Self-Lifting NaCl Crystals

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Supporting Information

Surface fraction calculation

The force generated by the crystallization is given by $F = P_{\text{crys}} A \varphi$, where $P_{\text{crys}}$ is the crystallization pressure, $A$ is the spherical area of the bottom of the crystal and $\varphi$ the surface fraction making contact. We analyzed the surface fraction $\varphi$ using Scanning Electron Microscopy(SEM). From this image, one can clearly distinguish smaller and smaller crystals that form when the capillary bridges become very small; these are ultimately the parts making contact with the bottom. The cubic structures in the background are the crystals formed in the earlier stages of the evaporation before the formation of the capillary bridges. Figure S1 shows a SEM measurement for a typical experiment and shows that the contact points are mainly at the edges of the 'legs' indicated in yellow in figure S1b. We find a value for the surface fraction of $\varphi \approx 6\%$.

Figure S1: Scanning Electron Microscopy images of the bottom of crystal assembly.

(a) SEM image from the bottom of the final crystal showing the amorphous legs making contact with the substrate and the earlier formed cubic strate. We see that legs mostly have their contact points at their edges.
Potassium chloride

We performed the experiments under the same conditions with KCl solutions at three different temperatures; 28°C, 50°C and 80°C. Droplets of 10 μL were deposited on fresh silanized glass slides with a contact angle of $\theta = 115 \pm 3^\circ$. The KCl solutions (Sigma-Aldrich purity $\geq 99.9\%$) were made at a saturation concentration of $S = 0.95$. A time sequence of the 28°C and 50°C experiments, as can be seen in figure S2, shows that with the KCl experiments, we find dynamics similar to NaCl.

![Figure S2: Time sequence of evaporating aqueous KCl droplets on silanized glass at T = 28°C and 50°C. All experiments were performed under a controlled relative humidity of RH = 40 ± 1 %.
](image)

Analyzing the growth rate, again we find a strong temperature dependence. The analyses of the activated process is shown in figure S3. As with NaCl, from plotting ln(K) over the inverse temperature, we obtain the activation energy and the attempt frequency. The slope gives an activation energy of $\Delta G_a = 67 \text{ kJ/mol}$. From the intersection with the y-axis, we obtain an attempt frequency of $C = 2.04 \times 10^{14} \text{ s}^{-1}$. 

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Figure S3: Plot of the overall growth coefficient vs. the inverse temperature for the KCl experiments. The activation energy $\Delta G_a$ is given by the slope of the linear fit (dashed red line). The intersection with the y-axis indicates the attempt frequency of the reaction and corresponds to $C = 2.04 \cdot 10^{14} \text{ s}^{-1}$. The inset shows the measured height for the different temperatures.

Mechanism of leg growth

It has been shown in microgravity\textsuperscript{18} and porous media\textsuperscript{19} experiments that when the evaporation rate is high and such thin films of solutions are present, lateral growth of steps start to develop from the liquid films. This kind of growth also shows up as the hopper growth of hollow pyramidal NaCl crystals. Figure S4 shows SEM images of lateral growth of steps and hopper growth in the legs. The special shape of the bottom of the legs (figure B) is therefore likely to be due to the conical shape of the liquid capillary bridges. Subsequently, just like in salt creeping, this leads to crystalline structures (image A) that can grow against gravity and push up the macrocrystal assembly in our experiments.
Figure S4: Scanning Electron microscopy images from the bottom of a crystal Assembly. The arrows from left to right show the hopper crystals, hollow cubes and lateral steps growth due to rapid evaporation. Image A and B show an enlargement of the legs formed by the lateral growth of steps from the thin films.