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Reaction networks and free-energy landscapes via a novel topological approach: ab initio Miller-like experiments and beyond

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The qualitative and quantitative description of chemical reactions is a formidable problem even in textbook cases. Determining the right pathways, their energetics and kinetics needs costly accurate quantum approaches. Typically this amounts to a simplification of gas-phase reactions into elementary chemical steps, which, in turn, are usually translated to the same reactions in solution. The topology-based method hereby presented [1] allows instead a fully unbiased exploration of the reaction space of both the gas-phase and the aqueous-phase regimes.

We recently reported on the first computer simulation of the celebrated Miller experiment [2], carried out with a standard, density-functional theory-based ab initio molecular dynamics approach. Quite unexpectedly, we observed that formamide is a crucial centerpiece towards the "Miller-like" synthesis of glycine. We presently adopt our new method to fully explore the reaction network of formamide, which reveals very different between the gas and the solution regimes, indicating, in the liquid phase, new pathways, intermediates, transition states, and reaction products. Even more importantly, the new scheme allows to demonstrate the comparable relative stability of formamide with respect to both hydrogen cyanide and formic acid, thus helping settle a heated current debate in the prebiotic chemistry community [3]. The generality of our method allows now applications in many disparate fields of materials synthesis and design.

References:

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