**Captions for Movies**

**Movie S1**. Direct observation of a tautomerization translocation reaction (TTR) involving three HI-proteoids that occurs as a consequence of Brownian fluctuations in a system of crowded proteoids, measured using time-lapse optical photomicroscopy. Initially, a lone monomer is surrounded by lock-and-key dimers. As a consequence of Brownian fluctuations, a dimer, located adjacent to the monomer, de-dimerizes, so three monomers in proximity are observed. The initial monomer then forms a lock-and-key dimer with one of the proteoids resulting from the de-dimerization event, leaving a different monomer than the original one in a substantially different spatial position as a reaction product. Thus, looking only at the initial and final frames of the movie, a monomer appears to have diffused a large distance in a highly crowded environment of dimers. Thus, TTR effectively increases the rate of monomer diffusion in the crowded system, thereby facilitating the expulsion of monomers from growing crystallites. One frame has been recorded every three minutes in the laboratory; the movie plays at a speed of two frames per second. Scale: the outer radius of a HI-proteoid is 2.5 μm. Average area fraction of proteoids in the field of view is $\phi_A \approx 0.41$. (AVI; 2.6 MB).

**Movie S2**. Direct observation of a tautomerization translocation reaction (TTR) involving four HI-proteoids in the crowded Brownian system, measured using time-lapse optical photomicroscopy. Initially, two monomers are proximate to neighboring lock-and-key dimers. Entropic fluctuations cause de-dimerization of a dimer adjacent to these monomers, yielding four monomers in proximity. The initial monomers combine with the proteoids resulting from the de-dimerization, leading to a final state that has two monomers that are significantly displaced from the location of the initial monomers. This 4-proteoid HI-proteoid TTR also increases the rate of expulsion of monomers from growing dimer crystals. One frame has been recorded every 30 minutes in the laboratory; the movie plays at a speed of two frames per second. Scale: the outer radius of a HI-proteoid is 2.5 μm. Average area fraction of proteoids in the field of view is $\phi_A \approx 0.38$. (AVI; 5.2 MB).