Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.931523

53 (2015) 2808–2815
March

Refining wastewater treatment using EGSB-BAF system

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Received 16 July 2013; Accepted 17 March 2014

ABSTRACT

The performance of an expanded granular sludge bed (EGSB) in combination with a biological aerated filter (BAF) system for petroleum refining wastewater treatment was investigated. The system was monitored for two months at a hydraulic retention time of 17.5 h and a digestion temperature of 34 ± 1˚C. The results showed that the total chemical oxygen demand (COD_{Cr}) and oil removal efficiencies were up to 90 and 87%, respectively, with the average COD_{Cr} and oil concentrations of 85 and 11 mg/L in the system effluent. Moreover, almost 97% of suspended solids (SS) were removed by the system and the effluent SS concentration was only 15 mg/L. The sludge yield coefficient of 0.0036 mg/mgCOD_{Cr} showed a low excessive sludge production for the EGSB reactor. The excellent treatment performance indicated that this EGSB-BAF system could be appropriate for refining wastewater treatment. Besides, methane yield was only about 0.21 mLCH_4/mgCOD_{Cr} in the EGSB reactor, lower than the theoretical yields. The poor methane production together with the low oil concentration and increased biodegradability of effluent indicated that the biochemical reaction of refining wastewater mainly remains in hydrolytic acidification phase in the EGSB reactor. For this reason, it was proposed that a highly efficient anaerobic process such as EGSB could be used as a pre-treatment process to improve the biodegradation performance of the following aerobic biochemical treatment.

Keywords: Petroleum refining wastewater; EGSB; BAF; Methane; Pre-treatment process

1. Introduction

Petroleum refining wastewater is characterized by a high concentration of oily organic pollutants, toxic to the micro-organisms and refractory to biochemical treatments. Conventional activated sludge processes cannot treat this kind of wastewater to meet tighter standards for emission. Therefore, it is imperative to improve the biodegradability of the petroleum refining wastewater and upgrade the treatment technologies. Anaerobic digestion, which is considered to be cost-effective and high-efficiency in the treatment of high-strength organic wastewater [1–3], has become a

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Presented at 2013 International Environmental Engineering Conference and Annual Meeting of the Korean Society of Environmental Engineers (IEEC 2013), Seoul, Korea, June 11–13, 2013

rapid-growing technology in recent years. However, when it comes to petroleum refining wastewater with fluctuant quality, poor biodegradability and low concentration, successful application of such technologies has rarely been achieved [4].

Expanded granular sludge bed (EGSB), the third generation of anaerobic reactor, was developed by Lettinga [5] of the Netherlands Wageningen Agricultural University in the early 1990s. Owing to the application of a high recirculation rate and a high height/diameter ratio of around 20 or more [6], the high up-flow liquid velocity is achieved in the reactor. In this way, granular sludge is expanded and the mass transfer between organic pollutants and micro-organisms are improved contributing to the increase of the biochemical reaction speed and the improvement of the treatment performance of the reactor [7, 8]. EGSB reactor has been successfully applied to treat various kinds of wastewater mainly from brewery, starch factory, pulp mill, slaughter house, palm oil mill effluent plant and so on [9–11]. Biological aerated filter (BAF) is a high-rate aerobic filter with reduced volume and fixed biofilm. BAF is a type of immobilization reactor, which integrates the biological contact oxidation, and an interception function of filtration process into one reactor. It has been widely applied all over the world as a novel wastewater treatment technology, due to its lots of advantages including high organic volumetric load and hydraulic load, short hydraulic retention time (HRT), excellent performance on organic removal, less infrastructure investment and low energy consumption and operation cost.

Integrating the advantages of anaerobic EGSB and aerobic BAF may be a solution to treat petroleum refining wastewater because of its high solid loading, good resistance to shock-loading, and low sludge production. In this paper, a hybrid process using an EGSB reactor combined with a BAF reactor was established. The possibility and performance of the integrated system to treat the petroleum refining wastewater were evaluated.

2. Materials and methods

2.1. Materials

The petroleum refining wastewater was taken from a refining sewage treatment plant in city Daqing, Heilongjiang province, China. The characteristics of the wastewater are shown in Table 1. The phosphate nutrients were added in the form of NaH_2PO_4 to obtain a C:N:P ratio of 200:5:1. In addition, the essential nutrients for an optimum anaerobic microbial growth were also prepared according to Ghangrekar et al. [12]. Then, 0.5 mL/L of the nutrients liquid was added into petroleum refining wastewater. The water-soluble organic products were analysed by a gas chromatograph-mass spectrometer (GC-MS) (SSQ710, Thermo-Finnigan, USA) computer equipped with a flame ionization detector.

The seed sludge for EGSB consisted of granular sludge and returning activated sludge with a volume ratio of 1:1.5. The granular sludge was obtained from a full-scale UASB plant treating brewery wastewater in city Daqing, Heilongjiang province, China and returning activated sludge was obtained from an anaerobic digester of a chemical sewage treatment plant in city Liaohe, Liaoning province, China. The biological carrier of BAF was the returning activated sludge-soaked ceramsite and a small amount of activated sludge was packed as stated before, with a packing height of 55 cm.

2.2. EGSB-BAF system

The schematic diagram of the experimental apparatus is shown in Fig. 1. The EGSB reactor was made of plexiglass column of 60 mm in internal diameter and 2000 mm in height. The total effective volume was 6.2 L. There was a gas–liquid–solids (GLS) separator at the top of EGSB reactor. The GLS separator had an internal diameter of 240 mm with a height of 400 mm. Besides, six sampling ports were arranged every 200 mm along the height of the reactor. The entire reactor assembly was housed in a vertical thermostat heater maintained at 34 ± 1℃. The methane produced was measured by a gas meter. The details of the reactor assembly were described elsewhere [13]. The BAF reactor had a working volume of 0.6 L (except the filler volume), height of 900 mm and an internal diameter of 60 mm. Four sampling ports were installed along the height of the reactor at intervals of 100 mm.

The flow diagram of EGSB-BAF process is shown in Fig. 1. The wastewater in the sink was pumped into the bottom of the EGSB reactor, and the effluent overflowed at the top of the reactor. Portions of the effluent from the EGSB were recycled to the bottom of the reactor at a suitable rate to maintain fluidization in the reactor. The total volumetric flow rate of methane was measured with a gas flow meter after flowing through a water seal bottle and a 3.0% sodium hydroxide solution bottle to absorb hydrogen sulphide and carbon dioxide. Some degradable organics were removed, and persistent organic pollutants were pre-processed effectively by micro-organisms at the same time in the EGSB reactor. Effluent from the top of the EGSB was pumped continuously to the BAF reactor. Residual organic pollutants were degraded further during this period. The effluent of the BAF reactor was pumped

Table 1
Characteristics of petroleum refining wastewater

Parameter	Range	Average value ± S.D. of 20 samples
pH	6–9	7.6 ± 0.8
COD_{Cr} (mg/L)	679–1,327	878.8 ± 171.7
Oil (mg/L)	90–130	115.8 ± 13.5
SS (mg/L)	400–500	453.6 ± 35.4
NH_3-N (mg/L)	40–50	44.9 ± 3.1

Table 2
The operating parameters of the EGSB-BAF hybrid system

Parameter	EGSB	Parameter	BAF
pH	6.5–7.8	HRT, h	1.5
HRT (h)	16	Temperature (℃)	20
Temperature (℃)	34 ± 1	Gas water ratio	9
Reflux ratio	6	Dissolved oxygen (DO) (mg/L)	3.6–4.6
Liquid upflow velocity (m/h)	0.94	Height of media (mm)	550
Inoculation ratio	1:1.5(V:V)	Backwash strength L/(m^2s), Frequency and duration	1.5 10 min per 15 d

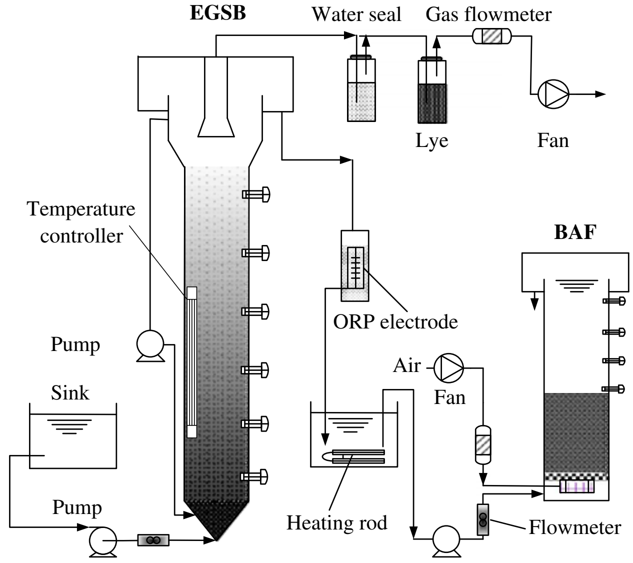

Fig. 1. Schematic diagram of the EGSB-BAF hybrid system.

out from an effluent weir at the top of the reactor. The operating parameters of the EGSB-BAF hybrid system are shown in Table 2. The experimental start-up procedure was finished in 94 d when about 70–64% chemical oxygen demand (COD_{Cr}) removal efficiencies were obtained in the EGSB reactor and BAF reactor, respectively. The details of the start-up procedure and affecting factors were shown in the previous researches [13,14].

2.3. Analytical methods

The influent and effluent samples of each reactor were analysed. The definite analytical methods are as follows: pH values were measured by a FE20 pHmeter (METTLER, TOLEDO) and the other physic-chemical parameters, COD_{Cr}, ammonia nitrogen (NH_3-N), oil concentration, SS, total suspended solids, mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were determined according to the standard methods for the examination of water and wastewater (SEPA, 2002) [15]. Total methane production was measured by water displacement method. Sample preparation: the water sample separated by microfiltration through a 0.45 μm membrane was prepared by acid and alkaline extraction, respectively. First, pH was adjusted to over 12 with 1 M NaOH or less than 3 by adding 1 M HCl. Then, 300 mL of the sample was transferred to a separatory funnel and 20 mL dichloromethane was added. The mixture was shaken for 15 min and the organic phase was collected. The extraction process was repeated three times. Then, a rotary evaporator concentrated the extracts. The GC-MS system involved a gas chromatograph with a SE-54 silica capillary column (60 m × 0.25 mm × 0.25 μm) and a SSQ 710 quadrupole mass spectrometer (Finnigan, MAT, USA). Finally, 1 μL of concentrated liquor was injected to GC-MS system to analyze the water-soluble organic matter. Instrumental conditions were as followed: initial column temperature was 40℃ and then increased to

270℃ at 10℃/min; gasification and transmission line temperature was set at 300–250℃, respectively. Nitrogen was used as a carrier gas, and column pressure was 414 kPa. Electrospray ionization (EI) was adopted. Ion source and interface temperature was 200 and 280℃, respectively. The electron energy was 70 eV, and mass scan range was from 35 to 400 amu.

3. Results and discussion

3.1. Overall performance of the hybrid EGSB-BAF system

3.1.1. Performance of the EGSB reactor

The COD_{Cr}, oil and SS removal efficiencies data of the EGSB reactor are depicted in Fig. 2. The influent COD_{Cr} concentration fluctuated greatly in the range of 679–1,327 mg/L and the effluent COD_{Cr} was in range of 186–299 mg/L. An average influent oil concentration was 116 mg/L and a mean of effluent oil concentration was 34.8 mg/L. The results clearly revealed that the EGSB reactor achieved a substantial reduction of COD_{Cr} and oil, resulting in average removal efficiencies of 72.72 and 69.85%, respectively. The effluent qualities were improved and the capacity of resistance to shock loading was excellent in EGSB reactor. Kumar et al. [16] had drawn the same conclusion: when treating low strength industrial cluster wastewater, the anaerobic hybrid reactor performed very well and proved its capability to withstand hydraulic shocks that occur in industrial applications. It may be a contribution of the effluent cycling system to the excellent operation effect, which improved liquid upflow velocity (V_{up}) and stirring intensity inside the EGSB reactor. For this reason, the mass transfer process between the substrate and the micro-organisms was strengthened and the toxic effect of oil on micro-organisms was reduced at the same time. Therefore, this efficient operation can be attributed to the higher impact resistance of the combined system. Similar reasons have been summarized by Ruiz et al. [17] when discussed about the methanogenic toxicity in anaerobic digesters treating with different concentration of municipal wastewater. However, the removal efficiencies of SS were relatively variable from 50 to 72.29%, and the average data was 60.16%. It may be the result that the effluent contained a portion of suspended sludge in the EGSB reactor. So the effluent needs a further treatment by aerobic technology.

3.1.2. Performance of the BAF reactor

The COD_{Cr}, oil and SS removal efficiencies data of the BAF reactor are depicted in Fig. 3. Owing to the anaerobic process, most toxic organic pollutants have been degraded so that the biodegradability of refining wastewater has been improved significantly. The average COD_{Cr} concentrations in influent and effluent were 237 and 85 mg/L, respectively. An average effluent oil concentration was 11 mg/L and a mean of effluent SS concentration was 15 mg/L. The results clearly revealed that the removal of COD_{Cr}, oil and SS was improved greatly in BAF reactor resulting in average removal efficiencies of 63.67, 66.55 and 92.39%, respectively. Some organic pollutants in refining wastewater, difficult to be degraded by aerobic microbes, can be used directly and indirectly by anaerobic bacteria. The refractory organics were converted to the biodegradable organics, NH_3, and biogas in the anaerobic digestion phase, so that the organic pollutant removals were improved and the total energy consumption for aeration and handling excess sludge were reduced. In addition, most of SS were removed by the interaction of physicochemical absorption and interception of the filter material layer in BAF reactor.

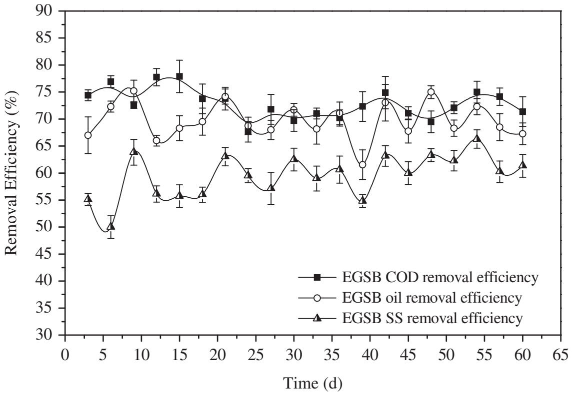

Fig. 2. The removal efficiencies of COD_{Cr}, oil and SS in the EGSB reactor.

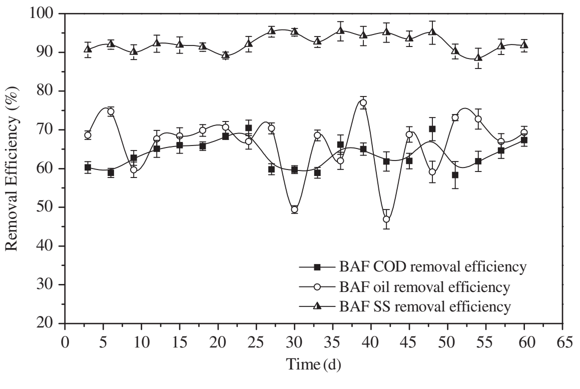

Fig. 3. The removal efficiencies of COD_{Cr}, oil and SS in the BAF reactor.

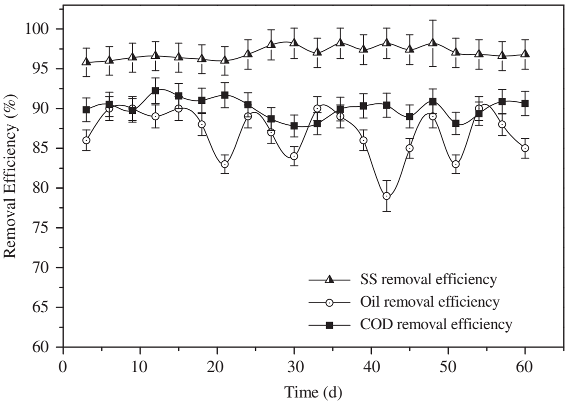

Fig. 4. The removal efficiencies of COD_{Cr}, oil and SS in the EGSB-BAF hybrid system.

3.1.3. Performance of the hybrid EGSB-BAF system

Such hybrid system achieved a substantial reduction of COD_{Cr}, oil and SS resulting in average removal efficiencies of 90, 87 and 97%, respectively (Fig. 4). The results clearly revealed that although the refining wastewater qualities fluctuated greatly, the effluent quality index reached the first grade standard of GB8978-1996. The hybrid EGSB-BAF system was relatively efficient and stable throughout the experiment and seemed to be not significantly affected by influent fluctuation. According to the overall removal of sewage indicators, the hybrid system is suitably applied to treat the petroleum refining wastewater (Fig. 4).

The water-soluble organic products of the influent and effluent in the EGSB process were analysed by GC-MS (Figs. 5 and 6). The analysis results (as shown in Tables 3 and 4) indicated that water-soluble organic products of the effluent for decomposition in the EGSB process include hydrolysis products such as acids, esters and aldehyde, which were easy to be degraded by the following aerobic biochemical treatment. It was suggested that a high-rate anaerobic process, such as EGSB, could be used as a pre-treatment process by controlling the running parameters to serve as high-rate hydrolysis and acidification reactor to improve its biodegradation performance in the following aerobic biochemical treatment.

3.2. Sludge production

Anaerobic sludge yield coefficients of the EGSB reactor were calculated in Eq. (1).

$$\text{Sludge yield coefficient } Y(t_1) = \frac{V\frac{dX}{dT} + Q_e X_e}{Q(C_i - C_e)} = \frac{V\frac{X(t_1)-X(t_0)}{t_1-t_0} + Q_e X_e(t_1)}{Q(C_i(t_1) - C_e(t_1))} \quad (1)$$

where V is the EGSB reactor volume (L), T is the experiment time (d), Q and Q_e are the influent and effluent flow rates (L/d), respectively, X and X_e are the influent and effluent MLVSS (mg/L), respectively,

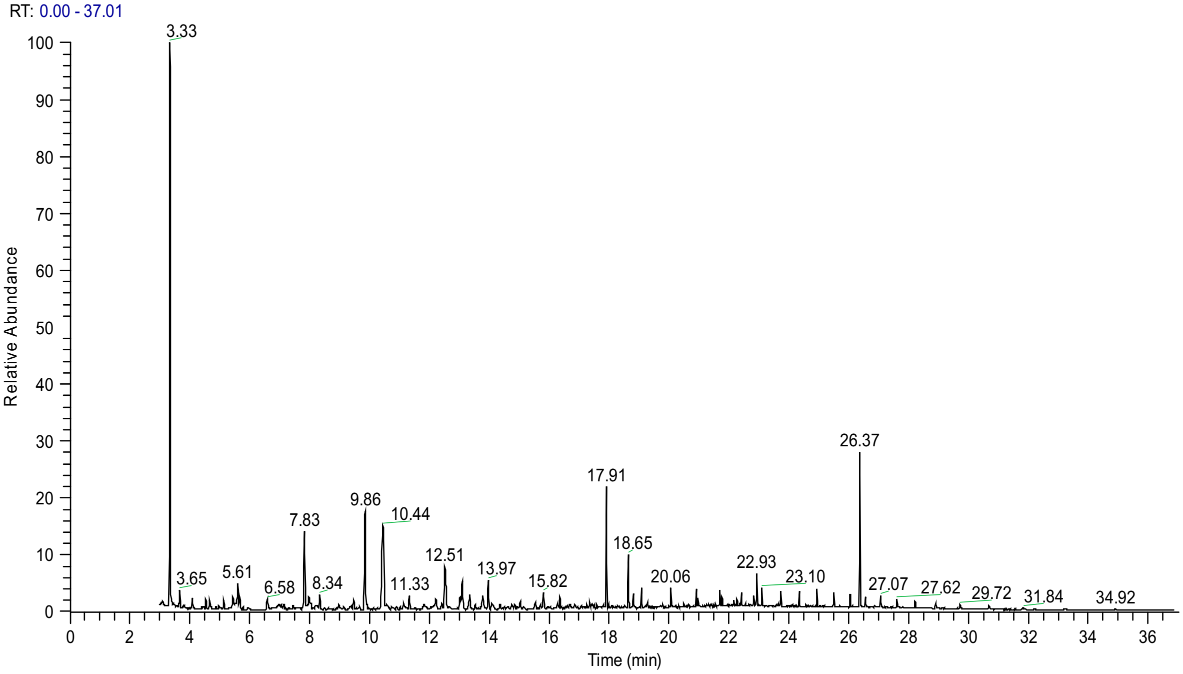

Fig. 5. The total ion chromatogram of GC-MS for the petroleum refining wastewater.

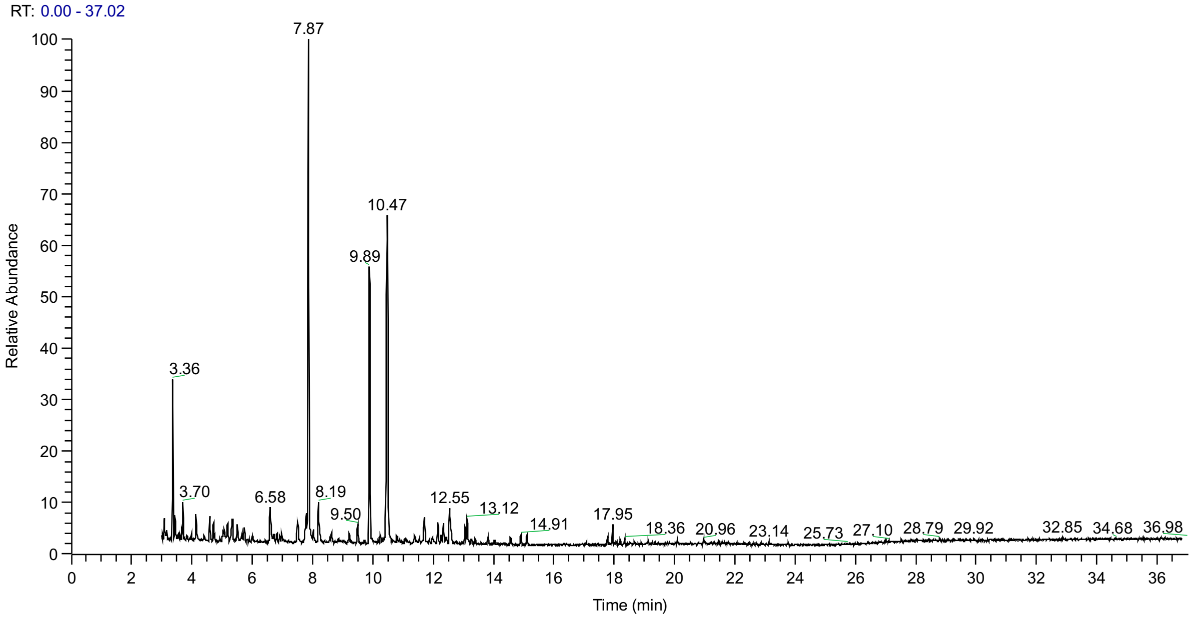

Fig. 6. The total ion chromatogram of GC-MS for the effluent from EGSB reactor.

Table 3
Toxic organic pollutants of petroleum refining wastewater

No.	Species	Molecular formula	Relative contents (%)
1	Valeric acid	$C_5H_{10}O_2$	3.15
2	Cyclopentanone	C_5H_8O	0.65
3	2,3-Dimethyl-2-cyclopenten-1-one	$C_7H_{10}O$	0.60
4	1,2-Benzenedicarboxylic acid dimethyl ester	$C_{10}H_{10}O_4$	0.25
5	1,2-Benzenedicarboxylic acid dibutyl ester	$C_{16}H_{22}O_4$	0.04
6	2,4-Dimethylhexane	C_8H_{18}	5.34
7	Toluene	C_7H_8	1.59
8	Ethybenzene	C_8H_{10}	2.67
9	1.2-Dimethyl-benzene	C_8H_{10}	1.20
10	P-Xylene	C_8H_{10}	12.4
11	1-Ethyl-2-methyl-benzene	C_9H_{12}	6.78
12	1-Ethyl-4-methyl-benzene	C_9H_{12}	6.62
13	1.2.4-Trimethylbenzene	C_9H_{12}	2.30
14	4-Hydroxy-benzenesulfonic acid	$C_6H_6O_4S$	3.50
15	4-Methyl-octane	C_9H_{20}	5.80
16	1.2.3-Trimethyl-benzene	C_9H_{12}	3.00
17	1.3.5-Trimethyl-benzene	C_9H_{12}	4.46
18	Indane	C_9H_{10}	4.44
19	Phenylethylene	C_8H_8	0.13
20	2-Methyl-phenol	C_7H_8O	9.12
21	3-Methyl-phenol	C_7H_8O	2.38
26	4-Hydroxytoluene	C_7H_8O	2.37
27	2.6-Dimethyl-phenol	$C_8H_{10}O$	4.50
22	1-Octadecanethiol	$C_{18}H_{38}S$	5.10
23	1-Methyl-4-(2-propenyl)-benzene	$C_{10}H_{12}$	3.01
24	2-Methyl-2-2-propenyl-benzene	$C_{10}H_{12}$	7.09
28	2,4-Dimethylpyridine	C_7H_9N	0.68
29	2,4,6-Collidine	$C_8H_{11}N$	0.39
30	Indole	C_8H_7N	0.44

Table 4
Toxic organic pollutants in the effluent of the EGSB reactor

	Species	Molecular formula	Relative content (%)
1	Valeric acid	$C_5H_{10}O_2$	25.2
2	Hexanoic acid	$C_6H_{12}O_2$	19.6
3	Heptanoic acid	$C_7H_{14}O_2$	12.3
4	Cyclopentanone	C_5H_8O	6.80
5	2,3-Dimethyl-2-cyclopenten-1-one	$C_7H_{10}O$	10.7
6	1,2-Benzenedicarboxylic acid dimethyl ester	$C_{10}H_{10}O_4$	2.25
7	1,2-Benzenedicarboxylic acid dibutyl ester	$C_{16}H_{22}O4$	3.04
8	Ethybenzene	C_8H_{10}	0.35
9	1.2-Dimethyl-benzene	C_8H_{10}	0.12
10	P-Xylene	C_8H_{10}	0.76
11	1-Ethyl-2-methyl-benzene	C_9H_{12}	1.06
12	1-Ethyl-4-methyl-benzene	C_9H_{12}	0.98
13	1.2.4-Trimethylbenzene	C_9H_{12}	1.11
14	4-Hydroxy-benzenesulfonic acid	$C_6H_6O_4S$	0.26
15	4-Methyl-octane	C_9H_{20}	0.40
16	1.2.3-Trimethyl-benzene	C_9H_{12}	1.10
17	1.3.5-Trimethyl-benzene	C_9H_{12}	1.60
18	Indane	C_9H_{10}	2.70
19	Phenylethylene	C_8H_8	2.30
20	2-Methyl-phenol	C_7H_8O	1.50
21	3-Methyl-phenol	C_7H_8O	0.43
22	4-Hydroxytoluene	C_7H_8O	0.37
23	2.6-Dimethyl-phenol	$C_8H_{10}O$	1.50
24	1-Octadecanethiol	$C_{18}H_{38}S$	1.20
25	1-Methyl-4-(2-propenyl)-benzene	$C_{10}H_{12}$	0.80
26	2-Methyl-2–2-propenyl-benzene	$C_{10}H_{12}$	0.86
27	2,4-Dimethylpyridine	C_7H_9N	0.22
28	2,4,6-Collidine	$C_8H_{11}N$	0.19
29	Indole	C_8H_7N	0.30

and C_i and C_e are the influent and effluent COD_{Cr} concentrations (mg/L), respectively, t_1 and t_0 are the day of measure time and the experiment start time (d), respectively.

The results showed that the sludge yield coefficients were all extraordinary low even negative (from −0.1 to 0.1 mg MLVSS/mg COD_{Cr}), with an average of 0.036 mg MLVSS/mg COD_{Cr}. The sludge production of this hybrid system was greatly reduced. And, granular sludge with high concentration of 40 g MLSS/L demonstrated high settling velocities resulting in good liquid separation. In summary, the hybrid system produced a small amount of excess sludge easy to be handled.

3.3. Methane yield

The methane production was measured during the whole experiment. The average rate of methane was ca 0.94 L/d, fluctuating between 0.84 and 1.77 L/d under the fluctuant influent conditions. However, methane yield was only about 0.21 mL CH_4/mg COD_{Cr} in the EGSB reactor, much lower than the theoretical yield (0.35 mL CH_4/mg COD_{Cr}) [18]. It may be due to the high concentration of refractory organic components and the restraining substances such as sulphate in the refining wastewater. Under anaerobic conditions, competitive reaction between the sulphate-reducing bacteria and methanogens was occurred, and the reductive products such as H_2S inhibited the growth of methanogenic bacteria.

Based on GC-MS analysis results of the EGSB reactor, the toxic pollutants are degraded to biodegradable products and the biodegradability of effluent is significantly improved. When it is limited, methane yield to a low level treating petroleum refining wastewater, the EGSB reactor can be kept in the hydrolysis and acidification stage by controlling the operating parameters. So, high molecular weight and refractory compounds can be broken down into small biodegradable molecules, the bio-degradability of petroleum refining wastewater consequently could be greatly improved.

4. Conclusions

A hybrid EGSB-BAF process can be used well to treat the petroleum refining wastewater with a high removal efficiency of oil, SS, COD and a low sludge yield. Nevertheless, the methane yield was only about 0.21 $\mathrm{mLCH_4/mgCOD_{Cr}}$ in the EGSB reactor, lower than the theoretical yields. A high efficient anaerobic process such as EGSB could be used as pre-treatment process by improving biodegradation performance of wastewater for the following aerobic biochemical treatment.

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