Journal of Environmental Management 181 (2016) 146–156

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Research article

Synthesis and application of green mixed-metal oxide nano-composite materials from solid waste for dye degradation

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ARTICLE INFO

Article history:
Received 17 November 2015
Received in revised form
26 May 2016
Accepted 5 June 2016

Keywords:
Electrochemical sludge
NCMs
Fe_2O_3
Catalytic activity
Zeta potential

ABSTRACT

Present study demonstrates reutilization of electrochemical (EC) sludge as a potential low-cost green catalyst for dye degradation. Hexagonal Fe_2O_3 type phase with trevorite ($NiFe_2O_4$)-type cubic phase nanocomposite material (NCM) was synthesized from solid waste sludge generated during EC treatment of textile industry wastewater with stainless steel electrode. For NCM synthesis, sludge was heated at different temperatures under controlled condition. Various synthesized NCMs were characterized by powder X-ray diffraction (PXD), energy dispersive X-ray (EDX) spectroscopy and X-ray photoelectron spectroscopy (XPS) analysis. The synthesized NCMs were found to contain iron, chromium, nickel and oxygen in the form of α-Fe_2O_3 (metal: oxygen = 40:60), $(Fe,Cr,Ni)_2O_3$ and trevorite $NiFe_2O_4$, $(Ni,Fe,Cr)(Fe,Cr,Ni)_2O_4$ (metal: oxygen = 43:57). Field emission scanning electron microscopy (FE-SEM), transmission electron microscopy (TEM), pore size distribution, and atomic force microscope (AFM) analysis showed distribution of grains of different shapes and sizes. Catalytic activity of NCM was studied by the methylene red dye degradation by using the catalytic wet peroxidation process. Zeta potential study was performed under different pH so as to determine the performance of the NCMs during dye degradation.

1. Introduction

Dyes and colors released in significant quantities in the effluents from textile industries cause a lot of environmental pollution. Different technologies have been employed for the treatment of these effluents (Bhatnagar et al., 2014a, 2014b; Singh et al., 2013a). Some of these methods require chemicals for treatment and generate solid waste. Some advance oxidation methods also require catalysts for treatment (Zinatloo-Ajabshir and Salavati-Niasari, 2015; Das and Srivastava, 2015a). Use of nano-materials (NMs), such as nano-sized transition metal oxides as catalysts has increased recently in industrial wastewater treatment largely due to their higher activity (Gao et al., 2015; Gu et al., 2012; Zinatloo-Ajabshir et al., 2015). Nano-sized metal oxide based catalysts need to be prepared by simple, versatile and environmentally friendly methods so as to get desirable properties in terms of texture, chemical composition, etc. in these catalysts. Several methods have been reported in the literature for the preparation of nanocomposite material (NCM) (Priyanka, 2013; Das and Srivastava, 2015b; Zinatloo-Ajabshir et al., 2016; Mortazavi-Derazkola et al., 2015; Singh et al., 2014a).

Some investigators have also worked on preparation of useful materials such as composites, catalysts, etc. from different solid wastes (Fan et al., 2008; Kargbo, 2010; Matos et al., 2011; Esparza et al., 2011; Zhuo et al., 2012). Sewage sludge-based composite materials have been used as an adsorbent for adsorbing NO_2 (Pietrzak and Bandosz, 2008). Municipal sludge has also been used for biodiesel production (Matos et al., 2011). Iron oxide NCMs possessing meso- and micro-pores have received much attention in wastewater treatment (Eriksson et al., 2005). Iron oxide NCMs with variable surface area and pore volume facilitate the adsorption and transport of various species in the reacting system. Moreover, they contain a large number of active sites and therefore, provide superior adaptability and sustainability for various applications in the field of energy and environment, semiconductors, medicinal use,

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http://dx.doi.org/10.1016/j.jenvman.2016.06.007
0301-4797/

and inorganic and organic contaminant degradation (Cao et al., 2007; Oha and Park, 2011; Xu and Wang, 2011). Various costlier methods have been used in the literature which require toxic and hazardous chemicals, and sophisticated instruments for the preparation of different iron oxide NCMs. However, to the best of our knowledge none of the methods have been used any type of waste material for the preparation of iron NCMs. Electrochemical (EC) method is one of the recently developed simple, cost effective and reliable methods with short treatment time for industrial wastes. Theory of EC methodology is described in the supporting information (SI).

In our previous studies (Singh et al., 2013c, 2013b, 2014b), detailed mechanism including changes in the zeta potential and schemes of dye degradation during EC treatment of dye bath effluent is reported. Despite small amount of sludge generation in the EC method as compared to conventional coagulation and biological methods (Zhuo et al., 2012; Singh et al., 2013c, 2013b, 2014b), still the sludge containing the electrode material needs to be disposed off.

In the present study, for the first time to the best of the authors' knowledge, preparation of iron oxide NCMs was carried out by heating the EC solid waste at different temperatures. Calcined samples obtained at different temperatures were further characterized using various methods to evaluate their composite nature in terms of structural and textural properties. The NCMs were further used as catalysts for dye degradation in order to explore their potential use in dye degradation.

2. Experimental section

2.1. Materials

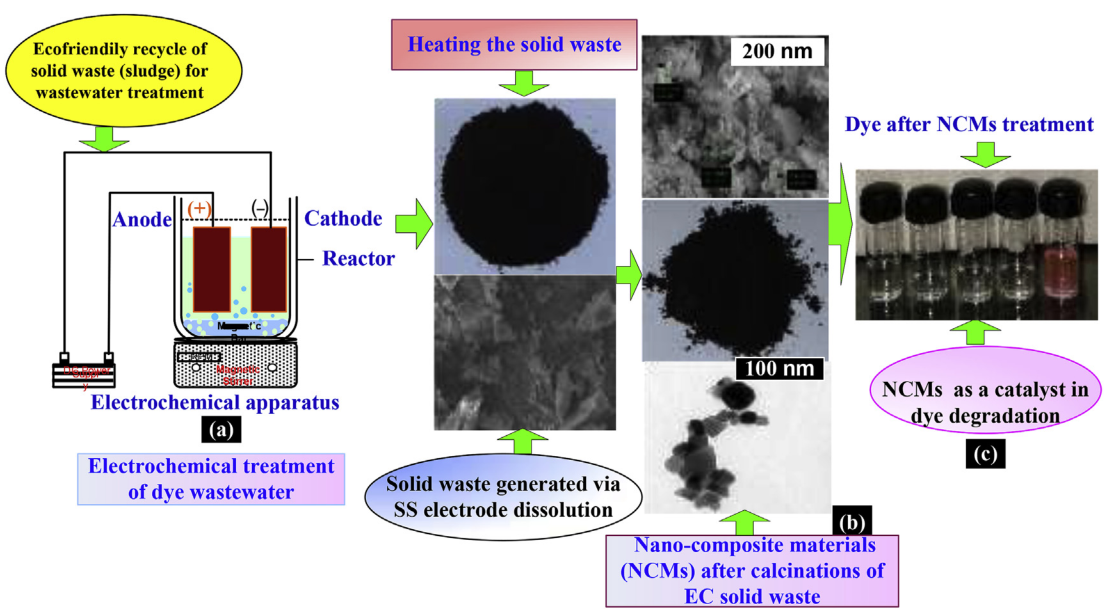
All the chemicals used in present study were of analytical grade. Methylene red (MR) was purchased from Yogesh Dyestuff Product Pvt Ltd., India. Detailed characteristic of dyes are reported in Table S1 (in supporting information). 30 wt% H_2O_2 was purchased from Ranken, India. Solid waste sludge was obtained from EC treatment of dye. Details are given in previous publication (Mondal et al., 2013; Singh et al., 2013c, 2013b, 2014b).

2.2. Synthesis of NCMs

The iron oxide based NCMs were prepared from the EC solid waste. Sludge sample was spread over glass plate and sun dried for 2–3 days time (in which its' mass became constant) (Matos et al., 2011). Thereafter, 5 g of the solid waste was taken in a silica crucible and calcined in a muffle furnace at different temperatures for 3 h. Scheme 1 shows the graphical representation of the preparation of iron oxide NCMs from EC solid waste. Experimental procedure for EC treatment and generation of sludge is given in supporting information (SI). EC sludge samples were heated starting from 100 °C and gradually increasing the temperature in steps of 100–300 °C up to 1000 °C. Finally, the samples were furnace cooled and coded as follows according to the calcination temperature: original sample (S(Fe-0), sludge calcined at 400 °C (SFe-4), 600 °C (SFe-6), 700 °C (SFe-7), 800 °C (S(Fe-8) and 1000 °C (SFe-10).

2.3. Characterization of NCMs

Perkin Elemer Pyris Diamond thermo-gravimetric (TG) analyzer was used for thermogravimetric and differential thermal analysis (TGA and DTA) in air atmosphere at the heating rate of 10 °C/min with air flow rate of 20 ml min^{-1} in the temperature range 20 °C–1000 °C. Powder X-ray diffraction (PXD) patterns were recorded on a Bruker AXS D8 advance powder X-ray diffractometer operating at 40 kV and 30 mA using Cu Kα radiation (λ = 1.5418 Å) in the angular range 10–90°. The data were recorded at a scan rate of 0.02°/step and 0.5 s per step. The observed PXD patterns were analysed by comparing with the powder diffraction files available in the joint committee on powder diffraction standards (JCPDS) database. Field emission scanning electron microscope (FE-SEM: QUANTA 200-FEG) was used to examine the surface topography. The elemental analysis of the prepared iron oxide NCMs was performed using energy-dispersive X-ray analysis (EDX) accessory by 5–10% error of element contents. FEI-TECNAI 300 kV digital transmission electron microscope (TEM) having resolution of 0.19 nm used for electron micrographs and selected area electron diffraction. Atomic force microscope (AFM: M/s Molecular Tools and Devices)



Scheme 1. Schematic representation of iron oxide nanocomposite materials (NCMs) from electrochemical solid waste sludge.