



Composites based on PET and red mud residues as catalyst for organic removal from water



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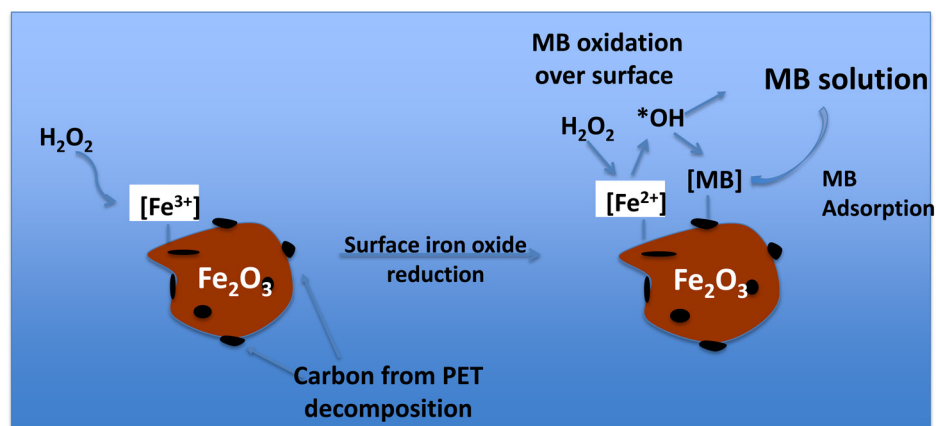
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HIGHLIGHTS

- Composite based on carbon/iron oxide from PET and red mud wastes for organic contaminants oxidation.
- Composites are mainly composed of hematite and a carbon matrix from PET decomposition.
- RM/PET-15 presents the highest methylene blue (MB) removal from water, by combined adsorption and oxidation processes.
- The dye oxidation was confirmed by ESI-MS studies.
- The RM/PET catalysts can be reused for at least four batch runs.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, we obtained a composite based on carbon/iron oxide from red mud and PET (poly(ethylene terephthalate)) wastes by mechanical mixture (10, 15 and 20 wt.% of PET powder/red mud) followed by a controlled thermal treatment at 400 °C under air. XRD analyses revealed that the α - Fe_2O_3 is the main phase formed from red mud. TPR analyses showed that the iron oxide present in the composites undergoes reduction at lower temperature to form Fe^{2+} species present in Fe_3O_4 , indicating that the iron oxide in the composite can exhibit greater reactivity in the catalytic processes compared to the original red mud. In fact, catalytic tests showed that the composites presented higher capacity to remove methylene blue dye (MB), presenting about 90% of removal after 24 h of reaction. The MB removal was also monitored by mass spectrometer with ionization via electrospray (ESI-MS), which demonstrated the occurrence of the oxidation process, showing the formation of MB oxidation products. The stability of the composites was confirmed after four reuse cycles. The results seem to indicate that PET carbon deposited over the iron oxide from red mud promotes adsorption of the contaminant allowing its contact with the iron atoms and their consequent reaction.

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1. Introduction

The polyethylene terephthalate (PET) and aluminum industries both generate a large amount of residue. The aluminum industry

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is recognized to produce an important residue known as red mud, which is a highly alkaline, insoluble residue generated during the bauxite beneficiation to produce alumina (Al_2O_3) in the Bayer process. The red mud is mainly composed of a mixture of oxides and hydroxides such as Fe_2O_3 , TiO_2 , SiO_2 , Al_2O_3 , CaO , Na_2O , $\text{Al}(\text{OH})_3$ and very small amounts of other metal oxides, depending on the geographical origin of the bauxite ore [1–4]. Moreover, the significant growth of PET utilization in soft drink bottles and containers leads to a considerable increase in residue generation [5,6]. Part of the post-consumer PET is collected and recycled. According to Brazilian PET industry association [7], 57.1% of the used PET was recycled in 2011 in Brazil. During recycling, the grinding of the bottles produces PET fragments or “PET flakes” and also generates a large amount of undesired PET powder residue that hinders the processing of the flake to produce PET resin [6]. Therefore, the integration of these two wastes (red mud and PET powder) to produce value added materials such as catalysts and adsorbents are of great industrial interest. The valorization of low-cost materials could contribute to economic development and reduce the environmental impact from these industries. There are several works describing the development and modification of industrial residues in order to remove contaminants from aqueous medium [8–10]. Many papers have shown that untreated, activated and reduced red mud can be used as catalyst for environmental applications, wastewater treatment, and biofuel production [1,3,11–15]. In addition, few works have revealed that activated carbons with high surface area could be produced from PET residues, showing high adsorption capacity of contaminants from water [16–18]. Therefore, the catalytic potential of the oxides present in the red mud and the adsorptive properties of the PET carbon, associated with the high availability of these residues have motivated this study. Here, we investigate the co-processing and transformation of both residues into a composite material that can be used for environmental application. The composites based on PET carbon/red mud can be potentially applied as catalysts during water treatment for organic contaminant oxidation. The composites were produced via mechanical mixture and thermal treatment that constitutes a low cost process and environmental friendly route without use of solvent. The materials were characterized and evaluated as catalysts in the degradation of methylene blue dye in the presence of hydrogen peroxide.

2. Material and methods

2.1. Composites synthesis

The red mud-PET composites (RM-PET) were prepared by the mechanical mixture of the residues in different weight proportions (10, 15 and 20 wt.% of PET). The materials were calcined at 400°C for 1 h in an oven under air atmosphere. The red mud (RM) was also calcined, under the same conditions. It is important to highlight that the residues were used as generated in the industries, without additional treatment or use of solvent.

2.2. Composites characterization

The materials were characterized by powder X-ray diffractometry (XRD) in a Shimadzu spectrometer using $\text{Cu K}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$). The crystalline phases were identified using the Powder Diffraction File (PDF) database (JCPDS, International Centre for Diffraction Data). The thermal stability of the precursors and catalysts was studied by thermogravimetric analysis (TGA, Shimadzu DTG-60AH) under N_2 atmosphere (gas flow of $30 \text{ cm}^3 \text{ min}^{-1}$) with a heating rate of $10^\circ\text{C min}^{-1}$. TPR experiments were performed in a CHEMBET 3000 equipment under 80 mL min^{-1} of a mixture of

H_2 (5%)/ N_2 with heating rate of $10^\circ\text{C min}^{-1}$. The surface area was determined with the BET method using N_2 adsorption/desorption at 77 K in an Autosorb iQ2 Quantachrome 2.0 instrument. The catalyst morphology was studied by scanning electron microscopy and the elemental composition by energy dispersive X-ray spectroscopy (SEM-EDS) on a Philips XL-30 FEG microscope.

2.3. Catalytic oxidation tests

The methylene blue dye (MB) oxidation experiments were performed according to the procedure described in [19]. Briefly, the reaction occurred in batch reactors of 20 mL volumetric capacity, using the oxidant agent H_2O_2 (0.1 mL, 50% v/v) added into 9.9 mL of MB solution (10 mg L^{-1}). The MB removal from water was monitored by UV-vis spectroscopy at 665 nm (EduTec – 752/752 N). In order to identify the intermediate species formed during the oxidation of MB a mass spectrometer with ionization via electrospray was used, (heated capillary temperature of 275°C , dry gas (N_2) at a flow rate of $10 \text{ dm}^3 \text{ min}^{-1}$, spray voltage of 4 kV, capillary voltage of 25 V, and tube lens offset voltage of 25 V).

2.4. Stability tests

The stability of the catalysts was studied through reuse cycles. The reactions were performed in a batch reactor of 30 mL of volumetric capacity under the same conditions described for the oxidation experiments but using a large amount of the reaction mixture. After 24 h reaction, the MB solution was analyzed by UV-vis spectroscopy and the catalyst was recovered by filtration, oven dried and used in a following cycle, keeping the same proportions of catalyst, MB solution and H_2O_2 .

3. Results and discussion

3.1. Characterization of the materials

The BET surface area of the red mud was $7 \text{ m}^2 \text{ g}^{-1}$ and for the PET powder was lower than $2 \text{ m}^2 \text{ g}^{-1}$. The thermal stability of the residues was investigated by TGA analysis under air atmosphere (Fig. 1). For red mud (Fig. 1a), a total weight loss of about 28% occurred and it is possible to observe two defined regions of weight loss. The first at a temperature lower than 100°C , can be attributed to physically adsorbed water and the second, in the range of $250\text{--}300^\circ\text{C}$, can be attributed to the gibbsite decomposition to produce alumina [$2 \text{ Al}(\text{OH})_3 \rightarrow \text{Al}_2\text{O}_3 + 3 \text{ H}_2\text{O}$] [20]. In this last range of temperature, the loss of the chemically adsorbed water or the goethite decomposition to produce hematite [$2 \text{ FeOOH} \rightarrow \text{Fe}_2\text{O}_3 + \text{H}_2\text{O}$] is also possible [21].

In the case of PET powder (Fig. 1b), the TGA profile showed an abrupt weight loss after 350°C . As expected, this sample was totally decomposed at high temperature ($>500^\circ\text{C}$) and the ash content was quite low (about 1.0%), since the PET polymer is essentially composed of organic matter.

For red mud (Fig. 1a), we also observed that at temperatures higher than 400°C , a negligible weight loss occurs ($\approx 1\%$), and this is the initial decomposition temperature of PET (Fig. 1b). Thus, 400°C was selected as the calcination temperature of the RM/PET materials in order to obtain composites containing the oxides present in red mud and a carbonaceous material from PET.

The thermal stability of the calcined RM and RM-PET composites was also investigated by TGA/DTG. The results are presented in Fig. 2.

From TGA (Fig. 2a) it is possible to observe that the weight loss is under 10% for all the composites. This indicates that during the composites calcination at 400°C , a significant weight loss occurred.

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