Carbon Dots: How many if any?

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Carbon dots (CDs) are a class of nanomaterials, discovered in 2004, characterized by their intense photoluminescent properties, low toxicity, photostability and simple preparation methods. These properties enable the application of CDs across many scientific fields, such as bioimaging, photocatalysis, and theranostics [1-3]. Photophysical properties of CD materials are directly influenced by the structure of CD nanoparticles as well as by the composition of post-synthetic reaction mixtures [4]. However, the structure and composition of CDs are still a matter of debate given the high heterogeneity of these systems [5]. A general consensus is that CDs consist of three main components: a core, a shell, and molecular fluorophores which are either free, or attached to the shell or embedded in the core [6].

The primary aim of our work was to investigate the structure of carbonized components of CDs prepared via bottom-up methods and to determine whether they have the character of the crystalline graphene-like lattice or can be better described as clusters of polymeric hydrocarbon chains. To this end, carbonized components of CDs prepared from citric acid and urea were separated from post-synthetic reaction mixtures by filtration and extended dialysis.

For the preparation of the CD materials, we employed hydrothermal treatment of the precursors at different reaction temperatures: 180 °C, 200 °C, 220 °C, and 240 °C. Later, we focused on the sample prepared at 240 °C assuming that the higher temperature will yield more carbonized cores. The raw CD solution was filtered through a syringe filter, and as-prepared CD material was dialysed for prolonged periods of time (minimum 10 days) in order to remove all low-molecular-weight impurities.

After 11-day dialysis, the yields of the product were ca 0.01-0.02% of the initial mass of precursors. There are two possible explanations of this outcome: either the chosen synthesis method and reaction conditions are inadequate for producing significant amounts of carbonized nanoparticles, or the carbon framework disaggregates over time and passes through the membrane. Overall, our findings highlight the need for a comprehensive mass balance analysis of the individual components of the reaction system and for a critical evaluation of systems referred to as carbon dots.

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