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Short communication

Effective treatment of oily scum via catalytic wet persulfate oxidation process activated by Fe^{2+}

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ABSTRACT

Oily scum, a hazardous by-product of petroleum industry, need to be deposed urgently to reduce environmental risks. This paper introduces catalytic wet persulfate oxidation (CWPO) process in the treatment of oily scum to realize risk relief. Under the activation of heat and Fe^{2+}, persulfate (PS) was decomposed into sulfate radicals and hydroxyl radicals, which played a major role on the degradation of petroleum hydrocarbons. The effects of wet air oxidation (WAO) and CWPO process on the degradation of oily scum were compared. In CWPO process, the total petroleum hydrocarbons (TPHs) content of oily scum was decreased from 92.63% to 16.75%, which was still up to 70.19% in WAO process. The degradation rate of TPHs in CWPO process was about 3.38 times higher than that in WAO process. The great performance of CWPO process was also confirmed by elemental analysis, which indicated that the C and H contents of oily scum were reduced significantly by CWPO process. These results indicated that CWPO process has high potential on the degradation of oily scum for environmental protection.

1. Introduction

Oily scum is a hazardous and complicated contaminant containing various recalcitrant petroleum hydrocarbons (PHCs), which are toxic, mutagenic and carcinogenic (Guan et al., 2017; Nkhalambayausi Chirwa et al., 2017). Hence, oily scum has been classified as priority environmental pollution, and taking relevant disposal is urgent (Cerqueira et al., 2011; Zhang et al., 2017). Recently, a variety of methods such as biodegradation, incineration and landfill have been applicated in the treatment of oily scum (Gholami-Shiri et al., 2017; Guan et al., 2018; Liu et al., 2010). As a simple, promising and reliable technology, biodegradation has been widely used for organic pollutant treatment. However, biodegradation is not very ideal in treating oily scum due to its relative long degradation period and poor degradation performance. As shown in the study of Jasmine and Mukherji, only about 5.8% of oil was degraded after 30 day biodegradation (Jasmine and Mukherji, 2015). Incineration is a valuable waste disposal method according to its high efficiency in reducing waste volume. Unfortunately, the production of toxic gases such as low molecular polycyclic aromatic hydrocarbons and fly ash may lead to air pollution. Besides, the high cost of the addition of auxiliary fuels and dewatering pretreatment may restrict its application (Hu et al., 2013). Although landfill is a cheap and straightforward method, oily scum without appropriate pretreatment may reduce the service life of landfill site and contaminate soil and ground water (Cui et al., 2009).

For treating oily scum swiftly and efficiently, wet air oxidation (WAO), one of the popular advanced oxidation technology, has attracted wide attention owing to its high efficiency in treating high-risk organic pollutants (Jing et al., 2012b). In WAO process with high temperature (423–603 K) and pressure (3–25 MPa), oxyen (O_2) in air was used as oxidant to degrade organic pollutants into simple soluble organic compounds, which are further decomposed into carbon dioxide (CO_2), water (H_2O) and other innocuous final products (Rocha et al., 2017; Weber et al., 2015). Considering the pursuit of greater treatment efficiency, the improvement of WAO process is necessary.

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Recently, persulfate (PS) has achieved wide concern due to its high efficiency, great solubility and extensive operative pH range (Yuan et al., 2015). Transition metal ions such as Fe^{2+}, Ni^{2+}, Cu^{2+}, Co^{2+} and Ag^{+} have great activation ability on PS for better treatment effect. However, the utilization of heavy metals may increase the risk of environmental pollutant and human healthy (Dong et al., 2017; Li et al., 2016). Hence, Fe^{2+} is a better choice according to its non-toxic, environmental friendliness and cheap price (Ji et al., 2014; Xiong et al., 2014). In PS activation by Fe^{2+}, PS is discomposed to oxidative sulfate radical ($SO_4^{\bullet-}$) and hydroxyl radical ($HO^{\bullet}$) (Eqs (1)–(3)), which have great performance in degrading organic pollutants. In the study of Tan et al., 90% of diuron was degrade by $SO_4^{\bullet-}$ and $HO^{\bullet}$ (Tan et al., 2012). Hence, it is possible to treat oily scum by catalytic wet persulfate oxidation (CWPO) process using PS as oxidant and Fe^{2+} as activator.

$$Fe^{2+} + S_2O_8^{2-} \rightarrow SO_4^{\bullet-} + SO_4^{2-} + Fe^{3+} \quad (1)$$

$$SO_4^{\bullet-} + H_2O \rightarrow SO_4^{2-} + H^{+} + HO^{\bullet} \quad (2)$$

$$SO_4^{\bullet-} + OH^{-} \rightarrow SO_4^{2-} + HO^{\bullet} \quad (3)$$

In this work, we are first to introduce CWPO process in the treatment of oily scum. The effects of WAO and CWPO process on the degradation of oily scum were compared. Besides, we investigated the effects of residence time, temperature, the molar ratio of Fe^{2+}:PS and PS concentration in degrading oily scum, which were analyzed by one-way analysis of variance (ANOVA) with 95% confidence level. The PHCs and element composition of oily scum after treatment were investigated. Compared with WAO process, CWPO process showed high application potential in the treatment of oily scum.

2. Experimental section

Oily scum was generated from the flotation process in the first sewage treatment plant, Sinopec Changling branch water operations department (Yueyang, China). Specific characteristics of oily scum are presented in Table 1. Specific experimental procedure and analysis methods have been shown in support information.

3. Results and discussion

3.1. Effect of residence time

The changes of TPHs contents were studied with different experimental conditions. To ensure the efficiency of CWPO process in degrading oily scum, the effect of residence time on oily scum treatment was depicted in Fig. 1. With the extension of residence time, the total petroleum hydrocarbons (TPHs) content in CWPO process showed a decrease trend and became stable at 45 min. After 45 min reaction, the TPHs contents of oily scum in WAO and CWPO process were 70.30% and 35.55%, respectively, which indicated the high efficiency of CWPO process in degrading PHCs. The whole change of TPHs contents indicated that the degradation efficiency of oily scum was slowed down continuously with the increase of residence time. Previous research showed that the degradation of PHCs by $SO_4^{\bullet-}$ and $HO^{\bullet}$ was concentrated in the beginning of reaction, and then radicals were reduced rapidly by continuous reaction with PHCs (Jing et al., 2012a).

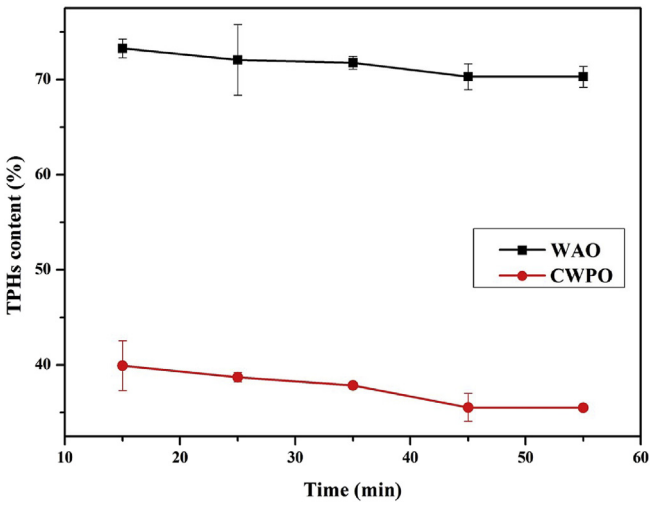

Fig. 1. Effect of residence time on TPHs degradation. Experimental condition: temperature = 583 K, Fe^{2+}:PS ratio = 1:1.2, PS = 63 mM.

3.2. Effect of temperature

As shown in Fig. 2, a significant decrease of TPHs content was observed in CWPO process, whereas fewer change of TPHs content was seen in WAO process. The TPHs content in CWPO process decreased from 60.67% to 35.55% with temperature increasing from 493 K to 583 K. High temperature promoted PS ($E^0 = 2.01$ V) to decompose to $SO_4^{\bullet-}$ ($E^0 = 2.50$–3.10 V) and $HO^{\bullet}$ ($E^0 = 1.80$–2.70 V).

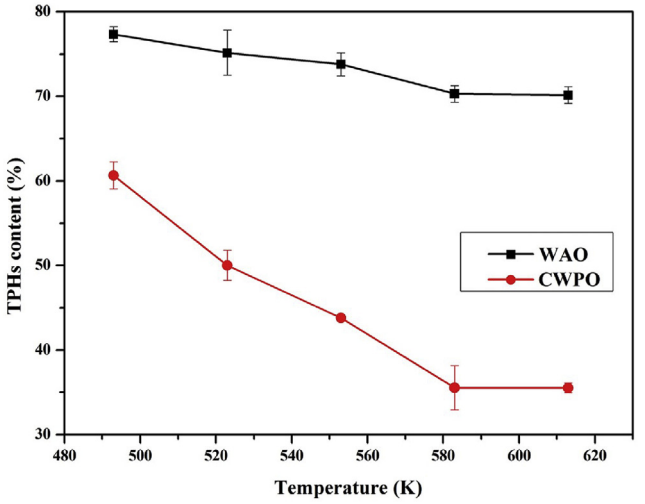

Fig. 2. Effect of temperature on TPHs degradation. Experimental condition: residence time = 45 min, Fe^{2+}:PS ratio = 1:1.2, PS = 63 mM.

Table 1
The charcateristics of original oily scum.

Characteristics	COD (mg/L)	Wet basis (%)			Dry basis (%)		Heavy metals contents (mg/kg)				
Original oily scum	7.8×10^4	Water content 60.68	Oil content 36.54	Solid content 2.78	Oil content 92.63	Solid content 7.07	Zn 73.79	Mn 18.37	Ni 3.93	Cr 0.76	Cu 0.69