Messy Chemistry Approach to Origins of Life

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While life is yet to be defined, the concept is typically associated with well-organized chemical reaction networks capable of sustainability, replication, and evolvability. It has been long noted that chemistry in prebiotically plausible model systems generate intractable heterogeneous mixtures of compounds. Messy chemistry hypothesizes that life started with complex reaction networks that are common to prebiotic chemistry, evolved as a complex reaction network into biochemistry that itself constitutes a complex reaction network. The goal of our work is to decipher the mechanisms and the principles of organization in the prebiotic messy reaction networks.

As a case in point, we focus on the abiotic synthesis of hyperbranched polyesters (HBPE). The topology of HBPE enforces globular structure and allows for a controlled microenvironment surrounding the core similar to globular protein structures present in enzymes. Similar to the way enzymes govern and organize the biochemical networks, we hypothesize that prebiotically plausible biomimetic HBPE-based complexes approximated the enzymatic function at the early stages of chemical evolution. We demonstrate that tertiary amine-bearing HPBE form hydrophobic pockets as a reaction-promoting medium for the Kemp elimination reaction [1]. Furthermore, we probe the ability of HBPE to support metal-sulfide particles that catalyze prebiotically and biologically relevant redox reactions.

The formation of HBPE is an attractive case in point for investigation of selectivity and memory in chemical systems. HBPE can be synthesized by subjecting multifunctional organic acids and alcohol mixtures to mild heating under solventless conditions [2]. This method, however, produces a multitude of polymeric products, linear, branched and crosslinked. Subjecting the polyesterification system to wetting-drying environmental cycles results in the selective predominant formation of branched products [3]. Finally, by invoking the transamidation process, we investigate the potential transition between HBPE to peptide systems while retaining the functionality of HBPE templates.

[1] Mamajanov & Cody (2017). *Phil. Trans. A* **375(2109)**, 20160357. [2] Mamajanov et al. (2014). *Macromolecules* **47**, 1334-43. [3] Mamajanov (2018). *ALife* **2018**, 580-581