

## Supplementary information

### Dielectric constant enhancement of Poly 4-Vinylphenol (PVPh) via Graphene flakes incorporation through electro spray atomization for Energy Storage

Adnan Ali<sup>1\*</sup>, Sosiawati Teke, Ghayas Uddin Siddiqui, Young Sun Mok

Department of Chemical Engineering, Jeju National University, Jeju 63243, Republic of Korea

#### 1. Electro spray Atomization Set-up:

In thin film fabrication techniques, electro spray atomization is one of the most cost-effective technique in which the liquid flow out of a nozzle with control flow rate is brought under the influence of a high potential electric field at desired temperature.<sup>1-4</sup> A cone is developed at the nozzle orifice by gradually increasing the applied potential and a stable jet forms at the tip of the cone. Due to coulombic forces, the stable jet breaks down further into extremely-tiny droplets, carrying nanoparticles/functional material. The electro spraying droplets are deposited on a substrate to form a homogenous film of the solute which is present as precursor in the solvent.

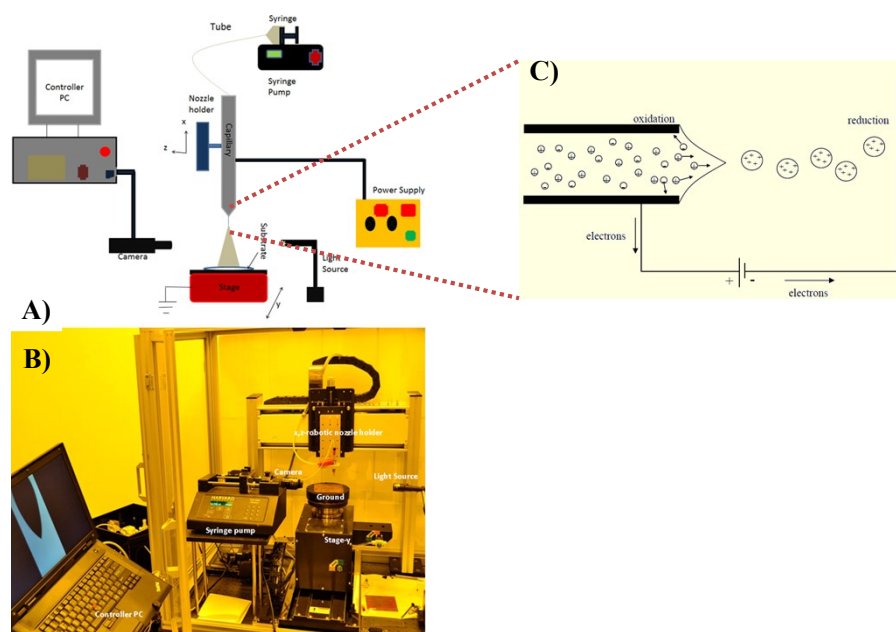
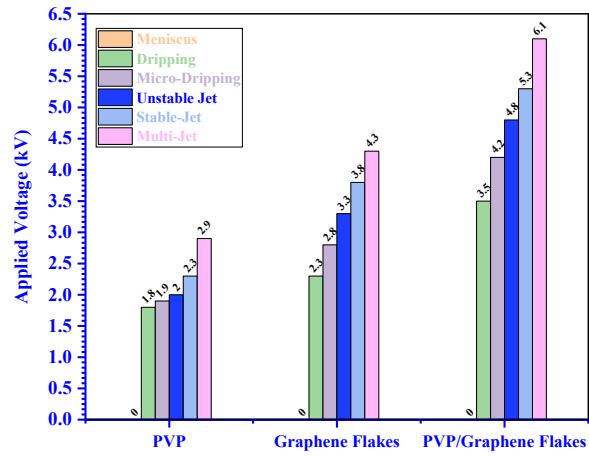
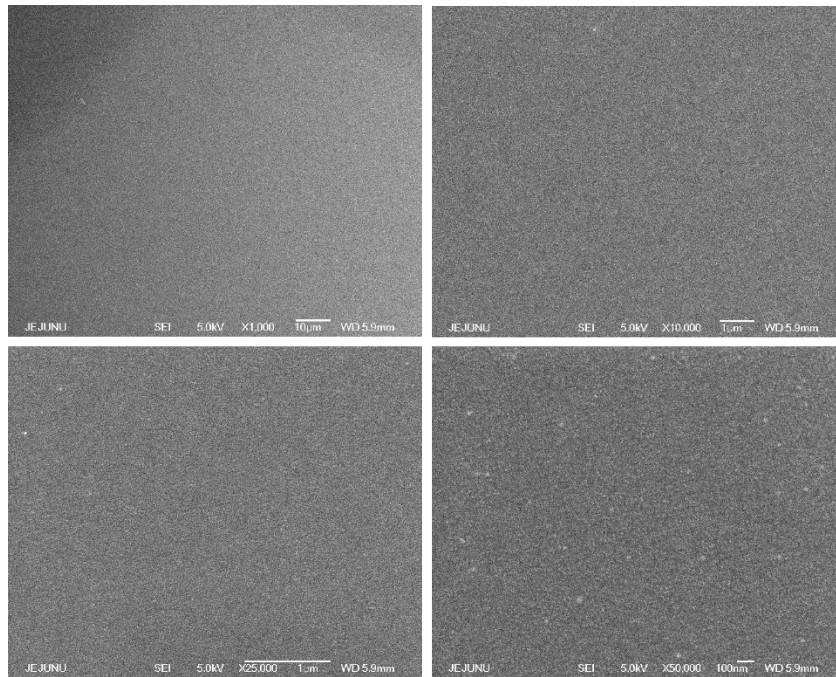


Fig. S1. A & B) Electro spray Atomization set-up, C) Electro spray Atomization deposition mechanism.

## 2. Potentials comparison for atomization modes of PVPh, Graphene and PVPh/Graphene dispersion



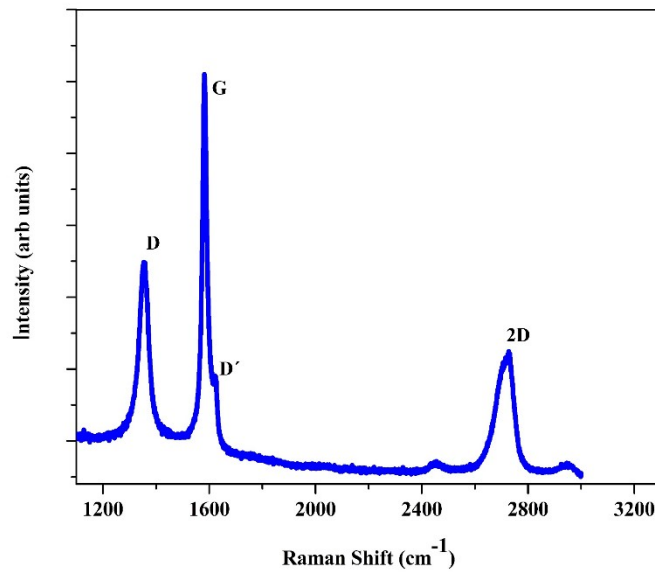
**Fig. S2.** Potential required for different electro spray modes at 300  $\mu\text{l/hr}$  flow rate of PVPh, Graphene and PVPh/graphene dispersions.



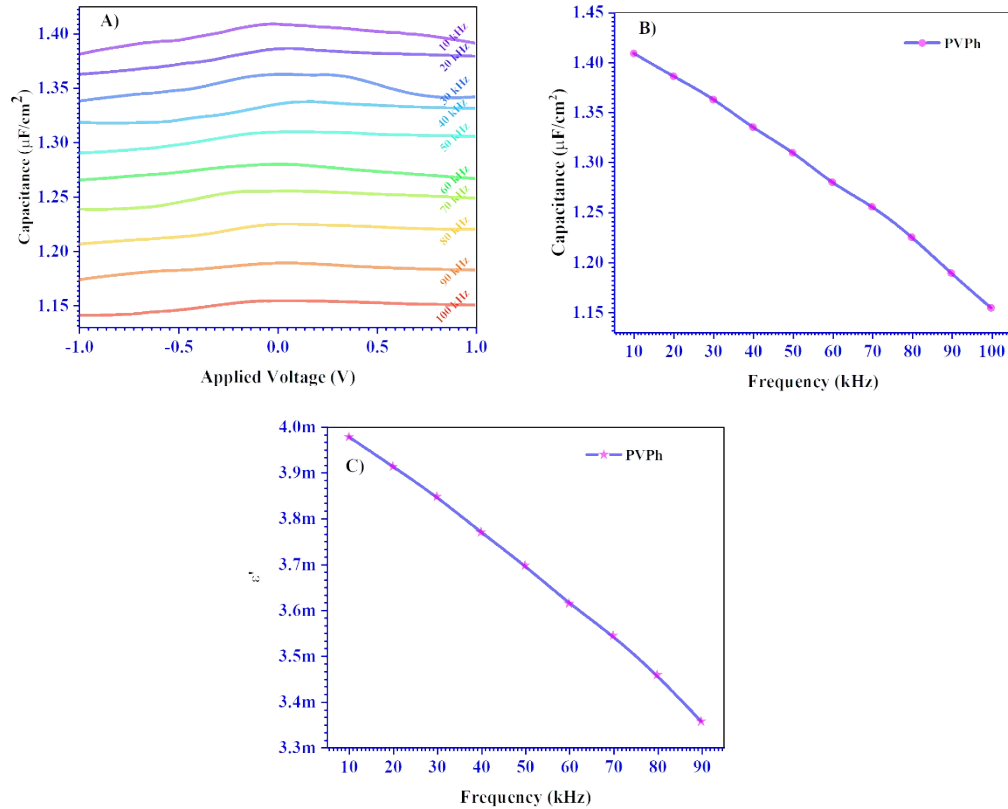
**Fig. S3.** Lower to higher magnification FESEM surface morphology analysis of PVPh thin films

### 3. Raman Spectroscopy Analysis of Graphene Flakes

Raman spectroscopy of the deposited graphene flakes film is carried out by using the LabRam HR800 microRaman spectroscope. The Raman system is operated at the 10 mW laser power and an excitation wavelength of 514 nm with Ar<sup>+</sup> ion laser. **Fig. S4.** shows the Raman spectrum of the deposited graphene flakes film. The major signature peaks, commonly observed in all chemically processed graphene are D band at 1,354/cm, G band at 1,580/cm and 2D band at 2,725/cm. The G band at 1,580/cm corresponds to E<sub>2g</sub> mode which is related to the sp<sup>2</sup>-bonded carbon atoms vibration in a 2D hexagonal lattice. And the D band at 1,354/cm arises from a breathing mode of k-point phonons of A<sub>1g</sub> symmetry. The high intensity of D band indicates the presence of sp<sup>2</sup> C with defects. It is also reported that the D band arises from the reduction in size of in-plane sp<sup>2</sup> domains as well as the larger surface-to-volume ratio. Deposited graphene flakes film has strong in-plane sp<sup>2</sup> bonds in two-dimensional system.



**Fig. S4.** Graphene Flakes Raman spectroscopy analysis, showing corresponding signature peaks of few layers graphene.



**Fig. S5.** A) Capacitance vs applied voltage (CV) at frequencies ranging 10 kHz to 100 kHz, B) Frequency dependent capacitance ( $C_f$ ) and C) Frequency dependent dielectric constant ( $\epsilon_f$ ), characterization of PVPh applied as dielectric layer in the MIS capacitor.

## References

1. Y. Z. N. Htwe and M. Mariatti, *Journal of Science: Advanced Materials and Devices*, 2022, **7**, 100435.
2. Y. H. Kim, K. H. Kim, M. S. Oh, H. J. Kim, J. I. Han, M. K. Han and S. K. Park, *IEEE Electron Device Letters*, 2010, **31**, 836-838.
3. L. F. Alexander and N. Radacsi, *CrystEngComm*, 2019, **21**, 5014-5031.
4. X. Liu, T.-J. Tarn, F. Huang and J. Fan, *Particuology*, 2015, **19**, 1-13.