Three Stages of Development of Electrical Percolation of Carbon Nanotubes in Nematic Liquid Crystals

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Liquid crystals (LCs) are ideal dispersing media for carbon nanotubes (CNTs), allowing to align ensembles of the tubes and their assembling in the structures imposed by LC host. On the other hand, loading of CNTs improves electro-optical characteristics of LC layers and extends range of their properties. In both cases, it is crucially important to take into account interaction of CNTs, their self-assembling and formation of percolating structures that lead to critical behavior of different system parameters.

We report peculiarities of electrical percolation of multi-walled CNTs in nematic LC 5CB in wide concentration range of CNTs (C_{CNT}=0-2 wt. %). The composites were made by ultrasonic mixing of LC 5CB (Merck) and multi-walled CNTs from Cheap Tubes Ltd. with average length of 0.5-2 μm. The resulted mixtures were loaded in the plane-parallel cells for planar alignment with a thickness of 50 μm having copper electrodes for in-plane application of electric field. The inter-electrode distance was 200 μm. The studies were conducted using dielectric spectroscopy method supported by polarization microscopy. The spectra of real ε' and imaginary ε'' parts of dielectric permittivity measured in the frequency range f =10-10^5 Hz allowed us to determine the range between several relaxations corresponding to polarization and charge transfer in the LC bulk. For the frequency f=10^3 Hz, which falls in the mentioned range of all composites, the electrical conductivity σ and permittivity constant ε' were determined. Based on these data, $\sigma(f)$ and $\varepsilon'(f)$ plots were obtained. The advantage of the cells construction used in this research is that dielectric method and optical microscopy can detect percolation of CNTs in the same direction (electrical and visual percolation, respectively).

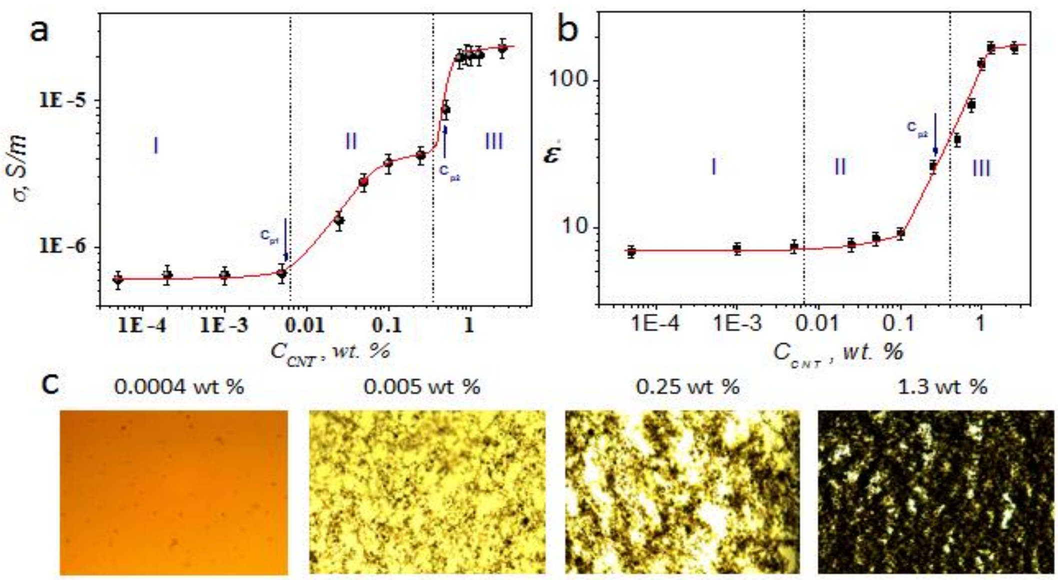

Fig. 1. Plots of electrical conductivity σ (a) and permittivity ε' (b) vs. concentration of CNT C_{CNT} . (c) Micro-photographs of the cells loaded with composites of different concentration of CNTs.

Fig. 1a shows electrical conductivity of 5CB-CNTs composites σ as a function of CNT concentration C_{CNT}. Three sections of this curve can be distinguished. At low CNT concentrations (C_{CNT}≤1*10^{-3} wt. %), the conductivity only slightly grows with C_{CNT} that might be attributed to contribution of separated conductive particles or their small aggregates (Maxwell mechanism [1]). The following growth of C_{CNT} leads to growth of aggregates and formation of branched interpenetrating structure (Fig. 1c) that causes percolation growth of conductivity. Critical concentration of this percolation is C_{p1}=4*10^{-3} wt.%. Even higher increase of C_{CNT} leads to dense packing of CNTs that in turn results in new percolation stage with critical concentration C_{p2}≈0.5 wt.%. The two-stage character of conductivity percolation was successfully described within the core-shell model of percolation [2,3]. The permittivity curve grows with C_{CNT} too (Fig. 1b), but weak percolation behavior is manifested only near second critical point, C_{p2}. Near this point ε' increases in one order of magnitude, but this growth is not steep. This is because the short circuiting of LC cell by clusters of CNTs is not a prerequisite for permittivity growth as in case of $\sigma(C_{CNT})$ curve. Due to this fact, the $\varepsilon'(C_{CNT})$ curve detects compression of CNTs aggregates in the rather wide (0.1 - 1 wt. %) concentration range of CNTs. Saturation of this curve as the $\sigma(C_{CNT})$ curve indicates formation of dense percolation cluster extending between the cell electrodes.

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