

Research Paper

Sequence Analysis of Trimer Isomers Formed by Montmorillonite Catalysis in the Reaction of Binary Monomer Mixtures

GÖZEN ERTEM,^{1,2,3} ROBERT M. HAZEN,¹ and JASON P. DWORKIN²

ABSTRACT

Oligonucleotides are structurally similar to short RNA strands. Therefore, their formation via non-enzymatic reactions is highly relevant to Gilbert's RNA world scenario (1986) and the origin of life. In laboratory synthesis of oligonucleotides from monomers, it is necessary to remove the water molecules from the reaction medium to shift the equilibrium in favor of oligonucleotide formation, which would have been impossible for reactions that took place in dilute solutions on the early Earth. Model studies designed to address this problem demonstrate that montmorillonite, a phyllosilicate common on Earth and identified on Mars, efficiently catalyzes phosphodiester-bond formation between activated mononucleotides in dilute solutions and produces RNA-like oligomers. The purpose of this study was to examine the sequences and regiospecificity of trimer isomers formed in the reaction of 5'-phosphorimidazolides of adenosine and uridine. Results demonstrated that regiospecificity and sequence specificity observed in the dimer fractions are conserved in their elongation products. With regard to regiospecificity, 61% of the linkages were found to be RNA-like 3',5'-phosphodiester bonds. With regard to sequence specificity, we found that 88% of the linear trimers were hetero-isomers with 61% A-monomer and 39% U-monomer incorporation. These results lend support to Bernal's hypothesis that minerals may have played a significant role in the chemical processes that led to the origin of life by catalyzing the formation of phosphodiester bonds in RNA-like oligomers. **Key Words:** Origin of life—RNA—HPLC—Mineral catalysis—Montmorillonite—Oligonucleotides. *Astrobiology* 7, 715–722.

INTRODUCTION

THE POSSIBLE ROLE OF MINERALS as a catalyst in the processes that lead to the abiotic formation of bio-molecules from monomers was first proposed by Bernal as early as 1949 (Bernal, 1949). Research carried out to test this hypothe-

sis, namely, the role of mineral catalysis on the formation of RNA-like oligomers, has been reviewed (Ertem, 2004; Ferris, 2006; Shapiro, 2006). Model studies demonstrate that the clay mineral montmorillonite catalyzes the condensation of activated monomers in an aqueous electrolyte solution at pH 8, which produces RNA-like

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Oligonucleotides are structurally similar to short RNA strands. Therefore, their formation via non-enzymatic reactions is highly relevant to Gilbert's RNA world scenario (1986) and the origin of life. In laboratory synthesis of oligonucleotides from monomers, it is necessary to remove the water molecules from the reaction medium to shift the equilibrium in favor of oligonucleotide formation, which would have been impossible for reactions that took place in dilute solutions on the early Earth. Model studies designed to address this problem demonstrate that montmorillonite, a phyllosilicate common on Earth and identified on Mars, efficiently catalyzes phosphodiester-bond formation between activated mononucleotides in dilute solutions and produces RNA-like oligomers. The purpose of this study was to examine the sequence and regioselectivity of trimer isomers formed in the reaction of 5'-phosphorimidazolides of adenine and uracil. Results demonstrated that regioselectivity and sequence specificity observed in the dimer fractions are conserved in their elongation products. With regard to regioselectivity, 87% of the linkages were found to be RNA-like 3',5'-phosphodiester bonds. With regard to sequence specificity, we found that 86% of the linear trimers were heterotrimers with 47% A monomer and 39% U monomer incorporation. These results lend support to Berna's hypothesis that minerals may have played a significant role in the chemical processes that led to the origin of life by catalyzing the formation of phosphodiester bonds in RNA-like oligomers. **Key Words:** Origin of life—RNA—HFIC—Mineral catalysis—Montmorillonite—Oligonucleotides. *Astrobiology* 7, 715–722.

INTRODUCTION

It is generally accepted that the role of mineral catalysis in the formation of RNA-like oligomers, has been demonstrated (Loren, 2004; Hazen, 2006; Hazen et al., 2006; Dworkin et al., 2006). Model studies demonstrate that the clay mineral montmorillonite catalyzes the condensation of activated monomers in an aqueous electrolyte solution at pH 9, which produces RNA-like

oligonucleotides. In particular, the role of mineral catalysis in the formation of RNA-like oligomers, has been demonstrated (Loren, 2004; Hazen, 2006; Hazen et al., 2006; Dworkin et al., 2006). Model studies demonstrate that the clay mineral montmorillonite catalyzes the condensation of activated monomers in an aqueous electrolyte solution at pH 9, which produces RNA-like

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