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CONTRIBUTIONS TO THE CLAY MINERALIZATION AND ZEOLITIZATION OF THE MIOCENE TUFFS IN THE MECSEK MTS., HUNGARY

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Abstract: Rhyolite, dacite and rhyodacite tuff and tuffite layers of Miocene age in the Mecsek Mountains, Hungary, partly or completely altered to containing clay (mostly smectite) and zeolite (clinoptilolite) minerals as dominant alteration product. The (authors') model for this alteration process is:
Hydrolysis → increase in pH → precipitation of smectite with d value of >14 Å → increase in pH → precipitation of clinoptilolite → decrease in pH → ion exchange between pore fluid and smectite (Ca is replaced by Na, which causes decrease in smectite d value to <14 Å) → precipitation of K–feldspar. The excess silica precipitated as Opal–C at different stages.

Key words: Mecsek Mts., Miocene, alteration model, smectite, clinoptilolite, opal–C.

Introduction

Clay has been used for a long time for industrial purposes, waste disposal site preparation, in the environmental protection, etc. In the recent decades zeolite has become also widely used in similar areas. So far their joint application has not happened, because their interaction in different conditions is not well studied. In the nature they can be found as coexistent minerals, so studying these occurrences we gain our understanding on their joint behavior.

Partly or completely altered tuff and tuffite layers of Miocene age can be found in the Mecsek Mountains, Hungary. Smectite (clay mineral) and clinoptilolite (zeolite mineral) occurs together in many samples. The authors give a model for this particular alteration, in which they describe the interrelationship between the formation of smectite and clinoptilolite.

Geological setting

In the Mecsek Mountains (southwest Hungary) the Paleo- and Mesozoic suits are partly covered and surrounded by Neogene (Miocene, Pannonian) and Quarternary sedimentary rocks (Fig. 1). These Miocene sedimentary rocks contain two tuff horizons (Fig. 2, formations no. 5 and 7). The K-Ar age of the lower horizon ranges from 19.1±1.8 to 22.0±2.4 Ma (Árva-Sós & Máthé 1992). These data confirm well with that of the so called "lower rhyolite tuff" in Hungary (Hámor et al. 1978), in average 19.6±1.4 Ma. The K-Ar age of the upper horizon varies between 15.3±0.6 and 18.1±1.0 Ma (Árva-Sós & Máthé 1992), which matches well with the average age of the so called "middle rhyolite tuff" in Hungary, i. e. 16.4±0.8 Ma (Hámor et al. 1978).

The tuff and tuffite layers deposited in various environments. The lower tuff horizon deposited between terrestrial–fluviatile sediments, or immediately on the top of the Mesozoic crystalline basement (Hámor 1970). This is a well distinguishable horizon (Fig. 2).

The upper horizon deposited into terrestrial, fluviatile, flood-plain, paludal, brackish, lagoonal, shallow marine carbonatic, littoral detrital, saline environment (Hámor 1970, Fig. 2). This horizon consists of two layers (mostly in the marine environment).

Based on the textural and structural characteristics the genetic types of the tuffs are:

Lower tuff horizon:

— ash-flow tuff. In the northern part of the Eastern Mecsek Mts. it outcrops on greater areas, while west of this area

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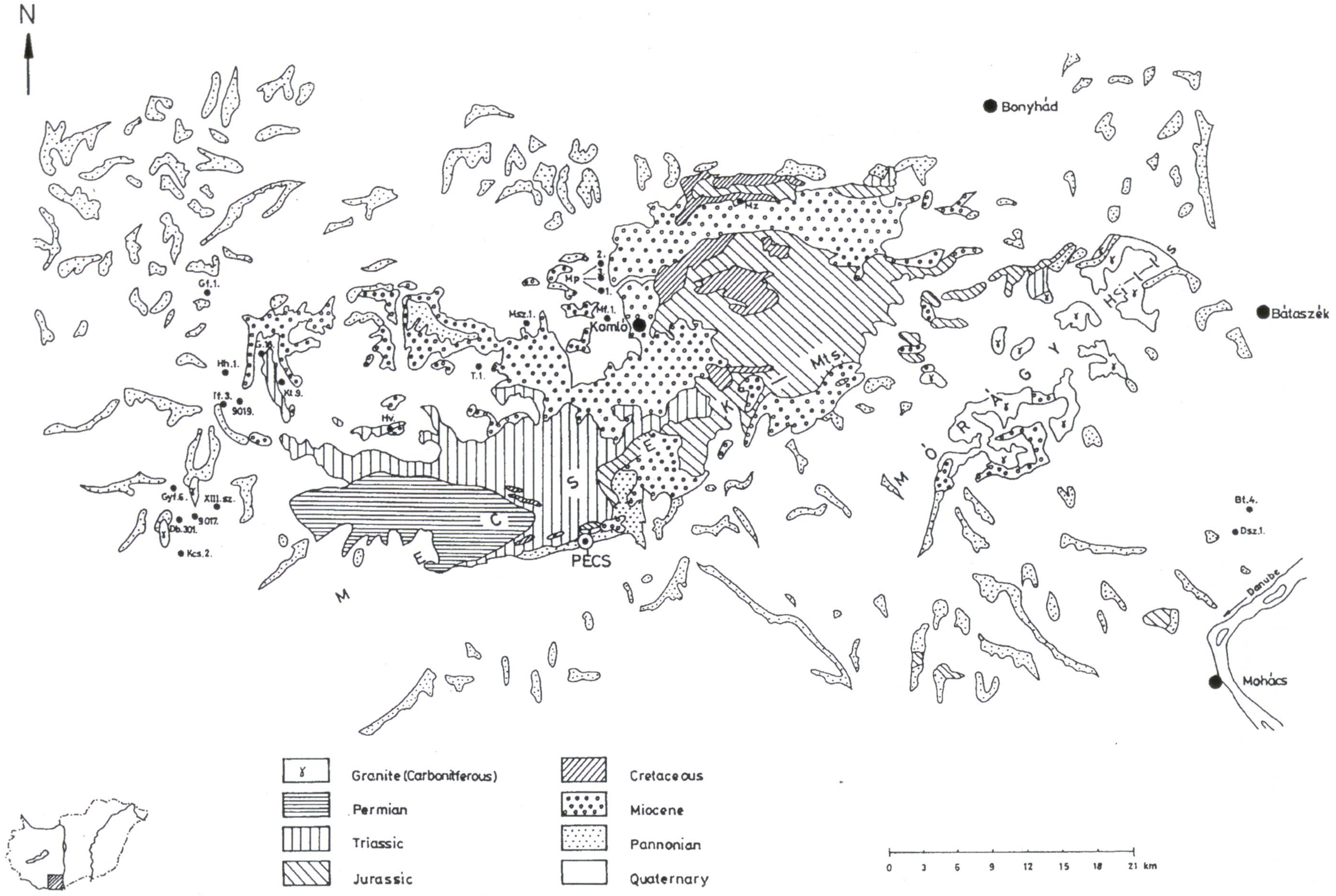

Fig. 1. Sketch map of the area of the Mecsek Mountains with the boreholes. Abbreviations: **Bt.4.** — Báta-4; **Db.301.** — Dinnyeberki-301; **Dsz.1.** — Dunaszekcsô-1; **Gf.1.** — Gálosfa-1; **Gyf.6.** — Gyurufu-6; **Hh.1.** — Horváthertelend-1; **If.3.** — Ibafa-3; **Kt.9.** — Kán mapping; **Mf.1.** — Mecsekfalu-1; **Mp** — Mecsekpölöske; **Msz.1.** — Magyarszék-1

it can be found in the depth of around 1000 m (e.g. Mecsekpölöske–1 borehole, Fig. 1).

— ash-fall tuff: can be found in depth in the Western Mecsek Mts.

The *upper tuff horizon* consists of ash-fall tuff only.

The average thickness of the ash-flow tuff of the lower horizon occurring only in the northern area of the mountains varies between 5 and 10 m. The extension of ash-fall tuff of the lower horizon is much greater, its thickness is similar to that of the ash-flow tuff (1 to 10 m). The ash-flow tuff is considerably zeolitized while the ash-fall variety of the lower horizon as well as that of the upper horizon are less altered, and are less zeolitized or without zeolite. The quantity of zeolite-bearing tuff can be estimated to be about 3.5 million tons in the near-surface extension of the Eastern Mecsek Mts. (Puzder & Badinszky 1982, 1983).

Mineralogy and petrology

Minerals formed prior to the explosion in both types of tuffs are biotite, plagioclase, porphyric quartz, green amphibole, sanidine (detailed description see in Ravasz-Baranyai 1973, and Árva-Sós & Máthé 1992). The main difference between the lower and upper tuffs regarding their phenocryst contents, the latter contains lesser amount of quartz and more green amphibole, and rarely pyroxene, and their plagioclase are more basic. Based on the phenocryst assemblage and refraction index of glass of the upper horizon Ravasz-Baranyai (1973) determined the rock type of the upper horizon as rhyoda-cite, and the type of the lower horizon as plagioclase rhyolite. This classification is supported by the chemical composition of plagioclase phenocrysts determined by electron microprobe (Árva-Sós & Máthé 1992). Projection of the whole rock compositions on the SiO_2–Zr/TiO_2 and SiO_2–Nb/Y diagrams proposed by Winchester & Floyd (1977) results in rhyolite–dacite series for the lower horizon (also called Gyulakeszi Rhyolite Tuff Formation), and rhyodacite–dacite series for the upper horizon (also called Tar Dacite Tuff Formation) (Fig. 3a, b, for detailed discussion see Máthé et al. 1997).

The alteration of ash-flow rhyolite tuffs accomplished by geoautoclave mode (on higher than 200°C), while the alteration of the other tuffs happened at low temperature and in open hydrologic system (Polgári et al. 1995, 1996).

Samples

For the present studies core samples were taken from the boreholes in the Western Mecsek Mountains and in the southeastern foreland of the Mórágy Hills.

In two boreholes both the rhyolite (lower) tuff and the dacite (middle) tuff horizons occurred, thus enabling detailed sampling from these horizons. In other boreholes either the lower or the middle tuff horizons occurred. Sampling localities are shown in Fig. 1. One of the main aspects of sampling was to sample both genetic types; the rhyolite tuff (ash-flow tuff and ash-fall tuff) and the characteristic types of dacite tuff (ash-fall type) occurring in several horizons. 59 samples were studied.

Methods

Petrological determination of the samples was carried out with the aid of polarization microscopy and scanning electron microscopy (SEM).

X-ray diffraction (XRD) analyses were made on whole rock samples, on the less than 2 μm fractions, and the measurements were repeated on glycolated samples. Methods used for identification of clay minerals are described in Thorez (1975). The samples were kept in desiccator for a week preceding the measurement. XRD studies were made with a Philips PW 1710 instrument (operated by APD software), recording conditions: CuKα, 45 kV, 35 mA, graphite monochromator.

The chemical composition of tuffs was determined by wet chemical analyses, that of trace elements by X-ray fluorescence (XRF) method. The chemical composition of zeolite and other authigenic minerals was studied by means of JEOL Superprobe 733 electron microprobe recording the energy dispersive spectra of the elements (focussed beam, 3 nA, samples were carbon-coated fragments). This instrument was used for qualitative mineral analysis of some polished samples.

Results

The mineral assemblage and the smectite d(001) values determined by X-ray diffractometry are shown in Table 1.

Minerals formed by alteration of the tuffs: smectite, clinoptilolite, mordenite, opal cristobalite, chlorite, illite, illite/smectite and kaolinite/smectite mixed layer structures, kaolinite.

Hence the type of cristobalite cannot be determined from the X-ray diffractogram of the whole rock (overlapping), cristobalite has been separated from sample Gálosfa–1 889.0 m (see Table 1) and its type has been determined by X-ray diffractometry as opal-C according to the nomenclature used by Graetch (1994). The main peak of cristobalite on the X-ray diffractograms of the whole rock samples show similar shape with that of the separated one, it is reasonable to suppose that cristobalite in the other samples are also opal-C.

Smectite is ubiquitous, it can be found in the highly zeolitized rhyolite ash-flow tuff as well as in the non zeolitized, but altered dacite tuff. The d value of the basal

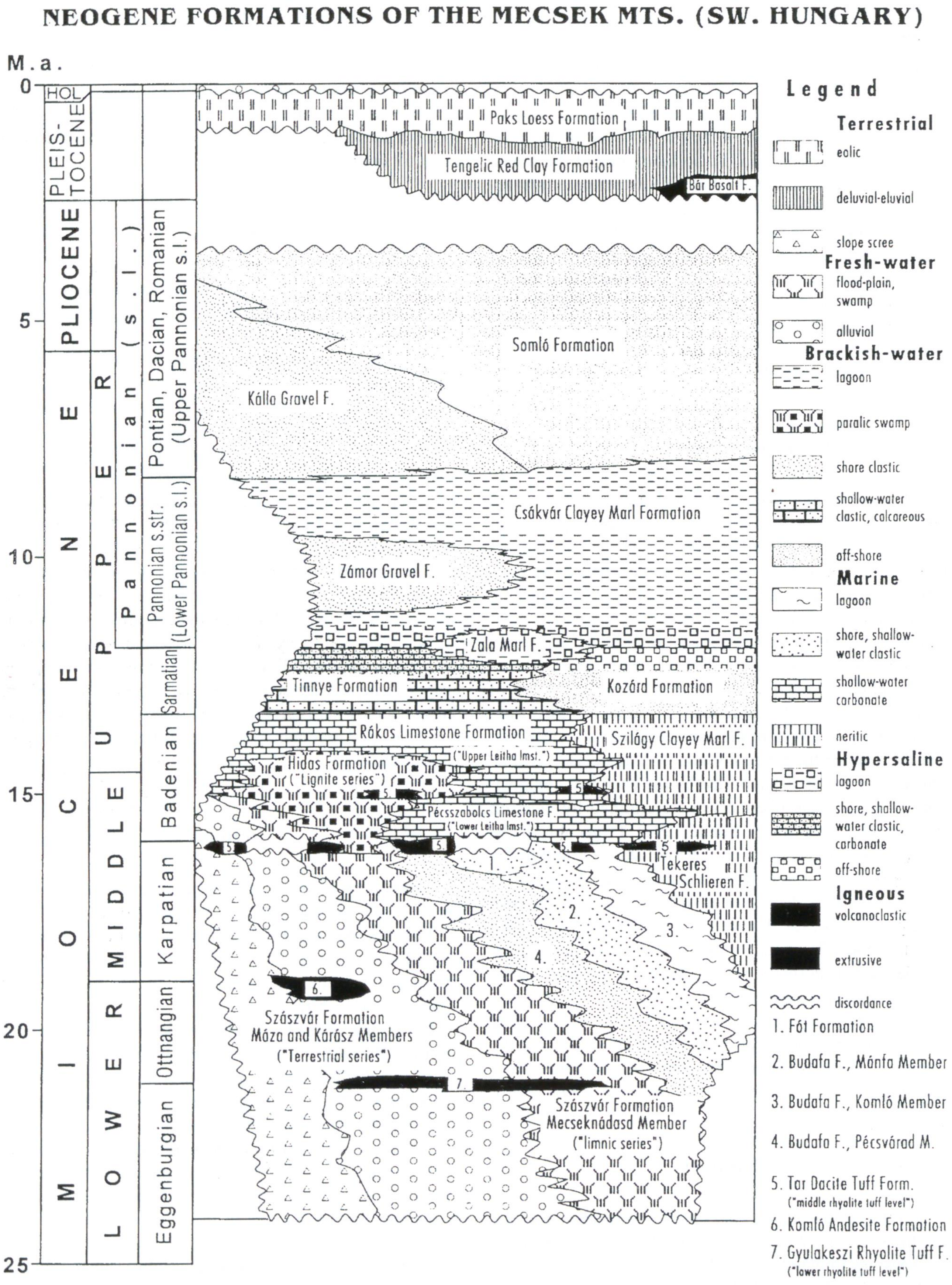

Fig. 2. Generalized (theoretical) stratigraphic column of the Mecsek Area (compiled by Barabás 1994).

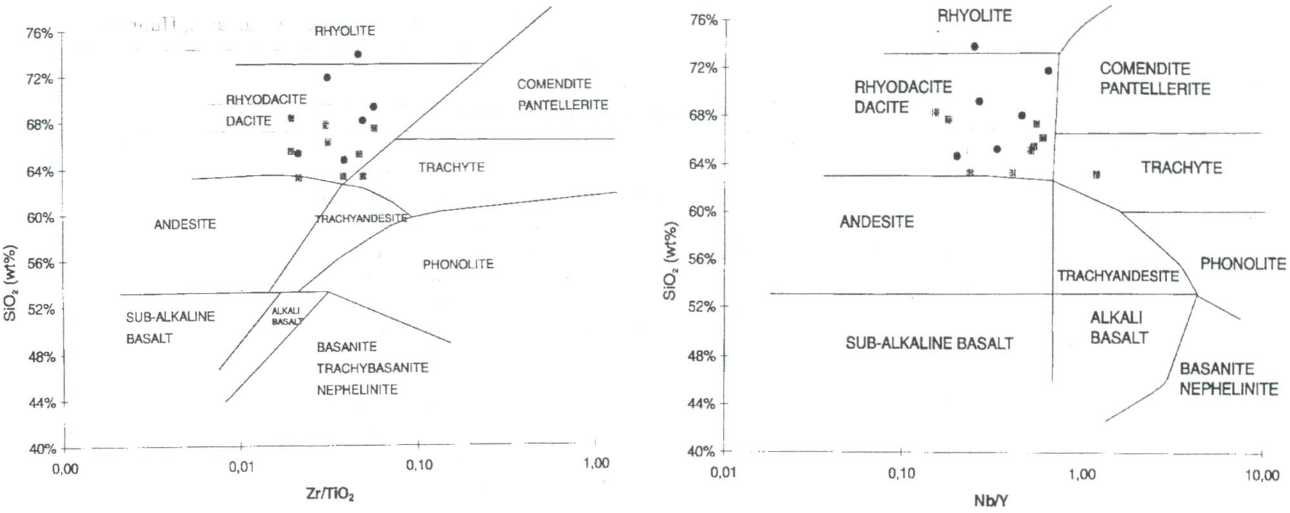

Fig. 3. Projection of the Mecsek Miocene tuffs on the a) SiO_2-Zr/TiO_2 and b) SiO_2-Nb/Y diagrams proposed by Winchester & Floyd (1977). Legend: **circle** — Gyulakeszi Rhyolite Tuff Formation, **square** — Tar Dacite Tuff Formation.

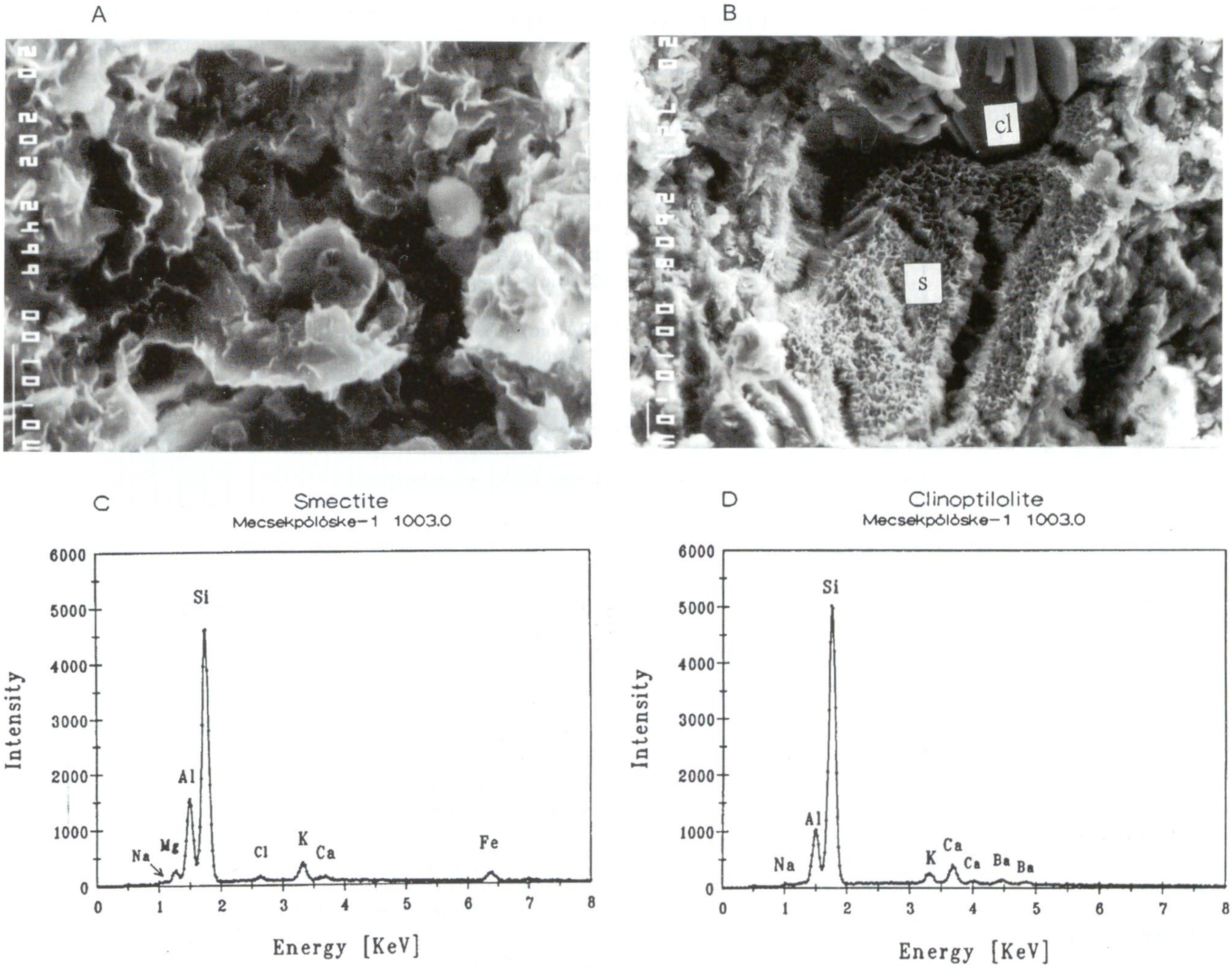

Fig. 4. Scanning electron micrographs of typical smectite and clinoptilolite occurrences and their EDS spectra. **A.** Typical smectite flakes in the sample Mecsekfalu-1 252.5 m. **B.** Tabular clinoptilolite (cl) crystals and smectite (s) flakes in the sample Mecsekpölöske-1 1003.0 m. **C.** EDS spectrum taken from about 10 mm^2 area of smectite flakes, position indicated on Fig. 4b by letter "s". **D.** EDS spectrum taken from a tabular clinoptilolite crystal, position indicated on Fig. 4b by letters "cl".

Table 1: Smectite d(001) values and XRD mineralogy of the Miocene tuff and tuffite layers in the Mecsek Mountains, Hungary.

Borehole samples	smectite d(001) Å	mineral assemblage
LOWER GYULAKESZI RHYOLITE TUFF		
Mecsekpölöske-1	tuff (ignimbrite)	
1002.1 m	12.19	Q>>clp>plg>mor(?)>10Å(bio)>smectite
1003.0 m	11.63	Q>>clp>10Å(bio)>smectite>>hematite>mor(?)
1017.4 m	12.45	Q>>clp>10Å(bio)>smectite>chl>mor(?)
1018.1 m	12.27	Q>>clp>10Å(bio)>smectite>plg>mor(?)
1018.5 m	12.42	Q>>clp>>10Å(bio)>smectite>plg(tr),glass(tr)
1019.2 m	12.27	Q>>smectite>10Å(bio)>plg>clp
1019.4 m	14.48	smectite>Q>plg>cr>glass(tr),10Å(tr),chl(tr)
(1021.3 m	*12.45*	*Q>smectite>10Å>plg>>calcite)*
Gálosfa-1	ash-fall tuff	
889.0 m	12.45	cr≥smectite>plg>Q>10Å(bio)>clp>chl>Kfp
896.0 m	13.39	cr>smectite>plg>10Å(illite)>clp>plg>Kfp>calcite
898.0 m	12.45	cr≥Q>smectite>plg>Kfp>10Å(illite)>clp>chl
904.2 m	12.45	cr>smectite≥plg>Q>clp>10Å(>calcite)
Horváthertelend-1	ash-fall tuff	
583.6 m	15.24	glass>>smectite>plg>Q≥10Å(illite)
584.0 m	13.59	glass>>smectite>plg>Q>dol>10Å(illite)>kaol
Kán-9	ash-fall tuff	
308.6 m	13.09	smectite>>plg≥10Å(bio)>Q≥chl
MIDDLE TAR DACITE TUFF		
Mecsekfalu-1	tuffite	
249.5 m	12.19	clp>smectite>cr>Q>10Å(bio)>calcite>plg(tr)
250.3 m	12.28	smectite>cr>clp>Q≥10Å(bio)>plg(tr)
251.4 m	12.27	smectite>cr>clp>10Å(tr),plg(tr),Q(tr)
252.5 m	12.36	smectite>cr≥clp>plg(tr),Q(tr)
256.0 m	–	10Å(bio)>plg>Q>clp>cr
354.1 m	12.62	smectite>cr≥Q>>10Å(bio)>calcite>clp
487.3 m	14.72	smectite>Q>10Å(bio)>plg>calcite>chl
488.7 m	14.85	smectite≥plg>Q≥10Å(bio)>clp>glass
489.5 m	14.85	smectite>10Å(bio)≥Q>plg≥Kfp>calcite>chl>clp(tr)
Mecsekpölöske-1	tuffite	
366.6 m	–	glass>>Q>10Å(bio)>plg>chl(tr),calcite(tr),siderite(tr)
367.0 m	14.73	glass>>10Å>Q>plg>siderite>calcite>dol>smectite>kaol
Mecsekpölöske-2	tuffite	
346.0 m	13.59	glass>>plg>Q>calcite>smectite>10Å(tr)
346.5 m	15.36	glass>>Q,plg,smectite
Mecsekpölöske-3	tuffite	
353.7 m	14.48	glass>>plg>smectite>Q>pyrite>gypsum
Ibafa-3	ash-fall tuff	
27.4 m	14.98	glass>plg≥smectite>10Å(I/S)>Q
9017	ash-fall tuff	
32.7 m	15.24	glass>plg>10Å(bio)>smectite>Q>kaol(tr)
34.6 m	16.21	glass>plg>10Å(bio)>smectite>Kfp>Q>kaol(tr), gypsum(tr)
35.2 m	15.24	glass>10Å(bio)>plg>Q≥smectite>chl(tr),gypsum(?,tr)
35.4 m	15.50	glass>10Å(bio)>smectite>plg>Q>gypsum(?,tr)
Kán-9	ash-fall tuff	
55.3 m	15.36	smectite>10Å(bio)>Q≥plg
55.8 m	15.23	smectite>10Å(bio)>Q≥cr>plg>glass
56.0 m	15.50	glass>plg>smectite>Q
57.0 m	15.23	smectite>10Å(bio)≥plg>Q>glass
57.3 m	15.24	smectite>10Å(bio)>plg>Q>kaol/smectite
Dinnyeberki-301	ash-fall tuff	
62.6 m	16.36	smectite>10Å(I/S)>kaol≥plg≥Q>calcite
62.7 m	15.64	smectite>Q>10Å(bio)>plg>kaol/smectite
64.6 m	16.07	smectite>plg>10Å(I/S)>Q>kaol/smectite>clp(tr)
Dunaszekcső-1	ash-fall tuff	
552.1 m	15.50	glass>Q≥smectite>plg>chl
552.3 m	15.70	glass>smectite>10Å(bio)>Q≥plg>amphibole
553.0 m	15.70	glass>smectite>10Å(bio)>plg>Q
554.6 m	14.72	glass>smectite>10Å(bio)>plg>Q>chl≥amphibole
556.0 m	16.99	glass>smectite>10Å(bio)>plg>Q>chl
556.2 m	16.06	glass>smectite>plg>10Å>Q>chl,kaol>amphibole

Continuation of Table 1

556.2 m	16.06	glass>smectite>plg>10Å>Q>chl,kaol>amphibole
559.9 m	14.90	glass>smectite>10Å(bio)>plg>kaol>Q>gypsum,pyrite
Magyarszék-1	tuffite	
264.8 m	11.60	glass>magnetite>clp>plg>Q>cr>10Å,smectite
267.4 m	13.49	glass>plg>>10Å(bio)>smectite>clp>kaol
268.8 m	13.01	glass>>>10Å(bio)≥smectite>Q>plg>Kfp(tr),kaol(tr)
269.3 m	–	calcite>Q≥plg,clp≥10Å(bio)>10Å/S>chl,cr,glass
485.6 m	12.27	10Å(bio)>plg>smectite>Q
Báta-4	ash-fall tuff	
415.1 m	14.72	smectite>kaol-d≥10Å(I/S)>Q
416.0 m	14.60	smectite>10Å(illite)>kaol-d>Q>glass
417.0 m	14.48	smectite>10Å(illite)>kaol-d>Q>glass
417.3 m	14.48	smectite>10Å(illite)>kaol-d>Q>glass
419.0 m	14.25	kaol-d≥smectite>10Å(illite)>Q(tr)>glass
419.2 m	14.72	kaol-d>smectite>10Å(illite)>Q(tr)>glass
Gyürüfü	ash-fall tuff	
91.7	15.24	glass>10Å(S/bio)>plg>Q>smectite>kaol/smectite(?)
92.8	15.24	glass>smectite>10Å(bio)≥plg>Q>kaol/smectite(?)

Abbreviations: bio=biotite, chl=chlorite, clp=clinoptilolite, cr=opal-C (cristobalite), dol=dolomite, I/S=illite/smectite mixed layer mineral, kaol=kaolinite, kaol/smectite=kaolinite/smectite mixed layer mineral, kaol-d=unordered (diffuse) kaolinite, Kfp=K-feldspar, mor=mordenite, plg=plagioclase, Q=quartz, tr=trace amount, 10Å=10Å phyllosilicate, ?=identification is uncertain

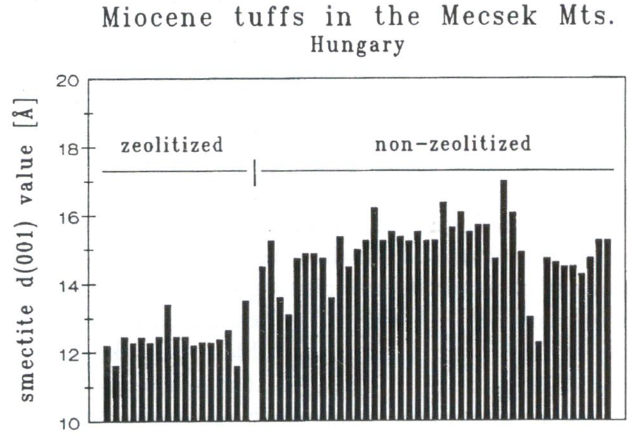

Fig. 5. Distribution diagram of d(001) values of smectite in the zeolitized and non-zeolitized rock samples of the Miocene tuffs in the Mecsek Mountains, Hungary.

reflection of smectite varies from 11.6 Å up to 17.0 Å. It is well known that this d value determined by the type of interlayer cations and the amount of water molecules between the TOT sheets. The d value is higher (14–17 Å) when the interlayer dominant cation is Ca, and it is lower (11–14 Å) when the interlayer dominant cation is Na.

A relation can be observed between the d value of the smectite basal reflection and the zeolitization. When the rock is zeolitized, the smectite d value ranges between 11.6 and 14 Å. When there is no zeolite mineral together with smectite, the latter's d value varies between 14 and 17 Å (Table 1, Fig. 5). There are only 4 exceptions when the d value is smaller than 14 Å and there are no zeolite minerals.

The electron microprobe study of the polished sections of samples Mecsekpölöske–1 1002.1 m and Mecsekfalu–1 249.5 m revealed that the cavity filling clinoptilolite shows compositional zonation. The Ca/Na ratio increases from the wall of the cavity to its center. The amount of K shows slight variation.

Discussion

The alteration of glassy material starts with hydrolysis. Ca^{++}, Na^{+} and H_4SiO_4 and other components go into solution, to the pore fluid. The concentration of the cations increases resulting in increasing pH. Then precipitation starts, usually the first crystallization product is smectite (cf. Dibble & Tiller 1981; Noh & Boles 1989; Tribble & Wilkens 1994; Christidis et al. 1995). The interlayer cations of this smectite are Ca^{++}, Na^{+} and K^{+}. The ratio of these ions depends on the chemistry of the glass precursor and the fluid/rock ratio. Precipitation of smectite yields increase in alkalinity and silica activity of the pore water, so pH further increases favoring the precipitation of zeolite minerals (cf. Christidis et al. 1995). In both horizons the zeolite mineral is clinoptilolite. The ratio of (Ca+Ba)/(Na+K) as a rough estimation from the EDS spectra is around 1:1 (no quantitative measurements have been made). The first clinoptilolite precipitation is rather Na-rich and Ca-depleted, the later crystallized ones are rather Ca-rich and Na-depleted. Electron microprobe investigations revealed that the Ca and Ba contents varies sympathetically (Polgári et al. 1996).

In the last stage the K content of the fluid increased, which resulted in illitization or in some cases precipitation of K-feldspar. During this period of alteration ion ex-

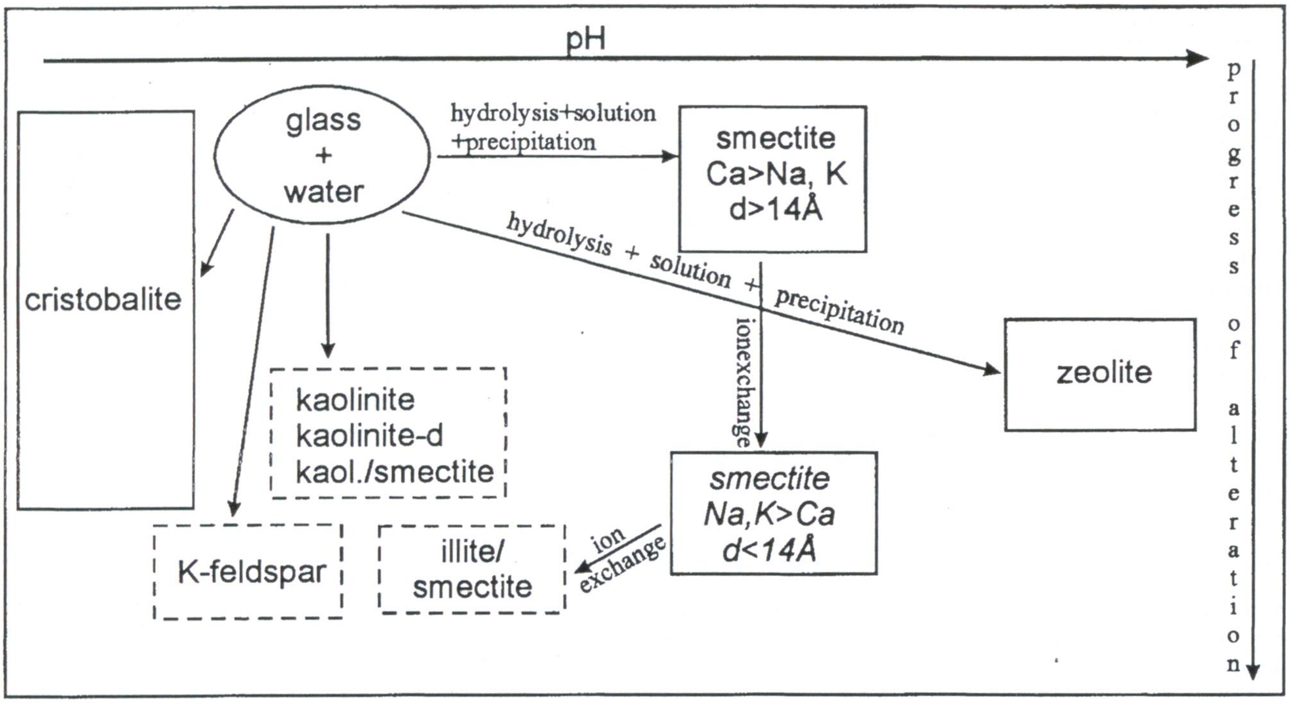

Fig. 6. Schematic model for the alteration process of the tuff and tuffite layers of Miocene age in the Mecsek Mountains, Hungary. Minerals framed by dashed lines occur sporadically in subordinate amount.

change occurs between the ions in solution and the interlayer cations of smectite, because the chemistry of the fluid was changed a lot from that one when smectite precipitated.

The excess silica precipitated as cristobalite during the different stages of the alteration.

The Mecsekpölöske–1 rhyolite tuff (Table 1) is well zeolitized except the very bottom of this layer (sample 1019.4 m). Smectite can be found in all the samples of this corehole and its d value is <14 Å when the tuff is zeolitized and >14 Å when the tuff is not zeolitized. The bottom of the tuff is at the depth of 1020.0 m. The sample 1021.3 m is sedimentary rock (aleuritic clay) containing a lot of smectite with a d value of 12.45. One can think that the most bottom part of the tuff layer is affect ed by the underlying layer, but this can be excluded because of the great difference between the d values of smectites in the two connecting layers. So as an initial d value of >14 Å can be inferred for smectite during its precipitation. This hypothesis is supported by the other non-zeolitized tuff samples (exceptions will be discussed later).

After the precipitation of clinoptilolite, ion exchange took place between smectite and pore fluid. Since the initial smectite d value was >14 Å, the dominant interlayer cation must have been Ca^{++}. During the ion exchange Ca^{++} might have replaced by

1) K^+: illitization starts (e.g. Dinnyeberki–301)
2) Na^+: smectite d value decreases to less than 14 Å.

Electron microprobe study of the samples Mecsekpölöske–1 1003.0 and Mecsekfalu–1 252.5 m revealed that the smectite mineral in these samples are Fe- and K-rich. At the same time no Fe peak can be observed on the EDS spectrum of the clinoptilolite crystal (Fig. 4).

The above described ion exchange process resulted in smectite d value <14 Å when the alteration reached the stage of clinoptilolite precipitation. Otherwise smectite d value remained >14 Å (Fig. 5). There are only few exceptions from this rule (Table 1, Fig. 5). Mecsekpölöske–2 346.0 m and Magyarszék–1 268.8 and 485.6 m samples are tuffites, so the initial chemical composition of these samples differed from that of tuff layers, and these samples can also contain inherited smectite.

Samples from the Mecsekfalu–1 borehole are tuffites and more or less zeolitized, and does not keep the above mentioned rule. Based on chemical as well as trace element compositions these samples fall into two groups (Árva-Sós & Máthé 1992). One group consists of sample numbers from 249.5 to 354.1, the other group consists of sample numbers from 487.3 to 489.5. Although the later group is partly zeolitized, its smectite d values are >14 Å. The reason why these d values remained over 14 Å can be a) clinoptilolite is in minor amount, so its precipitation had insignificant effect on smectite layer spacing, or b) the original chemical composition of this rock. The d value of smectite in the Horváthertelend–1 584.0 sample is also <14 Å, and there is no zeolite phase in the mineral assemblage, but this tuff layer is very thin, so the underlying and overlying layers could have affected significantly this tuff layer.

Conclusions

Miocene tuffs in the Mecsek Mountains, Hungary, partly or completely altered. The model, proposed by the authors, for this alteration process is summarized on Fig. 6.

Following the hydrolysis of the glassy material the first crystallization product is smectite with d value of greater than 14 Å and Ca as dominant interlayer cation. Precipitation of smectite resulted in higher pH favoring the precipitation of clinoptilolite. Following the formation of clinoptilolite ion exchange took place between the pore fluid and smectite. Ca was replaced by Na, so smectite d value decreased under 14 Å. If there was enough K in the solution, K-feldspar formed. The excess silicon precipitated as opal-C in the different stages of alteration.

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