

# Origin of pedogenic needle-fiber calcite revealed by micromorphology and stable isotope composition—a case study of a Quaternary paleosol from Hungary

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## Abstract

Pedogenic needle-fiber calcite was studied regarding its morphology, texture and stable isotope composition from the paleosol of the Quaternary Várhegy travertine (Budapest, Hungary). The needle-fiber calcite is composed of 40–200 µm long monocrystals. Smooth rods as well as serrated-edged crystals with calcite overgrowths were identified by SEM. Needles have several textural varieties: randomly distributed crystals in vugs and pores with calcite hypocoatings, bundles of subparallel crystals forming coatings around grains and alveolar structure with bridging needles in vugs.

The morphological study of needle-fiber calcite suggests that needles are calcified fungal sheaths and produced by fungal biomineralization, a common process in recent and fossil soils and calcretes. The stable isotope composition of needle-fiber calcite (average:  $\delta^{18}\text{O} = -7.1\text{‰}$  and  $\delta^{13}\text{C} = -7.3\text{‰}$  vs. V-PDB) indicates significant incorporation of organically derived  $\text{CO}_2$  and probably biological influence on needle genesis. Dissolved host rock travertine and/or atmospheric  $\text{CO}_2$  could also contribute some carbon to the acicular calcite.

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## 1. Introduction

Needle-fiber calcite represents one of the peculiar forms of pedogenic carbonates in soils, paleosols and calcretes. Its formation is thought to be related to organic or inorganic processes mostly independent from each other: needles-fibers may be products of fungal biomineralization or physicochemical precipitation from

soil solution (Verrecchia and Verrecchia, 1994). Determination of possible origin of this special type of carbonate is essential, since the presence of needle-fiber calcite may refer to former biogenic influence (fungi and associated higher plants) in paleosols.

An earlier study demonstrated the presence of needle-fiber calcite in a paleosol intercalated into the Quaternary travertine sequence of the Várhegy in Budapest (Bajnóczi et al., 2003). Fine calcite needles were detected in thin sections of the bulk carbonate material of the paleosol. During further sampling white and soft

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carbonate masses were collected from the brownish paleosol. These masses turned out to be enriched in pedogenic needle-fiber calcite. The macroscopic enrichment of this soil carbonate enables to perform stable isotope analysis. Stable isotope data from needle-fiber calcite are rather limited (Salomons et al., 1977; Strong et al., 1992; Verrecchia et al., 2003), therefore the case study of Várhegy paleosol with combined micromorphological and geochemical analyses may contribute to the better understanding of the genesis of this special type of pedogenic carbonate.

## 2. Characteristics and origin of needle-fiber calcite: a brief review

Needle-fiber calcite usually forms in the early phase of pedogenesis and precipitates as cement in soils and calcretes in vadose conditions (Verrecchia and Verrecchia, 1994). It can also be found in near-surface parts of travertine deposits (e.g. Janssen et al., 1999; Pentecost et al., 1997), in alluvial gravel banks (e.g. Heide et al., 1988) or on walls of caves (e.g. Borsato et al., 2000).

Needle-fiber calcite consists of elongated needles of low magnesium  $\text{CaCO}_3$ . Needles are a few  $\mu\text{m}$  wide and up to several hundred  $\mu\text{m}$  long. A morphological classification proposed by Verrecchia and Verrecchia (1994) distinguishes two basic groups: monocrystalline rods and polycrystalline chains. Monocrystalline rods can be (i) smooth, short, single micro-rods (M form, width  $<0.5\mu\text{m}$ , length  $<2\mu\text{m}$ ), (ii) smooth, long, paired rods (MA form, width  $=0.5\text{--}2\mu\text{m}$ , length  $<100\mu\text{m}$ ) and (iii) serrated-edged, long, paired rods (MB form, width  $=2\text{--}20\mu\text{m}$ , length  $=30\text{--}1000\mu\text{m}$ ). Polycrystalline chains are composed of rhombohedra of calcite joined together in different ways: e.g. “lublinite” consists of short crystals with *c*-axis parallel, but stacked slightly *en echelon* (Stoops, 1976); other types of polycrystalline needle-fiber calcite sometimes referred to as whisker crystals are chains of overlapping rhombohedra (Jones and Ng, 1988).

Needle-fiber calcite occurs in interparticle pores, desiccation cracks, dissolution vugs and fractures. Several textural arrangements of acicular crystals include randomly or subparallel-oriented meshes, fibers tangentially cemented to pore walls or grains and tubular voids walled by oriented fibers (e.g. Solomon and Walkden, 1985; Phillips and Self, 1987; Strong et al., 1992).

The origin of needle-fiber calcite has been discussed for many years and is usually interpreted in two ways: by purely physicochemical phenomena or in relation with organic material (roots, root hairs, bacteria, algae and fungi). The compilation of Verrecchia and Verrecchia (1994) indicates that the groups of needle-fiber calcite have similar morphologies, but do not seem to be related genetically. Each morphological type of calcite

crystal has a specific origin. Micro-rods can be calcified bacteria decomposing organic material (Phillips and Self, 1987; Loisy et al., 1999) or physicochemically precipitated calcite nuclei (Verrecchia and Verrecchia, 1994). Polycrystalline chains are generally considered to be purely physicochemical precipitations related to evaporation and desiccation (Jones and Kahle, 1993). However, MA and partly MB rods seem to have direct biological origin (Verrecchia and Verrecchia, 1994).

The textural arrangements of MA rods and their close association with mycelian strands of fungi coating plant rootlets suggest that they are products of fungal biomineralization (Phillips and Self, 1987; Wright, 1984, 1986). Calcite needles are embedded within mycelian strands and each double-rod probably forms inside a single fungal hypha (Phillips and Self, 1987). The MB rods form after liberation of MA rods from fungal sheaths due to decomposition of the organic material. The precipitation of serrated edges of MB rods occurs physicochemically and requires highly supersaturated solutions undergoing intensive evaporation (Wright, 1984; Solomon and Walkden, 1985; Jones and Kahle, 1993). Preferential crystal growth of calcite occurs on pre-existing MA rods (Phillips and Self, 1987; Verrecchia and Verrecchia, 1994).

Needle-fiber calcite was detected in recent and Quaternary soils-paleosols and calcretes in several cases (e.g. Phillips and Self, 1987; Becze-Deák et al., 1997; Bruand and Duval, 1999; Srivastava, 2001; Nash and McLaren, 2003; Alonso et al., 2004; Cailleau et al., 2005). It can also be preserved, however, in ancient soils and calcretes, e.g. in Miocene (Alonso-Zarza, 1999) and Palaeogene (Alonso-Zarza and Arenas, 2004) calcretes or in Lower Carboniferous paleosols (Wright, 1984, 1986). The needles are prone to disintegration or recrystallization to micrite, therefore their presence in paleosols or even in recent soils sometimes can be discovered only by using cathodoluminescence (Solomon and Walkden, 1985).

Preservation of needle-fiber calcite in soils indicates that the pedogenesis was weak (lack of leaching) and/or the climate of the environment was arid to semiarid (Wright, 1984). However, Strong et al. (1992) demonstrated its occurrence under cool and wet climate in a well-drained gravel deposit with abundant carbonate clasts and high degree of biological activity. According to the review of Verrecchia and Verrecchia (1994), the MA and MB forms of needle-fiber calcite seem not to have a special climatic or environmental significance.

## 3. Materials and methods

The Várhegy (Castle Hill, in English) is situated on the right side of the river Danube in Budapest

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