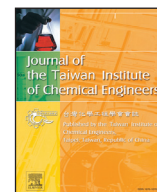




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Fabrication of novel magnetic zinc oxide cellulose acetate hybrid nano-fiber to be utilized for phenol decontamination

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ABSTRACT

An innovative magnetic nano-fiber composite was prepared via hybridization nano-magnetite zinc oxide and cellulose acetate (CA) using electrospinning technique. The affinity of synthesized composite nano-fiber for phenol adsorption from aqueous solution was monitored. Different parameters that effected on the fabrication of pure CA nano-fiber were optimized. The most proper fabricated CA nano-fiber was achieved using 10% CA that was dissolved at acetone and dimethylacetamide (DMAc) solution with mixing ratio 2:1 the electrospinning applied voltage was 20KV and the appropriate separation distance was 25 cm using 0.5 ml/h flow rate. Different weights of magnetic zinc oxide were immobilized onto the most proper fabricated CA nano-fiber (10% CA) to fabricate novel ZnO magnetic CA composite nanofiber. The various fabricated CA based nano-fibers either after and before magnetic ZnO immobilization were characterized using SEM, XRD and FTIR. The phenol sorption affinities of the various fabricated CA nano-fibers were tested using synthetic polluted wastewater in batch manner. It was recorded that magnetite ZnO-CA hybrid nanofiber with 0.1% magnetic zinc oxide immobilization represents the most efficient fabricated nanofiber compared with other studied fabricated CA based nano-fiber. This novel composite nano-fiber has ability to adsorb 64% of phenol within 2 h. The various parameters affecting the phenol sorption process onto this novel fabricated composite were optimized. It was evident that the increment at both the solution pH and temperature has negative impact on the phenol sorption process onto the novel fabricated magnetic ZnO-CA composite nano-fiber.

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1. Introduction

Biopolymers are type of polymers that are biodegradable and they do not produce any environment waste [1]. Cellulose acetate (CA) is organic esters derived from cellulose, known as a novel bio-material and it has wide industrial applications in its fibers form

[2]. Moreover, CA fiber characterized as good thermal stability, cost effectiveness, relatively easy manufacture, and renewable resource of raw material, non-toxicity and chemical resistance [3].

Oxide based materials such as Fe₃O₄, ZnO and their composites have a lot of interest where they are characterized by their high adsorption capacity, nontoxicity, cost-effective and environmentally friendly nanomaterials and have a potential application to the detoxification of groundwater, surface water, drinking water, and industrial effluents. Additionally, a lot of oxide-based nanocomposite/hybrid materials have been developed for water purification. As an attempt to increase the adsorption of contaminants on nano-fiber materials there is a new trend for fabrication hybrid materials. Furthermore, organic-inorganic hybrids are relatively new materials that combine inorganic particles governing the properties of

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hardness, brittleness and transparency, whereas density, free volume and thermal stability depend on the organic host polymer [4]. So far, the effect of ZnO NPs on crystallization properties of cellulose nanocrystal has been studied [5,6]. Cellulose nanocrystal-ZnO nanohybrids were successfully prepared previously using one-pot green synthesis, and they showed higher antibacterial activity and better thermal stability [6]. In addition another research was focus in fabrication cellulose nanocrystal-ZnO nanohybrids through one-pot route process and investigate the effect of incorporation cellulose nanocrystal-ZnO nanohybrids on the properties of poly(3-hydroxybutyrate-co-3-hydroxy-valerate) (PHBV) nanofibrous membranes as wound dressings without any dispersion agent [7]. However to date, no other studies have yet been published showing the influence of incorporation magnetite ZnO nanocomposite onto the properties of produced cellulose acetate-magnetite ZnO nanofibrous membrane and its performance for phenol decontamination from polluted water.

Phenols represent one from the top 45th priority hazardous substances that required being treated before its release into the environment that referring to Agency for Toxic Substances & Disease Registry, USA classification. Even at low concentration, phenols have a murder and destroying effect on the life form such as diarrhea, sour mouth, impaired vision, and excretion of dark urine [8].

This work is concerning with optimizing the fabrication of CA nanofiber-magnetic ZnO hybrid material and its usage in removal of phenol form polluted water.

2. Materials and methods

2.1. Materials

Cellulose acetate (CA), (MW 30000, India), acetic acid (99.8%, Alfa Aser), acetone (Sigma-Aldrich) N,N-dimethylformide (DMF) (Across), N,N-dimethylacetamide (DMAc) (Sigma-Aldrich), Zinc acetate (Sigma-Aldrich) and sodium hydroxide (Sigma-Aldrich). All chemicals were used without further purification.

2.2. Preparation of cellulose acetate nanofiber

The electrospun nano-fibers were fabricated through dissolving 10% (w/v) CA at various solvent mixtures with the following composition; (a) acetic acid and distilled water at different mixing ratio (100%, 75/25%, 50/50% and 25/75% (v/v)), (b) acetone and N,N-dimethylformamide (DMF) with ratios of 2:1 and 1:1 or (c) acetone and N,N-dimethylacetamide (DMAc) with ratios of 2:1 and 1:1 at 40 °C under vigorous stirring until a homogeneous colorless solutions were obtained. Subsequently, 3 ml of the pervious polymer solution was immersed in a 5 ml syringe and electrospun at the following conditions; (a) the separation distance between the tip-collector was fixed at 25 cm, (b) the pump flow rate was controlled at 0.5 ml/h and (c) the voltage was applied at values of 15, 20, 25, and 30 kV.

2.3. Preparation of magnetite zinc oxide

Magnetite zinc oxide was synthesized through immobilizing zinc oxide nanopowder with magnetite via co precipitation technique. Briefly, 0.5 g zinc oxide nano-powder was suspended in 50 ml mixed solution of iron (III) chloride and iron (II) chloride with molar ratio 2:1 until obtain homogeneous suspension. Two molar NaOH solutions was added drop wise to the previous suspension at 40 °C and maintained for 30 min under constant stirring until yield black precipitate. The precipitate separated by centrifuge, washed several time by distilled water and dried at 70 °C.

2.4. Fabrication of magnetite zinc oxide cellulose acetate composite nanofiber

A certain weight of nano-magnetite zinc oxide immersed in a cellulose acetate solution (acetone and N, N-dimethylacetamide (DMAc) with a ratio of 2:1) and kept stirring overnight at 40 °C until a homogeneous solution was obtained. The homogeneous magnetic-polymeric suspension was fed through the electrospinning at the above conditions.

2.5. Assessment of magnetite zinc oxide cellulose acetate composite nanofiber for phenol sorption

The affinity of fabricated nano-magnetite zinc oxide cellulose acetate nano-fiber toward phenol sorption was tested using phenol synthetic polluted water in batch manner. Firstly, the phenol adsorption behavior of the three different fabricated composite nano-fibers at different immobilized magnetic ZnO ratios (0.15%, 0.03%, 0.3%) was compared with that of free cellulose acetate nanofiber to select the most proper fabricated nano-fiber that attain the highest phenol sorption capacity. A set of stoppered conical flasks (50 ml) were filled with 40 ml solution with specific phenol concentration and mixed with fixed weight from the fabricated nano-fiber under shaking at 300 rpm for specific time. Various parameters that affected on the phenol removal process onto the most efficient fabricated nano-fiber such as contact time, material dosage, phenol concentration, solution pH, and temperature were examined. The concentration of phenol after treatment process was monitored colorimetry using UV spectroscopy through complex formation with phenol after mixed with amino antipyrine and potassium ferrous cyanide. After definite time interval, 1 ml of the treated solution was withdrawn then mixed with 2 ml from 2 ppm solution from 4-aimno-antipyrine and 2 ml of 0.3 ppm potassium ferricyanide. In order to adjust the solution pH, it was buffered with 6 ml of buffer solution (pH 10) in presence of 2 ml ammonia solution. The absorbance of the resulting solution mixture was measured using spectrophotometer at 510 nm. The percentage removal of phenol using tested nano-fibers was estimated from the following equation:

$$\% \text{Removal} = ((C_0 - C)/C_0) * 100,$$

where C_0 is the initial phenol concentration in solution (mg/l), and C is the final phenol concentration in aqueous solution after phase separation (mg/l).

2.6. Materials characterization

The different physico-chemical properties of the various fabricated cellulose acetate nano-fiber were examined using various techniques. The morphological structures of the various fabricated cellulose acetate based nano-fiber either before or after magnetic ZnO nano-powder immobilization were compared using Scanning Electron Microscope SEM (Jeol JSM-6360, Japan). The FTIR spectra of the fabricated cellulose acetate nanofiber and its magnetic ZnO composite were compared to elucidate the difference in chemical composition between the two nano-fibers using FTIR-8400 Shimadzu- Japan. Finally, the crystalline structures of pure cellulose acetate nano-fiber and the ZnO composite one were investigated using X-ray diffractometry (Schimadzu-7000, Japan).

2.7. Reusability of magnetite ZnO cellulose acetate nano-fiber

Regeneration of adsorbent and recovery of adsorbate are important aspects to minimize the waste and reuse of adsorbent material. In this concern, reusability of magnetite ZnO cellulose

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