Structural Models for Carbon Nanodots: Graphitic, Nitrogen-doped and Graphyne-Based systems.

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Graphyne and graphdiyne represent two-dimensional carbon allotropes structurally related to graphene. Unlike graphene, which is composed exclusively of sp^2-hybridized carbon atoms arranged in a honeycomb lattice, graphyne and graphdiyne incorporate sp-hybridized carbon atoms through acetylenic linkages. In graphyne, aromatic rings are connected by single –C≡C– units, while in graphdiyne they are linked via diacetylenic –C≡C–C≡C– bridges. These structural motifs introduce intrinsic porosity and significantly modify the electronic properties, leading to tunable band structures, enhanced charge transport characteristics, and well-defined nanoporous architectures suitable for energy conversion, sensing, and photocatalytic applications.[1] [2]

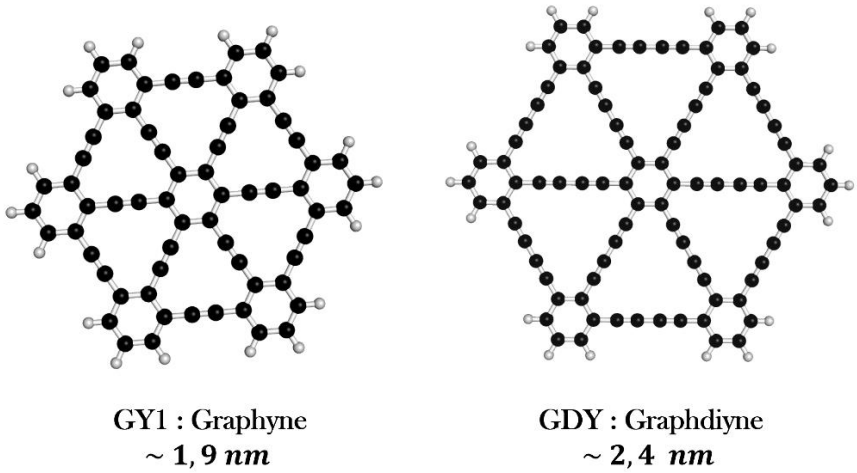

Figure 1: The two structures are shown in the figure, with graphyne on the left and graphdiyne on the right.

Carbon dots (CDs) are emerging luminescent carbon-based nanomaterials that have attracted significant attention due to their unique photoluminescence, low toxicity, chemical stability, and excellent biocompatibility. Typically considered quasi-zero-dimensional systems, CDs consist of a hybrid sp^2/sp^3 carbon core functionalized with oxygen- and nitrogen-containing groups, making them highly versatile for applications in bioimaging, optoelectronics, and photocatalysis.

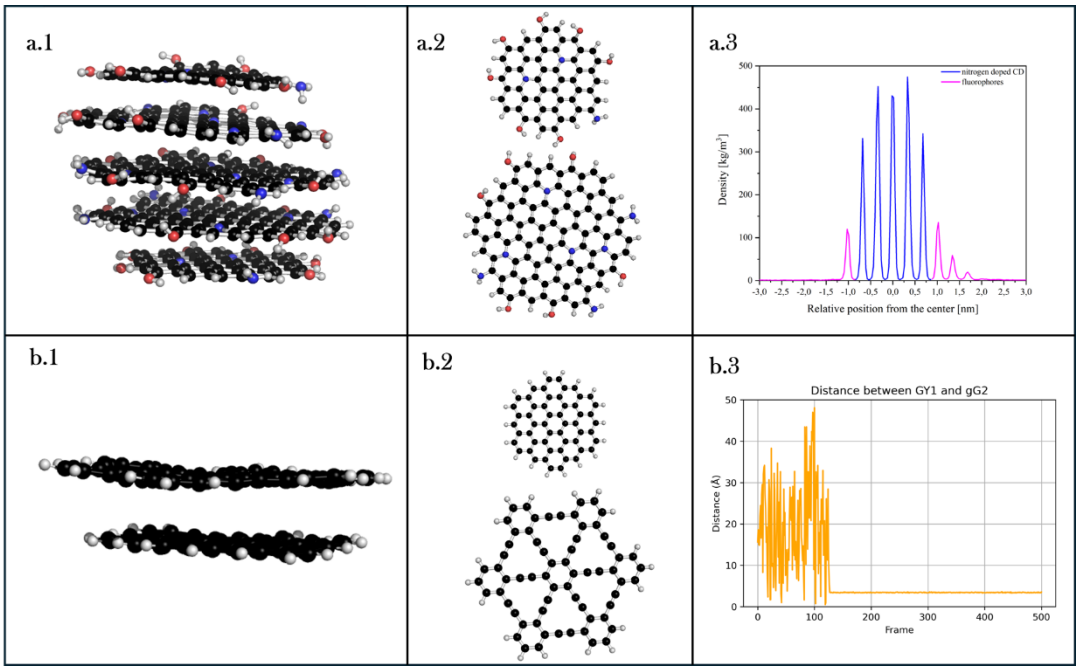

Figure 2: a.1 Nitrogen doped Carbon Nanodot, **a.2** Separate layers of the N-doped CD that composed its basic structure, **a.3** Measurement of the density distribution of the N-doped CD from which structural features such as interlayer distance and layer undulation are derived, **b.1** assembled structure of a **γ − graphyne** and a circumcoronene molecule, b.2 Individual structures of circumcoronene and a **γ**-graphyne; **b.3** Measurement of the distance between circumcoronene and a **γ**-graphyne as a function of time.

In this work, we develop and investigate atomistic models of carbon dots based on different structural building blocks, including:

- Nitrogen-doped graphitic layers,
- Graphitic carbon dots functionalized with hydroxyl groups on their surface,
- Hybrid assemblies combining circumcoronene units with graphyne and graphdiyne sheets.

For both the graphitic and nitrogen-doped carbon dot models, we also investigate their interactions with fluorophores belonging to the citrazinic acid family.

Classical molecular dynamics simulations are performed to investigate the interactions between these constituents and to elucidate their self-assembly mechanisms leading to carbon dot formation. Prior to the simulations, all structural components are carefully parametrized to ensure a consistent and reliable description of their interatomic interactions.

All simulations are carried out on high-performance computing (HPC) infrastructures, specifically the LUMI and Karolina supercomputers. Molecular dynamics simulations are performed using GROMACS, while the parametrization of all molecular building blocks is carried out using Gaussian. This computational framework enables an extensive exploration of different carbon dot design strategies, offering a systematic understanding of how structural motifs influence assembly behavior and ultimately govern the resulting morphology. The overall goal is to elucidate carbon dot formation mechanisms and identify optimal structural configurations that could lead to improved physical and functional properties.

REFERENCES

[1] O.A. Stasyuk, A.A. Voityuk, M. Sola, A.J. Stasyuk, γ-graphyne: A promising electron acceptor for organic photovoltaics, Mater. Des. 2023, 255, 111526. DOI: 10.1016/j.matdes.2022.111526.