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FABRICATION AND CHARACTERIZATION OF IRON COATED CARBON NANOTUBES/POLYMER COMPOSITE FOR MICROWAVE ABSORPTION

Electrical characteristics of iron coated multi-walled carbon nanotubes (MWNTs) along with ferromagnetic properties are very interesting nanomaterial for microwave absorption. In this research work, surface morphology, compositions and microwave absorption properties of polymer containing iron coated MWNTs have been investigated. Iron coated multi-walled carbon nanotubes composite were prepared by two simple steps method. In addition, microstructure and microwave absorption properties under frequency range 8÷13 GHz by means of FESEM, EDX &Vector network analyzer had shown. The maximum reflection loss is observed for Fe-coated MWNTs/polymer sample B is –20.86 dB and –18.13 dB at frequency 8.1 and 10.75 GHz respectively. And the maximum bandwidth window is available for sample C is 3.25 GHz from frequency 8.45 to 11.7 GHz with 3 mm thickness, which can be attributed to synergistic effect of improved impedance matching characteristic and superior microwave attenuation characteristic of the absorber. The reflection properties of the material enhanced with variations in the wt.% of Fe-coated MWNTs and polymer. In this research paper, Fe-coated MWNTs are analyzed as promising microwave absorbing material and combined utilization of dielectric loss and magnetic loss absorbent design shows great design flexibility and diversity in the frequency range 8÷13 GHz.

Keywords: multiwalled carbon nanotubes, microwave absorption, reflection loss, radar absorbing material

INTRODUCTION

Now's day, the world heavily depends on the technologies based on electromagnetic radiation. As our technological system grows electromagnetic technology advances, transferring data between terminals have become common in engineering applications and it leads to complexity and the unwanted interactions start to happen between its constituents. Due to Electromagnetic Interference (EMI), the electrical circuits of the devices are affected from other devices' electrical circuits due to electromagnetic radiation interference between them [1]. Due to use of millions of personal electronics it becomes essential to shield the circuits/ objects from unwanted interference in the broad frequencies range. Therefore, from past years, EM waves absorber materials become more commonly utilized in the fabrication of absorbing devices. Radar absorbing materials (RAMs) in the range of microwave frequencies band provide better attenuation to the EM waves and dissipate the absorbed wave energy in the form of heat. Basically, these RAMs are materials that increase electromagnetic radiations energy losses in certain frequency bands [2].

Nowadays, for development and fabrication of RAMs and microwave absorbing materials large numbers of different materials are used [3-14], but CNT is one of the best materials that is widely studied as radar absorbing material. In electromagnetic waves absorption field, CNTs show the advantages of better electrical properties and mechanical properties in broadband frequency range [15-20]. There are various methods of fabrication of metal filled and metal coated CNTs composite. The combination of magnetic loss materials and dielectric loss materials is an effective approach for absorption [21-24]. The combination of outstanding thermal, mechanical, and electronic properties as an advanced filler with better physical and chemical properties in different composites makes CNTs an ideal candidate for microwave absorption. Most of searches in this area are based on the exploration of microwave absorption properties of ferromagnetic materials in CNT/polymer and CNT/epoxy composites [14, 25-39].

Microwave absorbing properties of ferromagnetic nanoparticles such as cobalt, nickel, silver, and iron have been bounded and filled to the surface/cavity of CNTs were investigated. There are some earlier experimental reports for the studies of ferromagnetic properties of carbon nanotubes. Zhao et al. reported that, CNTs containing Ag nanowires provide most of absorption due to dielectric loss but microwave absorbing properties of nickel coated CNTs due to both dielectric properties of CNTs and magnetic properties of nickel [40]. EM wave absorbing behavior and electrical char-

acterization of radar absorbing materials were explored in the range of frequencies 2÷18 GHz [41]. Zhao et al. revealed that the improvement in absorption properties of iron filled CNT/epoxy composites is contribution of magnetic loss and dielectric loss of iron nanoparticles and CNTs, respectively [42]. In 2009, Chen Wang et al. investigated the surface morphologies, compositions, and reflection properties of iron filled CNTs with FeCo alloy nanoparticles [43]. Ki-Yeon Park et al. shown the improved magnetic characteristics of compound nano fillers, most favorable formation of nickel-iron coated CNFs/epoxy composites have absorption more than 90% and microwave absorption window of 3.7 GHz in X-band [44].

Research is dedicated to study the surface morphology, compositions and microwave absorbing properties for radar absorbing applications of polymer composites-based iron coated multi-walled carbon nanotubes (MWNTs) in the range of frequency 8÷12 GHz. During coating of the iron particles on MWNTs, the nitrogen atmosphere was required. Nitrogen gas was fed into flasks through syringe needle which was connected with syringe and then balloons. Main principle of this work was to coat the magnetic nanoparticles on the MWNTs and to study material characteristics i.e., morphology and elemental analysis of nanocomposites. Then polyester was added to Fe/MWNTs nanocomposites, to made the samples harder and to study electrical characteristics i.e., reflection loss performance in the frequency range of 8÷13 GHz.

EXPERIMENTAL

Multiwalled carbon nanotubes were purchased from Aritech Chemazon, Haryana, India and MWNTs have diameter range 2÷10 nm, length 2÷10 microns and the pure is more than 97%. For process synthesis, during refluxing first step was heating of the MWCNTs in nitric acid (68%) for 7 hours to extract the trace impurities from CNTs. Next, the mixture was filtered on a porous glass filter with distilled water to neutral pH (7pH). After that collected black precipitates were dried 50°C for 12 hour in a hot air oven [45-54].

TABLE 1. **Details of varying amount of MWNTs, ferric choloride, ferrous sulphate**

Sample No.	MWNTs [mg]	FeCl₃ [g]	FeSO₄ [g]
A	625	30.42	26.25
B	625	24.30	21.08
C	625	18.25	15.85
D	625	12.15	10.55

Acid treated MWNTs mixed with 9 milliliters of iron solution (FeCl₃ and FeSO₄.7H₂O) were poured to 90 mL of NaOH dropwise under the nitrogen environment at 80°C. The compositions of the four different

samples are given in Table 1. To each sample was washed 5-6 times with distilled water up to pH 7.0 and black precipitates were collected after filtration and then dry it at room temperature overnight.

Each iron coated MWNTs sample was uniformly mixed in anhydrous ethanol solution by sonication for 25 min and then the polyester polymer was put into each sample mixture. Then, each sample mixture was sonicated for 50 min and continuously stirred until the mixtures became sol like solution. Then the few drops of liquid hardener (MEKP) were put into each sample mixture. After uniformly mixing, the mixtures were rapidly dropped into the 30 mm x 30 mm size of the rectangular moulds and dried under room temperature naturally.

RESULTS AND DISCUSSION

Morphology of raw MWNTs & Fe-coated MWNTs

Fe-coated MWNTs are obtained in the type of black voluminous powder have ferromagnetic properties. Field emission scanning electron microscopy (FESEM) &Energy Dispersive X-Ray Spectroscopy (EDS) tools were used to understand the surface morphology and composition of iron coated MWNTs nanocomposites. Figure 1 represents the FESEM of raw MWNTs&acid purified MWNTs. The tubular MWNTs with diameter range of 5÷20 nm and length 2÷10 microns are clearly visible from the FESEM – Figure 1a.
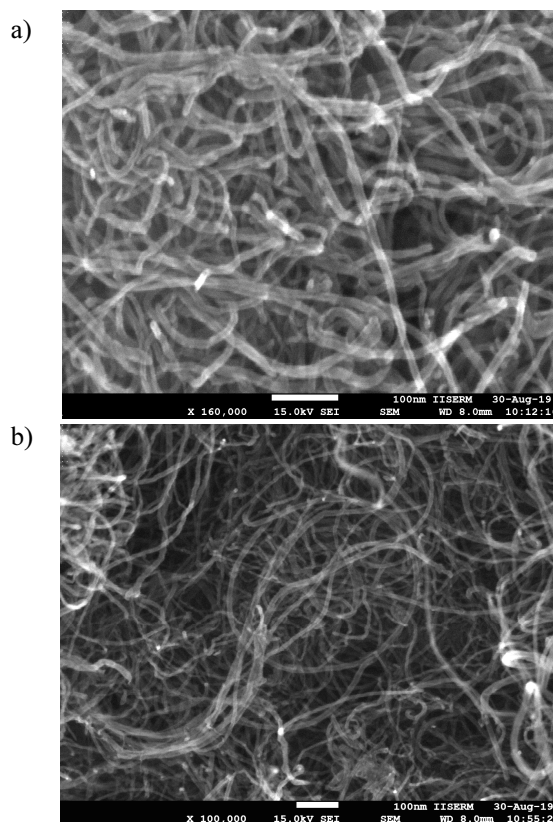

Fig. 1. FESEM of raw MWNTs (a) FESEM of oxidized raw MWNTs (b)

The results show that, the surface of raw MWNTs is very clean and smooth. And most of the nanotubes are curvy and twist together. The characterization of oxidized raw MWNTs is shown in Figure 1b. The MWNTs treated by HNO₃ acid revels that surface morphology of MWNTs is modified with some new defects. Because of acid purification it is simple to break C-C bonds, easy to open the sides of MWNTs, increase the dangling bonds in MWNTs and easy to attach the functional groups on MWNTs' surface. It is seen that the MWCNTs' surface is slightly damaged by purification treatment. The damage may be caused by excessive HNO_3 acid concentration. The sample composition graph of the raw MWNTs and oxidized MWNTs is shown in Figure 2. It is clearly visible from the graphs that MWNTs are pure form of carbon source.

FESEM images of each sample A to D are shown in Figure 3, respectively. Results shows, the iron nanoparticles are successfully coated on the walls of MWNTs.

From results it is clear that sample A and C are composed of mainly MWNTs. And FESEM images of sample B and D shows the thick coating of iron particle on MWNTs' walls.
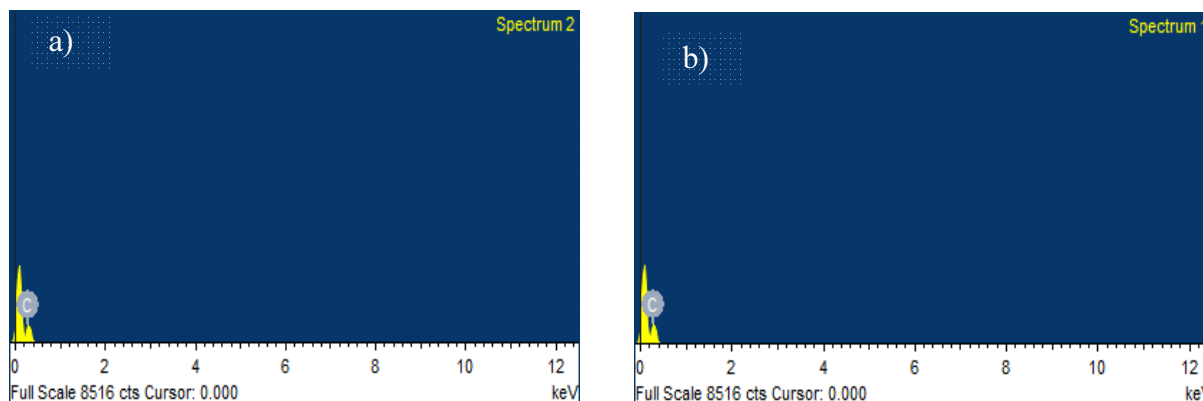

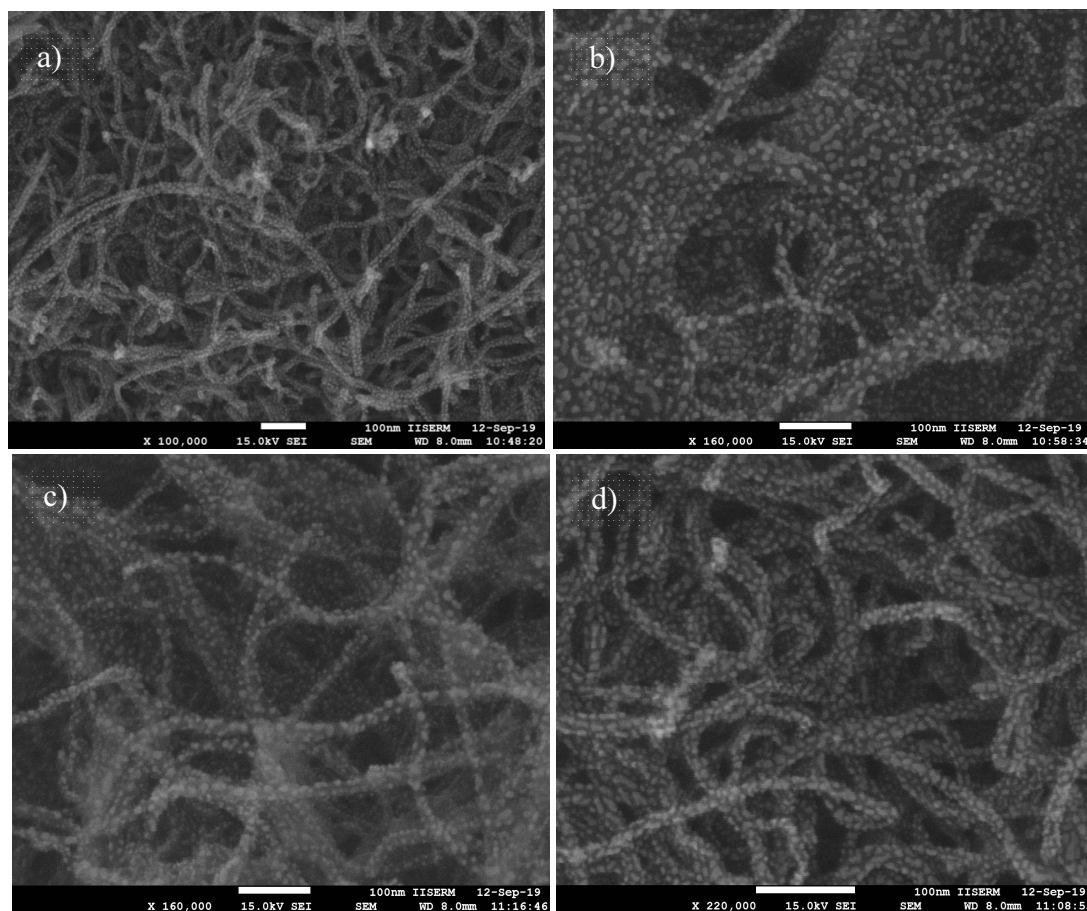Fig. 2. EDS of raw MWNTs (a) EDS of oxidized MWNTs (b)

Fig. 3. FESEM of iron coated MWNTs sample A (a), sample B (b), sample C (c), sample D (d)

EDX spectrum and element analysis, element weight percentage of and atomic percentage present of iron, carbon and unwanted impurities present in sample A-D are shown in Figures 4, 5, 6 and 7 respectively. Sample A, C and D contains carbon and iron but also contain impurities of silicon due to use of silicon crucible as shown in Figures 4, 6 and 7, respectively. Sample B has 48.88% carbon and 35.88% iron, also have impurities of oxygen arise due to the oxide's impurities entrapped in nanoparticles. Figure 7b shows that sample D have number of unwanted impurities present like oxygen, sodium and Silicon, but due to their low weight percentage it doesn't affect the microwave absorption performance.
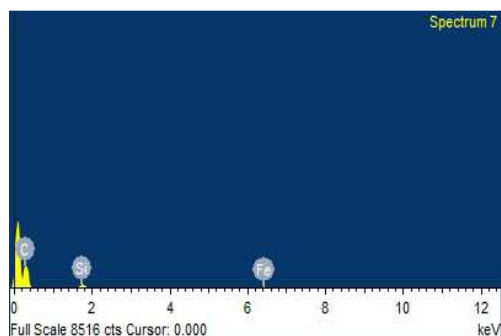

Element	Weight%	Atomic%
C K	72.64	38.63
Si K	5.08	6.31
Fe K	22.28	55.06

Fig. 4. EDX spectrum of sample A (a), EDX composition of sample A (b)

Element	Weight%	Atomic%
C K	48.88	20.8
O K	15.24	8.6
Fe K	35.88	70.6
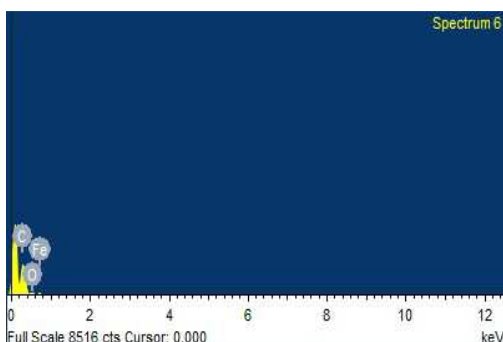
Fig. 5. EDX spectrum of sample B (a) EDX composition of sample B (b)

Element	Weight%	Atomic%
C K	50.12	23.36
Si K	13.04	15.13
Fe K	36.84	61.51
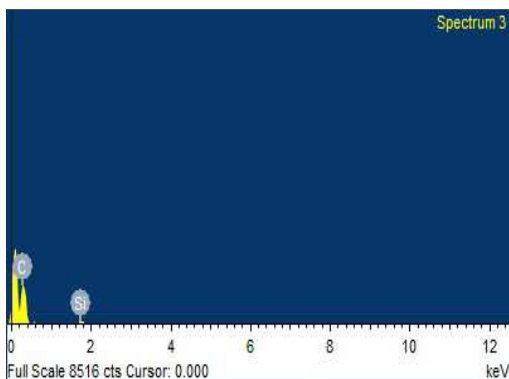
Fig. 6. EDX spectrum of sample C (a), EDX composition of sample C (b)

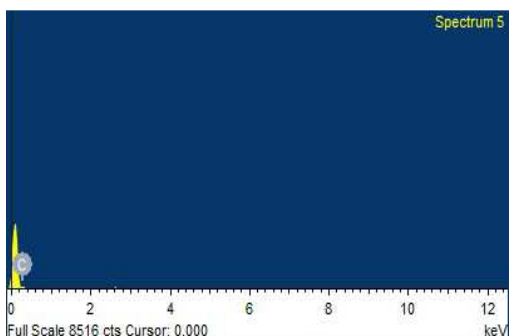| Element	Weight%	Atomic%
C K	60.55	30
O K	8.9	5.99
Fe K	25.33	58.6
Na K	3.50	3.33
Si K	1.72	2.08

Fig. 7. EDX spectrum of sample D (a), EDX composition of sample D (b)

Microwave reflection properties of iron coated CNTs/polymer composites

The reflection peak spectrum in the range of frequency 8÷13 GHz of composite with same wt.% MWNTs and different iron concentration are sum up in Figure 5. From the curves, the reflection loss peak of sample B has two maximum reflection peaks –20.86 dB and –18.13 dB at 8.1 GHz and 10.75 GHz, respectively. For sample A and D, the reflection loss peak reaches –14.39 dB and –15.41 dB at 12.72 and 8.52 GHz, respectively. Reflection peak for sample C reach about –17.3 dB at 12.6 GHz with maximum bandwidth 3.25 GHz. From the results it is clear that sample B have maximum reflection loss peak and maximum frequency absorption band is available for sample C.
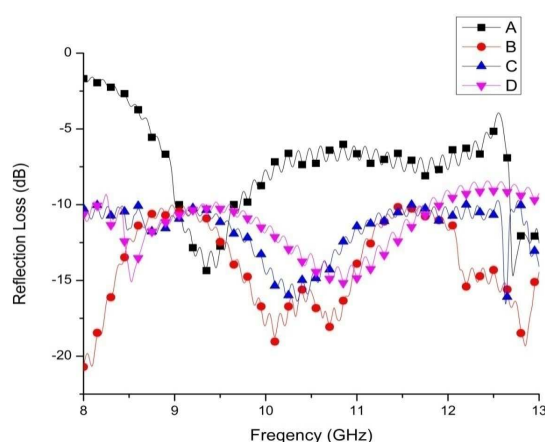

Fig. 8. Absorption characterization of MWNTs and different wt.% of iron (A, B, C & D)

Due to nitric acid treatment, more dangling bonds and defects are existing in Fe-coated MWNTs. Because of the interaction of charge multipoles at the interface between Fe-coated MWNTs and polymer with the microwave radiations, these dangling bonds and defects give rise to a greater number of scattering of the EM waves and interfacial electric polarizations, which provide a mechanism of microwave absorption. As stated by the Kubo theory, in the Fe coated MWNTs the energy levels are not continual but they are split due to the quantum confine effect. While an energy band of the Fe coated MWNTs are in the microwave energy range, the electrons at lower energy level will start to absorb the photons jump to higher energy level. The quantum confine effect greatly changes the magnetic and absorbing properties of Fe. The MWNTs and polymer have many interfaces between their layers and outer surface. Therefore, interfacial multipoles between MWNTs and polymer contribute to the microwave absorption.

CONCLUSIONS

Iron coated MWNTs/polyester composites have been investigated as a microwave absorbing material via simple method. The experimental results demonstrate that the reflection loss of MWNTs is increased up to a certain level with loading of iron, further increase in iron loading weaken the absorption abilities of MWNTs. The maximum reflection loss is observed for Fe-coated MWNTs/polymer sample B is –20.86 and –18.13 dB at frequency 8.1 and 10.75 GHz respectively. And the maximum bandwidth window is available for sample C is 3.25 GHz from frequency 8.45 to 11.7 GHz with 3 mm thickness. The reflection loss enhancement of the Fe coated MWNTs/polyester composites due to the contribution of both MWNTs and Fe. Due to the interfacial multipoles between Fe coated MWNTs and polymer, the microwave absorption of Fe coated MWNTs/polyester composites is increased. It is clear from the results that these Fe coated MWNTs composites can be potential composite for microwave absorption with wide absorption frequency window in X-band range.

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