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## Liquid-phase laser ablation synthesis of graphene quantum dots from carbon nano-onions: Comparison with chemical oxidation

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Graphene quantum dots (GQDs) have been synthesized reproducibly by chemical oxidation (CO) of carbon nano-onions (nCNOs) and a one-step pulsed laser ablation (LA) of nCNOs in deionized water. The photoluminescence (PL) spectra show that the LA-GQDs have blue shifted emission relative to the CO-GQDs which is attributed to the effects of both particle sizes and surface functional groups. The CO-GQDs have an average diameter of 4.1(8) nm and a thickness corresponding to two or three graphene layers, while the LA-GQDs have an average diameter of 1.8(6) nm and a thickness comparable to a single layer of graphene. The CO-GQDs favor the presence of carboxylic groups and have a higher fraction of  $sp^2$  carbons, while the LA-GQDs prefer the presence of hydroxyl groups and have a higher fraction of  $sp^3$  carbons. PL lifetime data suggests that surface functional groups are the main source of radiative deactivation and the  $sp^2$  carbon domains are mainly responsible for non-radiative decay. PL lifetimes are measured to be 7.9(6) ns for the emission from the carboxylic groups and 3.18(10) ns from the hydroxyl groups. Compared to CO, liquid-phase LA is a faster and cleaner one-step method for producing GQDs with fewer starting chemicals and byproducts.

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