## Supplementary Information

## Electrochemical and Electrical Performances of Cobalt Chloride (CoCl<sub>2</sub>) Doped Polyaniline (PANI)/Graphene Nanoplate (GNP) Composite

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## **Dielectric Permittivity**

The frequency dependent dielectric permittivity ( $\varepsilon'$ ) of the PGC composites was measured in the frequency region 50 Hz ~10<sup>7</sup> Hz at room temperature, as shown in figure 1. As observed, the dielectric permittivity of the PGC composites strongly depends on the frequency and rapidly decreased with the increase in frequency. From the figure, it was clearly shown that the dielectric permittivity of the PGC composites was high at the beginning stage and then gradually decreased with the increase in frequency at lower frequency region (50~10<sup>3</sup> Hz). This is the well-known characteristics of a dielectric material.<sup>1</sup> This kind of phenomena of any dielectric material could be well explained with the help of polarization effect.



Figure 1: Dielectric permittivity of the PGC composites vs. frequency at different GNP content.

Accordingly, the dielectric permittivity of a material is proportionally varied with polarizability.<sup>2</sup> The polarization effect is more prominent at low frequency region as the molecules of dielectric materials get sufficient relaxation time to orient themselves in the direction of the applied electric field. Thus, dielectric permittivity of the composites will become

more. With increase in frequency, polarization effect in the dielectric material become insignificant because the molecules are getting less relaxation time for orientation themselves in the direction of applied alternative electric field. This is the reason behind the sharp decrease in dielectric permittivity in the composites.<sup>3</sup> Thus, dielectric permittivity of the polymer composites reduced gradually after a certain frequency ( $\sim 10^3$  Hz).

The result in figure 1 shows that dielectric permittivity of the composites also depends on the filler concentrations and increases with increasing the concentration of GNP. This improvement in dielectric property indicated that the incorporation of nanofillers take a great role to increase the dielectric permittivity of the composites. The enhancement of dielectric permittivity of the composites with the incorporation of nanofillers can be related to several factors, such as (i) the large surface area of nanofillers modifies the morphology of the polymer in the composites, (ii) the large surface area and very high aspect ratio of nanofillers which creates a large interaction zone in the polymer composites, (iii) a reduction in the internal field due to nano-sized of the fillers in the composites, (iv) high electrical conductive nano-sized fillers changes the space charge distribution, and (v) also scattering mechanism of the composites. In the case of nanofiller filled materials, the nano-size of the fillers leads to an exceptionally large interfacial area in the composites. Since nanofillers have a greater volume to surface area ratio due to its nano-sized and high aspect ratio, they hold a much greater interface with their neighbors compared to the macrofillers. Thus, the degree of interfacial interaction among the nanofiller and the host polymer has been controlled by this interface and also controls the electrical and physical properties of the composites. Hill et al.<sup>4</sup> studied the dielectric permittivity of the polymer composites and found an increase in the value of  $\varepsilon'$  at low frequency region due to 'quasi-dc' conduction which is well explained by Lewis<sup>5</sup> with the help of O'Konski's model<sup>6</sup> and a double layer approach. According to the model, the charge carriers moved efficiently around the interface by the field leading to an induced polarization at the polar ends of the fillers.<sup>5</sup> This is the reason for higher dielectric permittivity. Since these double layer effects are likely to be prominent in composites, the slope of the permittivity is steeper than that for the microcomposites in the low frequency region.

## **References:**

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