Graphene oxide and simple molecules at high pressure: new perspectives for 2D nanoconfined chemistry of carbon based materials

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Graphene oxide (GO) has recently emerged as a versatile platform material for the large-scale synthesis of graphene and for the chemical functionalization of related 2D materials. In particular N-doping of these systems is a highly relevant topic for potential applications to energetic and environmental issues.[1] From the structural point of view, the intriguing properties of GO are related to its layered structure featuring sufficiently large interlayer spacing for molecular insertion.[2] Whereas a large variety of methods have been reported to explore the reactive behavior of GO at ambient pressure, mainly at high temperature in gas phase or in solution, with the only exception of a few structural studies, the high pressure chemistry of this material remains substantially unexplored.

The idea of this study is to use pressure for inserting molecule between the GO layers and realize high density conditions, where bond breaking and reconstruction can occur, leading to the incorporation of heteroatoms or to the functionalization of the carbon framework by molecular fragments. For this purpose the room temperature high pressure behavior of pure GO and of GO in presence of atomic and molecular systems of increasing reactive character, such as Ar, N_2 and NH_3, were studied using a membrane Diamond Anvil Cell (DAC) by means of X-ray diffraction, FTIR and Raman spectroscopy. Electronic photoexcitation was also employed to generate highly reactive species in high density conditions for activating chemical reactivity.

The presented results indicate the substantial stability of the underlying layered structure of GO at the investigated pressure and provide indication for the high pressure incorporation of nitrogen functionalities within the carbon framework, thus suggesting an innovative approach for the chemical functionalization of nanostructured graphene related systems and opening new perspectives for the synthesis of 2D advanced functional materials.[4]

References

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