**Novel "Carbon Nanotube/Graphene Layer" Nanostructures Obtained by Injection CVD Method for Electronic Applications**

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As it was predicted theoretically, a 3D network nanostructure, composed of parallel graphene layers stabilized by vertically aligned CNTs, when doped with lithium cations can be efficient structure for hydrogen storage [1], and, moreover, this nanostructure is considered as a novel material with tailored multidimensional thermal transport characteristics [2].

First practical realization of CNT/graphene nanostructures with vertically aligned CNTs grown in between the graphene layers by CVD method was reported in ref. [3]. The exfoliated graphene oxide was selected as the substrate to grow CNTs. These nanostructures have been successfully used as the electrodes in supercapacitors. The existence of CNTs in these nanostructures significantly enhanced the graphene property by, as believed, bridging the defects for electron transfer and increasing the basal spacing between graphene sheets.

However, the proposed method of CNT/graphene nanostructures realization is extremely complicated. The experimental fabrication of such nanostructures with the low cost processes is challenging.

Present investigation is devoted to the creation of composite nanostructures of the arrays of vertically aligned CNTs and the planar graphene (graphite) layers (PGL) located at the top of the CNT arrays (CNT-PGL nanostructures) by using the only one-step process - the most simple and low cost CVD process with the injected catalyst realized at ambient conditions. One-layer [4], as well as multi-layer nanostructures [5] were created. The last nanostructures we designated as CNT-PGL nanostacks.

Composite carbon structures were synthesized by the injection CVD method using xylene/ferrocene solution, as described in refs [4, 5]. Rate of injection was varied in the range 0,01-0,2 $cm^3$/min. The constant flow of Ar (100 $cm^3$/min) through a reactor was provided during the processes of reactor heating and cooling and CNTs synthesis. The content of ferrocene in the feeding solution was 1,0 (wt %). The process was carried out at the atmospheric pressure at the working temperatures of 850˚C. Wafers of *n*-type Si with 600 nm thermal oxide layer were used as substrates.

The elemental composition were investigated by Auger and EDX spectroscopy, structural characterization was performed using scanning and transmission electron microscopy, Raman Spectroscopy.

The growth mechanism of one- and multi-layer CNT-PGL nanostructures was proposed.

In Fig. 1 the SEM images of the fragments of one-layer CNT-PGL nanostructure are presented.

It was proved that the structure consists of carbon, i.e. represents CNT-PGL structure indeed. What is particular, PGL can be easily detached from the CNT array (Fig. 1a-d). The strips of graphene may be used for the production of different devices or for the physical experiments.

In Fig. 2 it is shown that by the developed technology one can produce any number of layers of CNT-PGL nanostructures. For example, three-layer (Fig. 2a) and four-layer (Fig. 2b) nanostructures are presented.

The CNT-PGL nanostructures presented in Figs. 1,2 are "ordered" nanostructures, because they demonstrate strongly organized configuration of CNT-PGL layers. Another type of CNT/graphene nanostructures, "disordered", obtained by the same method, but in different regimes are presented in Fig. 3.

In our approach the high-quality CNT/graphene nanostructures are produced by a very low cost process. We expect to observe extraordinary electrical properties of these structures and compatible commercialization conditions with any other approach. Moreover, the used CVD technique is versatile and scalable. The obtained nanostructures can enable many applications including high-performance elastic and flexible conductors, electrode materials for lithium ion batteries and supercapacitors, thermal management, catalyst and biomedical supports, electrical energy storage devices based on this new class of carbon material, and so on.

**References**

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## Figures

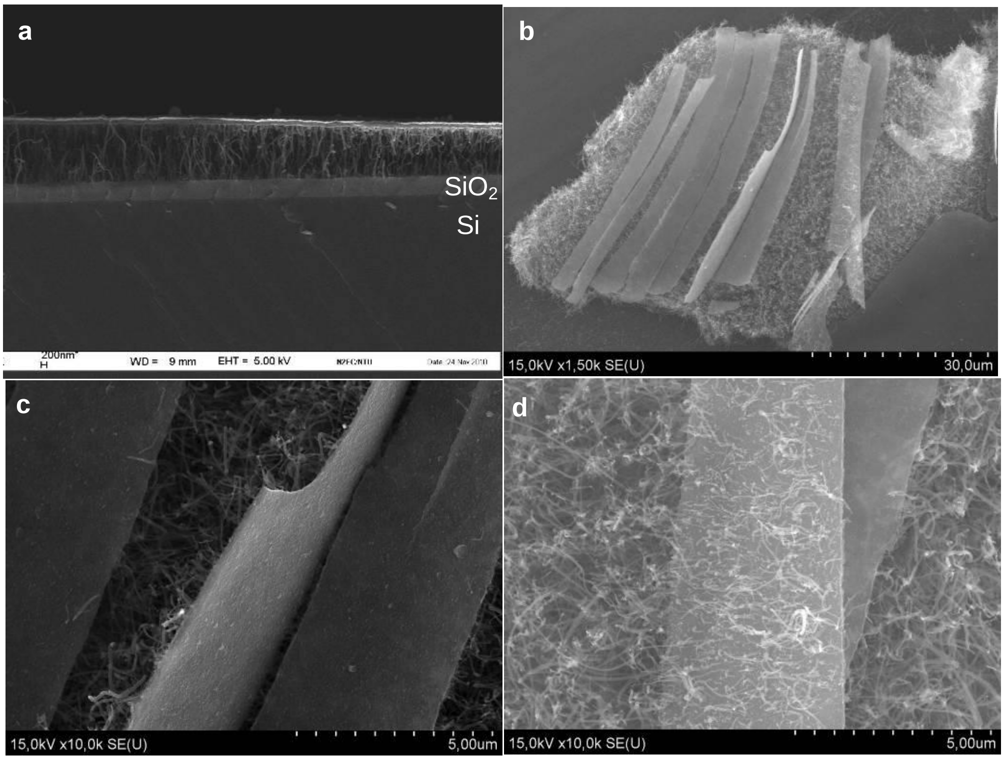


Fig. 1 SEM images of the fragments of the one-layer CNT-PGL nanostructure: (a) nanostructure formed on Si/$SiO_2$ substrate, (b) graphene strips detached from the surface of CNT array at different magnifications, (c) back side of a strip with the attached CNTs.

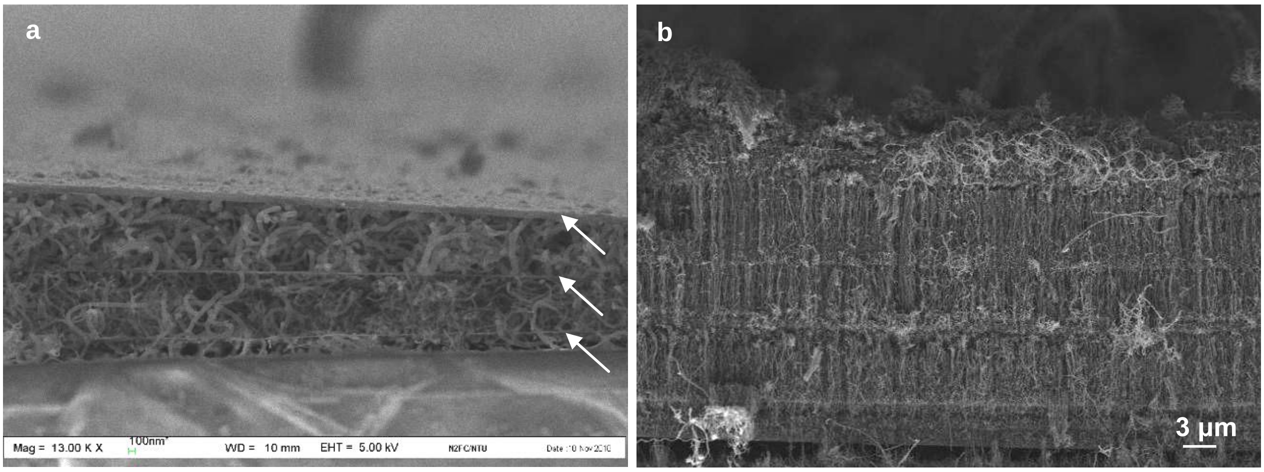


Fig. 2 SEM images of multi-layer CNT-PGL nanostructures: (a) three-layer (indicated with arrows) and (b) four-layer nanostructures.

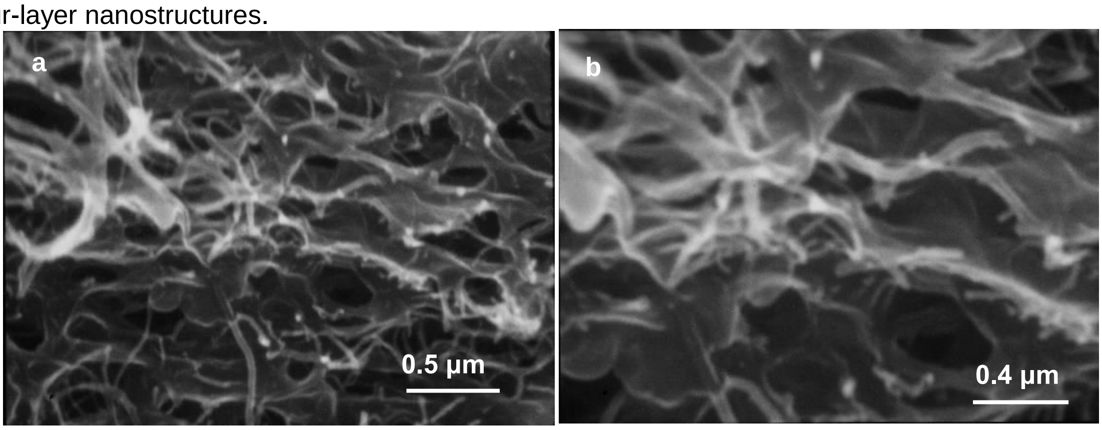


Fig. 3 (a, b). “Disordered” CNT/graphene nanostructures shown at different magnifications (SEM).