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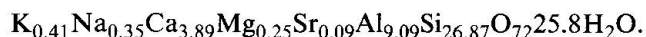
## Sr-bearing stilbite in a quartz-monzonite from Vathi, Kilkis, Northern Greece

By A. Filippidis<sup>1</sup>, C. Kougoulis<sup>2</sup> and K. Michailidis<sup>1</sup>

### Abstract

Sr-bearing stilbite occurring in veinlets, fracture and open space fillings of the Vathi quartz-monzonite (Kilkis, Northern Greece) has been examined in some detail by means of chemical and X-ray analysis.

The studied stilbite belongs to the "Intermediate Zeolites" with  $R = (\text{Si} / \text{Si} + \text{Ti} + \text{Al} + \text{Fe}^{3+}) = 0.747$  and has the following chemical formula:



The crystallographic parameters of the Sr-bearing stilbite and other seven different stilbites have been correlated with their chemical composition. The main influence to the cell parameters  $a$  and  $c$  of the stilbite comes from the monovalent cations  $\text{Na}^+$  and  $\text{K}^+$ . The Sr-bearing stilbite in the Vathi quartz-monzonite found associated with pyrite, calcite and quartz, has crystallized from hydrothermal solutions of low temperature, not exceeding 200°C.

**Keywords:** Zeolites, Sr-bearing stilbite, quartz-monzonite, crystallography and chemistry relations, Serbo-macedonian massif, Northern Greece.

### Introduction

Stilbite occurs commonly in association with other zeolites, in different rock types. It is found in sedimentary rocks (e.g. TSCHERNICH, 1972; STEPHENS and BRAY, 1973; NAKAMUTA, 1976), gneisses (e.g. PRIVETT, 1977), pegmatites (e.g. TENNYSON, 1960; ZABINSKI 1975), serpentinites (e.g. CHAUDHARI and SHARMA, 1971; ARGENTI et al., 1986), basalts (e.g. MASON and GREENBERG, 1954; WALKER, 1960; AUMENTO, 1966; COMIN-CHIARAMONTI et al., 1979), gabbros (e.g. KIMATA et al., 1979) and granitic rocks (e.g. RAADE, 1969; PRIVETT, 1977).

Stilbite is quite similar to stellerite and barerite in its X-ray diffraction trace. The most common of the three minerals is stilbite. The structure of stilbite was solved by GALLI and GOTTARDI (1966) and refinements were carried by SLAUGHTER (1970) and GALLI (1971).

Usually in natural stilbites the two sites (1 and 1P) are filled by Ca, and Na occupies in part one site (2P).

The SrO content of stilbites was always found very low, not exceeding 0.06 wt% (GOTTARDI and GALLI, 1985). In only one case a SrO content of 0.28 wt% is mentioned for a Hungarian stilbite (REICHERT and ERDELYI, 1935). The present work describes the mode of occurrence, the mineralogic properties and the genesis of a Sr-bearing stilbite in a quartz-monzonite from the Vathi area (N. Greece).

### Occurrence

The investigated area of Vathi belongs geotectonically to the Serbomacedonian massif (Fig. 1) and specifically to the Vertiskos series (KOCKEL et al., 1971). This series is composed

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of metamorphic rocks comprising biotite-muscovite, biotite-hornblende gneisses and mica-schists with concordant intercalations of amphibolites and ultramafic rocks. These metamorphics are of at least lower paleozoic age (KOCKEL and WALTHER, 1968). Several volcanic bodies of rhyolitic to andesitic composition are extruded on the metamorphics. Sub-volcanic to plutonic stock-like bodies, plugs and dykes ranging in composition from monzonite-monzodiorite to quartz monzodiorite and dacite-rhyodacite porphyries are also intruding the metamorphics. The magmatism in this area is considered to be of Upper Pliocene to lower quaternary age (MELIDONIS, 1972; KOCKEL et al., 1977; PANAGOS et al., 1978).

Drilling through an intrusive body (Fig. 1), which is surrounded by serpentinized ultramafic rocks, partly altered to talc and/or magnesite ceased at 504 m depth.

The study of thin sections of the recovered drill core revealed that the intrusive has the following mineralogic assemblage:

quartz, K-feldspar, plagioclase (An=35%), biotite, Ca-amphibole, apatite, sphene, opaques±chlorite±calcite.

Chemical analysis on representative samples of the intrusive rock (table 1) gave the composition of a quartz-monzonite, according to STRECKEISEN and LE MAITRE (1979) classification. The quartz-monzonite shows a strong potassic alteration and porphyry copper type mineralization, consisting of pyrite, chalcopyrite and traces of molybdenite and galena. These phenomena are connected with the latest stages of the same magmatic event which gave the quartz-monzonite.

Stilbite was found to be more widespread between 52 and 55 m depth, from which the studied samples have been collected.

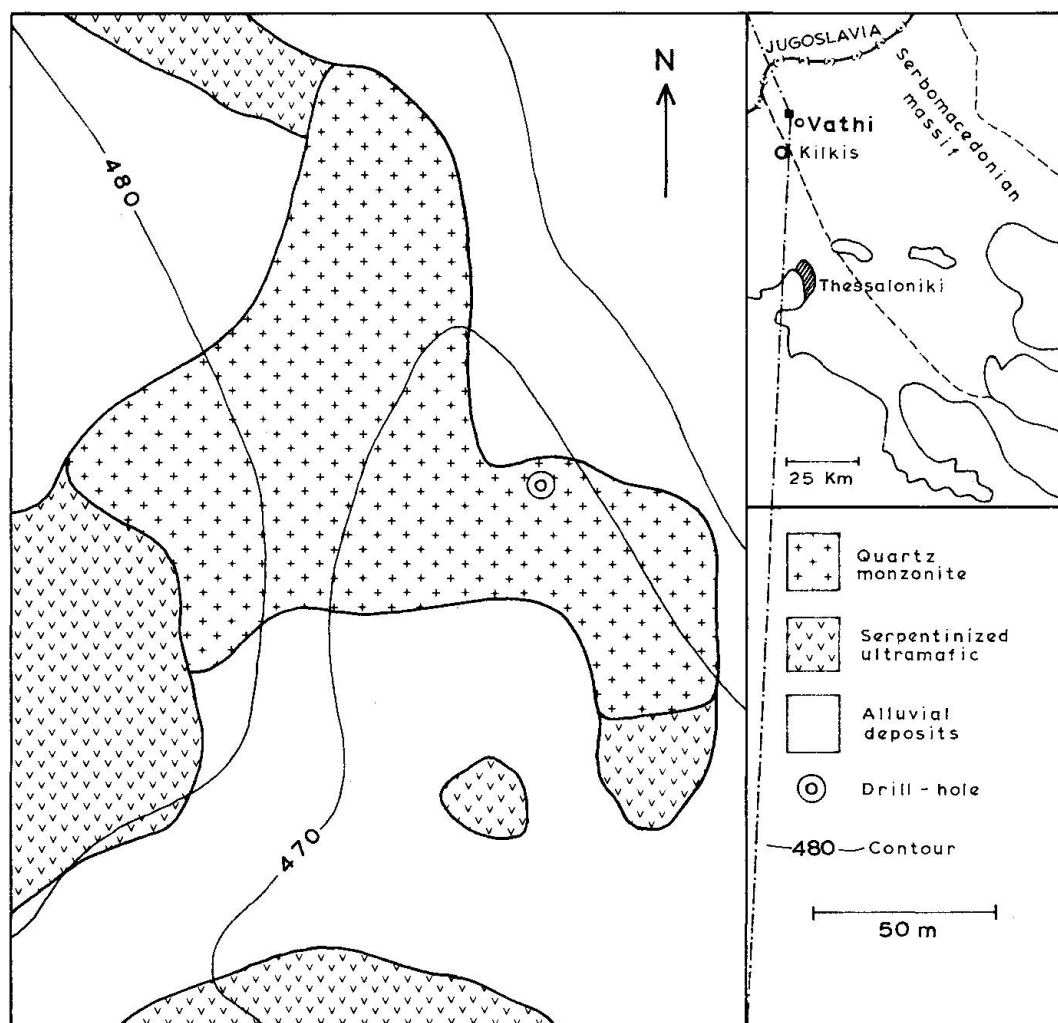


Fig. 1 Geological map of the investigated area.

*Tab. 1* Chemical composition of the quartz-monzonite from Vathi, Kilkis (N. Greece).

SiO <sub>2</sub> wt%	63.32
TiO <sub>2</sub>	0.35
Al <sub>2</sub> O <sub>3</sub>	17.58
Fe <sub>2</sub> O <sub>3</sub>	1.13
FeO	1.91
MnO	0.05
MgO	2.57
CaO	3.28
Na <sub>2</sub> O	4.00
K <sub>2</sub> O	5.60
P <sub>2</sub> O <sub>5</sub>	tr.
SO <sub>3</sub>	0.87
Total	100.66

Average of 3 analyses. Mixed sample from representative rock-samples selected from the drill-core (depth 180–240 m).

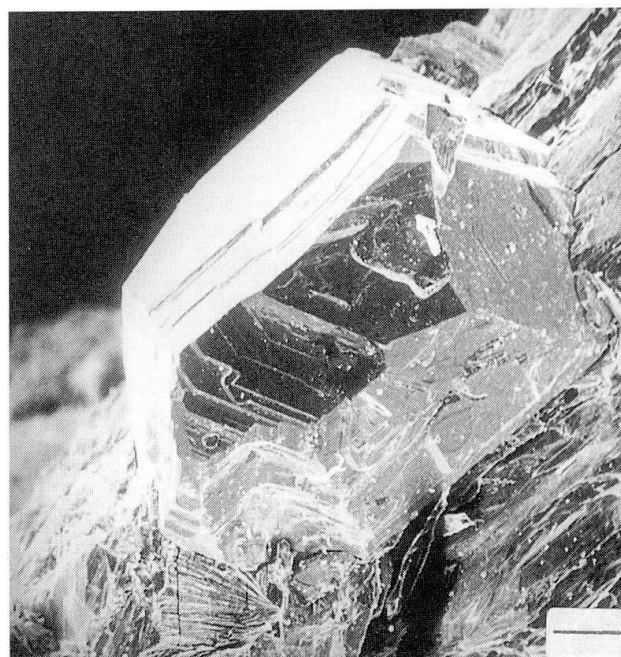


*Fig. 2* Scanning-electron microphotograph of Vathi Sr-bearing stilbite. Bar scale = 100  $\mu$ m.

It occurs in veinlets (up to 1 cm wide), fractures and open spaces (up to 10 cm wide) of the quartz-monzonite. In vein and fracture-fillings, stilbite develops granular masses or interlocking small crystals, while in open spaces it develops radiating aggregates of tabular crystals (up to 8 mm long), which are actually cruciform twins. The aggregates are grouped in nearly parallel position to form sheaf-like aggregates (Fig. 2). Thin section studies revealed that stilbite shows good cleavage and its characteristic twinning in polycrystalline aggregates. It is creamy yellow in colour with brown tint and occurs associated with calcite, quartz, pyrite (Fig. 3) and secondary alkali-feldspar and biotite.

### X-ray data

X-ray powder diffraction traces were taken with a Philips diffractometer, Ni-filtered CuK $\alpha$  radiation, using silicon as internal standard. The scanning speed was 1/1° and 1/4° per minute over the interval 4–104° of 2 $\theta$ . The X-ray pattern of Sr-bearing stilbite was carefully compared with the X-ray patterns of stilbite, stellerite and barrerite. The X-ray pattern differences between stilbite and stellerite indexed



*Fig. 3* Scanning-electron microphotograph of euhedral pyrite crystal as inclusion in the Sr-bearing stilbite. Bar scale = 71  $\mu$ m.

Tab. 2 Microprobe analyses and cell parameters of the Sr-Bearing stilbite from Vathi, Kilikis (N. Greece).

Sample No	Wt. %		Calculated to 72(0)		Cell parameters		
	V1	V2	V1	V2	V1	V2	
SiO <sub>2</sub>	57.29	57.57	Si	26.904	26.839	$a$ (Å)	13.607(6) 13.601(5)
TiO <sub>2</sub>	0.02	bdl	Ti	0.007	-	$b$ (Å)	18.259(8) 18.252(7)
Al <sub>2</sub> O <sub>3</sub>	16.34	16.62	Al	9.044	9.132	$c$ (Å)	11.286(5) 11.282(4)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.02	Fe <sup>3+</sup>	0.004	0.007	$v$ (Å <sup>3</sup> )	2210.7 2208.8
MgO	0.33	0.38	Mg	0.231	0.264	$\beta$	127° 96' 127° 94'
CaO	7.69	7.81	Ca	3.869	3.901		
SrO	0.31	0.34	Sr	0.084	0.092		
BaO	0.02	0.01	Ba	0.004	0.002		
NiO	0.03	0.03	Ni	0.011	0.011		
Na <sub>2</sub> O	0.35	0.41	Na	0.319	0.371		
K <sub>2</sub> O	0.83	0.53	K	0.497	0.315		
H <sub>2</sub> O*	16.78	16.28	H <sub>2</sub> O	26.28	25.31		
			Z	35.959	35.978		
Total	100.00	100.00	X	5.015	4.956		
			R	0.748	0.746		

V1) Average of 3 analyses. V2) Average of 7 analyses. \*) Estimated by difference. bdl = below detection limit. Z = Si + Ti + Al + Fe<sup>3+</sup>. X = Mg + Ca + Sr + Ba + Ni + Na + K. R = Si/(Si + Ti + Al + Fe<sup>3+</sup>). Figures in parentheses represent the estimated standard deviation; thus 13.607 (6) indicates estimated standard deviation of 0.006.

by PASSAGLIA et al. (1978) and GOTTARDI and GALLI (1985) were taken into consideration.

Stilbite is monoclinic with crystal symmetry C2/m (SLAUGHTER, 1970; GALLI, 1971), while stellerite and barrerite are orthorhombic with crystal symmetries Fmmm and Amma respectively (GALLI and ALBERTI, 1975). Natural stilbite crystals showing orthorhombic and monoclinic growth sectors have been mentioned by AKIZUKI and KONNO (1985).

The X-ray powder lines which are single in the orthorhombic system, split up in the monoclinic one (PASSAGLIA et al., 1978). In the most appropriate interval between 23° and 24° of 2 $\theta$  (CuK $\alpha$ -radiation), the studied stilbite exhibits two distinct peaks of the same height. According to PASSAGLIA et al. (1978) this is characteristic for monoclinic stilbite.

Cell parameter refinements (Table 2) on the Sr-bearing stilbite were performed with X-ray

powder diffraction data, using the 18 strongest and welldefined reflections and Si as internal standard. The values of the cell dimensions of the studied stilbite are close to those reported by other workers (e.g. GALLI and GOTTARDI, 1966; SIMONOT-GRANGE et al., 1970).

### Chemistry of the Minerals

Microprobe analyses of Sr-bearing stilbite and some of the associated minerals: alkali-feldspar, biotite and pyrite are given in Tables 2, 3 and 4.

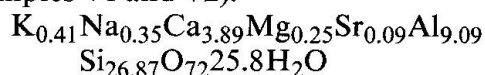
Stilbite from Vathi contains considerable amounts of Sr (on average 0.33 wt% SrO), which to our update knowledge is the highest Sr-content reported in literature. The major cation in the Vathi stilbite is Ca but considerable amounts of K, Mg and Na are present. The

Tab. 3 Microprobe analyses of alkali-feldspar (V2) and biotite (V1) from Vathi, Kilkis (N. Greece).

Sample No	Wt %		Calc. on 8(0)		11(0)
	V2	V1	V2	V1	
SiO <sub>2</sub>	64.97	37.22	Si	2.990	2.818
TiO <sub>2</sub>	0.02	4.68	Al	1.007	1.121
Al <sub>2</sub> O <sub>3</sub>	18.56	12.56	Ti	0.001	0.267
FeO*	0.15	14.87	Fe <sup>2+</sup>	0.006	0.942
MnO	n.a.	0.16	Mn	-	0.010
MgO	0.07	15.16	Mg	0.005	1.711
CaO	bdl	n.a.	Sr	0.016	-
SrO	0.61	n.a.	Ba	0.003	-
BaO	0.16	n.a.	Na	0.111	0.029
NiO	bdl	n.a.	K	0.846	0.942
Na <sub>2</sub> O	1.24	0.20	Or	88.4	-
K <sub>2</sub> O	14.40	9.75	Ab	11.6	-
			An	0.0	-
Total	100.18	94.60			

V2) Average of 3 analyses. V1) Average of 3 analyses.  
\*) FeO as total iron. bdl = below detection limit. n.a. = not analysed.

chemical parameter  $R = \text{Si} / (\text{Si} + \text{Ti} + \text{Al} + \text{Fe}^{3+})$ , which represents the percentage of the tetrahedra occupied by Si, varies between 0.746 and 0.748. Based on the R values, the investigated stilbite is classified as "Intermediate zeolite" ( $0.625 < R < 0.750$ ) after GOTTARDI (1978). The calculation of cation proportions on 72 oxygen atoms (Table 2) gives the formulae (average of samples V1 and V2):



Tab. 4 Microprobe analyses of pyrite from Vathi, Kilkis (N. Greece).

Sample No	Wt. %	Calc. on 2(S)	
	V1	V1	
Cu	0.04	Co	0.01
Ni	0.03	Fe	0.99
Co	0.43	S	2.00
Fe	46.01	Σ	3.00
S	53.56	Formula	Fe <sub>0.99</sub> Co <sub>0.01</sub> S <sub>2</sub>
Total	100.07		

V1) Average of 3 analyses.

PASSAGLIA et al. (1978), using the chemistry of many stellerites and stilbites on a triangular mole plot (Fig. 4), distinguish two separate fields for the minerals. Using the chemical data of table 2 and the triangular mole plot of PASSAGLIA et al. (1978), we see that the Vathi Sr-bearing stilbite falls in the stilbite area of Figure 4.

The alkali-feldspar (Or<sub>88.4</sub>Ab<sub>11.6</sub>An<sub>0.0</sub>) contains considerable amounts of SrO (0.6 wt%).

The associated mica (Table 3) is a common tri-octahedral mica, namely biotite. Partial microprobe analyses on calcite gave CaO = 47.5, MgO = 0.1, FeO = 0.0 and SiO<sub>2</sub> = 0.1 wt%.

The euhedral crystals of pyrite (Fig. 3) in association with the stilbite, contain around 0.4 wt% Co (Table 4).

### Relationship between crystallographic parameters and chemical composition.

In this study the crystallographic parameters  $a$ ,  $b$ ,  $c$ ,  $V$  and  $\beta$  of different stilbites have been correlated with their chemical composition. The following chemical variables were chosen:

$$\begin{aligned} Z &= \text{Si} + \text{Ti} + \text{Al} + \text{Fe}^{3+}, \\ R &= \text{Si} / (\text{Si} + \text{Ti} + \text{Al} + \text{Fe}^{3+}), \\ X &= \text{Mg} + \text{Ca} + \text{Sr} + \text{Ba} + \text{Ni} + \text{Na} + \text{K}, \\ M &= \text{Na} + \text{K} \text{ (monovalent cations)}, \\ D &= \text{Ca} + \text{Mg} + \text{Sr} + \text{Ba} + \text{Ni} \text{ (divalent cations)}, \end{aligned}$$

Na, K,  $Z/X$  and  $M/X$ .

The correlation diagrams have been made using data of nine different stilbites from various places (see footnotes to Fig. 5).

The correlations of the crystallographic parameters  $b$ ,  $V$  and  $\beta$  with the chemical composition show broad scattering. The crystallographic parameters  $a$ ,  $b$ ,  $c$ ,  $V$  and  $\beta$  seem not to be effected by the divalent cations (Fig. 5).

The correlations  $a - (X, M, M/X, Z/X)$  and  $c - (X, M, M/X, Z/X)$  seem to be the best ones. Both parameters  $a$  and  $c$  are positively correlated with  $X$ ,  $M$ ,  $M/X$  and negatively with  $Z/X$  (Figs. 6 and 7). Thus, the main influence to the cell parameters  $a$  and  $c$  comes from the monovalent cations ( $M$ ), these two parameters are increasing with increasing  $M$  (Figs. 6 and 7).

The correlations of  $\beta$ -angle with the chemi-



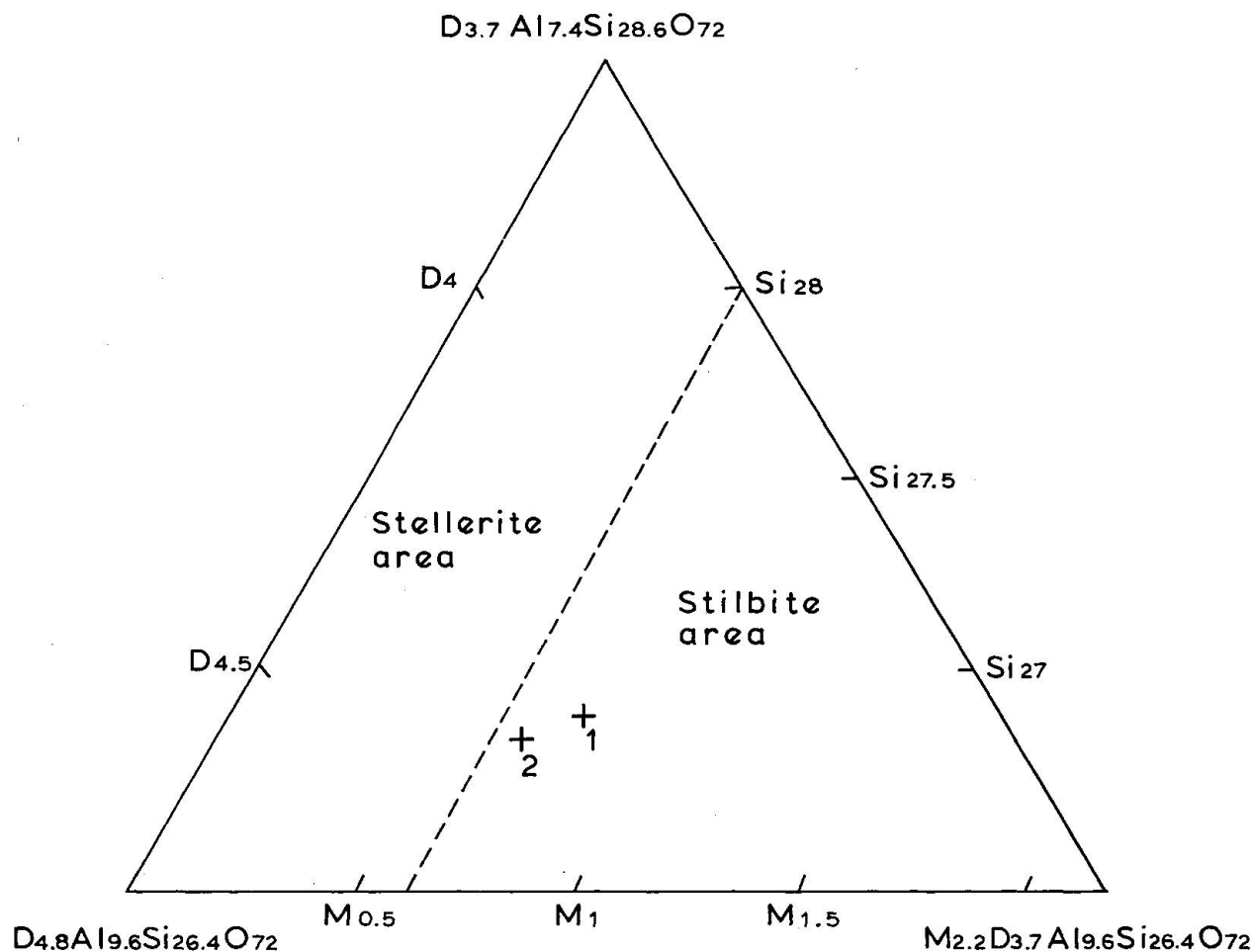


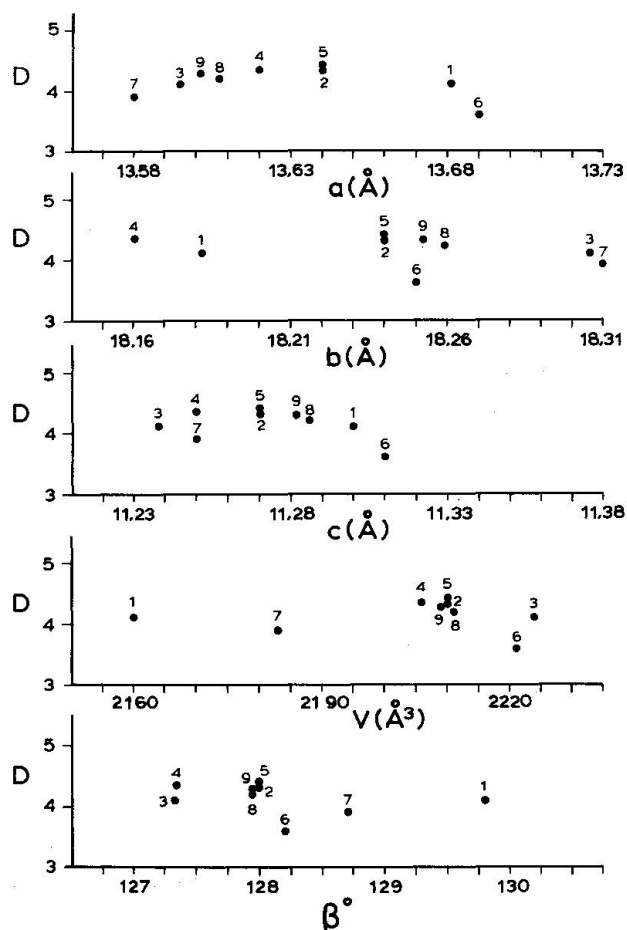
Fig. 4 The triangular mole plot  $D_{3.7}Al_{7.4}Si_{28.6}O_{72} - D_{4.8}Al_{9.6}Si_{26.4}O_{72} - M_{2.2}D_{3.7}Al_{9.6}Si_{26.4}O_{72}$  (after PASSAGLIA et al. 1978). The dashed line separates the area of stellerites and stilbites. 1) stilbite from Vathi, sample V1, 2) stilbite from Vathi, sample V2,  $M = Na + K$  (monovalent cations),  $D = Ca + Mg + Sr + Ba$  (divalent cations).

cal variables were not so discernible as in figures 6 and 7. Possibly the  $\beta$ -angle is correlated positively with the monovalent cations (Fig. 8). According to PASSAGLIA et al. (1978) the  $\beta$ -angle depends on the monovalent cations only, and K influences the  $\beta$ -angle in the same direction but less markedly than Na. The single correlations between cell parameters, Na and K show broad scattering except those of  $a$  with Na and K, which show that  $a$  increases with increasing Na and K content of stilbite (Fig. 8). These single correlations are based on data from fewer stilbites because some of them do not contain any Na or K.

#### Origin of the Sr-bearing stilbite

The mode of occurrence (veinlets, fracture and open space fillings) of the studied stilbite within the quartz-monzonite indicate an epigenetic origin. The association of the mineral with pyrite, calcite and quartz as well as the potassic alteration (secondary K-feldspar and biotite) and the porphyry copper mineralization of the quartz-monzonite suggest a hydrothermal origin.

The Sr content of the stilbite (0.31–0.34 wt% SrO, Table 2) derived mainly by hydrothermal activity on the Sr-bearing alkali-feldspars of



the quartz-monzonite, which contain around 0.6 wt% SrO (Table 3).

The stability field of stilbite was determined experimentally by JUAN and LO (1973). Their experimental study of the reaction  $\text{stilbite} = \text{Ca-analcime} + \text{quartz} + \text{H}_2\text{O}$  indicates that the equilibrium temperature is 213°C at 680 atms, 222°C at 1700 atms and 226°C at 2380 atms.

The reaction  $\text{stilbite} = \text{laumontite} + 3 \text{ quartz} + 3 \text{ H}_2\text{O}$  was experimentally studied by LIOU (1971), who suggested that stilbite is crystallized at temperatures lower than 200°C. The equilibrium temperature for their reaction is 170°C at 2 kbars, 185°C at 3 kbars and 185°C at 4 kbars. The existence of stilbite from ~180°C to ~226°C (JUAN and LO, 1973), is probably metastable in view of the results of

Fig. 5 Plot of D versus a, b, c, V and  $\beta$  in stilbite. D=Ca+Mg+Sr+Ba+Ni (divalent cations). Stilbites used: (1) from AUMENTO (1966), (2) from GALLI and GOTTARDI (1966), (3) from WISE (1969), (4) from ABBONA and FRANCHINI (1970), (5) from SIMONOT-GRANGE et al., (1970), (6) from SLAUGHTER (1970), (7) from LIOU (1971), (8) sample V1 and (9) sample V2 from this study.

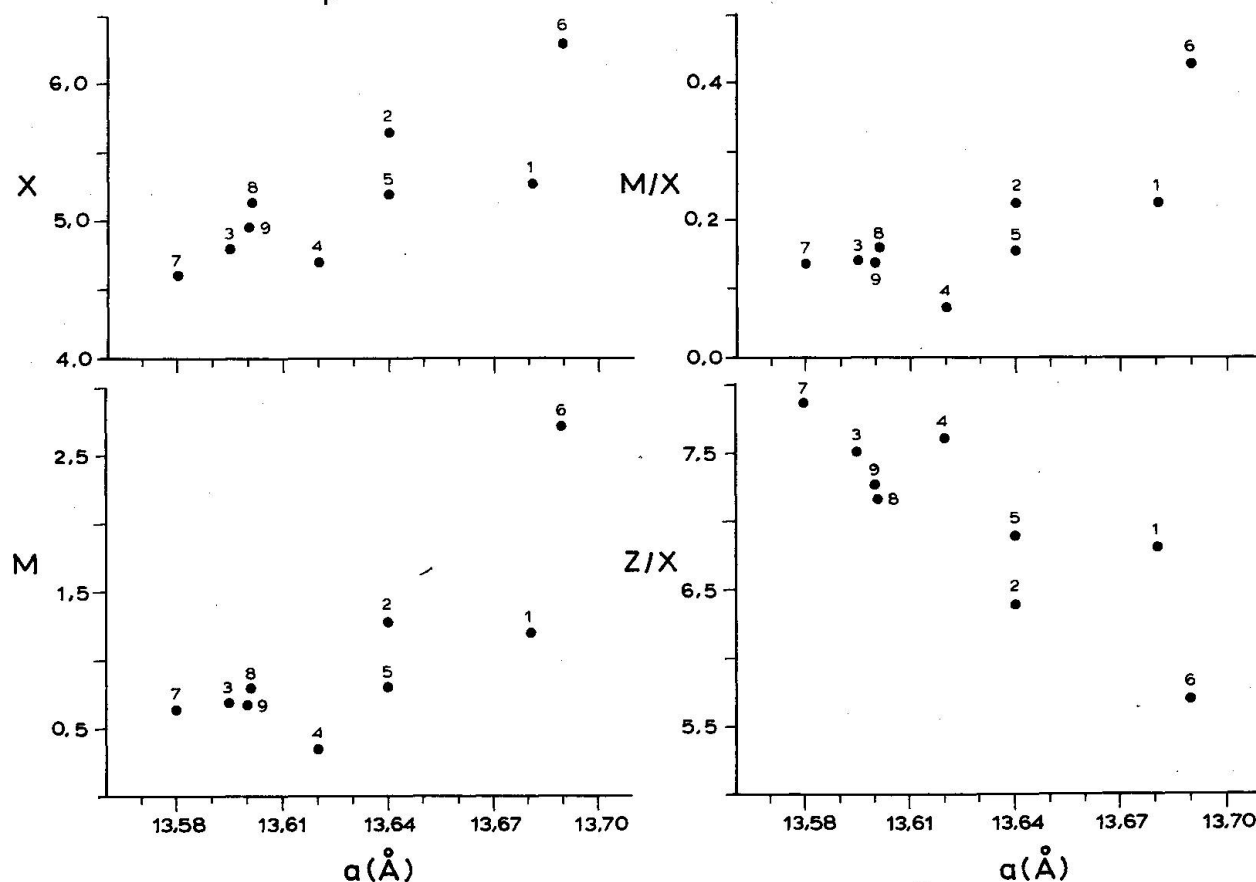


Fig. 6 Plot of a versus composition in stilbite. X=Mg+Ca+Sr+Ba+Ni+Na+K (divalent and monovalent cations), M=Na+K (monovalent cations), Z=Si+Ti+Al+Fe<sup>3+</sup>. Stilbites used: see footnotes to Fig. 5.



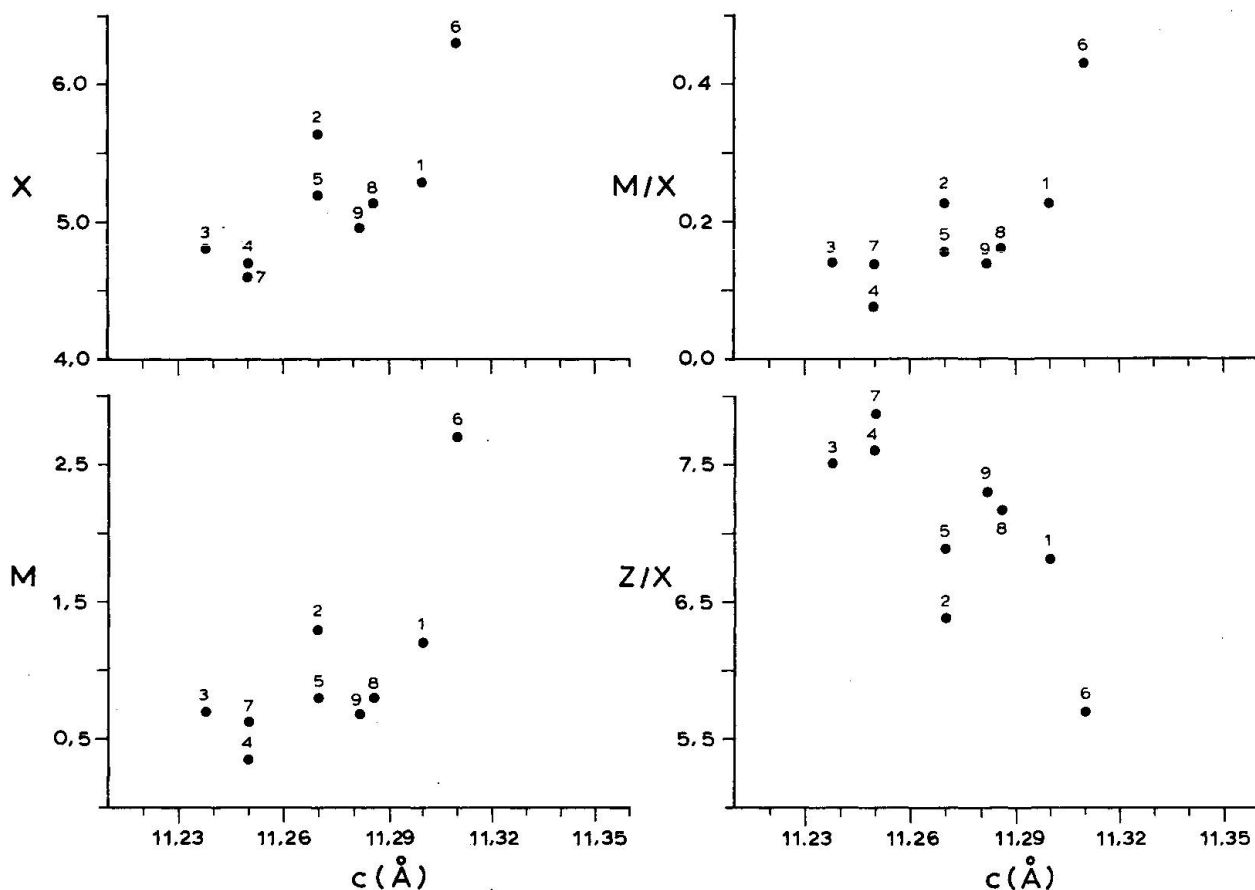
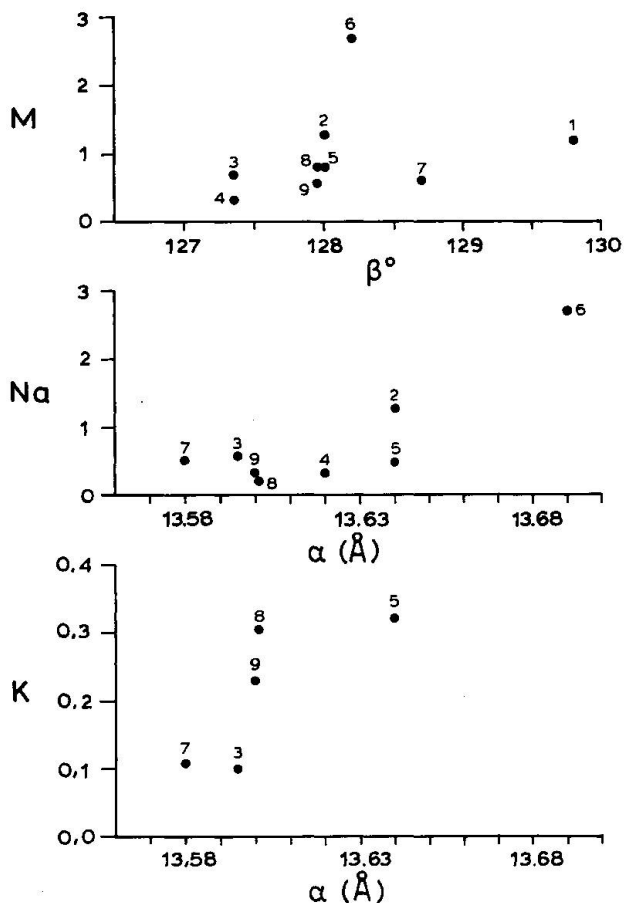


Fig. 7 Plot of  $c$  versus composition in stilbite. For abbreviations and symbols see footnotes to Figs 5 and 6.



LIU (1971). HARADA and TOMITA (1967) showed that stilbite above 200°C and water pressure >40 atms produced wairakite. A stability diagram for the above mentioned reactions is presented in Figure 9.

In the Vathi quartz-monzonite no Ca-analcime, laumontite, wairakite or other zeolites were observed. From the above it is concluded that the studied Sr-bearing stilbite has originated from a late stage hydrothermal activity, at temperature not exceeding 200°C.

Fig. 8 Plot of  $\beta$  and  $a$  versus composition in stilbite. For abbreviations and symbols see footnotes to Figs 5 and 6.

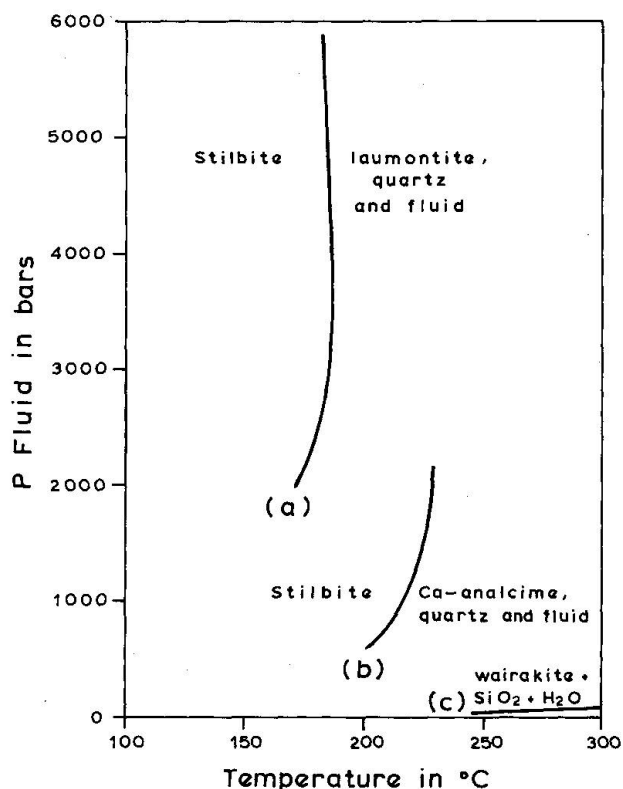


Fig. 9 P-T fields for stilbite. (a) stilbite = laumontite + quartz +  $H_2O$  (LIOU, 1971); (b) stilbite = Ca-analcime + quartz +  $H_2O$  (JUAN and LO 1973); (c) stilbite = wairakite +  $SiO_2$  +  $H_2O$  (HARADA and TOMITA, 1967).

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