Designing graphene for energy applications

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Graphene is currently considered one of the most promising materials for applications in the field of energy storage and harvesting [1,2]. However, for many of these applications, its interactions with different chemical species and/or with external stimuli must be considered.

The enhanced reactivity of rippled graphene has been theoretically studied [3] and proposed as a mean to create partially hydrogenated graphene structures [4] for nano-electronics [5]. We recently showed with Density Functional Theory (DFT) based simulations that this property could also be used for a hydrogen storage device working at room temperature [6]. The loading phase (chemisorption on convexities) has been already experimentally demonstrated using naturally rippled graphene grown on SiC [7]. However, to develop a device, two main issues must be addressed (i) the creation of a multilayer and (ii) the fine control of the local curvature. Here we present results concerning both of them, exploiting the interplay between decorated/functionalized graphene with external electric fields and/or optical stimuli.

We already explored the possibility of mechanical control of the curvature by means of coherent transvers phonons [6]. Other possibilities are optical control by functionalization with photosensitive molecules (which could be used also as spacers for multilayers structures), or electro-mechanical control, by means of external electric fields. This, however, imply enhancing the electro-mechanical coupling of graphene. Here we explore the possibility of realizing this by mean of B and N substitutional doping. This is done using a multi-scale simulation and modeling approach combining DFT, to evaluate the electronic, chemical and mechano-electric properties of pristine and substituted/functionalised graphene, with empirical force field based atomistic molecular dynamics simulations to evaluate the behavior on the large scale (100nm). A continuum description of the graphene sheet with physic-chemical properties mapped on it[8-9] is used to explore macroscopic scales and statistical behavior. This approach combines the accuracy with the large scale view. We reports evaluations of the macroscopic properties, such as the gravimetric capacity for H storage, which is seen to reach the required DOE goals, and the kinetics of response to external stimuli, which are predicted acceptable for room temperature use and compared with preliminary experimental results

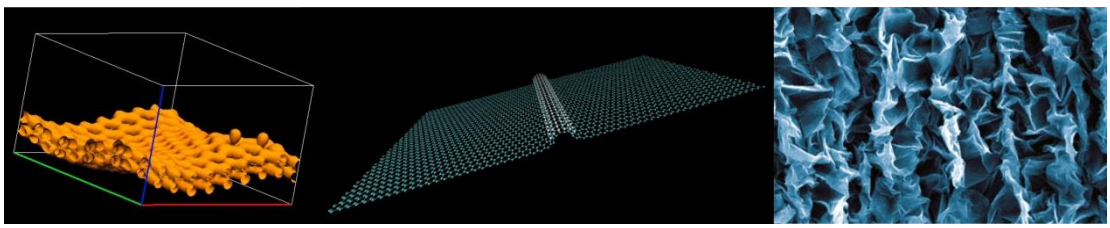

Fig 1 The three levels of the multi-scale representation of graphene. From left to right, a 2nmx2nm supercell of rippled graphene with a dimer of hydrogen atoms attached. A iso-charge surface of energy states near the Fermi level is represented in orange; a ~1000 atoms supercell of graphene with an hydrogenated ripple; a surface representation of buckled graphene.

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