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Disordered Graphene Materials: Atomistic Characterization and Performances

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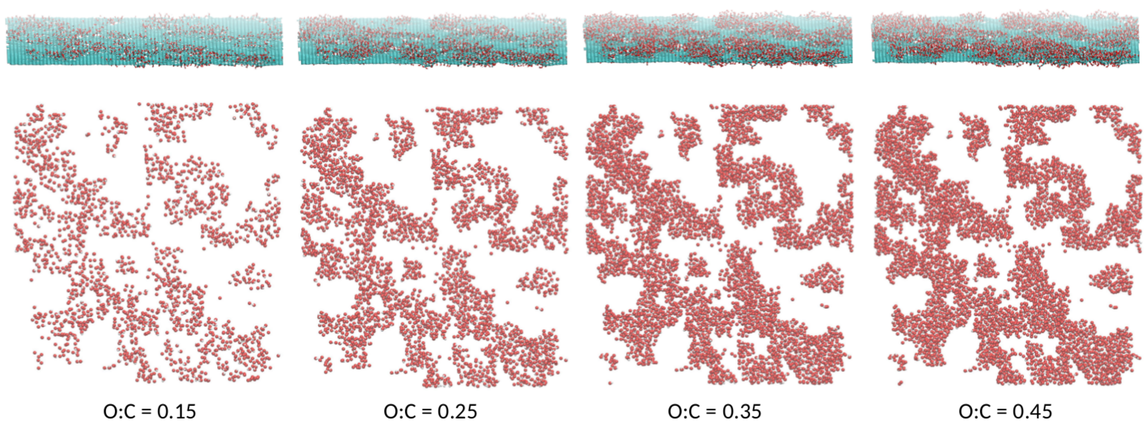
INTRODUCTION

Formidable progress has been recently achieved in the fabrication and characterization of disordered materials with unprecedented properties. In this context, particular forms of disordered graphene (reduced graphene oxides), obtained by chemical exfoliation techniques, have been found suitable to improve the performances of composite materials, with application in energy. Moreover, the recent demonstrated possibility to synthesize wafer-scale two-dimensional amorphous carbon monolayers, structurally dominated by sp2 hybridization has initiated a new platform of low-dimensional materials to explore as alternative forms of membranes with enhanced chemical reactivity which could serve as coating materials [1,2]. The excellent physical properties of the mentioned materials derive from the nature and degree oftheir disorder which, controlled at the fabrication level, represents the key ingredient to tune their physical/chemical properties for specific target applications. In this respect, new fabrication strategies to modify the degree of disorder and a systematic theoretical characterization of the impact of the material structural quality on the ultimate performance is urgent. In this poster we present the results of our theoretical investigation on possible strategies to improve the (thermal) reduction process of graphene-oxides and the consequent possibility to recover the quality of pristine graphene [3]. Moreover, we present a systematic analysis of the structural and vibrational properties of amorphous carbon monolayers as a function of the structural quality of the material, showing how disorder results in a tunable thermal conductivity varying by more than one order of magnitude [4]. Our simulations provide some recipe to design most suitable "amorphous graphene" based on the target applications such as ultrathin heat spreaders, energy harvesters or insulating thermal barriers.

Reduced Graphene Oxide: Impact of Thermal Reduction

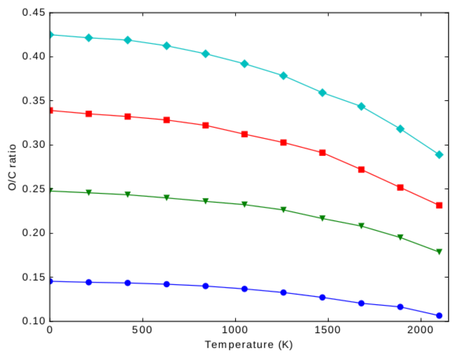
GO with varying O/C ratio in a fixed spatial distribution of oxidized surface



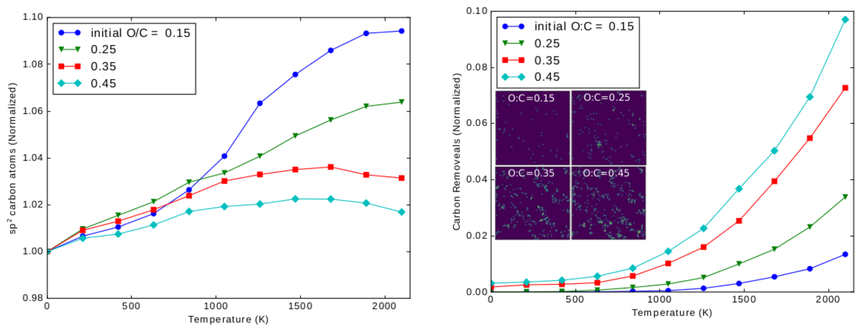
O:C = 0.15 O:C = 0.25 O:C = 0.35 O:C = 0.45

The initial O/C ratio is 0.15, 0.25, 0.35, 0.45 respectively. As a preliminary analysis, we monitored the amount of Oxygen adsorbed onto the surface at different annealing temperatures. Below 400K, heat energy is not sufficient to trigger the desorption of impurities from the graphene plane.

A stronger reduction of the oxygen content for higher functionalizing densities: for a given annealing temperature, the higher the initial density of oxygen-containing species, the higher the amount of desorbed oxygen.

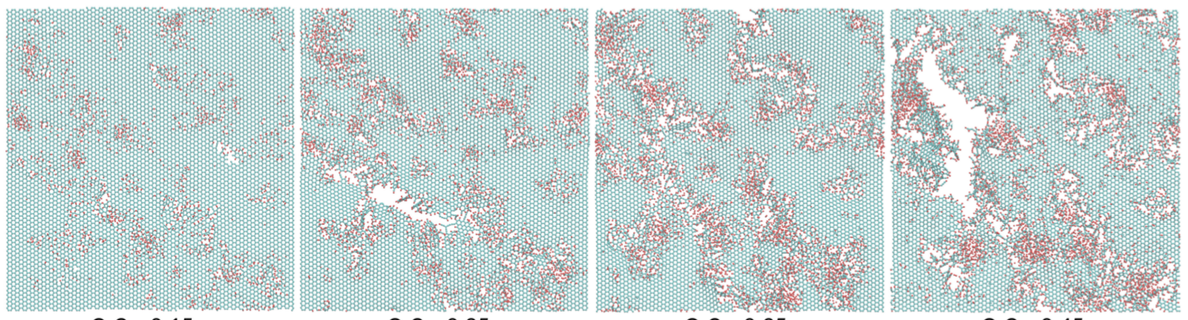


Systems with a higher concentration of oxygen groups undergo a weaker reduction process and only a small percentage of carbons goes back to the unoxidized state. The most probable reactions in highly-oxidized samples involve the removal of a carbon from the sheet.



The increasing number of vacancy formations could even engender the removal of the entire oxidized area leaving a hole

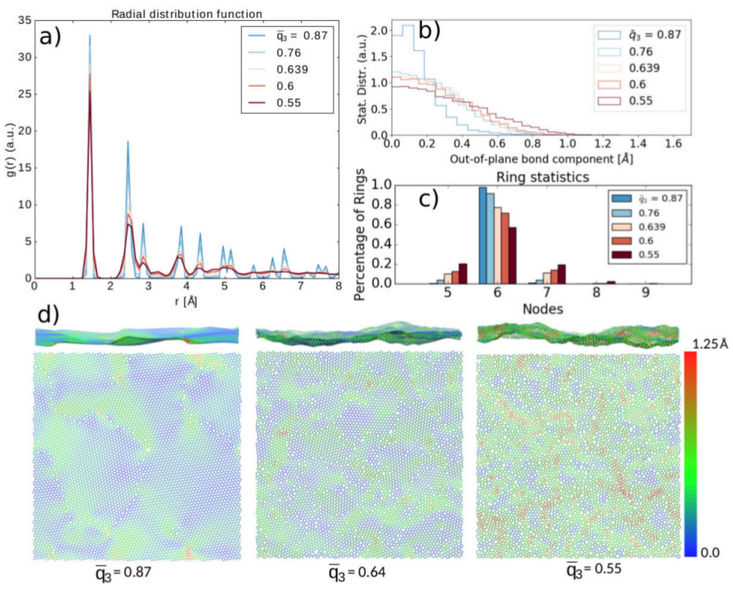
Samples after annealing at T = 1500K



O:C = 0.15 O:C = 0.25 O:C = 0.35 O:C = 0.45

Amorphous Graphene: Impact of Disorder on Vibrational and Thermal Properties

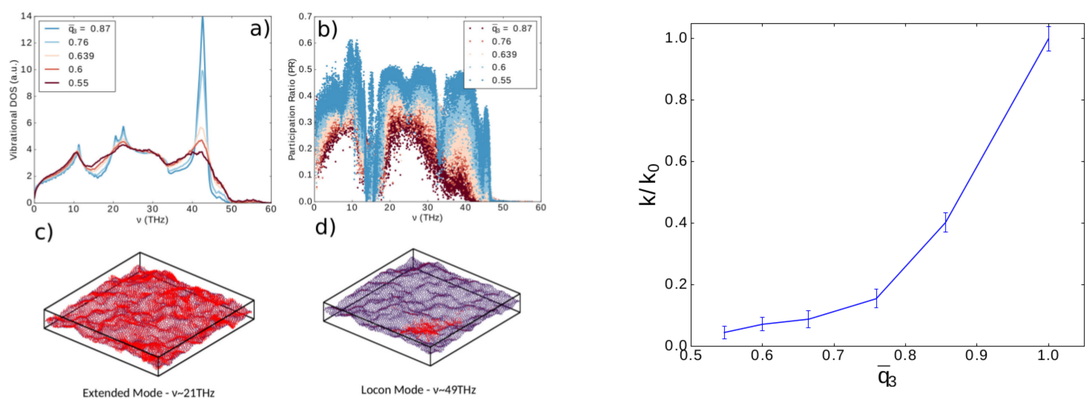
Amorphous Graphene Samples built via classical Molecular Dynamics: Structural Analysis



$\bar{q}_3$ = 0.87 $\bar{q}_3$ = 0.64 $\bar{q}_3$ = 0.55

The radial distribution functions g(r) (RDFs) of the samples as a function of the degree of amorphousness (a), quantified via the averaged triatic order parameter q3. Smaller values of q3 denote more amorphous systems. The same position of the first peak in the RDFs is found in any sample, suggesting that the average C-C bond length in the first coordination shell is similar in all the structures. There is a significant broadening in the second and third peaks of the RDF, denoting an increased disorder in the structure of a-G. a-G structures exhibit somestatic intrinsic ripples: specifically, strongly wrinkled structures correspond to the more amorphous ones.

Vibrational and Thermal Analysis



The VDOSs present peaks which are clearly reminiscent of the phonon bands of graphene. The higher the degree of amorphousness, the broader the peaks which eventually merge intoa continuous VDOS without frequency gaps.
Participation Ratio (PR) decreases with amorphousness > stronger mode localization with increasing disorder.
Thermal conductivity reduction of more than one order of magnitude is found wrt to pristine graphene.

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