

## Polyester microdroplets, coacervates, and other membraneless non-biological assemblies as primordial compartments

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Compartmentalization was likely essential for primitive chemical systems during the emergence of life, both for preventing leakage of important components, i.e., genetic materials, and for enhancing chemical reactions. Although life as we know it uses lipid bilayer-based compartments, the diversity of prebiotic chemistry may have enabled primitive living systems to start from other types of boundary systems. Here, we demonstrate membraneless compartmentalization based on prebiotically available organic compounds,  $\alpha$ -hydroxy acids ( $\alpha$  HAs), which are generally coproduced along with  $\alpha$ -amino acids in prebiotic settings. Facile polymerization of  $\alpha$  HAs provides a model pathway for the assembly of combinatorially diverse primitive compartments on early Earth. We characterized membraneless microdroplets generated from homo- and heteropolyesters synthesized from drying solutions of  $\alpha$  HAs endowed with various side chains. These compartments can preferentially and differentially segregate and compartmentalize fluorescent dyes and fluorescently tagged RNA, providing readily available compartments that could have facilitated chemical evolution by protecting, exchanging, and encapsulating primitive components. Protein function within and RNA function in the presence of certain droplets is also preserved, suggesting the potential relevance of such droplets to various origins of life models. As a lipid amphiphile can also assemble around certain droplets, this further shows the droplets' potential compatibility with and scaffolding ability for nascent biomolecular systems that could have coexisted in complex chemical systems. These model compartments could have been more accessible in a "messy" prebiotic environment, enabling the localization of a variety of protometabolic and replication processes that could be subjected to further chemical evolution before the advent of the Last Universal Common Ancestor. We also highlight other membraneless compartments generated from both biomolecules and non-biomolecules as a way to probe other possibilities of primitive compartments both on Earth and other extraterrestrial bodies.

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