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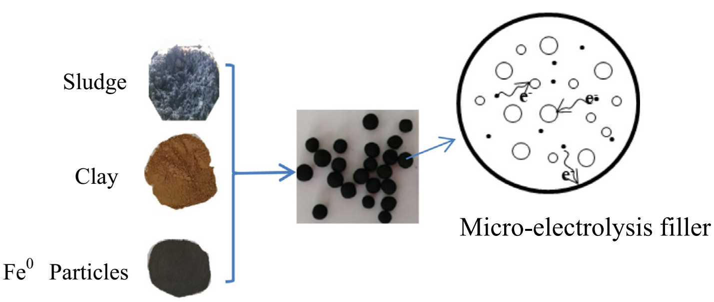
Regular Article

Sludge based micro-electrolysis filler for removing tetracycline from solution

Xiaoqian Cui, Ni Li, Guodong Chen, Hao Zheng, Xiaochen Li *

College of Water Conservancy and Civil Engineering, Shandong Agricultural University, Tai'an, Shandong 271018, PR China

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, a novel catalytic micro-electrolysis filler (CMEF) was prepared using waste sludge as the main raw materials. The preparation process was optimized in terms of the mass ratio of raw materials and sintering temperature based on the tetracyclines (TCs) removal efficiency. The physicochemical characteristics (e.g., surface area, morphology features, function groups, and valence state of Fe) of the prepared CMEF were determined. Finally, the removing performance and mechanism of CMEF were discussed by removing TCs from solution. The results showed that the optimum conditions for CMEF preparation were a sintering temperature of 1050 °C, and the mass ration for sludge:clay:Fe powder of 3:2:2. The CMEF exhibited a high degradation capacity for TCs with a removal rate up to 99.9% in 2.5 h, and the removal kinetics fit well with a pseudo-second-order model. Introducing Fe into the CMEF significantly promoted TCs removal, comparing to the removal of TCs by sludge based ceramic (without addition of iron). This study provides a novel cost-effective CMEF based on sludge, and is also of significance for co-friend recycling of waste sludge.

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1. Introduction

In recent years, catalytic micro-electrolysis fillers (CMEFs), which are based on traditional electrochemistry technology, have attracted much attention for treating wastewater recalcitrant to degradation [1]. CMEF systems are based on the potential difference between Fe^0 and activated carbon (AC), which is assumed to have a standard electrode potential difference of approximately 1.2 V [2]. Numerous micro-galvanic cells are formed when Fe^0

Abbreviations: CMEF, catalytic micro-electrolysis filler; AC, activated carbon; GAC, granular activated carbon; TCs, tetracyclines; FTIR, Fourier transform infrared spectroscopy; SEM, scanning electron microscopy; XPS, X-ray photoelectron spectroscopy; SCF, sludge-based carbon filler; HPLC, high-performance liquid chromatography; Qe, equilibrium absorption capacity; ROS, reactive oxidative species.

* Corresponding author.
E-mail address: lixiaochen02@163.com (X. Li).

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and C are immerged simultaneously into an electrolyte solution, in which Fe^0 and C acts as the anode and cathode, respectively. As electrons migrate from the anodes to cathodes, some products (e.g., active ·H, H_2O_2, and ·OH) are generated at the electrodes. Most of these products are highly reactive and can break the highly complex molecules of organic pollutants recalcitrant to degradation, enabling otherwise non-degradable organic pollutants to be degraded into simple molecules and inorganic [3,4]. CMEF technology has been applied extensively to treat refractory organic wastewater, including oil refinery wastewater [5], acrylonitrile wastewater [6], soy protein secondary wastewater [7], printing and dyeing wastewater [8,9], and other industrial wastewater due to the significant advantages of not requiring external power, low cost, and convenient operation and maintenance.

Traditional fillers used for Fe/C micro-electrolysis are usually made from physical mixtures of Fe chips and AC. The treatment efficiencies and practical applications of the fillers in CMEF systems largely depend on their inherent characteristics, such as porous structure, specific surface area and raw materials. Thus, the selection and preparation of fillers are key factors for the application of CMEF systems. A variety of materials have been used to produce fillers, including AC, sand, perlite, zeolite, lava rock, and sponge [10,11]. Granular activated carbon (GAC) is the preferred material due to its large specific surface area and porous structure. In recent decades, many waste materials such as sludge [12], oil palm [13], fruit stones, nutshells [14], and waste walnut shells [15] have been used as raw materials to produce GAC and ceramic fillers. Of all these raw materials, sludge is a byproduct of water and wastewater treatment processes, which contains toxic heavy metals, organic pollutants, soluble salts, and pathogenic microorganisms [16]. Thus, environmentally benign and economical methods for safe disposal and recycling of sludge are of great significance due to its hazardous properties and the large quantities produced [17]. Many efforts have been made to use waste sludge to produce wastewater treatment materials, such as GAC, ceramic particles, and other adsorbents. Wu et al. [7] prepared fillers with 75 wt% clay and 25 wt% sludge for soy protein secondary wastewater treatment using an up-flow aerated filter system. Yang [18] prepared ceramic fillers with a sludge-to-clay ratio of 1:(3–4) to treat tetracycline wastewater in a up-flow anaerobic bio-filter. Mymrin et al. [19] employed sludge as the principal component to enhance the mechanical properties of ceramics.

The main components of waste sludge can be divided into two groups, inorganic and organic components. The inorganic components of sludge include clay, sand, and chemicals salts (e.g., Fe, Al, Ca, Si, and other trace elements), which can be used as added substitutes in clay to form the ceramic skeleton of fillers. Meanwhile the organic components of sludge, including organic compounds and microorganisms, can be turned into porous carbonaceous matter via carbonization and volatilization under high-temperature and oxygen-deficit conditions [20]. Therefore, waste sludge is assumed to be recycled effectively into raw materials for producing catalytic micro-electrolysis fillers. Regardless, there is little research on this subject.

As pharmaceutical compounds, antibiotics are frequently used in human and veterinary medicine. However, the abuse of antibiotics has attracted increasing attention because of their considerable adverse effects on humans and ecosystems [21]. Tetracycline is one of the most common antibiotics in terms of both production and use, and is frequently detected in surface and ground waters [22]. Therefore, the removal of TCs has been deserved much concern because of their high persistence and potential ecotoxicity. Previous studies have demonstrated that conventional wastewater biological treatment processes are not as effective in removing TCs as previously expected because of their recalcitrance to degrade and lethal toxicity to most microorganisms [23]. Advanced oxidation processes, such as ozonation and Fenton oxidation, are promising approaches for antibiotics removal [2]. However, advanced oxidation processes are expensive due to the high cost of the required oxidants, especially considering that the co-existing bulk pollutants or other reductive substances in wastewater might consume more of other oxidants present in higher concentrations than TCs [24]. As another potential solution, catalytic micro-electrolysis may be theoretical feasible for decomposing TCs due to the generation of highly reactive compounds such as H_2O_2 and ·OH on the electrode.

The main objective of this study was to develop a cost-effective catalytic micro-electrolysis filler (CMEF) using waste sludge as the main raw material. The physicochemical characteristics of the prepared CMEF were investigated using Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and X-ray photoelectron spectroscopy (XPS). In addition, the ability of the prepared CMEF for removing organic matters difficult to degrade was assessed based on TCs removal in batch assays, since TCs are considered typical organic compounds difficult to biodegrade. The results will provide the basis for a novel CMEF using waste sludge as the raw material, and support a novel method for recycling waste sludge in an environmentally benign manner.

2. Material and methods

2.1. Pretreatment of the raw materials for CMEF

Waterworks sludge, sewage sludge, clay, and reductive Fe powder were used as the raw materials to prepare the CMEF. The waterworks sludge was obtained from the Sanhe Waterworks of Tai'an, Shandong Province, China. Sewage sludge was collected from the third municipal wastewater treatment plant of Tai'an, Shandong Province, China. Clay was collected from a farm in a suburb of Tai'an, Shandong Province, China. Reductive Fe powder was of analytical grade, and was made in Tianjin, China.

The waterworks sludge, sewage sludge, and clay were dried separately in an oven at 75 °C, until they reached a constant weight. Then, the sludge and clay were each ground using a pulverizer (JP-250A-8, China), and sieved through an 80 mesh stainless steel sieve with a diameter of 178 μm. Finally, the dry powders obtained from the raw materials were preserved in drying vessels to avoid humidity before use. The physicochemical characteristics of the sludge and clay used in this study are available in the supplementary material (Table S1).

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.jcis.2018.09.061.

2.2. Preparation of CMEF and sludge-based carbon fillers

An orthogonal experimental design was employed to optimize the multivariate analysis of the prepared CMEF, which were selected based on preliminary results. The most important parameters for the preparation of CMEF were the mass ratios of the raw materials and the sintering temperature, which influenced the characteristics and function of CMEF. Therefore, the sintered temperature, and dry weight of sewage sludge, clay and Fe powder were chosen as variables. The orthogonal experiment included five levels of four factors and was designed using SPSS 16.0 (SPSS Inc., Chicago, IL, USA). The designed orthogonal procedure is shown in Table 1.

The CMEF was prepared as follows:

Step 1: The prepared waterworks sludge, sewage sludge, clay, and reductive Fe powder were homogenized completely in the prescribed proportions. Deionized water was added to the mixture until the powdered materials readily clumped, and then raw uni-

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