

Combination of microwave demulsification, ozone oxidation and biological aerated filter for advanced treatment of oilfield wastewater with low biodegradability

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ABSTRACT

In this study, a combination process was developed in laboratory-scale including microwave demulsification, ozonation, and biological aerated filter (BAF) in series for the purpose of removing pollutants in oilfield wastewater with high microtoxicity. The microwave demulsification treatment removed 63.4% of oil. The ozonation treatment removed 79.3% of petroleum sulfonate, resulting in a substantial reduction of microtoxicity and great enhancement of biodegradability of the wastewater. During the following BAF process, the residual contaminants were effectively degraded by microbes. The final effluent could meet the emission standards for pollutants from petrochemical industry set by China's Environmental Protection Agency.

Key words | biodegradation, microtoxicity, oilfield wastewater, petroleum sulfonate

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INTRODUCTION

The majority of oilfields in China have now entered into the third oil recovery stage, which is carried out by injecting large quantities of water to flush out light oil or injecting steam to flush out heavy oil (Lu *et al.* 2009a). These approaches are considered to be the most efficient technique for tertiary oil recovery during the development of oilfields. However, more water is discharged along with the crude oil than is injected. This leads to the production of vast volumes of oily wastewater, which is called 'produced water' in the scientific literature (Ebrahimi *et al.* 2009). This wastewater must be treated properly, both to recycle the water resources and to prevent the pollution of underground water around farmlands (Lu *et al.* 2009a).

The treatment and remediation of oily wastewater through microbial degradation has been an important management process for many years (Lu *et al.* 2009a, 2011; Dong *et al.* 2011; Dias *et al.* 2012; Tong *et al.* 2013). However,

biological treatment alone is generally not sufficient to remove surfactants, polymers and recalcitrant organic fractions from oilfield produced water (OPW) to meet levels for direct discharge. For instance, water-soluble polymers and surfactants have been widely injected into petroleum reservoirs for the enhancement of oil recovery in Chinese oilfields (Li *et al.* 2014). These practices have produced a large quantity of OPW containing recalcitrant polymers and/or surfactants. Thereupon, additional specific steps are needed to improve the treatment performance of OPW.

Chemical oxidation can not only oxidize/mineralize organic substances, but also reduce toxicity associated with formulation ingredients and active agents. Iron (including zero-valent iron and iron salts)-based Fenton oxidation has been extensively applied to the pretreatment of bio-refractory wastewater such as OPW (Lu & Wei 2011), coking wastewater (Chu *et al.* 2012), dye wastewater (Chen

et al. 2013), cyanide wastewater (Kumar & Pal 2013), and pesticide wastewater (Pliego *et al.* 2014), etc. Nevertheless, these reactions could result in the production of large amounts of precipitation sludge and the reaction rates are not easy to control.

As a strong oxidant, ozone (O₃) reacts rapidly with organic compounds in water. More importantly, it does not produce any solid residue. The cost of ozone production declines gradually with the development and improvement of ozone generation technology. Thereupon, ozonation is increasingly widely used as a pretreatment or a final polishing step for advanced wastewater treatment (Lotito *et al.* 2012; He *et al.* 2013; Zhang *et al.* 2014).

In oilfield wastewater, the two basic types of emulsions are water-in-oil (W/O) and oil-in-water (O/W). More than 95% of petroleum emulsions formed in oilfield wastewater are of the W/O type (Ali & Alqam 2000). Because of its volumetric heating effects and faster processing rate over conventional heating methods, microwave energy has been increasingly used as an alternative heating technology with the aim of promoting the destabilization of petroleum emulsions (Kuo & Lee 2010; Xia *et al.* 2010; Ferreira *et al.* 2013). The demulsification efficiency of petroleum emulsions through microwave is different for each material and depends upon many factors including pH, inorganic salts, irradiation power and duration, etc. (Kuo & Lee 2010; Xia *et al.* 2010; Ferreira *et al.* 2013). Thus, it is necessary to optimize operational parameters and treatment conditions for microwave demulsification of petroleum emulsions through experimental designs.

This study evaluated the feasibility of combination of microwave demulsification, ozonation, and biological aerated filter (BAF) processes for the advanced treatment of OPW. A series of laboratorial experiments were conducted to investigate the impact factors and overall performance of the hybrid system.

MATERIALS AND METHODS

Chemicals

Standards of *n*-alkanes (C₈–C₄₀) and 16 US EPA priority polycyclic aromatic hydrocarbons (PAHs) were purchased

from Sigma (St Louis, MO, USA). All other solvents and chemicals used were of analytical grade. All solutions were prepared with high purity water (>18 MΩ).

Wastewater sample

The OPW used in this study was collected from Henan Oilfield, China. Table 1 shows the main physical and chemical parameters of the wastewater. As shown, this wastewater was characterized by high microtoxicity and low biodegradability (5-day biochemical oxygen demand (BOD₅)/chemical oxygen demand (COD)). The OPW contained a relatively high concentration of petroleum sulfonate (PS). This surfactant is synthesized by sulfonation with petroleum fractions above 200 °C. As one of the excellent surfactants, PS has been widely used for the enhancement of oil recovery in China (Zhao *et al.* 2010). However, it has been reported that PS may exert high ecotoxicity toward oil-degrading bacteria (Lu *et al.* 2009b). Thus, the high microtoxicity of the OPW in this study could be attributed to the presence of PS.

Microwave demulsification tests

Demulsification tests were conducted using a domestic microwave oven (frequency 2.45 GHz, Guangdong

Table 1 | Characteristics of the OPW used in the present study

Parameter	Initial value
pH	6.6
COD (mg/L)	648
BOD ₅ (mg/L)	45
BOD ₅ /COD	0.07
Oil (mg/L)	68
TPH (mg/L)	76
NH ₃ -N (mg/L)	52
Total nitrogen (mg/L)	126
Total phosphorus (mg/L)	1.21
SO ₄ ²⁻ (mg/L)	34
PS (mg/L)	58
Total dissolved solids (mg/L)	3,346
Microtoxicity (EC ₅₀ , %)	8.6

Galanz Group Co., Ltd, Foshan City, China). This oven can provide a maximum of 750 W of continuous microwave irradiation power. In a series of batch demulsification tests, 100-mL aliquots of OPW were placed in a 250-mL glass bottle, and mechanically mixed using a magnetic stirrer. The glass bottles, specifically made for performing gravitational bottle tests, are conical, transparent, and volume-graduated, with a screw cap. The sample bottle was placed in the middle of the tray, the region indicated as being subjected to the greatest microwave radiation. The effect of each parameter was studied by fixing the values of other parameters. HCl (1 M) and NaOH (1 M) solutions were used to adjust pH to desired levels before tests. After microwave irradiation, the OPW was allowed to settle down for 30 minutes at room temperature. The oil floating on the water surface was skimmed out by filter paper. All tests were triplicated, and the average values of the readings were used as the final results. The maximum fluctuation in the separation efficiency results, for a given set of experimental conditions, was found to be <5%.

Ozonation tests

The ozonation experiment was performed in a batch mode on the OPW treated under the optimum process conditions of microwave demulsification found above. The experimental setup consisted of oxygen gas, an ozone generator, a glass column reactor, and two washing bottles. An ozone generator (TLCF-G-300B, Xuzhou Tianlan Ozone Equipment Co., Ltd, Xuzhou, China) with a maximum capacity of 25 g O₃/h produced ozone from pure oxygen. The reactor had a glass column of 2.5 L with a porous distributor at the bottom through which ozone was introduced into the solution. The gas flow rate was controlled by a needle valve and was measured by a flow meter. The column was supplied with two washing bottles, each of them containing 500 mL of acidified 10% KI solution for determining unused ozone. The inlet concentration of ozone was measured before each ozonation test. The outlet ozone concentration was measured continuously during the experiments. Single-factor experiments were performed to investigate the effect of ozone dosage and contact time on treatment performance.

Biological treatment

The ozonation effluent obtained under the optimum conditions was continuously pumped into a BAF through a peristaltic pump. The BAF was made of plexiglass with an inner diameter of 6 cm and a working volume of 1.5 L with microporous air diffusers fixed on the bottom of the BAF column. The column was packed with ceramic media about 2–3 mm in diameter with an average specific surface area of 5.3 m²/g. Pebbles with diameter of 1–2 mm were packed in the bottom of the BAF to enhance the proportion of air bubbles. The activated sludge used for inoculation was collected from a petrochemical wastewater treatment plant. After inoculation, the BAF was filled with the OPW and supplemented with sugar (5 g/L). The air diffuser was turned on and dissolved oxygen was controlled to be 2–4 mg/L. After 2 days, the reactor was connected to a circulation pump and was recirculated at a rate of 1.0 L/h. Every 2 days, sugar (5 g/L) and nutrients (CaCl₂, MgSO₄, FeCl₃, each 0.1 g/L) were added into the BAF to sustain the growth of microbes. After 10 days, the OPW was continuously fed into the reactor. After about 15 days of operation, biofilm was observed on the surface of media, indicating the completion of start-up stage and successful biofilm formation. The overall operation temperature was 24–30 °C. The BAF was operated in up-flow mode, and the gas–water ratio was 4:1. The hydraulic retention time (HRT) of the reactor was adjusted in the range of 1–4 hours.

Analytical methods

Water quality parameters were determined according to standard methods (China EPA 2002). In brief, COD was determined with K₂Cr₂O₇ and H₂SO₄ in a 1:1 ratio by the open reflux method with AgSO₄ as a catalyst and HgSO₄ to remove Cl⁻ ion interference. BOD₅ was measured via the oxygen consumption of bacteria breaking down organics in the sample over a 5-day period under standardized conditions. NH₃-N was determined by Nessler's reagent colorimetry. SO₂⁴⁻ ions were analyzed using an ion chromatography system (HIC-10A, Shimadzu, Kyoto, Japan). Oil was determined by using an infrared spectrophotometry oil-measuring instrument (HD-1000, Beijing Purity

Instrument Co., Ltd, Beijing, China) after dichloromethane liquid–liquid extraction.

Microtoxicity of wastewater was measured by an SDI M500 analyzer (SDI, Michigan Seattle, WA, USA) (Dong *et al.* 2011). Results were expressed as EC₅₀ (15 minutes, 15 °C), which was defined as the effective nominal concentration of wastewater (volume percent) that reduced the intensity of light emission by 50%.

GC–MS analysis

Analyses of saturated and aromatic fractions were carried out using a simplified sample clean-up and a gas chromatography–mass spectrometric (GC–MS) system. After liquid–liquid extraction with dichloromethane, the extract was fractionated by silica-gel column chromatography to separate saturate and aromatic fractions according to Bastow *et al.* (2007). The two fractions were analyzed separately by internal standardization using an Agilent 7890-5975c GC–MS equipped with an Agilent HP-5MS fused silica capillary column (60 m × 0.25 mm × 0.25 μm).

RESULTS AND DISCUSSION

Effect of microwave power on oil removal

The effect of microwave power on oil removal efficiency of OPW was investigated. The irradiation time was set to 45 seconds. As shown in Figure 1(a), a remarkable increase in oil removal was observed with increasing microwave power from 200 to 400 W. Under microwave power of 400 W, oil removal reached 63.4%. As microwave power increasing from 400 to 600 W, however, oil removal continuously declined. When microwave power was raised, the electromagnetic field on emulsion could be strengthened, leading to higher rotational speed of polar molecules of the emulsion. Thus, electric double layers at oil/water interfaces could be more easily destroyed, and the zeta potential of the emulsion droplets may decrease significantly, resulting in higher demulsification efficiency of emulsions (Hesampour *et al.* 2008). However, when microwave power was increased above a certain value, the charge neutralization could decline and oil removal may decrease. In addition,

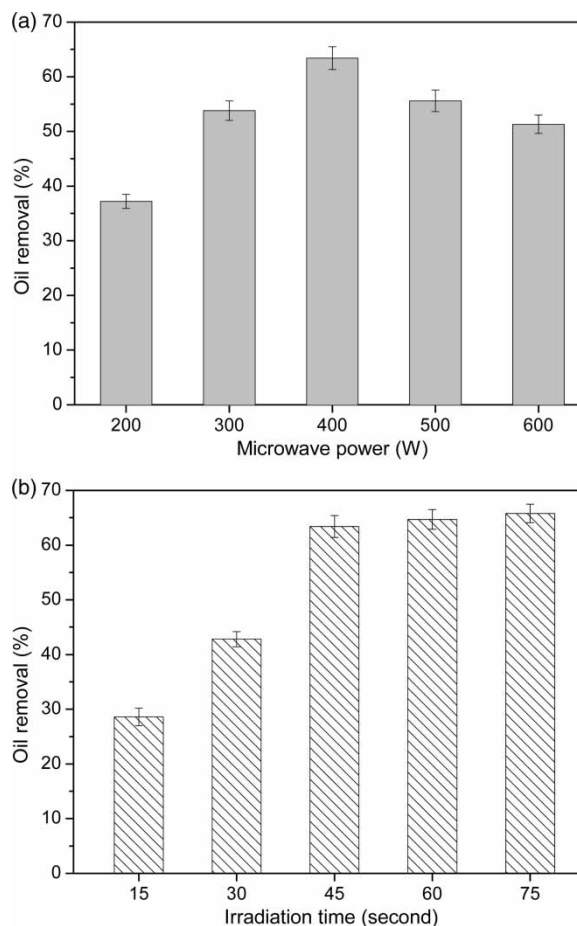


Figure 1 | Effect of (a) microwave power and (b) irradiation time on oil removal, using 100-mL aliquots of OPW with a settling time of 30 minutes and an initial oil content of 68 mg/L.

in the present study, it was found that the volume of the reaction solution expanded dramatically under higher microwave power due to strong exotherm. Thereupon, a microwave power of 400 W was used afterward.

Effect of microwave irradiation time on oil removal

To investigate the effect of microwave radiation time on the oil removal of OPW, batch experiments were performed at radiation time of 15–75 seconds, while fixing microwave power at 400 W. In general, an increase in irradiation time improved separation efficiency. As shown in Figure 1(b), the oil removal increased quickly with increasing radiation time from 15 to 45 seconds, and then increased little with time. By extending the radiation time, the interfacial film

of emulsion was gradually destroyed and the zeta potential of the emulsion droplets decreased. However, there existed limiting values for the destruction of interfacial film and the zeta potential of the droplets. Thus, the optimum operating conditions of microwave irradiation were 400 W of power and 45 seconds of irradiation. Under these conditions, the oil concentration decreased from 68 to 25 mg/L (Table 2), corresponding to 63.2% removal. This value was higher than the COD removal (41.1%), which was attributed to the fact that most of the dissolved pollutant fractions could not be removed by microwave irradiation.

The mechanism of demulsification by microwaves can be explained as below (Martínez-Palou *et al.* 2013): (1) a considerable reduction in the viscosity, which increases the temperature of the continuous phase and facilitates drainage of the interfacial film between the water droplets; and (2) microwaves induce molecular rotation, which tends to neutralize the zeta potential of the emulsion. Thereupon, the heating rate and the selective heating of the water are the two key factors for the high efficiency of microwaves to remove oil from emulsions. Microwave-assisted demulsification is a promising and economical technology as has been tested under field conditions (Morozov & Sedelnikov 2003).

Effect of ozone dosage on ozonation

Table 2 lists the water quality characteristics of OPW after treatment under the optimum operating conditions. As shown, except for COD, oil, and total petroleum hydrocarbons (TPH) were removed significantly, whereas other

parameters were changed little. Especially, the BOD₅/COD ratio was still very low, indicating that the microwave-treated effluent was not suitable for biological treatment. Thus, ozonation was performed on this effluent.

The ozonation tests were conducted for O₃applied/[PS]₀ ranging from 0.02 to 0.16 on the conditions of 15 minutes and natural pH (6.6). Since the major purpose of ozonation was to enhance the biodegradability of wastewater, only BOD₅/COD was continuously monitored and measured during the experiments. As shown in Figure 2(a), BOD₅/COD increased with increasing O₃ dosage up to 0.12 O₃applied/[PS]₀. At this O₃ dosage, the BOD₅/COD ratio was highest, close to 0.33. However, the BOD₅/COD ratio decreased when the O₃ dosage was further increased from 0.12 to 0.16 O₃applied/[PS]₀. This is attributed to the fact that an excess amount of O₃ could mineralize the biodegradable organics formed by ozonation. Thus, this O₃ dosage

Table 2 | Water quality parameters of the OPW after treatment

Parameter	After microwave treatment	After ozonation
COD (mg/L)	382	260
BOD ₅ (mg/L)	21	86
BOD ₅ /COD	0.08	0.33
Oil (mg/L)	25	16
TPH (mg/L)	28	18
NH ₃ -N (mg/L)	43	37
PS (mg/L)	58	12
Microtoxicity (EC ₅₀ , %)	8.8	31

The microwave and ozonation treatments were performed under their respective optimum conditions.

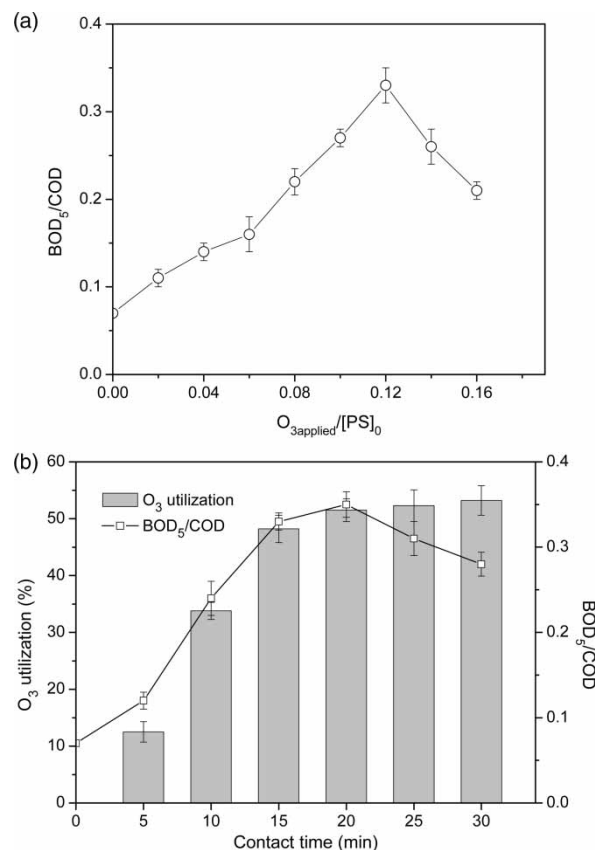


Figure 2 | Effect of (a) ozone dosage and (b) contact time on variations of BOD₅/COD of the microwave-treated effluent. [PS]₀ was the initial concentration of petroleum sulfonate (58 mg/L).

could be deemed optimum as it chemically yielded excellent BOD₅/COD improvement.

Effect of contact time on ozonation

Contact time is an important factor for ozonation because it can determine the size of the reactor in the practical applications. Thereupon, batch experiments of ozonation were performed corresponding to six different contact times ranging from 5 to 30 minutes. The optimum O₃ dosage (0.12 O_{3applied}/[PS]₀) was applied in all cases with the natural pH (6.6). The feed flow rate of ozone was regulated to adapt the duration of contact time during each test with the optimum O₃ dosage.

Figure 2(b) demonstrates changes of BOD₅/COD and O₃ utilization efficiency at different contact times. As shown, the BOD₅/COD ratio increased quickly during the first 15 minutes. For longer durations, a slow increment in BOD₅/COD was observed, even presenting a slight decrease during the ozonation time from 25 to 30 minutes. For instance, the BOD₅/COD ratio increased from 0.12 at 5 minutes to 0.33 at 15 minutes, and then decreased to 0.28 at 30 minutes. On the other hand, the O₃ utilization efficiency increased with an increase in the contact time. However, the ozone utilization efficiency increased slowly when the contact time was longer than 15 minutes. Therefore, the optimum operating conditions of ozonation were 0.12 O_{3applied}/[PS]₀ of O₃ dosage and 15 minutes of contact time.

Biological treatment by BAF

As shown in Table 2, the biodegradability of wastewater was enhanced to 0.33 after ozonation, indicating the wastewater was easily biodegraded (Lu & Wei 2011). In addition, it was found that the PS concentration and the microtoxicity decreased greatly (Table 2). Thus, the subsequent experiments were conducted using the BAF, and the ozonation effluent was continuously pumped into the BAF to remove the organic pollutants residue.

The effluent COD concentration of the BAF during the 60-day operation is plotted in Figure 3. The influent COD concentration was maintained at about 260 mg/L (Table 2). As shown, the effluent COD concentration was

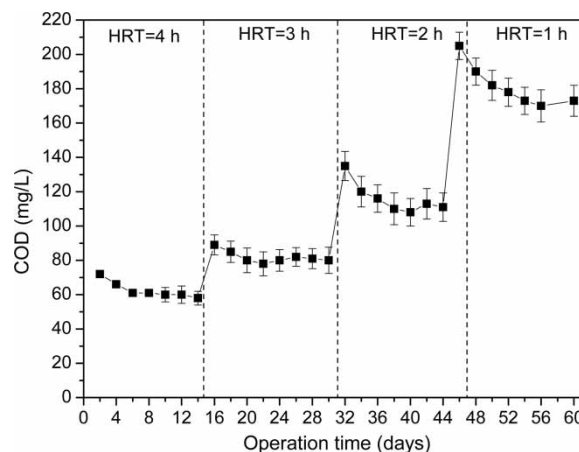


Figure 3 | Time course of COD concentration in the effluent of BAF.

averagely 65 mg/L, 83 mg/L, 116 mg/L and 178 mg/L at HRT 4 hours, 3 hours, 2 hours and 1 hours, respectively, corresponding to removal efficiencies of 75.0%, 68.1%, 55.4%, and 31.5%, respectively. At HRT 4 and 3 hours, the average effluent COD could satisfy the Chinese discharge standard for water pollutants from oilfield industry (GB8978-1996, grade I) (Table 3). Even at HRT 2 hours, the average effluent COD could satisfy the grade II discharge standard. Table 3 shows the average effluent oil and NH₃-N concentrations during the 60-day operation. As shown, at various HRTs, the effluent could satisfy the discharge standard concerning oil and NH₃-N.

The results obtained in the present study suggest that a BAF process has high adaptability to shock loading. This can be attributed to the higher biomass concentration that can be achieved in BAF systems (Liu et al. 2013; Tong

Table 3 | Overall effluent concentration (mg/L) of COD, oil and NH₃-N in the BAF and the Chinese discharge standard

	HRT (h)	Effluent level		
		COD	Oil	NH ₃ -N
Stage I	4	65	3.8	7.6
Stage II	3	83	4.5	9.5
Stage III	2	116	7.4	11
Stage IV	1	178	12	13
Discharge standard				
Grade I		≤100 mg/L	≤10 mg/L	≤15 mg/L
Grade II		≤150 mg/L	≤20 mg/L	≤25 mg/L

Table 4 | Total removal percentages of *n*-alkanes and PAHs after each treatment process

Hydrocarbon		Microwave treatment	Ozonation	BAF
<i>n</i> -alkanes	C8-12	36.4	68.3	100
	C13-16	43.3	62.6	100
	C17-20	51.2	79.5	100
	C21-24	62.6	82.9	98.6
	C25-28	63.1	84.6	96.5
	C29-32	68.4	81.0	95.4
	C33-40	74.5	82.8	93.7
PAHs	2-ring	34.2	52.6	92.4
	3-ring	42.3	61.5	85.8
	4-ring	47.4	68.3	84.0
	5-ring	50.6	66.5	78.3
	6-ring	55.2	67.2	75.6

The microwave and ozonation treatments were performed under their respective optimum conditions. The BAF process was sampled on the 28th day (HRT = 3 hours).

et al. 2013). BAF is characterized by high bioactivity, low energy consumption, little excess sludge, running stability and simple management, and has been successfully applied to OPW bioremediation (Liu *et al.* 2013; Tong *et al.* 2013).

Hydrocarbon dissipation

Table 4 lists total removal percentages of *n*-alkanes and PAHs after each treatment. The data obtained show that removal efficiency of individual components increased with the increase of molecular weight. This may be due to the fact that the solubility of hydrocarbon fractions decreased with the increase of molecular weight, and lower solubility led to higher separation efficiency by microwave demulsification. The BAF completely removed medium chain length *n*-alkanes (C8–C20) (100%) but incompletely degraded PAHs. The removal of 5- and 6-ring PAHs was lower in the BAF. It is known that biodegradation efficiency of hydrocarbons was negatively correlated with chain length or aromatic ring numbers (Lu *et al.* 2011).

CONCLUSIONS

The results of the present study demonstrate that the OPW with high microtoxicity could be well remediated by using the combined process of microwave demulsification, ozonation, and BAF under the optimum conditions. Under these

conditions, the effluent concentrations of COD, oil and ammonia could satisfy the discharge standard for petrochemical industry (China). This combination process has proved to be highly compatible and efficient in a laboratory-scale OPW treatment.

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