Carbon Dots structure and optical properties: experimental and theoretical approach

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Carbon Dots (CDs), and in particular N-doped CDs, are C-based rounded nanoparticles (diameter < 10 nm) with high emission quantum yield and large emission tunability across the visible range, depending on composition and structure. These optical properties, along with high chemical inertness, resistance to photobleaching, and biocompatibility, make CDs highly desirable for many technological purposes. However, a still open challenge is the correlation between their structure and photoluminescence. At present, the emission properties are not exclusively ascribed to a single mechanism: quantum confinement, due to hypothesized nanosized sp2 carbon core, and molecular states or surface states, due to fluorophores formation and surface traps, respectively, are both considered. In addition, the optical emission is in general excitation-dependent, suggesting the presence and possibly the interplay of more than one mechanism.

Carbon dots were prepared by solid-state synthesis of citric acid and urea in different molar ratio at 180° C at various synthesis time. Optical spectroscopy, NMR spectroscopy, XPS spectroscopy, structural and morphological analysis were considered to characterize the structure and properties of CDs. Quantum chemistry calculations were applied to model the emitting centres and their formation during the synthesis.

By changing the molar ratio of the precursors, CDs with main emission in the blue and green range can be prepared. The structural features indicate the presence of a carbon network with a large degree of disorder increased by the presence of N doping. The emission properties are related to both surface groups and molecular centres, whose formation starts from the very first minutes of the synthesis. The calculations allow ascribing the optical properties to some specific centres and to follow their genesis during the synthesis.

In these years we worked on the characterization of the role of N in CDs to ascertain how the doping modifies the structure and the optical properties of our samples aiming at correlating structure, morphology, and optical features by means of synergic experimental and computational approaches. The presentation will discuss our insights and perspectives to address the CDs' challenges and opportunities for a CD-based technology.

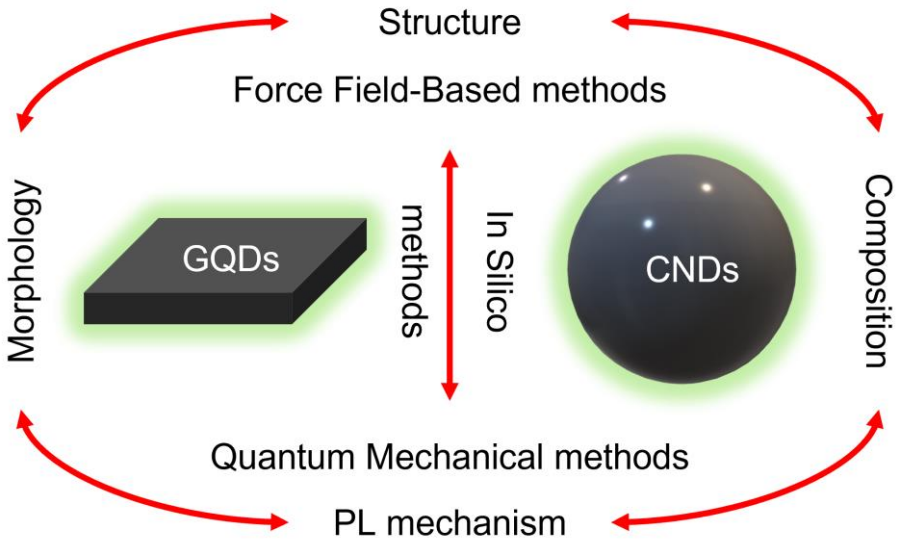

Figure 1 – In silico methods and features of carbon dots. Correlation between computational cost of in silico methods and features of graphene quantum dot (GQD) and carbon nanodot (CND) models [reproduced from ref 1, copyright Elsevier 2023]

REFERENCES

[1] Carbonaro C.M., Engelbrecht L., Olla C., Cappai A., Casula M.F., Melis C., Stagi L., Laaksonen A., Mocci F. (2023) "Graphene quantum dots and carbon nanodots: modeling of zero-dimensional carbon nanomaterials", *Zero-Dimensional Carbon Nanomaterials*, 411 – 482 (ed. by Woodhead Publishing, Elsevier) https://doi.org/ 10.1016/B978-0-323-99535-1.00014-7