

Water Microdroplets as Reaction Vessels: Abiotic Sugar Formation and Oligomerization

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This study investigates the role of aqueous microdroplets (between 200 nm to 1 μm in diameter) in the formation of monosaccharides and subsequent condensation reactions to produce disaccharides on a millisecond timescale, without the need for catalysts. The formose reaction, an abiotic pathway to monosaccharides, provides a mechanism of sugar formation from potential terrestrial aldehydes and ketones. Under laboratory conditions this reaction requires extreme pH, metal catalysts, and is prone to side reactions. The abiotic formation of more complex sugars, such as disaccharides, also requires extreme pH and produces a variety of products. Aqueous microdroplets, tiny water particles akin to small water aerosols have shown unique interfacial properties, where partial solvation of molecules at the surface, extreme surface pH, and increased mass transfer enable condensation reactions typically unfavorable in bulk water. The surface of these particles can expose chemicals encapsulated to a low water activity environment and extreme pH gradients. Microdroplets share chemical and physical properties with atmospheric and sea spray aerosols, where a high surface-to-volume ratio and water-air interface can facilitate reactions. **In addition, aerosols containing lipids have been shown to convert into vesicle structures upon landing in water, providing a link to protocell formation.** Using droplet generation techniques and mass spectrometric analysis, hexose sugars and pentose/hexose disaccharides were identified to be formed from prebiotic precursors in catalyst-free microdroplets. The synthesis of fructose and sorbose from glyceraldehyde or dihydroxyacetone, as well as that of xylose disaccharides from xylose and glucose disaccharides from glucose, was monitored via high resolution mass spectrometry and tandem mass spectrometry. The product of the glucose condensation reaction was found to be a mixture of six different disaccharides, including maltose and trehalose, and matches the reported product of a known acidic low water activity environment. Reaction yields were found to increase with decreasing droplet size for a maximum yield of 1.7 $\mu\text{g}/\text{min}$. This work has wide-ranging implications linking aerosols to biomolecule

formation. The discovered abiotic pathway produces molecules essential for metabolism and relevant to plant cell walls. Disaccharides' superior water stability suggests they could persist for further evolution, offering a novel route for future chemical evolution.

