

The law of attraction: computational insights into the role of non-covalent interactions in graphene-based sensing

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The field of chemical sensing with graphene-based materials (GBMs) is quickly developing. The high surface-to-volume ratio and the tunability of the functionalization of graphene allow for a large scope of analytes and the production of efficient electrochemical sensors [1]. One of the challenges is to rationalize how different functionalized graphene derivatives work, in order to optimize the detection performances. Computational chemistry is an important tool in understanding the mechanism of sensing and to unravel the main factors influencing it. In this work we consider the adsorption of organic analytes on the surface of the graphene-based sensor and the role that it plays in the detection performance. To understand how different functionalizations of GBMs influence the adsorption strength, computational methods are employed to analyse these non-covalent interactions. We chose to use two nitro-aromatic contaminants (NACs) as model analytes: di-nitro-toluene (DNT) and tri-nitro-toluene (TNT) [2]. The results show that the adsorption is mainly driven by dispersion forces rather than by electrostatic ones. These findings agree with experimental results on analytical performances [2-3], showing that the adsorption step of the sensing process is the one determining the limit of detection and the selectivity of an electrochemical sensor. We can now hypothesize that graphene derivatives with a high degree of aromaticity and high nitrogen content are good candidates for detecting nitro-aromatic pollutants, while highly oxidized GBMs are less effective.

References

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Figures

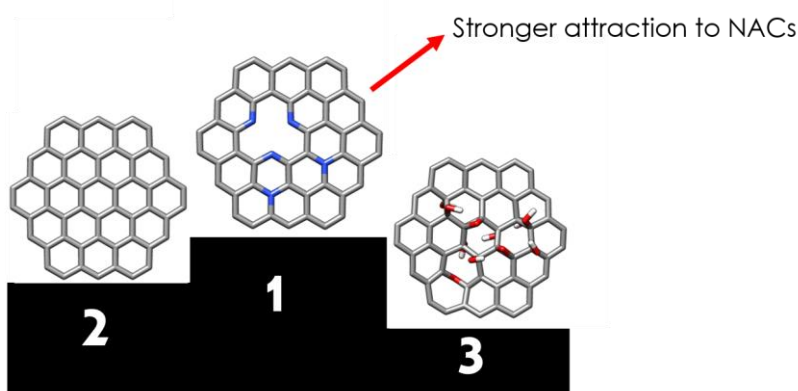


Figure 1: Different GBMs establish non-covalent interactions of different strengths with nitro-aromatic contaminants, leading to different analytical performances.