

From adsorption to reaction: A study of nucleotide-mineral interactions

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Interfacial chemistry has been proposed as a key element in the (geo)chemical evolution of prebiotic organic systems. In the context of the origin of life on early Earth, mineral surfaces are often invoked as multivalent inorganic supports for critical steps such as abiotic precursor synthesis, condensation and polymerization reactions, and vesicle formation. Among these minerals, phyllosilicates have received considerable attention. For example, montmorillonite clays are often used in prebiotic synthesis experiments to facilitate nucleotide polymerization in aqueous solutions. It is notable, however, that not all montmorillonite samples are catalytic, and those that are require chemical pretreatment. More recent experiments confirm that nucleotide polymerization is also induced by carbonate and sulfide minerals, suggesting that mineral catalysis could be more widespread than previously thought. Thus, interactions between organic monomers and mineral surfaces are a key aspect of prebiotic chemistry, though little is known about the mechanisms of adsorption, concentration, and subsequent reactivity of monomers on mineral surfaces.

Our recent studies focused on the adsorption of nucleotides on phyllosilicates under geochemically relevant conditions and demonstrated that (i) the canonical nucleotides exhibit homologous nucleotide-phyllosilicate interactions; (ii) ligand exchange is the main adsorption mechanism between the phosphate group and the metal hydroxyls of the lateral surfaces; (iii) the interaction of nucleotides on swelling clays, compared to non-swelling ones, is very sensitive to chemistry (pH, salinity). Here we expand on these results to consider the effects of mineral-monomer interactions on the condensation reactions that follow nucleotide adsorption. The efficiency of nucleotide polymerization is examined as a function of mineral characteristics, including cation exchange capacity, mineral surface area, mineral structure, hydration state, and the formation of minor secondary phases. The effects of environmental parameters, such as T, P, pH, and salinity, on both mineral characteristics and polymerization is used to develop a mechanistic interpretation of polymerization in water-rock systems that were ubiquitous on the early Earth.