

Direct correlation between the measured Electrochemical capacitance, wettability and surface functional groups of Carbon Nanosheets (CNS)

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Carbon Nanosheets (CNS) also called Carbon Nanowalls are the newest addition to the nanostructured carbon materials family. Carbon Nanosheets are built from layers of two dimensional sp²-bonded carbon atoms. A single sheet of the CNS consists of a few layers of graphene vertically standing on the substrate. CNS form a self-supported, high aspect ratio network with a sheet thickness ranging from a few nanometers to tens of nanometers. Carbon Nanosheets base material graphene has exceptional characteristics, high electrical¹ and thermal conductivity, high mechanical strength and large specific surface area². Study of CNS as an electrode material for a Supercapacitor application, also called Electrical Double layer Capacitor application has shown promising results³.

The supercapacitor performance is based on the charge accumulated at the double layer interface between an electrode and an electrolyte. The amount of charge in the double layer and thus the capacitance is proportional to the electrolyte accessible surface area. There can be an extra contribution to the accumulated ions in the double layer if reactive surface groups are present on the carbon electrode. Surface area accessibility depends on the wettability of the electrode, which is in its turn affected by its chemical composition and geometrical structure (roughness).

In this work, a series of CNS with heights ranging from few nanometer up to 1 micrometer were grown by RF plasma enhanced CVD from both C₂H₂-H₂ (1:10) and CH₄-H₂ (1:2) precursor gas mixtures. SEM imaging revealed that acetylene based CNS (A-CNS) had open porous structure, while the methane based CNS (M-CNS) had structure where fallen sheets were blocking the openings (Fig.1). The samples in M-CNS series were divided in two parts M-CNS-1 and M-CNS-2 depending on the growth history. M-CNS-1 and A-CNS were grown in the same period, while the M-CNS-2 sequence were grown several months after.

Electrochemical characterization of CNS was carried out by Cyclic Voltammetry and Electrochemical Impedance Spectroscopy (EIS) in aqueous and non-aqueous electrolyte solutions. The impedance behavior of all the CNS in both electrolytes could be fitted to the impedance behavior of an ideal capacitor in series with a resistor and capacitance density was determined as such. Fig 2 shows the capacitance density in aqueous electrolyte together with the static CA values in water versus the CNS height. The capacitance density for A-CNS and M-CNS-2 increased with height as expected for the increase in effective area. The capacitance density for M-CNS-1 were rather irregular versus the height in water, but increased regularly in the non aqueous electrolyte solution (not shown). A good correlation was found between the capacitance density in water and the Contact Angle (CA) values. FTIR measurements revealed distinct transmission peaks for hydrophilic Carbon-Oxygen groups on samples with low CA angle and absence of those peaks for high CA angle samples, indication the role of surface functionalization for wetting.

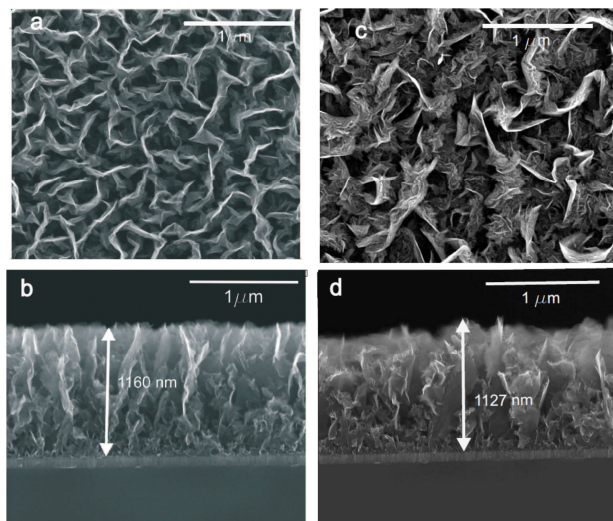


Figure 1: Top and cross-section Scanning Electron Microscopy images accordingly for CNS grown by acetylene based precursor (a,b) and methane based precursor (c,d)

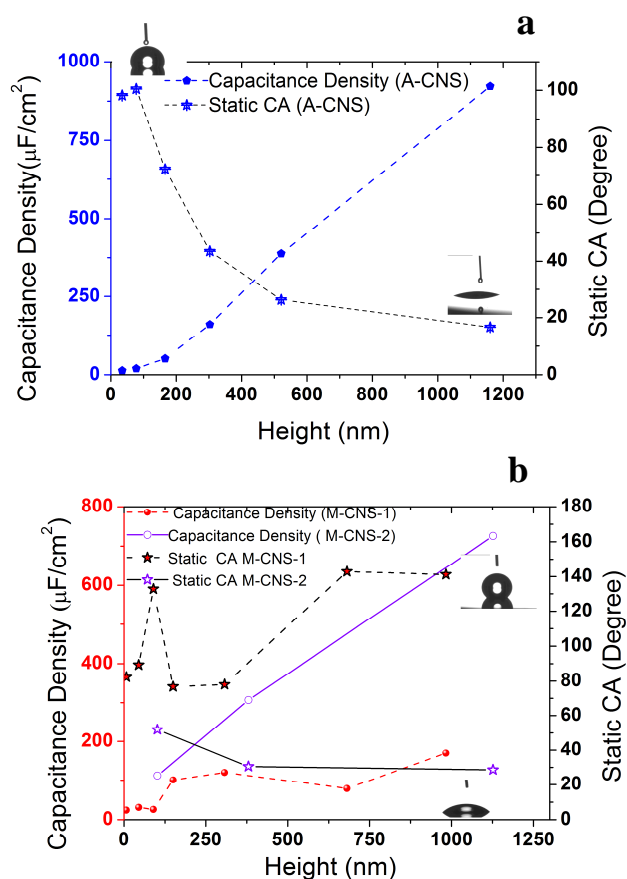


Figure 2: The capacitance density of A-CNS (a) and M-CNS (b) obtained in 1M Na₂SO₄ in H₂O at -064 V bias and the Contact Angle of water droplet on CNS versus the height.

[1] Novoselov, K.; Geim, et al *Science* 306 (2004.)

[2] Lee, C. et al. *Science* 321 (2008)

[3] Miller, J. et al *Science* 329 (2010)