Magnetic Nanoparticle - C Allotrope Composites: Physical and Electromagnetic Interference Shielding Properties

Abstract

Composites of magnetic nanoparticles and C allotropes are promising materials for spintronic applications, among many. Spintronic applications demand the composite to be semiconducting, apart from being magnetic. This, in turn, requires that the C-allotrope, which is usually the matrix, itself is semiconducting. For this, graphene is a good candidate, because a band gap in its linearly dispersing Dirac cone can be opened by quantum confinement or by adding surface functionalities. The latter is the simplest and most popular method, in which graphene is first converted to the insulating graphene oxide (GO), and then GO is reduced chemically to semiconducting reduced graphene oxide (RGO). The functionalization brings in vacancy defects, which are known to inherently possess magnetic moments. Attachment of $3d$ transition metal atoms at coplanar sites, or magnetic nanoparticles (NPs) at RGO surface, enhances the magnetic properties further via charge transfer between the C-plane and the attached atom or NP. Tuning the electronic transport and magnetic properties of such nanocomposites by the NP magnetism (possibly via charge transfer) is desirable, and studying the underlying mechanism, intriguing. Further, incorporating such C allotrope - magnetic nanoparticle composites additionally in a polymer matrix may result in conducting ternary nanocomposites that could reflect and absorb electromagnetic (EM) waves due to their electrical conductivity and magnetism, and could be used as efficient EM interference (EMI) shielding materials. This thesis is aimed at investigating (i) physical properties of magnetic alloy - RGO nanocomposites, and (ii) EMI shielding properties of magnetic nanoparticle/C-allotrope/polymer nanocomposites.

The thesis contains four contributory chapters (Chapter 3 - Chapter 6). **Chapter 3** constitutes the study of charge transfer, electronic transport and magnetic properties of chemically prepared nanocomposites of RGO and the alloy system $Cu_{1-x}Ni_x$ that has complete bulk solid solubility, and is known to undergo a composition-induced paramagnetic (PM) to ferromagnetic (FM) phase transition in bulk at $x = 0.44$. Raman spectra indicate a charge transfer between the RGO sheets and the nanocomposites. Density functional theory computations confirm the occurrence of charge transfer. Electrical resistivities of the nanocomposites are found to follow thermally activated band conduction mechanism, akin to disordered semiconductors. The nanocomposites are ferromagnetic, and the x-variation of activation energy of electronic transport is explicable in terms of the nanocomposite magnetization, which diverges at x_c. Thus, the magnetism of the nanoparticles is shown to tune the electronic transport and magnetic properties of the nanocomposites via charge transfer. **Chapter 4** is a brief study of electrical and magnetic properties of nanocomposites of RGO and $Ag_{1-x}Ni_x$, which is immiscible in bulk but miscible in nanodimensions, where it shows a PM to FM phase transition at $x = 0.75$. In this case, all the nanocomposites with $x > 0$ are found to be PM semiconductors, with conductivity akin to disordered semiconductors. Raman spectra confirm the presence of a considerable disorder in the nanocomposites. In **Chapter 5**, EMI shielding properties of nanocomposites with 0 - 5 weight % RGO/PdNi filler contents in poly(ethylene-co-vinyl) matrix are studied. Nanocomposites with 1 and 5 weight % filler content exhibit $\sim$ 12 - 30 dB shielding efficiencies (SE's), with maxima in S- and C-band, respectively. An analysis of the electrical conductivity, dielectric permittivity and magnetic permeability suggests that there is a scope to enhance the SE further by varying the Pd:Ni and RGO:alloy ratios. Mechanical and thermal characterisations reveal that the nanocomposites desirably have both the mechanical strength and the thermal stability better than the EVA matrix. Thus, these nanocomposites are found to have a potential in EMI shielding applications. **Chapter 6** is a brief study of EMI shielding properties of Fe_3O_4/C/polypyrrole nanocomposites in the form of core-shell structures. The SE of this structure in the S+C band is found to reach 28 dB. The nanocomposites are found to be FM, wherein an interplay between enhancement in coercivity and diminution in saturation magnetization indicates that the surface spin disorder plays a decisive role in determining the EMI shielding properties.

Keywords: Reduced graphene oxide, Magnetic nanoparticles, Charge transfer, Electronic transport, Magnetism, EMI shielding