Stepwise hydrogenation and fluorination of graphene towards graphane and fluoride

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A great efficacy of molecular quantum chemistry applied to basic graphene problems has been recently demonstrated by the authors when studying the formation of peculiar composites between carbon nanotubes and graphene [1] as well as considering tensile deformation and fracture of a graphene sheet in due course of a mechanochemical reaction [2]. The optimistic results obtained in the studies make it possible to shift attention from the solid state problems and consider chemical modification of graphene as a multistep addition reaction applied to the pristine molecule. The paper concerns the reactions that involve atomic addends such as hydrogen and fluorine. To proceed we have to answer the following questions: 1) what is a characteristic image of the addend atom attachment to the substrate; 2) which carbon atom (or atoms) is the first target subjected to the attachment and how carbon atoms are selected for the next steps of the adsorption; 3) is there any connection between the sequential adsorption pattern and cyclohexane-like conformers? First results obtained on this way are presented in the current paper. The calculations were performed within the framework of unrestricted broken symmetry Hartree-Fock approach by using semiempirical AM1 technique implemented in CLUSTER-Z1 codes.

Our study has convincingly shown that similarly to fullerenes and carbon nanotubes [3], the formation of polyhydride and polyfluoride graphene can be considered in the framework of stepwise computational synthesis, each subsequent step of which is controlled by the distribution of atomic chemical susceptibility in terms of partial numbers of effectively unpaired electrons on atom, N_{DA}, of preceding derivative over the substrate atoms. The high-rank N_{DA} values definitely distinguish the atoms that should serve as targets for the next chemical attack. The performed investigations have shown that there is a direct connection between the state of graphene substrate and the conformer pattern of the polyderivatives formed so that chair-like regular structures can be obtained only under particular conditions.

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[2] E.F. Sheka, N.A. Popova, V.A. Popova, E.A. Nikitina, L.Kh. Shaymardanova. *J. Exp. Theor. Phys*. **139**(4), (2011).
[3] E.F. Sheka. Fullerenes: Nanochemistry, Nanomagnetism, Nanomedicine, Nanophotonics. CRC Press, Taylor&Francis group: Boca Raton 2011.