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NONTRONITE AS THE WEATHERING PRODUCT IN THE ROCKS OF SITNO EFFUSIVE COMPLEX IN PUKANEC REGION

(Figs. 9)

Abstract: Iron-rich smectite mineral, nontronite was found as a product of the weathering of andesites and its pyroclastic equivalents belonging to the Sitno effusive complex. The presence of nontronite in the weathered decomposed rocks was proved using electron microscopy, X-ray diffraction and thermal analysis methods. The correlation between the character of the parent rocks and the weathering products composition was observed. The present work tries to solve a new problems concerning the secondary alteration processes connected with mineralization of the subvolcanic intrusive complex in Pukanec region.

Резюме: Железистый смектитовый минерал, нонтронит, был найден как продукт выветривания андезитов и их пирокластических эквивалентов принадлежающих Ситнянскому эффузивному комплексу. Присутствие нонтронита во выветрившихся разложившихся породах было доказано при помощи электронного микроскопа, рентгенографического изучения и термических аналитических методов. Была наблюдана корреляция между характером материнских пород и составом продуктов выветривания. Настоящая статья стремлится решать новые проблемы касающиеся вторичных процессов изменения в отношении к оруденению субвулканического комплекса в регионе с. Пуканец.

Introduction

Intensive geological survey in the wide-spread area of Pukanec environment offered a new ways in the study of mineralogy, petrography and metalogenesis.

Two geological units are in contact in this region:

1. the volcanoplutonic formation in the north belonging to the Nová Baňa—Kľakov volcanotectonic zone;

2. the isolated Pukanec Sarmat—Pannonian depression in the south, containing lignite ad clays.

The rocks of the volcanoplutonic formation (pyroxene and pyroxene-amphibol andesites as well as quartz diorite porphyries along with elder granodiorite porphyries) are secondarily altered. The strong alteration occurred especially near the phenomenon of mineralization. In this connection Forgáč (1980) considered the following important points: potassium metasomatism (adularization), chloritization, silicification and finaly argillitization and propylitization as the forms of the younger alteration. The altered rocks were desintegrated and re-

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deposited into the sedimentary basin in which during diagenetic process the beds of kaolinite and halloysite clays were formed together with the bulky accumulation of lignite.

The mineralogical composition of these sediments was studied many times previously (Harman, 1956, 1964; Mišík — Číčel — Marková, 1958 and others). Tomschey — Harman — Blaško (1986) using the method of the trace elements analysis consider the rocks of the volcanoplutonic formation (its north-western part) as the parent rocks of the sediments.

Complicated stage alteration of the rocks of the volcanoplutonic complex has started with the intensive chloritization and illitization (sericitization?). The presence of kaolinites, epidote and small amounts of carbonates was found near zones of mineralization. The presence of smectite-type minerals in the altered rocks of the volcanoplutonic formation has never been described.

We have found the iron-rich smectite, nontronite in the altered pyroclastic rocks occurring near the road to Uhliská (northern margin of the Pukanec depression), in the pyroclastic samples extracted from the bore-holes situated in this region as well as in the erosion gorge of the Sikenica brook in the southern part of the depression (see Fig. 1). The pyroclastic material from the northern part of the depression represents the fine- to medium-grained strongly altered tuff, light-green in the colour, containing only insignificant amounts of dark minerals. Towards south as well as in deeper beds it changes gradually to the coarse-grained pyroclastic material as much as to grey and greyish green intrusive breccias which are likely the equivalent of the pyroxene andesites in the Sitno effusive complex (Konečný — Lexa — Planderová, 1983). The question whether these rocks are forming also the bottom of the Pukanec basin is not solved. The bores situated in this area did not reach the bottom of the basin — they served for the study of the sedimentary filling. The drillings verified similar coarse-grained strongly altered pyroclastic rocks underlying kaolinite clays and lignite, containing limonite but no notronite. They represent likely the volcanoclastic redeposited material.

In the smaller extent we have found nontronite as a product of the weathering of the pyroxene andesites occurring in the northern part of the basin. But there is no notronite in the weathering products of amphibole-pyroxene andesite occurring in the southern part. This type of volcanic rocks belongs to Priesil effusive complex according to Konečný — Lexa — Planderová (1983) and its boundary with the pyroclastic rocks seems to be tectonic.

Two occurrences of nontronite as the product of volcanic rocks alteration in Slovakia was described previously. Forgáč — Číčel (1965) found nontronite and SiO_2 modification (chlor-opal) originated from pyroxene andesite and pyroclastic rocks in south-western part of Prešov-Tokaj Mts. near the Skároš village. Nontronite arised as a product of the secondary alteration of dark minerals (pyroxenes) due to influence of the circulating surface water. Kraus — Polakovičová (1967) described the mineralogical assemblage of nontronite and beidellite in the fissures and beds of pyroxene-andesites and pyroclastic rocks occurring in Badín valley (Kremnica hills region). The parent rocks were tectonically altered. Kraus — Gerthofferová (1968) have extended the knowledges about the occurrence of nontronites in Slovakia studying the iron-rich smectites comming from Sampor and Veľká Lúka regions. While nontro-

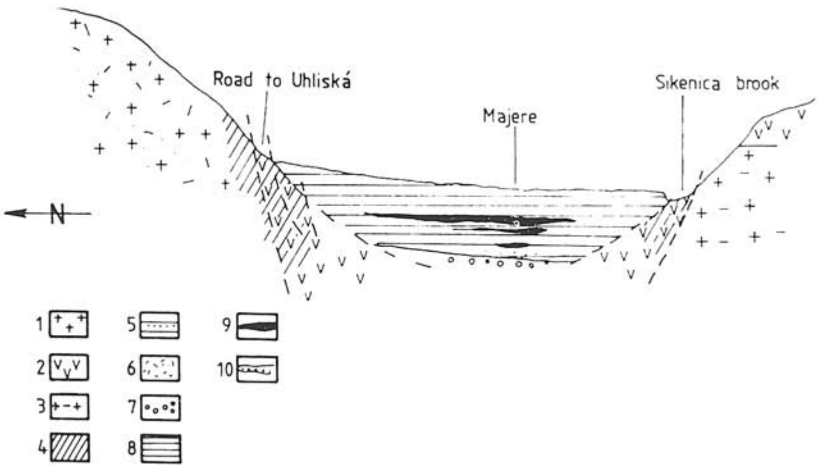

Fig. 1. Schematic profile of the nontronite occurrence at the margin of the Pukanec depression.
Explanations: 1. Volcanoplutonic formation of the southern part of volcanotectonic zone including subvolcano-intrusive complex, amphibole-pyroxenic andesites at the surface; 2. intrusive as much as autoclastic breccias and pyroclastic rocks: 3. amphibole--pyroxenic andesites of the Priesil effusive complex; 4. zone of nontronitization; 5. zone of illitization; 6. zone of chloritization; 7. zone enriched by Fe-oxides; 8. sediments of the Pukanec depression (Sarmat—Pliocene); 9. lignite; 10. diatoms.

nite-beidellite association from Badín and Sampor localities is the product of the surface weathering of pyroxene andesites and tuffs because of the effect of the circulating surface warter in the tectonically disturbed zone at pH more than 7, the occurrence of nontronite at Veľká Lúka locality is allochthonous.

The relation of the origin of nontronites and their chemical composition as well as their properties were summarizied by Vitovskaja (1983). Differences in the genesis of nontronite are reflected in its compositional and crystallochemical features. The most important signs are the oxidation state of Fe in the elementary unit and the isomorphic substitution in the sequence Fe—Al—Mg in the octahedral sheet of the structure. The presence of the ferrous (Fe^{2+}) ions is typical of the hydrothermal conditions of the origin (Bishoff, 1972), ferric (Fe^{3+}) ions are in the nontronite structure arising in the weathering processes. The composition of the octahedral sheet reflects to the great extent the chemical composition of the parent mineral. Nontronites derived from ultrabasic rocks contain in the structure more Mg, less Al and sometimes also Ni atoms. Example of a direct transformation mechanism from pyroxene to nontronite when the morphology of the parent mineral was preserved was described by Eggleton (1975). Using the combination of the optical and X-ray diffraction methods he has demonstrated the coherence of the sructural characteristics of pyroxene hedenbergite and nontronite. Defining the reaction conditions in the weathering of serpentinites to nontronites Vitovskaja (1978) has suggesed nearly neutral reaction environment (pH = 7) and medium presence of SiO_2 (35 ppm) and MgO (10 ppm) in the solution. The weathering of pyroxenes starts with the proton attack, release of Ca^{2+} ion and oxidation of Fe^{2+} to Fe^{3+} (E g g l e t o n, 1975).

On the problems of nontronite identification

The chemical analysis is the best method for the nontronite characterization in the case of the monomineral sample. In the sequence of the smectite type minerals

$$(Si, Al, Fe)_8 \ (Al, Fe, Mg)_4 \ O_{20} \ (OH)_4$$

nontronites are defined according to the composition of the octahedral layer as proposed by B r i g a t t i (1982):

$$(Fe_{3.34}Al_{0.66}Mg_{0.0}) — (Fe_{2.66}Al_{0.0}Mg_{1.34}) — (Fe_{2.00}Al_{1.34}Mg_{0.66})$$

what does not eliminate a possibility of the presence of also another atoms in the octahedral sheet (Ni, Zn and others). The occurrence of the monomineral samples of nontronite is very rare. Mostly we have to treat the mixture of nontronite with another smectites as montmorillonite and beidellite. Then its identification is more problematic. All smectites as a three-layered swelling minerals are giving similar diffraction patterns by means of the X-ray diffraction method, before all very similar basal reflections. The chemical composition of the structural unit of the smectite minerals is reflected by position of d(060) reflection (occurring in the 1.4—1.5 Å region) but, unfortunately, this diffraction line is not sharp enough (the peak is broad) making the troubles in the identification. Therefore the electron microscopy seems to be the most suitable method for identification of nontronite (H a r m a n, 1968; B e u t e l s p a c h e r— — V a n D e r M a r e l, 1968 and others).

Low thermal and chemical stability of nontronite compared with montmorillonite is another significant feature valuable for its identification. Dehydroxylation of nontronite starts immediatly after its dehydration and proceeds between 350—500 °C which is by 150—200 °C lower temperature range than for montmorillonites (H o r v á t h, 1980, 1983). Low chemical stability of nontronite, first of all in acid environment is even more significant. Č i č e l—N o v á k (1976), studying a half times of dissolution of smectites in hydrochloric acid, found by 500—600 times faster decomposition rate for nontronite compared with montmorillonite. These properties are important in the evaluation of further alteration of nontronite in the weathering and diagenetic processes.

Materials and methods

The mineralogical investigation was performed on the samples collected predominantly from the geological bores VPC 76, VPC 96, VPC 81, VPC 82 and VPC 93 (Geological Survey, Spišská Nová Ves, Banská Bystrica Geological Centre). The fraction less than 20 μm was prepared by sedimentation for the chemical analysis, electron microscopy, thermal analysis and X-ray diffraction method.

X-ray diffraction analysis was performed using Philips diffractometer with Ni-filtered CuK_{α} radiation (40 kV, 20 mA). Powder samples of clay were prepared by placing aqueous suspensions onto glass microscope slides and drying them at 60 °C. X-ray diffraction patterns are shown in Fig. 2.
The spacings at 15.5, 4.47 and 2.50 Å are dominant reflections indicating the presence of the smectite type mineral. The 15.5 Å spacing is changed to 18.4 Å after glycerol saturation (X-ray pattern No. 5, Fig. 2) which is attributed to the typical smectite-glycerol complex. Another basal spacings are unique, in exception of the patterns for the standard nontronite from East Ural (U.S.S.R.) in which the diffraction maximum at 7.33 Å represents d(002) spacing of nontronite (X-ray patterns No. 1 in Fig. 2).

Another spacings indicate the presence of the admixtures of plagioclase at 3.20 Å and 4.05 Å. Maximum at 6.4 Å occurs in case of higher concentration of feldspars in the sample indicating (020) reflection of plagioclase. The presence of SiO_2 modifications reflects maximum at 4.04 Å (cristoballite) and 3.75 Å (tridymite). The X-ray patterns of the sample VPC 96 (54.6 m) shows the presence of hydromica (10.06 Å) and chlorite (7.16 Å).

DuPont 990 Thermoanalyzer and TGA 951 module were used for TG analysis (sample size 15—20 mg, N_2 flow 1 $cm^3 . s^{-1}$, heating rate 10 °C . min^{-1}).

TG and DTG curves are shown in Fig. 3. Dehydration of the sample occurs between 25—210 °C at about 6 % of the weight loss representing the release of the physically bonded water. DTG curve shows that water release occurs in various stages which are typical of dehydration of smectites containing Ca^{2+} and/or Mg^{2+} cations in the interlayer space. Dehydroxylation proceeds at temperature interval 300—600 °C which is typical of the smectites containing a large amount of the Fe atoms in the structure. Dehydroxylation is accompanied by the weight loss of about 5 % (Fig. 3).

Taking into consideration the great similarity of the smectites X-ray diffraction data we took for very important the morphological study of the minerals. Electron micrographs were obtained by TESLA BS 500 electron microscope. The samples after ultrasonic desintegration (2 min. at 24 kHz) were studied using the method of the suspension.

Electron micrographs (Figs. 4—12) show significant differences in morphology of smectites: prolongated lath shaped crystals of nontronite and very fine dispersed foliage particles of beidelite. The crystals of nontronite despite their similar morphology exhibit some differences. Mostly they are splitted along prolongation, some individual laths are splitted across the prolongation (Fig. 4). These morphological differences reflect likely the pseudomorphosis after the parent mineral, most probably after pyroxene. The great part of the nontronite crystals exhibits the ledge-shaped morphology which are rolled up, especially in the samples of the weathered andesites. The length of the crystals reaches as much as 12 μm, but this size (with the exception of the pseudomorphic crystals) may be influenced by the preparation technique due to easy splitting properties of nontronite. The nontronite crystals are very well distinguished from another smectites. Their edges and boundaries are very sharp, part of them are corroded. This feature is connected likely with the low stability of the crystal structure of nontronite.

Fine dispersed, mostly sharp bordered particles with the size about 0.5 μm represent, in our opinion, the mineral beidelite which forms the smaller part

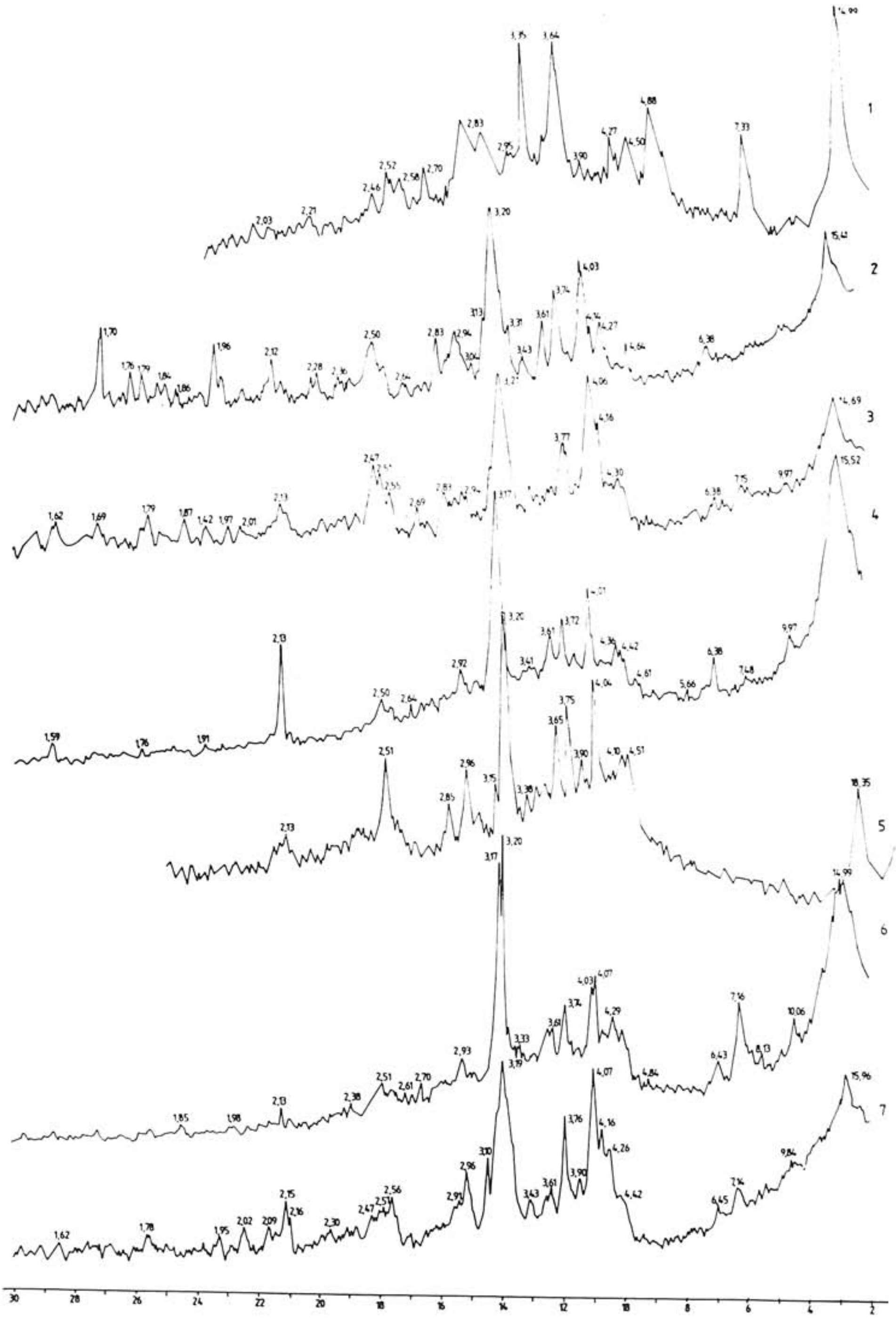
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Fig. 2. X-ray diffraction patterns of the altered pyroclastic rocks from the Pukanec region.
Explanations: 1. standard mineral nontronite — southern Ural (U.S.S.R.); 2. weathered amphibole-pyroxenic andesite, road to Uhliská; 3. weathered amphibole-pyroxenic andesite, Teplá Voda region; 4. decomposed vyroclastic rock (bore VPC 76, 23.4 m); 5. decomposed pyroclastic rock (bore VPC 76, 23.4 m), intercalated with glycerol; 6. decomposed pyroclastic rock (bore VPC 96, 54.6 m); 7. decomposed pyroclastic rock (road Majere-Uhliská).

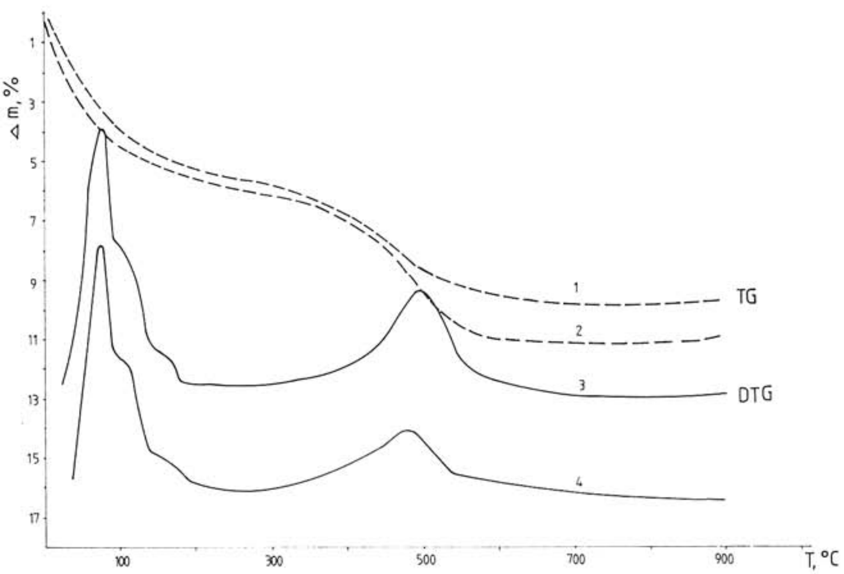

Fig. 3. DTG and TG traces of the altered rocks of the Pukanec region (bore VPC 76, 23.4 m, curves Nos. 1 and 3; bore VPC 96, 54.6 m, curves Nos. 2 and 4).

of nontronite-beidellite association (Figs. 4, 5, 6, 7). Globular forms of Fe-oxides as well as other secondary minerals are present in electron micrographs besides the smectites. Large, dark, irregulary bordered particles (3—13 μm in the size) which do not permit electronic rays belong to SiO_2 modifications and to the relics of undecomposed primary minerals.

The chemical composition of the typical samples: (wt. %)

Sample No.	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	TiO_2	K_2O	Na_2O	L.I.*)
1.	60.0	19.0	6.3	3.1	0.4	0.9	2.3	2.1	5.4
2.	60.3	20.4	6.0	1.8	0.3	0.5	0.5	2.6	6.2
3.	54.0	21.3	8.5	5.0	1.3	0.7	2.0	0.6	6.5

*) The loss on ignition at 1000 °C.

Sample No. 1. decomposed, altered pyroxene andesite Teplá Voda; 2. pyroclastic material of the pyroxene andesite, Majere, road to Uhliská; 3. intrusive breccia, bore VPC 76 (23.4 m). (Chemical analysis made by Geological Survey Spišská Nová Ves, Laboratory Centre Turčianske Teplice).

The results of the all methods have shown that the nontronite with the lower amount of beidellite are chief clay minerals in the studied samples. The most significant feature is their morphological characteristics. Nontronite affected the bulk chemical composition contributing to the increase of Fe_2O_3 content and to the decrease of dehydroxylation temperature. The content of nontronite in the studied rocks may be estimated to the 25—30 %. According to the chemical analysis and some genetic factors it may be supposed that nontronite present in the samples contains increased amount of Al and Mg atoms in its octahedral sheet close to the composition of the elementary unit ($Fe_{2.00}$ $Al_{1.34}$ $Mg_{0.66}$) defined by Brigatti (1982).

Discussion and conclusions

New information about the occurrence of nontronite in the Pukanec region extends the knowledge of one type of alteration of the rocks in volcanic complex starting with the potassium-metasomatic alteration (Forgáč, 1980; 1985), hydrothermal alteration trough the exogenetic alteration of the pyroclastic rocks especially and finishing with the diagenetic alteration of these rocks in the sediments of the Pukanec depression in the south-eastern part of the Bátovce basin. It is evident, that described features of the nontronitization are typical of the weathering of andesites and their pyroclastics which was affected by two points:

a) intensive tectonometamorphosis of the pyroclastic rocks occurring at the margin and at the bottom of Pukanec depression made possible the easy circulation of the surface water;

b) petrographical characteristic of the parent rock (glassy-like components in the pyroclastic rocks are easily weathered).

The mineralogical composition of the parent rocks gives the evidence that nontronite is an alteration product of dark minerals (pyroxenes and amphiboles). Intermediary feldspars and or volcanic glass from pyroclastic rocks were the sources of the origin of other smectites (beidellite, montmorillonite).

Slightly different kind of weathering of petrographically similar rocks resulting in the origin of Fe-rich montmorillonite as the weathering product, occurred at neighbouring locality near Brhlovce village (Harman—Horváth, 1987). The differences may be attributed to the different origin of the primary rocks: intrusive pyroclastic rocks from Pukanec region in distinction from the sedimentary pyroclastics deposited in the water basin at Brhlovce.

The boundary of nontronite occurrence in Pukanec region in the north is connected with the upper zones of pyroxene andesites and subvolcanic intrusive complex in which chloritization and illitization were dominant alteration processes. In the south nontronite occurrence is confined due to the action of the penetration of the surface water from the basin. This environment, containing carbonic acid (particularly HCO_3^- anions derived from its first stage of dissocia-

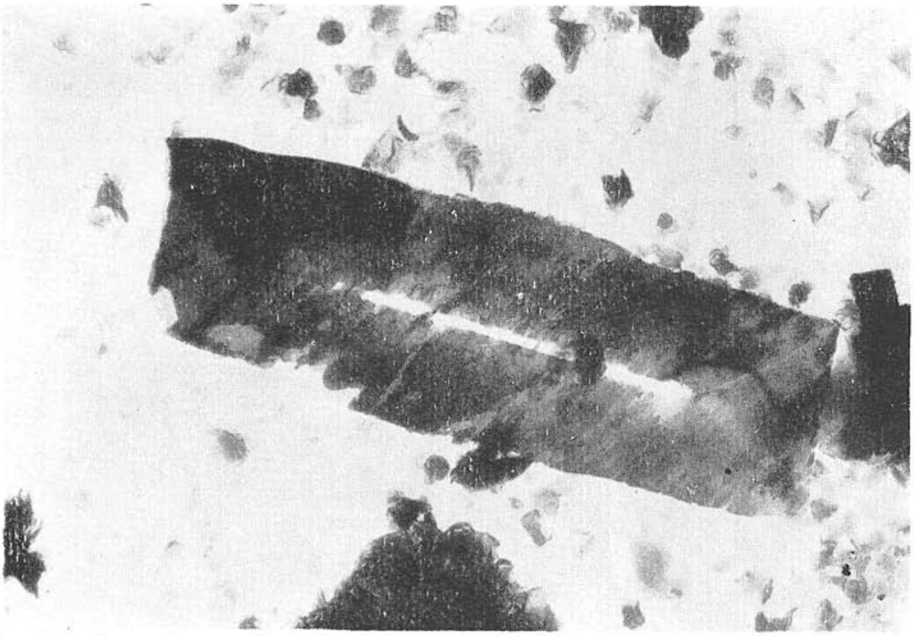

Fig. 4. Electronmicrograph of the nontronite crystal (pseudomorphosis after pyroxene), fine-grained beidelite (small particles), bore VPC 76, 23.4 (suspension, magnific. 15 000 ×).

Fig. 5. Prismatic morphology of the nontronite crystal with small particles of beidelite (sample — bore VPC 76, 23.4 m), suspension, magnific. 15 000 ×.

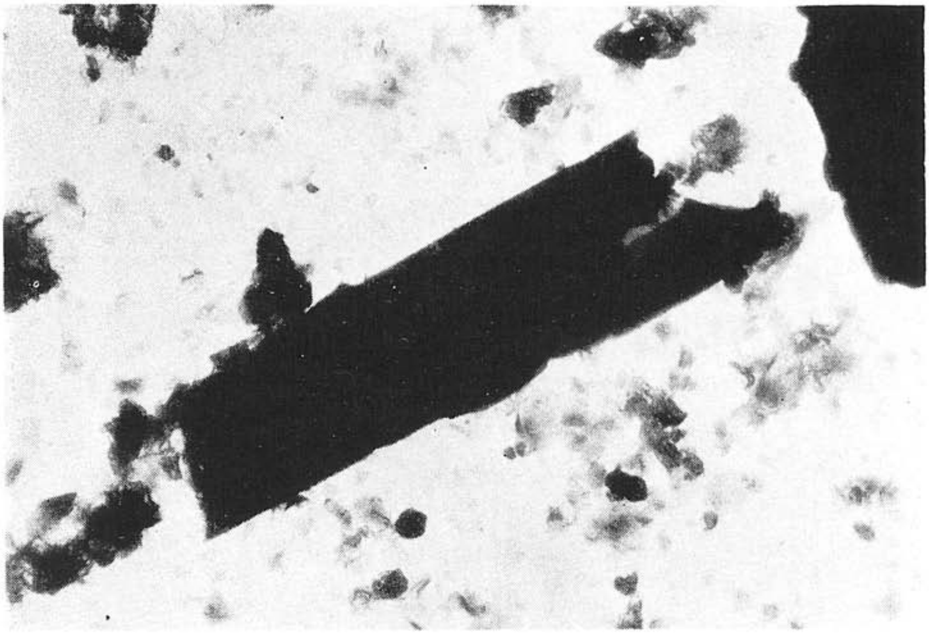

Fig. 6. Prismatic morphology of nontronite crystal with parallel splitting (sample — bore VPC 96, 54.6 m) suspension, magnific. 12 000 ×.

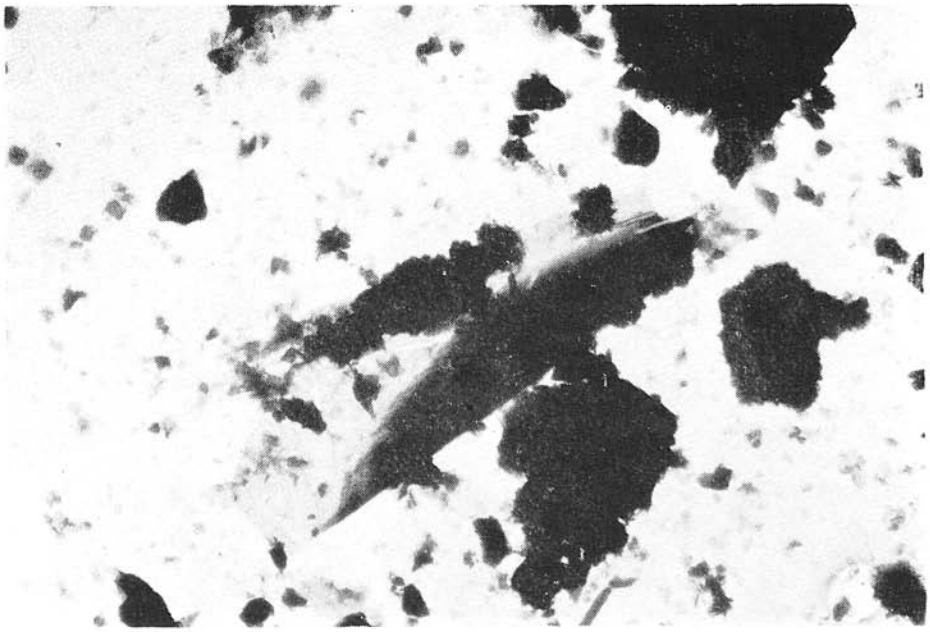

Fig. 7. Partly rolled up particles of nontronite (decomposed pyroclastic rocks, road Majere-Uhliská), suspension, magnific. 12 000 ×.

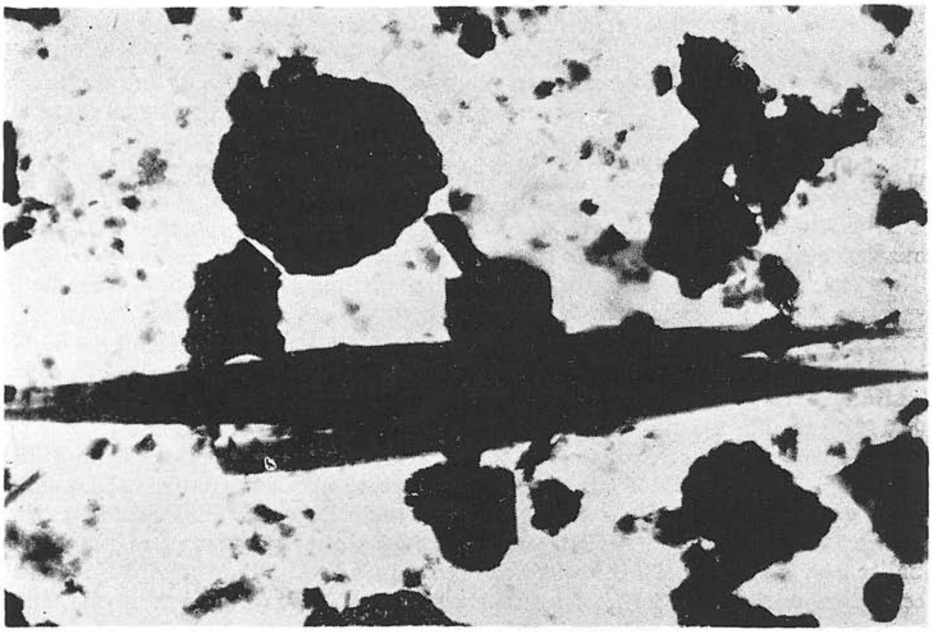

Fig. 8. Nontronite with the significant parallel splitting, dark particles of the parent minerals, fine-grained beidelite, (decomposed amphibole-pyroxenic andesite, sample — road Majere-Uhliská) suspension, magnific. 6 000 ×.

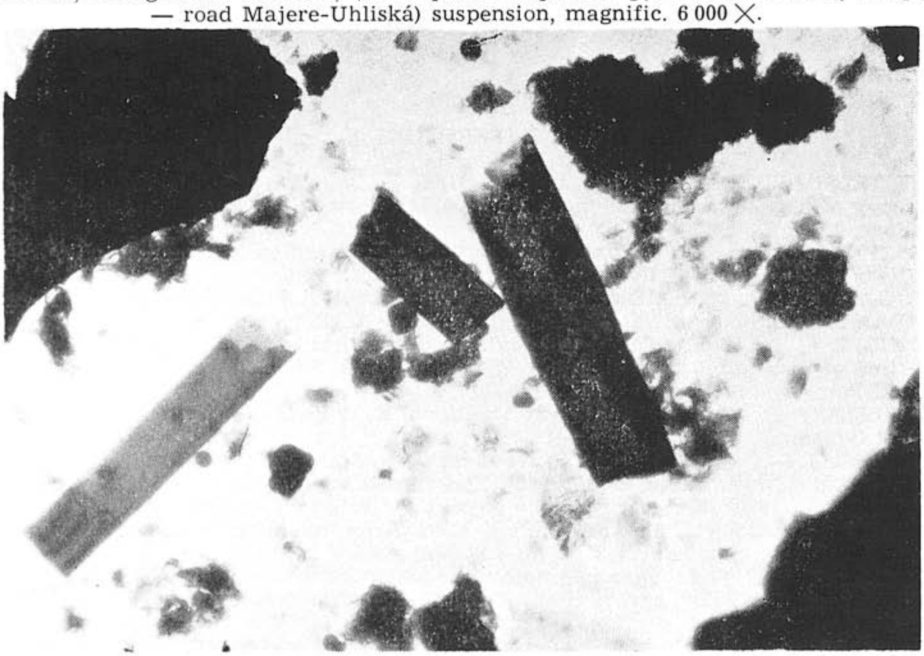

Fig. 9. Nontronite with the significant parallel splitting, dark particles of the partly decomposed parent minerals, (decomposed amphibole-pyroxenic andesite Teplá Voda) suspension, magnific. 8 000 ×.

tion) as well as organic acids originated from the oxidation of the organic substance present in the basin, produces pH = 4 and lower, makes nontronite structure very unstable. Decomposed nontronite produces Si- and Fe-rich phases, mostly amorphous, which have been found at the bottom of the basin (accumulations of Fe-amorphous oxides and diatoms).

Beidellite and other Fe-poor smectites occurring in the weathering products, were dissolved more slowly. They were, together with other relics of primary rocks, transported gradually to the basin and changed to the structurally various types of kaolinite minerals during diagenesis.

New knowledge of nontronitization of the volcanic andesite rocks in Pukanec region is in good accordance with the previous results on the alteration of pyroxene andesite in south-eastern part of Prešov-Tokaj Mts. (Forgáč—Číčel, 1965) and on the origin of nontronite-beidellite association in the region of Middle Slovakian volcanism (Kremnica hills) (Kraus—Polakovičová, 1967 and Kraus—Gerthofferová, 1968). Despite great similarities in the character of the parent rocks there are some differencies concerning SiO_2 admixtures and nontronite accumulation. There are no amorphous SiO_2 modifications (opal, chalcedone) in the weathered andesite rocks in Pukanec region. Weathering products contain the admixture of alpha-tridymite and alpha-cristobalite and it is supposed that part of it may be of secondary origin. Nontronite from Pukanec does not form nodules or veinlets, but it is dispersed in the whole volume of the parent rock.

Process of nontronitization in Pukanec region is not isolated but associated with the whole complex of the mineral alteration on the surface as well as with the succeeding alteration during diagenesis in the sedimentary basin.

Translated by V. Figusch

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