Fluid "self-purification" – Insight from the particle attachment processes during the growth of three-dimensional mineral dendrites

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Manganese (Mn) dendrites are a common type of mineral dendrites which can typically be found as two-dimensional branch-like patterns on rock surfaces, such as bedding planes and joints suggesting a fluid-rock interaction. Their three-dimensional (3D) counterpart has so far been massively overlooked, and thus little is known about 3D mineral dendrite growth processes and potential implications for fluid-rock interaction. Here, we combined high-resolution X-ray, electron-based micro-analyses with numerical modelling to show that the formation of natural 3D dendrites is an aggregation process of Mn-oxide nanoparticles in an aqueous environment. The dendrites form a < 15 mm high "forest" (Figure 1) in clinoptilolite-tuffs (zeolites), with trunks and branches, both having a core-rim structure and in the upper part of the forest, an alternating concentric core-rim layering. Secondary electron microscope (SEM) observations indicate dendrite growth reduced the rock´s original porosity from ~17% in the matrix to ~1% - 4% in the internal rims and 0% in the cores. High-resolution SEM shows dendrite-forming Mn oxides are built by sub-angular to rounded, several-nanometer- to 1-µm-sized particles that have been aggregated forming larger clusters. Using the lattice Boltzmann method we modelled the formation and evolutionary processes of the 3D Mn dendrites. This allowed us to track the diffusing population of Mn ions and oxygen molecules as well as the reaction between them that led to the formation of Mn-oxide nanoparticles. The mobility and aggregation of nanoparticle populations were then tracked.

Our numerical models suggest sensitive feedback between dendrite morphology and the volume of infiltrating fluids, as well as the concentrations of Mn ions. Our work provides three important findings. First, 3D mineral dendrites can offer a simple system to investigate the affecting physical parameters of particle attachment processes in nature, such as the interplay of diffusion and surface energy effects between particles on dendrite growth dynamics. Second, the growth of the 3D dendrites, aggregating the particles in the surrounding fluids, can be seen as a fluid "self-purification" process. Third, the formation of the banding structures of the 3D dendrites, as well as the sensitive growth of dendrites in relation to the volume of infiltrating fluid and concentrations of Mn ion, strongly suggest the dendrites encode the hydrogeochemical history of the hosting rocks.

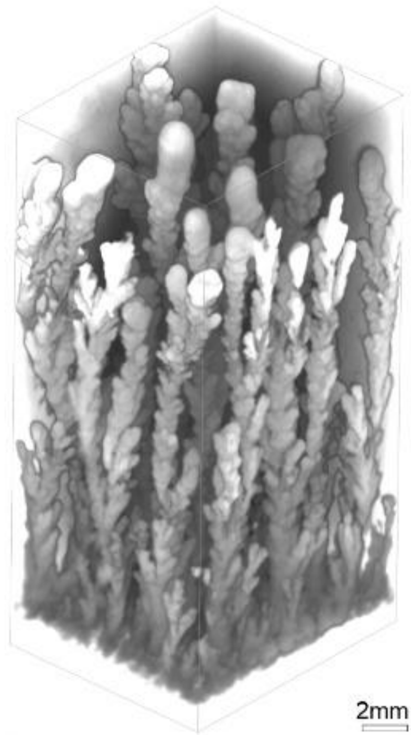

Figure 1. X-ray microtomography data show a 3D mineral dendrite forest.