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The understanding of the formation of biopolymers in the early ages of the planet Earth is crucial to rationalize the origin of life on our planet. In which conditions and how organic compounds polymerized in the primordial soup, before the emergence of the enzymes, remains a fascinating still unresolved issue in prebiotic chemistry. In particular the first peptide bond condensation reaction is still highly debated and controversial. The peptide bond formation is known to be disfavored in bulk water for both thermodynamic and kinetic reasons, and many reaction mechanisms in “exotic conditions” have been proposed.

Recently, the group of Prof V.Vaida in the USA has been able to observe the formation of polypeptides from Leucine ethyl ester amino acids at the air/water interface, the whole process being catalyzed by CuCl_2 salt. This work thus has suggested the surface of oceans and of marine aerosols as suitable environments for the birth of life on Prebiotic Earth. Both Leu-Ethyl esters and the CuCl_2 salt are compatible with known geochemical conditions of the primitive Earth, where air/water interfaces were furthermore ubiquitous in oceans or prebiotic salty water droplets.

What remains to be understood is the actual role of the air/water interface in catalyzing the peptide bond chemical reaction formation. In particular many organic reactions have been shown to be strongly accelerated at the air/water interface with respect to bulk water but microscopic insights are still lacking.

This is the goal of the first-principles molecular dynamics (FP-MD) simulations performed in this PRACE work, especially applying accelerated *ab initio* DFT molecular dynamics in order to directly assess energetics and mechanisms of the chemical reaction(s) at play.

For the first time to our best knowledge the peptide bond formation at the air/water interface and in the liquid water have been simulated in order to unravel the reasons for the air/water interface to be a key ingredient in this reaction.

Our simulations revealed the energetic stability of a necessary ‘reactive’ complex between the amino acids and the copper salt at both aqueous environments.

Coupling our simulations and in-situ experiments from Vaida’s group, we unambiguously detect and characterize a stabilization effect directly modulated by the interfacial water network.

The findings that the air/water interface stabilizes a ‘reactive’ complex different from the ‘one, stable in bulk water, is a novel ingredient in chemistry that can possibly be extended to other water ‘restraining/confined’ environments, allowing chemical reactions to occur.

Another key result is the essential-role of Cl^- enrichment and ion pairing at the air/water interface, for the peptide bond formation to occur at the interface. The CuCl_2 salt is found with a very important catalytic role all along the peptide bond formation.

The PRACE CPU 29.7 million hours of computing time on the GENCI-Curie Machine in France has allowed us to take into account “huge” quantity of water in the simulation boxes (512 water molecules not otherwise easily accessible to DFT-MD) in order to well reproduce the complex organization water at the interface along the peptide bond formation, and to unambiguously reveal its novel catalytic roles and associated salt effects.

Our next step will be to complement the present theoretical investigation by explaining possible stereo chemical effects on the unraveled prebiotic peptide bond condensation at the air/water interface that might explain some reasons for the development of the L-Chirality in biological systems.