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Hydrothermal laumontite from the Strzelin granitoids

ABSTRACT: Laumontite occurring in post-magmatic veins in the Strzelin granitoids, Lower Silesia (southern Poland), as it is evidenced by its physical and optical properties, chemical composition and crystallochemical formula, as well as by the X-ray powder patterns, *DTA* curve, *IR* absorption spectrum, and studies of fluid inclusions in associated quartz, originated jointly with associated minerals at the temperature range 340—175°C under the pressure lower than 730 ± 70 bars from diluted CO_2-bearing hydrothermal solutions. Full succession of the minerals in laumonite-bearing veins from the Strzelin granitoids is subsequently presented.

INTRODUCTION

Laumontite, the fibrous or prismatic calcic zeolite with C2 space group symmetry (Amirov & *al.* 1971), is known from veins and cavities in magmatic rocks; it also occurs in metamorphosed and metasomatosed rocks. Laumontite has been found in a large scale in sedimentary rocks as authigenic mineral (for references *see* Deer & *al.* 1963).

Occurrences of laumontite in the Sudetes Mts and Lower Silesia in southern Poland have been reported by Traube (1888). Of the several noted localities, appearance of laumontite in association with diverse minerals of drusy cavities in pegmatites from Strzegom granitoids is of a particular interest (*see also* Schwantke 1896. Gürich 1915, Michell 1941).

Laumontite from fractures and hydrothermal veins cutting granitoids in the main quarry at Strzelin occurs with quartz, prehnite, calcite, chlorite, epidote, stilbite, sphene and apatite (*cf.* Pl. 1, Fig. 8 and Pl. 2, Figs 1—4). The first report on this occurrence has been presented formerly by the present author (Stępisiewicz 1977).

Acknowledgements. The author is greatly indebted to Dr. A. Kozłowski and Dr. Ł. Karwowski for their kind help, advice and discussions in the course of the presented studies.

ANALYTICAL DATA

PHYSICAL AND OPTICAL PROPERTIES

The investigated laumontite is white with silk lustre, and in thin section it is colorless. It forms acicular and fibrous crystals up to 10 mm long with good (010) and (110) cleavages. Density of examined laumontite was measured by picnometer method as equal 2.254 g/cm³.

Optical properties, measured at room temperature in yellow light, are as follows: $\alpha = 1.504 \pm 0.001$; $\beta = 1.510 \pm 0.001$; $\gamma = 1.514 \pm 0.001$; $\Delta = 0.010$; $2V_\alpha = 34-38°$; $Z/\gamma = 30 \pm 2°$. The indices of refraction match those given by Coombs (1952) for leonhardite, a partially dehydrated variety of laumontite; the partial loss of water in the investigated minerals has probably been caused by its keeping under room conditions.

CHEMICAL COMPOSITION

The crystallochemical formula was calculated upon the chemical analysis (Table 1), accepting the number of 48 oxygen ions per unit cell, according to general formula (Coombs 1952) of laumontite $Ca_x(Na, K)_yAl_{2x+y}Si_{24-(2x+y)}O_{48} \cdot 16H_2O$. Low content of alkalies in the investigated laumontite results from difficulties to substitutions in its structure (Amirov & *al.* 1967, Miller & Ghent 1973). Small excess of Ca-ions is presumably caused by an impercible admixture of calcite in the analysed sample.

Table 1

Chemical composition and crystallochemical formula of laumontite from Strzelin

Component	in weight %	on 48 oxygens
SiO_2	51.01	15.92
TiO_2	0.05	0.01
Al_2O_3	21.21	7.80
Fe_2O_3	0.32	0.08
MnO	0.04	0.01
CaO	12.36	4.13
MgO	<0.03	-
Na_2O	0.20	0.12
K_2O	0.60	0.24
H_2O+	12.97	14.98
H_2O-	1.41	
Total	100.17	
$(Ca_{4.13}Na_{0.12}K_{0.24})(Al_{7.80}Fe_{0.08})Si_{15.92}O_{48} \cdot 14.98\ H_2O$		

As follows from Coombs' investigations, laumontite loses up to 1/8 of its water when exposed to the atmosphere providing the leonhardite form. The dehydration-hydration reaction is reversible, and the content of water in mineral structure may therefore vary, even with atmospheric conditions. Chemical analysis of a sample from Strzelin shows that the water content conforms to the intermediate variety. Since the specimens were stored uncovered, the partial loss of water cannot be precluded. The investigated mineral is believed to be called generally as laumontite.

X-RAY, IR ABSORPTION AND DTA INVESTIGATIONS

The *X*-ray analysis (Fig. 1 and Table 2) was carried out on *DRON-1* diffractometer using *CuKα* radiation. The refraction intensity values were calculated from the area under the peak, and the obtained *X*-ray data agree well with those given by Coombs (1952), Kaley & Hanson (1955), Lapham (1963) and Nativel (1974). Differences in intensity of some reflections are most likely due to the laumontite habit resulting in texture of the preparate.

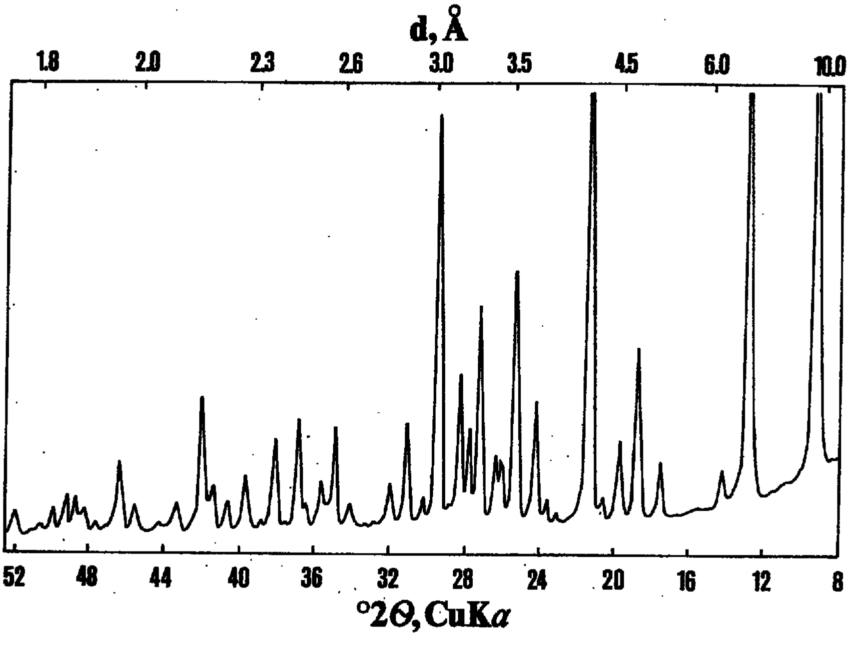

Fig. 1. *X*-ray powder diffraction pattern of laumontite from Strzelin

The *IR* absorption spectra were recorded on the *UR-20* spectrophotometer. The *IR* spectrum of the studied laumontite has several characteristic absorption bands (Fig. 2). The broad band in the region 3700—3200 cm^{-1} with peaks at 3300 and 3550 cm^{-1} is generated by asymmetrical and symmetrical stretching vibrations of the $(OH)^-$ groups in water molecules included into the crystal structure (Boldyrev & Povarennykh 1968). The explicit band 1665 cm^{-1} is due to bending vibrations of water molecules. The five distinct bands in the range 1200—900 cm^{-1} are caused by triple disorder asymmetric stretching vibrations of silica tetrahedra (Povarennykh & Gevorkyan 1971). The strong band at 775 cm^{-1} and bands in the range 650—430 cm^{-1} are generated from bending vibrations of bands in $(AlO_4)^{5-}$ and $(SiO_4)^{4-}$ tetrahedra. Carbonate ion vibrations result in the band at 1430 cm^{-1} and

Table 2

X-ray powder diffraction data of laumontite

(hkl)	Dillsburg, Penna., USA *Lapham (1963)*		Cilaos, Réunion *Nativel (1974)*		STRZELIN	
	d, A	I/I_0	d, A	I/I_0	d, A	I/I_0
110	9.49	100	9.50	100	9.50	100
200	6.86	36	6.91	30	6.86	80
020	6.54	2				
$\bar{2}01$ 011	6.19	2*	6.21	2	6.19	2
120	5.91	1.7				
111	5.052	6*	5.080	7	5.04	6
220	4.731	19.6*	4.751	25	4.72	16
221	4.500	8*	4.507	12	4.49	7
310	4.314	2.6			4.30	1
130 201	4.156	61	4.171	58	4.15	75
131	3.768	2.4	3.782	2	3.76	1
$\bar{4}01$	3.667	13	3.674	12	3.66	10
002 221	3.510	31.7*	3.517	32	3.50	30
131	3.411	7.7*	3.418	12	3.41	3
$\bar{3}12$ 012	3.367	3.7*	3.373	13	3.36	3
040	3.272	21	3.275	24	3.27	21
$\bar{3}31$ 311 $\bar{4}21$	3.205	7.8	3.215	22	3.20	5
320	3.152	16*	3.161	27	3.14	11
$\bar{4}02$	3.033	27	3.040	31	3.03	50
420 112	2.950	3.4	2.970	4	2.95	1
240 041	2.881	13.7	2.882	12	2.876	9
$\bar{5}11$ $\bar{2}41$	2.798	2.7	2.801	5	2.794	2
	2.629	3	2.630	4	2.626	1
	2.575	14*	2.572	12	2.572	8
	2.521	4	2.520	6	2.515	2
	2.463	3			2.463	1
	2.439	14	2.444	19	2.436	9
	2.361	12.3	2.365	14	2.357	7
600	2.278	2.7	2.273	7		
	2.268	6.5			2.264	4
	2.217	4.6	2.215	2	2.216	2
060	2.180	5.4	2.183	7	2.176	2
	2.153	18.6	2.154	15	2.147	14
	2.082	1.5	2.082	2	2.084	2
	2.060	1.5				
	2.042	1.5	2.040	2	2.044	1

*some preferred orientation

very slight band at 880 cm^{-1} testifying to a calcite admixture in the studied sample. Microscopic observations confirmed the presence of calcite intergrowths in laumontite crystals.

The *DTA* were carried out on Derivatograph *OD 102 A* in the temperature range 20—1000°C. Five endothermic effects were recorded on the *DTA* curve (Fig. 3) at the following temperatures of the peak maxima: 120°, 250°, 460°, 800° and 960°C. The mass decrease attends the first fourth endothermic effects and it is connected with the dehydration reaction.

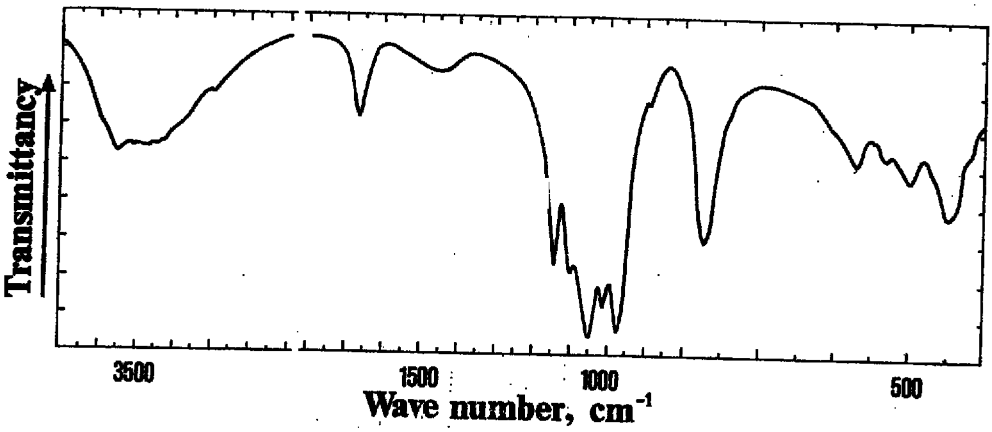

Fig. 2. *IR* absorption spectrum of laumontite from Strzelin

The *X*-ray patterns of a sample heated to 150°, 300°, 500°, 840° and 1000°C were made for elucidation of the thermal behaviour of laumontite and its transformation phases (Fig. 4). The sample heated up to 300°C has not demonstrated any substantial differences and it has been attributed to β-leonhardite. Heating of the sample to 500°C results in the arising of a new mineral phase called β-leonhardite *II* (Belitsky & *al.* 1966). Further heating to 840°C followed the formation of weakly crystalline substances, being the most likely a mixture of "amorphous" SiO_2 and oxides of Al and Ca. On the *X*-ray diffraction pattern of the sample heated to 1000°C, visible are one distinct (3.19 Å) and several weak reflections of anorthite, as well as a wide reflection typical of "amorphous" silica ($2\Theta = 16$—32°).

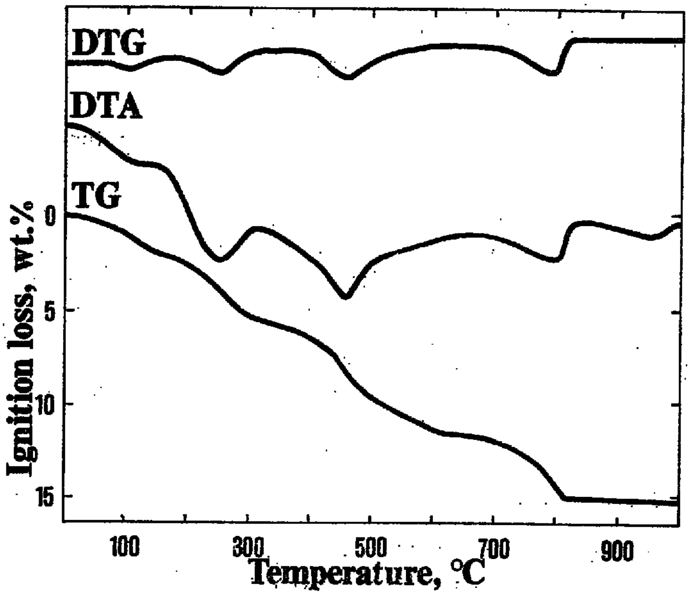

Fig. 3. *DTA*, *TG* and *DTG* curves of laumontite from Strzelin

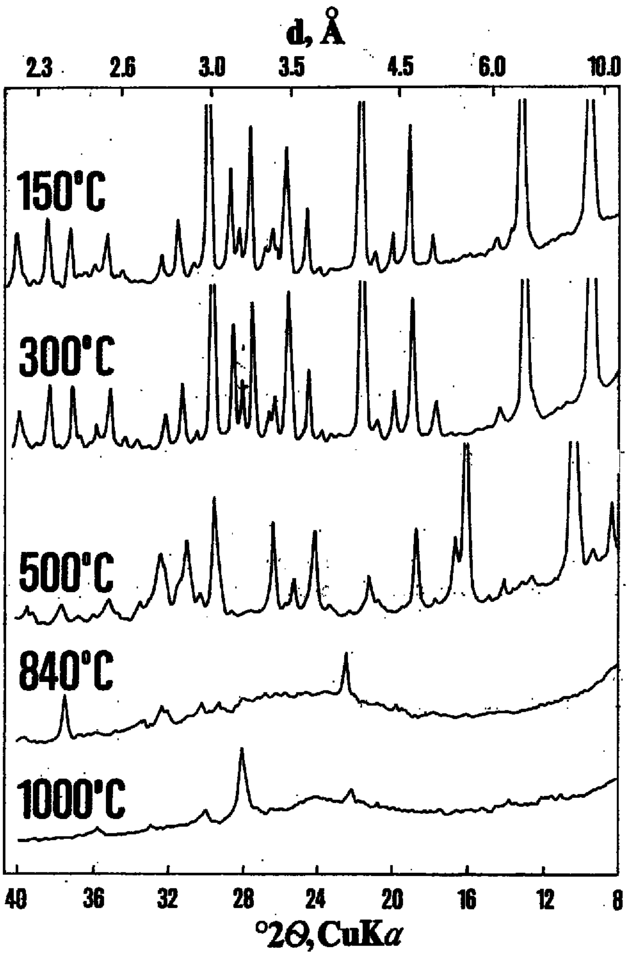

Fig. 4. X-ray powder diffraction patterns of products of the thermal destruction of laumontite from Strzelin

Conclusions based on the X-ray analysis were confirmed by the *IR* absorption studies of respective samples (Fig. 5). Generally, the dehydration reaction of the examined mineral lasts gradually until the temperature of about 800°C. Laumontite, being heated to 300°C still preserves its structure, forms β-leonhardite as a result of partial dehydration. In the temperature range 300—500°C the mineral transforms its lattice into β-leonhardite *II*. Between 700° and 800°C destruction of its structure takes place, and anorthite crystallizes from the products of such a destruction.

ENVIRONMENT OF LAUMONTITE CRYSTALLIZATION

Thermometric and barometric studies of fluid inclusions in quartz occurring with laumontite permitted to define indirectly conditions of crystallization of these two minerals.

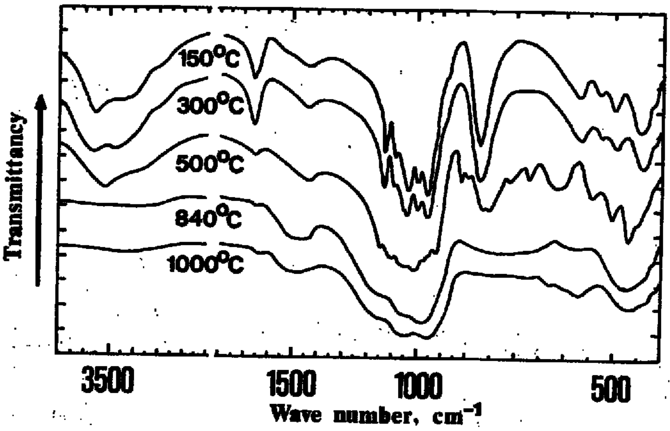

Fig. 5. *IR* absorption spectra of products of the thermal destruction of laumontite from Strzelin (transmittancy level 90% for each sample)

Primary gaseous and gaseous-liquid inclusions bearing liquid CO_2 as well as secondary gaseous-liquid inclusions were observed (Pl. 1, Figs 1—7; *see also* Stępisiewicz 1977). The inclusions homogenized in temperature ranges 280—245°C and 170—145°C, although they decrepitated in range 350—280°C. The pressure was determined by the Naumow & Malinin method (1968) availing oneself of inclusions with CO_2-phase as 730±70 bars at 280°C. The estimated value of the pressure at 170°C amounted about 500 bars. Considering the pressure correction to the homogenization temperatures (*cf.* Lemmlein 1973), the real temperatures of entrapping the inclusions into quartz crystals were determined to be 340—300°C and 200—175°C, respectively.

As it is apparent from the published experimental data (Thompson 1971, Ghent & Miller 1974), the studied laumontite presumably equilibrated with fluids having mole fraction of CO_2 about 0.0075. The absence of solid phases in the observed inclusions as well as data on chemical composition of inclusion fillings in related quartzes (*see* Stępisiewicz 1977) indicate low concentration of hydrothermal solutions.

The evaluated *P—T* values of quartz crystallization referred indirectly to laumontite are comparable with the published data (Coombs & *al.* 1959; Zen 1961; Liou 1970, 1971a, b). The *P—T* diagram of laumontite stability in the system $CaAl_2Si_4O_8$ — SiO_2 — H_2O — CO_2 was applied after Senderov (1973), taking into account the presence of CO_2 in fluids. The extent of the *P—T* field of laumontite from Strzelin on that diagram (Fig. 6; *P—T* values were limited on basis of mineral succession considerations) confirms reality of the estimated parameters obtained from the inclusion studies.

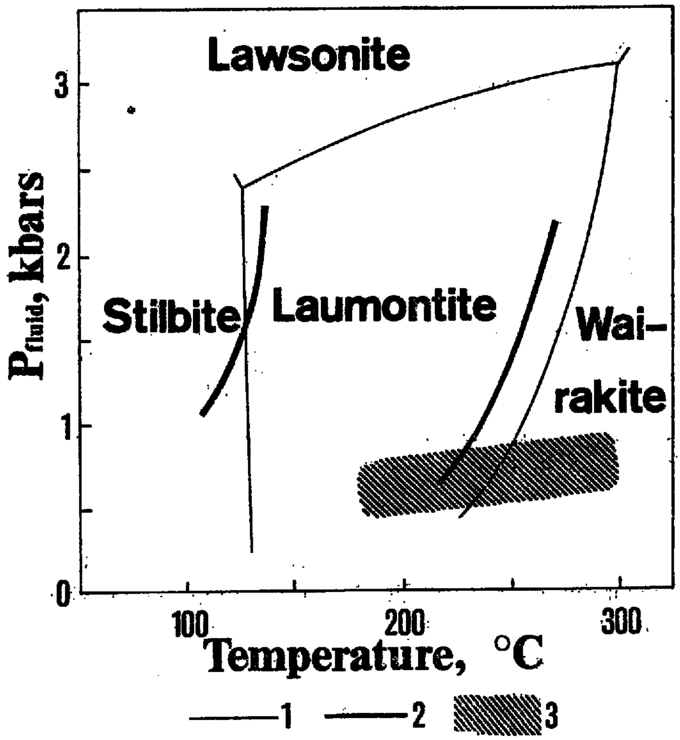

Fig. 6. *P—T* diagram of laumontite stability (*from* Senderov 1973) in the system $CaAl_2Si_2O_8$ — SiO_2 — H_2O (1) and $CaAl_2Si_2O_8$ — SiO_2 — H_2O — CO_2 (2)
P—T field for laumontite from Strzelin is shaded (3)

MINERAL SUCCESSION OF LAUMONTITE-BEARING ASSEMBLAGE

As follows from macro- and microscopic observations of the investigated laumontite and associated minerals, the main quartz mass of the investigated assemblage precipitated from CO_2-bearing fluids at temperatures 340—300°C under the pressure of 800—660 bars. Approximate *P—T* values presumably attended the formation of apatite and sphene. An insignificant decrease of temperature and pressure led to

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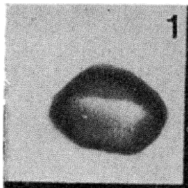
PLATE 1

1—7 Inclusions in quartz; magn. × 200 (except of Fig. 6 taken × 150)
1 One-phase gaseous inclusion
2—3 Primary three-phase inclusions bearing liquid CO_2 (T_H 280—245°C, T_HCO_2 25°C)
4—7 Secondary gaseous-liquid inclusions (T_H 170—145°C)
8 Hand specimen of hydrothermal laumontite-bearing mineral assemblage from Strzelin (*q* quartz, *ch* chlorite, *pr* prehnite, *lm* laumontite), ×1.5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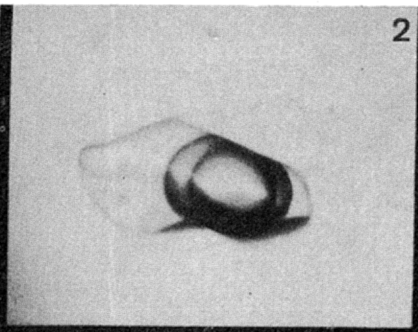
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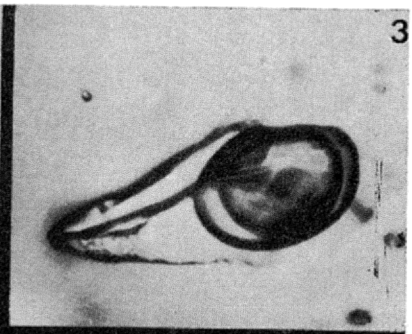
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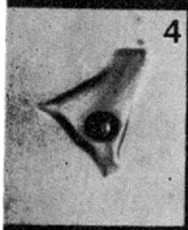
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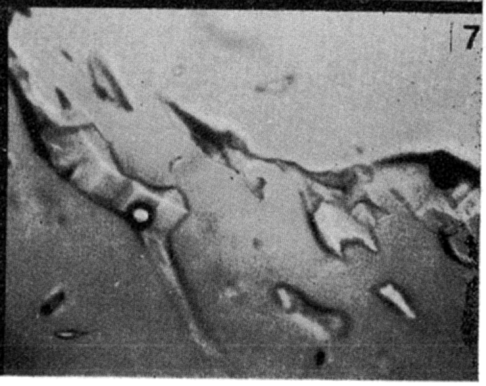
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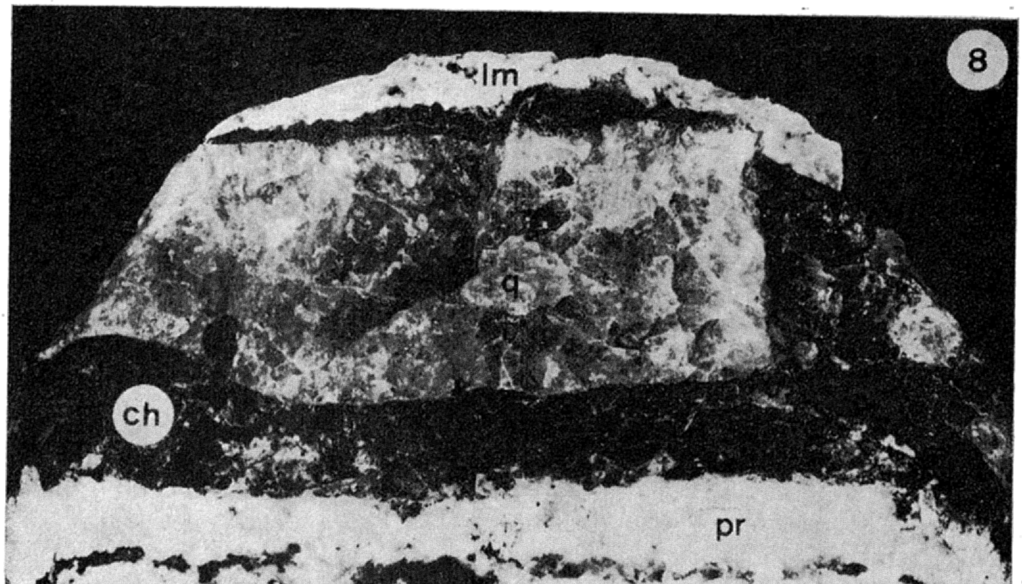
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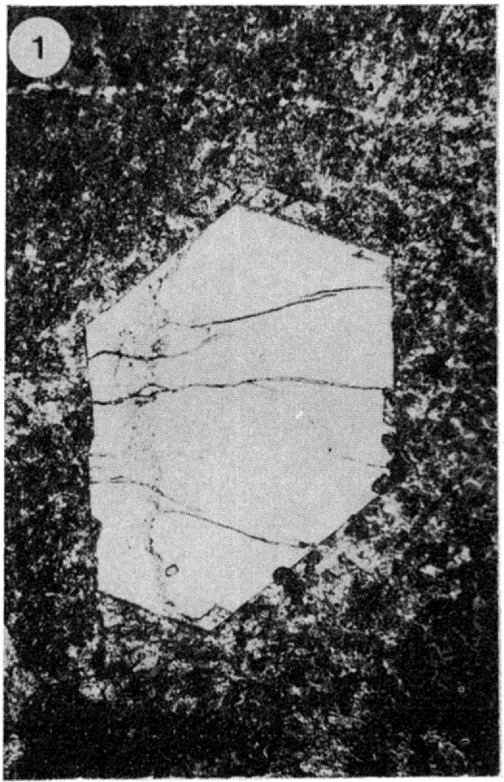
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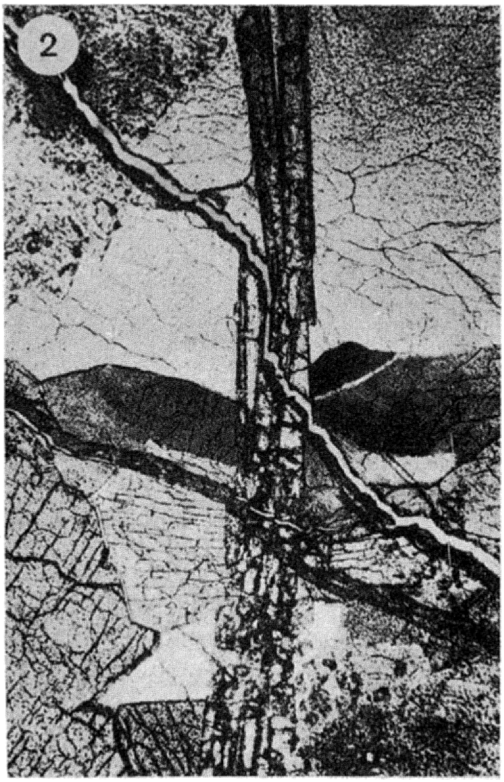
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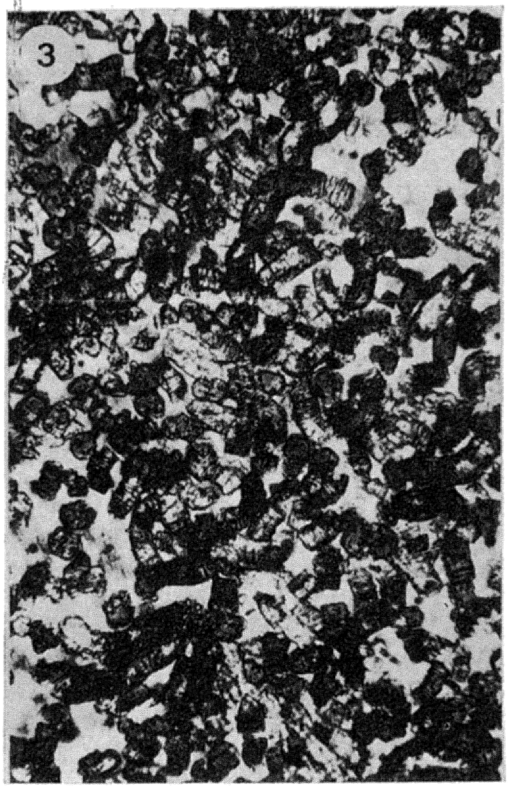
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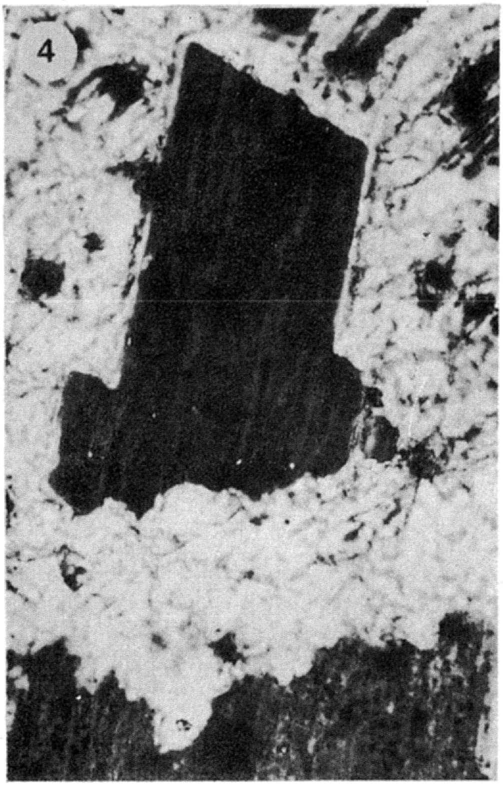
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crystallization of prehnite. Epidote most likely appeared at final stage of the proceeding mineralization. A next lowering of *P—T* parameters combined with changes of ion activities brought to the formation of laumontite and calcite (first generation), probably at the expense of prehnite and quartz, according to the reaction (*see* Winkler 1967):

$$\text{prehnite} + \text{quartz} + 3H_2O + CO_2 \rightarrow \text{laumontite} + \text{calcite}$$

The apparent stability of these association suggests that the natural conditions approaching the univariant relations were achieved. The fractures in quartz crystals were then closed up at the temperature range 200—175°C under the pressure of 500 bars, as it is indicated by the presence of fluid inclusions. Stilbite most likely originated subsequently to that latter process. The mode of contact of chlorite flakes with diverse minerals suggests a subsequent character of chloritization (Pl. 2, Figs 1 and 3) that developed within quartz, laumontite, and particularly within prehnite and calcite. It is also evident that finally the calcite mineralization of the second generation took place (Pl. 2, Fig. 4). It lasted until the temperature lower than 100°C (*cf.* Stępisiewicz 1977). Crystallization of calcite could perhaps be a consequence of the increase of CO_2 partial pressure in the discussed environment.

The examined assemblage of laumontite-prehnite-quartz plus calcite is rather rarely met since it requires special physico-chemical conditions (Zen 1961, Thompson 1971). Nevertheless, the coexistence of these minerals in the hydrothermal association at Strzelin excludes unstability of such an assemblage under the above determined conditions what previously was alleged by Rusinov (1965). The absence of kaolinite in the assemblage confirms Senderov's (1973) calculations claiming that kaolinite could not be in contact with CO_2-bearing aqueous phase under the above mentioned *P—T* conditions.

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PLATE 2

1 — Cracked automorphic quartz crystal in the first generation calcite with chlorite; corrosion of quartz by chlorite is also visible; nicols crossed, ×40

2 — Acicular crystal of epidote in prehnite, both cut by calcite veins; nicols crossed, ×40

3 — Vermicular aggregates of chlorite in laumontite; nicols crossed, ×40

4 — Chip of laumontite crystal surrounded by calcite of the second generation; nicols crossed, ×40

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HYDROTERMALNY LOMONTYT Z GRANITOIDÓW STRZELINA

(Streszczenie)

Przedmiotem pracy jest analiza lomontytu rozpoznanego po raz pierwszy wśród minerałów pomagmowych granitoidów Strzelina (*patrz* pl. 1—2). Cechy fizyczne i optyczne, skład chemiczny (tab. 1) oraz wyniki analiz: rentgenowskiej (fig. 1 oraz tab. 2), absorpcji w podczerwieni (fig. 2) i termicznej (fig. 3—5) badanego minerału wskazały na lomontyt lub jego częściowo odwodnioną odmianę (leonardyt).

Na podstawie badań gazowo-ciekłych inkluzji w kwarcu towarzyszącym lomontytowi stwierdzono, iż lomontyt wraz z towarzyszącymi mu minerałami krystalizował z rozcieńczonych roztworów hydrotermalnych zawierających CO_2 głównie w zakresie 340—175°C pod ciśnieniem rzędu 800—500 barów. Powyższe parametry są zbliżone do parametrów środowiska tworzenia się lomontytu podawanych przez innych autorów (*por.* fig. 6).

Ustalono następującą kolejność powstawania minerałów badanej paragenezy hydrotermalnej z utworów żyłowych granitoidów Strzelina: kwarc (główna masa), apatyt, tytanit, prenit, epidot, lomontyt, kalcyt (pierwsza generacja), kwarc (ilości podrzędne), desmin, chloryt i jako ostatni — kalcyt drugiej generacji.