Theoretical Mechanistical Study of CO2 Reduction to CH_4 by a Bio-Inspired NiFe Hydrogenase Model on graphite.

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In a recent article of ACS Energy Letters[1], the possibility of converting carbon dioxide into methane with the exception of any other carbon species was experimentally shown. This reaction represents the grail in the field of renewable energies allowing the recycling of CO2 in the most reduced source of carbon. This remarkable result has been achieved with a Nickel-Iron complex on graphite surface, shown in Fig.1. Notoriously difficult to achieve and faced with these results, the immediate question is: what is the mechanism?

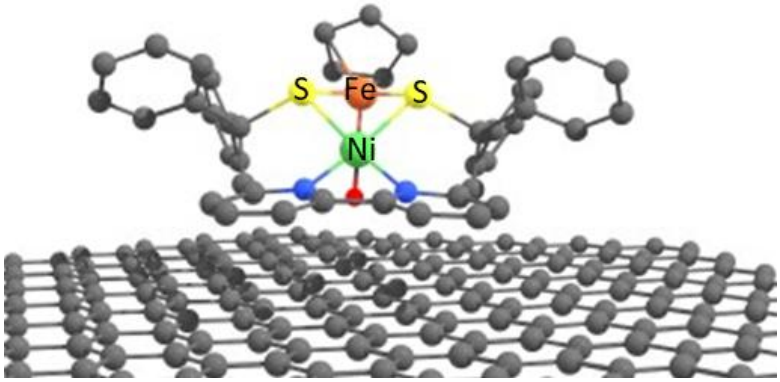

Figure 1.Ni-Fe complex on a graphene sheet: a model for computational studies

In this presentation, we will present our results about the first steps of this mechanism. First, we will show how the complex interacts with the graphite surface. For this, we have used the SurfOnDock[2] to generate the geometry for molecules on surfaces and find the most stables. In a second step, we will show how CO2 interacts and coordinates with the complex and we will study the possible sites of protonation. We will also present results on the competitive mechanism of H_2 formation.[3]

[1] M. E. Ahmed, S. Adam, D. Saha, J. Fize, V. Artero, A. Dey, C. Duboc *ACS Energy Lett.* **2020**, *5* (12), 3837–3842. https://doi.org/10.1021/acsenergylett.0c02002.
[2] C. Martí, S. Blanck, R. Staub, S. Loehlé, C. Michel, S.N. Steinmann DockOnSurf: *J. Chem. Inf. Model.* **2021**, *61* (7), 3386–3396. https://doi.org/10.1021/acs.jcim.1c00256.
[3] M. E. Ahmed, S. Chattopadhyay, L. Wang, D. Brazzolotto, D. Pramanik, D. Aldakov, J. Fize, A. Morozan, M. Gennari, C. Duboc, A. Dey, V. Artero *Angewandte Chemie International Edition* **2018**, *57* (49), 16001–16004. https://doi.org/10.1002/anie.201808215..