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Fungal quartz weathering and iron crystallite formation in an Alpine environment, Piz Alv, Switzerland

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Key words: Quartz weathering, iron crystallite, fungus, biodegradation, oxalate

ZUSAMMENFASSUNG

In der triassischen Hauptdolomit Formation der Unterostalpinen Berninadecke befindet sich in der Nähe des Piz Alv eine auffällig gelb angewitterte Quarzitlinse. Sie ist stark durchbohrt von Pilzhyphen der Klasse Basidiomycetes, die den Quarz aufgelöst haben und anschliessend von Eisenmineralien inkrustiert wurden. Die Hyphen erscheinen meistens entlang mechanisch schwacher Zonen des Quarzites. Gelegentlich findet man sie innerhalb einzelner Quarzkristalle, in die sie sich senkrecht zur Kristalloberfläche hineinbohrten, indem sie das SiO_2 auflösten. Die Pilzhyphen sind zwischen 3 und 9 µm im Durchmesser und bis zu 300 µm lang. Sie bestehen gewöhnlich aus einem hohlen inneren Teil und einem inkrustierten Rand.

Die Inkrustierungen bestehen hauptsächlich aus kugeligen Formen, die eine kettenähnliche Anordnung bilden. Diese Spherulite können aus verschieden geformten Kristallen bestehen, wobei nadelförmiger Goethit (FeOOH) weitaus am häufigsten vertreten ist. In kleineren Mengen tritt auch Hämatit (Fe_2O_3) auf, der aus kleinen Plättchen besteht, die rosettenförmig angeordnet sind. Diese Eisenphasen sind für die gelbe Farbe der Quarzitlinse verantwortlich.

Die Varietät und die Anordnung der Eisenspherulite entlang der Pilzhyphen zeigen eine starke Ähnlichkeit mit Kalziumoxalaten, die entlang von kultivierten Hyphen auftreten. Solche Kalziumoxalate bilden sich als Nebenprodukt nach der Ausscheidung von Oxalsäure durch die Pilze, wenn genügend Kalziumionen in der unmittelbaren Umgebung vorhanden sind. Auf Grund dieser Beobachtungen nehmen wir an, dass auch die Auflösung von Quarz und die Bildung von Eisenmineralen in Zusammenhang mit Oxalsäure steht, die durch die Pilze ausgeschieden wurde.

ABSTRACT

A quartzite lens occurring within the Triassic Hauptdolomite Formation of the Lower Austroalpine Bernina Nappe near Piz Alv, Switzerland, has been heavily eroded and altered by fungal dissolution of SiO_2 and subsequent precipitation of iron crystallites. Preliminary studies show that fungal hyphae of the class Basidiomycetes occur most abundantly along mechanically weak zones in the quartzite. Occasionally, hyphae occur within quartz crystals, into which they bored subperpendicularly from the quartz surface by dissolving SiO_2. The hyphae are approximately 3–9 µm in diameter and up to 300 µm long. They commonly consist of a hollow central part and an encrusted rim.

The encrustations consist mostly of spherical bodies forming a chain-like structure along the former hyphae. They show a variety of crystal structures and consist mainly of needle-shaped goethite (FeOOH) with lesser amounts of platy hematite (Fe_2O_3) arranged in rosettes. These iron phases account for the conspicuous yellow colour of the quartzite lens.

The variety and arrangement of the spherical iron crystallites along fungal hyphae show a striking similarity to calcium oxalate crystals arranged along laboratory grown fungal hyphae. Such calcium oxalates form as a by-product of fungal secretion of oxalic acid in the presence of calcium ions. Thus, we assume that quartz dissolution and iron crystallite formation in the investigated quartzites is associated with the presence of oxalic acid secreted by the fungal hyphae.

Introduction

In the 1950's, a small area of approximately 100 metres length and 50 metres width was discovered ~ 500 m NNE of the train station Bernina-Lagalb, Grisons, at an altitude of ~ 2300 m (Fig. 1) containing abundant conspicuously yellow debris (Fig. 2a). This loose debris derived from a quartzite lens which is covered by a humus layer a few centimetres thick and is intercalated in the upper Triassic Hauptdolomit Formation of the Austroalpine Bernina Nappe (see Schüpbach 1970; Schüpbach 1973). Siliceous lenses of quartzose chert are not uncommon in this formation, but do not show such an intense yellow colour in other locations. Preliminary studies show that this distinct colour is the product of

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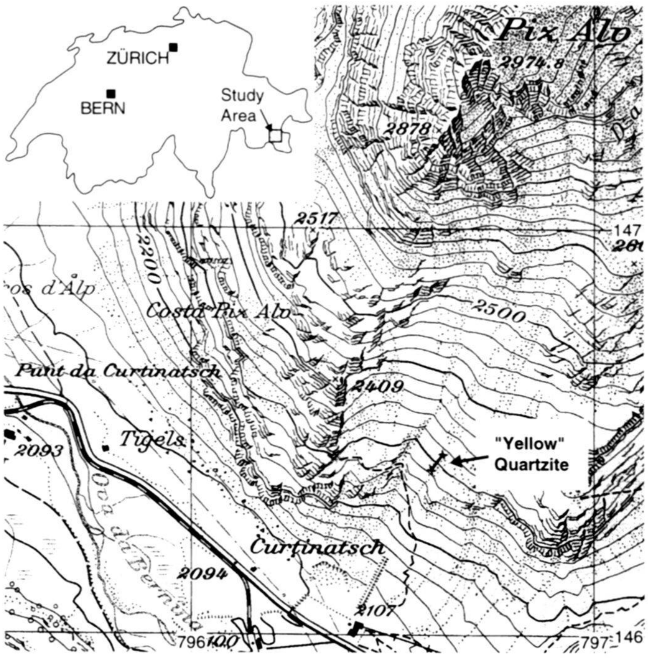

Fig. 1. Location of the "yellow" quartzite lens in the eastern Swiss Alps near Piz Alv. (Reproduced with permission of the BA für Landestopographie, CH-3084 Wabern, October 9, 1997)

continuous weathering of the quartzite by activity of fungi of the class Basidiomycetes (Fig. 2b), associated with the formation of iron oxide and oxihydrate.

Weathering of silicates by fungal activity has been observed in soils (Cromack et al. 1979) and associated iron crystallite formation has been described in studies of ancient rocks (Kretzschmar 1982). In this paper we illustrate a more recent example of fungal effects on quartz weathering associated with the formation of iron crystallites in an Alpine environment, which is buried under snow for up to 6 months per year during winter and spring.

Methods

Rock samples were mineralogically analyzed by X-ray diffraction (XRD) techniques on a Philips PW 1820 diffractometer using Söller slits and Cu-Kα radiation. Samples were air dried and coated with gold for SEM micrographs. SEM analysis was performed using a Cambridge Stereo Scan Mk II A coupled with an energy dispersive X-ray for element identification. Additional element identification was carried out using a Camera SX50 electron microprobe with a wave dispersive system (WDS) detection. Smear slides of powdered samples and thin sections were prepared for optical microscope analyses.

"Yellow quartzites"

The "yellow quartzites" generally have undulous lamination with laminae thicknesses of up to 1 cm (Fig. 3). Quartz crystal sizes typically range from 20 to 400 μm. Larger crystals are commonly hypidiomorphic in contrast to smaller crystals which appear to be xenomorphic. Individual laminae have an equigranular structure. Occasionally, quartz crystals have alternating yellow light and dark growth rims (Fig. 4). Within laminae, voids occur which are commonly filled with cryptocrystalline quartz (Fig. 5). These crystals are often associated with bulbous and rod-shaped structures (Fig. 6a–b). If the yellow quartzites occur as loose debris they contain abundant fossilized residues of fungal hyphae (Fig. 7). If the rock material is buried below a humus layer it is additionally colonized by a basidiomycete fungal mat. Structures of fungal hyphae typically occur along mechanically weak zones and within pore spaces (Fig. 8a–c). Individual hyphae often grew into quartz crystals (Fig. 9a–b; Feldmann 1997). Fungal hyphae are approximately 3–9 μm in diameter and up to 300 μm long. They commonly have a hollow centre part (approx. 1 μm in diameter), and an encrusted rim causing a yellow to brown appearance in thin-section (Fig. 10). Fungal structures commonly do not occur in voids filled with cryptocrystalline quartz (Fig. 11) and at the quartz-dolomite contact zone. There, quartz and dolomite crystals are often separated by a dark rim as observed in thin-section.

Quartz crystal faces are often heavily eroded and have distinct dissolution patterns, such as etch marks and borings (Fig. 12a–e). Borings oriented subperpendicularly to the crystal face were repeatedly observed (Fig. 13). In some cases, micro-reaction zones up to 5 μm across occur on the surfaces of quartz crystals (Fig. 14). Etched zones and the walls of borings are usually covered by a thin sheet of bulbous to needle-like structures (Fig. 14, 15).

XRD and microprobe analyses of encrusted fungal hyphae (Fig. 16a–c) show that the encrustations consist mostly of goethite (alpha-FeO(OH)) which accounts for the unique yellow colour of the quartzite (Fig. 2, 3). Other iron phases, such as hematite (Fe_2O_3), occur in lesser amounts with gradual transitions from goethite to hematite. Encrusted fungal hyphae often have chain-like structures consisting of spherical bodies (Fig. 17a–b) or appear to be amorphous (Fig. 18). The iron oxides and oxihydrates which repeatedly were observed in association with residues of organic material (Fig. 19a–b) have distinct forms. Goethite encrustations are typically needle-like (Fig. 20) whereas hematite appears to be platy (Fig. 21). Spherical goethite encrustations strongly resemble minute hedgehogs showing characteristic holes in their isodiametric bodies (Fig. 22) or radially-grown fan structures (Fig. 23a–b). Spheres consisting of hematite have minute rosette-arranged plates (Fig. 24). The diameters of the spheres are 2–4 μm. Spherulites of unknown composition were also found growing from hyphae of the basidiomycete mat (Fig. 25a–c).

Discussion

The origin of the particular quartzite lens forming the "yellow quartzites" near Piz Alv is not known. However, there is evidence that quartzite formation did not take place in association with the colonization of the fungal mats. This evidence includes, for instance, the occurrence of a petrographically similar quartzite lens in the same Hauptdolomite Formation near Piz Mezzaun (Schüpbach 1976) which does not contain fungal hyphae. The lack of fungal hyphae in the quartzite-dolomite contact zone and in the surrounding dolomite, as well as the existence of subperpendicular fungal borings in quartz crystals (Fig. 13), indicate that quartz formation occurred prior to the colonization by fungal mats. In addition, subvertical domino-style faults associated with subhorizontal tension fractures in the yellow quartzite (Fig. 8a), and the presence of a slight lineation in the quartzites from the Mezzaun area, indicate formation of the quartzite lens prior to the most recent deformation events affecting the Bernina Nappe. Thus, a common explanation suggests that circulating hydrothermal solutions supersaturated with respect to SiO_2 led to the formation of these quartzite lenses.

However, in thin-section samples from the quartz-dolomite contact zone, the degrading dolomites usually have a dark organic rim towards the newly forming quartz. SEM examination of such contact zones shows that there are colonies of a star-shaped, bacterial-like species which are only associated with the dolomite (Fig. 26a–b). The function of these microbes is not yet known, but they appear to be somehow associated with the degradation of the dolomite and/or the formation of quartz. A bacterially controlled, continuous replacement of dolomite by quartz seems possible. Although this observation does not provide conclusive evidence, a microbially induced origin of quartz, possibly associated with hydrothermal solutions, has to be reconsidered.

In the pH range of most natural waters, quartz dissolution is extremely slow (Bennett, 1991). However, in the presence of high concentrations of a few multifunctional organic acids, such as ascorbic, citric and oxalic acid quartz dissolution is accelerated and quartz solubilities are controlled by organisms (Silverman & Munoz 1970; Lauwers & Heinen 1974; Bennett 1991; Hiebert & Bennett 1992; Bavestrello et al. 1995). Boring and dissolution of the yellow quartzite can be explained by fungal secretion of oxalic acid (oxalate) which is considered to be an oxidation product resulting from the aerobic breakdown of carbohydrates that accumulate in fungus cultures (Foster 1949). It has been shown that basidiomycete fungal mats produce oxalic acid, which can accelerate quartz dissolution at near-neutral pH and low temperatures (Cromack et al. 1979; Chisholm et al. 1987; Bennett 1991; Arnott 1995; Franceschi & Loewus 1995). Through interactions with Al and Fe, oxalate plays a major role in plant nutrition by increasing the availability of P, K, Mg, and Ca in soils (Graustein et al. 1977; Cromack et al. 1979; Allison et al. 1995). Thus, we suggest that the borings observed in the quartz crystals of the yellow quartzite are the product of fungal oxalate production which dissolves SiO_2. However, it remains speculative whether the borings observed in the quartz crystals are simply a by-product of the oxalate, which has been secreted in order to increase the availability of nutrients for the symbiontic host plants, such as *Dryas octopetala* L. (Fig. 2a).

Although most of the quartz crystals are weathered by fungi there is evidence that quartz formation is a subordinate process. Cryptocrystalline quartz filling voids, for instance, is often not attacked by fungi (Fig. 11), indicating that quartz formation occurs after or independent of fungal colonization. Minute quartz crystals found in pore spaces appear to have formed in association with organic material as shown by their arrangement which resembles organic strings (Fig. 27). Unaltered rims of quartz crystals (Fig. 4) are also evidence for continuous quartz formation.

It appears that, at least partly, the formation of quartz is associated with bacterial activity as indicated by a number of minute rounded and rod-shaped bodies of less than 1 μm in diameter (Fig. 6a–b) found in the presence of newly formed quartz. However, it is not known whether this present quartz formation is associated with circulating supersaturated fluids or the contemporaneous dissolution of diagenetic quartz, which might lead to supersaturation with respect to SiO_2.

In fungi, most of the oxalic acid appears to be secreted during growth of the mycelia. The secreted oxalate may precipitate as crystals of calcium oxalate within or on the surface of cell walls (Arnott 1995; Franceschi & Loewus 1995). Unfortunately, microprobe and XRD analyses of rock material did not provide any evidence confirming the presence of Ca-oxalates and biological studies have not yet been carried out to demonstrate that the basidiomycete fungal mats from Piz Alv produce calcium oxalate. However, some fresh fungal hyphae clearly have bulbous structures (Fig. 25a–c) which strongly resemble Ca-spherulites from Messinian sediments (Franz et al. 1978) and calcium oxalates produced by laboratory culturing (Arnott 1995). Although these structures were not identified chemically, they indicate that, at least during an early stage of fungal growth, calcium oxalate might have been produced. The absence of Ca-oxalate in the investigated rock material may be due to the dissolution of Ca-oxalate in the presence of Fe^{3+}. Under favoured conditions, oxalate will react with this trivalent ion leading to an enrichment in iron crystallites, such as goethite and/or hematite, in the solid phase (Graustein 1976; Graustein et al. 1977). The spherical iron crystallites with their hollow centres (Fig. 22, 24) might have originated around Ca-oxalate which was continuously dissolved as the Fe^{3+} content increased in the circulating solution.

Although the presence of bacteria was not clearly recognized, there exist etch marks on quartz surfaces (i.e. Fig. 14), the origin of which possibly needs an explanation other than fungal activity. Similar features were also noted by Kretzschmar (1982) who interpreted them as "solution structures, presumably with roundish bacteria encrusted with iron". Hiebert and Bennet (1992) postulate for such features "that surfaceadhering bacteria created a reaction zone in their immediate vicinity in which organic acids, produced within the cell and released extracellularly, were concentrated at the cell solution-mineral interface. A gradient in chemical potential was produced between the cell surface and the surrounding bulk fluid. Within this microenvironment, complex organic acids chelated SiO_2 at the quartz surface and dissolved the mineral even though the bulk pore water was supersaturated with respect to the dissolving mineral". However, another explanation for such etched spots might simply be given by the reaction of the quartz crystal surface with small amounts of oxalic acid separated from the fungal hyphae.

Conclusion

The microbial dissolution of quartz (i.e. Lauwers & Heinen 1974; Bennett 1991; Hiebert & Bennett 1992) as well as the microbially induced precipitation of iron crystallites (i.e. Beveridge & Fyfe 1985; Ferris et al. 1986, 1987) are well known processes. However, the combination of both seems rarely to occur. Thus, only one fossil example from the Rhenish Massif (Kretzschmar 1982) is known to the authors. We suggest that quartz dissolution and iron crystallite formation near Piz Alv is associated with fungal production of oxalic acid which reacts stepwise with Fe^{3+} to form Fe-crystallites around organic hyphae and previously produced and dissolved Ca-oxalates. In this manner fungal hyphae are fossilized and preserved.

Although many factors concerning the formation of the quartzite lens near Piz Alv and its biodegradation by fungi are unknown, the yellow quartzite demonstrates the importance of microbial silicate weathering in an Alpine region. In contrast to tropical regions, where most biochemical processes are accelerated by elevated temperatures, the example from Piz Alv suggests that such processes can also take place to a significant degree under Alpine climate conditions.

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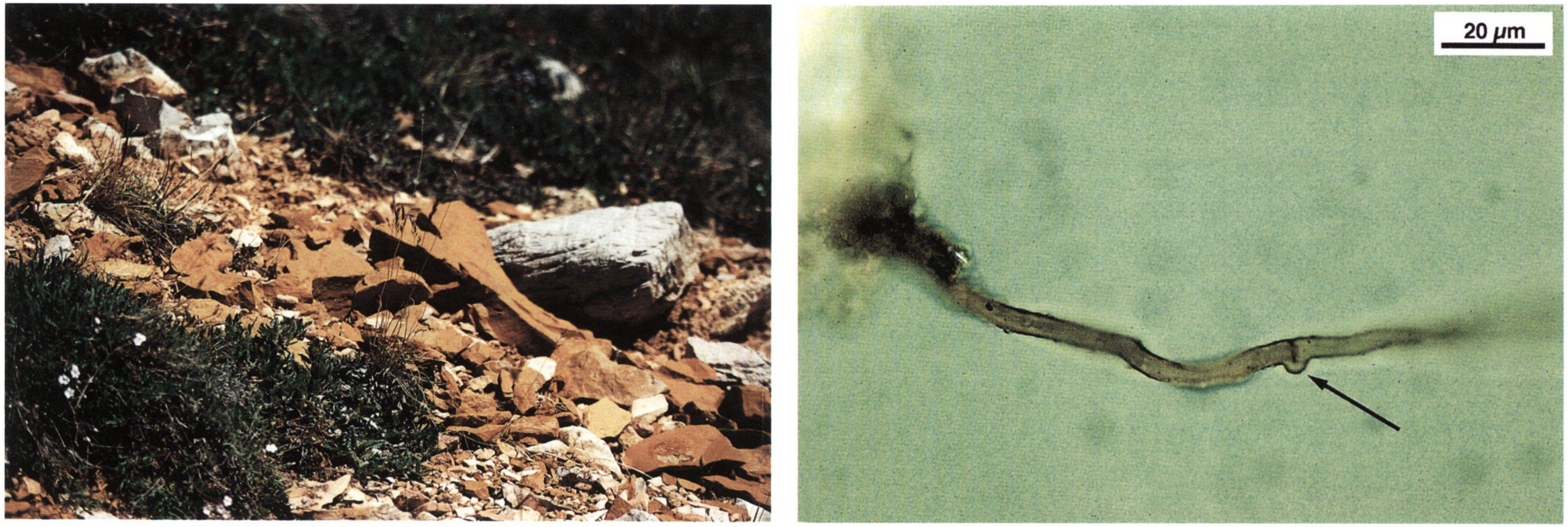


2a 2b

Fig. 2. Features associated with the "yellow" quartzite. **a)** The quartzite is mostly found as conspicuous yellow debris in an area of about 100 metres length and 50 metres width. Note the green plants of *Dryas octopetala* L. (foreground centre) a common plant within the yellow debris forming ectomycorrhizae with fungi of the class Basidiomycetes. The length of the grey dolomitic rock (centre right) is about 20 cm. **b)** Photomicrograph showing a fungal hypha of the class Basidiomycetes with its characteristic clamp connection (arrow). Such fungi alter the quartzite by dissolving SiO_2 with associated formation of iron oxide and oxihydrate.

Fig. 3. Cross-section of a quartzite hand sample showing the undulously laminated structure. Laminae have thicknesses up to 1 cm.

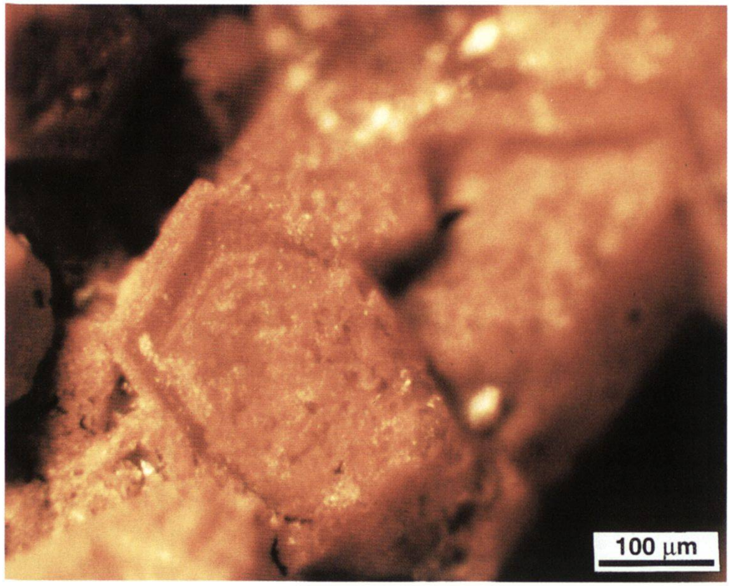


Fig. 4. Photomicrograph of a quartz crystal with several generations of growth rims. The light yellow parts contain organic encrustations.

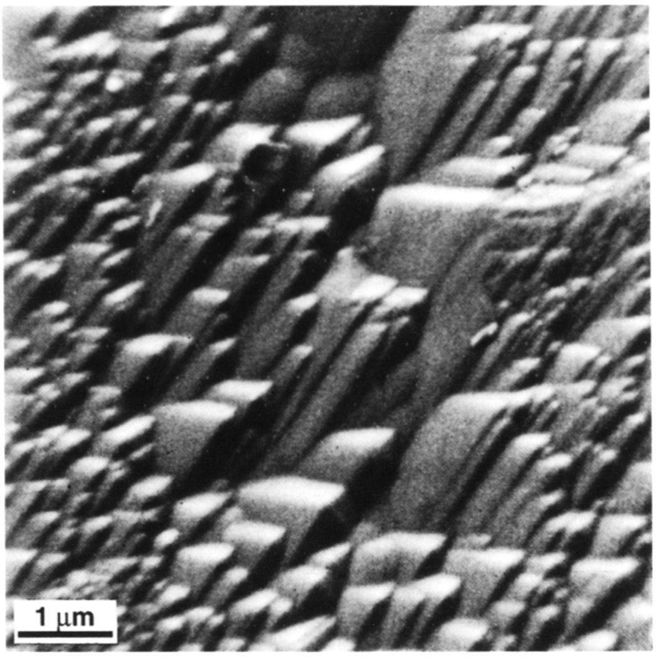


Fig. 5. SEM photomicrograph showing the complex rough surface of a void filling quartz.

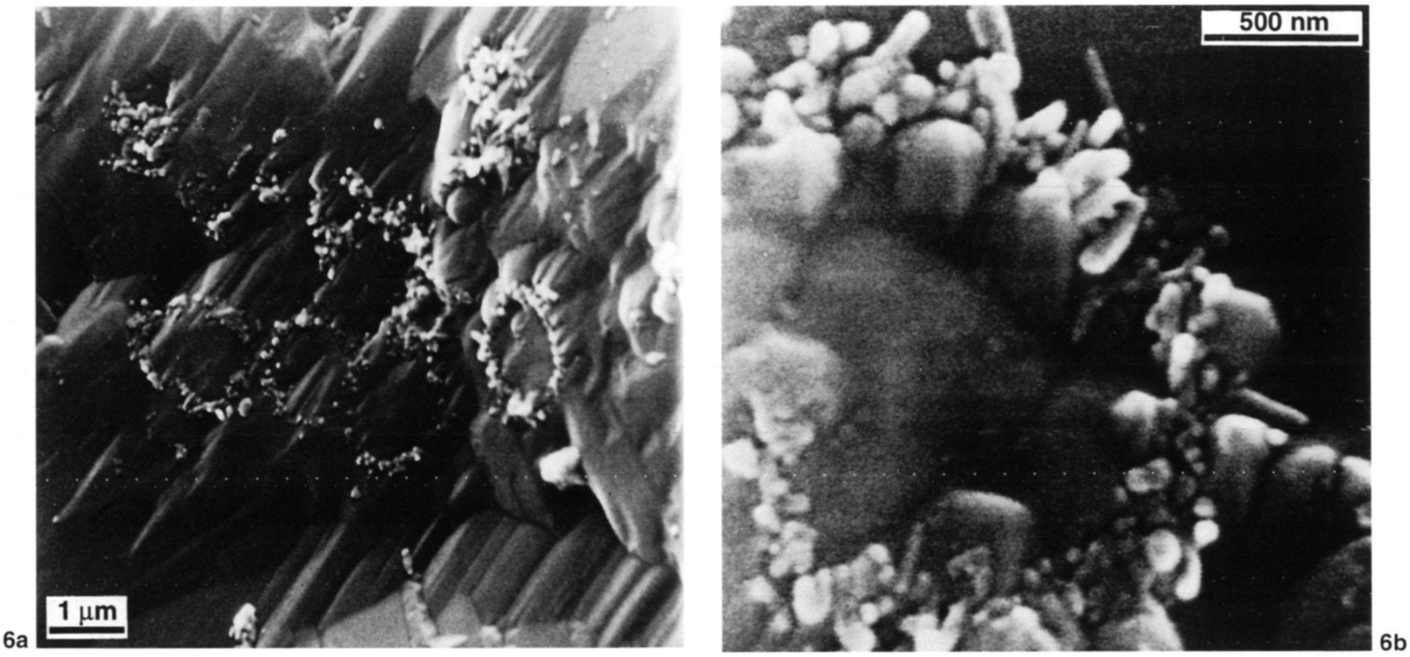


Fig. 6. SEM photomicrographs showing a possible microbial effect in quartz growth. **a)** Ring-like structures most-likely of microbial origin are often associated with rough quartz surfaces. **b)** Close up of a "microbial" ring consisting of bulbous and rod-shaped structures which strongly resemble bacteria. It appears that quartz growth is associated with these microbial structures.

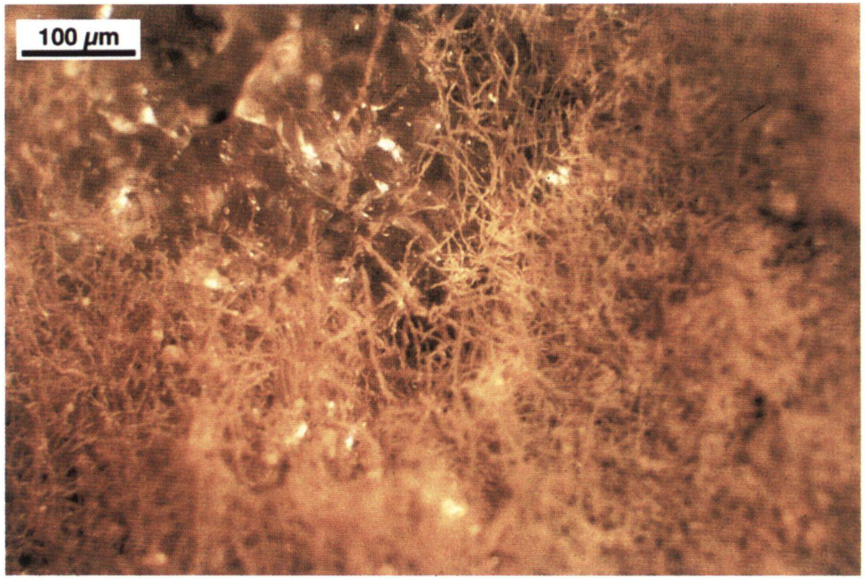


Fig. 7. Photomicrograph showing a colony of mineralized fungal hyphae of the class Basidiomycetes in quartz.

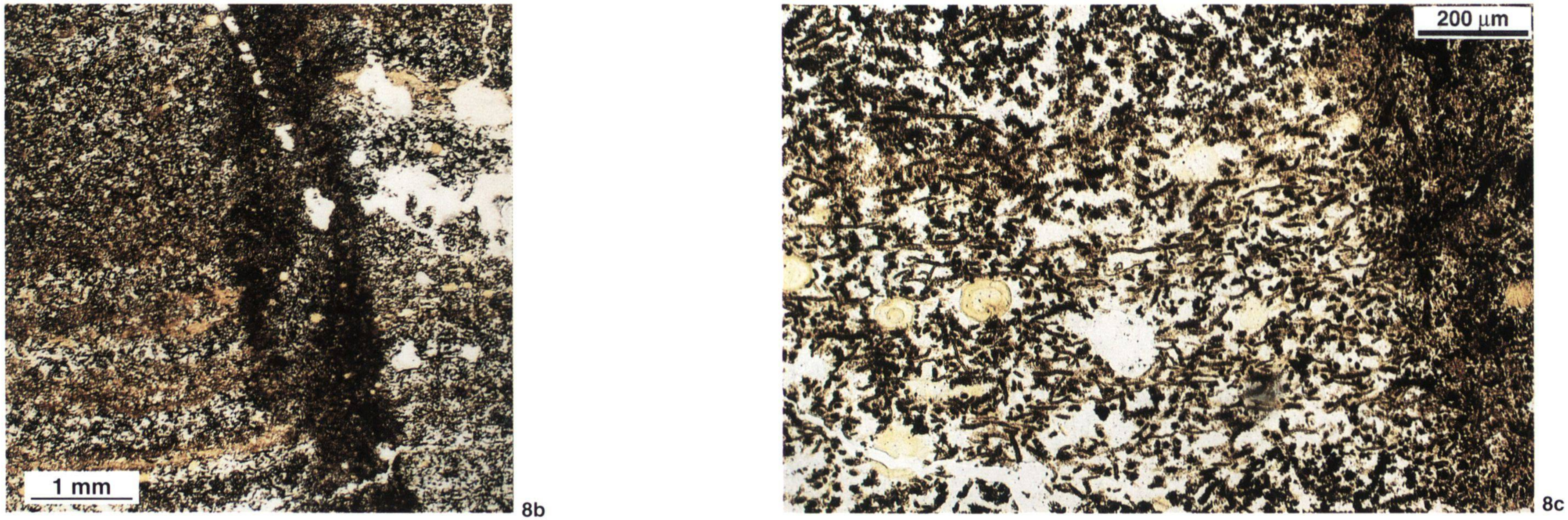
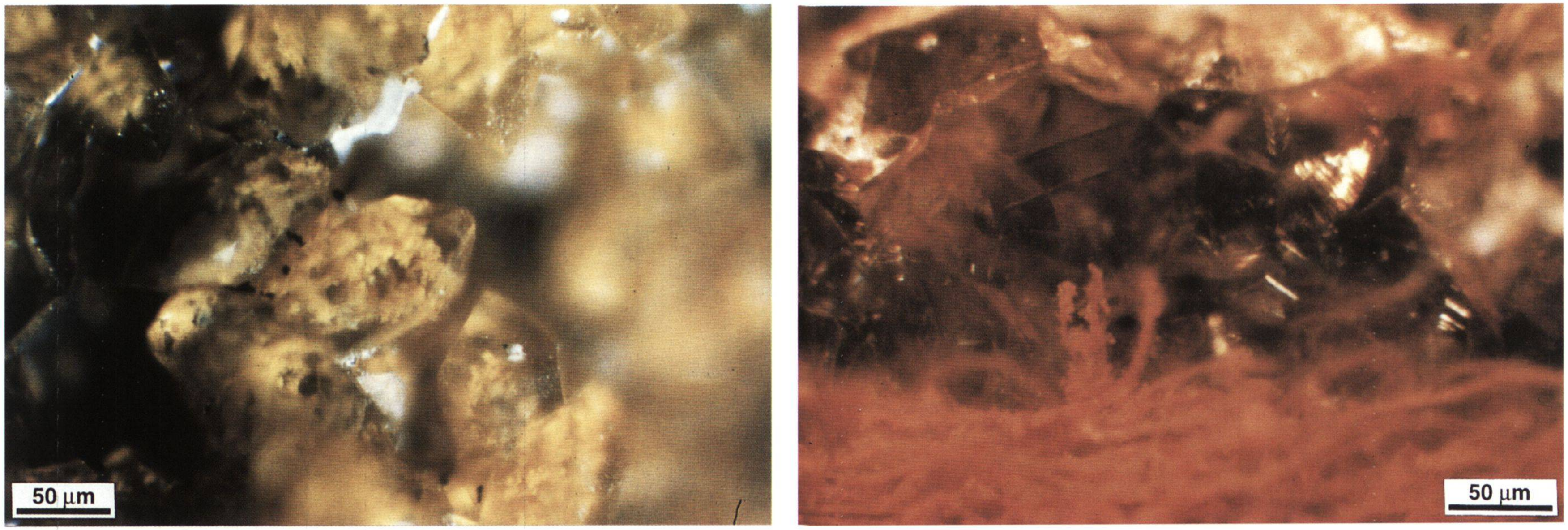


Fig. 8. Occurrences of filamentous structures. **a)** Cross-section of a quartzite hand sample showing subvertical faults separating tilted domino-style fault-blocks. Fungal hyphae preferentially occur along such fracture zones. Scale bar unit = 1 mm. **b)** Thin-section photomicrograph of a fracture zone of sample shown in Fig. 8a. The dark colour of the vertical fracture and the yellow colour of the horizontal fractures are the result of different concentrations of fungal hyphae. **c)** Thin-section photomicrograph showing the parallel orientation of fungal hyphae in horizontal fracture zones.



9a 9b

Fig. 9. Photomicrographs showing fossilized fungal hyphae in quartz. **a)** Quartz crystals with fuzzy hyphae. **b)** Three hyphae which grew perpendicularly into the quartz from a parallel oriented fungal mat.

Fig. 10. Thin-section photomicrograph showing several generations of mineralized fungal hyphae in quartz. Older hyphae appear darker, being more mineralized, whereas younger hyphae consist of a hollow internal part and an encrusted rim.

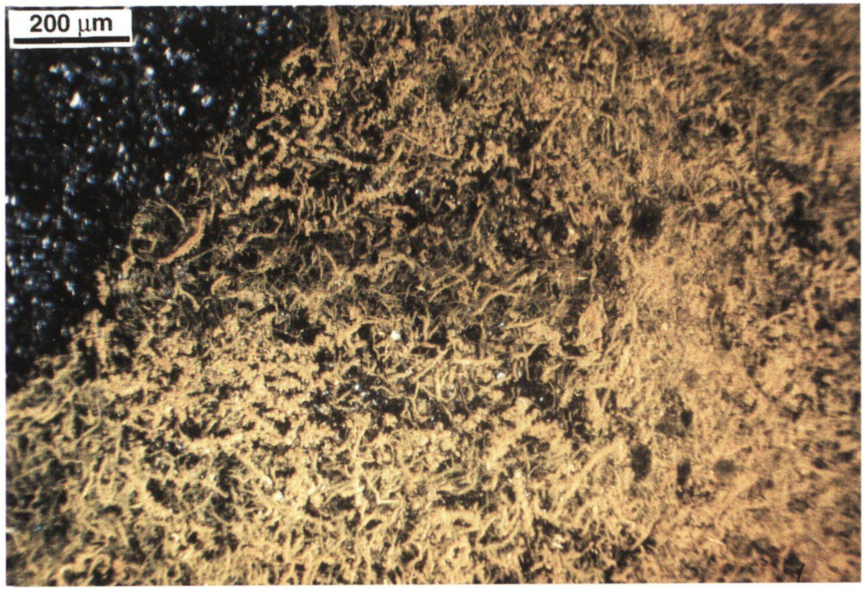
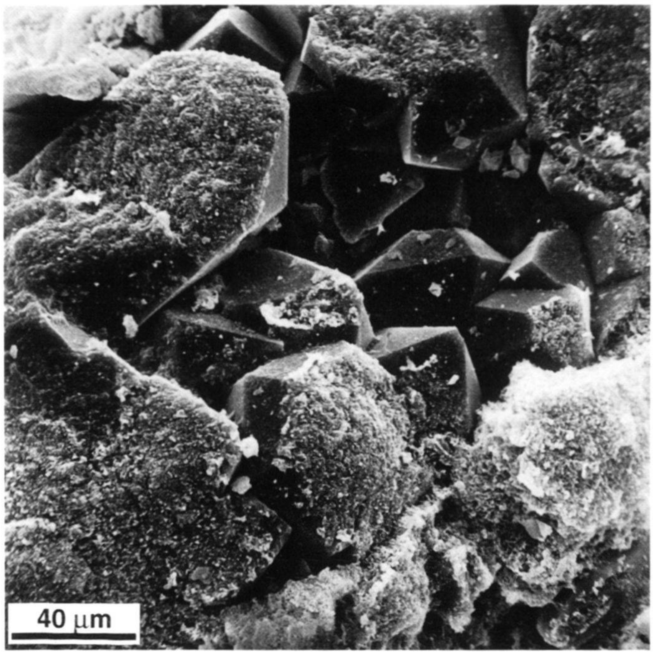


Fig. 11. Thin-section photomicrograph showing abundant fossilized fungal hyphae (yellow) in quartz. Void filling cryptocrystalline quartz (grey) is mostly not colonized by fungi.



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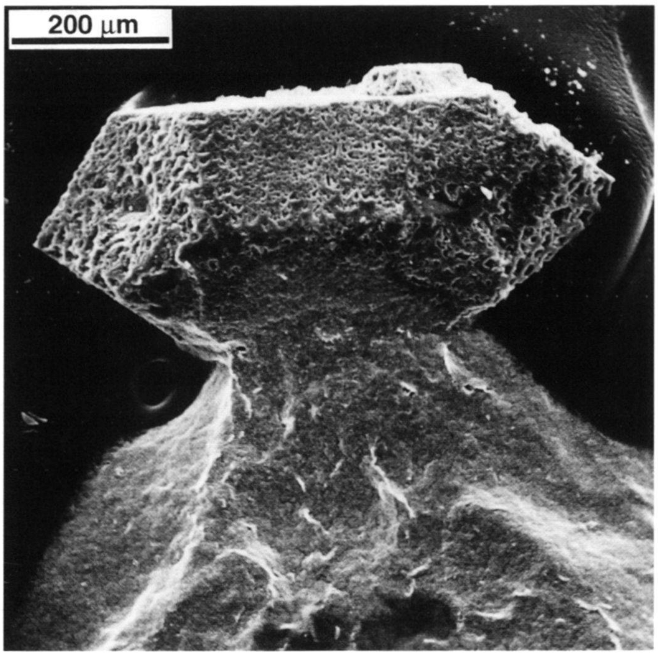

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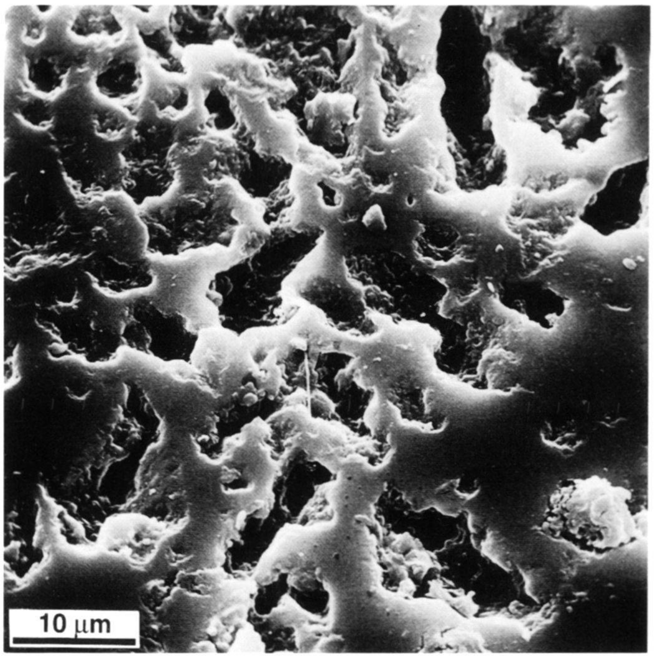

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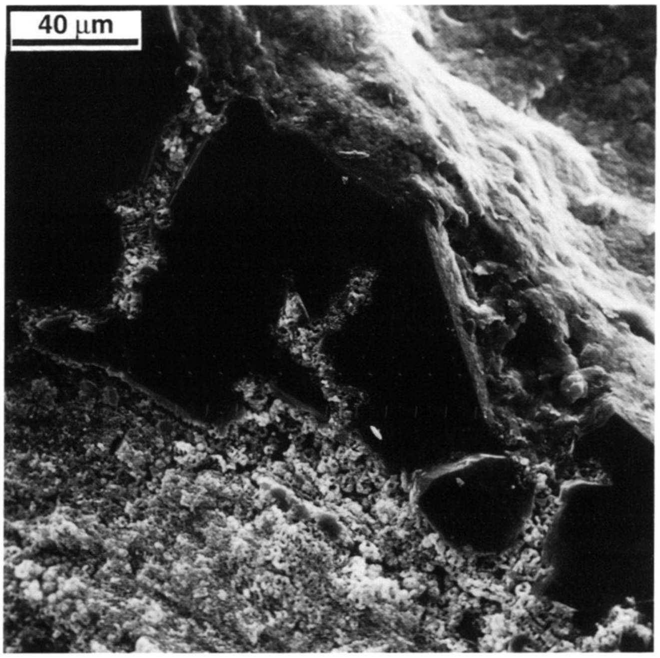
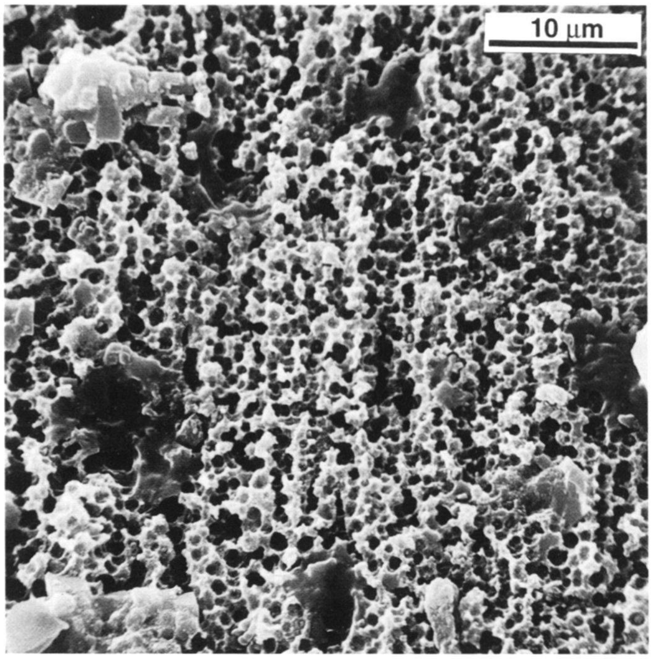


12d

Fig. 12. SEM photomicrographs showing microbial effects in quartz erosion. **a)** Heavily eroded surfaces of vein quartz crystals. **b)** Faces of individual quartz crystals which are covered by microbial borings. **c)** Close-up of Fig. 12b showing the irregular structure of the borings. **d)** Erosional pattern of a parallel oriented fungal colony in quartz (lower right) and an individual fungal boring into the quartz crystal (centre). **e)** Close-up of Fig. 12d showing the parallel oriented erosional pattern of a fungal colony.



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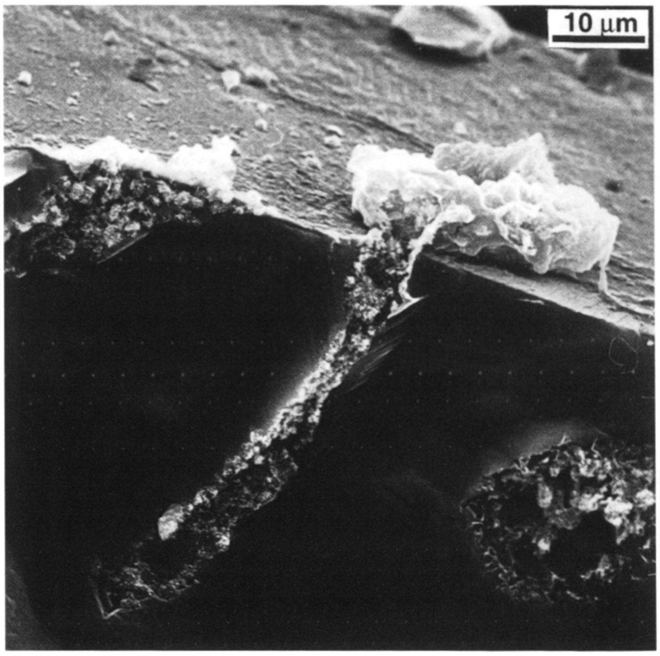

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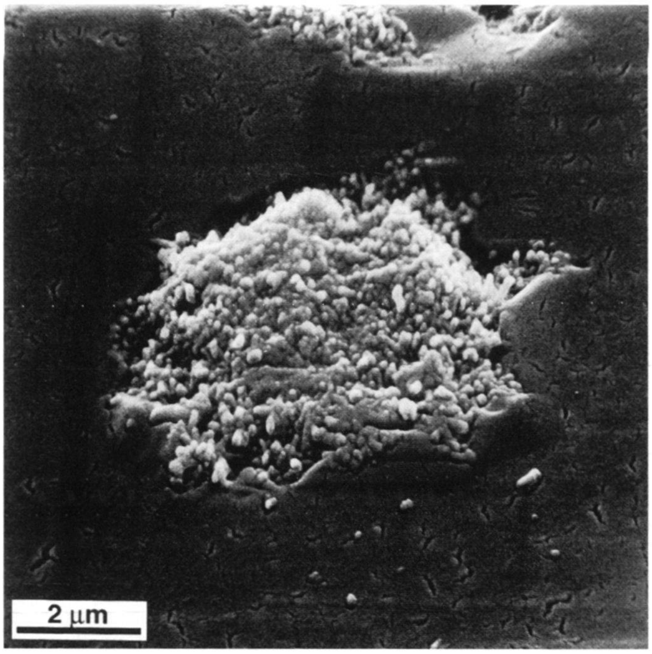

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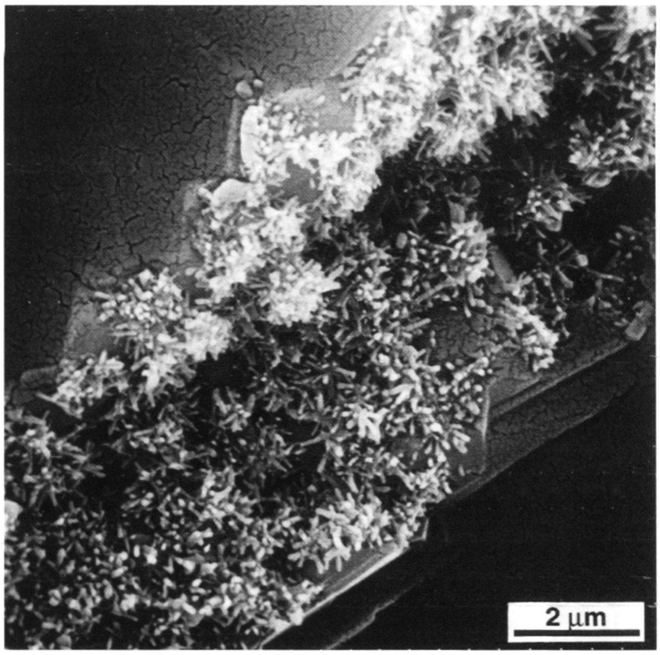

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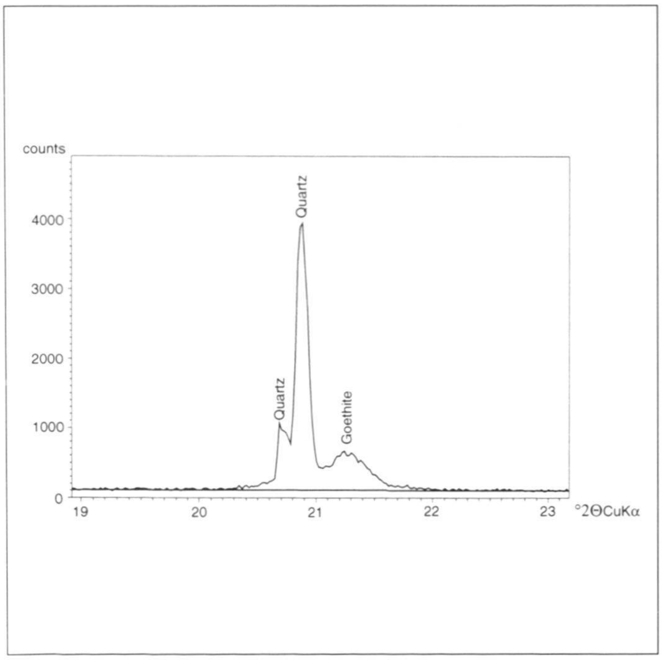
Fig. 13. SEM photomicrograph of the face of a quartz crystal with a subvertical boring.

Fig. 14. SEM photomicrograph of a micro-reaction zone covered by bulbous structures on the surface of a quartz crystal. It is not known whether such etch marks are the product of fungally secreted oxalate or bacterial activity.

Fig. 15. SEM photomicrograph showing a close-up of a fungal boring. The walls of such borings are generally covered by needle-like iron oxihydrate.



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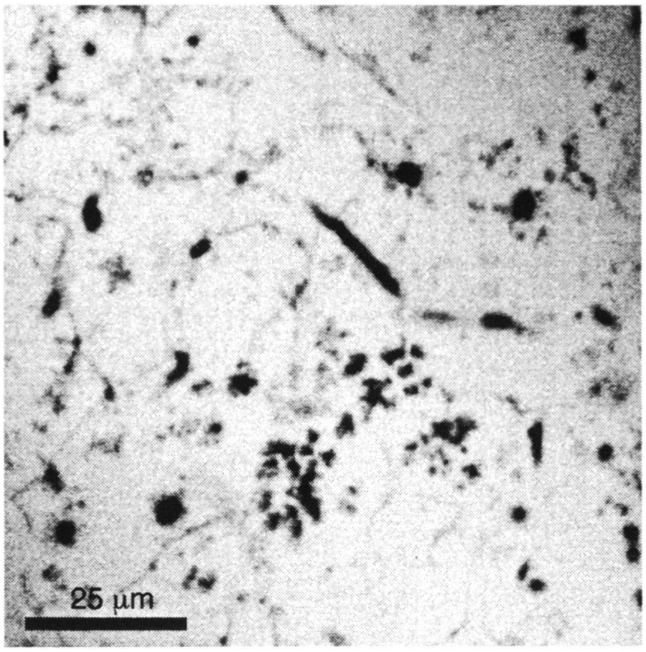

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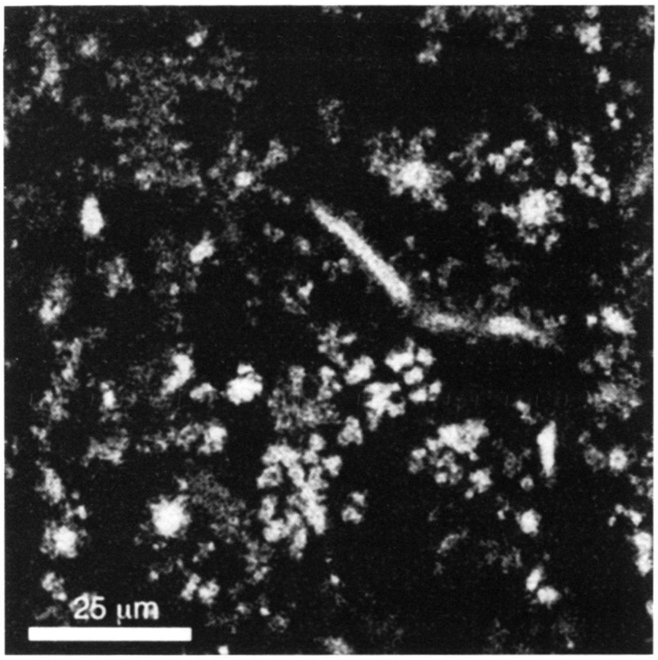
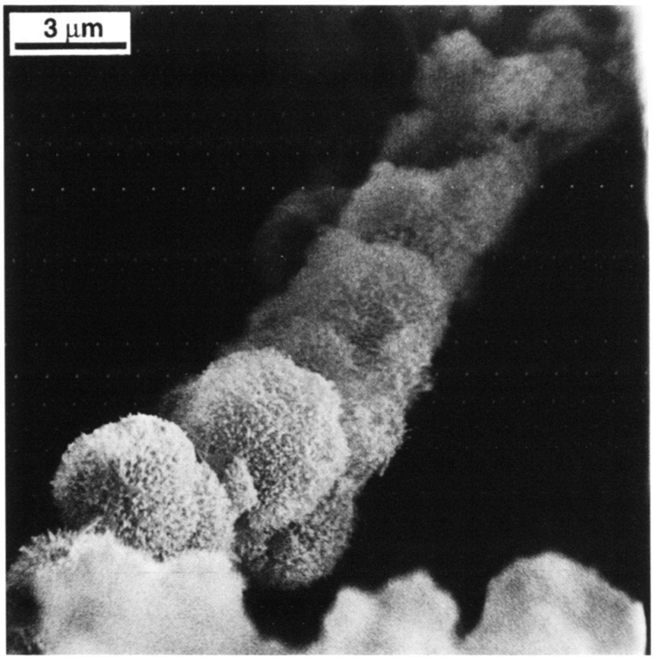


16c

Fig. 16. Chemical analyses of "yellow quartzite". **a)** XRD plot of a bulk sample showing that the rock material essentially consists of quartz and goethite. **b)** Electron microprobe elemental distribution of Si shows that the host rock consists of quartz and **c)** the mineralized fungal hyphae of the iron-phase.



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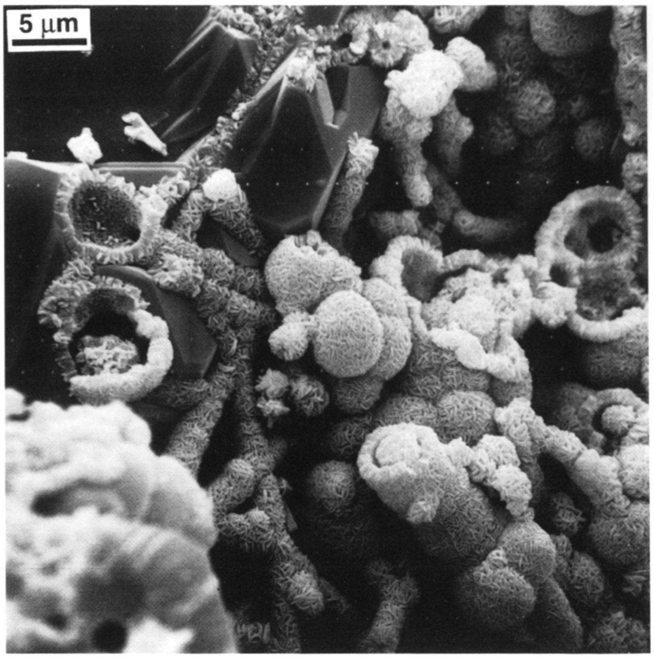

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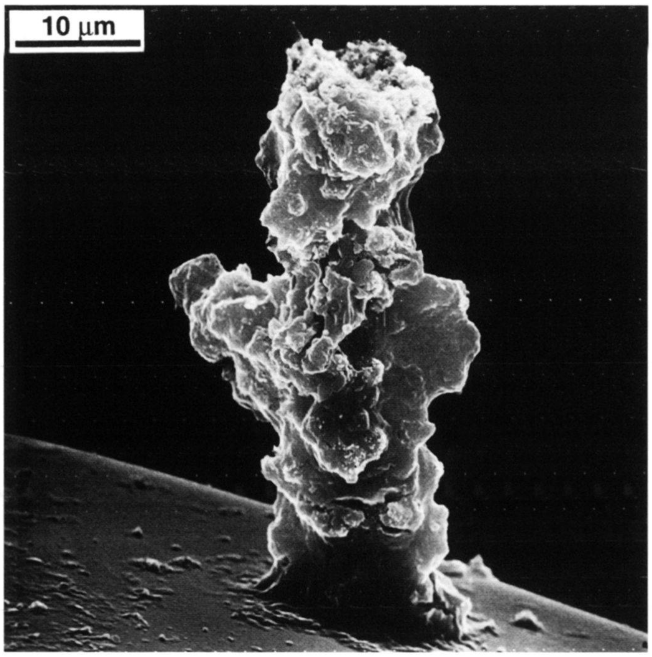

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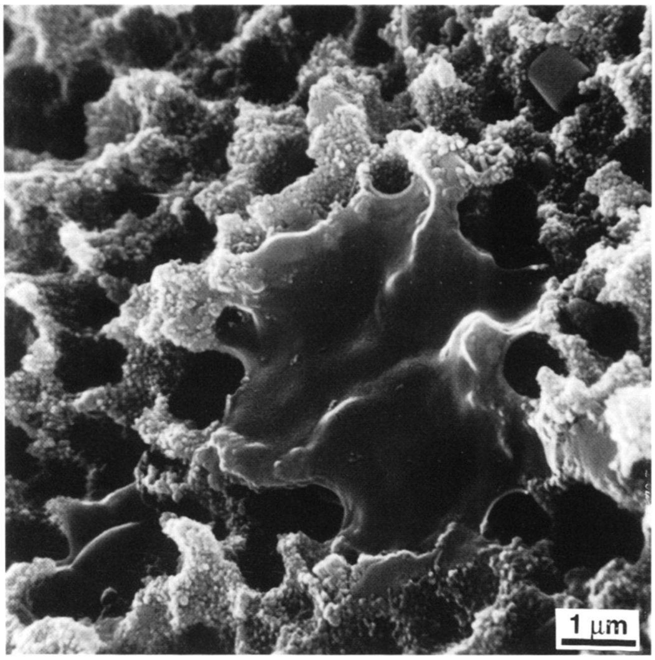
Fig. 17. SEM photomicrographs of encrusted fungal hyphae with chain-like structures. **a)** Individual hypha consisting of needle-like goethite. **b)** Fossilized colony of fungal hyphae consisting of platy hematite.

Fig. 18. SEM photomicrograph of an amorphous-appearing encrustation. The fungal hypha has grown vertically on a quartz crystal face.



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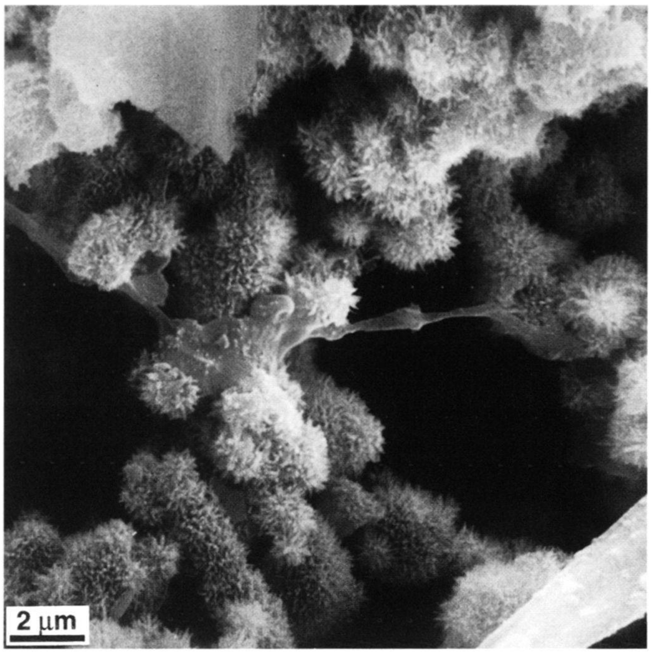

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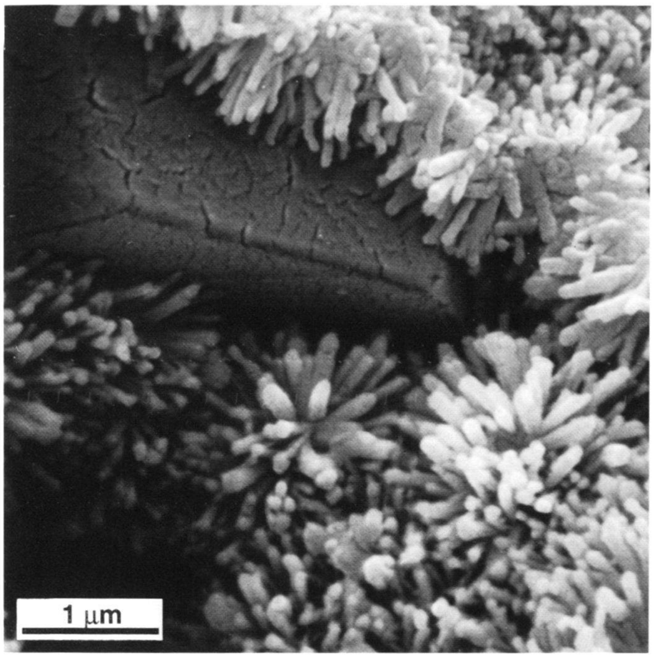

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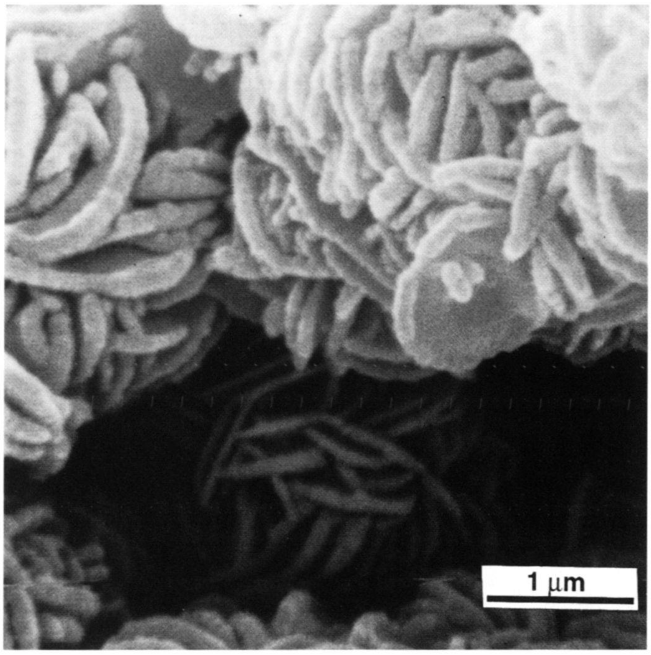

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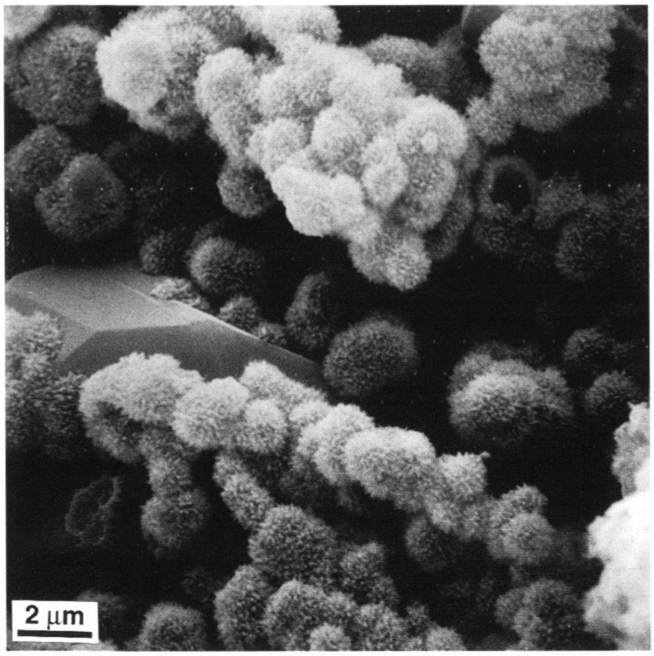
Fig. 19. Organic matter associated with the formation of iron oxihydrate. **a)** SEM photomicrograph of quartzite surface which is heavily eroded by a fungal colony. The whole zone is coated by a layer of bulbous Fe-crystallites the growth of which apparently is associated with a thin film of organic material (centre). **b)** SEM photomicrograph of goethite spherulites which appear to grow around an organic filament.

Fig. 20. SEM photomicrograph showing typical needle-like goethite encrustations.

Fig. 21. SEM photomicrograph of typical platy hematite encrustations.



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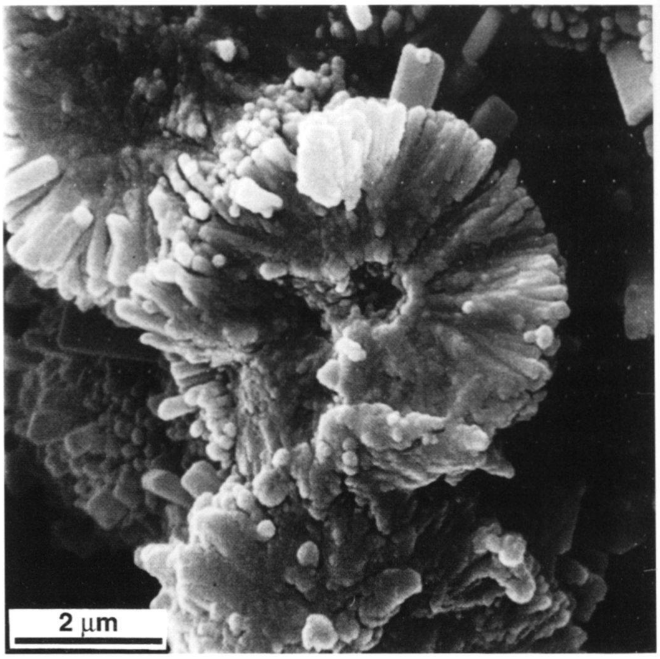

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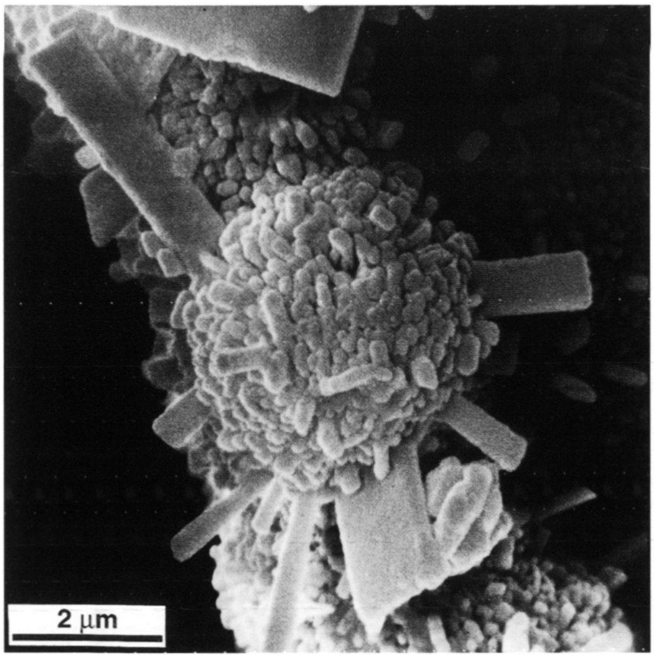

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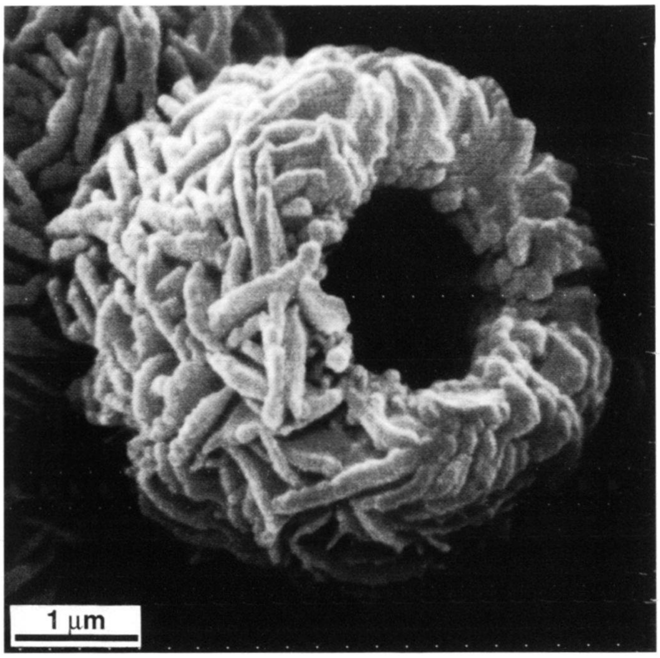

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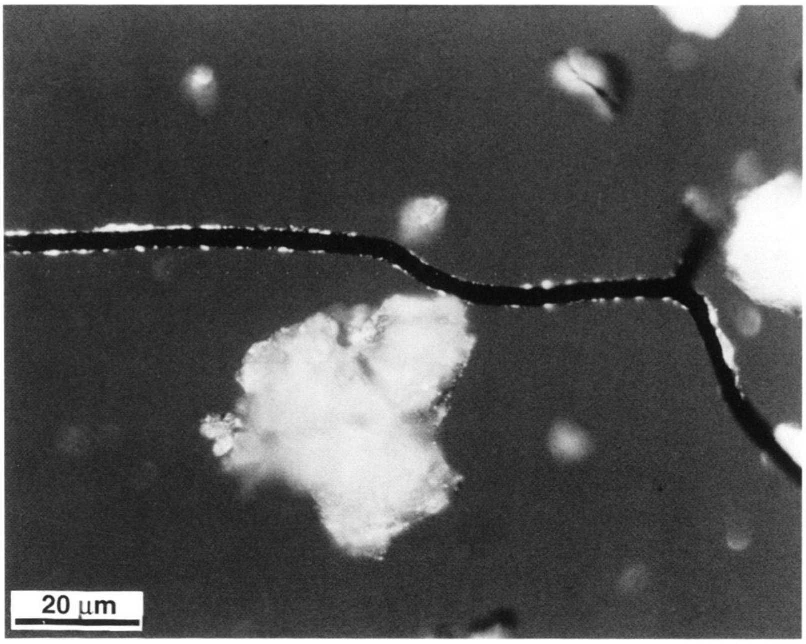
Fig. 22. SEM photomicrograph showing spherulites of goethite resembling minute hedgehogs with characteristic holes in their isodiametric bodies. Note the faces of the quartz crystal in the centre left.

Fig. 23. SEM photomicrographs of goethite spherulites. **a)** Radially grown spherulite with a relatively small hole in the centre indicating formation around the fungal hypha. **b)** Bulbous spherulite with distinct radially grown platy needles.

Fig. 24. SEM photomicrograph of an isometric spherulite consisting of platy hematites arranged in a rosette.



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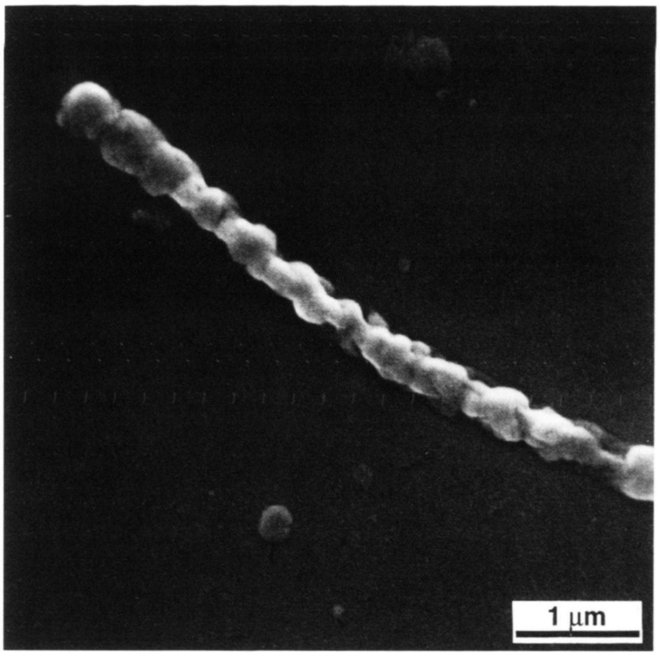

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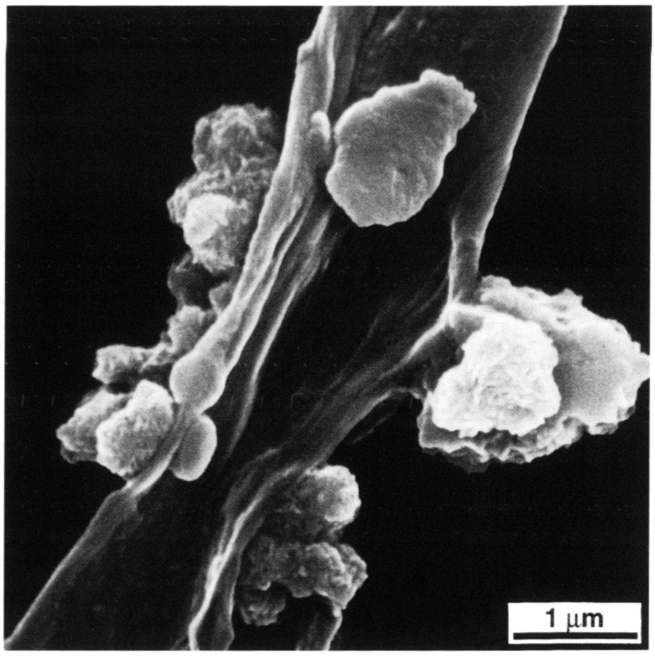
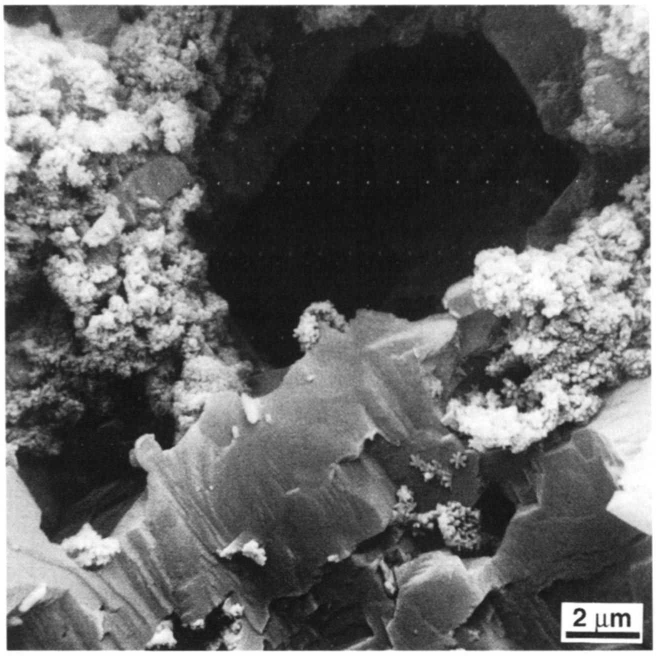


25c

Fig. 25. Growth structures around fresh fungal hyphae. **a)** Photomicrograph of a hypha from a fresh fungal mat. Note the white rim of unknown composition around the hypha indicating beginning mineralization. **b)** SEM photomicrograph of bulbous to spherical growth structures along a fungal hypha. These structures have not been mineralized. **c)** SEM photomicrograph showing a close up of mineralized bulbs developing on a fungal hypha. These bulbs strongly resemble Ca-oxalates from laboratory cultured fungal hyphae.



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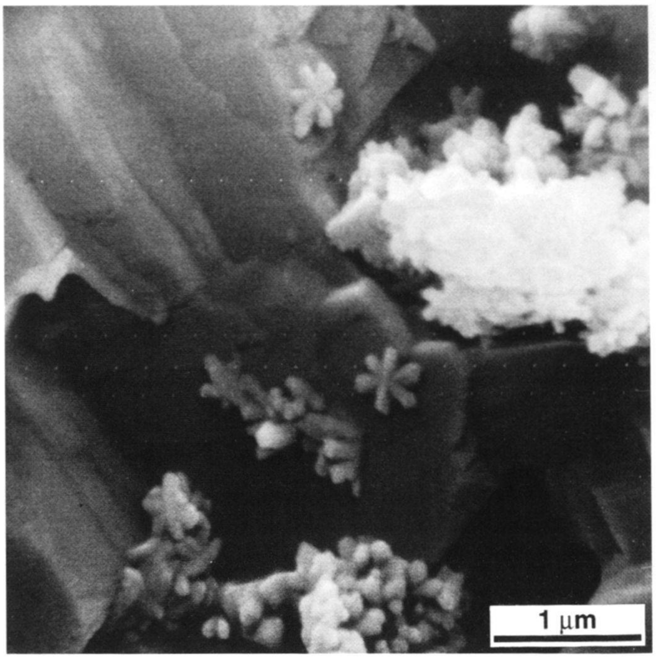

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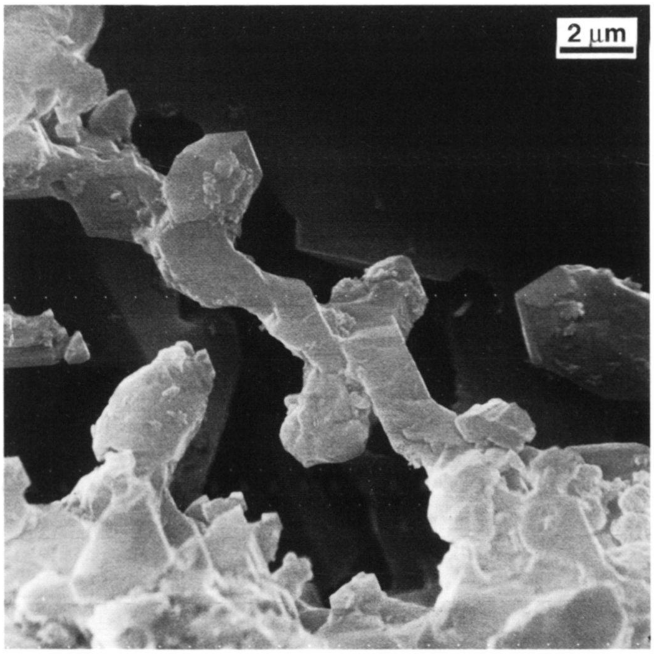

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Fig. 26. SEM photomicrographs of the dolomite surrounding the quartzite lens. **a)** Bacterial colonies on dolomite surfaces. It is suggested that these bacteria degrade the dolomite. **b)** Close up of Fig. 26a showing the distinct star-shaped form of these bacteria.

Fig. 27. SEM photomicrograph of minute quartz crystals occurring in pore spaces. Owing to their arrangement, which resembles organic strings, it is assumed that they formed in association with organic material. They represent a late generation of quartz.