Supporting information for

Tunable Crystallization via Osmosis-Driven Transport Across a Droplet Interface Bilayer

Experimental Details

Methods

We used an isolated individual aqueous microdroplet, which is manipulated by a micropipet under digital video microscopy (Olympus IX50 with the 40x phase contrast objective). A detailed description of our system has been reported.[i] In brief, commercially-available micromanipulation equipment with an inverted microscope has been used to generate and manipulate a microdroplet in an immiscible liquid environment. Once an individual microdroplet is generated by applying a slight positive pressure using a syringe, two droplets were made to contact without external force.

Materials and Preparations

All chemicals, of the highest purity available, were purchased from Sigma-Aldrich and used without additional purification. Aqueous solutions with osmolytes of various concentrations were prepared from purified, deionized water (18.2 MΩ•cm) using Millipore water purification system (Direct Q-3). For all our experiments, 1-Oleoyl-rac-glycerol [namely, monoolein, $CH_3(CH_2)_7CH{=}CH(CH_2)_7COOCH_2CHOHCH_2OH$] was used as the lipid and squalene or squalane or decane were used as the immiscible organic phase. Monoolein containing organic phase was prepared by dissolving directly into squalene or squalane followed by bath sonication for ~15 min at RT. All solutions were prepared immediately prior to use and filtered through a 0.2 μm Millipore syringe filter. A micropipet used for manipulation of aqueous microdroplets was prepared using a commercially available micropipet puller and subsequently hydrophobized using hexamethyldisilazane [$(CH_3)_3SiNHSi(CH_3)_3$] to inhibit wetting of the micropipet glass surface by the aqueous solution. All experiments were carried out at an ambient temperature of 24±1 °C. Vapor pressure osmometer (VAPRO: Wescor Inc.) was used to measure the osmolality.

Description of Movie

Crystallization of KPF_6 (6% w/v) droplet adherent to osmolyte containing NaCl 3.5M. A droplet pair is formed in squalene containing 9mM monoolein.

[i] K. Allain, R. Bebawee and S. Lee, *Cryst. Growth Des.*, 2009, **9**, 3183-3190.