Chemosphere 159 (2016) 293–299

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Photocatalytic discoloration of Acid Red 14 aqueous solution using titania nanoparticles immobilized on graphene oxide fabricated plate

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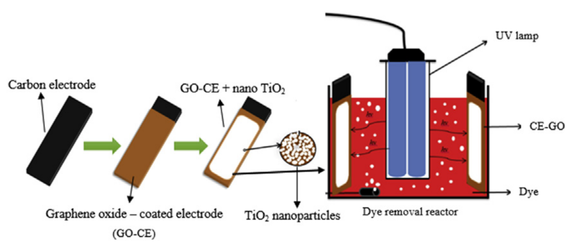
HIGHLIGHT

- Photocatalytic degradation of anionic dye effluent using immobilized TiO_2 nanoparticle.
- Immobilization of TiO_2 nanoparticle on graphene oxide (GO) fabricated carbon electrodes.
- Fabrication of GO nanoparticles on carbon electrode using electro-chemical deposition.
- Investigation of effect of pH, dye concentration and TiO_2 dosage on degradation efficiency.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 12 January 2016
Received in revised form
18 April 2016
Accepted 5 June 2016
Available online 13 June 2016

Handling Editor: Shane Snyder

Keywords:
Immobilized photocatalyst
Graphene oxide fabricated electrode
Decolorization
Nanoparticle
Anionic dye

ABSTRACT

Textile industry consumes remarkable amounts of water during various operations. A significant portion of the water discharge to environment is in the form of colored contaminant. The present research reports the photocatalytic degradation of anionic dye effluent using immobilized TiO_2 nanoparticle on graphene oxide (GO) fabricated carbon electrodes. Acid Red 14 (AR 14) was used as model compound. Graphene oxide nanosheets were synthesized from graphite powder using modified Hummer's method. The nanosheets were characterized with field emission scanning electron microscope (FESEM) images, X-ray diffraction (XRD) and FTIR spectrum. The GO nanoparticles were deposited on carbon electrode (GO-CE) by electrochemical deposition (ECD) method and used as catalyst bed. TiO_2 nanoparticles were fixed on the bed (GO-CE-TiO_2) with thermal process. Photocatalytic processes were carried out using a 500 ml solution containing dye in batch mode. Each photocatalytic treatment were carried out for 120 min. Effect of dye concentration (mg/L), pH of solution, time (min) and TiO_2 content (g/L) on the photocatalytic decolorization was investigated.

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1. Introduction

Water resources can always be contaminated through various pollutants including dyes, toxic metal ions, oil and so forth by different industries such as textile, food, cosmetics, plastics, paper, printing, etc (Sharma and Das, 2012; Thiam et al., 2015). Among the industries, textile has an important role in the consumption and pollution of water during dying, finishing, scouring and other processes. Color-containing wastewater originated from textile industries is one of the most important sources of pollution that causes dangerous effects on environment and human beings health (Radha et al., 2009; Nguyen and Juang, 2013). In addition, district regulations have been established by many governments in order to limit the releasing of organic compound such as colored effluents into environment (Caliman et al., 2007). Thus, for human and

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http://dx.doi.org/10.1016/j.chemosphere.2016.06.020
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environmental protection, proper treatment of wastewater prior to discharge is a vital duty.

The World Bank reported that nearly 17–20% of industrial water pollution is produced by textile dyeing and treatment. Furthermore, it has been reported that more than 100,000 kinds of textile dyes are existed and more than 7×10^5 tons per year are produced (Rondon et al., 2015). Approximately 10–15% of these dyes are wasted in dyeing process, which are low biodegradable with high chemical stability. These dyes may have many hazardous effects including: high toxicity, color depth, esthetic pollution, complex composition, highly carcinogenic and mutagenic effects on human beings (He et al., 2004). In addition, they can considerably affect the aquatic life by decreasing the penetration of sunlight, eutrophication phenomenon, and increase in suspended solids (Sauer et al., 2002). Therefore, restrict regulations have been established by the international environmental standards (ISO 14001, October 1996) in order to treat the colored wastewater effectively. Consequently, a proper dye removal before discharge into water streams seems necessary.

There are various conventional techniques such as trickling filters, active sludge, chemical coagulation, adsorption, ion exchange, biological methods and so forth which have been studied extensively for the removal of textile dyes and many other organic compounds (Sauer et al., 2002; Srinivasan and Viraraghavan, 2010). However these methods may not be suitable for effective wastewater treatment because of some disadvantages like being nondestructive, high sludge formation and in some cases high costs (Srinivasan and Viraraghavan, 2010; Bansal and Sud, 2011).

However, the recent progresses in wastewater treatment suggest an improved chemical process based on the oxidation of organic compounds named advanced oxidation processes (AOPs) (Chan et al., 2011). These methods are based on the production of free radicals (such as $OH^{\bullet}$ and $O_2^{\bullet -}$) by UV irradiation to photocatalyst. These free hydroxyl radicals are powerful oxidizing agents (with an oxidation potential of 2.80 V) that can degrade the organic structures (Vahdat et al., 2012). Among various photocatalysts TiO_2-based materials have been the most promising choice for photocatalytic mineralization (Jafari et al., 2012). Many researches were carried out to increase the photocatalytic oxidation performance of TiO_2 by making TiO_2 in different forms, e.g. nanoparticles, nanotubes, films and single crystals. These materials can be impressive in wastewater treatment, air purification, antibacterial treatment, detoxification, antifogging treatment, and self-cleaning purposes. Specially, the composite of TiO_2 and carbon are now being considered as a promising photocatalysts in the treatment of air and water (Ajmal et al., 2014). They can be generally classified into three kinds: TiO_2-mounted activated carbon (Tryba et al., 2003), carbon-doped TiO_2 (Ren et al., 2007), and carbon-coated TiO_2 (Inagaki et al., 2006), which give acceptable photocatalytic activities.

In photocatalytic activities, semiconductor receives energy greater than its band gap and an electron from the valance band (VB) moves to the conduction band (CB). Then the electron-hole pair can transfers to photocatalyst surface and take part in red-ox reactions with generating free radicals (Houas et al., 2001; Fotiou et al., 2013). If the reactions carry on entirely, total degradation happens and finally organic contaminants convert to HO_2 and CO_2 (Tayade et al., 2009).

If photocatalytic processes carry out in the form of photocatalyst powder suspended in aqueous solutions, we need an extra operation to separate the nanoparticles. This makes adverse effects such as increase in running cost, time and energy. Furthermore, this additional step may cause secondary contamination in solution because of incomplete recycling. However, there are methods that can eliminate the issues by photocatalyst immobilization on proper bed through effective operations such as electrophoretic deposition, thermal treatment (Lei et al., 2012), chemical vapor deposition (CVD) (Byun et al., 2000), sol-gel method (Gelover et al., 2004), electro spun nanofibers (Yousef et al., 2012) etc.

In recent years, graphene oxide(GO) has been widely attracted remarkable attention, due to its promising merits like having various oxygen functionalities on basal planes with this assumption that the oxygen containing functional groups such as hydroxyl, carboxyl and epoxy groups are uniformly distributed on the GO surface (Madadrang et al., 2012). Followed by excellent and tunable hydrophilicity and uniformity of dispersion in water. It can used as an absorbent for pollutants such as dyes and heavy metals (Haldorai et al., 2014). Other carbon-based materials are graphene, nanotube (CNT), activated carbon etc. immobilized carbon-based absorbents demonstrate wide application in many fields especially in wastewater treatment (Vadivel et al., 2014; Yu et al., 2016). Electrochemical deposition (ECD) of CNTs on graphite electrode and GO on stainless steel electrode has been reported by Pajootan et al. (Pajootan et al., 2014) and Sung Jin et al.(An et al., 2010), respectively.

In this present work, we present a novel and simple method in order to photocatalytic degradation of dye on immobilized graphene oxide (GO) substrate. Our novel composite substrate contains GO which is fixed on carbon electrode by using electrochemical process and TiO_2 nanoparticles which anchored on GO sheets by thermal process. The model dye chosen was Acid Red 14 which has a complex structure. We examined the photocatalytic effect of this immobilized composite on dye degradation value.

2. Materials and methods

2.1. Chemicals and materials

TiO2 nanoparticle (P-25) was purchased from (Degussa, Germany) and used as the photocatalyst. Graphite powder was purchased from Seraj Co. Sulfuric acid 96%, phosphoric acid 96%, HCl 30% and ethanol were obtained from Merck Co. Potassium permanganate ($KMnO_4$), Hydrogen peroxide (H_2O_2) and Cetyl Trimethyl Ammonium Bromide ($(C_{16}H_{33})N(CH_3)_3Br$, CTAB) were provided by Loba Chieme. Distilled water was used throughout the whole experiment. The azo dye, Acid Red 14 (AR 14) was purchased from Ciba Co. All reagents were analytical grade and they were used without further purification. The molecular structure of AR 14 is shown in Fig. 1.

2.2. Synthesis of GO

Graphene oxide (GO) was synthesized by the modified Hummer's method using an oxidizing process from natural graphite. Graphite powder (3 g) was dispersed in to a mixture of 360 mL of concentrated Sulfuric acid, 40 mL of phosphoric acids and 18 g of potassium permanganate. The mixture was heated to 50 °*C* and

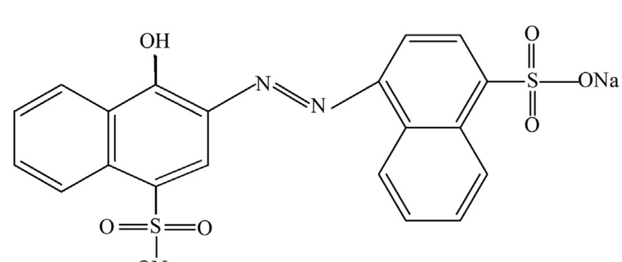

Fig. 1. Molecular structure of Acid Red 14 (AR 14).

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