Genesis of filamentary pyrite associated with calcite crystals

IVAN K. BONEV1*, JUAN MANUEL GARCIA-RUIZ2, RADOSTINA ATANASSOVA1, FERMIN OTALORA2 and SVETOSLAV PETRUSSENKO3

1Geological Institute, Bulgarian Academy of Sciences, 1113 Sofia, Bulgaria
2Instituto Andaluz de Ciencias de la Tierra, Universidad Granada, 18002 Granada, Spain
3National Museum of Natural History, 1 Tsar Osvoboditel Blvd., 1000 Sofia, Bulgaria

*Corresponding author, e-mail: bonev@geology.bas.bg

Abstract: The calcite of the hydrothermal Surneshko Kladenche copper vein deposit from the Rossen ore field, Bulgaria, sometimes encloses peculiar filamentary pyrite crystals. Three successive calcite generations were observed belonging to a low-temperature (<235°C) carbonate paragenesis formed in open cavities of the ore veins after the main chalcopyrite mineralisation. Three generations of pyrite crystals are associated with these calcite crystals: pyrite 1, with [001] elongated columnar crystals which crystallised in open space; pyrite 2, in groups of long (up to 10 mm) sub-parallel tortuous filaments of varying thickness (3-20 µm) which are oriented nearly perpendicular to the surfaces of a transient w[3145] scalenohedral calcite crystal zone; and pyrite 3, made of slightly elongated small crystals, located in the outermost v[2131] zone of the same scalenohedral calcite crystals. The columnar pyrite 1 is formed by subsequent thickening of thin straight whiskers rapidly grown under diffusional regime, whereas the filamentary pyrite 2 and 3 grew contemporaneously with the enclosing calcite crystal.

Key-words: crystal morphology, filamentary crystals, calcite, filamentary pyrite, contemporaneous growth.

Introduction

Pyrite, the most important and widespread sulphide mineral on the Earth surface, usually occurs in the form of crystals displaying three main equilibrium faces, namely {100}, {111}, and {210} (Sunagawa, 1957, 1987). Nevertheless, many other crystal forms and habits have been reported for pyrite, including skeletal, dendritic and acicular crystals, strongly deviating from the equilibrium morphology (Pabst, 1971; Pshenichkin & Korobeinikov, 1974; Strunz, 1976; Endo, 1978, etc.). White (1973) and Bideaux (1970) described whisker (hairlike), filiform, and even ring-like pyrite formations. Some rare thin pyrite needles formed inside small vesicles have the characteristics of straight or kinked whiskers (Kamb & Oke, 1960; Roberts et al., 1974). A review on pyrite filiform crystals with right-angle bends from different deposits was given by Henderson & Francis (1989). They show that the bending is a specific growth feature of these crystals, not a result of (110) twinning. Bonev et al. (1985) provided a systematic scanning electron microscopic (SEM) and high voltage electron microscopic (HVEM) study on pyrite whiskers and thin platelets from lead-zinc deposits in Bulgaria and discussed their growth mechanisms.

Syngenetic whisker inclusions of pyrite in quartz from Mangyshlak in Kazakhstan were investigated by Galuskin & Winiarski (1997). Zhabin (1986) described filamentary cinnabar inclusions in calcite. Examples of simultaneous growth of two minerals have been reviewed by Grigoriev & Zhabin (1975) (mostly from Russian literature), including inclusions of hematite in quartz (Laemmlein, 1973), cassiterite in fluorite (Lyakhov, 1966), etc.

We studied the formation of pyrite filamentary inclusions in large calcite crystals collected from the Surneshko Kladenche vein copper deposit in the central part of the Rossen ore field, Bulgaria. The pyrite inclusions were isolated by selective dissolution of the calcite matrix, and examined by optical and scanning electron microscopy. The results of these observations are reported in the present paper, followed by a discussion of the growth mechanisms.

Geological setting

The economically important Bourgas ore district is located in the easternmost part of the Cretaceous Srednogorie metallogenic zone. It is comprised of three important ore fields namely, Rossen, Vurly Bryag and Zidarovo. The Rossen ore field is related to the Senonian Rossen volcanic-intrusive structure of andesites and tuffs in which a central gabbro-monzonite-syenite plutonic core is intruded, followed by porphyritic subvolcanic bodies and dykes (Bogdanov, 1978). The numerous copper ore veins were intensively worked during 1945-1995 in several underground mines.
such as Rossen, Koru Cheshma, Meden Rid, Surneshko Kladenche, Propadnala Voda and Chiplaka (Fig. 1) and are now abandoned. The mineralisation formed mainly by open space filling includes an early pre-ore silicate stage (phlogopite, feldspars, pyroxenes, apatite, etc.), the main ore stage of several parageneses (with main minerals chalcopyrite, pyrite, quartz, chlorite and carbonates, together with magnetite, hematite, molybdenite, bismuthinite, etc.), and a final carbonate stage. According to Strashimirov & Kovachev (1992) the main ore stage was rather high-temperature (365-320°C), while for the late post-ore stage the T range is 300-100°C.

Pyrite is an important mineral in the copper ores and occurs in the early parageneses mainly as dense masses. In the late carbonate paragenesis pyrite is only a minor but characteristic component. Calcite is the main mineral of the final carbonate paragenesis. It forms massive coarse-grained aggregates, well-formed crystals and crystal druses. In a detailed morphological study accompanied by thermometric fluid inclusion measurements, Naidenova (1966) distinguished several crystal habits of calcite in the Rossen deposits: earliest pinacoidal plates (270°C and lower), scalenohedral v{213} (235-175°C), prismatic (170-160°C), barrel-form, rhombohedral, short-prismatic, and flat rhombohedral (140-135°C). Mincheva-Stefanova et al. (1993) described giant, up to 30 cm wide, scalenohedral calcite crystals containing central thin tabular {0001} seeds. The habit evolution of calcite crystals in these deposits mostly follows the general morphological dependence of calcite with temperature, as known from various mineral deposits (Kostov & Kostov, 1999).

Our samples were collected at the level 280 of the ore vein #185 of the Surneshko Kladenche mine. The vein consists of brecciated pieces of the main chalcopyrite-pyrite ore and altered wall rocks cemented by coarse-grained calcite and fine-grained quartz. In the central part of the vein irregular open cavities up to several meters in size occur. Their walls are partly overgrown by calcite crystals forming druses, which are the subject of this work.

**Methods**

Hand specimens of calcite druses, cleavage plates and doubly polished thin sections were examined with binocular microscope and optical transmission microscope. Calcite crystals were selectively dissolved to access to pyrite inclusions which were later studied by scanning electron microscopy. Low concentration acetic acid solutions (6% v/v) or hydrochloric acid solutions (5% v/v) were used as solvents. The detached thin pyrite crystals were very delicate and fragile. Therefore etching of calcite was performed under continuous inspection to stop it before reaching the bases of pyrite filaments, in order to preserve them attached to the calcite substrate. Scanning electron microscopic studies were performed after carbon or gold coating of crystals. JEOL Superprobe-733 and Philips 515 SEM devices were used. Electron probe microanalysis and X-ray diffraction study in Gandolfi and rotation cameras were also applied for characterising the mineral phases and crystal orientation. Some filaments were measured by optical goniometry.

**Sample description**

The filamentary pyrite crystals are associated to druses made of three successive generations of calcite crystals with different morphology and position (Fig. 2). The earliest gen-

![Fig. 1. Schematic geological map of the Rossen ore field, Bulgaria (adapted from Bogdanov, 1987).](image-url)
Table 1. Comparison of the filamentary crystals of the three pyrite generations (1, 2, and 3) associated with calcite 2 (Ca).

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Pyrite 1</th>
<th>Pyrite 2</th>
<th>Pyrite 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Position</td>
<td>Free on crusts of quartz or included within Ca</td>
<td>Always in distinct internal zone within Ca</td>
<td>In the peripheral zone of Ca</td>
</tr>
<tr>
<td>Habit</td>
<td>Straight columnar or with 90°-kinks</td>
<td>Single crystal tortuous filaments</td>
<td>Cubo-octahedral [100]=[111], elongated</td>
</tr>
<tr>
<td>Crystal elongation</td>
<td>// [001]</td>
<td>Roughly // [001], with deviations</td>
<td>Slightly elongated // [111] or [001]</td>
</tr>
<tr>
<td>Bounding; crystal faces</td>
<td>Smooth [100] ± [210], [111])</td>
<td>Striated [100] ± [210] and [111]</td>
<td>Rough [111] ± [100]</td>
</tr>
<tr>
<td>Crystal seeds</td>
<td>Small cubic crystals</td>
<td>Small complex crystals</td>
<td>Small cubo-octahedral crystals</td>
</tr>
<tr>
<td>Length</td>
<td>2-3 mm and more</td>
<td>Up to 1-2 mm</td>
<td>0.1 mm</td>
</tr>
<tr>
<td>Thickness</td>
<td>Permanant: 20-50 µm</td>
<td>Changeable: 3-20 µm</td>
<td>Up to 10-20 µm</td>
</tr>
<tr>
<td>Substrate</td>
<td>Quartz or Ca: [10T1]</td>
<td>Ca: [3145]</td>
<td>Ca: [2151] and [0332]</td>
</tr>
<tr>
<td>Orientation on the substrate</td>
<td>Random</td>
<td>Roughly ± 10-20°</td>
<td>Nearly ⊥</td>
</tr>
<tr>
<td>Distribution and density of crystals</td>
<td>Uneven dispersed with low density</td>
<td>Even distribution with high density: up to 50/cm²</td>
<td>High density: up to 100-120/cm²</td>
</tr>
<tr>
<td>Growth mechanism</td>
<td>Fast whisker growth and sub-sequent thickening</td>
<td>Contemporaneous growth with the enclosing Ca</td>
<td>Contemporaneous growth with the enclosing Ca</td>
</tr>
</tbody>
</table>

The crystals of the next calcite 3 generation form autoepitaxic sceptre overgrowths on the terminal parts of the large scalenohedral crystals of calcite 2 (Fig. 2). They are bounded by the prismatic faces m(10[10]) and a(1120), and the scalenohedron e(01T2).

The pyrite crystals belong to three successive generations which are related to three growth zones observed within the crystals of calcite 2. Their characteristics and relationships with calcite crystals are shown in Fig. 2b and Tables 1 and 2. According to microprobe analysis pyrite crystals have nearly stoichiometric composition, with some small (<0.5 wt.%) Co content: S 53.49, Fe 45.75, Co 0.46 (Ni, As <0.2), Σ 99.70 wt. %, and formula (Fe0.99Co0.01)S2.01. No measurable compositional differences between crystals belonging to the three pyrite generations have been detected.

Pyrite 1 appears as small, up to 1 mm in size, isometric cubic a[100] crystals with minor truncating octahedral o[111] faces. Often, these crystals serve as basis for straight filaments strongly following [001] directions (Fig. 4). The
length of these thin columnar pyrite formations ranges from 1 to 2 mm, and their width ranges from 50 to 100 µm. They are bounded by flat cubic faces and have square or rectangular cross sections. In some cases, irregular stepped areas with oscillating \( \{210\} \) and \( \{111\} \) faces form parts of their terminations or side faces (Fig. 5). Right angle bends are not uncommon. L-, T- and more complex frame-like skeletal shapes occur (Fig. 5a, d). The cubic surfaces have a fine-layered structure with slightly rounded outlines nearly parallel to the crystal edges. The thickness of the layers is smaller than 1 µm (Fig. 5b, c). Crystals of pyrite 1 can be found on two different locations either scattered over the fine-grained quartz crusts (Fig. 4) or embedded as inclusions in calcite 2 crystals on top of an internal phantom rhombohedral \( \{101\} \) surface (Figs. 2b and 6a). In both cases the orientation of the crystals is random and evidently they formed under similar crystallisation conditions.

Pyrite 2, the second generation of pyrite crystals, consists of numerous thin tortuous filaments embedded in crystals of calcite 2 forming sub-parallel groups (Fig. 6a-b and 7). These filaments nucleate and grow roughly perpendicular to the faces of an internal phantom scalenohedron \( \{3145\} \) of calcite until they reach the outer crystal surface (Fig. 2b). The filaments are not perfectly straight and show serrated surfaces and variable thickness with changes of growth direction. In some cases they look like a chain of small sub-crystals with crystallographic coherence (Fig. 7 and 8). As established by morphological, goniometric and X-ray studies the filaments are single crystals elongated along [001] direction. Their surfaces are strongly striated due either to
the oscillation of the thickness of the filament or to the change of the displayed crystal forms. Thus, cubic $a\{100\}$ together with $e\{210\}$, $o\{111\}$, and sometimes $d\{110\}$ and $s\{321\}$ forms are present. The striation is usually uniform (e.g. transverse on Fig. 8a-d and 9), but local changes in its orientation due to unequal development of additional faces sometimes also occur. Some crystal segments have clearly irregular or distorted octahedral $o\{111\}$ habit (Fig. 7 and 9a-b). Filaments formed by prevailing $a$ faces have the shape of an $[001]$ elongated prism (Fig. 9c). Complex branched forms also occur (Fig. 8e). The length of the filaments reaches up to 1-2 mm, while their thickness varies from 2-3 to 20 µm. The thinner section of the filaments corresponds to the connection between two sub-crystals. Interestingly, there is a correlation between the thicknesses of neighbouring filaments (Fig. 7, 8a-d). The density of filaments in some places reaches up to 50 filaments per mm². Neither epitaxic nor topotaxic relationships between calcite and pyrite exist.

**Pyrite 3**, the last generation of pyrite crystals, occurs within a narrow (<0.5 mm thick) peripheral growth zone below the scalenohedral $\{2131\}$ surface of the main calcite 2 crystals (Fig. 2). This zone is rather enriched of pyrite inclu-
sions in relation to all other internal calcite zones (Fig. 6a-b). Pyrite crystal terminations projecting on the calcite surface have cubic-octahedral habit and rather rough faces (Fig. 10a-b). The elongated parts of these crystals inside calcite, as revealed after slight surface dissolution (Fig. 10c) are heavily striated, mostly by growth rate oscillation. The crystals of pyrite 3 are similar to the filaments of pyrite 2 but are shorter and more monolithic, often elongated also in $[111]$ direction. The crystals are up to 10-15 µm wide and three to four times larger in length but much shorter than those of pyrite 2.

### Discussion

The three generations of pyrite obviously correspond to three nucleation events, which are associated with three growth zones of calcite 2 (Table 2). We did not find fluid inclu-
sions in the calcite crystals and therefore we do not have direct evidence of the $T$ at which the mineralisation took place. However, the fluid inclusions data of Naidenova (1966) for scalenohedral calcite crystals from the same de-
posit (i.e. calcite 2) established that their crystallisation from the hydrothermal fluids occur at a temperature between 235 and 175°C. For the prismatic and the other following habit types the temperatures are still lower, 170-140°C. This range of temperature discards a biologically induced precipitation mechanism which has been sug-

<table>
<thead>
<tr>
<th>Minerals</th>
<th>Habit</th>
<th>Succession</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chalcopyrite</td>
<td>base</td>
<td>++</td>
</tr>
<tr>
<td>Calcite 1</td>
<td>scalenohedral ${21\overline{3}1}$</td>
<td>--</td>
</tr>
<tr>
<td>Quartz, fine-crystalline</td>
<td>prismatic ${10\overline{1}1}$ + ${0\overline{1}1}$ + ${10\overline{1}0}$</td>
<td>=</td>
</tr>
<tr>
<td>Calcite 2, zoned crystals</td>
<td>rhombohedral ${10\overline{1}1}$ → scalenohedral ${314}$ → scalenohedral ${21\overline{3}1}$ + ${03\overline{3}2}$</td>
<td>--</td>
</tr>
<tr>
<td>Pyrite 1</td>
<td>isometric ${100}$, and columnar along ${100}$ and ${001}$</td>
<td>--</td>
</tr>
<tr>
<td>Pyrite 2</td>
<td>filamentary $\approx [001]$ elongated</td>
<td>=</td>
</tr>
<tr>
<td>Pyrite 3</td>
<td>isometric ${11}$+$(100)$, elongated $\approx [111]$ or $[001]$</td>
<td>=</td>
</tr>
<tr>
<td>Calcite 3</td>
<td>prismatic ${10\overline{1}0}$ + ${11\overline{2}0}$ + ${01\overline{1}2}$</td>
<td>--</td>
</tr>
</tbody>
</table>

---

*free growth (calcite, pyrite)*

*contemporaneous growth*
gested by Hofmann & Farmer (2000) for filamentous fabrics of some low-temperature minerals, including pyrite.

As evidenced by experimental studies (Shoonen & Barnes, 1991; Kozerenko et al., 1995; etc.), nucleation of pyrite in such hydrothermal solutions, is embarrassed and it can only form initially through a monosulphide precursor. In the presence of sulphur species (H$_2$S, etc.) this is a fast process. The absence of marcasite (an indicator for acid conditions) in the samples studied is evidence of close to neutral or alkaline conditions, representing the normal environment for carbonate precipitation. The deposition of scarce fine-grained hematite in the carbonate stage (Bogdanov, 1988) points to oxidised conditions.

The earliest straight prismatic [001] crystals of pyrite 1 grew from small isometric crystals which are bounded mainly by flat $a$ faces. They grew from solution in an open space with random orientation onto the crust of fine-grained quartz. They are also found inside calcite 2 crystals, on an internal rhombohedral phantom {10$ar{1}$1} surface, having the same morphology and random orientation at the support. These identical crystal characteristics are indicative of the same growth mechanism in an open space over the calcite.
Fig. 10. The superficial cubic-octahedral (a+o)c crystals of pyrite 3 on the outer scalenohedral {2131} faces of calcite 2: (a) light microscopy view at low magnification; (b-c) SEM views at higher magnification. The elongated inwards pyrite crystals observed in (c) are revealed after slight dissolution of calcite.

face in a period of cessation of its growth. Later, during the further growth of calcite, the embraced pyrite forms the first internal zone of solid pyrite inclusions.

The screw dislocation mechanism of formation of straight columnar crystals and whiskers from low-supersaturated solutions, as early proposed by several authors (e.g., Amelinckx, 1958, for NaCl) is accepted by many authors (Sunagawa, 1987). However, as Givargizov (1986) mentioned, our level of understanding of whisker growth from liquid phase is poor in comparison with vapour growth, where a proven VLS (vapour-liquid-solid) mechanism prevails.

Henderson & Francis (1989) hypothesised that the screw-dislocation mechanism is sufficient to explain both the acicular habit and the formation of right-angle bends observed in natural filiform pyrite. However, their conclusions are not supported by direct observation of dislocations. As regards the right-angle bends, a migration of dislocations or their bifurcation is even more hypothetical and unlikely.

The experimental works, known from the literature, established rather different growth conditions for pyrite whiskers. Yamada et al. (1979) studying the pyrite crystallisation by chemical vapour transport in Cl₂ gas have produced isometric polyhedral crystals under lower thermal gradients (e.g., 700 → 600°C), and blade-like crystals and whiskers under much higher gradients (700 → 400°C). These [001] pyrite whiskers, formed by two-dimensional growth at high supersaturation, have a high growth rate of 1 mm/day. Trufanov et al. (1986) also produced dendritic and hair-like whiskers of pyrite and chalocite from high-temperature chlorine solutions by abrupt, brief (in seconds) T and P decrease from 400-300°C down to 20°C.

The diverse morphology of pyrite crystals, grown at different hydrothermal conditions, was systematically studied by Murowchick & Barnes (1987). A hydrothermal-transport method at controlled T (range 450-250°C) and T gradients was used, followed by rapid (30 min) cooling of the glass-capsules before opening. As mentioned by these authors, the degree of supersaturation rapidly increased during the cooling period. Thus, transition from surface-controlled to diffusion-controlled growth was taking place, resulting in skeletal and dendritic forms and overgrowths. In one of the runs (7 250°C and low gradient of 0.35°C/cm) needle-like crystals were obtained. The authors presumed that their formation occurred at a very low degree of supersaturation by the screw-dislocation mechanism, but no direct evidence was given. It appears more realistic that the whiskers arose during the short cooling period by fast diffusion-controlled growth.

Bonev et al. (1985), in a detailed SEM and HVEM study on morphology and perfection of natural pyrite whiskers and thin platelets from hydrothermal Pb-Zn deposits, did not find direct support for dislocation growth mechanism. They concluded that whiskers as highly non-equilibrium crystals were formed by rapid growth under a diffusion-controlled regime from inhomogeneous highly supersaturated solutions by two-dimensional nucleation on the tip. In fact, the whiskers represent highly anisometric acicular skeletal crystals.

Thus, in the case of pyrite 1 described here, considering the available experimental data and analysis of Bonev (1990; Bonev et al. 1985), we assume that the growth process of this straight-linear pyrite is realised in two successive stages: (1) initial rapid directed growth of thin [001] whiskers at a diffusional regime and embarrassed nourishment, and (2) steady layer growth of the side faces of the whiskers and their uniform thickening, after overcoming the diffusional resistance of the surroundings. Some isolated single whiskers form kinks following the heterogeneity of the fluid environment.

Convincing natural cases of similar two-stage formation of columnar pyrite crystals by thickening of thin whisker leaders have been reported for Eureka, Pennsylvania, USA, by Henderson & Francis (1989, see their Fig. 19 and 20), and for Naica, Chihuahua, Mexico, by White (1973, his Fig. 4, 7 and 8). The apparent curvature of some pyrite columns, in the last case, is evidently inherited from the flexible bended primary thin whiskers.

Pyrite 2, with its peculiar filamentary shapes considerably differs from known morphologies of this mineral. Several of its characteristics and the relations with calcite are important for understanding its origin.

1) All filamentary pyrite 2 crystals are entirely enclosed in calcite and are not observed as free filaments out of it. In fact, only their terminal parts protrude on the calcite surface as small euhedral crystals (as seen on Fig. 10 for pyrite 3).
2) The pyrite filaments are located between an internal growth zone of calcite (a face of the scalenohedron w{3145}) and the outermost crystal faces (the scalenohe-
(2) The tortuous pyrite filaments have variable thickness, owing to the more autonomous growth of their individualised composing segments and the directions of the single filaments not being emphatically straight-linear and parallel (except when two adjacent filaments have a joint seed). However, a distinct correlation can be observed in neighbouring filaments between their thinnest and thickest parts (Fig. 7, 8a-d) when removed from the base at equal length from the base.

Such morphological characteristics and the control of the location, orientation and size of the pyrite filaments by the enclosing calcite clearly suggest the contemporary growth of calcite 2 and pyrite 2, and cannot be explained by sequential growth.

The most favourable orientation of pyrite can be considered to have occurred as a result of specific geometrical selection in the competition between the two growing phases in which the acicular crystals of pyrite 2 developed in their direction of fast growth [001] survive. Due to the restrictive influence of spreading around calcite growth layers, the unfavourably oriented pyrite crystal-nuclei hindered their growth (Fig. 8e). The changeable thickness of the needles is related to some fluctuations in the FeS$_2$ supersaturation in solutions, which slightly changed the ratio between calcite and pyrite growth rates.

Pyrite 3 in principle is similar to pyrite 2, being enclosed in calcite 2 and crystallising contemporaneously with it. The difference is that the new nucleation is accomplished only in a thin peripheral growth zone, closely below the outer crystal surface (Fig. 6a, b and 10). This is why the crystals of pyrite 3, also growing roughly perpendicular to this zone, are much shorter. These crystals have cubo-octahedral habit, and are elongated in [001] or in [111] direction. The large number of pyrite crystals in the peripheral calcite zone is indicative for a period of considerably increased supersaturation of FeS$_2$ phase. Of course, during this period the previously arisen acicular crystals of pyrite 2 continue their growth, although with some local thickening.

On the outer calcite surface the acicular inclinations of pyrite 2 and 3 are revealed as normal isometric crystals (Fig. 10a, b). No pyrite filaments protruding beyond the calcite faces occur. This is additional firm evidence for contemporaneous running and ending of growth of these two pyrite generations with calcite 2. The sceptre crystals of the later calcite 3, overgrowing the terminations of crystals of calcite 2 after some interruption of mineralisation, do not contain any pyrite inclusions.

The case of syngeneic thread-like pyrite inclusions in quartz, studied by Galuskin & Winiarski (1997), shows similar morphological features of the minerals. Other impressive cases are inclusions of cassiterite in fluorite (Lyakhov, 1966) and of cinnabar in calcite (Zhabin, 1986). Despite the differences in composition and structure, the similar characteristics of these assemblages are due to the contemporaneous growth at comparable quantitative relations of the mineral pairs.

Conclusions

- Two main types of pyrite crystal formations, clearly different in their morphological characteristics and growth mechanisms, were found: (1) straight columnar prismatic pyrite 1, formed by free growth in open space, and (2) chain-like and tortuous filaments (pyrite 2 and 3), formed as inclusions in calcite during their contemporaneous growth. No differences in the chemical composition of these pyrite crystals were established.
- It is assumed that the prismatic crystals of pyrite 1 are formed by subsequent thickening of thin whiskers, previously formed by fast 2D growth on the tip from highly supersaturated solutions under diffusion-controlled regime.
- The chain-like single crystals of pyrite 2 show characteristic features of contemporaneous growth with enclosing calcite: their axes are approximately perpendicular ($\pm 10-20^\circ$) to their starting growth zone in calcite; there is correlation in the variable thickness of neighbouring filaments; some needles are composed of more individualised crystal (e.g. octahedral) parts. Similar, but much shorter, are the crystals of pyrite 3, located only in the peripheral growth zone of calcite.
- The specific cross-orientation of the pyrite needles to the calcite growth zones is the result of specific geometrical selection between these two simultaneously growing crystal phases.

Acknowledgements: The authors are indebted to CSIC of Spain and the Bulgarian Academy of Sciences for the financial support and to D. Rickard, E. Libowitzky, E. Tillmanns and an anonymous referee for the useful and constructive comments.

References


Received 2 May 2005
Modified version received 30 May 2005
Accepted 9 June 2005