

Ultrasonic Assisted Preparation and Characterization of Conductive Polyaniline-Modified Magnetite Nanocomposites (PAni/Fe₃O₄ Nanocomposites)

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ABSTRACT

Polyaniline (PAni) is one of the most widely studied conducting polymers due to its easy synthesis method, reversible chemical reaction, and high environmental stability, low cost and flexible electrical conductivity. Blending PAni with some quantity of aniline dimer-COOH modified Fe₃O₄ nanoparticles can form novel composites with suitable electric and magnetic properties. Standard four-point probe apparatus were used to measure the conductivity of pure Aniline, Polyaniline (PAni) and PAni/Fe₃O₄ nanocomposites, while magnetic properties of the magnetite (Fe₃O₄) nanoparticles and PAni/Fe₃O₄ nanocomposites were recorded on Vibrating Sample Magnetometer (VSM). It was found that the electrical conductivity was increased and decreased with addition of Fe₃O₄ nanoparticles (1.8×10^{-9} S/cm, 6.32×10^{-1} S/cm, and 2.3×10^{-3} S/cm). It was also observed that the composites exhibit super paramagnetic performance with saturation magnetization (M_s) of $M_s = 22.5 \text{ emug}^{-1}$. The morphology and elemental composition analysis of the modified Fe₃O₄ nanoparticles and PAni/Fe₃O₄ nanocomposites was studied using Field Emission Scanning Electron Microscope (FESEM). However, the obtained Fe₃O₄ nanoparticles was found to be spherical and agglomerated, due to higher magnetic attraction between the particles, while the modified Fe₃O₄ nanoparticles embedded in the PAni nanotubes lead to the formation of the composites with clear nanofibers structure. The absorption analysis of the modified Fe₃O₄ nanoparticles and PAni/Fe₃O₄ nanocomposites studied by UV-visible found to be 370 nm and 650 nm respectively which can be attributed to π - π transition of benzenoid ring of PAni, suggesting that the conductive PAni was formed. The result was further supported by XRD and FTIR analysis.

Keyword: Polyaniline, Fe₃O₄ Nanoparticles, Composites, Conductivity, Field Emission Scanning Electron Microscope (FESEM).

1. INTRODUCTION

Nowadays, the functionalized electromagnetic nanostructure of the conducting polymers has become field of interest by many researchers, owing to their potential application in the field of science and engineering due to their unique optical, electric and magnetic properties [1, 2]. One of the major taxes is to design a nanostructure that would have a significant effect on the magnetic and dielectric properties. Blending magnetic nanoparticles with conducting polymers

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has become a promising and most considerable approach to achieve desired modulated electromagnetic functional nanomaterials [3, 4]. Polyaniline (PAni) is one of the outstanding conducting polymer with essential electrical conductivity, its good reversible redox reaction, excellent processibility and high environmental stability, as well as several potentials applications make it suitable candidate for many application [5, 6].

The magnetite (Fe_3O_4) nanoparticles have been extensively studied owing to their unique and tenable magnetic properties, drug/gene delivery, their magnetic feature is widely used in many applications such as environmental remediation [7], biomacromolecule separation, magnetic resonance imaging [8], catalyst separation [9], and magnetic recording release. Blending PAni nanotubes with certain quantity of modified Fe_3O_4 nanoparticles will form suitable matrix for achieving PAni/ Fe_3O_4 nanocomposites or absorption materials by doping with iron oxide nanoparticles. PAni is commonly prepared by oxidizing aniline monomer in acid solution via either chemical or electrochemical method. In comparison with electrochemical method, the chemical preparation of PAni is widely used for mass production [10,11]. Different class of oxidants such as $(\text{NH}_4)_2\text{S}_2\text{O}_8$, $\text{H}_2\text{Cr}_2\text{O}_7$, KIO_3 , and H_2O_2 can be used in the preparation of PAni. Among these oxidants $(\text{NH}_4)_2\text{S}_2\text{O}_8$ is the most commonly used oxidant [12,13]. Fe_3O_4 possesses very good ferromagnetic properties and resistance toward the corrosions due to acid solution. Since Fe_3O_4 is hydrophobic materials, chemicals reaction with PAni increase its dispersion in solution. It also exhibits good electrical behaviour when external electric field is applied to materials [14, 15].

In the current study, polyaniline/iron oxide (PAni/ Fe_3O_4) nanocomposites were prepared by ultrasonic dispersion method. The polymerization of aniline was chemically produced using hydrochloric acid (HCl) as acidic medium and ammonium persulfate $(\text{NH}_4)_2\text{S}_2\text{O}_8$ as oxidizing agent which lead to the formation of electromagnetic PAni/ Fe_3O_4 nanocomposites. The composites were synthesized using uniform dispersion of Fe_3O_4 nanoparticles in hydrochloric acid, Aniline and Fe_3O_4 nanoparticles were polymerized together, to obtained homogenous composites of PAni/ Fe_3O_4 nanocomposites. This work was based on ultrasonic technique to produce stable PAni nanotubes containing modified Fe_3O_4 nanoparticles. However, the electric and magnetic properties of the composites was further analyzed using standard four point probe apparatus and Vibrating Sample Magnetometer (VSM) respectively. Furthermore, the composites were characterized by Field Emission Scanning Electron Microscope (FESEM), Fourier Transformation Infrared Spectroscopy (FTIR), X-ray diffraction Spectra (XRD) and Ultraviolet (UV-VIS)

2. MATERIALS

In this work, PAni/ Fe_3O_4 nanocomposites were synthesized using ultrasonic techniques. (FeCl_3) (Guangdong Xirong Chemical Plan), ferrous chloride (FeCl_2) (Chongqing Beibei Chemical Reagent Factory), ammonium hydroxide (NH_4OH) (R & M Chemical), N-phenyl-1,4-phenylenediamine (Tianjin Tiantai Fine Chemical Co., Ltd), Succinic anhydride (Sinopharm Chemical Reagent Co., Ltd), Diethyl ether, and dichloromethane CH_2Cl_2 (R & M Chemical). Aniline ($\text{C}_6\text{H}_5\text{NH}_2$ R & M chemicals) ammonium peroxodisulfate $(\text{NH}_4)_2\text{S}_2\text{O}_8$ and hydrochloric acid (HCl R & M 37%) were used in the production of PAni/ Fe_3O_4 nanocomposites. The entire chemicals were used without further purification.

2.1 Aniline Dimer-COOH Preparation

The preparation aniline dimer-COOH was carried out under 200-ml round bottom flask using mechanical stirrer. 0.721 g of N-phenyl-1, 4-phenylenediamine and 3.5 g of succinic anhydride were dissolved into a 30 ml of dichloromethane (CH_2Cl_2) under a mechanical stirrer for a period

of 5 h. White grey precipitate was obtained. Finally, the precipitate was washed, filtered and then dried in a vacuum for 24 h.

2.2 Preparation of magnetite (Fe_3O_4) nanoparticles

The Fe_3O_4 nanoparticles was achieved when 1.82g of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ and 3.7 g of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ was dissolved in 60 ml distilled water under the influence of the magnetic stirrer. The reaction was allowed to be heated to 60 °C, then 10 ml ammonium (NH_4) was added into the reaction, followed by addition of 400 mg of aniline dimer-COOH in 5 ml of acetone. The reaction was then allowed to continue for 1.5 h at 90 °C under the mechanical stirrer to obtained stable water base suspension. The reaction was allowed to cool to ambient temperature, then filtered, washed and dried for 12 h in the vacuum.

2.3 Preparation of polyaniline/magnetite (PAni/ Fe_3O_4) nanocomposites

PAni/ Fe_3O_4 nanocomposites were synthesized by ultrasonic irradiation method. In the typical procedure, 500 mg of Fe_3O_4 nanoparticles modified with aniline dimer-COOH were successfully dispersed in 30 ml of HCl solution in 200 ml beaker under magnetic stirrer at 0-10 °C. However, 0.5 g of aniline was added to the above mixture. A solution of 1.22 g ammonium persulfate (APS) was dissolved in 30 ml of HCl solution and then added in drop wise to the to the above reaction for a period of 10 minutes. The reaction was stirrer under ultrasonic disperser for 7 h and then filtered. The precipitate was washed with distill water and methanol several times; the product was then dried under the vacuum at 50 °C for 24 h.

2.4 Characterization

The conductivity of PAni/ Fe_3O_4 nanocomposites and PAni were measured using standard four-point probe apparatus (Guangahou semiconductor materials) at ambient temperature. The magnetic properties of the modified magnetite (Fe_3O_4) nanoparticles, as well as PAni/ Fe_3O_4 , were measured using vibrating sample magnetometer JDM-1 in the magnetic field of 15K0e at room temperature. Fourier Transformation Infrared Spectroscopy (FTIR) was used to determine the molecular structure and functional group of Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanocomposites samples (Nicolet 560 FTIR spectrometer brand). The phase structure of the modified Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanocomposites was estimated using X-ray diffraction analysis (XRD) at a radiation wavelength of ($\lambda = 0154$ nm) on X-pert pro MPD diffractometer using Cu $\text{K}\alpha$ radiation. The optical absorption spectrum of modified Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanocomposites was taken through ultraviolet UV-VIS. The morphologies of PAni nanotubes containing modified Fe_3O_4 nanoparticles were investigated by Field Scanning Electron Microscopes (FESEM) (JEOL JSM-7600F) equipped with energy dispersive X-ray spectra.

3. RESULT AND DISCUSSION

3.1 UV-vis Analysis

The absorption spectra of the magnetite (Fe_3O_4) nanoparticles modified aniline dimer-COOH and PAni/ Fe_3O_4 nanocomposites were investigated using UV-VIS spectroscopy. The absorption band of the Fe_3O_4 nanoparticles modified aniline dimer-COOH was observed at a wavelength of 250 nm and 370 nm. That was corresponded to the π - π^* transition in the benzenoid ring of the aniline dimer-COOH [16, 17]. Fe_3O_4 nanoparticles sample modified with aniline dimer-COOH was repeatedly washed with acetone and ethanol which can easily dissolve aniline dimer-COOH completely. There is a strong indication that there is an interaction between aniline dimer-COOH and Fe_3O_4 nanoparticles. The absorption spectra of PAni/ Fe_3O_4 nanocomposites was

observed at 370 nm and 650 nm, as shown in Fig 1, the first absorption band can be attributed due to partial π - π^* transition of benzenoid ring and partly polaron band transition due to broad future Fe_3O_4 nanoparticles. The second transition can be assigned to the polaron band transition, suggesting that the conductive PANi was formed [18, 19].

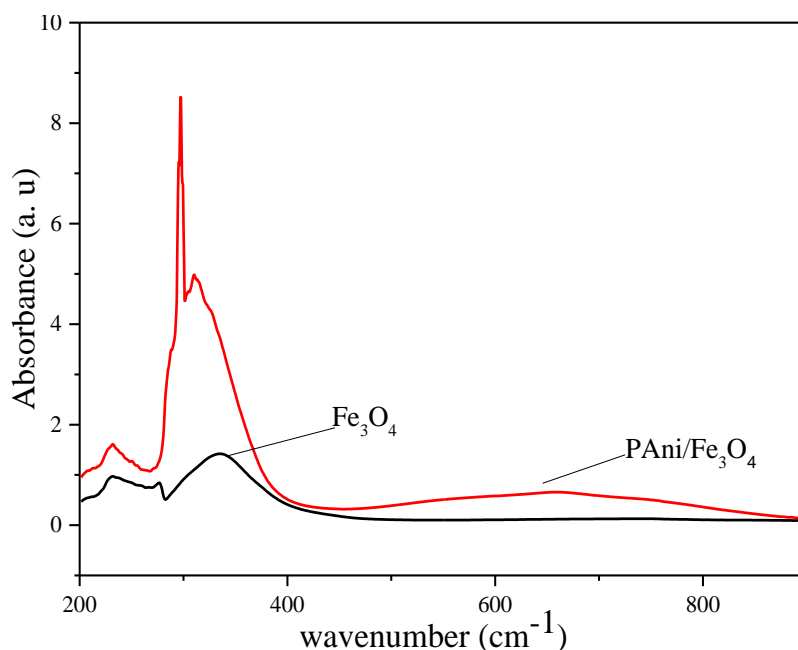


Figure 1. UV-vis of modified Fe_3O_4 nanoparticles and PANi/ Fe_3O_4 nanocomposites.

3.2 FTIR Analysis

The FTIR result is shown in Fig 2. The IR spectra of Fe_3O_4 nanoparticles modified with aniline dimer-COOH indicates the existence of peak at 3238 cm^{-1} , 2819 cm^{-1} and 1663 cm^{-1} , 1415 cm^{-1} , 1214 cm^{-1} , 1089 cm^{-1} and 832 cm^{-1} , which are assigned for O-H stretching, C-H stretching, C=C stretching, C=O stretching and C-O stretching bands respectively. This indicates that the acidic medium condition of Fe_3O_4 preparation. A peak at 557 cm^{-1} represents the characteristic peak of Fe-O stretching band [20]. In PANi/ Fe_3O_4 nanocomposites spectra, it was found that a peak at 3328 cm^{-1} (N-H stretching) is slightly disappeared due to a weak vibration, thus a chemical interactions of the nanocomposites attachment occurred. Peaks at 2828 cm^{-1} , 2258 cm^{-1} , 1767 cm^{-1} , 1588 cm^{-1} and 1588 cm^{-1} represent the C=O, C-H, C=N and C=C stretching of quinoid and benzenoid rings, respectively. Meanwhile, a peak at 1303 cm^{-1} assigned for C-N secondary aromatic amine stretching [21]. The C-H deformation bond at 1,4-disubstituted benzene ring is confirmed at peaks 943 cm^{-1} and 825 cm^{-1} . The differences of peaks shifting on both Fe_3O_4 nanoparticles and PANi/ Fe_3O_4 nanocomposites spectra are due to the weakening in the PANi bonds [22]. Most of those peaks are slightly diminished in the PANi/ Fe_3O_4 nanocomposites spectra, thus showing that the blending of Fe_3O_4 on the PANi matrix was successfully formed [23]. A scheme representation of modified Fe_3O_4 nanoparticles and PANi/ Fe_3O_4 nanocomposite prepared by ultrasonic irradiation is presented in Fig 3.

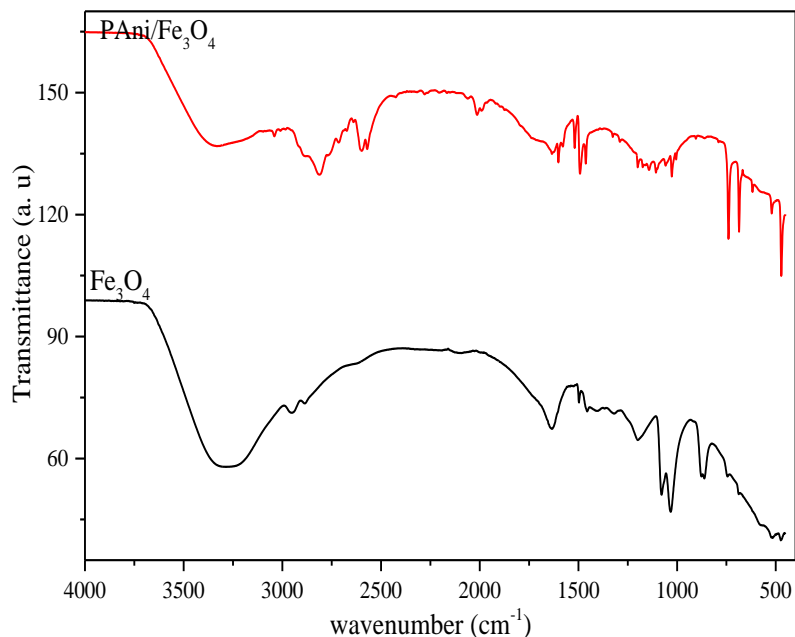


Figure 2. FTIR analysis of modified Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanocomposites.

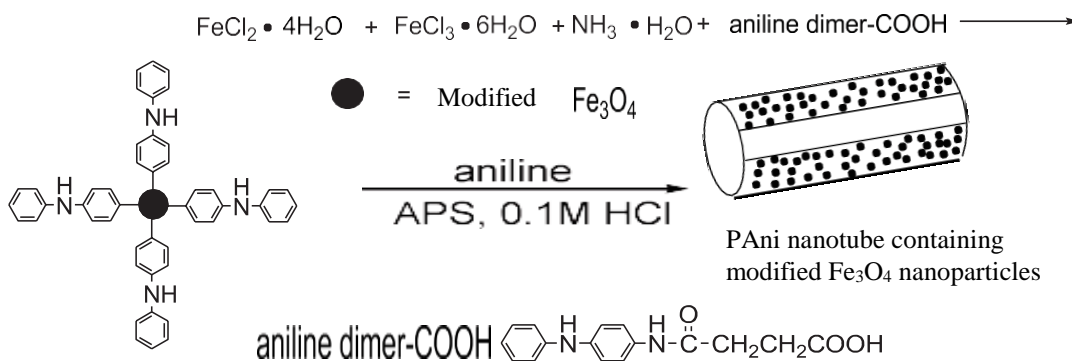


Figure 3. Scheme representation of modified Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanocomposite prepared by ultrasonic irradiation.

3.3 VSM Analysis

The magnetic properties of the modified Aniline dimer-COOH magnetite (Fe_3O_4) nanoparticles and PAni/ Fe_3O_4 nanocomposites was determined using the standard vibrating sample magnetometer (VSM) apparatus at a room temperature. A typical magnetization curve was plotted as a function of applied field at ambient temperature (300K) as shown in Fig 4. The Fe_3O_4 nanoparticles exhibit soft magnetism with a low remanence (13.85 emu/g) and low coercive force (25.95 Oe). The saturation magnetization of the magnetic nanoparticles was found to be 58.10 emu/g, which is much higher than that of the nanoparticles prepared by the common microwave irradiation method (about 40–55 emu/g) [21]. The PAni/ Fe_3O_4 nanocomposites was observed at 22.5 emu/g. The high drying temperature in the modified Fe_3O_4 nanoparticles results in a higher crystallinity and consequently leads to a higher saturation magnetization [24]. The hysteresis loop of the particles exhibits essentially no remanence or coercive force. This interesting result can be compared with a previous conclusion, where ferromagnetic particles can be dispersed in aqueous media. The remanence and the coercive force of the Fe_3O_4 nanoparticles dispersed in aqueous solution were found to decrease to 0.042 emu/g and 1.94 Oe, respectively.

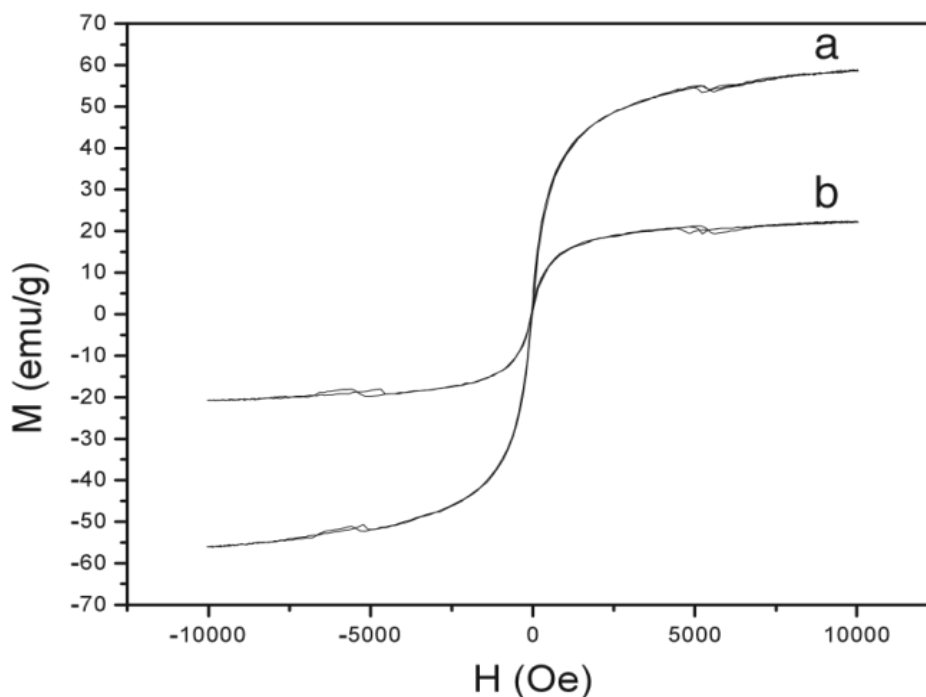


Figure 4. Magnetization versus applied magnetic field at room temperature for the (a) aniline dimer-COOH capped Fe_3O_4 nanoparticles and (b) PANi/ Fe_3O_4 nanocomposite.

This phenomenon indicates that, if suitable surface modification is performed to decrease the magnetic force between the adjacent nanoparticles, the ferromagnetic particles can rely on the Brownian motion to disrupt the chain-like structure induced by the magnetic moment. Then, a transformation from ferromagnetism of the particles to a hysteresis loop with much small remanence and coercive force of the particles suspension appears. In this case, there will be no significant magnetic interactions remaining after removing the external magnetic field, which is important in achieving a reproducible and reversible optical response [25].

3.4 XRD Analysis

The phase structure of modified Fe_3O_4 nanoparticles and PANi/ Fe_3O_4 nanocomposites were estimated using X-ray diffraction spectra (XRD), shown in Fig 5. The polymeric composite was obtained under chemical state of iron oxide after being incorporated into the PANi matrix. The characteristic peaks for the composites were identified, the peaks at $2\theta = 24.2^\circ, 32.3^\circ, 35.65^\circ, 41.3^\circ, 43.4^\circ, 47.3^\circ, 49.6^\circ, 54.2^\circ, 58.42^\circ, 63.2^\circ, 64.3^\circ, 72.12^\circ$ and 78.7° correspond to the (101), (110), (311), (202), (302), (222), (122), (331), (112), (401), (501), (411), (143), and (201) Bragg reflection respectively. Absence of peaks from 10° to 20° indicated the amorphous behaviour of PANi containing Fe_3O_4 nanoparticles [26, 27]. Furthermore, these results indicated that Fe_3O_4 nanoparticles are still present in the structure. The average size of the particles was calculated from half-width of the diffraction peaks using Equation 1.

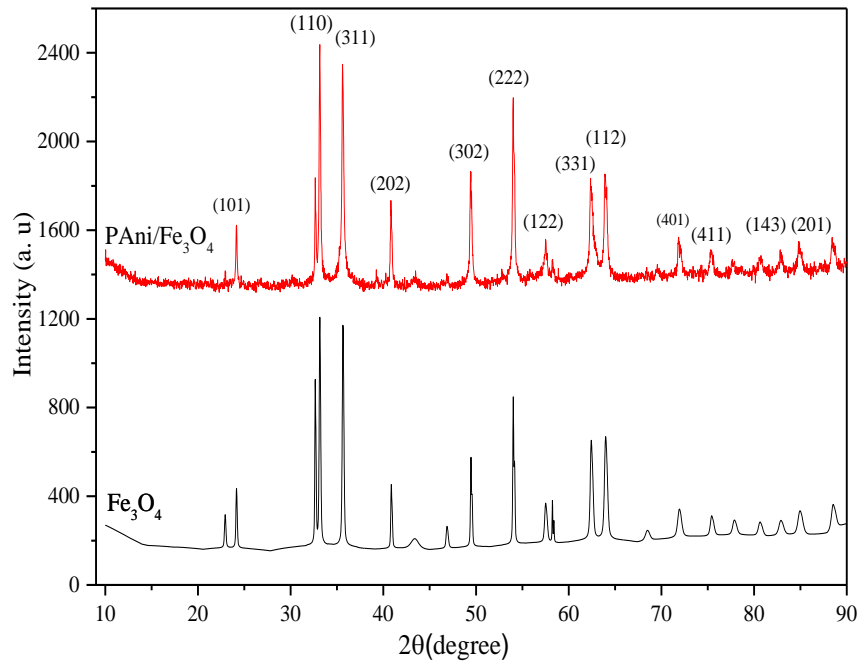


Fig. 5: XRD analysis of modified Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanocomposites

$$D = \frac{K \cdot \lambda}{\beta \cdot \cos \theta} \quad (1)$$

Where K (0.94) is a dimensionless quantity, λ is the X-ray wavelength, β is the line broadening at half-maximum intensity (FWHM) and θ is the Bragg angle. The obtained particles size was calculated using Equation 1. The average particle size was calculated using Scherer's formula and was found to be 30 nm.

3.5 FESEM Analysis

The morphologies of the Fe_3O_4 nanoparticles and PAni/ Fe_3O_4 nanoparticles were investigated using Field Emission Scanning Electron Microscopes (FESEM) equipped with energy dispersive X-ray (EDX). From Figure 6, it was observed that the morphology of Fe_3O_4 nanoparticles modified aniline dimer-COOH was found to be spherical. However, the spherical nanoparticle has higher internationalization rate and high cellular take up than another shape like nanorod or nanodisk [29]. The tendency of agglomeration of nanomaterials is a direct consequence with enhancement of surface energy and small grain sizes of the particles [28]. The Fe_3O_4 nanoparticles exhibit small grain size and consequently enhanced surface energy. Hence, their agglomeration is expected and clearly observed in the FESEM analysis. The agglomeration could be attributed to magnetic dipole interaction with neighbouring Fe_3O_4 nanoparticles [27]. Reduction in surface energy and magnetic dipolar interaction prompt the modified Fe_3O_4 nanoparticles to surround the PAni nanotubes and settled at the surface of the PAni matrix [25].

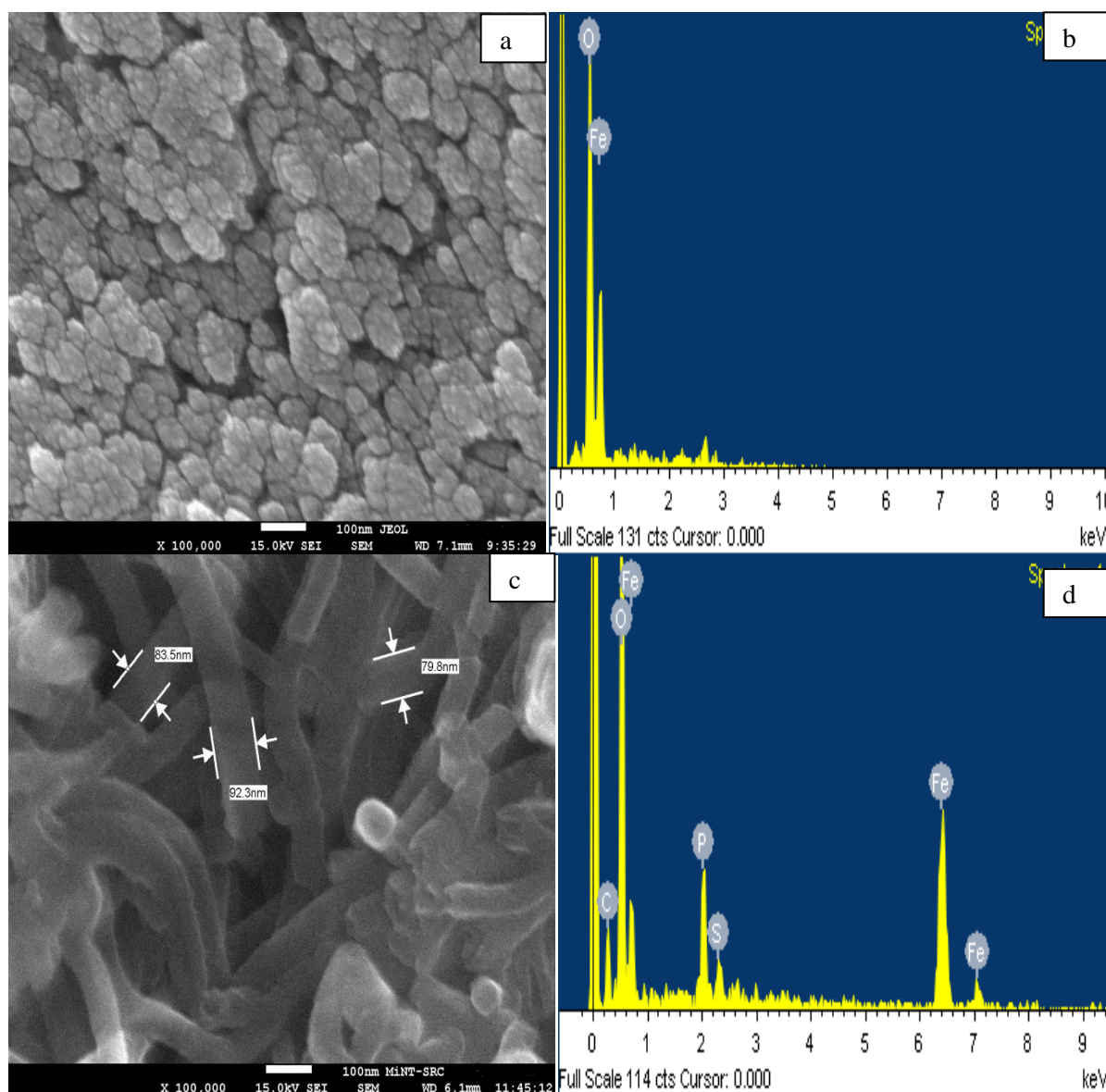


Figure 6. FESM image of a & b) modified Fe₃O₄ nanoparticles and EDX spectra c & d) PANi nanotubes containing Fe₃O₄ nanoparticles.

Embedding PANi nanotubes with certain quantity of modified Fe₃O₄ nanoparticles leads to the formation of PANi/Fe₃O₄ nanocomposites with very clear nanofiber structures. Due to possible interaction between PANi and modified Fe₃O₄ nanoparticles the aniline molecules may be absorbed on the surface of Fe₃O₄ nanoparticles [30]. The PANi/Fe₃O₄ nanocomposites was successfully synthesized through ultrasonic irradiation method. Fig 6 presents the typical FESEM and EDX image of Fe₃O₄ nanoparticles and PANi/Fe₃O₄ nanocomposites. The result shows the appearance nanofiber in the PANi/Fe₃O₄ nanocomposites, while the EDX result indicates the present of Fe₃O₄ nanoparticles and PANi/Fe₃O₄ nanocomposites.

3.6 Electrical Conductivity Study

The electrical conductivity of pure aniline, polyaniline (PANi) and PANi nanotube containing modified Fe₃O₄ nanoparticles was estimated using four-point probe method. It was found that the conductivity of pure aniline (acid free) is 1.8×10^{-9} S/cm. While the electric conductivity of polyaniline (PANi) was found to increase to 6.32×10^{-1} S/cm, further addition of modified Fe₃O₄ nanoparticles decrease the conductivity of PANi/Fe₃O₄ nanocomposites to 2.3×10^{-3} S/cm. This

fluctuation of the conductivity values is due to insulation behaviour of Fe_3O_4 nanoparticles introduced in to PANi matrix [31]. Fig. 7 shows the variation of conductivities of aniline, Polyaniline and PANi/ Fe_3O_4 nanocomposites.

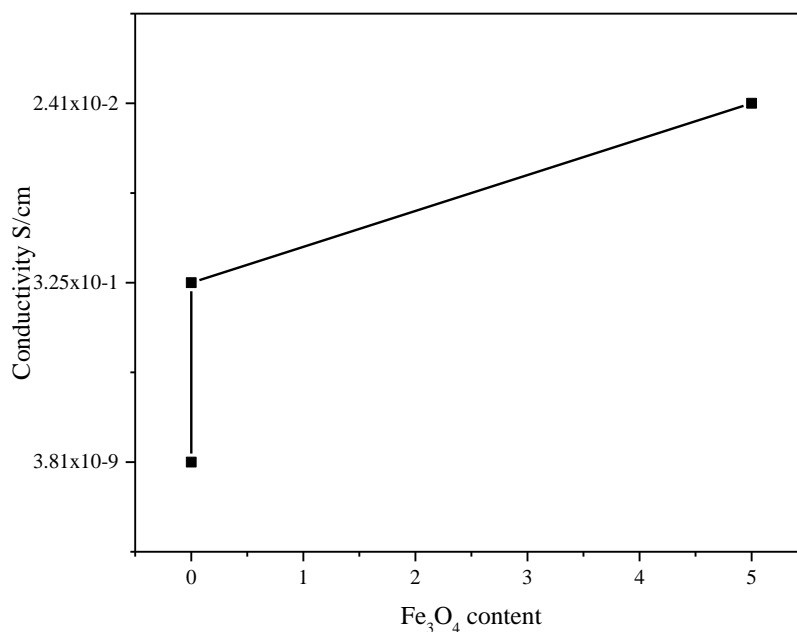


Figure 7. variation of conductivities of Aniline, Polyaniline and PANi/ Fe_3O_4 nanocomposites.

4. CONCLUSION

Polyaniline (PANi) containing modified Fe_3O_4 nanoparticles was successfully prepared via ultrasonic irradiation method. The polymerization of aniline was carried out in the presence of HCl and ammonium persulfate (APS) as oxidants. The electrical conductivity of aniline, polyaniline and PANi/ Fe_3O_4 nanocomposites was measured on four-point probe apparatus. It was found that the conductivity was initially increased and then decreased due to addition of Fe_3O_4 nanoparticles in the composites (1.8×10^{-9} , 6.32×10^{-1} and 2.3×10^{-3} S/cm). Furthermore, the magnetic properties of the magnetite nanoparticles and PANi/ Fe_3O_4 nanocomposites were measured using VSM. As a result, the Fe_3O_4 nanoparticles were found to be within the range of ferromagnetic magnetic nanoparticles and decrease with addition of polyaniline in the matrix. Moreover, the morphology analysis from FESEM shows that Fe_3O_4 nanoparticles are spherical in nature while PANi nanotubes containing modified Fe_3O_4 nanoparticles show the morphology of nanofibers. The composites were further characterized by FTIR, XRD and UV-VIS. The obtained XRD results indicate the presence of PANi containing Fe_3O_4 nanoparticles were further supported by FTIR and UV-VIS analysis.

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