

Adsorption of CO₂ and OCCO on Graphene Surface with Defects

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Abstract

Graphene doped with N acts as a metal-free electrode, it has up to three times more catalytic activity than pure graphene and achieves almost the catalytic activity of the Pt. So, it has been proposed as a future alternative to Pt since it would be cheaper and easier to produce on an industrial scale.

We report a computational analysis using standard DFT calculations and Molecular Dynamics simulations. In specific, we studied the adsorption properties, electronic properties, geometries, and stabilities of the adsorption of CO₂ and OCCO molecules on graphene surfaces with induced defects, using DFT- PBE methodology with the dispersion correction Grimme D3 and ultrasoft pseudopotentials. The selected defects are a graphitic-N type, one vacancy in the graphene sheet, Pyridinic-N type. All systems are compared with the pure graphene.

It can be seen that graphene with different types of defects has weak interactions with CO₂ and OCCO molecules. These weak interactions are marked by the type of defect of the surface. Although the defects studied are not strong enough to induce greater attraction to the molecules. The most stable systems are graphitic-N based systems, which present *n*-type doping, pulling electrons from the CO₂ and OCCO molecules.

Nevertheless, the system with greater reactive activity after the adsorption of

CO₂ and OCCO is the pyridinic-N based-systems, inducing an increase of available states in the Fermi energy and in the DOS virtual zone. This makes the OCCO molecule an excellent candidate to be used in catalytic processes, like transform CO₂ to ethanol or other industrial product. This is feasible since N-doped graphene based systems are the best systems for the adsorption of the OCCO molecule and its possible electrochemical conversion to ethanol.

References

- [1] Song Y., Peng R., Hensley D.K., Bonnesen P.V., Liang L., Wu Z., Meyer III H.M., Chi, M., Ma C., Sumper B., Rondinone A.J., *Chemistry-Select*, 1, (2016), 6055-606.