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# The effect of sand grain roughness on the grain-scale spatial distribution of grain-surface precipitates formed by evaporation



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#### GRAPHICAL ABSTRACT



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#### ABSTRACT

This work examined the precipitation patterns formed by two solutes (sucrose, sodium chloride) on individual sand grains after evaporation from aqueous solution. Experiments were conducted by placing droplets of solution on individual grains, allowing them to evaporate, and then imaging the resulting precipitates. Grains were cleaned and used for multiple experiments with different solution concentrations. Precipitation patterns were studied through a combination of scanning electron microscope (SEM) imaging and energy dispersive x-ray spectroscopy (EDS). The results of the work indicate that sodium chloride precipitates were formed on ridges on the grain surfaces, while sucrose precipitates were formed in valleys. The difference in the behavior was consistent with the hydraulics of film flow out of low points on grain surfaces during evaporation (a phenomenon sometimes referred to as the coffee-ring effect), and the different viscosities of the two solutes near their solubilities. Simulations were able to produce precipitation patterns on grain surface consistent with experimental observations. Based on the results of the work, it is likely that most solutes will exhibit precipitation on ridges; only those with extremely high viscosities at their solubility (e.g. hundreds of mPa s) might be expected to be trapped in valleys.

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#### 1. Introduction

Evaporation of a solvent in natural or engineered porous media can cause the concentrations of nonvolatile dissolved solutes in porewater to increase progressively until the compounds precipitate out of solution onto grain surfaces and in the surrounding pores. In an environmental context, the spatial configurations of precipitates can potentially impact the dynamics of redissolution in porous media by impacting the accessible interfacial area across which dissolution can occur, potentially influencing the ultimate mobility of the compounds in the environment. In an industrial context, an understanding of factors impacting precipitation on rough surfaces could be useful in a range of situations, such as controlling unwanted surface deposition during processes involving evaporation. The focus of this work was on studying how grain-surface roughness impacts patterns of chemical precipitation on individual grain surfaces, with the ultimate goal of developing a more quantitative understanding of how precipitates form on rough surfaces.

Noninvasive imaging methods such as dual energy gamma radiation, x-ray micro tomography and magnetic resonance imaging (MRI) have been used to study processes occurring in porous media [1–5]. A significant amount of work has been reported studying the patterns of salt precipitation in sands as a result of evaporation. The effects of factors such as grain size, grain angularity, grain hydrophobicity, grain size distribution, ambient temperature, and salt type on precipitation patterns have been studied [6–11]. While surface precipitation is widely observed to occur, many of these studies have focused on patterns of precipitation formed in pore spaces, in part because of their impact on permeability and evaporation patterns.

Across various disciplines, the related phenomena of surface precipitation and deposition on surfaces has been studied using imaging technology like scanning electron microscopy (SEM) and transmission electron microscopy (TEM) [12–15]. For example, research has shown that rougher surfaces tended to retain a higher number of nanoparticles in the environment [4]. In the medical sciences, the effect of surface morphology on precipitation patterns was studied and it was found that roughness was critical to bioactivity of metals to be used as implants. The ability of apatite to precipitate out from simulated body fluids onto treated metal strips was greatly enhanced by surface roughness [16]. Studies have shown that for ceramic materials to be used as bone implants, surface roughness of the ceramic material greatly improved the rate of apatite formation from simulated body fluids in vitro [17]. Surface roughness also increased precipitation of calcium phosphate on phospholipid layers [18].

Despite the extensive studies performed on evaporation/precipitation in saline rich soils, the mechanisms behind the process are not yet fully understood [19]. While numerous studies have been performed on representative elementary volumes (REVs) of sand, thus giving some understanding of precipitation in pores, more limited research has been performed to understand the evaporation/precipitation process at the grain scale [20]. In the same vein, a plethora of data exists on precipitation of NaCl and other inorganic salts on rough surfaces but relatively fewer studies have been completed on precipitation of organic substances as a result of evaporation. Studies of precipitation patterns formed on rough surfaces in saturated environments exist, but these surfaces are usually reactive with precipitation occurring as a result of some chemical reaction or adsorption of ions to the surface. Little work has been reported examining surface precipitation in unsaturated systems due only to evaporation.

The hypothesis driving this work was that the evaporation process would cause thicker precipitates to form in the deeper valleys on grain surfaces and thinner precipitates on the plateaus, as a result of solute concentration increasing as capillary held water films recede to low points on the grain. As the water evaporates, the air-water interface recedes gradually towards the grain surface, forming a liquid film which initially bridges the high points on the grain. As evaporation





Fig. 1. SEM images of the two quartz sand grains used in the study, Grains A and B.

progresses, regions of thick film on solid surfaces shrink to occupy hydraulically disconnected regions with lower-elevation surface roughness features [21]. It was hypothesized that greater solute mass present in these regions would ultimately lead to greater precipitate mass.

The work described here involved single-grain evaporation experiments, where the spatial distribution of precipitates formed following evaporation was observed through SEM imaging of thick precipitates, combined with Energy Dispersive X-ray Spectroscopy (EDS) to quantify the spatial distribution of thinner precipitates. A model of the evaporation-precipitation process was developed and used to test the hypotheses of the work.

#### 2. Methodology

#### 2.1. Experimental procedures

Sodium chloride (NaCl, MW = 58.44 g/mol) and sucrose ( $C_{12}H_{22}O_{11}$ , MW = 342.3 g/mol) were chosen for the study as representative high-solubility inorganic (sodium chloride) and organic (sucrose) solutes. The solubilities of the compounds are approximately 26% w/w and 70% w/w for sodium chloride and sucrose, respectively. Experiments were conducted at 0.1 and 0.2 M for sodium chloride, and 0.05 and 0.1 M for sucrose. These concentrations correspond to approximately 2.2 and 4.4% of the solubilities of the two compounds.

Sand grains used for experiments were taken from a beach sand sample collected from Sandwich, Massachusetts. The sand sample was Download English Version:

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