Generalization of the Schottky Barrier Postulate at Carbon-Metal Interfaces for Pure, Aligned, Suspended Horizontal Carbon Nanotubes and Hot-press Laminated Graphene Thin Films

M. T. Cole¹, K. Ying¹, S. Haque², K. B. K. Teo³, W. I. Milne¹

 ¹ Cambridge University, Centre for Advanced Photonics & Electronics, 9 JJ Thomson Avenue, Cambridge, UK
² Nokia Research Centre, Broers Building, 21 JJ Thomson Avenue, Cambridge, UK
³ AIXTRON Nanoinstruments Ltd., Swavesey, Cambridge, UK mtc35@cam.ac.uk

Roll-to-roll, large-area production of flexible transparent metallic and semiconducting thin films is becoming increasingly important in opto-electronic devices. Discrete carbon nanostructured materials, such as carbon nanotube (CNT) and graphene based thin films, offer a potential alternative to the rigid and increasingly expensive industry standard material; indium tin oxide.

In this study, hot-press lamination (HPL) was used to transfer graphene films, synthesised by thermal chemical-vapour-deposition, from their source Cu foil substrates onto polycarbonate (PC) supports (Fig. 1 (a-d)). [1, 2]. Areas in excess of 30 cm² have been achieved with typical sheet resistances of ~3 k Ω/\Box . Multi-Walled carbon nanotube (MWNT) forests (Fig. 2(g)) were also synthesised in the same reactor, and were aligned and suspended across metallic contacts via an extruding technique [3, 4] (Fig. 2 (a-f)).

The HPL-graphene films showed a nominal optical transmittance of 77% (81% substrate transmittance, Fig. 1(f)), and 76% for the freestanding S-MWNT films (Fig.2(h)), comparable values to that achieved through matured ITO processing. Macroscopic investigations into the mechanical robustness of these films showed maximum tensile strains of 35% and >140% for the HPL-graphene and S-MWNTs, respectively; values significantly higher than that of state-of-the-art PC/ITO (1.15%). These exceptionally high strains are attributed to the combined effects of intra-flake/CNT expansion and inter-flake/CNT separation, where this second contribution is significantly enhanced in CNT-based thin-films. The HPL graphene flakes have an exposed area of the order of 400 μ m² (Fig. 1(e)).

Low temperature *I-V* measurements were undertaken to compare the underlying conduction mechanisms in these discrete nanostructured carbon films. Back and top-gated thin film transistors were also fabricated based on a Si/SiO₂ support using a S-MWNT channel and parylene-C dielectric on PC support with a HPL-graphene channel. Al, Pd and Mo source-drain contacts were deposited in each case to investigate the generality of the Schottky barrier postulate, previously identified to explicate the observed electric-field gating effect in SWNTs [5]. Here it was found that the work function of source/drain contact metals played a key role in determining the p-type, n-type or ambipolar behaviour observed.

References

- [1] V. P. Verma, S. Das, I. Lahiri, & W. Choi, *App. Phys. Lett.*, **96**, (2010) 203108.
- [2] I. Lahiri, V. P. Verma & W. Choi, Carbon, (2011).
- [3] M. Zhang, S. L. Fang, A. A. Zakhidov, S. B. Lee, A. E. Aliev, C. D. Williams, & R. H. Baughman, *Science*, **309**, (2005) 1215.
- [4] K. Liu, Y. H. Sun, L. Chen, C. Feng, X. Feng, K. L. Jiang, Y. G. Zhao, S. S. Fan, Nano Lett., 8, (2008) 700.
- [5] M. H. Yang, K. B. K. Teo, D. G. Hasko, & W. I. Milne, *App. Phys. Lett.*, 87, (2005) 253116.

Figures



FIG. 1. Hot-pressed laminated graphene fabrication. (a) Laminating of graphene (synthesized on 12.5 µm Cu foil) at 120 °C on polycarbonate (PC) support. (b) Removal of underside PC laminate. (c) Cu etching using 22.5 % wt. FeCl₃. (d) Remaining graphene coated PC substrate. Inset: Optical image of a 4.5 x 7.0 cm graphene/PC sample. Typical (e) histogram of the exposed flake Normal area. distribution fitted (Mean flake area of 400 µm, std. 1498 μm²). (f) UV-Vis transmission spectrum for competing technologies (PEDOT/PSS:IPA, ITO), the bare PC substrate and the graphene/PC platform

showing a uniform optical transparency of 77%. (g) Raman spectrum of CVD synthesized graphene transferred to Si/SiO_2 (300 nm).



FIG. 2. (a-f) Suspended MWNT array fabrication process. (g) Typical sample Raman Spectrum. (h) UV-Vis spectrum showing uniform optical transmittance.