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H_2S removal in landfill leachate treatment using UASB reactor

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Abstract: Leachate from a sanitary landfill site in Chengdu, China is treated using a hybrid-UASB reactor at pilot scale. H_2S, resulting from the anaerobic bioconversion process of sulfate-reducing bacteria (SRB), inhibits the growth and activity of methane-producing bacteria (MPB) and poses serious problems of pollution, so $FeCl_3$ is used for H_2S removal. The results show that the system performs well in the treatment process. COD removal generally increases with the increase in the organic loading rate (OLR), while the sulfate removal decreases slowly. As the OLR is higher than 7 kgCOD/($m^3 \cdot d$), both COD and sulfate removal tend to be stable. When the reactor is operated at the design load of 9 kgCOD/($m^3 \cdot d$), COD and sulfate removal remain about 79% and 91%, respectively. At the same time, the percentage of COD removed by SRB (COD_{SRB}) also decreases from 8.9% to 4.0%. With $FeCl_3$ addition, COD removal increases to 83%, while sulfate removal and COD_{SRB} further decrease to 89% and 1.89%, respectively. According to the mass balance, nearly 82% of the sulfur is prevented from converting into H_2S. Moreover, when the $FeCl_3$ dosage is more than 1.6 g/L leachate, H_2S can be removed totally from the biogas. Therefore, the application of $FeCl_3$ for H_2S removal in leachate treatment using the UASB reactor is very suitable and viable.

Key words: landfill leachate; upflow anaerobic sludge blanket (UASB); H_2S; $FeCl_3$; sulfur balance

As a mixture of organic and inorganic compounds, as well as a mixture of pathogenic microorganisms, landfill leachate has a hazardous impact on the environment and human health. Though many treatment processes have been adopted to control the pollution caused by leachate, a combination of physical, chemical and biological methods has been gaining a wider popularity. The biological treatment method, especially the anaerobic technology, is mainly the first stage in an integrated process. Among others, the upflow anaerobic sludge blanket (UASB) reactor is regarded as one of the most successful and extensively applied systems due to its high treatment efficiency and short hydraulic retention time (HRT)[1].

However, the major problem of the anaerobic technology is the conspicuous end product hydrogen sulfide (H_2S) when there is sulfate in the influent. Microbial sulfate reduction is a conventional method of anaerobic digestion. During the oxidation of organic matter, specific bacteria using sulfate as the terminal electron acceptor produces H_2S as the final product, which can be described as[2]

$$SO_4^{2-} + \text{nutrients} + H_2O \rightarrow H_2S + HCO_3^- \tag{1}$$

This process is known as the dissimilatory sulfate reduction and the bacteria involved are known as sulfate-reducing bacteria (SRB). Simple substrates, such as hydrogen, lactate, ethanol, acetate, methanol and propionate, are often used as electron donors[3], and complex substrates, such as cellulose, starch and molasses, can also be used[4]. Not only does the end product (H_2S) inhibit the growth and activity of methane-producing bacteria (MPB), but it poses serious problems of odor, toxicity and corrosion. Some plants and projects using biogas as bioenergy have been shut down due to the corrosion problem of H_2S[5]. Furthermore, when burning with CH_4, H_2S exhausts in the form of SO_2, which is more dangerous than H_2S because of its detrimental role in the atmosphere. Therefore, it is necessary to remove H_2S prior to its use for the sake of safety, health and the environment.

H_2S can be removed through a physical or a chemical method such as the absorption and oxidation process[6]. The use of iron species for H_2S removal is a common and extensively documented practice[7-8], and the $FeCl_3$ solution has been used to remove H_2S in dairy manure treatment[9]. The main advantages of iron precipitation for H_2S removal include high removal efficiency, low sludge generation and nontoxic product formation[10]. During the treatment process, H_2S is absorbed and oxidized to elemental sulfur (S^0), and simultaneously Fe^{3+} is reduced to Fe^{2+} and precipitated in terms of FeS. In addition, part of the H_2S can also be precipitated with heavy metals in wastewater, which can be illustrated by

$$2FeCl_3 + 3H_2S \rightarrow 2FeS + S + 6HCl \tag{2}$$

$$H_2S + M^{2+} \rightarrow MS + 2H^+ \tag{3}$$

where M^{2+} represents divalent metallic cations such as Pb^{2+}.

However, there are few studies which have evaluated the process performance and behaviors of the UASB reactor when $FeCl_3$ is used for H_2S removal in landfill leachate treatment. Based on this background, this paper is conducted to test H_2S removal with $FeCl_3$ additions on a pilot scale. A specific focus is paid to evaluate the performance of the hybrid-UASB reactor, removal efficiency of the H_2S and sulfur balance during the process of leachate treatment.

1 Materials and Methods

1.1 Leachate analysis

The landfill leachate used in this paper is collected from the Chang'an sanitary landfill site, located in Chengdu,

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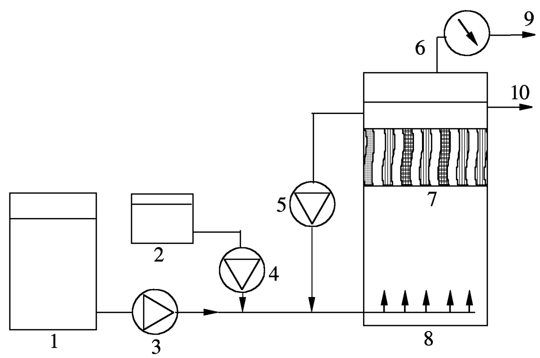
China. The main physicochemical characteristics of the leachate are shown in Tab. 1.

Tab. 1 Characteristics of influent leachate

Parameter	Mean	Range
pH	7.6	6.7 to 8.5
COD/($mg\cdot L^{-1}$)	18 300	11 850 to 24 870
BOD_5/($mg\cdot L^{-1}$)	6 810	5 270 to 8 350
BOD/COD	0.39	0.34 to 0.44
SS/($mg\cdot L^{-1}$)	3 860	920 to 6 800
NH_3-N/($mg\cdot L^{-1}$)	1 700	1 300 to 3 100
NO_3-N/($mg\cdot L^{-1}$)	138	96 to 180
TN/($mg\cdot L^{-1}$)	10 750	7 000 to 14 500
TP/($mg\cdot L^{-1}$)	147.5	122 to 173
SO_4^{2-}/($mg\cdot L^{-1}$)	2 120	1 790 to 2 450
As/($mg\cdot L^{-1}$)	0.04	0.02 to 0.06
Cd/($mg\cdot L^{-1}$)	0.045	0.03 to 0.06
Cr/($mg\cdot L^{-1}$)	0.57	0.35 to 0.79
Cu/($mg\cdot L^{-1}$)	0.48	0.39 to 0.57
Hg/($mg\cdot L^{-1}$)	0.02	0.01 to 0.03
Ni/($mg\cdot L^{-1}$)	0.955	0.81 to 1.10
Pb/($mg\cdot L^{-1}$)	0.77	0.39 to 1.15
Zn/($mg\cdot L^{-1}$)	13.915	10.95 to 16.88

1.2 Reactor setup

Fig. 1 shows a schematic diagram of the experimental setup used in this paper. The pilot scale hybrid-UASB reactor is cylindrical in shape and fabricated using carbon steel with an internal diameter of 1.95 m and an overall height of 3.5 m. The total and active volumes of the reactor are 10.6 and 9.0 m^3, respectively. Two peristaltic pumps are used separately for leachate feeding and $FeCl_3$ addition, and the velocity of up-flow liquid in the reactor is kept at 0.52 m/h through a recirculation pump. In addition, the influent flow and gas production are measured by using an electromagnetic meter and a wet gas meter, respectively.



1—Feed tank; 2—$FeCl_3$; 3—Influent pump; 4—Adding pump; 5—Recirculation pump; 6—Wet gas meter; 7—Carrier; 8—Hybrid-UASB reactor; 9—Biogas; 10—Effluent

Fig. 1 Schematic diagram of the hybrid-UASB system

The soft polyurethane belts (0.05 cm × 2 cm × 100 cm) are perpendicularly suspended in the reactor as carrier material. The top of the belts is fixed to a metal net which is parallel to the cross section of the reactor and the lower end of the belts is connected to counterbalance weights, which can make the belts swing slowly in up-flow liquid. Besides being a place where microbes attach and grow, the belts act similarly to a three-phase separator. The gas-carried biomass, after crashing onto the carrier, has a chance of falling back down to the sludge bed. The higher the carrier is in the packing zone, the less chances the biomass can be easily washed out.

1.3 Inoculation and operation

Digested slurry collected from a sewage treatment plant located at Wuhou district, Chengdu, China is used as the inoculum. The total solids (TS) and volatile suspended solids (VSS) content of the inoculum are found to be 60 230 and 10 750 mg/L, respectively. This reactor setup is placed inside a temperature-controlled room maintained at 35 ℃. 30% (wt) $FeCl_3$ solution is introduced into the UASB reactor from 101 to 191 d.

1.4 Analytical methods

Parameters, such as chemical oxidation demand (COD), SO_4^{2-}, H_2S and $FeCl_3$, are checked every day. COD and SO_4^{2-} are determined using colorimetric tests on an HACH-DR 2800 photometer. $FeCl_3$ manufactured by Guotao Chemical Company (Chengdu, China) is of industrial grade and measured by the phenantroline method (Standard Methods, 1998)[11]. Biogas is analyzed via gas chromatography (BFRL-SP-2100, Beijing Analytical Instrument Factory, China).

1.5 Calculation methods

1.5.1 H_2S removal efficiency (RE) and percentage of COD removed by SRB (COD_{SRB})

RE(%) is calculated by

$$RE = \frac{C_0 - C}{C_0} \times 100 \tag{4}$$

where C_0 and C are the concentrations of H_2S in biogas before and after $FeCl_3$ addition, respectively. COD_{SRB} is obtained from the amount of sulfate reduced and the total amount of COD removed[12].

1.5.2 Sulfur mass balance

According to Eqs. (1), (2) and (3), the influent sulfur is distributed into different sulfur species (Ⅰ to Ⅴ) through the UASB process (see Fig. 2). Parts Ⅰ and Ⅱ are directly measured, and parts Ⅲ and Ⅳ are calculated based on Eq. (2) and the consumed $FeCl_3$, so part Ⅴ can be deduced from a mass balance of all other sulfur species.

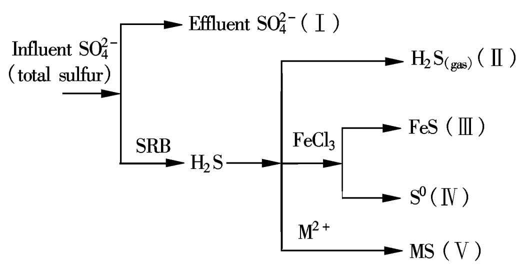


Fig. 2 Distribution of sulfur during the UASB process

2 Results and Discussion

2.1 Performance of UASB reactor

The hybrid-UASB reactor is operated continuously for a period of 191 d. Figs. 3 and 4 give the process performance of the UASB reactor, in terms of removal efficiency and loading rate, the profiles of COD and sulfate are demonstrated, respectively.

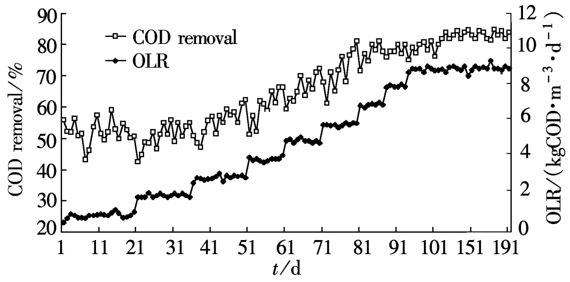


Fig. 3 Variation of COD removal efficiency and organic loading rate (OLR) with operation time

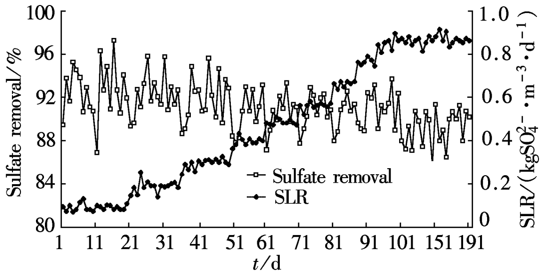
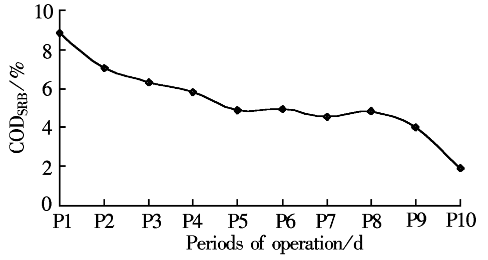


Fig. 4 Variation of SO_4^{2-} removal efficiency and sulfate loading rate (SLR) with operation time

When landfill leachate is introduced into the UASB reactor, organic matter is removed via both sulfate reduction and methanogenesis. In the first period (1 to 50 d), with OLR increasing from 0.5 to 3 kgCOD/(m^3·d), the effluent COD increases to above 7 200 mg/L while COD removal varies from 43% to 62%. Methane production is suppressed in this period, and the methane content of biogas is lower than 40%. The biomass in the reactor seems not to be able to handle the increased OLR. However, at 0.08 to 0.36 $kgSO_4^{2-}$/(m^3·d) loadings, the sulfate removal is relatively higher (87% to 93%), and a certain amount of COD elimination (6.3% to 8.9%) is accomplished through sulfate reduction (see Fig. 5). It is believed that the activity of SRB inhibits the growth and activity of MPB to some extent, though the inhibition is not strong due to the higher COD/SO_4^{2-} ratio of 8. Generally, there are three major mechanisms of MPB inhibition by SRB: 1) Sulfide toxicity; 2) Competition for the substrate; 3) Precipitation of trace elements by sulfide. The direct toxicity correlates with the amount of free hydrogen sulfide, especially at alkaline pH values[13].



P1—1 to 20 d; P2—21 to 35 d; P3—36 to 50 d; P4—51 to 60 d; P5—61 to 70 d; P6—71 to 80 d; P7—81 to 87 d; P8—88 to 93 d; P9—94 to 100 d; P10—101 to 191 d

Fig. 5 COD_{SRB} in different periods of the operation

In the following period (51 to 80 d), when OLR increases from 3 to 6 kgCOD/(m^3·d), COD removal increases more quickly than that in the first 50 d, indicating the improvement in microbial activity of the population. Additionally, at each stepwise increase in OLR, COD removal increases correspondingly until the reactor accommodates to the existing OLR. During the period of 81 to 100 d, however, COD removal does not change much, remaining at about 79%, though OLR increases from 7 kgCOD/(m^3·d) to the design load 9 kgCOD/(m^3·d). On the other hand, when SLR increases from 0.4 to 0.99 $kgSO_4^{2-}$/(m^3·d) during the period of 51 to 100 d, the sulfate removal remains steady (around 91%). The results indicate that the competition process between SRB and MPB seems to reach a state of equilibrium.

During the last period (101 to 191 d), the UASB reactor is operated at design load for H_2S removal tests. H_2S obtained from SRB, mainly in the undissociated form, can inhibit the activity of MPB and SRB, simultaneously[14]. However, $FeCl_3$ addition relieves this kind of inhibition and results in the increment of COD removal efficiency which attains a level approximate to 83%. The decrease of COD_{SRB} to 1.89% also indicates that MPB is more competitive during this period. However, $FeCl_3$ retards the dissimilatory sulfate reduction owing to its Lewis acid (electron deficient) nature because during metabolism, it also accepts electrons along with sulfate ions[9]. This is the reason why sulfate removal decreases slightly (to 89%).

Throughout the experiment, both total gas and methane production generally increase with the increase in OLRs, though there are small fluctuations sometimes. The increase also indicates that the sludge in the reactor becomes more active, and, hence, can handle the greater throughput of the leachate.

2.2 H_2S removal using $FeCl_3$

Tab. 2 shows the results of H_2S removal using $FeCl_3$. In most cases, the removal efficiency is 100%, which is better than what has been reported[15]. With $FeCl_3$ addition of 1.6 g/L and beyond this dosage, H_2S can be removed completely. The optimum $FeCl_3$ dosage in this study is defined as the lowest $FeCl_3$ dosage at which the maximum H_2S removal efficiency is achieved. So we think the dosage of $FeCl_3$ added should not be less than 1.6 g/L under the experimental conditions.

With regard to methane content and biogas yield, it can be seen from Fig. 6 that they both rise with the increase in $FeCl_3$ dosage, reaching the maximal value, and then decrease with further $FeCl_3$ addition. In fact, iron content itself is not toxic to methanogens, but in high concentration, it affects the activity of microorganisms indirectly as it forms $Fe(OH)_3$ by hydrolysis in medium[9], which results in the increase in pH values and the amount of free hydrogen sulfide, and thereby affects the specific methanogenic activity and biogas yield. This is the reason why the methane content and the biogas yield decrease with an addition of $FeCl_3$ beyond 1.6 and 1.4 g/L, respectively. On the other hand, as a kind of coagulating agent, $FeCl_3$ can make the biomass coagulate and, hence, improve the activity of the microor-

Tab. 2 Concentration and removal efficiency of H_2S with $FeCl_3$ dosage

$FeCl_3$/ ($g \cdot L^{-1}$)	Parameter	Operation time/d					
		101 to 115	116 to 130	131 to 145	146 to 160	161 to 175	176 to 191
0.6	H_2S/($mg \cdot L^{-1}$)	519	557	616	772	631	600
	RE/%	68	67	65	59	64	67
0.8	H_2S/($mg \cdot L^{-1}$)	308	338	388	546	403	345
	RE/%	81	80	78	71	77	81
1.0	H_2S/($mg \cdot L^{-1}$)	0	118	164	320	193	109
	RE/%	100	93	91	83	89	94
1.2	H_2S/($mg \cdot L^{-1}$)	0	0	41	94	32	0
	RE/%	100	100	98	95	98	100
1.4	H_2S/($mg \cdot L^{-1}$)	0	0	0	13	0	0
	RE/%	100	100	100	99	100	100
1.6	H_2S/($mg \cdot L^{-1}$)	0	0	0	0	0	0
	RE/%	100	100	100	100	100	100
1.8	H_2S/($mg \cdot L^{-1}$)	0	0	0	0	0	0
	RE/%	100	100	100	100	100	100

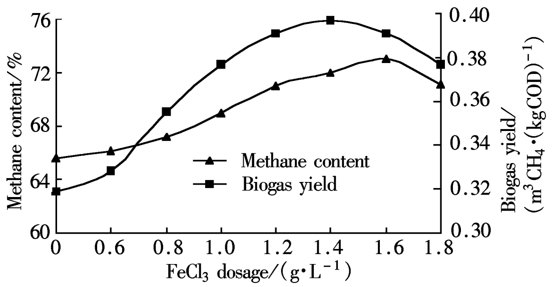


Fig. 6 Variation of methane content and biogas yield with $FeCl_3$ dosage

ganism, which contributes to greater production of methane and better anaerobic treatability[16].

In this study, 100% H_2S removal efficiency has been achieved by using a small quantity of $FeCl_3$, which is practical and economical compared to caustic scrubbers that are expensive and are used in large quantities. Therefore, the technique of biogas desulfurization using $FeCl_3$ is very suitable and feasible.

2.3 Sulfur mass balance

The distribution of the sulfur in the UASB process is demonstrated in Fig. 2. Except discharge in effluent(Ⅰ), most of the sulfate is reduced to H_2S by SRB, and further distributed into four parts(Ⅱ, Ⅲ, Ⅳ and Ⅴ) after $FeCl_3$ addition. Tab. 3 gives the sulfur balance calculated for different periods of operation.

Tab. 3 Sulfur balance in the UASB reactor and COD_{SRB}

Period/d	Total sulfur/ ($g \cdot d^{-1}$)	Sulfur balance/%					COD_{SRB}/%
		Ⅰ	Ⅱ	Ⅲ	Ⅳ	Ⅴ	
101 to 115	571.78	14.42	1.61	51.85	27.48	4.64	1.90
116 to 130	638.32	9.61	2.00	54.18	29.13	5.08	1.56
131 to 145	714.15	8.26	4.66	48.58	29.02	9.48	1.78
146 to 160	1 076.78	7.43	2.36	55.17	30.07	4.97	2.53
161 to 175	756.68	8.09	1.48	49.16	36.17	5.10	1.79
176 to 191	684.98	11.99	0.93	55.56	28.72	2.80	1.78
Overall	740.45	10.80	2.17	52.42	29.26	5.35	1.89

It can be inferred from Tab. 3 that the overall sulfate removal is about 89% from 101 to 191 d, which is slightly lower than that without the $FeCl_3$ addition (see Fig. 4). Owing to the $FeCl_3$ addition, nearly 82% of sulfur (parts Ⅲ and Ⅳ) is prevented from converting into H_2S (gas) which may cause potential air pollution. There is still 2.17% of sulfur (part Ⅱ) released into the air in terms of H_2S(gas); however, if the $FeCl_3$ dosage is more than 1.6 g/L, H_2S (gas) can be removed completely from the biogas (see Tab. 2).

The main product of sulfur removed is FeS, which not only precipitates in the reactor and prevents stripping of H_2S to biogas, but also offers protection against oxygen contamination, which is the basic requirement of this anaerobic bioconversion technology[9]. Furthermore, FeS is insoluble and nontoxic to the anaerobic digestion process, which plays an important role in the performance of the UASB reactor.

In most cases, with an overall averaged value of 1.89%, COD_{SRB} is lower than that during the period of 1 to 100 d (see Fig. 5). This can be explained by the increment of methanogenic activity; i.e., after an adequate acclimation period, the active biomass is adaptable to the operational conditions. Especially, $FeCl_3$ removes most of the H_2S and alleviates the direct toxicity to the MPB, which contributes to the improvement of methanogenic activity and performance of the UASB reactor.

3 Conclusion

Landfill leachate is a potential pollution source for environmental and human health, and the UASB reactor is most extensively used in leachate treatment. However, as the final product of this anaerobic process, H_2S has a negative effect on treatment performance and causes secondary pollution. In this paper, a chemical process for H_2S removal using $FeCl_3$ is performed on a pilot scale. The results show that the hybrid-UASB reactor can perform well in leachate treatment. At the design load of 9 kgCOD/(m^3·d), COD and sulfate removal efficiency remain above 79% and 91%, respectively. $FeCl_3$ alleviates the inhibition of H_2S, increases the COD removal to 83%, and thereby contributes to the high performance of the UASB process. Furthermore, the complete removal of H_2S from the biogas is achieved when the $FeCl_3$ dosage is more than 1.6 g/L. Therefore, the technique of biogas desulfurization using $FeCl_3$ is very suitable and viable.

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UASB 反应器处理垃圾渗滤液过程中的 H_2S 去除

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摘要: 以成都市某生活垃圾填埋场渗滤液为研究对象, 采用 UASB 反应器对其进行处理. 由于渗滤液中的 SO_4^{2-} 被硫酸盐还原菌(SRB)转化成 H_2S, 会造成污染并对厌氧反应中的产甲烷菌(MPB)产生抑制作用, 因此添加 $FeCl_3$ 溶液对 H_2S 进行去除. 实验结果表明,该工艺运行效果良好. 随着进水负荷(OLR)的增加, COD 去除率逐渐增加而硫酸盐去除率则缓慢下降, 当 OLR 高于 7 kgCOD/(m^3·d)时, 二者趋于稳定. 当系统达到设计负荷 9 kgCOD/(m^3·d)时, COD 和硫酸盐的去除率分别稳定在79%和91%, SRB 所去除的 COD(COD_{SRB})也由启动初期的 8.9% 下降到 4.0%. 添加 $FeCl_3$ 后, COD 去除率上升并稳定在 83%, 而硫酸盐的去除率和 COD_{SRB} 则分别进一步降为 89% 和 1.89%. 质量平衡显示, $FeCl_3$ 去除了近 82% 的硫, 并且当 $FeCl_3$ 用量达到 1.6 g/L 时, 产气中的 H_2S 被完全去除. 因此, $FeCl_3$ 用于去除 UASB 处理垃圾渗滤液过程中产生的 H_2S 是切实可行的.

关键词: 垃圾渗滤液; 升流式厌氧污泥床(UASB); H_2S; $FeCl_3$; 硫平衡

中图分类号: X506