SWCNT structural and chiral origin studied by computer simulation

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Since 25 years, significant progress has been achieved in the controlled synthesis of Single Walled Carbon Nanotubes (SWNTs), but we are still facing difficult issues concerning the yield and selectivity of their synthesis by Catalytic Chemical Vapor Deposition. The choice of a catalyst is critical, and hitherto made by trial and error. In fact, we don't know what are the required properties of a "good" catalyst for a selective SWNT growth, partly because currently available models *[1,2]* did not directly address this issue.

We answer this question by developing a statistical thermodynamics model, that in the case of a perpendicular growth figure.1.a,b *[3,4]*, relates the stable (n,m) tube structures, to the tube/catalyst interfacial energies for zigzag and armchair edges and the temperature. This model shows that, at low temperature, only zigzag or armchair tubes are stable. Chiral tubes become stable at higher temperature because of the configurational entropy of the tube edge in contact with the catalyst, that is a key element of the model. This enables us to produce chiral stability maps and phase diagrams figure.1.c, that relate the catalyst interfacial properties and the temperature with the resulting equilibrium chiral distribution. It explains under which conditions, a near armchair distribution can be obtained, and accounts for the temperature evolution of the chiral distributions reported in a number of experiments. The technical aspects of the model *[5]* and possible further developments will be discussed.

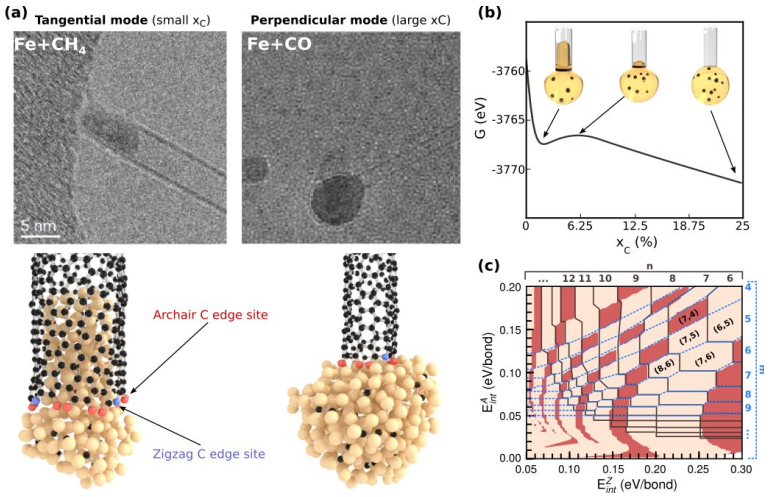

Figure 1. *a) TEM images and simulated tangential and perpendicular growth modes depending on carbon concentration inside the catalyst. b) Free energy depending on carbon concentration. Chirality diagram depending on contact energy between zigzag and armchair C edge sites with a Ni catalyst. Brown area denotes metallic (n,m) tubes chirality, ligth grey denotes semiconducting (n,m) tubes chirality.*

References

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