very close to each other thus it is not necessary to present all the 17 individual measurements.

We can see on the diagram that the run-off curves for 3 groups are fairly similar, only group IV (comprising one sample only, marked 4R) shows different concentration values and the run-off for the curve is also different from the rest of the groups. The separation of sample 4R of group IV is also evident in case of cluster analysis and non-linear mapping procedures performed for 11 trace elements (La, Ce, Sm, Eu, Tb, Tm, Yb, Lu, U, Th, Sc) in 17 samples too (*Figs. 2, 3, 4* and Table 2). The cluster analysis and the nonlinear mapping was performed in the Hungarian Geological Survey by the help of L. O. KOVÁCS and P. G. KOVÁCS. Cluster analysis was made in two versions according to a weighted-pair group average and an unweighted average. The aim of applying different methods was to separate types within the rhyolite on the basis of trace element composition. As a result of the statistical analysis, four well separated units could be established on *Figs. 2, 3* and *4.* Especially remarkable here the unique character of sample 4R, representing group IV.

Table 2 gives a comprehensive account on the location of the samples. It is observable that all outcrops yielded samples for each groups proving that the rock

Table 1.

REE and other trace element content of the rhyolyte samples in the Rudabánya Mts (ppm).
For location of the samples, see Table 2.

	La	Ce	Nd	Sm	Eu	Tb	Tm	Yb	Lu	U	Th	Hf	Ta	Rb	Cs	Sc	Y	Nb	Be	Zr
2R	18	38	33	10	0.24	2.0	1.0	4.5	n.a.	6.7	20	4.3	2.2	166	n.a.	3.3	50	16	6	130
3R	19	43	n.a.	9.0	0.23	1.5	0.8	3.6	0.54	2.9	19	3.8	1.8	120	n.a.	3.0	60	20	10	160
4R	83	215	n.a.	39	0.93	9.4	5.5	33	6.5	155	4.7	1.1	0.7	n.a.	n.a.	1.0	600	40	5	150
5R	27	64	40	13	0.55	2.3	1.3	5.8	0.85	6.6	25	4.7	1.8	153	n.a.	2.7	100	20	10	200
6R	37	76	48	21	0.66	3.0	1.2	5.8	0.91	4.4	24	4.9	1.8	176	n.a.	4.0	60	<16	6	160
7R	21	41	n.a.	9.5	0.20	1.5	0.9	3.2	0.5	3.3	19	4.1	1.7	n.a.	n.a.	2.8	60	16	4	160
8R	11	27	n.a.	6.1	0.15	1.3	0.8	3.2	0.51	2.3	15	3.4	1.2	n.a.	n.a.	2.6	n.a.	n.a.	n.a.	n.a.
10R	29	63	41	12	0.29	1.8	0.6	3.6	0.61	4.2	24	5.1	2.1	n.a.	n.a.	3.5	40	<16	10	160
11R	10	23	n.a.	5.1	0.08	1.0	0.6	2.7	0.47	6.6	13	2.5	1.3	80	n.a.	1.9	60	<16	4	130
13R	16	28	n.a.	6.9	0.15	1.2	0.8	3.0	0.44	3.2	26	2.6	1.4	2.7	66	n.a.	60	16	7	160
14R	9.8	22	n.a.	6.4	0.07	1.1	0.9	3.2	0.49	9.5	13	2.7	1.7	2.3	103	n.a.	25	<16	n.a.	100
15R	14	26	n.a.	6.8	0.11	1.6	1.1	4.9	0.82	13	13	2.6	n.a.	2.3	205	n.a.	40	<16	n.a.	100
16R	35	76	68	14	0.37	2.6	1.4	3.8	0.6	3.8	26	6.0	1.6	3.8	88	3.4	25	<16	10	250
18R	10	31	22	6.0	0.08	1.4	1.0	3.0	0.44	1.8	14	2.7	1.5	2.1	53	4.4	60	20	14	160
19R	10	23	n.a.	5.9	n.a.	1.4	0.9	2.5	0.37	1.8	14	3.0	1.6	2.4	66	4.1	25	<16	10	100
20R	15	34	n.a.	14	0.21	1.4	1.4	5.4	1.0	15	11	2.1	1.1	1.7	41	2.8	60	16	8	100
21R	8.8	22	n.a.	5.7	n.a.	1.1	1.1	2.7	0.42	2.5	14	3.2	1.6	2.2	81	4.5	10	<16	n.a.	60
22R	15	33	37	9.5	0.32	1.6	1.6	4.4	0.42	2.5	18	3.7	1.7	2.6	119	5.2	40	16	10	160
23R	23	47	51	11	0.46	1.9	1.3	3.1	0.48	4.8	19	4.0	1.6	62	2.5	2.5	n.a.	n.a.	n.a.	n.a.

n.a.: not analysed
Analysis performed by J. Bérczi at the Nuclear Technical Laboratory of the Budapest Polytechnical Institute
(REE, Th, U, Ta, Hf, Sc, Rb, Cs) by NAA, and Mrs. B. Nagy, Petrological-Geochemical Department (Nb, Z, Be, Zr) by OES
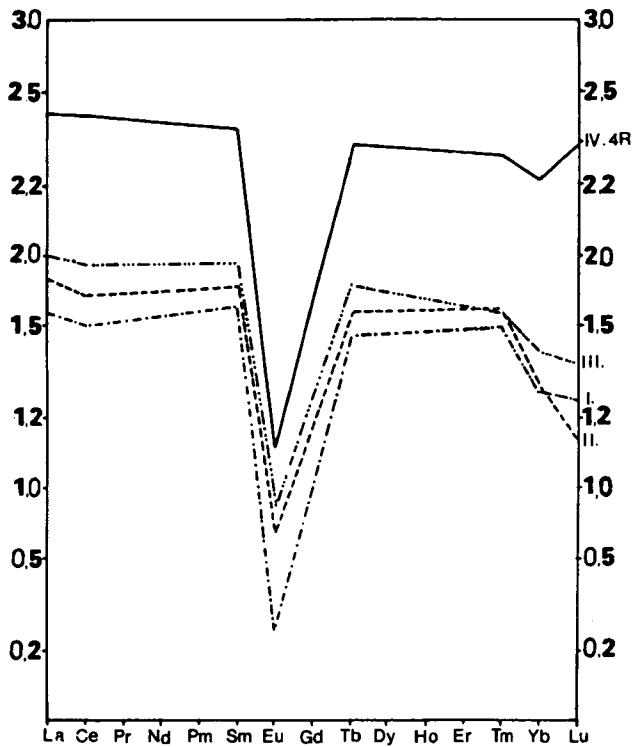
Fig. 1. Mean values normed to chondrite (after HASKIN *et al*. 1968) for the four groups formed on the basis of the REE content of rhyolites occurring in the Rudabánya Mts. For the location of the samples, see Table 2.
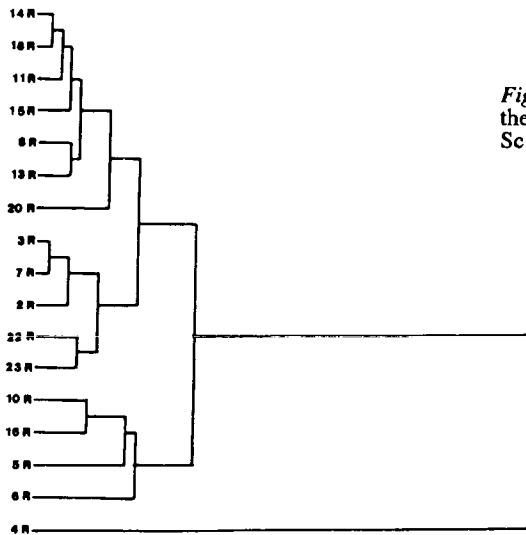

Fig. 2. Result of the cluster analysis using the concentration values of REE, U, Th and Sc (weighted-pair group average).
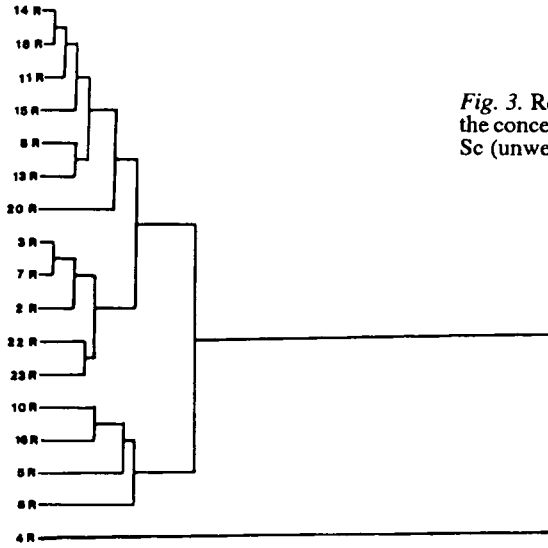
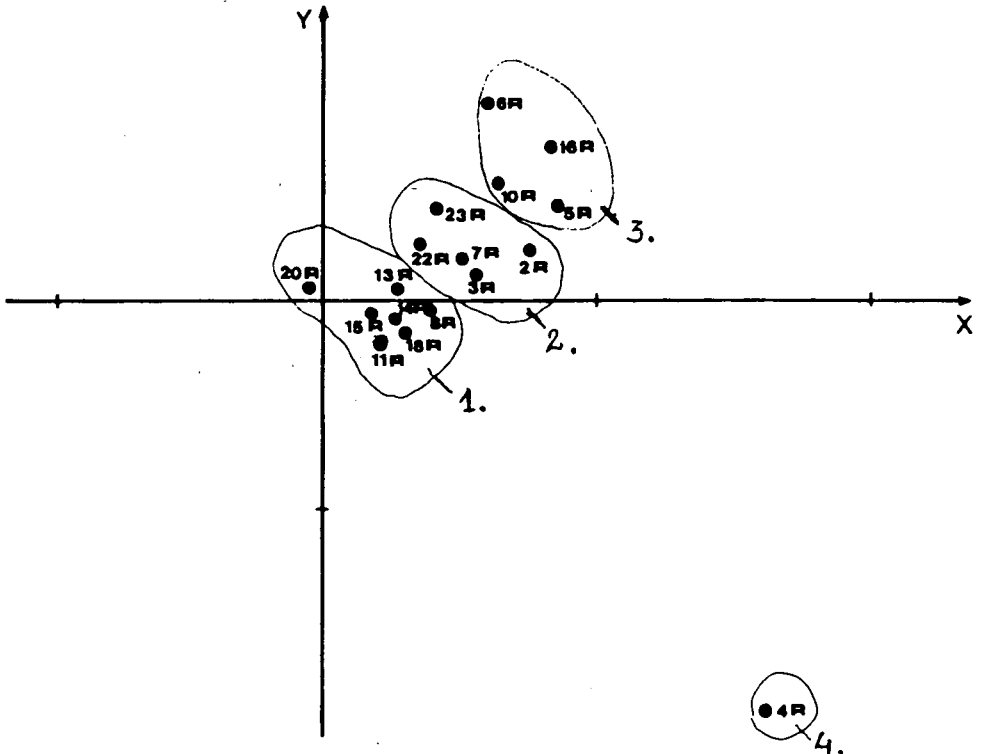*Fig. 3.* Result of the cluster analysis using the concentration values of REE, U, Th and Sc (unweighted average).

Fig. 4. Representation of the results using the concentration values of REE, U, Th and Sc by non-linear mapping method

bodies separated on the area belong for one lithological unit and reached their recent position due to posterior tectonic events. Vertically, we can observe some differences within the rhyolite; the trace element composition can help us to separate different levels within the rhyolite body. Sample 4R assigned to group IV is distinctly different from the samples of the other three groups, therefore it was analyzed in details at a later stage.

Groups formed by cluster analysis and the location of the samples

Group 1.	14R —	hill slope to the NW of Vadászháza
	18R —	S foothills of the plateau Balázstető
	11R —	Borehole Sz-10 at 31.8 m
	15R —	hill slope to the NW of Vadászháza
	8R —	SW side of the outcrop lying to the SE of Vadászháza
	13R —	outcrop lying to the SE of Vadászháza
	20R —	Dunnatető
Group 2.	3R —	Borehole Sz-10 at 55.8 m
	7R —	NE side of the outcrop lying to the SE of Vadászháza, close to the contact with the slate
	2R —	Borehole Sz-10 at 66.0 m, on the rhyolitic side of the contact zone
	22R —	Borehole Sz-10 at 121.7 m, on the rhyolitic side of the contact
	23R —	on the rhyolitic side of the contact zone at the outcrop to the SE of Vadászháza
Group 3.	10R —	silicified rhyolite, outcrop lying to the SE of Vadászháza
	16R —	hill slope to the NW of Vadászháza
	5R —	Borehole Sz-10 at 121.8 m, on the rhyolitic side of the contact
	6R —	outcrop lying to the SE of Vadászháza
Group 4.	4R —	Borehole Sz-10 at 44.5 m

GENETICAL CONCLUSIONS

The groups separated on the basis of analytical data are genetically related as proved by the stable ratio of incompatible elements (*Fig. 5.*, see TREUIL *et al.* 1977). The separation of the genetically related groups can be explained by means of another diagram (*Fig. 6.*), following the data on a considerably incompatible (Th) and a less incompatible (Tb) element within the rock sequence. On one axis, the ratio of the two elements are plotted against the concentration of the more incompatible element on the other axis. We can observe on the diagram that members of the sequence formed by partial melting are dispersed along a line of general position, while those of fractional crystallization along a line described by the following equation:

$$\frac{Th}{Tb} = constant$$

We can see on *Fig. 6.* that one part of the rhyolite in the Rudabánya Mts. was formed by partial melting of the crust (groups 1., 2. on Table 2 except for samples
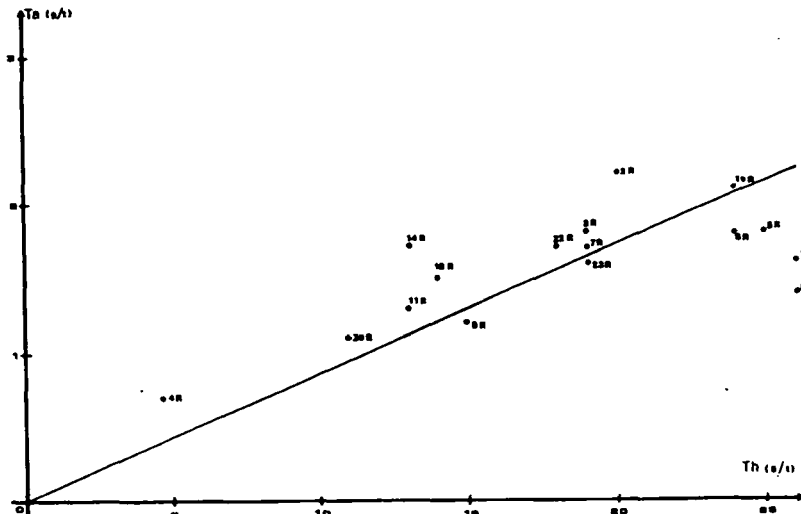
Fig. 5. The Th-Ta diagram of the rhyolites occurring in the Rudabánya Mts, (based on the method of TREUIL *et al.* 1977)
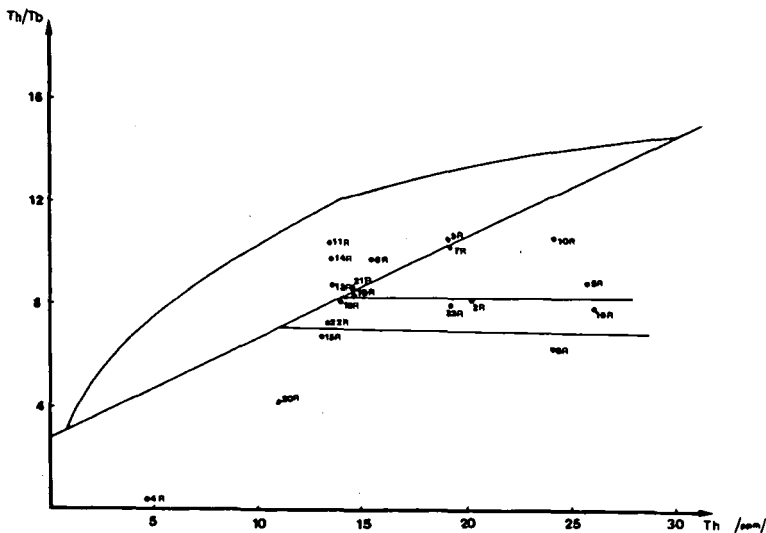

Fig. 6. The Th/Tb-Ta diagram of the rhyolites occurring in the Rudabánya Mts. (based on the method of TREUIL *et al.* 1977)
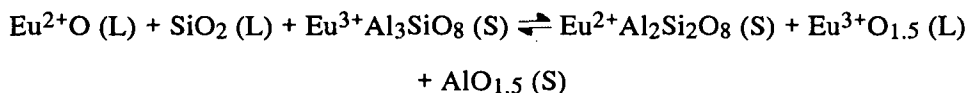
2R and 23R). Trace element concentration in these samples is generally very low. In case of another part of the samples (group 3. in Table 2), the values are scattered along the line Th/Tb = constant, that is these rocks were formed as a result of differentiation processes within the magma chamber. Highest concentration of trace element (REE, Sc, Ta, Th, Hf) were found in these samples, (see Table 1).

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In course of the evaluation of the REE analytical results, we found a considerable Eu-anomaly in the rock (*Fig. 1*).

Europium can occur in the rock in two forms, namely as Eu^{2+} and Eu^{3+}. Data published by WEIL and DRAKE (1973) as well as GERASIMOVSKY *et al.* (1976) suggest that in course of the cryistallization of magmatic melts, REE are seemingly accumulated in minerals rich in Ca (mainly, plagioclase) where they can subsitute Ca in an isomorphic way. This can even be furthered by Na occurring in the melt, by following way:

$$REE^{3+} Na^+ \xrightarrow{\text{Isomorphic substitution}} 2\ Ca^{2+}$$
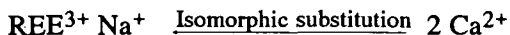
It is generally stressed that the formation of Eu-anomaly is basically related to crystallization of plagioclase. According to WEIL and DRAKE (1973) the distribution of Eu^{2+} and Eu^{3+} between the plagioclase (S) and the melt (L) can take place according to the following reaction:

$$Eu^{2+}O\ (L) + SiO_2\ (L) + Eu^{3+}Al_3SiO_8\ (S) \rightleftharpoons Eu^{2+}Al_2Si_2O_8\ (S) + Eu^{3+}O_{1.5}\ (L)$$

$$+ AlO_{1.5}\ (S)$$

This is one of the possible ways describing the Eu^{2+}—Eu^{3+} distribution behaviour in the plagioclase — melt system. On the basis of crystal chemistry of feldspars it is presumable that Eu^{2+} is preferred in place of large cations in the feldspar structure than Eu^{3+}. That is, Eu^{2+} can substitute Ca in the plagioclase. The model propesed by WEIL and DRAKE (1973) as well as practical observations show that negative Eu anomaly tends to increase in the plagioclase by the increase of Na or the decrease of Ca concentration, that is, we have a strong positive correlation between the negative Eu anomaly and the albite content of the plagioclase.

WEIL and DRAKE (1973) emphasized that the extent of Eu-anomaly is considerably related to the oxidation potentials of the magma during crystallization. For the formation of a substantial amount of Eu^{2+}, a suitable reductive medium is needed, i.e., the Eu-anomaly hints at the oxidation-reduction relations of the environment during rock formation. In other words, the greater the negative Eu-anomaly, the crystallizing medium was possibly more reductive.

The presence of a negative or positive Eu-anomaly as well as the lack of this phenomenon has a litho-genetical significance. Basaltic magmas of primary /mantle/ origin have no Eu-anomaly, therefore wherever we find this phenomenon in a rock we can suppose certain differentiation processes in course of it's formation. FREY *et al.* (1984) and GERASIMOVSKY *et al.* (1976) considered — on the basis of rock sequences ranging from basalt to rhyolite — a negative Eu-anomaly of rhyolites as a sign of plagioclase fractionation. While plagioclase is present as a fractionating phase, it has a distinguished (favourable) position, in respect of the uptake of Eu compared to other REE. In the plagioclase feldspars of early crystallization Eu is selectively enriched, therefore its relative quantity is essentially decreased in the residual melt.

The extremely high negative Eu-anomaly of rhyolites in the Rudabánya Mts. proves that the rock must have originated by the crystallization of a residual melt. Plagioclase as the dominant mineral for the enrichment of Eu was already separated from the system (that is, the fraction rich in plagioclase had already been crystallized previously). During the crystallization of the rhyolite the environment was reductive, marked by the relatively larger quantity of Eu^{2+} substituting the Ca in the crystal lattice of the plagioclase.

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Previous studies SZAKMÁNY *et al.* (1989) demonstrated that there is only a very small amount of primary plagioclase (albite, oligoclase) present in the rock. This has been corroborated by chemical analyses as well. Secondary albitization is basically proved by the very low CaO content and the relatively high amount of Na_2O. CaO is more abundant in those rocks only, where a considerable amount of calcite appeared due to secondary processes.

Another apparent feature of the REE analyses is the small enrichment of heavy lanthanids (*Fig. 1*). According to FLEISCHER (1965) these elements are mainly enriched in granate, zircon, apatite and xenotim. WATSON (1980) investigated the role of zircon in the enrichment of lanthanids by means of crystallization experiments from melts. He could determine experimentally a distribution coefficient (D) for zircon / melt for the different rare earth elements and found that for the heavy lanthanids, the value of D was higher; that is, zircon enriched heavy REE more that the light ones. Zircon is relatively frequently found in the rhyolite of the Rudabánya Mts. At the same time the samples contained a considerable amount of fluor-apatite as well (in some samples, especially in 4R, in essential quantities as treated in details below). Consequently, the heavy REE encountered are concentrated mainly in these two minerals.

The triangular diagram containing the values Th-Ta-Hf/3 was applied first by WOOD *et al.* (1979). The basic advantage of the diagram compared to previous similar ones is, that — apart from the distinction of basic rocks — it can be effectively used for the separation of acidic rocks of different genesis as well. More precisely, acidic rocks of calc-alkaline and crustal origin can be separated from pure (uncontaminated) acid rocks produced as a result of fractional crystallinization (as well as from E-type ridge basalts and intra-plate basalts, too). Another advantage of the diagram, and the reason of its applicability to acidic rocks is based on the fact that the points representing the individual acidic magmas are situated near to the relevant points of those basic melts from that they were derived through differentiation processes.

During fractional crystallization the Th content of the residual melt is relatively enriched, i.e, points tend to shift within the triangular diagram towards the apex of Th. The reason for this can be found in the fact that oxides of Fe and Ti, notably clinopyroxene and other minerals (where Hf and Ta are accumulated typically better that Th) appear among the first solid phases in acidic and intermediate magmas. The increase of the ratios of Th/Hf, Th/Ta etc. in the residual melts, however, do not obscure their relation with the original basaltic melt.

Presenting the analytical data on the rhyolites of the Rudabánya Mts. in a Th-Ta-Hf/3 diagram (*Fig. 7*), the points range into field D and its immediate
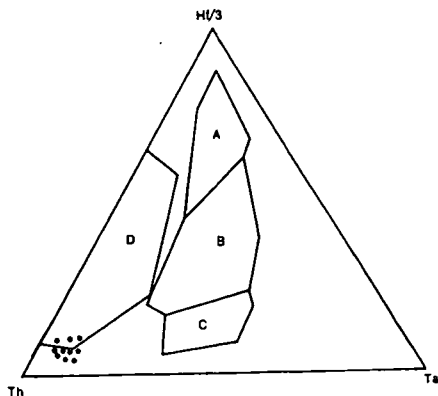

Fig. 7. The Th-Ta-Hf/3 triangular diagram of the rhyolites occurring in the Rudabánya Mts. (based on the method of TREUIL *et al.* 1977):
A: N-type ridge basalt
B: E-type ridge basalt
C: intra-plate basalt
D: consummated plate margin (subduction)

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surroundings. Accordingly, the rhyolite can be connected to the volcanism of a consummated plate margin (subduction), that is a typical calc-alkaline formation.The points are very far from field C, which represents intra-plate basalts and their derivatives.

The results obtained agree well with the geological model set on the formation of the mountains and its geohistory by GRILL *et al.* (1984). RÉTI (1987) could prove the presence of an ophiolite sequence within the mountain. According to the geological model the oceanic crust was formed here in the Middle Triassic. The most active phase of the spreading could be supposed for the Ladinian-Karnian period. By the Norian stage spreading got much slower (possibly stopped) because the Hallstatt and Pötschen type limestones — denoting a sedimentary basin in state of equilibrium — are found in large areas. The authors suppose the subduction of the oceanic crust, proceeding from South to North, by the Jurassic as well as the consequent volcanic activity along the consummated plate margin.

The age of the slate embedding the rhyolite was determined by the help of Radiolaria analyses as Middle Jurassic (GRILL and KOZUR 1986). Analysing the contact zone between the rhyolite and the slate SZAKMÁNY *et al.* (1989) could detect a thermal contact, thus the age of the rhyolite can be fixed to Middle Jurassic as well, corroborating the evolution scheme outlined above.

EVALUATION OF THE ANOMALIC TRACE-ELEMENT CONTENT OF SAMPLE 4R

Studying Table 1 as well as *Figs. 1—4* carefully, we are immediately faced with the different character of the rhyolite sample marked 4R (borehole Szalonna—10, sample taken at 44.5 m). In this sample, the concentration of light lanthanids is 3—4 times, and that of heavy lanthanids as well as Y about 10 times more than in the rest of the samples. Enrichment of U in the sample is also considerable (155 g/t) while the concentration of Th and Hf is decreased to third-quarter of the quantity observed in the other samples. It is worth to note that the quantity of Zr is not considerably decreased, that is, the geochemically strongly related Zr and Hf are detached from each other. This is possibly rooted in the fact that the bulk of zirconium in this sample is stored not in the zircon but other minerals (this was supported by microscopic analysis of the sample where zircon was found in very small quantities).

For tracing the reason of the essential enrichment of REE, Y and U, we performed a more detailed analysis using X-ray diffraction and electron microprobe analysis as well. In course of evaluating the thin section obtained from the sample it was found that the sample contains an essential amount of apatite, present as accessory mineral, in the other rhyolite samples as well (*Fig. 8*).

For a more detailed analysis, the apatite was enriched by separating heavy minerals and the remnants were used for X-ray diffraction analysis. The evaluation of the X-ray diffractogram could unambiguously prove that this mineral is fluorapatite. This is also supported by the chemical analysis of the sample (high phosphorous and fluorine content, Table 3). (The result of the chemical analysis was rendered to our disposal by Prof. J. KISS). We can suppose that this mineral is the host for the great amount of REE, Y, and U found by trace element analysis. The role of apatite in REE enrichment is uniformly stressed by literature as well (FLEISCHER 1965, HENDERSON 1984). Rare eart elements seemingly show special affinity to fluorine as proved by BALASOV (1976) (in PANTO 1980). REE-fluorides, -phospates and -carbonates are relatively ill-soluble compounds, and this fact can also explain the role of apatite in the enrichment of REE.

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TABLE 3.

Chemical composition of the rhyolite sample marked 4R.

SiO_2	52.40 %
TiO_2	0.05 %
Al_2O_3	4.40 %
Fe_2O_3	0.77 %
FeO	0.78 %
MnO	0.04 %
MgO	1.60 %
CaO	19.30 %
Na_2O	1.30 %
K_2O	0.27 %
P_2O_5	12.40 %
$-H_2O$	1.20 %
$+H_2O$	2.40 %
CO_2	2.30 %
F	0.96 %
$(RFF)_2O_3$	0.08 %
	100.45 %

The analysis was performed at the analytical laboratory of the MÉV.

Electron probe microanalysis of the sample has shown — apart from apatite — some traces of monacite as well which is also well known as a mineral important in the accumulation of REE, Y and U (*Figs. 9—12*).
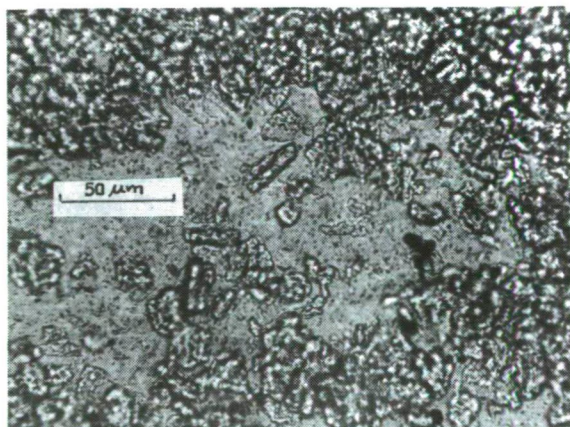
Fig. 8. Crystals of apatite in the rhyolite sample 4R (borehole Sz—10 at 44.5 m, 1N, scale 300x)
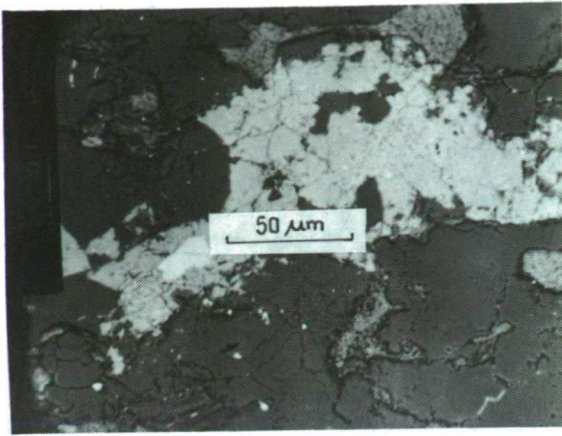
Fig. 9. Composition electron micrograph on apatite (a) and monacite (b) from sample 4R, scale 315x
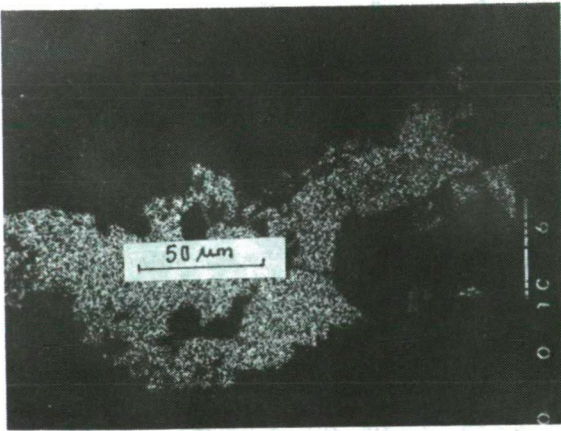

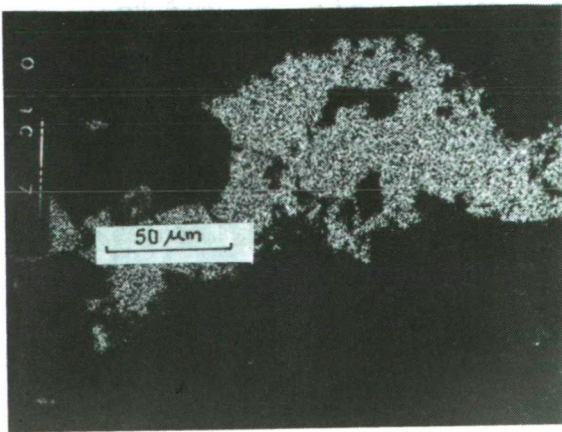*Fig. 10.* P distribution on the area presented on *Fig. 9.* K_α (scale 315x)

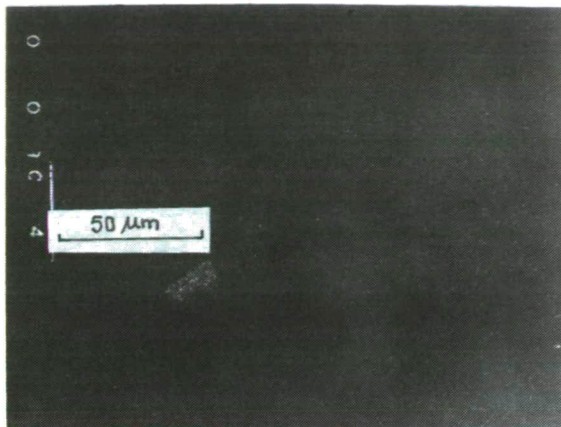*Fig. 11.* Ca distribution on the area presented on *Fig 9.* K_α (scale 315x)

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Fig. 12. Ce distribution on the area presented on *Fig. 9.* K$_\alpha$ (scale 315x)

SUMMARY

On the basis of trace element analysis of the rhyolite occurring in the Ruda-bánya Mts. we can deduce the following genetical conclusions:
— According to the diagram introduced by WOOD *et al.* (1979) the rock can be considered as the product of a volcanism in consummated plate margin (subduction zone).
— The rhyolite was formed, following the partial melting of the crust during differentiation processes. This fact is corroborated by the points fitting to relevant places on the diagrams presented by TREUIL *et al.* (1977) as well as by the considerable Eu-anomaly.
— At a certain level of the rock, a considerable enrichement of REE, Y and U was encountered.
— Results of the geochemical interpretation seem to fit well to the geological model and evolutionary scheme proposed by GRILL *et al.* (1984).

A more detailed view on the genetics of the rock, notably, the original composition of the primary melt and its evolution can be expected from further detailed studies. First of all other differentiation products of the melt (basic-neutral rocks) should be spotted within the mountain as well as its surroundings. Considering the nappe structure of the region this may not be an easy task because these units can now be several hundred kilometres far from each other due to considerable lateral movements.

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