



## Research article

# Activated electric arc furnace slag as an effective and reusable Fenton-like catalyst for the photodegradation of methylene blue and acid blue 29

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## ABSTRACT

In this work, an activated electric arc furnace slag (A-EAFS) was investigated as an effective Fenton catalyst for the photodegradation of methylene blue (MB) and acid blue 29 (AB29). Fourier transform infrared spectroscopy and UV–visible absorption analyses indicated that A-EAFS offers additional Fe<sub>3</sub>O<sub>4</sub> because of the changes in the iron oxide phase and the favorable response to visible light. It has been found that the highest degradation efficiency can reach up to 94% for MB under optimal conditions of 1 g L<sup>-1</sup> of A-EAFS, 20 mM H<sub>2</sub>O<sub>2</sub>, and pH 3. The optimal conditions for AB29 were 0.1 g L<sup>-1</sup> A-EAFS, 4 mM H<sub>2</sub>O<sub>2</sub>, and pH 3 to reach 98% degradation efficiency. Visible light enhanced the degradation of both dyes. In addition, A-EAFS, could be easily separated magnetically, exhibited good chemical stability after seven successive photodegradation cycles.

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## 1. Introduction

The dyestuff are allegedly responsible for the release of large quantities of highly colored pollutant and textile industry corresponding to more than 15% of the total world production of dyes (Módenes et al., 2012; Fayazi et al., 2016). Among organic dyes produced, anionic and cationic dyes have prevail at about 50%–70% of the total market in global production (Konstantinou and Albanis, 2004). Industrial dyes effluent have consider toxic and carcinogenic compound. In fact, recalcitrant dyes effluent contains high value of TOC (2900 mg L<sup>-1</sup>), BOD (>80 mg L<sup>-1</sup>) and COD (>150 mg L<sup>-1</sup>) has led to reduce aesthetic value in water resources (Buthiyappan et al., 2016; Holkar et al., 2016). Therefore, these dominance dyes has contributed to the environmental challenge (Guimarães et al., 2012). Among the treatment methods used (Türgay et al., 2011; Yu et al., 2010; Shan et al., 2015), the advanced oxidation process has been proven efficient and economical (Chong et al., 2016; Buthiyappan et al., 2016).

Iron oxides are widely used as heterogeneous catalysts because of their good oxidation–reduction potential and the generation of

highly active radicals ( $\cdot\text{OH}$ ) after the addition of an oxidizing agent (e.g., H<sub>2</sub>O<sub>2</sub>) (Pereira et al., 2012; Rahim Pourn et al., 2014). All these materials are considered environmentally friendly and high performing, but the low separation efficiency and high cost limit their widespread usage.

Rich mineral resources, such as clay (Hassan and Hameed, 2011), electric arc furnace dust (Wu et al., 2014), blast furnace slag (Zhang and Chai, 2014), residue mud (Jones et al., 2012; Liu and Naidu, 2014), and other low-cost raw material, have attracted substantial research attention. Electric arc furnace slag (EAFS) consists of a combination different of metal oxides, the majority of which contain various iron oxides (de Araújo and Schalch, 2014), such as magnetite (Fe<sub>3</sub>O<sub>4</sub>), maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ), hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ), and goethite ( $\alpha\text{-FeOOH}$ ). The unique combination of iron oxide and other transition metals has transformed this material to a potential Fenton-photocatalyst. The use of EAFS as Fenton-like catalyst offers several benefits in overcoming the limitation of homogeneity, good catalytic activity, easy separation, cost effectiveness, and reduced waste disposal issues. Among the minerals found in EAFS by our previous study (Nasuha et al., 2016), Zn and TiO<sub>2</sub> (E<sub>g</sub>: 3.2 eV) were found the most potent even in small quantities (Hossain et al., 2015; Lam et al., 2013) and photoactivity under the UV light region only (Saleh and Djaja, 2014; Sood et al., 2015). However, given the

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energy crisis, narrow-band-gap catalysts are preferred as new responsive visible-light materials. Therefore, combination with other minerals significantly increases the red shift and hence favors performance in the visible-light region. Few studies have optimized  $\text{TiO}_2$  and  $\text{ZnO}$  catalysts with the aid of a photocatalyst combined with a Fenton-like catalyst. Visible-light-mediated photo-Fenton oxidation by employing  $\text{ZnO}$  or  $\text{TiO}_2$  doped with Fe as narrow-band-gap semiconductors have been reported in literature (Li et al., 2016; Palanisamy et al., 2013; Mardani et al., 2015). The synergistic effect between Fenton oxidation and photocatalysis also enhances  $\cdot\text{OH}$  production in the degradation of various pollutant types. Therefore, EAFS, by its own unique chemical properties, is believed to be a good candidate Fenton-type photocatalyst.

A previous work (Nasuha et al., 2016) has shown that magnetically activated EAFS is an excellent Fenton-like catalyst with abundant availability source and good chemical stability. However, improving its catalytic activity remains necessary. Therefore, this work mainly aims to improve the degradation of dyes, analyze the optical properties of activated EAFS (A-EAFS), and compare the material's catalytic activity with visible assistance in the degradation of acid blue 29 (AB29) and methylene blue (MB) dyes.

## 2. Materials and methods

### 2.1. Materials

The following materials were purchased from Merck and used without purification: high-quality (98%) AB29 and MB, hydrogen peroxide (30%), and terephthalic acid (TA; 99%). The main material EAFS was collected from a local steel factory in Penang, Malaysia. Alkaline (0.1 M NaOH) and acid solutions (0.1 M HCl) were prepared and used to regulate pH.

### 2.2. Catalyst preparation and characterization

#### 2.2.1. A-EAFS preparation

A-EAFS was prepared as described in a previous study (Nasuha et al., 2016). Briefly, raw EAFS (R-EAFS) of 40–50  $\mu\text{m}$  size was obtained by grinding and sieving. Then, R-EAFS particles were soaked in 0.1 M NaOH for 4 h and washed several times with distilled water until neutral pH to remove adhering dust and fine materials. The resulting samples were decanted by centrifuging at 500 rpm for 10 min, filtered, and dried at 100  $^\circ\text{C}$  for 24 h. Finally, the prepared samples were calcined at 400  $^\circ\text{C}$  for 2 h before use as A-EAFS.

#### 2.2.2. Characterization of catalysts

The prepared catalysts were characterized by UV–visible (UV–vis) diffuse reflectance spectroscopy (DRS) using a Perkin Elmer Lambda 35 UV–vis spectrometer. The spectra were recorded timely in the range 300–600 nm using  $\text{BaSO}_4$  as the reference standard. Fourier transform infrared (FTIR) measurement was conducted using a Thermo Scientific Nicolet IS10 FTIR spectrometer within the range 400–4000  $\text{cm}^{-1}$ . Photoluminescence (PL) measurement with TA as probe was performed using a Perkin Elmer Lambda S55 spectrofluorometer with an excitation wavelength of 325 nm using a Xe lamp.

#### 2.2.3. Analysis of hydroxyl ( $\cdot\text{OH}$ ) radical

PL technique with TA as probe molecule was conducted to detect the formation of  $\cdot\text{OH}$  radicals on the surface of photo-irradiated A-EAFS. The basic principle is as follows. As TA readily reacts with  $\cdot\text{OH}$  to produce a highly fluorescent product, 2-hydroxyterephthalic acid (HTA) gives a PL signal at 425 nm. The procedure was run similar to the experiment on photocatalytic activity except that the AB29 and MB aqueous solutions were

replaced by the  $5 \times 10^{-4}$  M TA aqueous solution dissolved in  $2 \times 10^{-3}$  M NaOH solution. All generated HTA samples were observed by PL analysis using a Perkin Elmer Lambda S55 spectrofluorometer at 425 nm excited by 315 nm light.

### 2.3. Catalytic activity procedure

The photodegradation of MB and AB29 in a heterogeneous Fenton-like reaction was carried out using a 250 mL conical flask with 200 mL of different initial concentrations (25–150  $\text{mg L}^{-1}$ ) and pH (2–7) (Nasuha et al., 2016). Prior to batch runs, the reaction was initiated by adding a certain amount of  $\text{H}_2\text{O}_2$  (2–12 mM) into the aqueous solution. The adsorption experiments were performed in the dark for 30 min to obtain the equilibrium adsorption of MB and AB29. Withdrawn samples at regular intervals were analyzed using a UV–vis spectrophotometer (UV-1700 Pharmaspec Shimadzu). The MB and AB29 concentrations were examined by measuring the absorbance of the solution at  $\lambda_{\text{max}}$  668 and 602 nm, respectively. After 30 min under dark conditions, the dye solution was irradiated with  $1 \times 8$  W fluorescent lamp visible light for 180 min photodegradation reaction. This lamp was fixed at about 5 cm above the reaction solution.

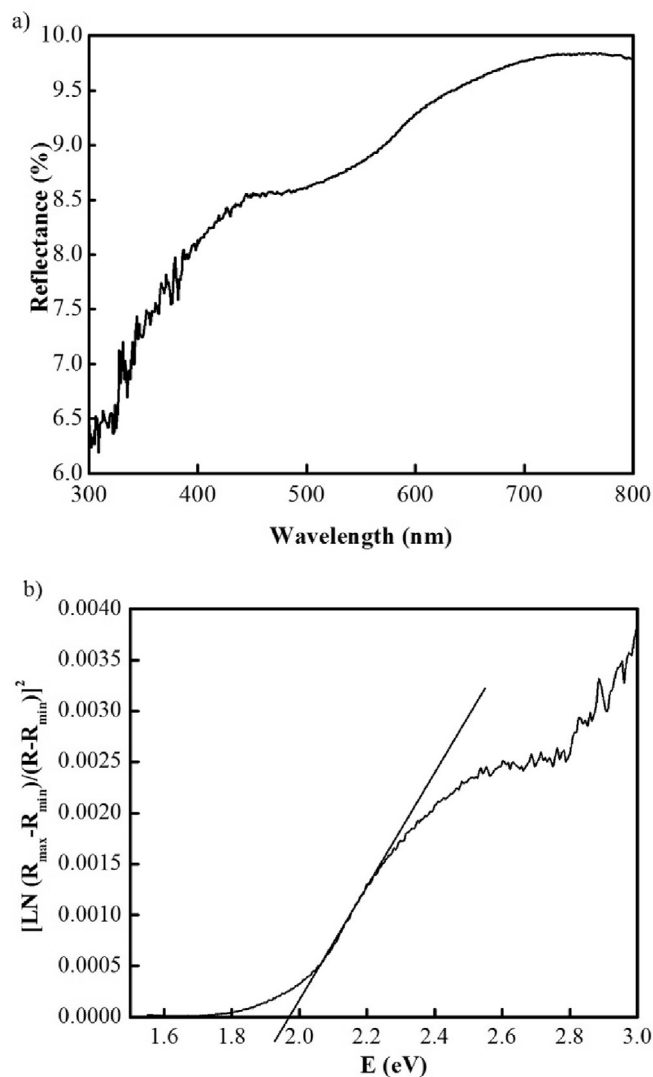


Fig. 1. (a) UV–vis DRS spectra of the A-EAFS. (b) Corresponding  $[\text{LN}(R_{\text{max}} - R_{\text{min}})]^2$  vs.  $E$  plots of A-EAFS.

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