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THAUMASITE — A RARE MINERAL CONTAINING $Si(OH)_6^{2-}$ GROUPS FROM DUBIE (THE DĘBNIK ANTICLINE, S POLAND)

Abstact. Detailed mineralogical investigation of exo- and endocontact rocks formed due to the emplacement of late Palaeozoic rhyodacite laccolith in carbonate sedimentary rocks, drilled in the Dębnik anticline near Dubie, revealed the presence of thaumasite $Ca_3[Si(OH)_6 \cdot 12H_2O](CO_3)(SO_4)$. The thaumasite veins cut the rocks of the proximal zone of the highest temperature metamorphism, which extends few tens of meters within exocontact rocks (marbles) and one meter within rhyodacite. Apparently, the thaumasite formed as result of retrogression reactions from minerals contrived at the peak of metamorphism. High temperature calcium silicates reacted, at low temperatures, with basic waters containing carbonate and sulphate ions, allowing the changes of silicon coordination from Si^{IV} to Si^{VI}. The thaumasite, calcite, aragonite and sepiolite veins belong to the last mineral generations formed in the Dębnik anticline. Thaumasite forms mineral paragenesis with calcite. To the best of our knowledge this is the first finding of this unique mineral in Poland.

Key-words: thaumasite, IR spectrum, contact metamorphism, retrogression, Dębnik anticline

INTRODUCTION

At the Dębnik anticline (Fig. 1) the late Palaeozoic rhyodacite intrusion (Muszyński, Pieczka 1994) formed a 300 m thick contact aureole in the Devonian carbonate rocks (Narkiewicz 1983). Contact metamorphism of dolomites led mainly to the formation of calcite-brucite marbles (Harańczyk, Wala 1989; Lewandowska 1989, 1998). The dolomites primarily contained a few intercalations enriched in siliceous, probably terrigenic material. In these siliceous zones neomineralization of lizardite, chrysotile (Lewandowska 1991), hydrotalcite (Muszyński, Wyszomirski 1998), grossular (Lewandowska, Rospondek 2000), and sepiolite (Lewandowska 1996) was observed. Rhyodacite itself is affected by the contact alterations only within a thin outermost zone where primary quartz-plagioclase-feldspar matrix was replaced by minerals of analcite group (Lewandowska et al. 2000). These exo- and endocontact rocks were cut by white,

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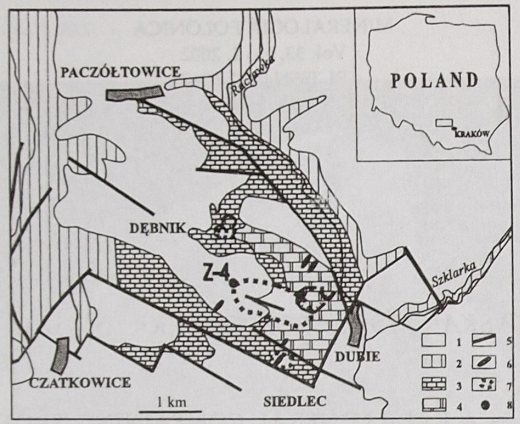

Fig. 1. Geologic map of the Dębnik anticline
1 — Jurassic limestones and sandstones, 2 — Carboniferous limestones, 3 — Devonian limestones, 4 — Devonian dolomites, 5 — faults, 6 — volcanic rocks, 7 — alteration zones, 8 — borehole (according to Kozłowski 1955 and Łaptaś 1982, modified)

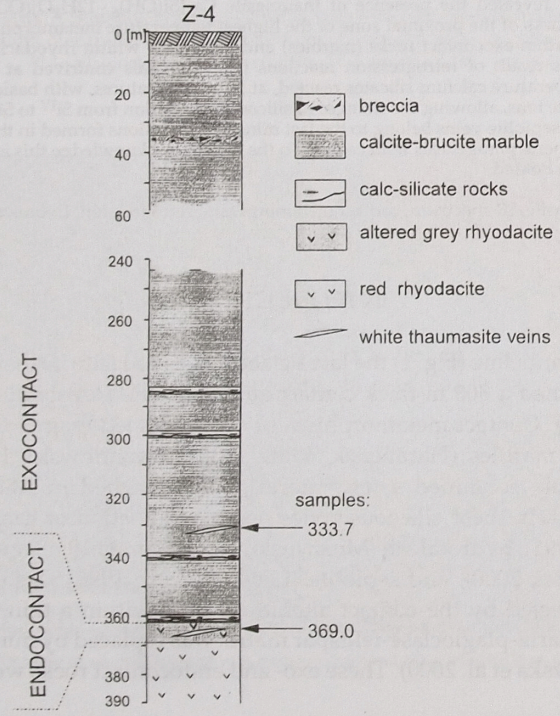

Fig. 2. Lithologic column of the Z-4 borehole with the localization of thaumasite veins (based on the description by Boleński 1981)

thin veins, filled with fine crystalline mineral. From these veins, samples were selected (Fig. 2) for detailed mineralogical investigations, resulting in the first description of thaumasite in Poland.

SAMPLES AND METHODS

Samples were collected from the Z-4 borehole core. The borehole was drilled between Dębnik and Dubie villages near Kraków (Fig. 1). At the depths of 333.7 m and 369.0 m the rocks were cut by veins, from which mineral samples were handpicked for further investigations (Fig. 2). The samples come from the veins cutting the exo- (333.7 m) and endocontact rocks (369.0 m), respectively.

The samples were analysed by powder X-ray diffractometry (XRD) using a horizontal goniometer (HZG-4B), Cu-Kα radiation and Ni-filter. The observation of crystals' habit and elemental composition was performed with a JEOL 5410 scanning electron microscope (SEM) coupled with an energy dispersive spectrometer (EDS) EDAX V-9800. Infrared absorption spectra (FTIR) were obtained at ambient temperatures with 2.0 cm^{-1} resolution in a range of 400–4000 cm^{-1}, using a BIO-RAD Fourier Transform Spectrometer (FTS 135) equipped with a Michelson interferometer. The air-dried, powdered samples were vacuum-pressed into KBr discs.

RESULTS

White, small crystals found are visible in rare veins (1–2 mm thick) at the depth 333.7 m and 369.0 m. In the sample from 333.7 m, i.e. 36 m away from the contact, the vein, parallel to the axe of the core, is hosted by calcite--brucite marble (predazzite) over the length of 20 cm. In this sample 1 cm wide nest was found (Phot. 1). In the sample from 369.0 m, collected about 10 cm down from the contact, another vein is hosted by the altered rhyodacite.

The veins are filled with white crystals with vitreous luster. The crystals are up to 0.1 mm long and up 0.03 mm wide. Their habit is well visible in scanning electron microscope images (Phot. 2 and 3). The hexagonal prismatic crystals are doubly terminated with partly developed hexagonal pyramids in the sample from the exocontact void (Fig. 3). The pyramids are better developed for the large specimens, while smaller are entirely prismatic.

In thin section the mineral is colourless. Cleavage was not observed. Crystals are often automorphic, uniaxial (at Phot. 4 some crystals were cut perpendicularly

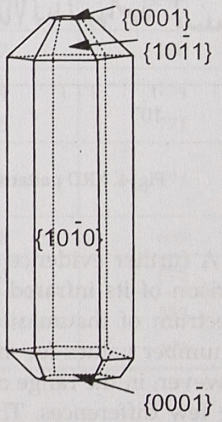

Fig. 3. Habitus of the thaumasite crystals separated from voids (sample from 333.7 m)

to z-axis and therefore appear isotropic) and negative. They exhibit negative relief against the mounting medium (n ~1.54). The described geometrical and optical features are consistent with those of thaumasite. Thaumasite refractive indices are $n_\varepsilon = 1.468$ and $n_\omega = 1.506 \pm 0.002$, and the birefringence $\Delta = 0.04$ (e.g. Carpenter 1963). The birefringence herein observed was in this range.

This identification was positively confirmed by the XRD analysis. X-ray diffraction pattern (Fig. 4) is analogous to that of the specimen from Ballyalton, Northern Ireland (card 23–0128 JCPDS), with the main reflections at 9.55 Å (100), 5.51 Å (40) and 3.41 Å (20). Additionally, a small admixture of calcite was detected.

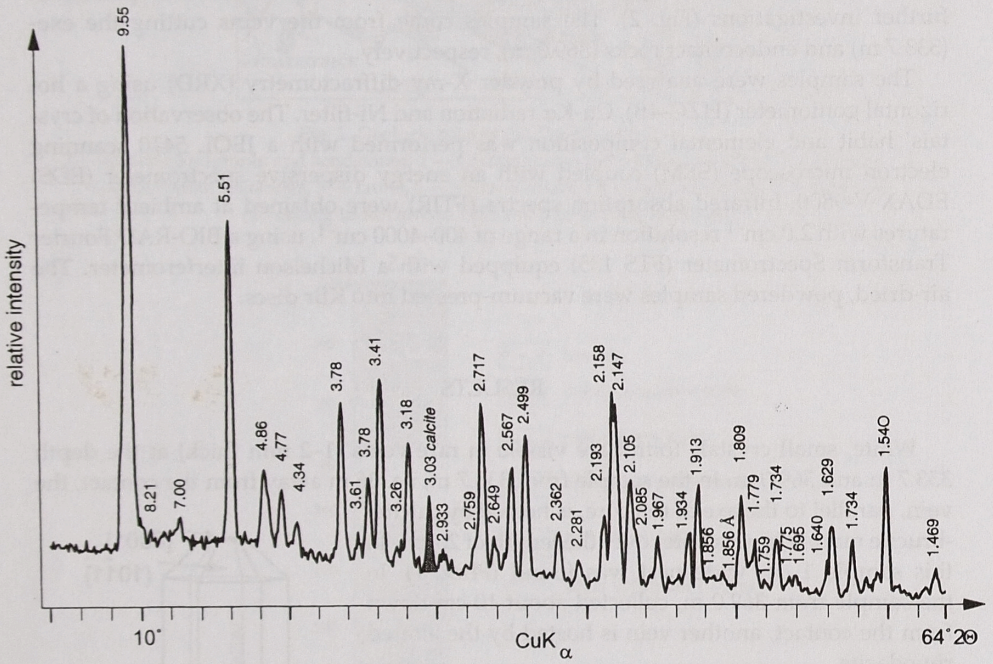

Fig. 4. XRD pattern of the thaumasite, numbers denote d_{hkl} [Å] (sample from 333.7 m)

A further evidence for thaumasite identification in Dubie comes from the comparison of its infrared absorption spectrum (Table 1 and Fig. 5) with the reference spectrum of thaumasite from Paterson, New Jersey, USA (Moenke 1966). The wavenumber values for most of the absorption bands are in good agreement (Table 1), however, in the range of the O-H bond stretching frequencies (3600–3400 cm^{-1}), there are few differences. The band at 3560 cm^{-1}, described by Moenke (1966), was not observed in the Dubie thaumasite here (Table 1 and Fig. 5). This difference can be related to the presence of ettringite in the reference material due to the existence of solid solution between these minerals (Kollmann, Strübel 1981). For mixed thaumasite-

Position of the IR absorption bands in the Dubie thaumasite and in the reference minerals (in brackets)

Localization	Dubie (Z-4 depth 333.7 m)	Dubie (Z-4 depth 369.0 m)	Paterson, New Jersey, USA*	Paterson, New Jersey, USA***
	Absorption bands			
Assignment**	[cm^{-1}]			
ν (O-H)	not observed	not observed	3560	not observed
ν (O-H)	3502 3473	3503 3470	3500 3470	3503, 3470
ν (O-H)	3427 3405	3430 3404	3420	3427 3403
ν (O-H)	3240	3235	3240**	3230
ν (O-H)	3050	3038	not mentioned	3040
ν (O-H) ?	2860	2855	not mentioned	2850
δ H-O-H	1700	1696	1705	1696
δ H-O-H	1647	1647	1660	1648
ν (a) CO_3	1398	1398	1400	1396
ν (a) SO_4	1100	1101	1102	1100
ν (a) SO_4	1070	1075	1075	1070
ν Si-O SiO_4	907, 937 (weak)	934	935	none
δ CO_3	883	877	887	886
ν Si-O in $Si(OH)_6$	749 771	747 765	750 765	746 771
ν Si^{VI}-O in $Si(OH)_6$	673	669	673	674
δ SO_4	638	639	638	639
δ SO_4	592	591	592	590
δ Si-O in $Si(OH)_6$	550 501	500	555 500	553 499

* Moenke (1964, 1966).

** Varma, Bensted (1973) and Bensted, Varma (1974).

*** Thaumasite from the collection of Mineralogical Museum, Institute of Geological Sciences, Jagiellonian University.

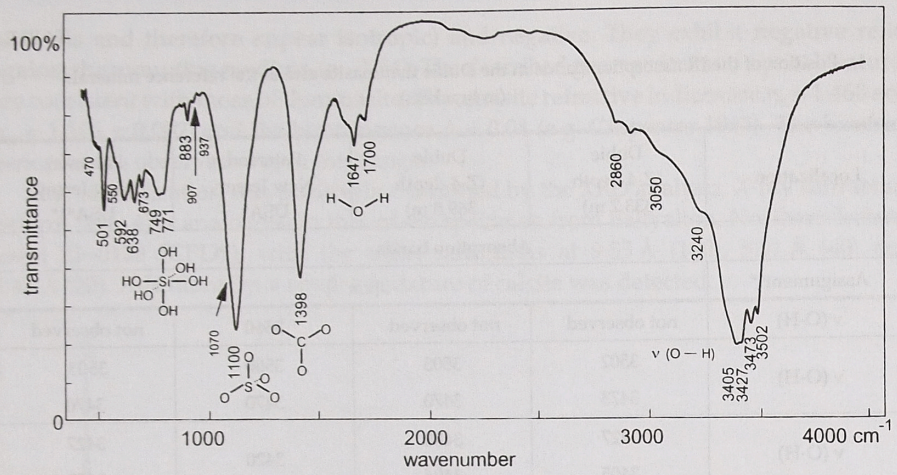

Fig. 5. Infrared (FT-IR) absorption spectrum of the thaumasite (sample from 333.7 m)

-ettringite crystals such an absorption band at ca. 3600 cm^{-1} was observed indeed, whereas pure thaumasite had no such a band (Kollmann, Strübel 1981). This band becomes prominent in the spectrum of ettringite. Such a band is also noted in hydrated calcium silicates. Unfortunately the data on the composition of the reference thaumasite (Moenke 1966) are not available. In addition to these bands there are bands at ca. 2860, 3050, 3240 cm^{-1} repeatedly observable for the Dubie thaumasite. These bands were also recorded for a specimen from Paterson, New Jersey, from the institute museal collection. This range is usually omitted in the data published, however some of these bands (specially at ca. 3250 cm^{-1}) are easily recognizable at the published spectra (e.g. Bensted, Varma 1974; Kollmann, Strübel 1981).

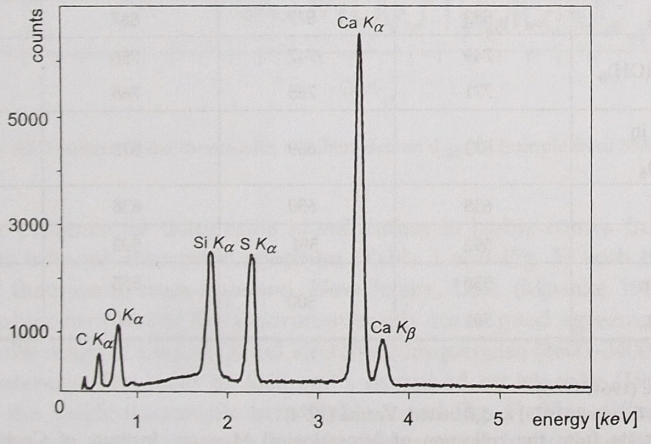

Fig. 6. EDS spectrum of the thaumasite (sample from 333.7 m)

The spectrum of thaumasite in the data base (Win IR Search provided with the BIO-RAD IR spectrometers) is inconsistent neither with those published by Moenke (1966) and Kollmann and Strübel (1981) nor with the data obtained herein.

The elemental analysis (EDS) showed the presence of Si, S and Ca (Fig. 6) confirming that the mineral does not form solid solution with ettringite.

DISCUSSION

Thaumasite is a hydrated calcium silanol-carbonate-sulphate mineral. Its name is derived from the Greek "*thaumasion*" — surprising (Knill, Young 1960). Surprisingly, it is one of a few known substances where Si coordinates six hydroxyl groups. Due to its features Strunz (1978) classified thaumasite into a separate class of nesosilicates. Thaumasite crystal structure was determined by Edge and Taylor (1969, 1971) revealing that it contains silanol groups $Si(OH)_6^{2-}$. However, even before the thaumasite X-ray structure determinations had been accomplished (Edge, Taylor 1969; 1971), Moenke (1964) concluded from the comparison of the stishovite SiO_2 and thaumasite infrared spectra that the Si was six-coordinated. This coordination records in the IR spectra as absorption bands at 501, 673 cm^{-1} resulting from Si^{VI}-O bending and a broad band at 749–771 cm^{-1} from Si^{VI}-O stretching (Bensted, Varma 1974) for the Dubie thaumasite. Furthermore, presence of an extremely tiny amount of four-coordinated silica (Si^{IV}-O) is recorded as a weak band at 937 cm^{-1} (Table 1) (Varma, Bensted 1974). This is apparent that the Dubie thaumasite is very pure phase.

Thaumasite consists of $Ca[(H_2O)_4(OH)_4]^{2-}$ and $Si(OH)_6^{2-}$ polyhedra, sharing common edges, running parallel to the z-axis, which are connected to sulphate and carbonate groups by hydrogen bonding (Effenberger et al. 1983; Jacobsen et al. 1996). Probably due to this bonding sulphate and, specially, carbonate stretching vibration are shifted towards relatively low frequency ranges. The water particles of $Ca[(H_2O)_4(OH)_4]^{2-}$ polyhedra are unequal, one of the four has planar threefold coordination of oxygen surrounded by Ca and two H atoms, whereas three other have oxygen tetrahedrally coordinated with Ca, two H of water and another from hydrogen bond (Effenberger et al. 1983). Due to the existance of strong hydrogen bonding of the water molecules, the (H-O) stretching vibrations are shifted towards relatively low frequency ranges. These are recorded as bands at ca. 2860, 3050, 3240 cm^{-1}. The differences in structural position of water must have resulted in the split of the water bending vibrations observed in a range of 1650–1700 cm^{-1}.

Due to the structural features of thaumasite, the chemical formula is often written: $Ca_3[Si(OH)_6 \cdot 12H_2O](CO_3)(SO_4)$ (Effenberger et al. 1983). The mineral is hexagonal belonging to the space group P6/m (hexagonal dipyramids).

Thaumasite is a rare mineral noted from sulphide deposits and skarns. In Sweden it is known from thermally metamorphosed carbonates hosting sulphides in Långban, Skottvångs and Viscaria (Knill, Young 1960). In similar rocks, it was found in England (High Sedling Mine, Weardale, County Durham, and in Shap, Cumbria) (Young et al. 1998) as well as in the Baikal region of Russia [in veinlets of the wollastonite-garnet

skarn hosting sulphides (Krivolutskaya 1984)]. Again in skarns (spurrite-merwinite-gehlenite) oriented overgrowth of thaumasite on ettringite was reported from Crestmore in California (Carpenter 1963). Additionally, an existence of the solid solutions between thaumasite-ettringite were described (Kollmann et al. 1977; Kollmann, Strübel 1981). Thaumasite is also known from veins in volcanic and volcanoclastic sediments affected by volcanic intrusions at the Pacific floor, e.g. Lau Basin at the Tonga fore-arc basin (Blanc et al. 1995, Schöps, Herzig 1994), tuff from Mariana Basin (Karpoff et al. 1992) and Mururoa Atoll, French Polynesia (Noack 1983).

The thaumasite occurrence in Dubie can be assigned to the skarn type, which is also commonly named sulphide deposit type. The thaumasite occurs in veins cutting exo- and endocontact rocks. This type of vein occurrence is limited only to the zone of the highest temperature alterations, which extends to a few tenths of meters within marbles and up to one meter within rhyodacite. Apparently, the thaumasite formed as a result of retrogression reactions from minerals formed at the peak of metamorphism, probably such as calcium silicates. These, high temperature stable, calcium minerals reacted later with waters containing carbonate and sulphate ions at low temperatures, allowing change of silicon coordination from IV to VI in the basic environment. The sulphate ion, which is necessary in low concentrations below ranges of gypsum stability, may be derived from oxidation of sulphides. The temperature of thaumasite stability, i.e. temperature at which dehydration would not have occurred at geologicaly relevant time, was estimated at <60°C based on the dehydration experiment of thaumasite at ambient pressure (Giampaolo 1986). At higher temperatures thaumasite decomposes to calcite and gypsum (Giampaolo 1986). This is further supported by the empirical observation of concrete structures where thaumasite forms only as a result of deleterious sulphate attack on buried concrete (e.g. Lukas 1975) at low temperatures. Its formation in laboratory experiments was generally observed below 15°C, however the thaumasite synthesis was usually conducted at 5°C (Kollmann, Strübel 1981; Aguilera et al. 2001 and references cited therein). Furthermore, in concrete the deleterious formation of thaumasite is promoted by the presence of dolomite aggregates (Gouda et al. 1975). This all is in accordance with the Dubie thaumasite occurrence in veins representing the last, low temperature stages of alterations. The last mineral generation beside thaumasite comprises sepiolite (Lewandowska 1996) and aragonite (Lewandowską 1989). Thaumasite forms mineral paragenesis with calcite in the exocontact rocks. To the best of our knowledge, it is the first record of this mineral in Poland.

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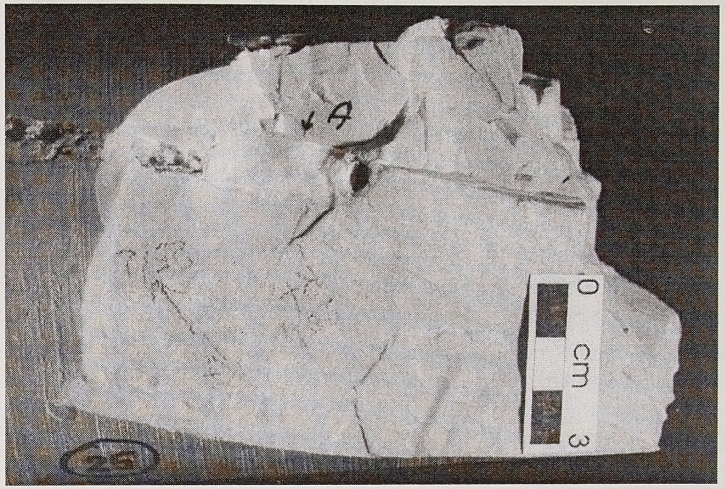
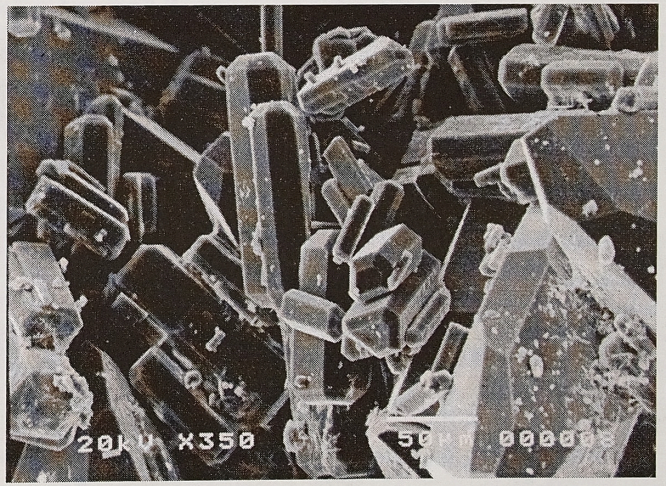
THAUMASYT — RZADKI MINERAŁ ZAWIERAJĄCY GRUPY $Si(OH)_6^{2-}$ Z DUBIA (ANTYKLINA DĘBNIKA, S POLSKA)

Streszczenie

Szczegółowe badania mineralogiczne skał egzo- i endokontaktowych, powstałych w wyniku oddziaływania późnopaleozoicznej intruzji ryodacytowej na dewońskie skały węglanowe, doprowadziły do identyfikacji thaumasytu $Ca_3[Si(OH)_6{\cdot}12H_2O](CO_3)(SO_4)$ występującego w tnących te skały żyłach. Żyły thaumasytu przecinają utwory strefy najwyżej temperaturowych przeobrażeń, która to strefa obejmuje kilkadziesiąt metrów aureoli egzokontaktowej oraz około jednego metra skał endokontaktowych. Thaumasyt powstawał w procesach retrogresywnych przeobrażeń, kosztem minerałów uformowanych w temperaturowym piku metamorfizmu. Wysokotemperaturowe krzemiany wapnia reagowały w niskich temperaturach z zasadowymi wodami zasobnymi w jony wapnia oraz jony siarczanowe, umożliwiając zmianę koordynacji krzemionki z Si^{IV} na Si^{VI}. Thaumasyt w paragenezie z kalcytem, oraz aragonit i sepiolit tworzą ostatnie generacje minerałów żyłowych w skałach przeobrażonych antykliny Dębnika. Niniejsza praca dokumentuje pierwsze stanowisko występowania tego unikalnego minerału w próbkach geologicznych w Polsce.



Phot. 1. Veins and voids hosting thaumasite in the calcite-brucite marble (the Z-4 core from 333.7 m)



Phot. 2. Authomorphic thaumasite (ca. 100–200 μm) in paragenesis with calcite from the exocontact rocks (sample from 333.7 m) — SEM image

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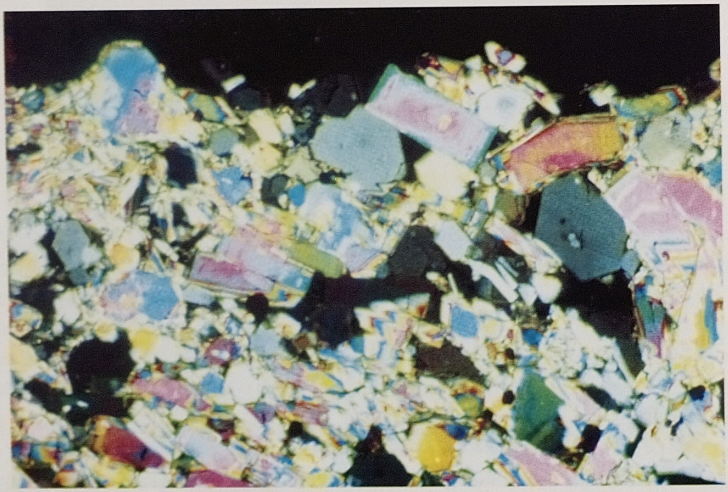
Phot. 3. The thaumasite from vein (sample 333.7 m) — SEM image



Phot. 4. The thaumasite crystals (100–200 µm) in polarized light (crossed polars)

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