Development of Graphene-Based Field Emitter for THz Device Application

A.K. Singh [a,b], S.K. Shukla [a,b], V. Rawat[b], D. Kumar[b], P. Prince[b], M. Chauhan[b], and R. K. Barik[a,b]
(a) PhD Engg. AcSIR, Ghaziabad, (b) CSIR-CEERI, Pilani

Abstract

Development of graphene-based field emitter for application in THZ VEDs have been the main objective. Modified Hummers method, Vacuum Filtration and thermal annealing technique has been adopted to develop a ~ 5μm film. The characterization results using SEM, Raman and XRD have also been discussed herewith.

Introduction

Recently, the development of a compact terahertz (THz) Vacuum Electron Devices (VEDs) for application like communication, remote sensing, security, medical imaging, etc has gained immense pace. When compared to the existing THz solid state devices, the VEDs offer high energy conversion efficiency, thermal robustness and radiation hardness [1]. One of the critical challenges in the development of the THz VEDs is the requirement of very high current density electron emitters. Though, most of the effort in this area has been directed towards the development of scandate cathodes, capable of delivering current density in excess of 100 A/cm2, still a higher current density, of the order of 1000 A/cm2, would be required at frequencies > 200 GHz [2].

Till date, in almost all the high-power VEDs employ the thermionic cathode, however its use could be limited at high frequencies as the device size scales down with frequency. In that case, the field emission cathodes will be the best possible candidate to cater to such high current density needs. The field emitters are generally characterized with instant turn-on, high current density, lower power dissipation and Micro-Nano fabrication technology. In principle, these cathodes will be very useful in realizing the high frequency compact and light-weight VEDs however, the reliability and the life are yet to be established. In the past, numerous dedicated efforts have been made to develop the high current density field emitter arrays. Arrays of cathodes with 107 tips /cm2 could be produced, using micro fabrication technology, and the emission densities of 1000 A/cm2 could be achieved from 10000 tips [3]. Arrays of carbon fibre coated with CsI have been reported to yield 50 A/cm2 over one million pulses at 165 kV [4]. The use of the FEAs has been limited for two reasons: (a) the surface of the FEA tip is highly dynamic as adsorption of the ambient gases changes the work function of the surface, and (b) the ion back bombardment changes the radius of curvature of the tip during the operation. Recently, graphene-based field emitters have attracted immense interest due to its high aspect ratio (thickness to lateral size ratio), high carrier density, the larger carrier mobility, excellent electrical and thermal conductivity and stability. Graphene, a single /multiple layer (< 10 layers) of carbon sheets, can be synthesized using various methods such as chemical vapor deposition, chemical exfoliation, electrophoretic deposition, screen-printing and chemical techniques. The criticality of the film is the it should able to bear the thermal load due to the joule heating caused by the electron drift and high machinal strength to hold the film firmly at such high temperature operation.

GO film Synthesis

For the synthesis of graphene film suitable for very high current density applications, large yield is required and hence the choice of synthesis is modified Hummers method [5]. The synthesis flowchart has been depicted in the fig. 1.

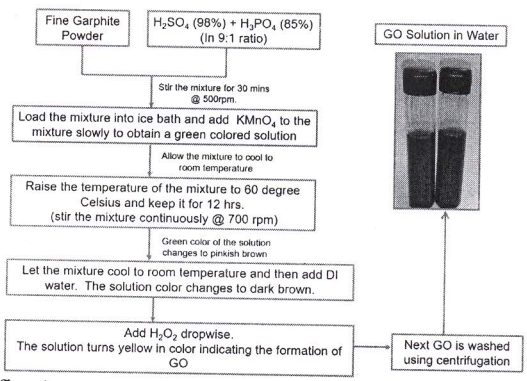


Fig. 1. Modified Hummers Method for GO synthesis.

GO film was prepared using Vacuum filtration technique [6] as shown in fig. 2 (a). The thickness of the film depends on the concentration of the GO solution. 0.45μm PTFE membrane was used as the filter paper on which the film was obtained. The film was the left overnight for drying in a vacuum oven at 60 ^{0}C. The GO film was then fished out by dissolving the PTFE membrane in acetone. The film was prepared out of 20 ml GO solution in water (1mg/ml) as shown in fig. 2 (b). The GO film could not be used directly for high current density application as due to the high oxygen content, the conductivity is very low. So, the film was further hydrogen fired 1300 ^{0}C for 10 mins. The dark brown GO film turns to shin black rGO film after hydrogen reduction.

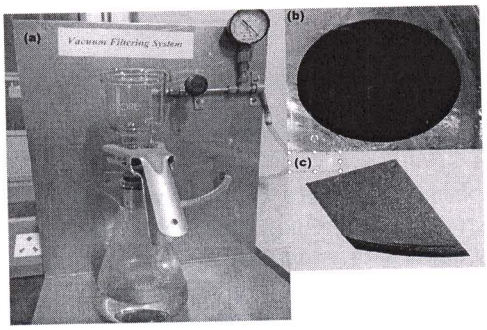
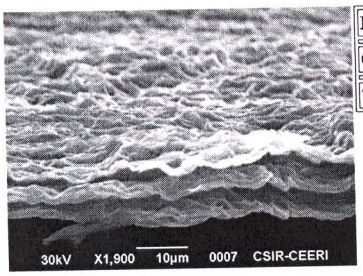


Fig. 2. (a)Vacuum Filtration System, (b) GO fim, (c) thermal annealed rGO film

Characterization Results

SEM and EDAX analysis were carried to see the morphology and the oxygen content in the rGO film. The film was placed vertical such that the plane of the film is in line the electron beam axis to image the cross-section. As seen from the SEM of the film cross-section in fig. 3 (a), the thickness of the film obtained is ~ 5μm and the EDAX results shows that nearly all the oxygen content has been removed after hydrogen firing.



Element	Weight%	Atomic%
C K	100.00	100.00
Totals	100.00	

Fig. 3. SEM image of the GO film and EDAX analysis.

The Raman spectrum of graphene and related materials generally exhibit three primary bands namely the D, G and 2D bands which appears around 1350cm^{-1}, 1600cm^{-1} and 2700cm^{-1} respectively. The D band supports the presence of defects within the graphene lattice whereas the G band from sp2-hybridized graphitic carbon atoms. The intensity ratio of I_D/I_G is a measure of lattice dis-orderness. Fig. 4 (a) & (b) is the Raman spectrum of the GO film and the thermally reduced GO film. The I_D/I_G ratio in the case of GO and rGO has been found 0.8 and 1.6 (due to the evaporation of carbon atoms) respectively. Fig. 4. (c) & (d) shows the X-ray diffraction (XRD) patterns of GO and rGO, respectively. GO exhibited a sharp peak at 11° with an interlayer distance of 0.8 nm, which is larger than the interlayer distance of graphite (0.34nm), revealing that many different oxygen-containing groups were intercalated within the interlayer space. After reduction, a broad peak at 26° is observed with a d-spacing of 0.4 nm, implying that the successful reduction of GO to reduced graphene oxide.

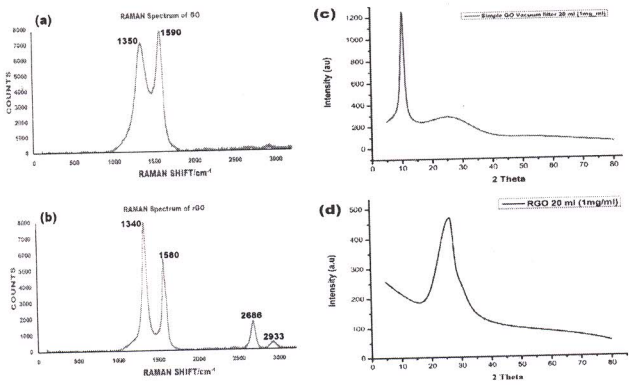


Fig. 4. (a) Raman Spectrum of GO, (b) Raman Spectrum of rGO, (c) XRD Spectrum of GO, (a) XRD Spectrum of rGO.

Future Work

A lot of scope for the improvement in properties of the film still remains. It is also proposed to dope the film with metallic nano-particles to improve the thermo-mechanical properties of the film. Also, the characterization of the film for its field emission capability and emission stability have to established for usability in any real device.

References

[1] Srivastava, V., "Vacuum Microelectronic Devices for THz Communication Systems," in Annual IEEE India Conference (INDICON), Dec. 2015.

[2] Han, S.T., Jeon, S.G., Shin, Y.M., Jang, K.H., So, J.K., Kim, J.H., Chang, S.S. and Park, G.S., "Experimental Investigations on Miniaturized High-Frequency Vacuum Electron Devices," IEEE TRANSACTIONS ON PLASMA SCIENCE, vol. 33, no. 2, pp. 679-684, April 2005.

[3] C. A. Spindt, C. E. Holland, A. Rosengreen, and Ivor Brodie, "Field-Emitter Arrays for Vacuum Microelectronics", IEEE Transaction on Electron Devices, vol. 38, no. 10, pp. 2355-2363, October 1991.

[4] L. F. Drummy, S. Apt, D. Shiffler, K. Golby, M. LaCour, B. Maruyama, and R. A. Vaia, "Nanostructural evolution During Emission of CsI-Coated Carbon Fiber Cathodes", Journal of Applied Physics, vol. 107, no. 11, June 2010.

[5] Chen, J., Yao, B., Li, C., & Shi, G., "An improved Hummers method for eco-friendly synthesis of graphene oxide". Carbon, vol. 64, pp. 225–229, 2013.

[6] Chen, J., Guo, Y., Huang, L., Xue, Y., Geng, D., Liu, H., Wu, B., Yu, G., Hu, W., Liu, Y. and Zhu, D., "Controllable fabrication of ultrathin free-standing graphene films", Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 372(2013).