



## Research paper

# Conductive thin films based on poly (aniline-co-*o*-anthranilic acid)/magnetite nanocomposite for photovoltaic applications



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## ABSTRACT

In this research, magnetite particles in a nanoscale were coated with three various weight percentages of poly (aniline co-*o*-anthranilic acid) copolymer. Aniline and *o*-anthranilic acid monomers were copolymerized in the presence of Fe<sub>3</sub>O<sub>4</sub> nanoparticles via surface initiated polymerization method. Characterization of nanocomposites was carried out by several analysis techniques including; FT-IR, FE-SEM, TEM, XRD, TGA and UV–vis-Near infrared. FT-IR results showed that pended COOH groups of PANAA are physically interacted to magnetite nanoparticles. Magnetite nanoparticles enhanced the thermal strength of the nanocomposites in the range of 300–800 °C. The XRD analysis revealed that the average crystallite size ranges from 7.59 to 11.14 nm depending on PANAA weight percentage. Thin films of uniform spherical shape of PANAA/magnetite nanocomposite were fabricated by thermal evaporation technique. The values of E<sub>g(direct)</sub> and E<sub>g(indirect)</sub> decreased with increasing the thickness of PANAA, while the conductivity increased with increasing of PANAA thickness coated magnetite particles. The conductivity value ranged from 3.73 to 26.98 Ω<sup>-1</sup> cm<sup>-1</sup> at a photon energy 3.65 eV. Using the nanocomposites in photovoltaic applications was highlighted.

## 1. Introduction

Optically transparent and electrically conductive polymers have been attractive in the past two decades due to their benefits in various applications including solar cells, electrostatic dissipation, electromagnetic shielding, sensors, touch-sensitive screens, and alarm devices [1,2]. Furthermore, conducting polymers/magnetic nanocomposite have attracted great attention of the scientific community currently due to its high potential of technological industrial applications in various areas, such as electromagnetic interference shielding [3], rechargeable batteries [4], corrosion protection coatings [5], electrodes [6], gas separating membranes [7,8], microwave absorption [9], sensors [10–12], and microelectronic devices [13,14]. PANI as any conjugated polymer has some characteristics related to its molecular structure and bond alternation make the polymer infusible, insoluble in organic solvents and cannot be handled like thermoplastic polymers [15,16]. The problem of difficult solubility in organic solvent has been solved by using various functionalized dopants [15–18]. Other researchers enhanced the PANI solubility by introducing another *o*-anthranilic acid monomer through PNAI backbones [19–21]. Unique magnetic and electrical

properties can be achieved by preparing PANI/Fe<sub>3</sub>O<sub>4</sub> nanocomposites where polyaniline nanocomposites can be reinforced with different weights of Fe<sub>3</sub>O<sub>4</sub> nanoparticle. Wet chemical method and surface initiated polymerization (SIP) method have been used for this purpose. The resulting magnetic polyaniline nanocomposites have a higher saturation magnetization of 72 emu/g and a conductivity around 10–4 S/cm. PANI and its nanocomposites display a negative permittivity [22–25]. Poly(aniline co-*o*-anthranilic acid)/Fe<sub>3</sub>O<sub>4</sub> nanocomposites have prepared by applying surface initiated polymerization (SIP) method for wastewater applications [26]. In the current study the aim is to cover magnetite nanoparticles with different amounts of PANAA using surface initiated polymerization method (SIP) and to prepare PANAA/magnetite nanocomposite thin films. The nanocomposite consists of organic (PANAA) and inorganic (magnetite) parts, the presence of magnetite in the composite will improve the dielectric constant and improve the efficiency if the material is used in photovoltaic cells. The study aims also to investigate the applicability of the thin films in photovoltaic applications. Photovoltaic material must fulfil two conditions, namely: (i) absorb incident photons through the promotion of electrons to higher energy levels, and (ii) contain an internal electric

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**Nomenclature**

A	Absorbance
$A_o$	Energy-independent constant
a, b, c	Lattice constants
B	Full width at half maximum intensity
C	Speed of light
D	Magnetic particle diameter
$D_m$	Magnetic domain size
$D_c$	Crystal size
d	Interplanar spacing
$E_g$	Band gap
$E_I$	Constant
$E_U$	Urbach energy
H	Magnetic field strength
$h_\nu$	Photon energy
$h, k, l$	Miller indices
K	Constant (0.94)
$k_B$	Boltzmann constant
$k_e$	Extinction coefficient

M	Magnetization
$M_s$	Saturation magnetization
m	Constant
$n_r$	Refractive index
R	Reflectance
T	Temperature
$\alpha$	Absorption coefficient
$\alpha_o$	Constant
$\epsilon_o$	Free space dielectric constant
$\epsilon_1$	Real dielectric constant
$\epsilon_2$	Imaginary dielectric constant
$\theta$	Bragg angle
$\lambda$	X-ray wavelength
$\mu$	True magnetic moment
$\rho$	Nanocomposite density
$\sigma$	Optical conductivity
$\sigma_1$	Real part of optical conductivity
$\sigma_2$	Imaginary part of optical conductivity
$\omega$	Angular frequency

field that accelerates the promoted electrons in a particular direction, resulting in an electrical current. To this end the optical and electrical properties of the nanocomposites were studied.

## 2. Experimental

### 2.1. Materials

All chemical substances were used as received without any additional treatment excluding aniline. Double distillations of aniline were carried out before use and it was kept in a dark bottle. Aniline and *o*-anthranilic acids were bought from Aldrich, potassium dichromate (Merck), ammonia solution and hydrochloric acid (Aldrich). Iron (II) chloride tetrahydrate, iron (III) chloride hexahydrate, hydrochloric acid (HCl) and ethanol of analytical grade were purchased from Shanghai chemicals.

### 2.2. Synthesis of magnetite nanoparticles

Typically, 50 ml of 0.2 M  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  were mixed with 25 ml of 0.2 M  $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$  solution under magnetic stirring. Separately, 40 ml (33%v/v) ammonia solution was diluted with 20 ml distilled water. Subsequently, diluted ammonia solution was added to previous iron ( $\text{Fe}^{3+}/\text{Fe}^{2+}$ ) salts drop by drop at room temperature and 800 rpm. Gradually, a black precipitate of magnetite was gradually appeared. The resulting magnetite dispersion was diluted with additional 200 ml of distilled water. The obtained dispersion was left to settle down under gravitational force. Washing process was carried out by decantation method for several times. A magnetite nanoparticle was kept in the mother liquor (total volume 40 ml distilled water) as a dispersion. The magnetite dispersion in the mother liquor was kept for using in the next step of the polymerization process with aniline and *o*-anthranilic acid co-monomers. A separate experiment to estimate the weight of magnetite was conducted following the same aforementioned procedure and the resulting magnetite was separated by filtration and then dried. The weight of the resulting magnetite (blank sample) was found to be 1.4 g.

### 2.3. Synthesis of PANAA/Magnetite nanocomposite

Three different weights of PANAA were polymerized onto the magnetite surface. The first weight of PANAA was obtained as follows: 8 ml conc. HCl was added to 0.93 g aniline under magnetic stirring at

700 rpm at room temperature. Separately, 1.37 g *o*-anthranilic acid was dissolved in 30 ml ethanol then added to aniline solution. 40 ml purified water were added to the co-monomers. The co-monomers' solution was directly mixed to the magnetite dispersion obtained from the previous step. The magnetite/co-monomers mixture was kept in an ice box to maintain the temperature 0–5 °C under magnetic stirring at 700 rpm for 30 min. 60 ml 1 M  $\text{K}_2\text{Cr}_2\text{O}_7$  initiator was added drop wisely to the magnetite/co-monomers mixture for 1 h under the above conditions. During the polymerization process the copolymer was gradually observed to coat the magnetite nanoparticles. The resulting PANAA/magnetite nanocomposite was filtrated using a Buchner funnel and washed with distilled water frequent times, followed by ethanol to remove excess oxidant, unreacted monomers and oligomer. Washing continued until the filtrate (liquid) become almost colorless and then the sample was oven dried at 80 °C for 24 h. The resulting composite was labeled by  $L_1$ . The second weight of PANAA prepared to coat magnetite nanoparticles was obtained according to the same procedure of the first weight except using twofold amount of the aniline and *o*-anthranilic acid in comparing with the first weight (1.86 g aniline and 2.74 g *o*-anthranilic acid) and subsequently the twofold amounts of HCl and initiator used as well. The resulting PANAA/magnetite composite was labeled by  $L_2$ . The third weight of PANAA was also obtained according to the same previous procedure of the first weight except using twofold amounts of the aniline and *o*-anthranilic acid as for the second weight to be (3.72 g aniline and 5.48 g *o*-anthranilic acid) and of course, twofold of amounts HCl and an initiator was used as well. The resulting nanocomposite was labeled by  $L_3$ . The typical composition of the three nanocomposites are listed in Table 1.

### 2.4. Preparation of PANAA/magnetite nanocomposite thin films

Thin films of PANAA/magnetite nanocomposites were fabricated by thermal evaporation method using a high vacuum coating unit

**Table 1**  
Composition of three different PANAA/magnetite nanocomposites.

Co-polymer	Ratio (aniline and <i>o</i> -anthranilic acid)	Weight of the resulting polymer composite (gram)	% weight of $\text{Fe}_3\text{O}_4$
PANAA ( $L_1$ )	(0.93 g and 1.37 g)	2.43	36.55%
PANAA ( $L_2$ )	(1.86 g and 2.74 g)	3.12	30.97%
PANAA ( $L_3$ )	(3.72 g and 5.48 g)	4.76	22.73%

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