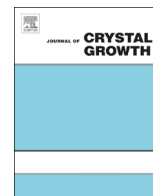




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Evaporation with sodium chloride crystallization in a capillary tube

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ABSTRACT

Sodium chloride crystallization induced by evaporation of aqueous solution is studied from visualization experiments in a circular capillary tube. In agreement with recent studies, the onset of crystallization is observed with a significant supersaturation. Detailed information on the precipitation kinetics and transport of ions is obtained from numerical computations of the ion mass fraction field during the evaporation process. It is shown that the precipitation kinetics is fast compared to transport so that the crystal growth is mostly controlled by the transport. This offers possible simplifications for analyzing more involved situations such as crystallization in porous media. The fact that the significant supersaturation does not lead to tube damage is explained.

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1. Introduction

Capillary tubes can be seen as model systems for the study of porous media, [1,2]. For instance, a single capillary tube can often be considered as the elementary component in pore network models, e.g. [3]. Studies with an elementary geometry such as capillary tube are also of interest for the domain of microfluidics, e.g. [4]. In this context, the present article is devoted to a study of evaporation of a sodium–chloride aqueous solution from a single capillary tube of circular cross section.

The study of evaporation in a porous medium in the presence of a dissolved salt with possible crystallization has been the subject of several studies in recent years, e.g. [5–8] and references therein. A motivation is the study of damage on porous stones and other building materials that can result from the crystallization process, e.g. [9]. As reported in [9], the so-called crystallization pressure, which is the key-concept to analyze the generation of stresses due to the growth of crystal in pores, is directly dependent on the supersaturation at the onset of crystallization, i.e. the amount of ions in excess compared with a solution in equilibrium with salt crystals. In this paper, the supersaturation, denoted by S , is defined by:

$$S = \frac{m_{o_{cr}}}{m_{o_{sat}}}, \quad (1)$$

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where $m_{o_{cr}}$ is the molality (moles of solute by unit mass of solvent) when crystallization occurs and $m_{o_{sat}}$ is the molality at saturation ($m_{o_{sat}} = 6.1$ M for NaCl).

It should be pointed out that the question of supersaturation in sodium chloride aqueous solutions is somewhat controversial. Until recently, it was widely admitted that supersaturation was negligible with NaCl, i.e. $S \sim 1$. This was notably supported by experiments where the ion concentrations during the evaporation process were measured using NMR, i.e. [10–13]. By contrast, the experiments with capillary tubes reported in [14], indicated supersaturation as high as 1.6, a level of supersaturation largely sufficient to explain for instance the damages on porous rocks observed in the experiments reported in [15]. A significant supersaturation was also indirectly reported from the analysis of drying experiment with packings of glass beads, [5]. Owing to the crucial impact of supersaturation on stress generation, it is important to evaluate the supersaturation. One major objective of the present experiment is therefore to confirm the findings reported in [5] and [14] regarding the level of supersaturation that can be expected from evaporation of a sodium chloride aqueous solution. As reported in [16] and [13], the supersaturation can be quite significantly increased by performing deliquescence/recrystallization cycles. Here, we do not perform such cycles and are only interested in the basic situation where the solution evaporates from the capillary tube without previous purification of the solution through deliquescence/recrystallization cycles. We note that the estimate of supersaturation reported in [14] was obtained from a global mass balance (details are given below) whereas here we also compute numerically the ion mass fraction distribution

during the evaporation process so as to take into account the spatial variations of the ion mass fraction within the solution during the evaporation process.

A second objective, which was not addressed in the above-mentioned papers, is to discuss the crystal growth by considering the balance between the precipitation kinetics and the convective–diffusive transport processes. This allows us to distinguish two main phases: a very fast one controlled partly by the precipitation kinetics followed by a much longer one controlled by the ion transport. This provides interesting indications for the modeling of the crystal growth process in more complex situations, such as in porous media.

The paper is organized as follows. The experimental set-up is described in Section 2. Experimental results are presented and discussed in Section 3 together with some numerical simulation results. A conclusion is presented in Section 4. The numerical model is presented in the Appendix A.

2. Experiments

Evaporation experiments are performed using a circular capillary tube. A circular tube is preferred to a tube of polygonal cross-section so as to avoid complications associated with the development of capillary films along the corners of the tube, i.e. [17,2]. The tubes are borosilicate glass tubes of 1 mm interior diameter (Hilgenberg®). This diameter is denoted by d in what follows. The tube is held either in vertical or horizontal position. The tubes are soaked in an ultrasound bath with detergent solution before use. Then, they are rinsed in deionized water and ethanol before passing in an O_2 plasma oven. The sodium chloride solution is prepared with a mass fraction of salt equal to 25% (25 g of salt for 100 g of solution), close to the saturation (solubility) value (26.4%). Salt is provided by Sigma-Aldrich®, with a purity ensured to be higher than 99.5%. It is diluted in deionised water prepared by using an activated carbon filter and 3 deionizer filters. In order to avoid wetting the top of the tube, the tube is filled from the bottom as sketched in Fig. 1a. As can be seen from Fig. 1, the tube is filled in such a way that a volume of solution is confined between two menisci. Because the top of the tube is surrounded by dry air and the bottom one is in contact with a gas plug saturated in water vapor, evaporation takes place only at the upper meniscus. To form the liquid and gas plugs visible in Fig. 1, the syringe is first connected to a flexible tube. A volume of solution is sucked into the flexible PTFE tube, then again a volume of

solution. The flexible tube is then glued to the entrance of capillary tube and the adjacent liquid and gas plugs are slowly injected into the capillary tube to obtain the phase distribution shown in Fig. 1b.

The tube is set in an enclosure at controlled temperature ($T \sim 22^\circ\text{C}$) and relative humidity ($\text{RH} \sim 7\%$) thanks to a lithium bromide (LiBr) solution covering the bottom of the enclosure. The temperature and relative humidity in the enclosure are measured and recorded during the experiment. All the data are obtained from successive images of the capillary tube during an evaporation experiment. Images are taken with a Nikon® D70 camera with an AFS micro Nikkor 105 mm lens for the low acquisition frequency (typically 0.0033 Hz), and a film camera CCD Senciscam PCO® for higher frequency (typically 0.5 Hz). These devices are interfaced to a computer thanks to Camera Control Pro 2® and Camware® software, respectively. The objective is to follow the receding meniscus during evaporation in order to: i) measure its kinetics, ii) determine the size of liquid plug, which gives access to values of saturation. Also the visualization enables us to measure the crystallization kinetics. As can be seen from Fig. 1, gas and liquid phases appear in light gray and wall and interface in dark with the ombroscopy method used for this experiment.

Results are presented for seven experiments (Table 1), four with the tubes held in a vertical position and three with the tube in a horizontal position. This is basically the same experiment, which is repeated seven times. The differences lie in the position of the tube, vertical or horizontal, the initial position z_{m0} of the receding meniscus within the tube and in the initial volumes of gas and liquid plugs.

Looking at Fig. 1b, one may wonder whether the dark thick lines along the vertical edges of the tube image in the regions of the tube occupied by the gas phase corresponds to liquid films along the tube inner wall. These dark lines are, however, due to an optical effect associated with the curvature of the tube wall. They are also observed in a perfectly dry tube. Note that the contact angle of a salt solution on glass is on the order of $40\text{--}45^\circ$ [18]. Thus we are far from a situation of perfect wetting. A very thin film, of a few nanometers, might be present. However, we do not believe that such a film, if any, can play a role in the investigated problem.

3. Results and discussion

3.1. Evaporation kinetics

A first step is to control that the evaporation kinetics before the onset of crystallization is in good agreement with the available

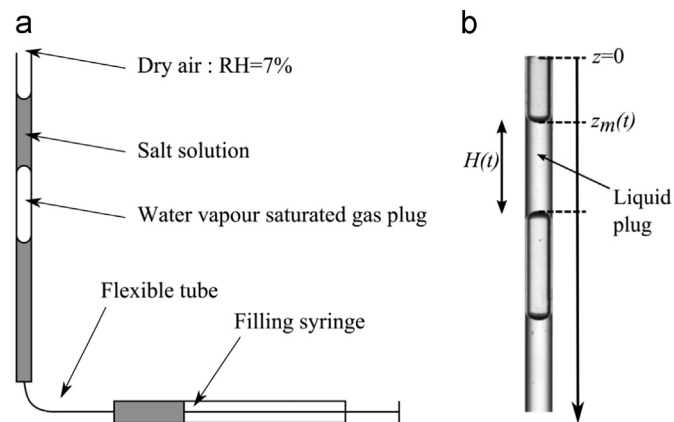


Fig. 1. (a) Schematic of experimental setup, (b) image of the tube obtained by ombroscopy. Liquid and gas phases are in light gray and walls and interfaces appear in dark. The liquid plug of solution is in contact with a dry atmosphere at its top and with a saturated atmosphere at its bottom. The volume of solution at the bottom is connected to the filling syringe.

Table 1

Global and local supersaturation obtained for each experiment (see Section 3.4). The global supersaturation is obtained from an average mass balance whereas the local supersaturation is obtained from determining numerically the ion mass fraction distribution within the tube; Meaning of letters is: V: vertical, H: horizontal, T: top, B: bottom, M: middle; z_{m0} is the initial position of the receding meniscus measured from tube entrance, H_0 is the initial length of liquid plug; D_s is the molecular diffusion coefficient of the ions in the liquid phase ($D_s = 1.3 \cdot 10^{-9} \text{ m}^2/\text{s}$). The Peclet number Pe is defined and discussed in §3.2. Note that the images in Fig. 1b and Fig. 3 do not correspond to one of the experiments mentioned in the table. They correspond to an experiment performed with a shorter initial liquid plug designed to visualize the fall of the crystal within the plug (see text).

Experiment #	1	2	3	4	5	6	7
Tube orientation	V	V	V	V	H	H	H
z_{m0} (mm)	1.4	3.7	1.3	1.9	3.5	8.8	4.0
H_0 (mm)	27.3	19.4	19.4	16.9	18.6	18.2	20.5
Crystal growth site position	T	B	B	B	T	M	T
Initial Peclet number $Pe_0 = \frac{dz_m(0)H(0)}{D_s}$	3.73	1	2.86	1.7	1.01	0.4	0.98
Global supersaturation	1.35	1.34	1.46	1.32	1.22	1.40	1.62
Local supersaturation	1.59	1.53	1.62	1.52	1.41	1.51	1.78

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