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The Characterization of Graphene and CNTs Based Magnetic Composites

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***Abstract*—The magnetic composites of CNTs and graphene have been prepared by thermal decomposition of the ferrocene. The comparative study of their properties has been done using X-ray diffraction and electron paramagnetic resonance (EPR) spectroscopy. The results revealed that the crystallite size and magnetic nature of composites have been changed. The variations of calculated uncompensated spin of free electron in graphene, CNTs and composites have shown their varying magnetic nature.**

***Keywords*—Composite materials, Magnetic materials, Chemical vapour deposition, Magnetic properties**

I. INTRODUCTION

The carbon nanotubes (CNTs) after their discovery by Iijima, they became a promising candidate for versatile applications due to excellent mechanical, optical and electrical properties [1-3]. As such CNTs were generally neutral in nature, but they have been functionalized and coated with different metal ions or particle through various chemical and physical methods. The metal such as lead in molten state could be filled into the open ended carbon nanotubes through capillary action to fabricate nanowires [5]. The carbon nanotubes have also been encapsulated with cobalt nanopaticles by decomposition products of $Co(CO)_3$ NO [6-7]. Stoffelbach and co-workers have been prepared the MWNTs-Fe_3O_4 composites through solution treatment of positively charged Fe_3O_4 nanoparticles with negatively charged carbon nanotubes [8]. At some place, Korneva has been filled the MWNTs with ferrofluid and produced the magnetic tubes with magnetic materials including Fe [9], Ni [10], Co [11], and FeCo [12] have also been encapsulated in CNTs. Sun has fabricated the composites of different morphologies, from CNTs and magnetic nanoparticles through decomposition of ferrocene in presence of MWCNTs, at different temperatures [13-14]. These magnetic composites of coated CNTs could found the great importance in field of magnetic data storage and xerography. In the present study, it was explored that the magnetic composites have been prepared by thermal decomposition of ferrocene in presence of CNTs and graphene at 450°C temperature. The weight ratio of CNTs and graphene with ferrocene was one by one. These prepared samples were characterized by X-ray diffraction and electron paramagnetic resonance (EPR) spectroscopy techniques to investigate the structural and magnetic nature.

II. EXPERIMENT

Synthesis of CNTs

CNTs were synthesized by chemical vapor deposition method [15]. The solution of toluene and ferrocene [$Fe(C_5H_5)_2$] was injected in inert argon atmosphere at 750°C to occur the chemical reaction. The pristine CNTs had various carbon impurities; those were removed by the air oxidation and acid treatment.

Synthesis of magnetic composites

The 0.2g purified CNTs with 0.2g ferrocene and 0.15g graphene with 0.15g ferrocene, were mixed and heating at 450°C for 2hrs at the heating rate of 5°C per min. The samples were collected and washed repeatedly with absolute ethanol to remove excess unreacted ferrocene. Thus composites of Fe_3O_4 with CNTs and graphene were synthesized separately. In figure, they were represented as CNTs by *a*, CNTs-ferrocene by *b*, graphene–ferrocene by *c* and graphene by *d*.

III. CHARACTERIZATION

The synthesized composites were analyzed by Rigaku make powder X-ray diffractometer (Model: XRG2KW) using CuKα radiation (λ=1.54059Å). The X- ray diffraction patters were recorded in 20^o to 70^o range of 2θ at scanning rate of 0.02^o/s, at 40 kV, 30 mA. The free electrons spin concentration and nature of magnetic interaction was observed from their EPR spectra. The EPR spectra were recorded by Bruker BioSpin GmbH at 9.841GHz microwave frequency with 23.0dB attenuation, 8000G sweep width and 100 kHz modulation frequency, at ambient temperature.

IV. RESULT AND DISCUSSION

The composites of Fe_3O_4 with graphene and CNTs have been synthesized through thermal decomposition of ferrocene with CNTs and graphene at 450°C temperature. The X-ray diffraction pattern of CNTs, graphene and their composites has shown in fig. 1. The composite *b* has higher crystalline nature than *c* and *d* both. The diffraction (002) peak at 26.2° was corresponds to graphitic phase of carbon, peaks at (220), (311), (400), (511) and (440) corresponds to magnetite phase [16] and peaks at (012), (104), (110), (113), (024), (116), (214) and (300) were due to maghemite γ-Fe_2O_3 phase [17] by standard (JCPDS Card No-019-0629). The crystallite size was

calculated by Scherrer formula $D=0.9\lambda/\beta\cos\theta$ [18]. The average crystallite sizes calculated from (002) peak varied in 9-15 nm range. It was observed that composite *c* had larger crystallite size than composite *b* as given in table1. The induced strains in *a* and composite *b* have been increased which attributed to surface bonding of magnetic nano-particles with CNTs through magnetic interaction between $Fe^{2+}$and/or Fe^{3+} ions and free electrons of CNTs. The EPR spectra of a, composite *b* and composite *c* were shown in fig. 2. It was observed that CNTs have the small peak at 3500G which attributed to presence of Fe within the CNTs. The composite *c* have symmetrical spectral peak, while in composite *b*, there was slightly broadening of spectral peak. That broadening of spectral peak attributed to change in lattice-lattice relaxation time, which may be due to different structure and magnetic interaction of $Fe^{2+}$and/or Fe^{3+} ions with CNTs, which might be due to the presence of γ-Fe_2O_3 phase. The different EPR parameters like ΔHp-p, g-values and free electrons spin concentration has been calculated and given in table 1. Thus XRD and EPR investigations confirmed that CNTs and graphene based composite had γ-Fe_2O_3 phase, while graphene had Fe_3O_4 phase alone and thus affecting the magnetic interaction.

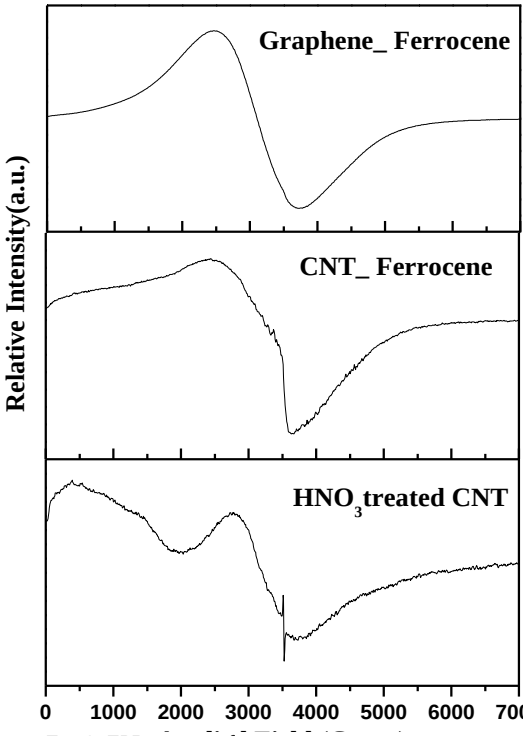

Fig. 2. EPR spectra of composites.

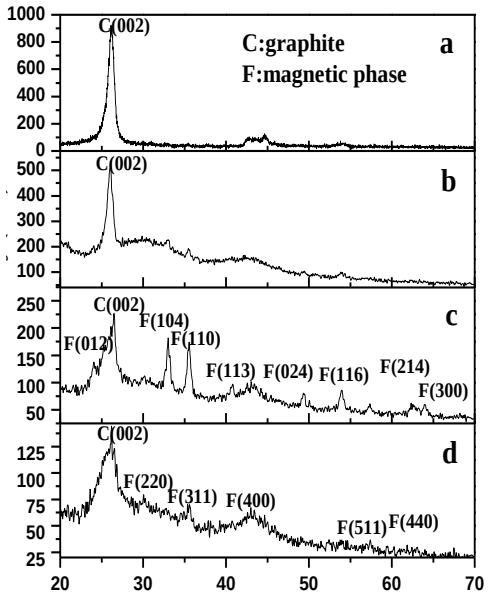

Fig. 1. X-ray diffraction patterns of composites.

TABLE 1.

Composite	2θ-values	d-value	Crystallite size (nm)	ΔHpp (Gauss)	g-value	Spin concentration (Spins / g)
CNTs	26.100	3.411	9.45	939.8	2.25921	1.489 X 10^5
CNTs-ferrocene	26.061	3.4164	13.30	1232	2.18299	2.093 X 10^5
graphene-ferrocene	26.481	3.3632	14.27	1232	2.25914	1.788 X 10^6

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