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THE OCCURRENCE OF ZEOLITES IN VARIEGATED DEPOSITS OF THE RYBNIK COAL BASIN {UPPER SILESIA}

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A b s t r a c t. The paper discusses the occurrences of analcime and harmotome within the zone of pre-Miocene weathering accompanied by underground fires of coal seams. The genesis of zeolites is to be associated with the epigenetic process of "burial diagenesis" under the conditions of high pH and high concentration of Si, K and Na ions. Clay rocks have been found to be most susceptible to zeolitization whilst rocks that have been subject to epigenetic silicification are zeolitized only in fracture zones and in zones of silica leaching.

INTRODUCTION

An analysis of all the available borehole profiles from the southern part of the Rybnik Coal Basin (Kowalski 1977, in press 1) has shown that a regionally developed zone of pre-Miocene weathering of the Carboniferous rock mass occurs in this area. It has also been observed in other outcrops of the Carboniferous rock mass (Alexandrowicz, Bogacz 1971; Alexandrowicz, Siedlecki 1961; Lipiarski 1969) under an overburden layer consisting of Permian, Triassic or Miocene rocks. In the Rybnik Coal Basin it is covered by Miocene marine sediments with the locally preserved relics of Triassic rocks that were not eroded before the Miocene. Weathering took place both before and after the Triassic, reaching the greatest intensity in the Upper Palaeozoic and Palaeogene. The weathering processes operated in Carboniferous sedimentary rocks, as well as in the local volcanite intrusions.

The regional weathering processes were locally accompanied by underground fires of coal seams in the zone of pre-Miocene outcrops, which expedited the oxidation of coal and caused the coal seams to disappear in this zone. The weathering-fire genesis of zones of disappearance of coal seams in the Ostrava – Karvina area (Czechoslovakia) has been advocated by Králik (pers. comm.), who found mullite there. In fire zones the structures of clay minerals were destroyed under the influence of elevated temperatures (Kowalski 1971, 1973, 1977, in press 2).

During weathering siderite, pyrite and marcasite passed to haematite and

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maghemite, and sulphuric acid liberated during the decomposition of sulphide minerals favoured local precipitation of gibbsite. Recrystallization of the clay material burnt out in the zone of outcrops, and maybe also the resilicification processes, led to the formation of hydrohalloysite and kaolinite.

The progressive concentration of silica in intergranular water of an initially low pH promoted resilicification of gibbsite and the removal of iron. The investigations show that hydrohalloysite was partly replaced by genetically younger smectite. The increase in silica content in the intergranular water led to partial silicification of the fossil weathering zone.

Partial silicification of the rocks studied, which were being gradually buried and covered by younger sediments during the formation of the Carpathian foredeep, was followed by alkalization of the environment of epigenetic alterations. This resulted initially in the replacement of hydrohalloysite and smectite by illite, and then in the crystallization of fissure illite in the silicified rocks. Illitization was accompanied by an increase in pH, which is reflected in the leaching of silicic rocks and the formation of cavities or caverns. In time the pH exceeded 9.0, which at the high concentration of SiO₂, led to the formation of zeolites. The processes similar to those described are defined by Richard (1978) as "burial diagenesis".

THE OCCURRENCE OF ZEOLITES IN VARIEGATED DEPOSITS

Among a variety of minerals occurring in the variegated deposits, analcime and harmotome, as well as small amounts of phillipsite and natrolite, have been found. Analcime was described from the Jastrzebie mine (Fig. 1, site 4), from the zone of the Orlowo – Michałkowice disturbance, where it appears in paragenesis with harmotome, montmorillonite and quartz (Kowalski 1977). A similar paragenesis, reported by Králik (1971), was investigated in samples collected together with the cited author from the Fučik 5 mine (Fig. 6) and from the zone of disappearance of the seam 504 in the Knurów mine near Gliwice (Kowalski 1977). All these localities belong to the same tectonic unit. Local occurrences of analcime have also been ascertained in the zones of disappearance of the seams 415/1 and 415/1 – 2 in the Moszczenica and Jastrzebie mines (Fig. 1, zones 5, 6).

Harmotome is the commonest zeolite in the variegated deposits. It may appear in the form of idiomorphic crystals white, honey-yellow, grey-yellow and orange-yellow in colour, elongated along the X-axis, frequently exhibiting cross-twinning. These crystals are developed in cavities within volcanic and silicic rocks subject to leaching. Harmotome veinlets crosscut the silicified rocks, also occurring within large recrystallized plates of epigenetic illite. This shows that harmotome is younger than the silicification process and also younger than illite. Harmotome was found in the zone of disappearance of the seam 415/1 in the Moszczenica mine, where it is locally accompanied by phillipsite (Kowalski 1977), as well as in the zone of disappearance of the seams 505/1, 505/2, 505/3 and 510 (Fig. 1, sites 2, 7). Harmotome and, locally, small amounts of natrolite were found in the zone of disappearance of the seam 504 in the Knurów mine. This is evidenced by the interplanar spacings 6.72 (1), 5.87 (1), 4.38 (3), 4.17 (2), 3.16 (1), 3.10 (1), 2.98 (1), 2.87 (1) and 2.79 Å (1), determined in one of the samples.

The present paper discusses new occurrences of analcime in the zone of disappearance of the seam 415/1-2 in the Moszczenica mine (Fig. 1, site 1) and the signs of zeolitization in the disappearance zones of the seams 505/1 (Fig. 1, site 2)

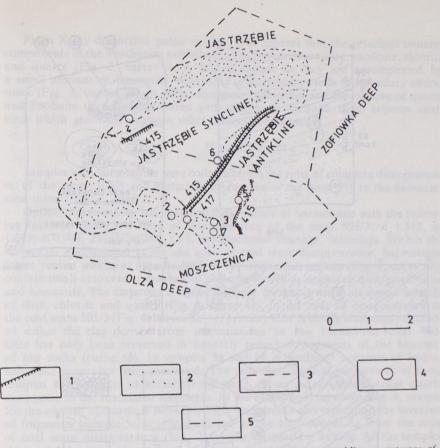


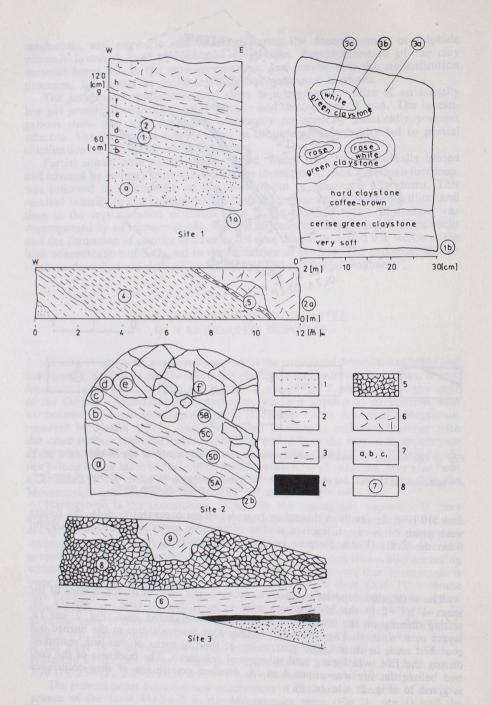
Fig. 1. The location of analcime and harmotome occurrences related to pre-Miocene outcrops of fossil weathering zones

I- strike and dip of some coal seams, 2- outcrops of fossil weathering zones, 3- boundaries of mine fileds, 4- sites with zeolites, 5- boundaries of mantles of waste relics

and 510 (Fig. 1, site 3) in this mine. In order to get comparative data, the results were given of X-ray diffraction studies of volcanic rocks subject to zeolitization from the Žofie (Fučik 5) mine in Czechoslovakia (Fig. 6).

Analcime

The variegated deposits in question occur in the zone of disappearance of the seam 415/1-2 in the Moszczenica mine (Fig. 1, site 1; (Fig. 2, site 1a, b), resting directly on the bottom sandstone of the eliminated seam (Fig. 2, site 1a, layers a, c, d, e, f). They are relics of clay rock intercalations in the burned-out coal bed and, in their upper part, relics of the top rocks of this bed, reworked during the fire, weathering and subsequent epigenesis. The thickness of the coal bed before the fire was about 4 m. A detailed description of this occurrence is given in (Fig. 2, site 1a, b).



From X-ray diffraction patterns (Fig. 3) it appears that the principal mineral components of the weathering sandstones and claystones are illite, kaolinite, chlorite and quartz (Fig. 3, curves 1, 2, 3a). In sample 3a they are accompanied by a small amount of haematite. Within the concretionary zones of secondary alterations (Fig. 3, curves 3b, c), illite disappears altogether and the content of quartz and kaolinite decreases compared with sample 3a. Instead, there appears analcime which gradually replaces other mineral components.

Harmotome

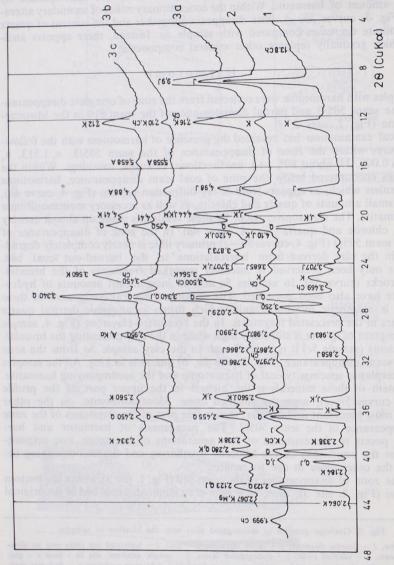
Samples with harmotome were collected from the zone of complete disappearance of the seam 505/3 and partial disappearance of the seam 510 in the Moszczenica mine (Fig. 2, sites 2, 3).

Optical examination has revealed the presence of harmotome with the following features within the zone of disappearance of the seam 505/3: n 1.513, n 1.505, $\Delta 0.008$, 2V about 80° , (+), unidirectional cleavage, twinnings. Within the claystones encountered below the zone of coal seam disappearance, harmotome forms venlets which, as appears from X-ray diffraction pattern (Fig. 4, curve 4b), contain small amounts of quartz and chlorite, as well as accessory montmorillonite and haematite. The claystone crosscur by these veinlets is made up almost entirely of illite, chlorite and quartz (Fig. 4, curve 4a). In the zone of disappearance of the coal seam 505/3 (Fig. 4, curves 5a-c) primary illite is nearly completely degraded within the clay derived from intercalations in the burned-out coal bed. Illite has only been preserved in intensely reworked fragments of the brecciated top rocks (curve 5b). In samples 5a and 5b insignificant amounts of hydrohalloysite have also been preserved. The dominant mineral component of these samples is epigenetic montmorillonite which forms as allogenic detrital quartz disappears in the brecciated fragments. In the brecciated claystone (Fig. 4, sample 5b) the content of quartz is still fairly high while in the clay cementing the brecciated fragments (sample 5c) it is insignificant. In the clay sample 5a from the zone of coal seam disappearance (Fig. 2, site 2a, b) quartz is lacking. All the samples show interplanar spacings typical of harmotome and the accompanying haematite. The content of these minerals is the highest in the upper part of the profile (Fig. 4, curves 5b, c), near the silicified zone. Montmorillonite, on the other hand, is most abundant in the lower, quartz-free part of the claystones of the zone of disappearance of the seam 505/3. The paragenesis of haematite and harmotome points to the existence of two generations of haematite, one originating under the conditions of pre-Miocene weathering and the accompanying fires, and the other close in age to zeolites.

In the zone of disappearance of the seam 510 (Fig. 1, site 3), above the bottom sandstone (Fig. 2, site 3), there is a relic of an eliminated coal bed of the original

Fig. 2. Geologic profiles of investigated sites with the location of samples

I - sandstone, 2 - aleuritic claystone, 3 - pelitic claystone, 4 - coal, 5 - brecciated top rocks over an eliminated coal seam, 6 - silicified rocks, 7 - distinguished layers, 8 - sample numbers; site Ia - layer a - grey sandstone, b - clay of epigenetic origin, c - green-grey sandstone, d - light-green sandstone, e - dark-grey sandstone, f - sandstone with haematite veinlets, g - green-cherry-red soft clay, h - coffee-coloured top claystone of the eliminated coal seam 415/1-2. The Moszczenica mine; site Ib - fragment of the layer g (as above), 3a - fairly soft, friable pelitic rock with an oriented structure, 3b - concentric zone of green claystone, 3c - concentric zone of white colour, sometimes with a red core; site 2a - sample 4 - claystone with veinlets; site 2b - layer a - soft cherry-red claystone, b - soft pale green and white claystone, c - hard dark-green claystone, d - cherry-red clay, e - fragments of brecciated claystone, f - rocks subject to silicification in the top of the eliminated coal seam 505/3; site 3 - zone of partial disappearance of the seam 510 m in the Moszczenica Mine



Moszczenica the in seam Ha - 1Q - qzone of disappearance of the coal h – hydrohalloysite, H – haematite, manganese calcite, P – plagioclase, samples from the diffraction patterns of X-ray cme, Au ... M - 1

Fig. 4. X-ray diffraction patterns of samples from the zone of disappearance of the coal seam 505/3 in the Mosz-czenica mine. Symbols as in Fig. 3. D_{MI} values in Å

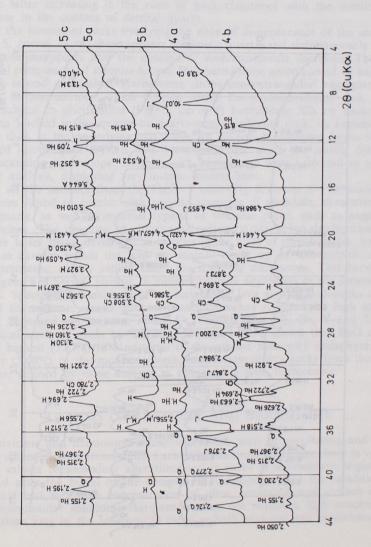
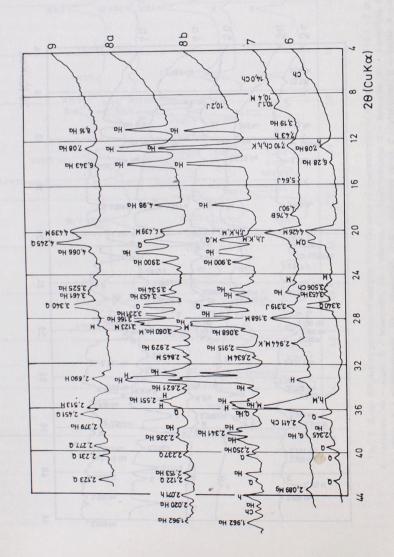


Fig. 5. X-ray diffraction patterns of samples, from the zone of disappearance of the coal seam 510 in the Moszczenica mica mine. Symbols as in Fig. 3. Dz., values in A



thickness of about 9 m. In the area under study it was burnt out during an underground fire. The relict fragment of the bed is overlain by clays showing an oriented structure and by brecciated top rocks that have been partly subject to epigenetic homogenization in the roof of the mine working.

X-ray diffraction studies (Fig. 5) have revealed that the dark laminated rocks in the top of the relict coal bed (curve 6) contain allogenic detrital quartz as well as montmorillonite, hydrohalloysite and chlorite as dominant mineral components whereas harmotome occurs sporadically. Optical examination has confirmed the presence of orange-yellow harmotome in the rock fissures. Within the superjacent pink claystones (sample 7) quartz is lacking, having been removed from this zone, and the pigment of clay rock is made up of haematite and maghemite. Both varieties of claystones contain hydrohalloysite and montmorillonite, the content of the latter increasing in the zone of pink claystones with the simultaneous

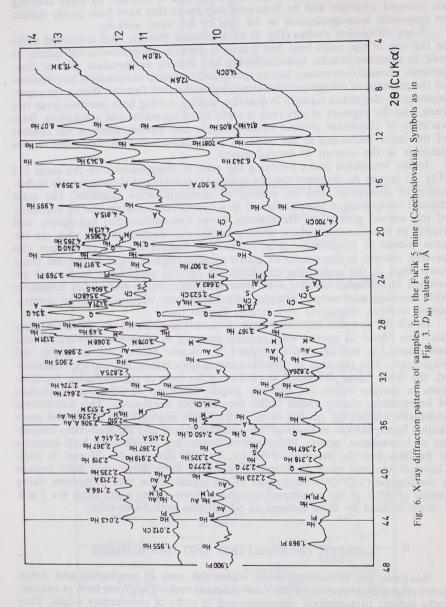
decrease in the content of detrital quartz.

In the brecciated rocks overlying the zone of disappearance of the seam 510 (Fig. 5, curves 8a, b) there appears epigenetic quartz, and simultaneously hydrohalloysite disappears and the content of montmorillonite decreases. The main mineral component of samples 8a and 8b is harmotome accompanied by haematite of the second generation. Harmotome forms concentric-radial aggregates, well visible in thin sections, which develop within the montmorillonite rock. In the zone subject to silicification (Fig. 5, curve 9) harmotome fills up fissures and caverns leached in the quartz or quartz-montmorillonite rock, its content being lower than in the underlying claystones. In caverns harmotome forms well developed idiomorphic crystals with cross-twinnings, which are readily detectable with scanning microscope. Samples with harmotome were subjected to spectroscopic analysis, displaying very intense barium lines.

In all the samples from the Fučik 5 mine (Fig. 6, curves 10-14) harmotome has been found to coexist with varying amounts of analcime, montmorillonite and quartz, as well as minerals typical of volcanic rocks, such as augite and plagioclase. The presence of chlorite and epigenetic siderite has also been ascertained, the latter being developed in cavities in the form of rhombohedra. Incrustations in caverns within the lava are mainly made up of idiomorphic crystals of harmotome, clearly visible under the scanning microscope. This harmotome undergoes weathering, passing to montmorillonite aggregates of the second generation. Spectroscopic analysis of the melaphyric lava and the wall-rocks consisting of harmotome, quartz and montmorillonite has revealed the high intensity of Ba lines and the presence of Sr, Cr, Cu, Pb, Mn, Ni, Ga, V and Ti lines. Chemical analyses made by Dopita and Králik have shown that BaO content is the highest (5.87%) in sandstones of the contact zone with melaphyre, being similar (5.09%) in the montmorillonite-harmotome-quartz rocks from the Fučik 5 mine, defined by these authors as porcelanites with harmotome.

GENESIS OF ANALCIME AND HARMOTOME

Analcime and harmotome occur within the zone of weathering and subsequent epigenetic alterations of the Carboniferous rocks. They form both in volcanic rocks subject to secondary alterations and in rocks of sedimentary origin. They formed after the burial of the complex in question, under the conditions of high pH and at a high concentration of silica, potassium and sodium ions in the intergranular solutions. Harmotome does not require a high concentration of barium ions in the solutions to form, as both analcime and harmotome form



at a low concentration of these ions in the solutions. Zeolitization proceeds in zones of complete elimination of coal seams (Fig. 1, site 2). Where a relic of a seam remains (Fig. 1, site 3), zeolitization does not operate in claystones of the zone of direct contact with coal but moves upwards into the zone of brecciated rocks. The high pH favoured the precipitation not only of harmotome but also of fissure haematite. In silicified rocks the content of harmotome decreases. While in clay rocks it replaces metasomatically the groundmass, in silicified zones it only fills up fissures and caverns in the silicic rocks being leached (Fig. 2, sample 9; Fig. 5, sample 9). This indicates that harmotome is younger than the silicification process and that extreme alkalization of the environment of epigenetic alterations followed silicification.

Since analcime and harmotome crystallize from solutions of a high pH (more than 9.0), the acid environment of the direct coal seam contact favours in this zone the preservation of the genetically older products of aggradational transformation of minerals of the weathering and fire zone, characteristic of en-

vironments with a low pH.

Silicification processes, which generally operate at some distance from coal beds, are older than zeolitization. They cause clay minerals that readily convert to zeolites to be replaced by quartz which is unsusceptible to structural transformation. Therefore, zeolites form throughout smectite rocks whereas in silicified rocks, only within fissures and caverns.

Zeolites encountered in the discussed rocks may be Miocene or post-Miocene in age and seem to be coeval with the rock salts of Zory and Rybnik. According to Peterson *et al.* (1970), phillipsite does not already occur in pre-Miocene sediments, and the presence of this mineral has been ascertained in the Moszczenica mine. The present author is in favour of the hypothesis of "burial_diagenesis" advanced by Richard (1978).

Translated by H. Kisielewska

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WYSTĘPOWANIE ZEOLITÓW W PSTRYCH UTWORACH RYBNICKIEGO OKRĘGU WĘGLOWEGO (GÓRNY ŚLĄSK)

Streszczenie

W pracy opisano wystąpienia analcymu i harmotomu w obrębie strefy przedmioceńskiego wietrzenia połączonego z podziemnymi pożarami pokładów węgla. Genezę zeolitów wiąże się z epigenetycznym procesem "diagenezy pogrążenia" w warunkach wysokiego pH i wysokiego stężenia jonów Si, K, Na. Najbardziej podatne na zeolityzację są skały ilaste, natomiast skały poddane epigenetycznej sylifikacji są zeolityzowane jedynie w strefach spękań i ługowania krzemionki.

OBJAŚNIENIA FIGUR

- Fig. 1. Rozmieszczenie badanych wystąpień analcymu i harmotomu na tle podmioceńskich wychodni reliktów kopalnych stref wietrzenia.

 1 bieg i upad pokładów węgla, 2 wychodnie kopalnych stref wietrzenia, 3 granice pól kopalnianych, 4 stanowiska zeolitów, 5 granice reliktów pokryw wietrzeniowych
- Fig. 2. Profile geologiczne stanowisk badań szczegółowych z zaznaczeniem położenia próbek I piaskowiec 2 iłowiec aleurytowy, 3 iłowiec pelitowy, 4 wegiel, 5 zbrekcjowane skały stropowe nad zanikającym pokładem wegla, 6 skały poddane sylifikacji, 7 wyróżnione warstwy, 8 numery próbek; site Ia warstwa a szary piaskowiec, b ił o epigenetycznej genezie, c szarozielony piaskowiec, d jasnozielony piaskowiec, e ciemnoszary piaskowiec, f piaskowiec z żyłkami hematytu, g wiśniowozielony miękki ił, h kawowy iłowiec stropowy zanikłego pokładu wegla 415/1 2. Kopalnia Moszczenica; site 1b fragment warstwy g(j.w.), 3a skała pelitowa, o teksturze kierunkowej, krucha, dosyć miękka, 3b koncentryczna strefa iłowca zielonego, 3c koncentryczna strefa barwy białej, niekiedy z czerwonym rdzeniem: site 2a próbka 4 iłowce z żyłkami; site 2b warstwa a miękkie iłowce wiśniowe, b miękkie iłowce bladozielone i białe, c twarde ciemnozielone iłowce, d wiśniowy ił, e fragmenty zbrekcjowanego iłowca, f skały poddane sylifikacji w stropie zanikłego pokładu 505/3; site 3 strefa częściowego zaniku pokładu 510 w kopalni Moszczenica
- Fig. 3. Dyfraktogramy próbek ze strefy zaniku pokładu 415/1-2 w kopalni Moszczenica
 A analcym, Au augit, B brammalit, Ch chloryt, h hydrohaloizyt, H hematyt, Ha harmotom, J illit, K kaolinit, Ka kalcyt, M montmorillonit, Mg maghemit, MnK kalcyt manganowy, P plagioklaz, Q kwarc, S syderyt
- Fig. 4. Dyfraktogramy próbek ze strefy zaniku pokładu 505/3 w kopalni Moszczenica. Objaśnienia jak do fig. 3
- Fig. 5. Dyfraktogramy próbek ze strefy zaniku pokładu 510 w kopalni Moszczenica, Objaśnienia jak do fig. 3.
- Fig. 6. Dyfraktogramy próbek z kopalni Fučik 5 (CSSR). Objaśnienia jak do fig. 3

Вачлав Марян КОВАЛЬСКИ

РАСПРОСТРАНЕНИЕ ЦЕОЛИТОВ В ПЕСТРЫХ ПОРОДАХ РЫБНИКСКОГО УГОЛЬНОГО ОКРУГА (ВЕРХНЯЯ СИЛЕЗИЯ)

Резюме

В работе описано распространение анальцима и гармотома в зоне домиоценового выветривания, сопровождавшегося подземными пожарами угольных пластов. Образование цеолитов относится к эпигенетическому процессу "диагенеза погружения" в условиях высокого рН и сильной концентрации ионов Si, K, Na. Наиболее благоприятны к цеолитизации глинистые породы, а породы, подвергшиеся эпигенетическому силицифицированию, цеолитизированы лишь в зонах трещиноватости и выщелачивания кремнезема.

ОБЪЯСНЕНИЯ К ФИГУРАМ

- Фиг. 1. Распространение исследованных проявлений анальцима и гармотома на фоне подмиоценовых выходов остатков ископаемой коры выветривания 1 простирание и падение угольных пластов, 2 выходы ископаемой коры выветривания, 3 контуры шахтных полей, 4 проявления цеолитов, 5 границы распространия остатков коры выветривания
- Фиг. 2. Геологические профили точек детального исследования с отмеченными местами опробования

 1 песчаник, 2 алевритовый аргиллит, 3 пелитический аргиллит, 4 уголь, 5 брекчированные породы, перекрывающие выклинивающийся угольный пласт, 6 силицифицированные породы, 7 выделенные слои, 8 нонера образцов; нестонахождение 1a: слой a серый песчаник
 - породы, 7 выделенные слои, 8 номера образцов; местонахождение 10: слой а серый песчаник b глина эпигенетического происхождения, c серовато-зеленый песчаник, d светлозеленый песчаник, e темносерый песчаник, f песчаник c гематитовыми прожилками, g вишнево-зеленая мягкая глина, h темнокоричневый аргиллит, перекрывающий выклинивающийся угольный пласт 415/1—2 (шахта Мощеница); местонахождение 1b: часть слоя g (см. выше), 3a пеличческая порода c орментированной текстурой, рыхлая, довольно мягкая, 3b концентрическая зона зеленого аргиллита, 3c концентрическая зона белого цвета, местами c красным центром; местонахождение 2c: слой a нягкие вишневые аргиллиты, b мягкие светлозеленые и белые аргиллиты, c твердые темнозеленые аргиллиты, d вишневая глина, e глыбы брекчированного аргиллита, f силицифицированные породы, перекрывающие выклинивающийся пласт 505/3; местонахождение 3c: зона частичного выклинивания пласта 510 в шахте Мощеница
- Фиг. 3. Диффрактограммы образцов из зоны выклинивания пласта 415/1—2 месторождения Мощеница А—анальцин, Аи—авгит, В—браммалит, Сh—хлорит, h—гидрогаллуазит, Н—гематит, На—гармотом, l—иллит, К—каолинит, Ка—кальцит, М—нонтмориллонит, Мд—магхемит, МпК—марганцевый кальцит, Р—плагиоклаз. Q—кварц, S—сидерит
- Фиг. 4. Диффрактограммы образцов из зоны выклинивания пласта 505/3 месторождения Мощеница. Объяснения как к фигуре 3
- Фиг. 5. Диффрактограммы образцов из зоны выклинивания пласта 510 месторождения Мощеница. Объяснения как к фигуре 3
- Фиг. 6. Диффрактограммы образцов из шахты фучик 5 (ЧССР). Объяснения как к фигуре 3