Preparation of sludge derived magnetic porous carbon and their application in Fenton-like degradation of 1-diazo-2-naphthol-4-sulfonic acid

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A magnetic porous carbon containing Fe$_3$O$_4$ (FPC) has been synthesized by a novel activation and carbonization process of sewage sludge without extra addition of ferric ions. Properties of FPC carbonized at 600, 800 and 1000 °C were studied using N$_2$ adsorption and desorption isotherms, X-ray diffraction (XRD), scanning electron microscopy (SEM) and vibrating sample magnetometer. The results indicate that FPC carbonized at 600 °C has a superior porous structure and high pore volume (0.504 mL/g). Further study found that Fe$_3$O$_4$ is dominating in the presence of iron in FPC carbonized at 600 °C. The resulting chars show higher catalytic activity in 1-diazo-2-naphthol-4-sulfonic acid (1,2,4-Acid) oxidation than commercial Fe$_3$O$_4$ MNPs. The 1,2,4-Acid and TOC removal efficiency can reach 96.6% and 87.2% after 260 min Fenton-like treatment. The mechanism in FPC-H$_2$O$_2$ system may include a Haber–Weiss type reaction between the active sites (e.g. Fe$_3$O$_4$) in FPC and hydrogen peroxide.

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1. Introduction

An increasing amount of sewage sludge generated by wastewater treatment plant has become an issue of particular concern (Wurz et al., 2011). Common disposal options for sewage sludge include landfill, application to farmland and forestry and incineration (Stuber et al., 2011). The reuse of sewage sludge is an attractive alternative that is being widely studied. The studies using sludge as a precursor are now primarily centred on the production of activated carbons and much work has been done on sludge derived carbons for sorbing organic pollutants and heavy metals (Smith et al., 2009; Wen et al., 2011). Nevertheless, the application of sludge derived products as efficient catalysts has not been reported in literatures. It is reported that sewage sludge is rich in Fe species and Mohamed et al. observed the presence of a large amount of iron in sewage sludge, about 100 times more than that found in commercial activated carbons (Mohamed et al., 2011). Therefore, the effective recycling of the sewage sludge as effective materials is of great interest and needs to be investigated.

Recently, the catalytic abilities of Fe oxide have been found most effective in heterogeneous Fenton-like oxidation of various organic pollutants due to their superior catalytic behavior and properties that could work over a wide pH range (Manh et al., 2011; Zhang et al., 2010). However, the decaying catalytic activities of iron oxide and constantly exhausted natural resources have posed challenges to searching for alternatives. Herein, we propose a novel strategy to synthesize a porous carbon containing magnetite Fe$_3$O$_4$ from sewage sludge by using a simple physiochemical activation and carbonization processes. These carbon materials combined the advantages of high surface area and good magnetic separability. 1-Diazo-2-naphthol-4-sulfonic acid (1,2,4-Acid), which is an important dyestuff intermediate and used as diazo component for synthesizing a whole range of textile and leather dyes (Gu et al., 2011, 2012), was used as a model target pollutant in the Fenton-like reactions in the presence of FPC. FPC catalyst thermally treated at 600 °C shows good catalytic property and can be easily separated by magnet at the end of degradation process.

2. Experimental

2.1. Preparation of FPC

(1) The concentrated sludge was pretreated by impregnating into 0.5 M potassium hydroxide solution, and the pH value was controlled at 12.0. After alkaline addition, the sludge was further stirred for 30 min and then placed in a temperature-controlled cabin at 50 °C for 20 h.

(2) The activated wet sludge was then centrifuged at 4000 rpm for 10 min, dried at 110 °C for 24 h, crushed with distilled water several times, and finally sieved into a granular size less than 20 mesh.

(3) The pretreated sludge was then subjected to immerse in the HNO$_3$ solutions at pH 1 under microwave digestion at room temperature for 2 h and then dried at ambient temperature..