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(G*) The role of ammonium ions in prebiotic RNA polymerization in hot-cold and hydration-dehydration cycles

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The RNA world proposed by Gilbert in 1986 envisions that the first genetic polymers that formed on the early pre-biotic Earth were based on RNA polymerization and replication. Ever since, researchers have tried to synthesize RNA without enzymes by using catalysts such as clay, lipids, and inorganic salts. We investigated the formation of RNA by using hot-cold and hydration-dehydration cycles in the presence of different compounds that have been reported or suggested for their catalytic function. This study uses a novel simulation chamber to mimic hot-cold and hydration-dehydration cycles with unprecedented resolution and duration. When analyzed by gel electrophoresis and Molecular Dynamics (MD) computer simulations, we found that ammonium salts are an excellent catalyst for RNA synthesis and can form RNA polymers of >150bp. Efficiency of different ammonium salts can be related to their molar Gibbs energy of formation (ΔG): the smaller ΔG the better due to the formation of smaller crystallites that allows for more uniform mixing. Importantly, we find that the length of RNA-polymers is not directly related to the number of cycles. Irregular cycling, with a more prolonged hot dehydration phase, increases the polymerization rate while prolonged humid phases were detrimental to polymerization. From MD simulations, nucleotides form tight clusters in the aqueous phase in the presence of ammonium ions, driven by hydrogen bonding between ions and nucleotides. The ability to form these hydrogen bonds makes ammonium different from other inorganic salts. Nucleotides in these clusters form 'pre-polymers'when adsorbing and stacking on a substrate during the dry phase, ready to form phosphodiester bonds via condensation reactions during the hot and dry phases of the cycles.

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