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Template-directed crystallization of sulfates onto calcite

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Epitaxial crystallization strategies involve the use of organized substrates that facilitate crystal nucleation and are successfully employed in industry to control size, habit and orientation of crystals. This inductive effect is associated with the structural matching between the substrate and the overgrowth. Organic templates may help overcome eventual structural mismatching between substrate and overgrowth. In nature, organic surfaces often direct the nucleation and growth of inorganic phases, a process known as biomineralization. Here we present investigations on sodium sulfate crystallization onto an inorganic substrate (calcite), directed by organic molecules (phosphonates). *In situ* crystallization experiments were carried out on an environmental scanning electron microscope (ESEM) and using an X-ray diffractometer. These studies show that oriented sodium sulfate grows on a DTPMP (diethylenetriamine-penta-methylene phosphonic acid) monolayer template adsorbed onto Iceland spar cleavage surfaces. Mirabilite nucleates onto the DTPMP template with $(001)_{mirabilite}//(10\bar{1}4)_{calcite}$. In contrast, different sodium sulfate phases crystallized in the absence of an organic additive, and crystals grew with no preferred crystallographic orientation. These results give experimental evidence of template-assisted sulfate crystallization on calcite surfaces in the presence of phosphonates. The organic template, whose formation was observed with atomic force microscopy (AFM), may contribute to the heterogeneous nucleation of mirabilite ($Na_2SO_4{\cdot}10H_2O$) at very low supersaturation. This effect will significantly reduce crystallization pressure exerted by this salt when growing within a porous calcitic substrate. It is therefore suggested that this process may help minimize damage to carbonate stones due to salt crystallization, which is one of the main hazards for historic architecture and statuary. These results also suggest that the ex-

tensive use of phosphonates as crystallization inhibitors to prevent pore plugging due to Ca and Ba sulfate crystallization in oil fields, could in fact enhance such a problem in carbonate reservoir rocks due to template-assisted sulfate precipitation.