Supporting Information:

Thermal Preconditioning of Membrane Stress to Control the Shapes of Ultrathin Crystals

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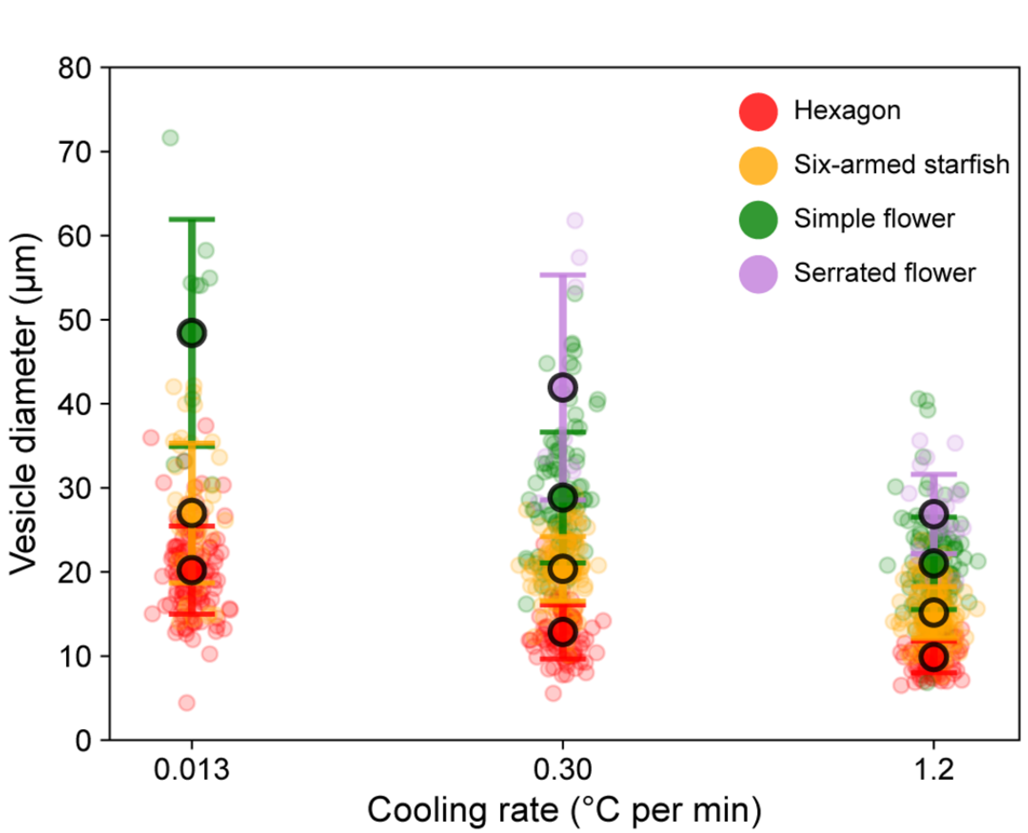
1. Distribution of vesicle sizes for different crystal shapes, measured at different cooling rates.



2. Summary of Solid Area Fractions

Table SI-1 summarizes the solid area fractions of the solid domains (one solid domain is present in each vesicle.). The solid area fractions were calculated as described in the supporting information of our prior work.[1]

Table SI-1. Summary of solid area fractions of solid domains produced at different cooling rates.

	1.2 ℃ per min	0.30 ℃ per min	0.013 ℃ per min
Mean	0.127	0.125	0.134
Standard Deviation	0.015	0.019	0.025
Sample Size	14	44	12

3. Impact of membrane property parameters on tension evolution, from the engineering model.

Below we show the impact of membranae properties on the tension evolution for an hour of cooling at the rate of 0.3 °C/min. The findings generally show that despite substantial variations in properties, the small vesicles, 20 µm in diameter or less, reach a steady state plateau that varies only slightly with the membrane property of interest. Therefore the impact of the membrane property on the plateau level and timing of the plateau would be very difficult to distinguish in experiments with small vesicles.. The greatest impact of membrane properties is seen for the largest vesicles, approaching 60 µm in diameter. Here the rapid rise in tension was sensitive to membrane properties but, because the large tensions approached lysis conditions, burst and reseal dynamics may dominate in for the largest vesicles.

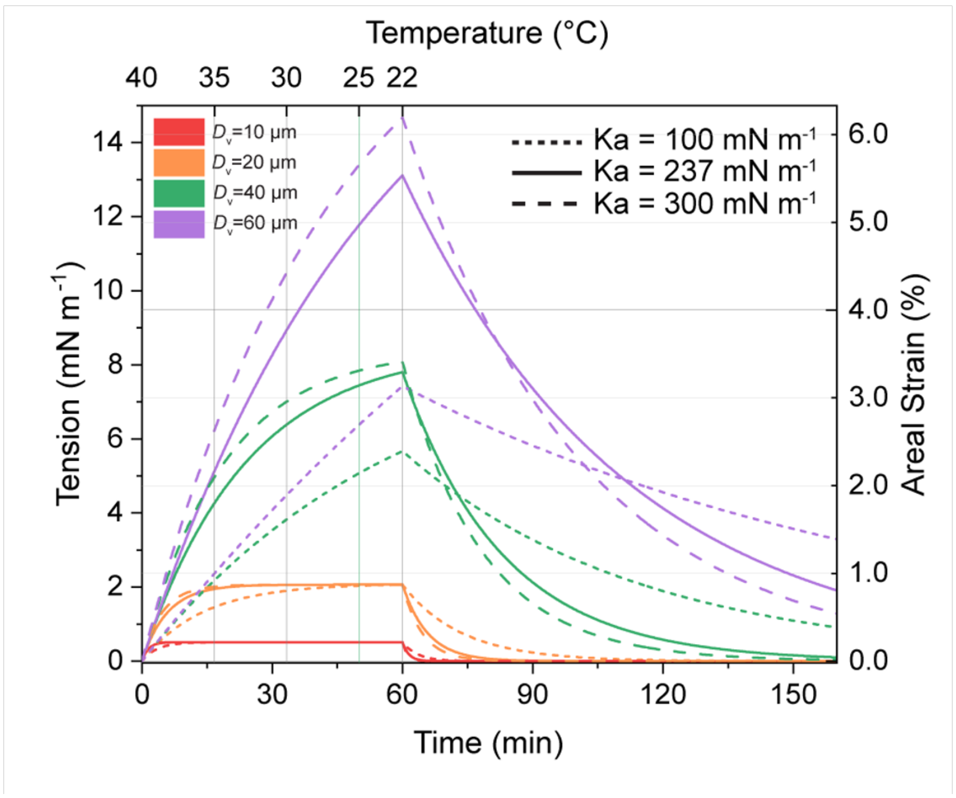


Figure S-2. Impact of K_a, area expansion modulus, on tension evolution during cooling and subsequent temperature hold near room temperature. Each color family represents a vesicle size while the line types show variations in K_a. Similar to the main paper, the upper axis estimates the temperature history for the case where vesicles become spherical and first develop tension at 40°C.

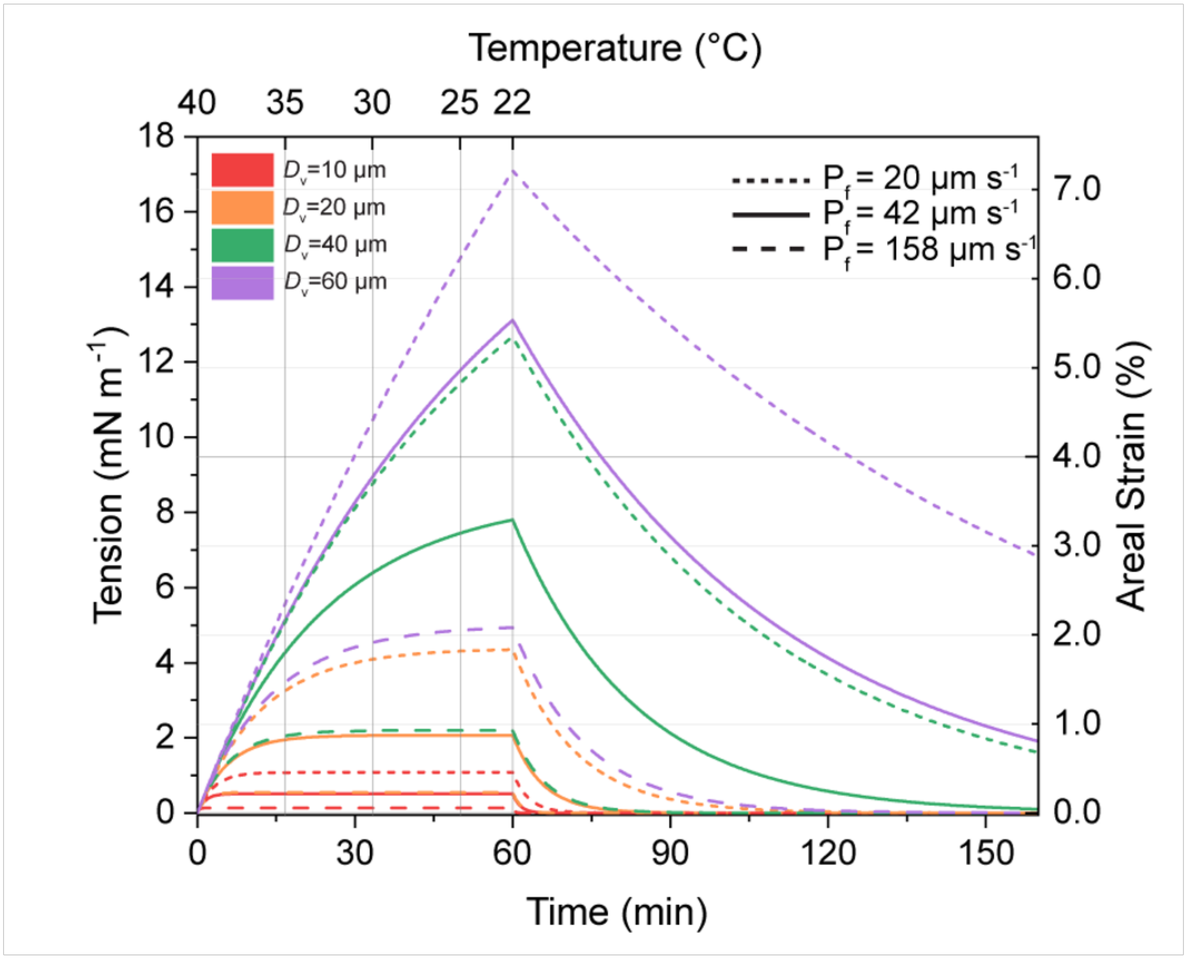


Figure S-3. Impact of membrane permeability, on tension evolution during cooling and subsequent temperature hold near room temperature. Each color family represents a vesicle size while the line types show variations in permeability. Similar to the main paper, the upper axis estimates the temperature history for the case where vesicles become spherical and first develop tension at 40°C.

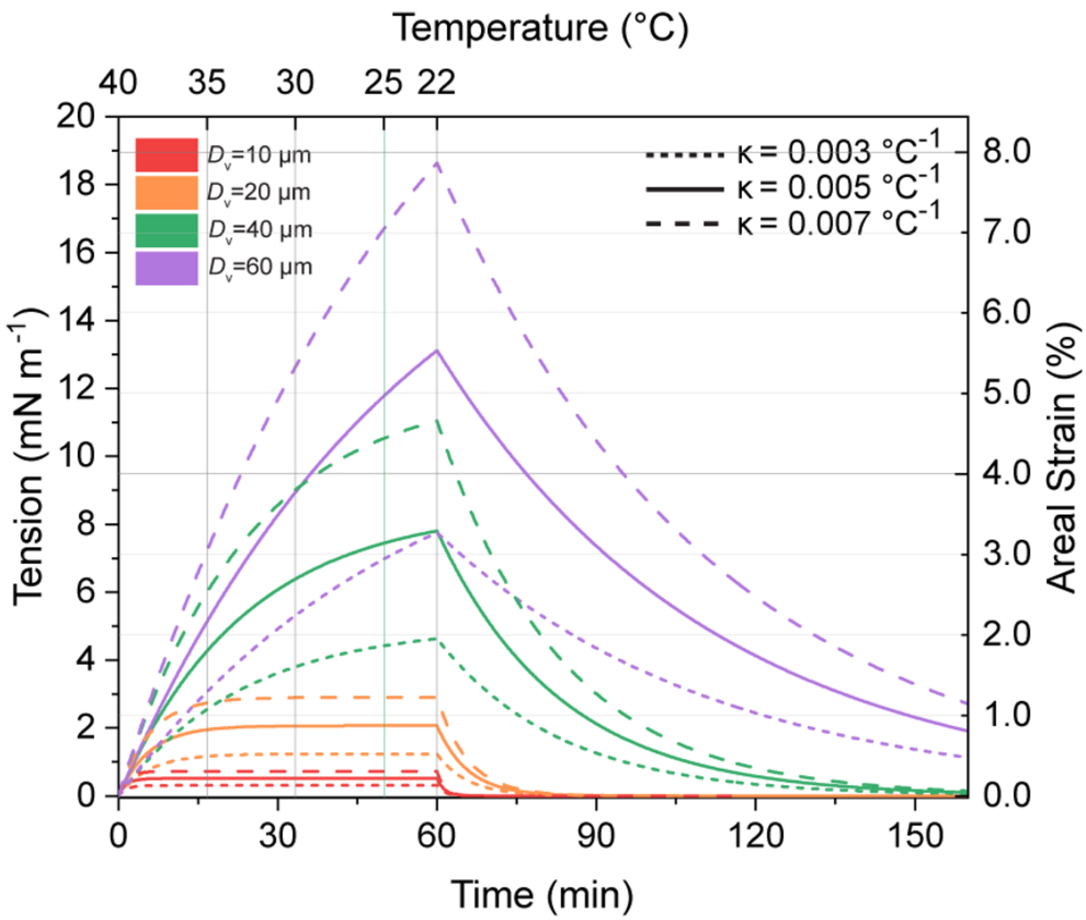


Figure S-4. Impact of coefficient of thermal expansion, on tension evolution during cooling and subsequent temperature hold near room temperature. Each color family represents a vesicle size while the line types show variations in coefficient of thermal expansion. Similar to the main paper, the upper axis estimates the temperature history for the case where vesicles become spherical and first develop tension at 40°C.

References

1. Wan, H., Jeon, G., Xin, W. Y., Grason, G. M. & Santore, M. M. Flower-shaped 2D crystals grown in curved fluid vesicle membranes. *Nature Communications* **15**, 3442 (2024).