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# ZEOLITIC TUFFS FROM COSTIUI ZONE - MARAMURES BASIN

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**Abstract**: The Badenian deposits from the Coştiui zone form a small sedimentary basin in the north of Maramureş depression. The Badenian is disposed discordantly over the Eocene deposits and includes dacitic tuffs having a thickness of about 250m with globigerines. Sarmatian formations represented by marls, marls-sandstone, friable sandstone and sands. In the Coştiui zone the Badenian tuffs consist of the basis of lapilic tuffs over which crystaloclastic and vitroclastic tuffs are disposed. The primary magmatic minerals are represented by plagioclase feldspars, quartz, alkaline feldspars, micas (biotite and muscovite), pyroxene, amphiboles. The volcanic glass of the Costiui tuffs shows a high degree of devritrification in glassy groundmass with formation of diagenetic minerals represented by zeolites. By using the X-ray diffraction analyses the presence of the following members from the zeolites groups has been pointed out: clinoptilolite, mordenite and heulandite. The clinoptilolite is the predominant zeolite and is characterised by an increased crystallinity degree. The celadonite and the cryptocrystalline silica appear besides the zeolites. The chemical composition of the zeolitic tuffs from the Coştiui zone emphasised a dacitic character. The average cationic exchange capacity is: $Ca^{2+}$=62 mvali/g, $Mg^{2+}$=6 mvali/g, $Na^{+}$=18 mvali/g, $K^{+}$ =24 mvali/g. The surface area varies between 12.4 – 21.9 $m^2$/g. The density is of 1.99% g/$cm^3$. The zeolitization of the tuffs has a pervasive character and has affected especially the volcanic glass. From the point of view of the mineralogical composition and of the physical and chemical properties the tuffs from the Coştiui zone are similar with those from the Iza zone. The deposits of zeolitic tuffs from the Coştiui zone are included in the “open system deposits” type.

**Key Words**: Badenian, zeolitic tuffs, clinoptilolite, cationic exchange capacity, density, zeolitization.

In Romania there are many occurrences of Badenian zeolitic tuffs, (Bedelean & Stoici 1998). A lot of occurrences of zeolites appear in the Badenian zones from the Transylvania depression known under the name of Dej tuff or Perşani tuff (Mârza, et al. 1991). Badenian zeolitic tuffs are also known in the Simleu depression, (Pop et al. 1982) and in the Maramureş depression (Damian et al. 1991, 2002).

In the north part of Maramureş depression is placed a small Badenian sedimentary basin known under the name Coştiui. Here are widely developed the deposits of zeolitic tuffs that are less known than the rest of the occurrences along the Iza Valley, (Cochemé et al. 2003).

## 1. GEOLOGY OF THE MARAMUREŞ DEPRESSION

The Maramureş depression, situated in the northern part of the Eastern Carpathians is the first from the series of depressions that make up the big groove (Sârcu, 1971). Most of the authors admit the origin of volcanic barrier of the Maramureş depression because, without the westwards volcanic chain, its depression character would not have manifested itself. Geologically, the Maramureş depression includes the Transcarpathian zone of the flysch, (Mutihac, 2004) over which the Miocene and Quaternary formations are disposed.

The oldest sedimentary formations belong to the Eocene, the Strâmtura and Voroniciu sandstone which crops out in the average course of Iza and in its affluents from this zone. The oligocene deposits represents the biggest part from the Transcarpathian zone from Maramureş area. In the facies of the Borşa sandstone, (Dicea 1980), the Oligocene deposits include: the Caselor Valley formation; the marl-sandstone formation; the Borşa sandstone formation and the superior marl-sandstone formation.

The Badenian is disposed discordantly over the Paleogene deposits. The deposits are tightly folded and have in their structure an alternation of marls and sandstone and intercalations of dacitic tuffs finer in the upper part and coarser at the lower part.

The deposits attributed to the Sarmatian are disposed in continuity of sedimentation over the Badenian ones, forming, usually, the filling of some synclinals. The Pannonian deposits, which finish the Miocene sedimentary cycle, are disposed discordantly over the Sarmatian formations. These deposits of variable thickness include mainly grey marls, and subordinately thin intercalations of sandstone with bivalves remains and intercalations of sands with cross stratification.

## 2. GEOLOGICAL DATA OF THE COŞTIUI ZONE

In the Coştiui zone, over the paleogene deposits, are present the Miocene formations, which develop in a synclinal (figure 1) with a length of about 2.5 km and a width of about 0.3-0.6 km which extends in south-eastern part of the salt massive from Coştiui and include the Badenian and Sarmatian deposits.

According to Atanasiu (1956) the Badenian is disposed discordantly over the Eocene deposits and includes dacitic tuffs of about 250m thickness with globigerines developped south of Coştiui. At north-eastern part of Coştiui, respectively east of Rona, crop out coraligene limestones of Leitha type, which have at bottom remaniated paleogene sandstone, with remains of Pectinide şi Lithothamnium. On Dumbrava Valley tuffs having a thickness of 0.30m are disposed, therefore we can consider that here the limestones are an equivalent of the tuffs.

Over the tuff and limestone deposits are disposed marls with sandstone intercalations with development west of Coştiui. At Coştiui, the inferior Badenian is bent 45º north-east and supports the salt body over which are deposited the marl-sandstone deposits of the superior Badenian.

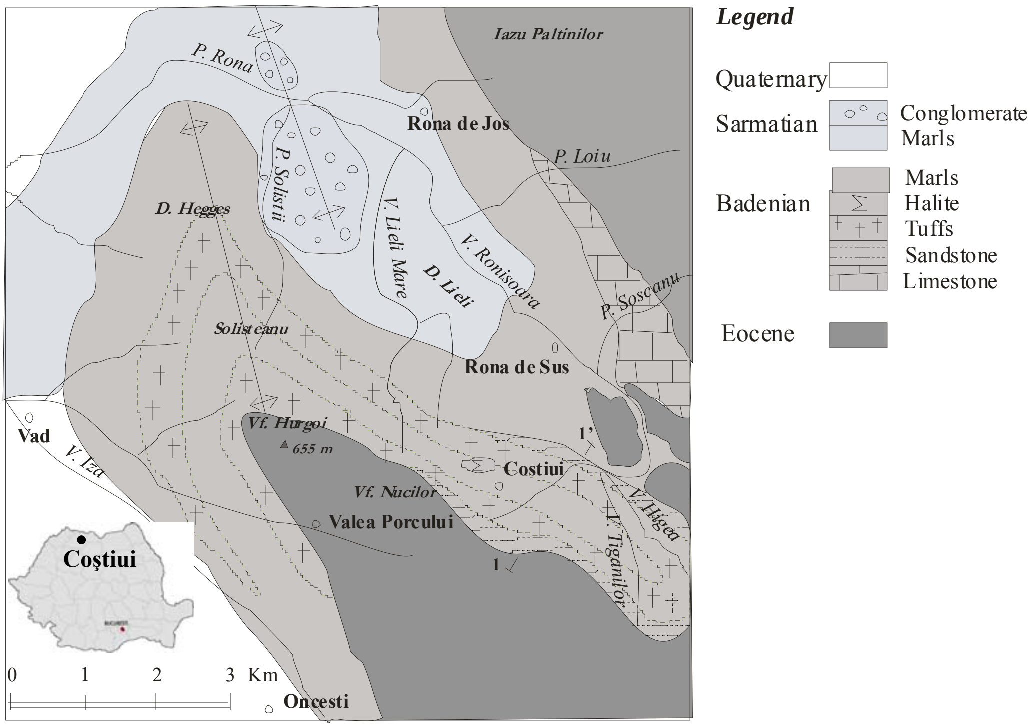

Fig. 1. Geological map of the Rona de Jos – Vad Cuştiui zone

According to Antonescu (1979) the Picuiul tuff from the Porcului Valley zone - Năneşti - Bârsana (Iza Valley) is connected with the intercalated tuffs in the formation of the salt breccia from Coştiui.

The Sarmatian formations are disposed as continuity of sedimentation over the Badenian ones. Lateral variations of facies are presented, especially in the lower part. Lithollogically one can separate marls, sandstone marls, friable sandstones and sands. The transition from Badenian to Sarmatian is done gradually, the possibility to separate the two levels on lithological bases being in most cases difficult.

The observations have been carried out on three cross profiles on the bottom level of Badenian tuffs. The profile from the Senes hill (the most western one) is the only one that allowed the sampling of the tuff from the northern flank of the synclinal. The bottom level is made up of coarse grained tuffs with pumices (15.5m), at the upper part is present an alternation of sandstone tuffs and greenstone.

The profile on the left affluent of Higea Valley I-I' (figure 2) opens the level of the Bârsana tuffs on a real thickness of 68m. The lower sequence has a thickness of 32m, being made up of coarse grained tuffs with pumices. The upper sequence has a thickness of 36m and is made up of fine tuffs with intercalations of medium grained tuffs, (figure 3).

The profile at the origin of the Higea Valley includes a 19m sequence of coarse grained tuffs sometimes with pumices at the bottom (lower sequence) and an upper sequence of about 30m of fine tuffs. From the correlation of the profiles results a constancy of the lower sequence of coarse grained tuffs with pumices, which suggests their proximal position. The upper sequence presents an increased variation over a distance of only 2.2 km. In the western part (Seneş hill) the tuffitic sequences are frequent while in the eastern part (origin Higea Valley) there are fine homogenous tuffs with an appreciable thickness of 31m.

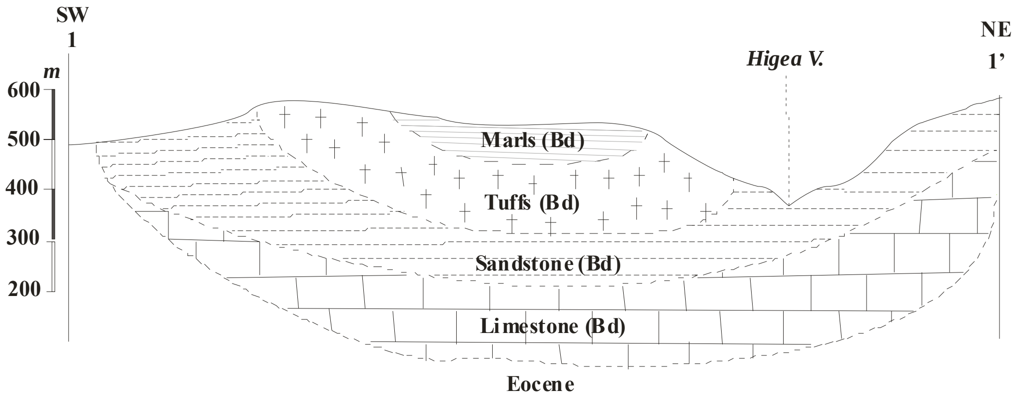


Fig. 2. Geological cross section in South – Eastern part of the Coştiui zone; Bd - Badenian

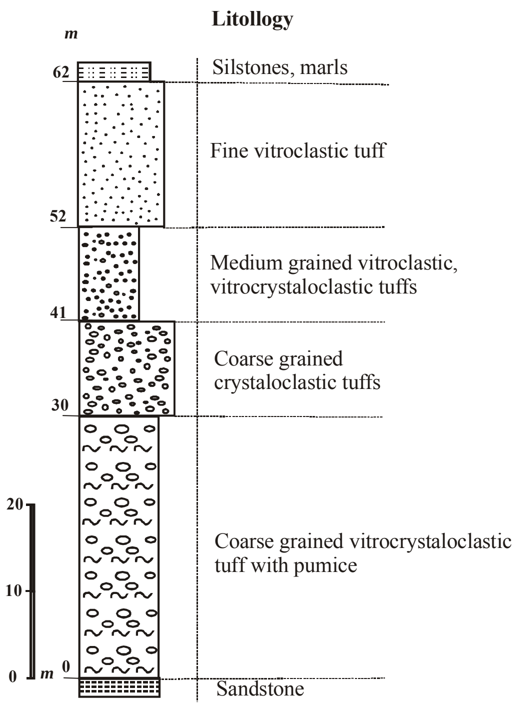


Fig. 3. Lithologic column of the Badenian tuffs

The upper sequence shows an east-west variation, which suggests the presence of an emission centre in the eastern part. The differences remarked between the variations in the lower and upper sequences suggests that the lower sequence is due to an emission centre situated very close to the investigated area, and that the products of the upper sequence have as origin an emission centre situated more in the eastern part. Which means that in the Coştiui zone the products of some different emission centres overlap; an older one, situated nearby, and a newer one, with a more eastern position. Another important observation is that the variations in the grading of the explosive products over relatively small distances reflects the existence of several local emission centres with a relatively extension existence in the inferior Badenian.

### 3. PETROGRAPHIC DESCRIPTION OF THE TUFFS

The samples from the volcanic tuffs that outcrop on Ţiganilor Valley and especially on one of its left affluents, at about 2.5 – 3 km upstream the confluence with Coştiui Valley have been studied. On Ţiganilor Valley the tuffs form banks apparently open thickness along the valley over several hundreds of meters. The succession of the tuffs from downstream to upstream has the same grain size as the Bârsana tuffs, (Damian et al. 1991)

The colour of the tuffs is green, green-blue, white-grey or with stained aspect. The green colour is due to the celadonite, (Pop et al. 1982). Macroscopically quartz, feldspar, amphiboles, lithic fragments can also be noticed besides biotite in the coarser levels; sometimes the micro-stratification is obvious and it is given by the alternance of some bands with finer grain size.

#### 3.1. Crystaloclastic tuffs

The crystaloclastic tuffs have been identified at the bottom of the tuff level. There is a gradual succesion from these tuffs to fine vitroclastic tuffs. There are hard, massive, compact rocks, without any obvious stratification and very homogenous. At the superior part of the fine vitroclastic tuffs alternations of crystaloclastic tuffs and fine tuffitic material appear. The texture is massive psamitic, the grain size of the components is rarely larger than >2 mm.

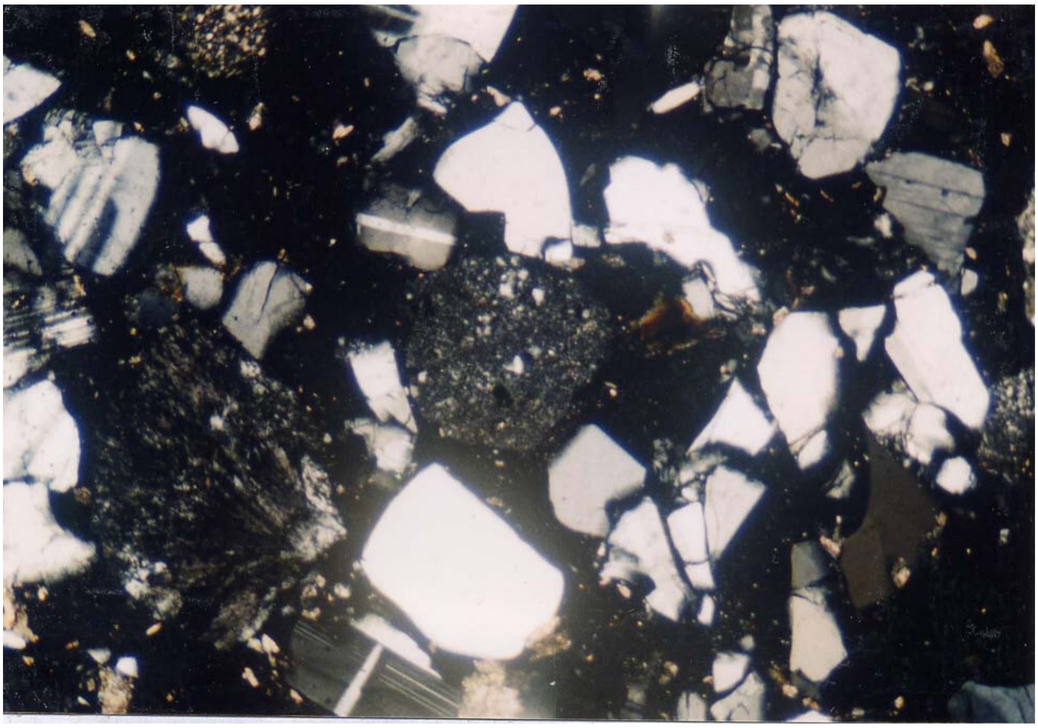

Fig. 4. Crystaloclastic tuff, crossed polarized light image, 33X

**The crystalocllasts** have sizes of up to 1-1.5 mm and represents more than 45% up to 63%, (figure 4). The crystalloclasts are represented by: quartz, feldspar (plagioclase and orthoclase), besides which appear biotite, pyroxene (augite), amphiboles (especially green hornblende), muscovite and small quantities of zircon, rutile, leucoxene, illmenite. The frequent presence of some zircon crystals also suggests a sedimentary material origin.

**The vitroclasts** have large sizes of over 0.2 mm resulted from a good hydraulic sorting. The outline of these glass fragments is obvious due to the fine clusters of clay minerals. Besides the glass fragments rare fragments of pumices also appear. The vitroclasts are substituted by zeolites, clay minerals and celadonite. The fragments of pumices are very easily noticed macroscopically due to the green colour resulted from the substitution with celadonite and chlorite in high proportion.

**The lithoclasts** are represented by fragments of sedimentary rocks: siltite fine sandstones, claystone with rounded forms demonstrating the influence of their terrigenous deposition. Besides them appear fragments of metamorphic rocks (quartzite quartz-sericite schist) and eruptive rocks.

**Diagenetic minerals** are represented especially by the zeolites that appear frequently inside the vitroclasts. The clay minerals, especially the montmorillonite substitute the micro-pumices. The authigenic calcite are also frequent. The celadonite can be easily noticed by the green colour. It is associated with the zeolites and the clay minerals in micro-pumices. The nests with secondary silica represented by quartz and chalcedony have a non-homogenous distribution in the rock groundmass.

### 3.2. Lapillic tuffs

The lapillic tuffs occurs at the bottom of the level of crystaloclastic tuffs. They can be recognised easily by the high grain size and a large amount of pumices up to 50% of the total components of the rock, and the crystalloclasts can be in some samples up to 35%. The green colour is due to the substitution of pumices by celadonite.

**The vitroclasts** are represented by glass fragments substituted by zeolites in the greatest part and represent up to 43% from rock. The vitroclastic fragments are isometric or slightly elongated or flattened being disposed in the free spaces between the pumices.

**The pumices** are elongated and include fragments of biotite (figure 5). In the highest part the fragments of pumices are devitrified and transformed into celadonite and clayey minerals and less into zeolites. (figure 6). Pumices present elongated or flattened forms, having a mainly fibrous texture.

**Chrystalocllasts** present in about 35%, are represented by fragments of quartz, feldspar, biotite, muscovite. **Lithoclasts** also have an important share being represented by sedimentary rocks (siltite fine sandstone, marls, claystone) and eruptive rocks, especially andesitic ones. They are well represented in some samples, in which they can exceed 5%, rounded and sub-angular fragments of quartzite and eruptive rocks of andesitic composition, with the basic mass of brown-reddish colour, with average crystallinity and pilotaxitic or hyalopilitic structure.

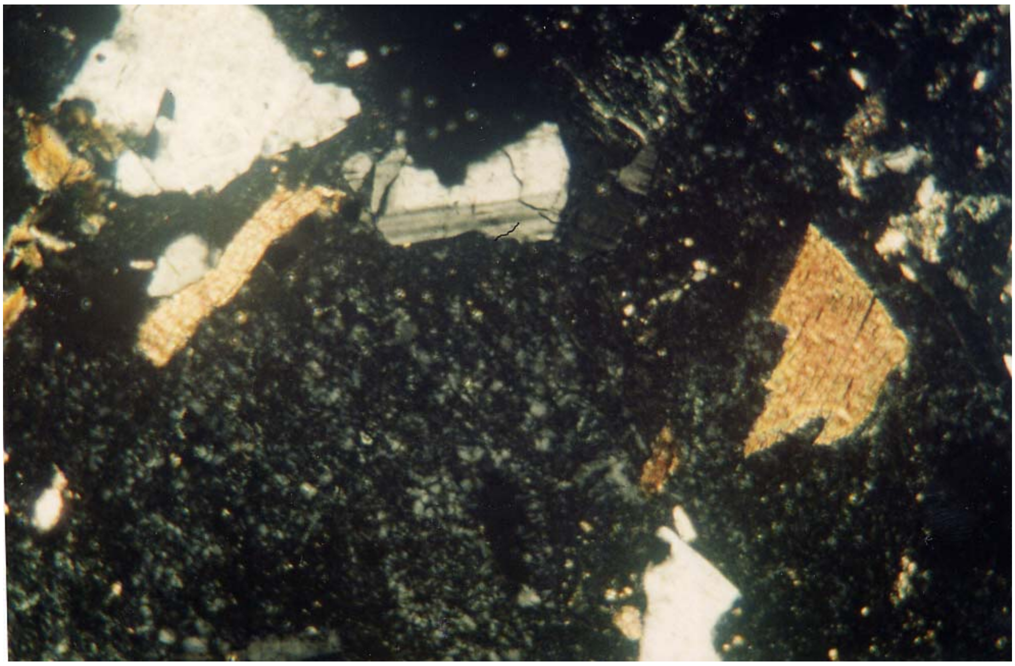

Fig. 5 Biotite crystals in lapillic tuffs with matrixes of zeolitic pumices N+, 130X.

Fig. 6 Substituted pumices of celadonite in lapillic tuffs, NII, 130X.

**The diagenetic minerals** are represented especially by smectites, celadonite, zeolites. The carbonates appear sporadically as fine pellicles and in nests. Iron hydroxides substitute the biotite fine crystals.

### 3.3. The zeolitic vitroclastic tuffs

The colour of the tuffs is green, green blue, white grey or with stained aspect. The micro-stratification is given by the alternation of some bands with finer or coarser granulation. Compositional the studied tuffs are equivalent to the rhyodacites-dacites.

**The crystalloclasts** are represented (figure 7) by plagioclase feldspars and orthoclase, quartz, micas, especially biotite, amphiboles (green hornblende) and sporadic apparitions of muscovite and pyroxene (augite). From the accessories minerals the garnet, the rutile, the zircone, the turmaline, the orthite, the leucoxene, the ilmenite have been pointed out.

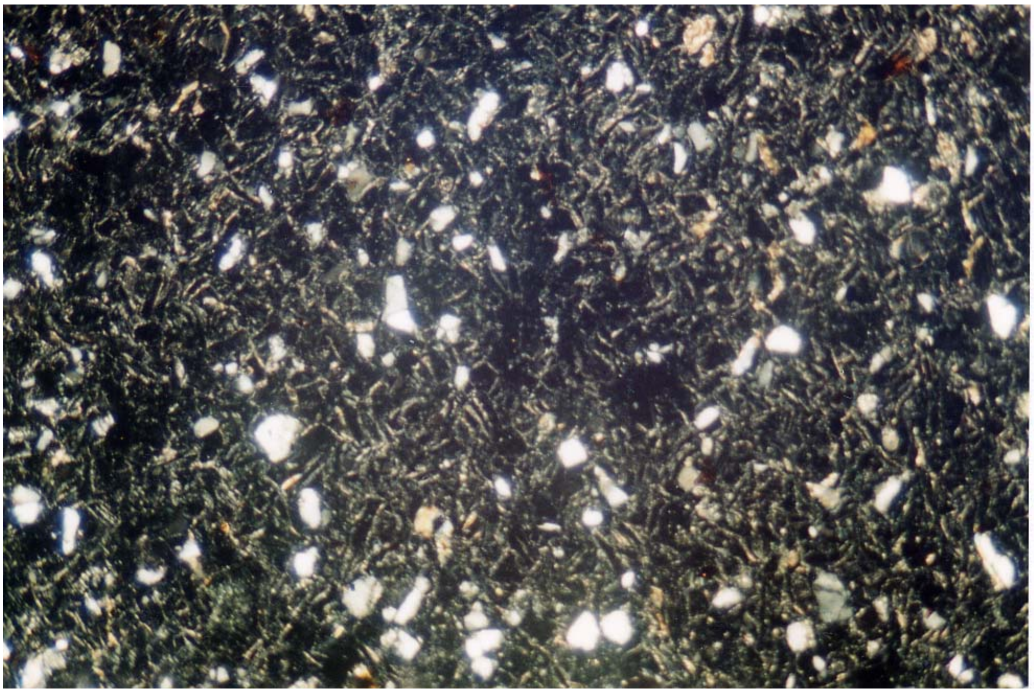

Fig 7. Vitroclastic tuff N+, 250 X

**The vitroclasts** have a participation of about 80 – 85 % in samples. They are devitrified into zeolites and clay minerals (montmorillonite). (figure 8).

**The lithoclasts**, in some samples, they can exceed 5%, rounded and sub-angular fragments of quartzite and eruptive rocks with andesitic composition, with pilotaxitic or hyalopilitic structure are present.

**The diagenetic minerals** are also well represented this case by zeolitic minerals. Besides the zeolitic minerals are also present the smectites and the carbonates.

### 4. MINERALOGY OF THE ZEOLITIC TUFFS

The mineral components have been separated according to the genetic criteria: primary magmatic minerals, represented by crystalloclasts; diagenetic minerals and minerals of neo-formation.

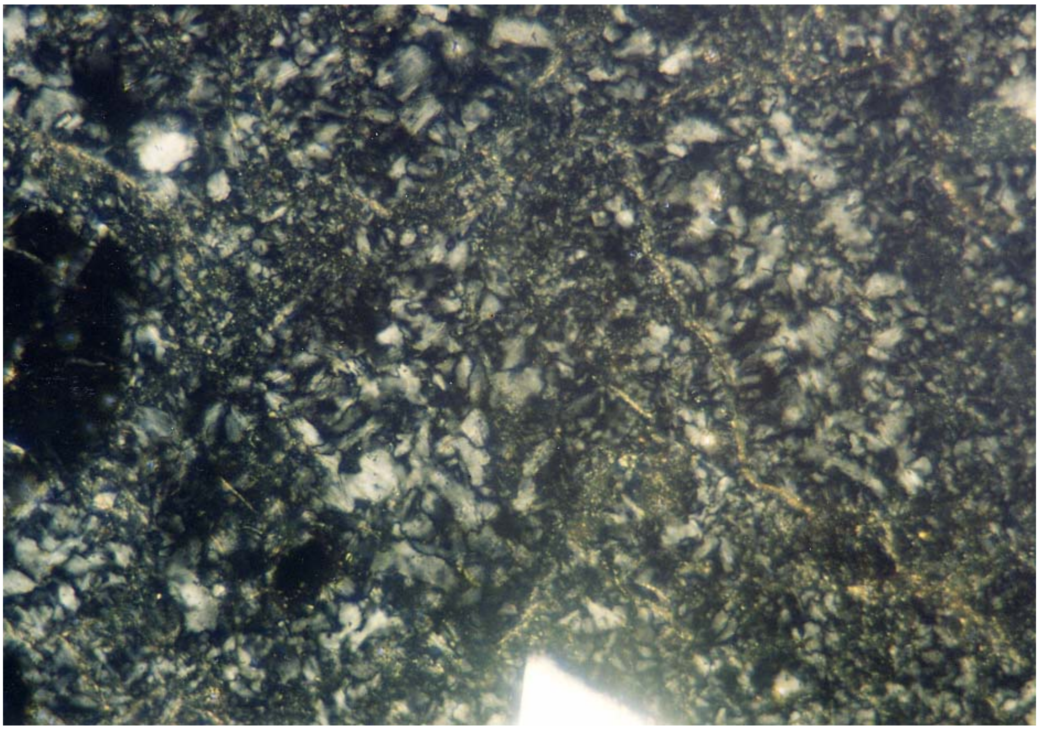

Fig. 8, Zeolites with prismatic habitus as nests in a fine zeolitized groundmass associated with the montmorillonite, N+, 250X.

**The primary magmatic minerals** existing previously to the volcanic explosion, transported and deposited in a sedimentation basin, in this case underground marine, are represented by: plagioclase feldspars, quartz, alkaline feldspars, biotite and muscovite, the amphiboles, the pyroxenes and the accessory minerals.

**The plagioclase phenocrysts** are present either as broken crystals, or as euhedral crystals, which suggests a magmatic origin. The euhedral feldspar phenocrysts existed in the magma body as isolated crystals before to explosion of the eruption, (Best & Christiansen, 1997).

The plagioclase crystals are polysynthetically twinned according to the Albit Periclin laws; they are frequently zonated. They present corrosion under the action of glass, marginally they can be surrounded by an albitic rim, sometimes they are partly sericitized (connate aspects from the pre-explosive stage). The plagioclase feldspars also contain the glass inclusions that are generally irregular in outline and in arrangement within the crystal, or are invaded by isotropic glass or glass re-crystallized in zeolites. The presence of plagioclase phenocrysts range between 1–2 % to over 15%. They are important components, especially in the coarse levels. The optical data of the plagioclase crystals suggest a range of compositions between albite, oligoclase and sometimes of the andesine.

**The alkaline feldspars** are present in quantities much subordinated to the plagioclase, being represented by albite and sanidine.

**The quartz** is present as grains of different sizes and forms, the most complex aspects being encountered in the crystaloclastic tuffs. Lots of quartz crystals are fragmented, angular, which demonstrates their magmatic source. The sub-rounded and sometimes rounded quartz crystals demonstrate a possible sedimentary source. There are fragments of crystals, rounded or magmatic corroded, cracked or broken with the zone of increase of crystobalite of high temperature. They contain inclusions of volcanic glass, re-crystallized in zeolites. The grain size is variable from 0.1mm, representative for the fine tuffs, to over 1 mm in case of coarse tuffs.

The micas are represented by biotite and muscovite. The biotite is quite frequent, it is presented as slightly undulated lamellar crystals. The muscovite occurs as elongated fine crystals. A representative development is recorded by the biotite in the coarse tuffs, where the grain size of the biotite sheets can be of 1mm. The biotite fine crystals are substituted by iron hydroxides.

The chlorites have been pointed out, but with a sporadic presence. The amphiboles are components with much subordinated participation. They are represented by the green hornblende. The pyroxenes have a sporadic presence, being represented by augite.

The diagenetic minerals were formed during the process of sedimentation the volcanic ash in the marine environment. Reactions were initiated between the glassy component and the sea water, which continued in the stage of hydrated sediment. The volcanic glass suffered a mass devitrification process with recrystallizations. The zeolites were formed in these conditions. The mineral assemblage of the zeolite group is composed of clinoptilolite, mordenite and heulandite, by using the analysis of diffraction of X rays.

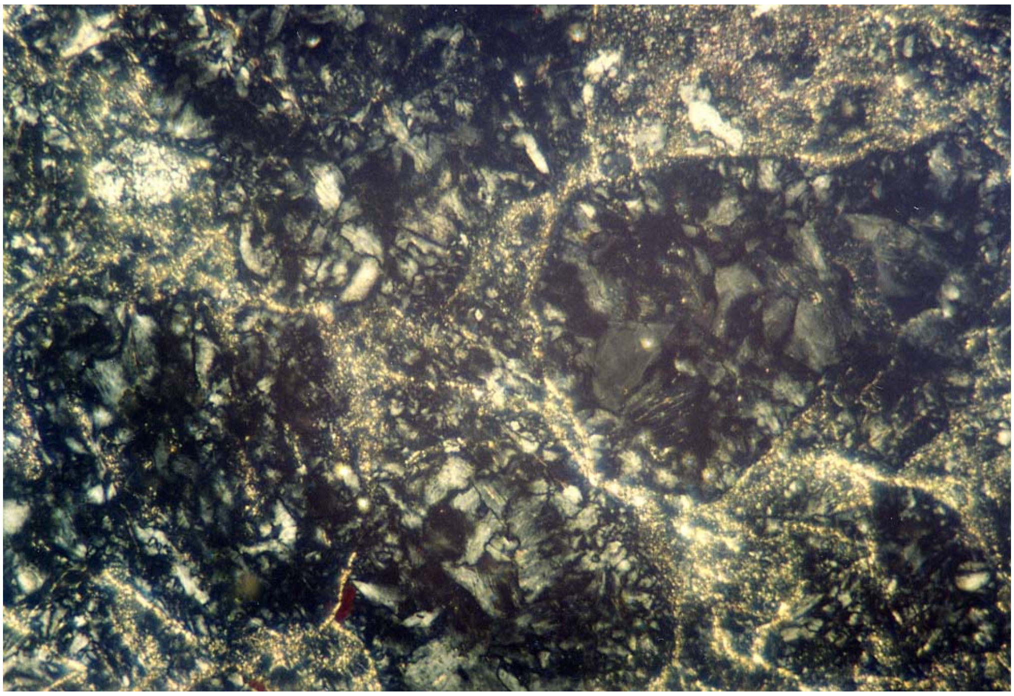

Fig. 9. Zeolites with prismatic habitus from the vitroclasts, N+, 250X

The distribution of the zeolitic minerals in tuffs is controlled by the ratio between the glassy ash that makes up the matrix and the vitroclasts. In case differences of granulation appear between the two components the apparition of two different morphologic types of clinoptilolite is registered, one that was named clinoliptolite of first type, characterised by a very fine grain size of the order of 1 – 5 microns, frequently at the limit of optical detectibility, resulted from the re-crystallization of the fine ashes that makes up the matrix of the rock, and a second type, within the outline of the vitroclasts (figure 9), with their strict observance, which have a high degree of crystallinity and sizes of 20 microns up to 40 microns.

The zeolitic minerals are distributed in nests or inside the frame of the vitroclasts. The general microscopic aspect of their distribution is non-homogenous. Besides these two ways of presentation of the zeolitic component the presence of some zeolite crystals has also been pointed out in the micro-vacuoles or in the cracks from the rock.

The clinoptilolite from Coştiui presents a X ray diffraction spectrum (figure 10) with a very strong and sharp reflex of 8.9Å and the reflexes of 8.91 Å, 2.96Å, 3.96Å, which correspond to the diffraction data presented in literature for clinoptilolite are well marked (Valiter et. al .1975).

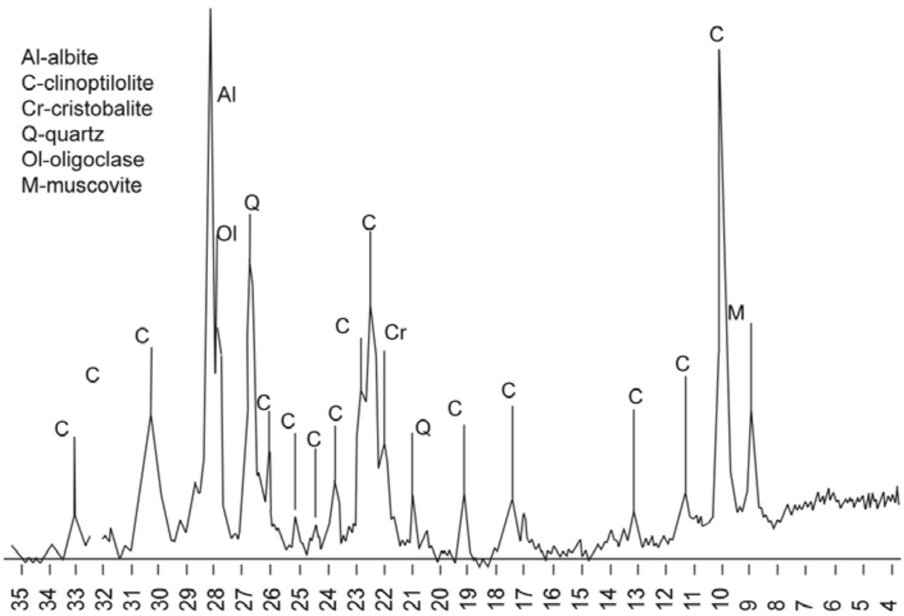


Fig. 10. The spectre of X ray diffraction of the clinoptilolitic tuff from Coştiui

The thermal curves carried out on zeolitic tuffs (figure 11) are characterised by a single endothermic effect with the maximum between 140-200ºC, to which corresponds a continuous and progressive loss of weight because of eliminating water. The total loss in weight is of about 115 and it is close to the theoretical content of water from the structural formula of the clinoptilolite. It results that the fine tuffs from the Coştiui zone have a high content of zeolites. Also the weight loss is identical with the water content resulted from the chemical analysis.

The presence of the heulandite has been pointed out optically and confirmed by the diffraction analyses. It is generally present in nests as more elongated crystals. The quantities in which it occurs are very reduced.

The mordenite appears associated with the clinoptilolite as some fine fibrous needle-like crystals and was pointed out by diffraction analysis. Other re-crystallisation products of glass are represented by smectite minerals, mainly montmorillonite, present in two aspects: forming micro-nests individual or without a well-defined crystal shape around the vitroclasts.

The cryptocrystalline silica is individualised as micro-nests or by forming spherulitic aggregates made up of cryptocrystalline silica, with undulatory extinction.

The celadonite is present in all the samples of studied tuff, the characteristic green colour due to its presence. It is present as rounded grains, sometimes with zonal structure. The grain sizes of the celadonite are of the order 0.078-0.055mm.

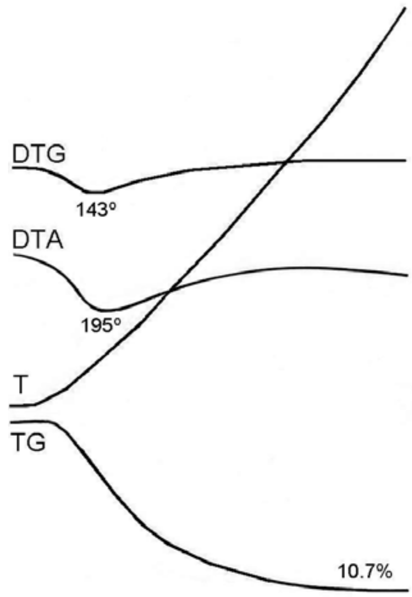


Fig. 11 Thermal curve of the zeolitized tuff from Coştiui, DTA 1/10, TG- 200, T° - 1000°C, Total loss =10.7%.

**The minerals of neoformation** are represented by the iron hydroxides, carbonates and a part of the cryptocrystalline or amorphous silica.

The iron oxyhydroxides occur as fine grain impregnated into the micro-fracturing or infiltrated in mechanical micro-discontinuities. A part of the iron hydroxides have a secondary character resulted from the alteration of the biotite. Sometimes it develops as a rim at the contact surfaces of the crystalloclasts or of the vitroclasts with the matrix of the rock.

The carbonates have a character of neoformation forming impregnations in the rock, as a result of the circulation of some downward solutions over zones from the microfractures. The carbonates appear especially in coarse grained tuffs and can have a zonated structure, the central part consists of dolomite and calcite surrounded by a sideritic material.

The colomorphous silica appears as micro-veins, with thicknesses of 0.5-1.5 mm reflecting mobilization of the coloidal silica.

## 5. PHYSICAL – CHEMICAL CHARACTERISATION OF THE ZEOLITIC TUFFS

The chemical composition of the volcanic tuffs from the zone Coştiui is presented in figure 12. The contents of $SiO_2$ with a restricted range between 63.19-68.04%, with an average of 66.78% are similar with the contents from the other occurrences from the Maramureş basin (Damian et al.1991, Cochemé et al. 2003). The high content in $SiO_2$ is related with the volcanic glass devitrified in zeolite. The chemical composition of the Costiui tuffs correspond with the dacitic character.

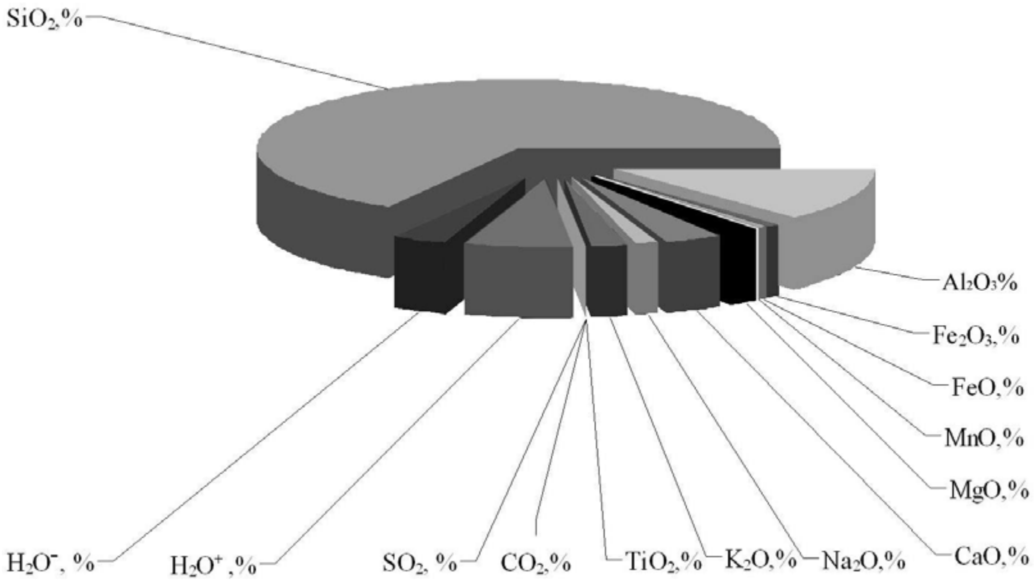


Fig. 12 Distribution of the chemical composition of the zeolitized tuffs in the Coştiui zone.

$Al_2O_3$ present a wide range between 9.73-15.01%, with an average of 12.59%. $Fe_2O_3$ presents values in a restricted domain 0.46-1.32% with an average of 0.91%, and FeO varies between 0.27-0.83%, with an average of 0.46%. MnO is present in small quantities, between 0.01-0.18%, with an average of 0.05%. MgO varies within rather big limits 0.86-2.40%, with an average of 1.74%.

CaO presents restricted limits of variation between 3.24-5.39% with an average of 3.84%. The content of CaO is rather high, giving the zeolites a predominantly calcic character. $Na_2O$ varies between the limits 0.67-2.77%, with an average of 1.40%, and $K_2O$ generally has rather homogenous values, with rather restricted variation limits, included between 1.59 – 2.19%, with an average of 1.99%.

Water ($H_2O^+$) has values between 4.91 – 8.80%, the average content being of 6.62, at which $H_2O^-$ is added.

The cationic exchange capacity has been determined both for the monovalent cations ($Na^+$, $K^+$) and for the divalent ones ($Ca^{2+}$, $Mg^{2+}$). For the tuffs in the Coştiui zone the cationic exchange capacity (figure 13) have higher values for $Ca^{2+}$ and lower values for $Na^+$. The average for $Ca^{2+}$ is rather high 63.11 mvali/g, but the extreme values are between very large limits 1,25 to 105,15 mvali/g. The cationic exchange capacity for $Na^+$ is low, with large variation limits 3.03 to 81.58 mvali/g, but the average is rather low 18.24 mvali/g, which demonstrates that the low values are frequent. For $K^+$ the values are between 14.00 to 33.92 mvali/g with an average of 25.75 mvali/g. For $Mg^{2+}$ the values of the cationic exchange capacity are maintained between the limits of 1.48 – 11.94 mvali/g, with an average of 5.45mvali/g.

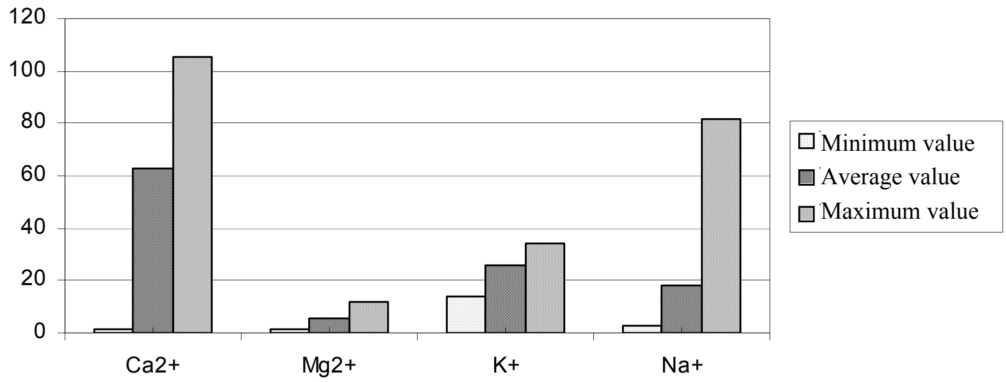


Figure 13. Distribution of the cationic exchange capacity, Coştiui zone

The zeolitic tuffs from the Coştiui zone have a surface area that varies between 12.4 – 21.9 $m^2/g$, with an average of 18.25 $m^2/g$ (figure 14).

The determined values of the densities are included between 1.83 – 2.16 $g/cm^3$ and the average is of 1.99$g/cm^3$.

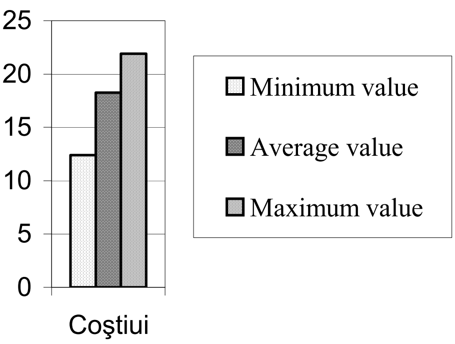


Figure 14. The distribution of the measurements of the surface area

### 6. DISCUSSIONS AND CONCLUSIONS

The zeolitization process of the tuffs has a pervasive character and it has affected especially the volcanic glass, deposited in a sedimentation basin with fluctuating salinity. Zeolitized tuffs can be formed during emplacement of volcanic material and the water with a certain salinity which also continued after the volcanic material has been covered with sediments, in the stage of diagenesis of the hydrated sediment.

The zeolitic minerals have been formed by the in situ diagenesis of the ryodacitic volcanic glass. The clinoliptolite is the zeolitic mineral predominant in the tuffs from the Coştiui zone, being similar from this point of view with those from the Iza basin (Damian et al. 1991, Cochemé et al 2003). The zeolitization process is identical with the one from the Transylvania basin described by Bedelean & Stoici (1984). The tuffs from the Transylvania basin contain the same authigenic minerals or zeolites. The sinking of the post-Badenian sediments was not very big because one did not reach the limit of metamorphism of low intensity characteristic for the facies laumontite pumpellyte – prehnite, which was not pointed out. The physical characteristics of the zeolitic tuffs do not correspond with the debut stage of the metamorphism, because they have a high porosity, they are not strongly hydrated and have a reduced degree of crystallinity.

Related to the well-known genetic models, the deposits of zeolitic tuffs from the zone Costiui belong to the "open system deposits" type (Steppard & Gude 1973, Mumpton 1978) and are placed in the zone with clinoptilolite montmorillonite (Hay 1978).

The tuffs from the Coştiui zone have an appreciable content of zeolites and, due to their physical and chemical characteristics they can be used in different industrial domains and for environment protection, according to Mumpton (1999).


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