

Treatment of heavy oil wastewater by a conventional activated sludge process coupled with an immobilized biological filter



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ABSTRACT

A field pilot study had been constructed in the Liaohe oilfield, China to treat heavy oil wastewater enriched with large amounts of dissolved recalcitrant organic compounds and low nutrient of nitrogen and phosphorus by conventional activated sludge process (CAS) coupled with immobilized biological aerated filter (I-BAF). After biological treatment, the chemical oxygen demand (COD) was removed around 64% when the hydraulic retention time (HRT) was 18 h. The average effluent COD reached approximately 75 mg L⁻¹, which met the national discharge standard. Gas chromatography–mass spectrometry (GC–MS) indicated that the CAS could completely remove phenolic, alkenes, aldehydes and organic acid compounds from the wastewater and the alkane components were removed by the I-BAF. Environment scanning electron microscopy (ESEM) disclosed that bacteria flourished in both reactors during the operating period and most of them resemble rods and filaments. The bacterial community structure analysis based on Polymerase Chain Reaction–Denaturing Gradient Gel Electrophoresis (PCR–DGGE) technology revealed that the predominant bacteria in the CAS reactor belonged to the *Pseudomonas*, *Planococcus* groups and the *Agrococcus*, *Acinetobacter* groups that were major degraders in the I-BAF reactor. Although some high molecular weight n-alkanes (C₁₅–C₂₃) were found to be refractory in our biotreatment systems, it could be improved by optimizing the process.

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1. Introduction

A large amount of heavy oil wastewater is produced during oil extraction in oil fields. Since the wastewater contains complex recalcitrant organic pollutants such as polymers, surfactants and various kinds of heavy mineral oil (Ji et al., 2002), it leads to severe contamination of water, soil and even air systems if it is discharged directly (Stromgren et al., 1995; Chen et al., 1999, 2003; Jerez et al., 2002). Therefore, an effective treatment for such wastewater is necessary before release into the environment.

Although physicochemical methods such as de-mulsification (Patrick et al., 2003), flotation (Zeng et al., 2007), micro-electrolysis (Li et al., 2010) and advanced oxidation processes (Li et al., 2006) can be used to treat such wastewater, high energy consumption and

secondary pollution restrict their application to wastewater treatment engineering. In contrast, biological methods had become increasingly popular in treating oilfield wastewater due to the high efficiency, cost effectiveness, and environment friendliness (Margesin and Schinner, 2001; Jiménez et al., 2006; Eusébio et al., 2007; Chavan and Mukherji, 2008). For example, the conventional activated sludge (CAS) process was commonly used in petrochemical wastewater treatment (Shokrollahzadeh et al., 2008; Ma et al., 2009) after solving sludge bulking (Ramothokang et al., 2003; Fikar et al., 2005). Recently, immobilization of microorganisms had been successfully used to remove toxic and refractory substances such as 2,4-dinitrotoluene (2,4-DNT) (Wang et al., 2010a,b), 2,4,6-trinitrotoluene (TNT) (Wang et al., 2010a,b), phenol (Banerjee and Ghoshal, 2011), surfactant aided (Sarma and Pakshirajan 2011) and *p*-nitrophenol (Sreenivasulu et al., 2012). Owing to advantages such as high hydraulic loading rates, high biomass concentration, less sludge formation, and protection of microorganisms (Zhao et al., 2006), it had also received considerable attention in the treatment of industry wastewater.

Since heavy oil wastewater had low chemical oxygen demand (COD) and nutrient (N and P) characteristics, a common conventional activated sludge (CAS) process was difficult to effectively treat

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the wastewater in order to meet the discharge criterion (Ji et al., 2009). Hence, a combination of biological techniques may be required for the treatment (Lu et al., 2009). The research aimed at investigating the feasibility and capacity of the treatment of the heavy oil wastewater using a CAS process coupled with immobilized biological aerated filtering (I-BAF) technology. In the work, a pilot instrument was constructed and operated continuously for 185 days. The COD was used as an indicator of the biodegradation rate and gas chromatography–mass spectrometry (GC–MS) was used to analyze the change in the organic components in the wastewater before and after treatment. The microbial morphology was observed by environment scanning electron microscopy (ESEM). The polymerase chain reaction–denaturing gradient gel electrophoresis (PCR–DGGE) technique and sequencing analysis were used to identify the dominant microorganisms in the bioreactor.

2. Materials and methods

2.1. Materials

2.1.1. Heavy oil wastewater

The heavy oil wastewater used in this study was obtained from the Liaohe Oilfield located in Liaoning Province, northeastern China. The wastewater was composed of produced water, steam assisted gravity drainage (SAGD) wastewater, scum wastewater, and filtered and ion-exchanged wastewater. Thus, its composition was very complex and our measurements indicated that oil separated from the wastewater had a density of 0.94 g cm^{-3} (20°C), freezing point of -5°C , wax content of 1.6%, and high viscosity of 4500 mPas (20°C).

2.1.2. Seed activated sludge and microorganisms

The seed activated sludge was taken from an aerobic activated sludge pond of a super heavy oil produced water treatment plant in the Liaohe Oilfield, Liaoning Province, China. It had good settling characteristics ($\text{MLSS } 15,500 \text{ mg L}^{-1}$; $\text{MLVSS } 9500 \text{ mg L}^{-1}$) and showed a dark brown color.

Microorganisms (B350 and B925) were purchased from BIO-SYSTEM Co. (USA) and BIONETIX Co. (Canada), respectively. Each microorganism contained 28 special microorganisms, cellulase, lipase and hydrolytic enzyme. Its bulk density was $0.6\text{--}0.8 \text{ g cm}^{-3}$ and the number of the microorganisms was $3\text{--}5 \times 10^9 \text{ g}$. The B350 and B925 were used to degrade alkane compounds and aromatic compounds, respectively.

2.1.3. Carrier used for microorganism immobilization

A polycin urepan (FPU) carrier (Ye and Ni, 2004) was used to immobilize the microorganisms. The carrier consisted of

micro-pores and macro-pores. The macro-pores guaranteed better three-phase mixing of air, wastewater and carrier, and added three-phase mass transfer propulsion. The micro-pores could immobilize microorganisms via active chemical groups on the carrier surface such as $-\text{OH}$, $-\text{NH}_2$, $-\text{COOH}$, $-\text{CH}_2$, and $-\text{CHOCH}_2$. It had a size of $10\text{--}20 \text{ mm} \times 10\text{--}20 \text{ mm} \times 10\text{--}20 \text{ mm}$ in volume. The carrier density was about 1.0 g cm^{-3} and the specific surface area was $35,000 \text{ m}^2 \text{ m}^{-3}$ with a ratio of surface area to weight between 80 and $120 \text{ m}^2 \text{ g}^{-1}$. The water content and porosity were 2500% and 96%, respectively.

2.1.4. CAS-I-BAF system

Fig. 1 showed a schematic of the CAS-I-BAF pilot system. The system consisted of a tank (2.5 m^3), CAS system with a reactor ($\sim 10 \text{ m}^3$), settling tank ($\sim 5 \text{ m}^3$), and an I-BAF reactor ($\sim 27 \text{ m}^3$) where the FPU carriers were filled (60%, v/v). Air was released from the aerator pipes ($\varnothing 5 \text{ mm}$) located on the bottom of each aerobic reactor. The influent was pumped into the reactor from the bottom and the treated effluent exited from the top.

2.2. Methods

2.2.1. Experimental procedures

The test was divided into two stages, including the culturing, adapted to the specific wastewater to be treated and immobilization of microorganisms, and stable reaction operation stage. In the beginning stage (about 5 days), the CAS and I-BAF reactors were filled with pipe water (9 m^3), seed activated sludge (0.8 ton), microorganisms B350 (250 g) and B925 (250 g), sugar (240 g), carbamide (30 g), Na_2HPO_4 (5 g), and 2 ml trace element stock solution (Wang et al., 2010a,b). The air blowers were turned on and dissolved oxygen (DO) was controlled to be $2\text{--}4 \text{ mg L}^{-1}$. After culturing for 5 days, the heavy oil wastewater was directly added to the biological reactors, but it was added partly each time. Briefly, it was added to replace one third of the pipe water in the biological reactors, and keep aeration for three days for microbial adaptation. This operation was repeated for three times until the tape water in the biological reactors was completely replaced. During this process which lasted about 10 days, the same amount of the nutrients and microorganisms were added to the reactors every two days and the heavy oil wastewater was continuously pumped into the reactors for domestication and immobilization of microorganisms. The flow rate was $0.5 \text{ m}^3 \text{ h}^{-1}$. When the removal efficiency of the final effluent COD was over 40%, the first stage was considered to be complete and stable operation began.

2.2.2. Wastewater quality detection

The biochemical oxygen demand after 5 days (BOD_5) was determined according to the Chinese standards GB 7488-87.

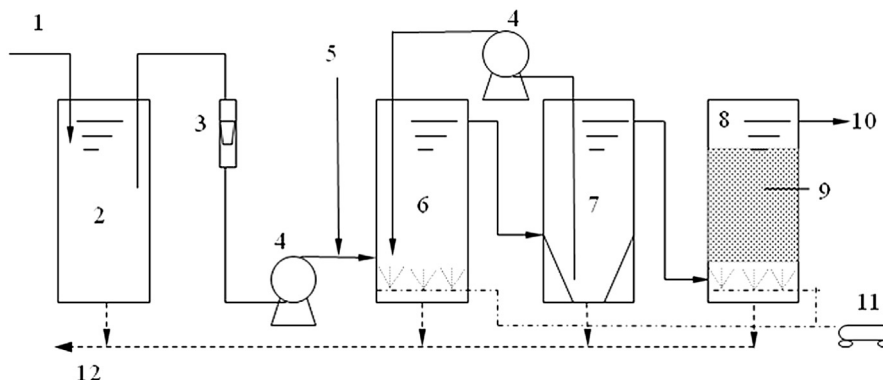


Fig. 1. Heavy oil wastewater treatment process. (1) Influent (2) Adjust tank (3) Flow meter (4) Water pump (5) Nutrients (6) Activated sludge reactor (7) Settling tank (8) I-BAF reactor (9) Carrier (10) Effluent (11) Air pump (12) Sludge.

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