

GEOCHEMICAL STUDY OF LEPTINITES FROM STEJERA (ROMANIA)

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Abstract. In the Țicău Mts., (NW Transylvania, Romania) an extended level of mica-poor, quartz-feldspar gneisses (so called leptinites) occurs. In this paper, the analytical data, obtained during the exploration of the Stejera kaolin deposit for an extensive geochemical study were utilised. After a short introduction in the geological setting and in the petrography of leptinites, the authors, using the Niggli's and other discrimination diagrams, demonstrate the igneous origin of these rocks. The statistical treatise of the analytical data and the petrochemical calculations resulted, that there were two or more petrographic and genetic rock-types. These rock-types were originated from magmatic differentiation processes (i.e. between dacitic and rhyolitic ones and their pyroclastic products), from mixing up with epiclastic materials and from further (postmagmatic) transformations (loss of alkalies and residual enrichment in Al_2O_3).

Comparing the normative minerals, which are stable in medium stage of the metamorphism with the real mineralogical composition of the leptinites, it seems to be sure, that the chemical-mineralogical equilibria was accomplished. Finally, the use of some immobile elements permits to separate the original magma types, the same ones, which were recognised by petrochemical calculations. With the immobile elements (namely Rb, Y and Nb), the geotectonic setting of these rocks determined the separation line between the syn-collisional and volcanic-arc domains.

Key words: NW Transylvanian Crystalline Islands, Sylvanides, Țicău Mts, Metaigneous Rocks, Leptinites, Chemical analyses, Trace elements, Discrimination diagrams.

The Țicău Mts. are situated in the NW part of Romania, at a 30 km distance from Baia Mare. It is one of the metamorphic island-like massifs of NW Transylvania (fig. 1). In our previos papers (Kalmár, 1994; Szederkényi et al., 1995; Kalmár and Kovács-Pálffy, 1996), we have identified this area with the border zone of the Tisia-realm, where the metamorphic basement of the Great Hungarian Plain outcrops. Thus, the detailed, petrographic and geochemical studies broaden the information about the basement, among others for identify some prevariscan structures. For this reason, we considered, that it is important to study the rock series with presumptive igneous

origin, which appear in the Țicău Mts.



Fig. 1. The island-like crystalline masifs of NW Transylvanie. 1. Preluca; 2. Bâc; 3. Ardud; 4. Heghieș; 5. Măgura Șimleului; 6. Mezeș; 7. Rez. Cr. Outcropping metamorphic rocks. In nframe, Țicău Mts.

One of the peculiarity of the Țicău Mts. is the large extension of quartz-feldspar (and small amount of mica)-bearing, massive or banded rocks, defined in the metamorphic petrology as Quartz-feldspar gneisses, Aplite-gneisses, Leuco-gneisses or Leptinites. Genetically, the term „orthogneiss” could be accepted, if the igneous origin of the protolith was proved unequivocally. In this matter, both in the past (Dimitrescu, 1963) and in the present (Denuț, 1999), some uncertainties have arisen, so we considered to be necessary to start a detailed geochemical study of this rocks based on a great amount of analytical data. The aim is not only to demonstrate the igneous origin, but after knowing this origin, to evaluate the geotectonical role of this igneous event in the basement's history.

1. HISTORY OF THE RESEARCHES

The Țicău Mts. appear in the regional map of Hauer & Stache (1863) and in the studies of Hofmann (1887), Koch (1898) and Szádeczky (1930). It is important to mention the studies about all of the crystalline massifs made by Kräutner (1938, 1940) and Dimitrescu (1962, 1963). Following the prospection of Țicău (Kalmár, 1969), with

the occasion of the exploration of the Stejera kaolin deposit, hundreds of complete chemical analyses were executed, in the majority about the material of the weathered crust of the leptinites (Pop et al., 1977). The mineralogy and geochemistry of the alteration processes were accomplished by Kovács-Pálffy & Földvári (1993) and Kovács-Pálffy (1994).

The question of the location of the Țicău Mts. in the North-Transylvanian area, respectively in the Carpathian one, was raised first by Lăzărescu (1965). In the structural interpretation of Săndulescu (1984), the NW-Transylvanian crystalline massifs were included in the Western Dacite Unit of the Carpathian Great Structural Unit. The paper of Zincenco et al. (1990) accepts this option, separating the „Sylvanide” subunit („Sylvanic Belt”) to which the Hercynic metamorphic formation of the Țicău Mts. belongs.

2. THE GEOLOGICAL SETTING

As we said, we consider to Someș Platform, with the island-like crystalline massifs, as well together with the northern part of the Apuseni Mts. as parts of Tisia Realm or Tisia microplate.

Based on the detailed mapping data, Kalmár & Kovács-Pálffy (1996) subdivided the metamorphic rock succession of the Țicău Mts., the part of Sylvanide belt, i. e. the Țicău Formation in two members.

Considering the first member, the *Stejera-Cheud* magmatogene one, the leptinites which constitute the subject of our study, form the lowest level. Between the next mica-schist and micaceous quartzite levels, other two leptinite levels are imbedded. In the upper, *Chelița-Hagău* member, few thin leptinite levels occur, too.

Resetting the blocks which were displaced by the (far the greatest part) Tertiary aged fault system, the metamorphic formations are being arranged in a northward sinking perisyncline. Now, it is difficult to decide, whether this one should be a part of a normal structure or represents a reverse, overfolded succession. The detailed field observations show only, that the primary schistosity (S_1) develops parallel with the surfaces which separate the several rock types (S_{str}), i. e., the schistosity has followed the stratification surfaces ($S_{str} \approx S_1$).

The detailed petrographic research show at least two progressive metamorphic recrystallizations, separated by a regressive phase. Despite of this intensive processes, the strata, built up from different protoliths, preserved their identity, in other words, the metamorphism has not „mixed” the initial stratiform build-up.

During the Upper Cretaceous – Paleogene continental phase, the metamorphic succession was submitted at intensive weathering processes. By the alteration of the leptinites, an argilised crust was formed, which constituted the Stejera kaolin deposits.

The crystalline basement was covered by Senonian, Eocene and Badenian sedimentary deposits.

3. SAMPLES AND METHODS

During the exploration of the Stejera kaolin deposit, by the gallery and pit

sampling, approximately 250 channel samples were analysed. In conformity with the Romanian raw material standards, the samples were analysed on the following basis:

- a). In the dried and milled samples: Fe_2O_3 , whiteness index;
- b). In the dried, crushed and leached samples, the separation of the <0.064 mm fraction;
- c). In the $\phi > 0.064$ mm fraction, total chemistry and whiteness index;
- d). In the $\phi < 0.064$ mm fraction, total chemistry and in each case, trace element analysis;
- e). In 16 fresh leptinite samples, we have determined, also, the concentration of immobile elements.

We selected the analyses of the $\phi > 0.064$ mm fraction, in which the loss of ignition did not surpass 2%. The X-ray analyses demonstrate, that in this samples, the secondary minerals occur in an insignificant amount, thus, they are adequate for geochemical studies as fresh, unaltered leptinites.

The L.I. $> 2\%$ resulted also by the petrochemical calculation, since all of the rock samples contain a little amount of micas with OH^- .

The oxydic components of the analyzed main elements were determined by classic, gravimetric and titrimetric methods (at the Laboratory of the Non Metalliferous Mining Research Institute, Cluj Napoca, 1978-1984).

The analytic data were submitted on the statistical and petrochemical calculations. Some petrochemical and discrimination diagrams were drawn, too.

4. THE PETROGRAPHY OF THE LEPTINITES

The leptinites of the Țicău Mts., especially which occur on either side of the Stejera valley are massive or slabby rocks, with thin schistose inbeddings. Both on a macroscopic or microscopic scale, the leptinites show oriented aspect, on the one hand, because the presence of the micas which form plainy or slightly undulated films; on the other hand, due to alteration of the parallel or lens-like quartz- and feldspar-rich layers. In the thick banks, rather the no oriented, massive rock type appears, in which the mica sheets have a randomly disposition.

The structure of the rocks is granoblastic, mosaic-like, „aplitic”, (it is so called *Pflasterstruktur*). The grano-lepidoblastic structure appears subordinately, only in the mica-rich zones (Pl. I., photo 1).

The leptinite is constituted by 80–95% quartz+potash feldspar+plagioclase.

The quartz appears as 0.2–0.5 mm large, limpid, polygonal, undulatory extincting grains or as coalescent, 0.5–2 mm thick and few cm large, plate lens-like, polycrystalline separations, with wavy separation surfaces, which bear many dark inclusions, microfractures oriented perpendicularly at the surfaces and present curtain-like or mosaic-like undulatory extinction. In a few samples at the quartz-feldspar border, myrmekite concrescences were observed.

The majority of the feldspars appear as 0.1–0.3 mm large, mosaic-like, polygonal grain assemblage, with straight borders which form 120° one with other. They are represented by microcline (Pl. I., photo 3) and oligoclase in equal amounts. In both feldspars, drop-like quartz and small, platy muscovite inclusions appear.

Into the thin granular quartz-feldspar groundmass, mainly in the biotite-bearing leptinites, 1–5 mm large, often broken feldspar-porphyroblasts appear: they are the so-called *checkboard-albite* (Pl. I., photo 2). It should be mentioned, that the albite represents only the smaller part of these porphyroblasts: they are a complex crystal-agglomeration, in which the oldest, dull, island-like plagioclase was substituted by crossbarly twinned microcline, and later, the microcline was replaced by albite, on the borders and along the fracture lines. The porphyroblasts include quartz (drops and veinlets), biotite sheets, and on the border of the albite, small epidote grains and chlorite appear. The twins of microcline and albite show a checkboard-like display, that partly, justifies the name of this structure.

In some fractured zones of the rock, or in the pressure-shadow zone of the quartz lenses, large crystallised albite appears, in association with limpid quartz veins. The albite shows fine-as-hair twins, rather with grass-green chlorite rosetts. This albite is a secondary formation, that likely has the same age as the albite which substitutes microcline.

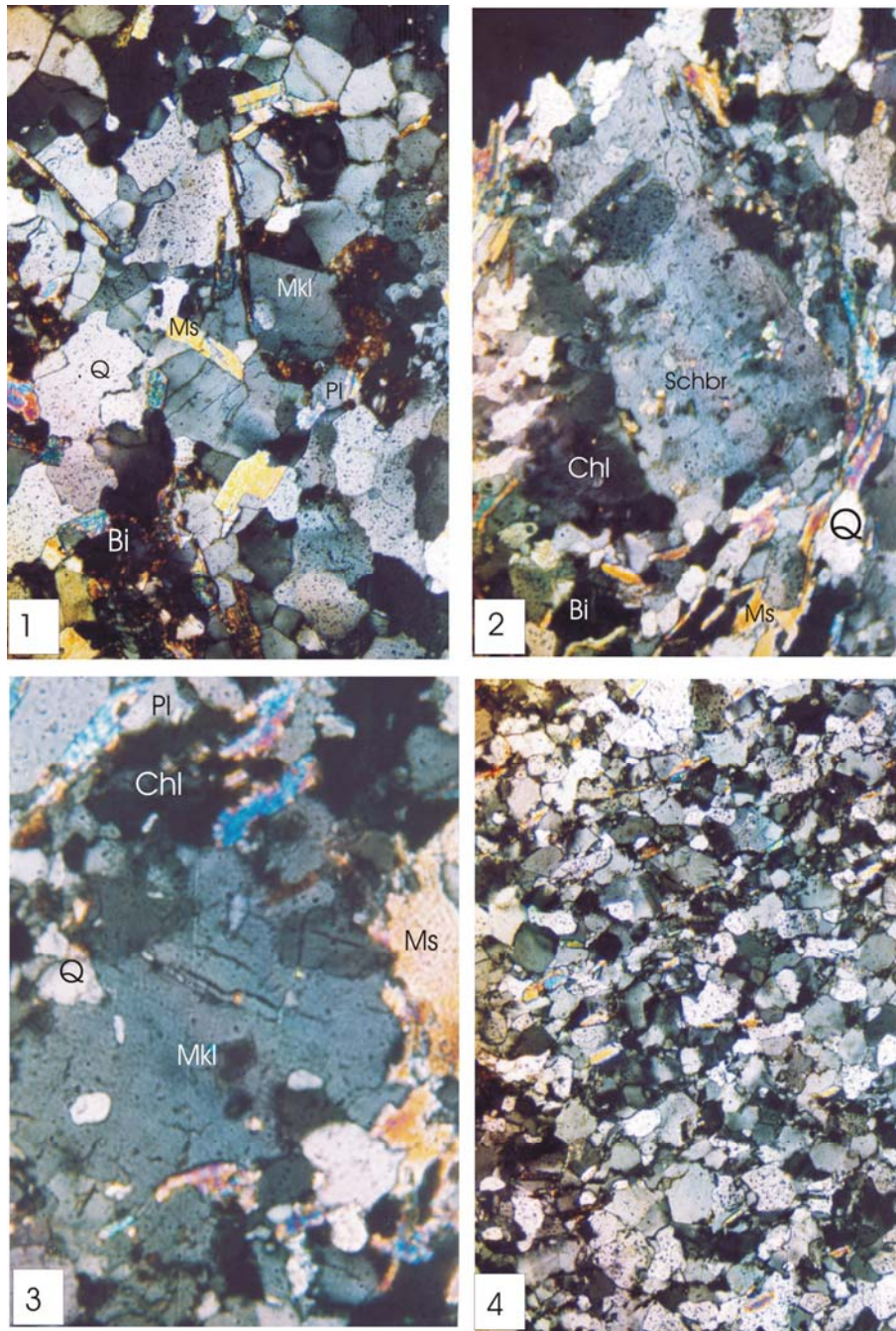
The amount of the micas rarely surpasses 10%. They appear as individual, oriented or randomly disposed sheets, weakly waved films, thin layers on lenses, frequently on the border of the quartz lenses. The greater part of the muscovite appears as inclusions in the feldspar grains, or as biotiteless films. The biotite is light brown, medium pleocroic, unaltered, often with opaque, kelyfitic rims, with chloritisation along the basal cleavage. Rutile and pleocroic zircon inclusions are rarely.

In the mica bearing gneiss, 2–5 mm large, plate-like, greenish, strongly pleocroic, limpid, neomorphic biotite appears, crosscutting the schistosity plane at 25–45° (“*Querbiotit*”). In the same rock, near to the mica lenses, small, clear garnet-grains (almandine) appear, forming nests or strings. The accessory minerals are represented by zircon, apatite, rutile, titanite, ilmenite and magnetite; the secondary ones, by sericite, chlorite, calcite, clay minerals, gypsum, pyrite, chalcedony and opal.

Based on the quantity of the micas and the structural features, two types of the leptinites could be distinguished:

The first type is the *biotite-muscovite leptinite*, in which the amount of the biotite reaches to 12%; the granoblastic and the grano-lepidoblastic structure predominate, as well as the massive or weakly schistose textures. It forms a continuous, 10–20 m thick banks, with 10–50 cm thick slabs or lenses separated by 1–3 cm thick, mica-rich gneisses or micaschist imbeddings. The colour of the rock is grey or greenish-grey.

Imbedded into the biotite-muscovite leptinites, 10–50 m large, platy, mushroom-like lenses of white, biotiteless, (often micaless) „*aplite-gneiss*” appear. They show a granoblastic-microblastic structure (Pl. I., photo 4) and massive, banded texture, due to



the alternation of greyish quartz layers and the white, porcelain-like feldspar ones. The amount of the micas is below 1%.

In the Stejera leptinite level, some graphite-bearing quartzites, micaceous quartzites and two-mica+garnet bearing micaschists are imbedded. The boreholes No. 302 (Vâlcea Secării), placed on the leptinites, traversed a 4 m thick amfibolite with green hornblende, plagioclase, titanite and garnet. In the micaschists which constitute the footwall of the leptinites, not so far from the Stejera kaolin deposit (Vâlcea Groșilor), a micaschists level with staurolite porphyroblasts appears.

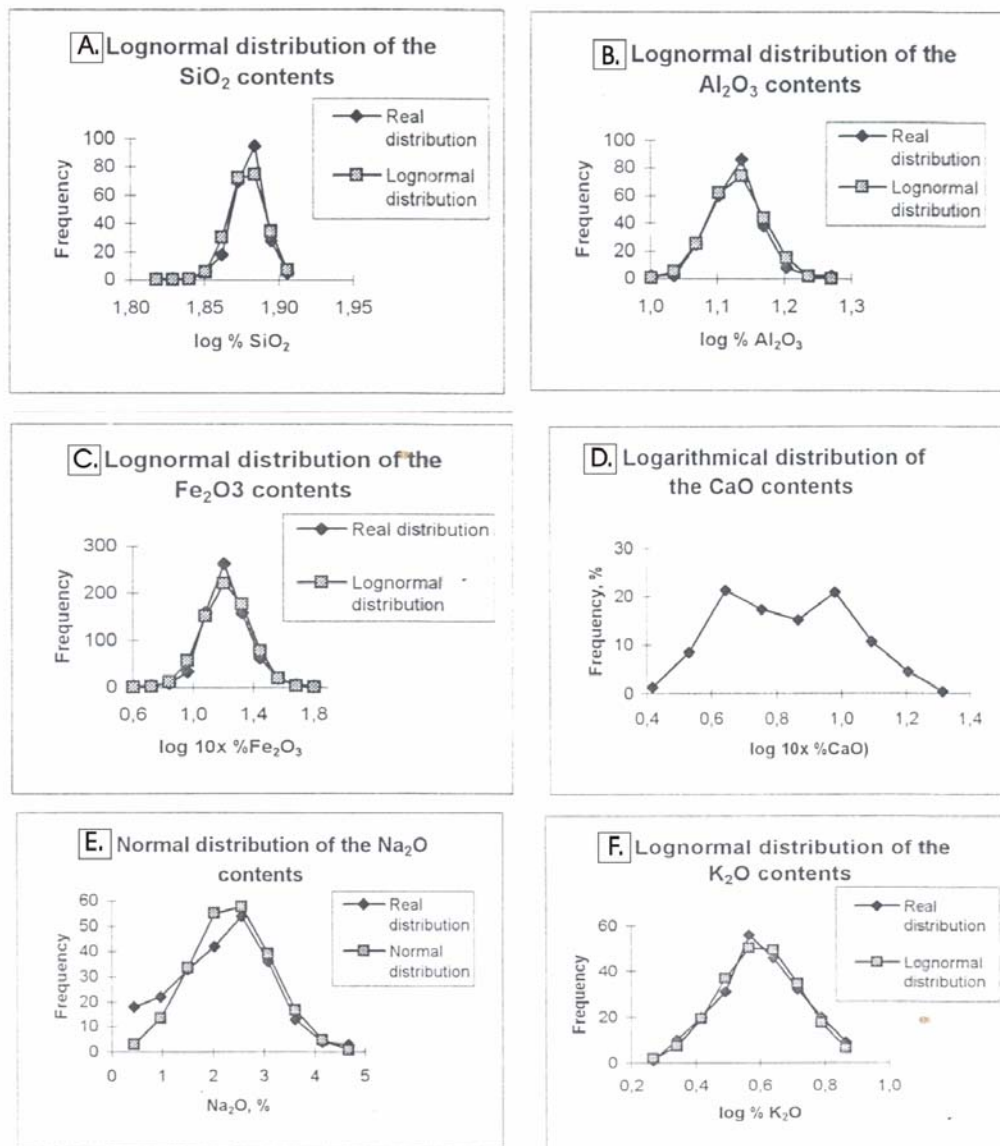


Fig. 2. Distribution of the main elements

Based on the association of the above presented minerals, the main phase of the metamorphism was the Barrowian type, medium stage, that culminate between the *Biotite-in*, *Staurolite-in*, *Almandine* and *Hornblende+30%An* isograds. The main metamorphic event was followed by a slightly regressive phase (chlorite+epidote +albite±phengite mineral assemblage) and by late, postmetamorphic fractural deformation of the metamorphic rocks (Kalmár, 1994a).

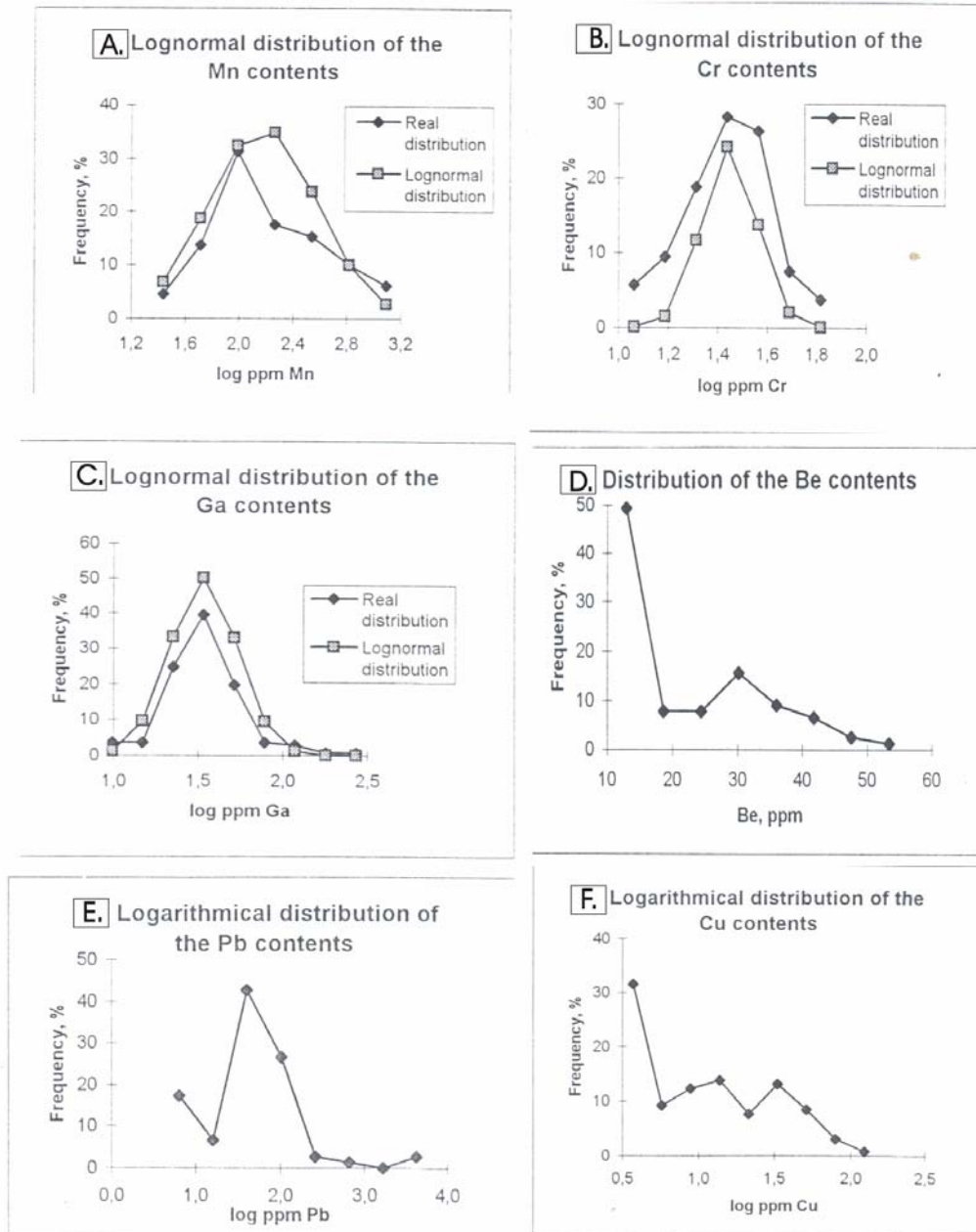


Fig. 3. Distribution of some trace elements

5. THE STATISTICAL DISTRIBUTION OF THE ROCK-FORMING AND TRACE ELEMENTS

The aim of the statistical analyze was to obtain an image about the homogeneity (or inhomogeneity) of analytical data, explaining the distribution of them using our field and mineralogical observations

The main data of the analyses of the rock-forming components and the trace elements are presented in the tables 1. The distribution of the classes of the contents, calculated with the Sturges formula were examined in the distribution diagrams of fig. 2. In case of the normal and lognormal distributions, we applied the χ^2 test.

Table 1. Statistical parameters of the rock-forming components

Component	Min, %	Max, %	Aver. %	Dispersion.	χ^2 calc.	χ^2 admiss.	Distribution
SiO ₂	64.88	81.16	75.71	2.07			
log SiO ₂	1.81	1.91	1.88	0.01	17.04	18.75	lognormal
Al ₂ O ₃	9.66	19.10	13.50	1.19	26.34	18.48	near normal
log Al ₂ O ₃	0.98	1.28	1.13	0.04	16.93	18.48	lognormal
Fe ₂ O ₃	0.35	6.36	1.76	0.67			
log Fe ₂ O ₃	0.54	1.80	1.22	0.15	38.70	21.67	lognormal
MgO	0.01	1.72	0.51	0.40			bimodal
log MgO	0.70	3.24	2.41	0.69			polymodal
CaO	0.23	2.25	0.75	0.34			polymodal
log CaO	0.36	1.35	0.83	0.19			bimodal
Na ₂ O	0.17	4.84	2.16	0.94	23.23	18.48	normal
log Na ₂ O	0.23	1.68	1.27	0.26			bimodal
K ₂ O	1.70	7.75	4.14	1.22			bimodal
log K ₂ O	0.23	0.89	0.60	0.13	5.22	18.48	lognormal

Number of analyses: Fe₂O₃ 717; the other components :225.

From the rock forming components, SiO₂ and Al₂O₃ show a regular, normal and lognormal distribution, similar to an ideal Gaussian one. The iron-oxide shows a lognormal distribution. The Na₂O presents a normal distribution, but in the logarithmic scale two maxima appear, in the domain of the small contents (below and above 0.7% respectively). In turn, the logarithmic scale „smooths” the two maxima of the K₂O, thus a regular lognormal distribution appear. Calcium and magnesium oxide show bi- or polymodal distributions.

Although, some trace elements, as the nickel, the zinc, the arsenic and the boron, among the 16 analysed trace elements (table 2), are important indicators of the igneous and, respectively, of the postmagmatic activities, they appear only in few samples above the detection limit. The distribution of manganese, chrome and gallium (fig. 3) approaches to the lognormal distribution. The small inflexion of the distribution line of manganese (around the 200 ppm value) probably covers a bimodale distribution. The bimodale distribution shows in the logarithmic distribution of the

beryllium, vanadium, cobalt and lead contents. The titanium, the tin and the copper have trimodal distribution

The polymodal distribution of some rock-forming and trace elements support the petrographic observations, i. e. two or three different petrographic and genetic type of the leptinites.

Table 2 Trace elements in the Stejera Leptinites

Element	Number of analyses	Min, ppm	Max, ppm	Average, ppm	Dispersion	χ^2	χ^2 admissible
Be	77	10	52	22	11.93		
B	8	36	190	93	56.30		
Ti	139	100	3300	2005	1161.95		
V	33	10	99	38	26.46		
Cr	53	10	70	32	11.44	19.30	13.39
Mn	131	20	3300	312	475.56	17.53	15.03
Co	67	3	130	27	28.29		
Ni	15	5	295	69	98.61		
Cu	130	3	100	18	20.02		
Zn	22	20	3000	265	620.15		
Ga	137	8	230	40	27.42	17.08	16.66
As	11	30	100	43	28.32		
Sn	72	3	130	26	30.44		
Pb	75	4	3300	146	510.87		
Sb	11	100	1000	278	256.74		

Analysed in the laboratory of the Geological Institute of Romania, Bucureşti

6. THE CHEMISTRY OF THE FORMER IGNEOUS ROCKS

For accepting (or rejecting) the igneous origin for leptinite, we effectuated a complete petrochemical calculation, to know the nature of the original volcanic rocks, the differentiation processes, the metamorphic changes and the tectonomagmatic setting of rocks. The nature of the original volcanic rocks could be known, plotting the analytical data in some ternary diagrams (fig. 4).

The QLM-diagram was drawn by the calculation of the normative Q (quartz), L (leucocrates) and M (melanocrates), using Niggli's normative minerals. The samples, due to their high quartz contents, are plotted in a well concentrated field, in the upper corner of the rhyolite-dacite-andesite one.

We calculated the CIPW normative minerals also, and that resulted high feldspar and low ferromagnesian mineral contents. Plotting these results in Streckeisen's QAP-diagram, the samples form an irregular surface, in the quartz-rich granitoid (quartz-keratophyre?), syeno-granite (rhyolite), monzogranite (rhyodacite) and granodiorite (dacite) fields.

The differentiation processes of the original magma are emphasized by the diagrams C and D of fig. 4. The samples plot in the subfemic and salic fields of the *al-*

fm diagram, and in the alkaline and subalkaline fields of the *al-alk* diagram (fig. 4C). In the *k-mg* diagram, the two leptinite types may be separated: the Mg-rich, biotitic leptinite and the Mg-poor, „aplite-gneiss” ones. The well concentrated field in the upper right side of the *k-alk* diagram emphasize the predominance of the potash minerals (orthoclase) despite of the sodic ones (fig. 4D).

Plotting the *al*, *fm*, *c* and *alk* parameters versus *si*, the differentiation diagram shows three, clearly separated sectors, all of them with increasing *si*, slightly increasing *al*, quasi-constant *alk* and slightly decreasing *fm* and *c*. In the first sector with 300–350 *si* plot the intermediary-acide rocks, mainly the biotite-rich leptinites, in the second sector (350–420 *si*) the massive, quartz-rich, biotitic and muscovitic leptinites and in the third sector, above 430 *si*, the „aplite gneisses”. Thus, their protoliths may be dacite, rhyodacite and rhyolite, respectively.

The meso-norme show the same mineralogical associations which appear in thin sections (muscovite-K-feldspar-biotite-almandine-staurolite), i.e., the chemical-mineralogical equilibrium was accomplished, and the further (postmetamorphic, weathering) transformations do not have a remarkable influence on the composition of these rocks (i.e. the <0,063 mm fraction of the samples).

Table 3. Immobile elements of Stejera leptinites

Sample	SiO ₂ , %	TiO ₂ , %	Rb, ppm	Y, ppm	Nb, ppm	Ga, ppm	Zr, ppm
223	62,81	1,02	305	20	2,0	5	199
227	82,41	0,10	193	18	1,3	15	234
229	82,40	0,14	167	27	1,3	30	325
230	70,82	0,16	243	15	1,0	33	90
235	80,21	0,45	146	33	1,0	11	949
236	84,84	0,16	333	14	2,3	19	494
240	77,12	0,10	139	33	2,0	23	152
244	76,80	0,10	100	37	3,0	23	120
246	71,91	0,34	104	39	3,0	33	200
247	79,26	0,18	136	33	2,0	27	362
249	69,73	0,62	205	17	1,0	33	319
251	68,96	0,64	202	18	1,3	15	218
255	81,61	0,01	157	29	1,0	21	230
256	83,83	0,06	269	16	1,0	28	183
260	61,32	0,87	1,59	29	2,0	14	139

Analysed in laboratories of University of Concepción, Chile (1998)

Beside the usual trace element analyses, six immobile elements (Winchester & Floyd, 1977) for 15 natural leptinite samples were analysed (Table 3). These discrimination diagrams (Pearce et al, 1984) permit a classification of these rocks which have suffered deep mineralogical and chemical transformations, as metamorphic processes for example. In the Zr/TiO₂–Ga diagram the samples are plotted in the fields of rhyolite, rhyodacite-dacite and andesite. In the SiO₂–Zr/TiO₂ diagram the samples are arranged in a quasi straight line between the andesitic and rhyolitic composition. The SiO₂–Nb/Y diagram (fig. 5A) shows a similar feature. Thus, using the immobile

elements, we confirmed the results of the classical petrochemical calculation and the field and petrographic observations: the leptinites derived from acid (and subordinately, intermediary) volcanic rocks, including their alteration products and/or pyroclastites (Ewart et al., 1968).

The Cr–Be diagram (fig. 5B) shows that the samples leptinites are products of initial magmatites, in the keratofiric series. In the Rb+Y+Nb diagram (fig. 5C), the range of the points traverses the separation line between the syn-collisional and volcanic arc settings.

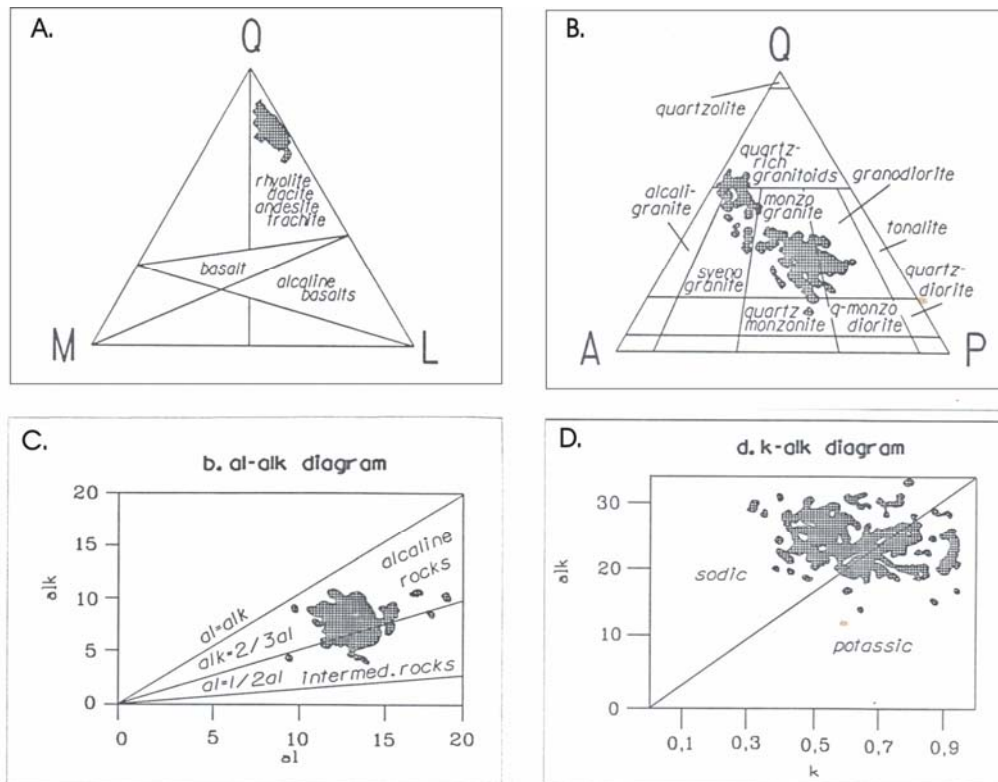


Fig. 4. Petrochemical diagrams of Stejera leptinites

7. THE PROTOLITH

The mineralogical composition, the structural and the textural peculiarities of the leptinites, and — admitting the above emphasised $S_{str} \approx S_1$ hypothesis — also the mode of the imbedding, infer their igneous origin. The geochemical study of these rocks, as well as, the use of the discrimination diagrams, makes possible to get information about the nature of the protolith.

In the discrimination diagram of Debon and LeFort (1983), the values are plotted in a elongated, hemi-elliptical field which cuts cross the separation line between the igneous and sedimentary field.

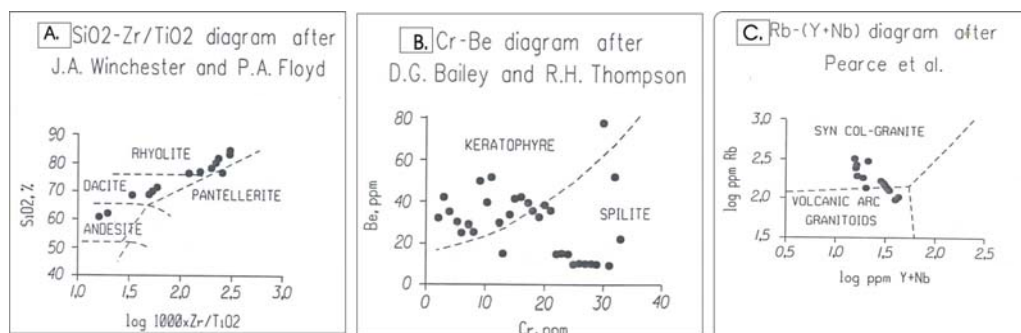


Fig. 5. Some discrimination diagrams of Stejera leptinites

In both discrimination diagrams, the samples which are plotted out of the igneous field differ from the rest by increased Al₂O₃ contents and, in each case, by a weak deficit of alkaline oxides.

Thus, the discrimination diagrams indicate that (i) part of the analysed rocks are igneous origin and the others have a sedimentary (or mixed, sedimentary+pyroclastic) origin, forming a strong imbedded succession, or (ii) the whole succession of the protolith was igneous, but before the metamorphic recrystallisation, a secondary process had altered a part of these rocks, subtracting alkalis and consequently, enriching it in alumina (i.e. hydrothermal or halmirolitic argilisation). The presence of the mica-rich imbeddings into the leptinite succession support the first hypothesis; however, the petrochemical calculations (see below) support the second one.

8. DISCUSSION

The magmatic origin of the leptinites, inferred by the field and petrographic observations, was confirmed (partially) by our petrochemical and geochemical studies. Thus, the protolith of the leptinites was an acid, alkali-rich volcanic rock, with calcalkaline, subordinately alkaline composition, corresponding to rhyolite, rhyodacite, dacite and in each case, andesite. Some of them have suffered certain alteration processes *before* the metamorphism or were mixed with some pyroclastic and/or terrigenous sediments.

Both the statistical treatise of analytical data and the petrochemical diagrams indicate the presence of two or three different petrographic types, as a result of the differentiation of the same magma. In the field, they correspond to biotitic leptinites and aplitic gneisses respectively.

It is very likely that the meta-igneous rocks from the basement of the Great Hungarian Plain (Tóth, 1995) and the Stejera leptinites may be linked to an old volcano-tectonic line, which starts in the Inău and Preluca Mts., crosses the Țicău and Codru (Bacă) Mts., the basement of the Sălaj basin (Kalmár, 1994b), the Great Hungarian Plain and reaches the river Dráva (Szederkényi et al., 1995). In this order, the leptinites may be used as a good marker horizon in the basement of the Tisia Realm.

The metamorphic processes, which have induced strong changes in the composition of the surrounding meta-pelites, in the case of these acid volcanic rocks, have not affected their main petrographic and chemical features. Despite of the advanced metamorphic recrystallisation, in condition of the chemical–mineralogical equilibrium, the feature and the extension of the several rock bodies are still recognisable: the characteristic „rhyolite-banding” in some aplitic gneisses, as well the phenocrystals of dacites transformed in the so called „checkboxboard-albite”. The chemical composition, as well the correlation between the rock-forming and trace elements respectively, remains, also, near to the initial one.

9. ACKNOWLEDGEMENTS

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