RNA Adsorption and Polymerization: Mineral Surface-Catalyzed Non-enzymatic Polymerization of Ribonuleotides

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Non-enzymatic RNA polymerization is a major challenge in understanding the origin of life on early Earth. The polymerization of activated mononucleotides in the presence of montmorillonite clay was achieved three decades ago but the molecular interactions promoting this reaction remain unknown. It is generally assumed in the literature that minerals with a high adsorption capacity for mononucleotides would favor polymerization efficiency; however, this relationship has never been shown. Here we examined the relationship between ribonucleotide adsorption affinity and the polymerization catalytic efficiency of minerals, and also explored the mechanism of polymerization. Results showed that the activated mononucleotide, 2-methylimadzolide of adenosine monophosphate (2-MeImPA), has an ~10 times higher adsorption capacity on zincite (ZnO) than on montmorillonite, but only montmorillonite acts as a catalyst for 2-MeImPA polymerization. The orientation of the adsorbed activated mononucleotide was determined on zincite and montmorillonite using Attenuated Total Reflectance FTIR and Cross Polarization-Magic Angle Spinning ^{31}P NMR. Spectroscopic results showed that 2-MeImPA mononucleotides adsorb strongly on ZnO via the phosphate moiety, making it unavailable for polymerization. In contrast, mononucleotides adsorbed on montmorillonite by weak H-bonding or van der Waals interactions involving the amine group, thus leaving the phosphate moiety available for polymerization. Thus, providing a favorable orientation of the monomer, rather than a high adsorption capacity, is important for a mineral catalyze RNA polymerization. Furthermore, the mineral should also provide a nanoconfined environment in order to promote polymerization.