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## Sodium chloride crystallization in confinement: The mechanism of cubic to hopper crystal growth transition

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Sodium chloride crystallization is a key factor in many industrial and geological processes such as for deicing salts, in oil well drilling and CO<sub>2</sub> sequestration, in the erosion of rocks, monuments and for the sodification of soils. Generally, salt crystallization in confined space, as encountered in porous media, can significantly change the porosity and permeability, and as such restrict flow and mass transfer; in addition it can even deform the solid matrix due to the build-up of a crystallization pressure. Therefore, the ability to influence the rate of the crystal growth and the resulting morphology can have a significant impact on various applications.

We report on the transition mechanism from cubic to hopper growth of sodium chloride and the supersaturation at which the transition is observed. For this we have investigated experimentally at the microscale the kinetics of crystal growth at different degrees of supersaturation  $S$  in confinement, up to its metastability limit which happens at  $S=1.6\pm0.2$  times the saturation concentration [1]. The growth rates at the onset of precipitation are quantified together with the corresponding supersaturation. Our results show that the growth rate of the cubic crystal increases linearly with increasing supersaturation up to  $S=1.4$ , which corresponds to the lower limit of the metastability. Surprisingly, above this supersaturation, the cubic growth rate levels off to a maximal rate of  $6\pm2\ \mu\text{m s}^{-1}$ . By combining the global growth coefficient derived from our measurements with those in the literature at different temperatures, we corroborate the activation energy for the growth of sodium chloride.

For supersaturations above 1.4, as the cubic crystal cannot grow any faster, the hopper growth is initiated in order to consume the supersaturation rapidly; the hopper growth shows up as a chain-like structure of small cubic crystals. We show that the speed of the total hopper growth is much faster than the determined maximal speed of the growth of each cubic unit. Our experimental results of growth speed as a function of supersaturation above 1.4 can be fitted with a power law with an exponent of 3, underlining a two dimensional nucleation growth mechanism [2]. The hopper morphology disappears at a later stage of growth if there is enough salt solution to supply the crystal with ions to continue the cubic growth at very low supersaturation. Although hopper growth has been reported for different salts, this study is the first one in which the mechanism of such growth morphology is clearly identified and experimentally measured. Understanding the crystallization pathways allows to better control crystal design or to elucidate on pore-space changes as occurring during e.g. salt weathering of rocks and building materials or CO<sub>2</sub> sequestration, and to enrich geochemical models.

### References

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- [2] Mullin, J. W. *Crystallization*; 4th ed.; Butterworth-Heinemann: Oxford, 2001.

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