Detailed Status Information

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Title	CryoEM Reveals the Complex Evolution of a Thermodynamically Unstable Phase in the Chemically Fueled Dissipative Assembly of a Disulfide Hydrogel
Manuscript Type	Article
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Authorship	Yes
Abstract	Inspired by the adaptability of biological materials, a variety of synthetic fuel-driven dissipative processes have recently been developed. A general hypothesis in the field is that that dissipative systems form unique structures compared to conventional self-assembly processes due to the non-equilibrium nature of dissipative chemistry. However, comparing dissipative and non-dissipative processes in chemically-fueled systems is challenging, which has limited our understanding of how these processes differ. Here, we use a chemically-fueled redox system where we can fully separate out the forward activation and backward deactivation reactions. We study the forward and backward reactions sequentially and synchronously (dissipative) using time-resolved cryoEM. The data shows that the dissipative process is more complex and heterogenous than the sequential process. Our key finding is that a thermodynamically unstable stacked nanorod phase observed in the backward reaction is sustained for ~ 6 hours in the dissipative process. Quantitative analysis of cryoEM data and kinetic Monte Carlo modelling show that the dissipative process is driven by multiple cycles of activation, deactivation, assembly, and disassembly, and diffusive kinetics and concentration gradients. The data shed light on how dissipative systems create unique structures and provide plausible design principles to develop and optimize dissipative materials with unique functions.
Subject Terms	Physical sciences/Materials science/Soft materials/Self-assembly Physical sciences/Chemistry/Materials chemistry/Soft materials/Self-assembly
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In Review	No, my co-authors and I would not like to benefit from *In Review*
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