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Advanced anaerobic expanded granular sludge bed (AnaEG) for treatment of coal gasification wastewater

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ABSTRACT

A state-of-art technology, advanced anaerobic expanded granular sludge bed (AnaEG) was developed for the anaerobic treatment of coal gasification wastewater, a typical industrial wastewater having poor biodegradability and high toxicity. Three batch tests were conducted to check the efficiency of the reactor, 330 days using acidification, hydraulic retention time (HRT) of 45 hours. With the influent conc. of COD 1400 mg/L, total phenol 320 mg/L, volatile phenol 150 mg/L, the effluent COD, total phenol, volatile phenol could decrease to 800 mg/L, 200 mg/L and 40 mg/L. AnaEG shows, COD removal efficiency 50%, with loading rate of 0.806(kg COD/m^3.day) and removal rate of 0.357(g COD/day), total phenol removal efficiency 50%, volatile phenol removal efficiency 80%, respectively. Besides being able to remove 70-95% of organic matter from the wastewater, this technology generates less sludge. Finally, scanning electron microscopy (SEM) revealed has long-chain filamentous bacteria; coccus and rod-shaped bacteria, were dominant microorganisms. CH_4 production rate of 227.23 (ml CH_4/L.d) during loading rate of 626.25 (mg COD/L-d) and removal rate 87.68 (mg COD/L-d) was observed. Significant reduction in amount of sludge produced, so lower sludge management cost. It does not have to reflux; this equipment is simple as less accessory equipment, low power consumption and high effluent quality. All the results demonstrate that AnaEG could be used efficiently for treatment of coal gasification wastewater containing high COD, phenol concentration.

Keywords: Coal gasification wastewater, total phenol, volatile phenol, sludge, Advanced Anaerobic Expanded Granular Sludge Bed (AnaEG)

1. Introduction

For organically polluted industrial waste streams anaerobic wastewater treatment is considered the most cost-effective solution [1] and due to increasing energy prices it has gained interest. Anaerobic treatment processes are known for the unique ability to convert highly objectionable wastes into useful products [2]. Treatment of industrial wastewater in anaerobic bioreactors has grown especially its importance since the introduction of the Up flow Anaerobic Sludge Blanket (UASB) reactor about more than 30 years ago [3]. UASB reactors are worldwide still the most common even though a variety of additional anaerobic bioreactor designs has now been developed [4].

Expanded Granular Sludge Blanket (EGSB) and Internal Circulation (IC) reactors are currently replacing more conventional UASB systems due to improved performance and economic efficiency [4]. EGSB systems have a comparable design as UASB reactors, but contain an expanded granular sludge bed allowing more circulation and interaction between the micro-organisms and the organic compounds in the sludge granules.

The UASB system dominated the industrial wastewater treatment worldwide in the 1980’s and before 1997. All anaerobic reactors that were sold during 2002 to 2007 worldwide EGSB type reactors were 52% while UASB reactors only 34% [4-6]

The present study combines the advantages of UASB and EGSB technology. The simplicity and high treatment efficiency of the AnaEG reactor in comparison to EGSB is shown in Table 1. It's a hybrid reactor. AnaEG is the third generation of high efficiency anaerobic biological process invented[7] by our research group it is a further step as an advanced anaerobic system. AnaEG is a state-of-the-art technology for treatment of organic or biological waste.

First of all, in anaerobic treatment process (AnaEG) the hydrolysis acidification and methanogenic process are placed in one unit as seen in Fig. 1. The bottom section (1/5 -1/3 height of the sludge bed) is mainly in acidogenesis condition，while above it，the upper section is methanogenesis zone; the effluent of anaerobic reactor need not to be recycled back to influent to maintain a high upward velocity, and the wastewater flows upward in a plug flow pattern, meanwhile the organic matters in wastewater is decomposed by acidogenesis to methanogenesis process in upward direction, which achieves a two-phase anaerobic process in one reactor. Alkalinity requirement in operation may not be needed, and the desired pH range is relatively wide, generally between 6 and 9.

During well-dispersed influent flows upward, the organic matter was degraded and biogas (mainly methane) is produced, which causes the granular sludge bed to expand. The grade of expansion increases from bottom to top, and creates the fluidization condition at the upper section.Therefore, in our process it does not have bad odour in the hydrolysis pool as process takes place in an enclosed reactor, significantly saving the land usage area as treatment plant is smaller, strict control system over influent pH.

The biggest feature is that it has no water cycle. Water in the reactor was pushed forward by the streaming flow pattern. The reactor can be divided into three parts as the inlet area, reactor area, gas-solid-liquid separation area. Wastewater enters the reactor from the reactor base and flows through the reactor in an upward direction. As it flows through the reactor, organic matter was biodegraded in an anaerobic process. Organic acid and methane gas was formed in two different layers in the AnaEG™ reactor.

The AnaEG™ treatment system removes 70% – 95% of the organic matter from the wastewater. A simple aerobic process easily removes remaining organic matter.

It was designed to overcome the shortcomings of the existing anaerobic reactor and then it was employed for a case study to treat the coal gasification wastewater which is characterized by complicated composition, high concentrations of pollutants and high toxicity [8-13] The organic components present in CGW wastewater mainly include volatile phenols, polyphenols, polycyclic aromatic hydrocarbons (PAHs), aromatic and heterocyclic compounds, most of which are toxic, mutative, carcinogenic, teratogenic, and may produce long-term adverse effects in the environment [9, 14-19]. Therefore, in China as coal gasification has become an efficient way to provide clean energy in recent years; the pollution caused by it has brought a serious problem faced by China [18, 20, 21].

In this paper, AnaEG was employed to evaluate its effectiveness and feasibility for the treatment of coal gasification wastewater.Effect of AnaEG operating parameters were investigated in detail. The variations of pH and methane production were also determined. Finally, the morphological and microbial structure of the anaerobic bacteria was studied during the course of coal gasification wastewater treatment at stable state. This study will not only help to understand the process and performance of the AnaEG for treatment of coal gasification wastewater treatment, but also contribute to provide information for treatment of other high strength industrial wastewater by AnaEG.

2. Materials and method

2.1 Characteristics of coal gas wastewater

The coal gasification wastewater was received from the Coal Long Hua Harbin Coal Chemical Industry Co. Ltd, Harbin, China. The raw water had pH 8.5-9. COD conc. 4400mg/L, BOD conc. 700 mg/L, Total phenol (TP) 950 mg/L, Volatile phenol (VP) 450 mg/L, ammonia conc. 300 mg/L. In addition, the BOD/COD ratio was 0.15. The high conc. of ammonia and phenol had an inhibitory effect on biological treatment [22, 23] but the AnaEG treatment has the capability to effectively treat a wide range of wastewater and is particularly efficient for problematic wastewater with very high organic content.

2.2 Pilot scale anaerobic reactor

The experiments were performed in the apparatus as shown in Fig.1. It consist of inlet, outlet zone, a control unit, a three phase separator, a gas dome, a expanding granular sludge layer, a water dispensing unit, and a main body for wastewater treatment. The main body was made of transparent rigid Plexiglas with an inner diameter of 100 mm and a height of 1500mm. The effective volume of the reactor was 13.4L, its shape was cylindrical. It does not had to reflux; this equipment is simple as less accessory equipment, low power consumption and high effluent quality. The grade of sludge expansion in AnaEG depends upon the influent and the biogas production, and automatically adjusted by loading rate. Circulating pump is not required for maintaining the expansion purpose. Therefore, power consumption is smaller. It is a plug-flow reactor, so its adaptability of shock-load is stronger, Top phase separation zone is the three phase separation (gas-liquid-solid separation). The reactor was operated under mesophilic conditions (35^{o}C) and the temperature was maintained by the recycling of hot water by a thermostatic water bath. The CO_2 and H_2S and other acid gases will be absorbed by the base liquid.

2.3. Pre-start up operating conditions

The inoculated sludge was taken from sewage wastewater treatment plant in Wuxi Ashimura, china. Digested sludge inoculum anaerobic reactor effective volume of 40%. The test water for preoperational stage was tap water and CGW (1:1). The tap water was added in order to lower down the COD of coal gasification wastewater.

2.4. Start-up and operating conditions

Inoculated anaerobic granular sludge was made in our own Laboratory (inoculum: anaerobic reactor effective volume of 40 %).The main aim of start-up was to develop the most appropriate microbial culture for wastewater treatment, so a certain amount of glucose was added for the activation of anaerobic microorganism, after that gradually the amount of glucose was reduced and ultimately without the glucose.

AnaEG reactor was run for 330 days. Start-up was divided into three stages, first start-up run for 87 days the HRT was 96hr, the wastewater flow efficiency was control at 3.4L/d. according to the amount of glucose it was divided into five phases, gradually the amount of glucose was reduced, inorder to make the anaerobic microorganism grow and adapt the coal gasification wastewater environment. The second stage was to reduce HRT and stability (88-200 days) it was operated for 110 days. HRT was reduced from 96hr to 48hr and to achieve stable running for about 60 days. The III stage was the second start-up with stabilization phase (201-333 days) HRT was 48hr and it was operated for 133 days.

2.5. Analytical methods

Biogas production was measured daily with a wet glass flow meter making correction for atmospheric pressure and temperature. Methane concentration were determined by GC2010A gas chromatography (shimadzu, Japan) with stainless steel column (300cm x 0.3 cm) packed with active carbon (30-60 mesh) using thermal conductivity detection (TCD).

The effluent and influent pH values were measured using a pH meter; COD, Volatile phenol, total phenol analyses were carried out according to the standard procedures [24]. The concentrations of total phenols and volatile phenols were measured by the titration method [25].To further understand the nature of wastewater GCMS 2.0 (Shimadzu, Japan) was used. SEM (HITACHI TM3000 Tabletop Microscope)

3. Results and discussion

3.1. Pre-start up operational stage of anaerobic reactor

The pre-start up stage was run for 148 days, during the operation (1-39 days and 40-62 days) the tap water was used as dilution water. For 63-104 days the reactor showed removal efficiency for COD 40-45%, total phenol 50-55%, and VP 70-85%. When the reactor operated from 105-145 days , COD removal was about 45%, total phenol removal was 30%, VP removal efficiency was 50-55% (aerobic effluent was taken as dilution water). TP was controlled at 300-350mg/L, pH at 7.0-7.5 for the further stage of anaerobic influent. Thus when the raw water was diluted with tap water, anaerobic reactor performance was excellent.

3.2. Start-up stage of AnaEG

For 87 days, anaerobic reactor first start-up stage, was divided into five phases of operation according to the amount of glucose. With HRT remained at 96hr, operating time of each stage and glucose amount are shown in Table 2 and the degradation trends of COD and COD removal changes, TP removal, VP removal and changes in pH are shown in Fig. 2.

In first phase of the start-up stage for 34 days due to the higher seeded sludge activity (adding a sufficient amount of metabolites as glucose), initially the total COD removal was higher 73%, CGW COD removal 54% , total phenol removal efficiency was about 40 %, VP removal efficiency was about 20%. But with the extended operatinal time, highly unfavourable toxicity of coal gasification wastewater effluent hinders the removal efficiency of COD, total phenols and volatile phenol, the total COD removal efficiency decreased to about 40%, coal gasification wastewater COD removal efficiency dropped to about 10% , total phenol removal efficiency decreased to about 20% , removal of volatile phenol decreased to about 20%.

For the second phase (35-54 days), glucose amount was reduced to 800 mgCOD/L, COD and phenol removal efficiency gradually increased, the total COD removal was 51.5%, coal gasification wastewater COD removal efficiency was 24%, TP removal efficiency was 34% , VP removal efficiency was 37.8% .Glucose amount was reduced to 500mgCOD/L in the third phase (55-65 days), the amount of glucose was further reduced, a brief transient inadaptability of anaerobic microorganisms occurs but they soon resumed their activity. The total COD removal was 47%, coal gasification wastewater COD removal efficiency slightly increased to 29%, total phenol removal efficiency was 24%, volatile phenol removal efficiency 27%.

The fourth phase (66-76 days), the amount of glucose was further reduced to 300mgCOD/L, COD removal efficiency was reduced after showing a brief upward trend, the total COD removal efficiency was 47%, compared with the previous glucose lowering, coal gasification wastewater COD removal efficiency from 29 % to 37%. After a brief decline phenol removal also showed a gradual upward trend, the TP removal efficiency was 30%, VP removal efficiency was 30%. The fifth phase (77-87 days) glucose was not added so the microorganism cannot behave well as the removal of COD was very less but after a short period of time they regain and the removal of organic matter gradually increased.

Fig. 2 (e) and Table 2, the first start-up stage the reactor run in the normal range of basic pH value. Only four times the system observed less than 6.8 pH value but after a timely adjustment of the reactor quickly returned to the normal range.

After 87 days of operation, the successful completion of the first start-up stage takes place. HRT was 96hr from the start till the end of stage. With the decrease of glucose amount, coal gasification wastewater COD removal reached a gradual upward trend, indicating that anaerobic microorganisms continue to be domesticated. Effluent COD was about 680mg/L, COD removal efficiency was about 50%; total phenol concentration was about 170mg/L with removal efficiency about 44%; volatile phenol concentration about 40-60mg/L, with removal efficiency of about 50-70%

3.3. Increasing system loading and stabile running stage (88-200 days)

At this stage, the reactor was running for 123 days. Since the removal ratio of organic compounds in anaerobic systems have achieved relatively high levels, two increasing loads was carried out at this stage, from first start-up stage of 96hr HRT was reduced to 78hr, running for 30 days (88-117 days). In the second loading stage (118-142 days) HRT was further shorten to 48hr, close to the optimum operating parameter obtained from shaker test. From 143-200 days, the loading was maintained and the reactor performed stably. Fig. 3 (a) and Table 2 shows that the influent of COD was between 800-900mg/L (fluctuates). COD removal efficiency was in decreasing trend but still more than 30% in first increase of loading. This indicates that the increase of loading had some effect on anaerobic microorganisms but did not have a serious impact on the activity of anaerobic microorganisms it can be overcome by acclimatization. The COD removal ratio was in between 30-40 %. During the stable operation for 53 days, the effluent of anaerobic process exhibited a decreasing trend. Between 47 and 38 days, the effluent COD was 800mg/L or less.

From Fig. 3 (b), 3(c) and Table 2, TP conc. was 200-260mg/L; VP fluctuated between 100-140mg/L after increasing the loading. During the stable running TP, VP conc. showed a degradation trend. At the first half stage TP conc. was maintained at

170-200mg/L, VP 80-130mg/L. In the latter half, TP concentration was only 150mg/L, VP 20-40mg/L with removal efficiency 50% and 70%, respectively.

3.4. Biogas and Methane production

Fig. 4 (a) shows the biogas production (CH_4 60%, CO_2 40%, H_2S <1%). With the continuous reduction of glucose amount (Table 2), biogas production was gradually reduced and biogas generation was less than 0.4L/d as the glucose was not added. Fig. 4 (b) shows the methane production rate during the course of the first start-up stage of the reactor. The reactor showed CH_4 production rate of 227.23 (ml CH_4/L.d) during loading rate of 626.25 (mg COD/L-d) and removal rate 87.68 (mg COD/L-d). The increase of CO_2 in biogas indicates that the acidifying microorganisms are prevailing on the methanogens which may leads to volatile fatty acids (VFA) accumulation. The coal gasification wastewater (poor biodegradability) has low B/C ratio (0.15), high conc. of phenol, ammonia and other refractory materials which exhibit an inhibitory effect on the anaerobic bacteria, which results in a low bio gas production rate.

3.5. Anaerobic reactor second start-up and stable running stage

During the wastewater treatment, there may be circumstances where, for various reasons waste water treatment system had to stop (running). In order to understand the influence of stopping the anaerobic reactor, we stopped the entire wastewater treatment system for 10 days, and after 10 days second start-up was done. Since the hydrolysis acidification process was adopted in anaerobic treatment system. We maintained the previous load of 48 HRT in the second start-up. Fig. 5 and Table 2 shows that during the first 10 days of second start-up stage as the reactor was stopped for more than one week it had some adverse effects the removal ratio of COD, TP, VP removal efficiency were very low ,10-20 %, but had a tendency to recover .

As shown in Table 2 and Figure 5 (a) , COD removal ratio increased to more than 30% after 10 days of recovery; After 20 days ,COD removal ratio recovered to 40% , and showed a increasing trend ; effluent COD conc. maintain at about 500-600mg/L. Fig. 5(b) and Table 2, On the 15th day of second start-up stage, TP removal ratio hasd been recovered to 40-50%, TP effluent conc. was about 100-130mg/L. Degradation of volatile phenols and total phenols roughly the same as shown in Fig. 5(c) and Table 2 , at the 18th day its removal restored to about 70% , VP effluent conc. was about 15mg/L. The maximum COD, total phenol, volatile phenols removal rate were found to be 0.483 g COD/L.day, 0.08335 g/L.day, 0.063 g/L.day, respectively, for total loading rate of 0.6835 kg COD/(m^3.day), total phenol loading rate of 0.151 kg/(m^3.day) and volatile phenol loading rate of 0.0725 kg/(m^3.day). Thus can be considered, the second anaerobic reactor start-up takes about 22 days, its anaerobic effluent will meet the water requirements of the influent of subsequent aerobic biological treatment.

3.6. Microbiological analysis of anaerobic sludge

The microbial composition of anaerobic bioreactors is rather complex [3]. In order to observe the microbial structural characteristics in the anaerobic reactor, the internal and external structures of the sludge was analyzed through SEM after 128 days of running shown in Fig. 6. As can be seen from the SEM, There are mainly some micrococci; filamentous bacteria can be seen they intertwined randomly throughout the cross-section. Fig. 6a, Fig. 6 b Zoogloea shape, the internal structure of bacteria is not obvious. According to the SEM (×2000 times), Figure 6c, 6d shows there are long-chain filamentous bacteria, coccus and rod-shaped bacteria, presenting typical shapes of acid-producing bacteria. Meanwhile, there are some filamentous bacteria and bacillus intertwined together. Many colonies consisting of cocci and bacillus were also observed.

3.7. Advantages of AnaEG reactor

It has several key benefits

- Capable of treating a wide range of wastewater (even with high organic content)
- High removal rate for organics
- does not require oxygen supply in the treatment process
- Produces large amount of methane – generate up to 10,000 kJ of energy for every 1 kg of COD removed
- Significant savings in energy costs
- Significant reduction in amount of sludge produced – lower sludge management cost as the removed COD was mainly converted into CH_4 in AnaEG, while the amount of COD which is utilized for the multiplication of anaerobic bacteria

generally accounts for 10% of the COD removed. During more than 200 days of continuous running, the sludge quantity in the bioreactor does not show obvious increase.

- Effectively reduced odour production as process takes place in an enclosed reactor
- Increased capacity for organic volumetric load
- It combined with perfect principles of modern reactor engineering theory and anaerobic microbiology.

Conclusions

Anaerobic treatment plays an important role both in treatment and optimization treatment efficiency of coal gasification wastewater. The results shows that start-up time was 90 days when we add glucose as co-metabolite and the second start-up time was 20 days without metabolites. During the stable running of anaerobic reactor its COD removal efficiency reaches 50 %, TP removal ratio also reached 50% and the TVP removal efficiency reach to 80%. The SEM result shows that the anaerobic sludge has long-chain filamentous bacteria, coccus and rod-shaped bacteria, presenting typical shapes of acid-producing bacteria and methanogens leading to efficient methanogentic activity. AnaEG has distinct advantages over other systems currently available because of its compact design, which occupies a relatively small footprint. It boasts the ability to work on higher organic loading rates and variable hydraulic loads. The system also does not emit noise or odour, and the biogas produced can be captured and converted into energy, hence reducing the emission of greenhouse gases to the atmosphere

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Figure Captions

Figure 1. **S**chematic diagram of experimental apparatus of anaerobic reactor (AnaEG)

Figure 2. Anaerobic reactor initialization phase degradation trends (a) COD degradation trends (b) COD removal efficiency (c) Total phenol (d) Volatile phenol (e) pH variations.

Figure 3. Anaerobic system loading trends during stable running phase (a) COD degradation (b) Total phenol degradation (c) Volatile phenol degradation

Figure 4. (a) Biogas production (b) Methane production

Figure 5. Anaerobic reactor second start-up and stable running (a) COD degradation trends (b) Total phenol degradation trends (c) Volatile phenol degradation trends

Figure 6. Anaerobic sludge scanning electron micrograph (a), (b), (c) and (d) filamentous bacteria and bacillus intertwined together.

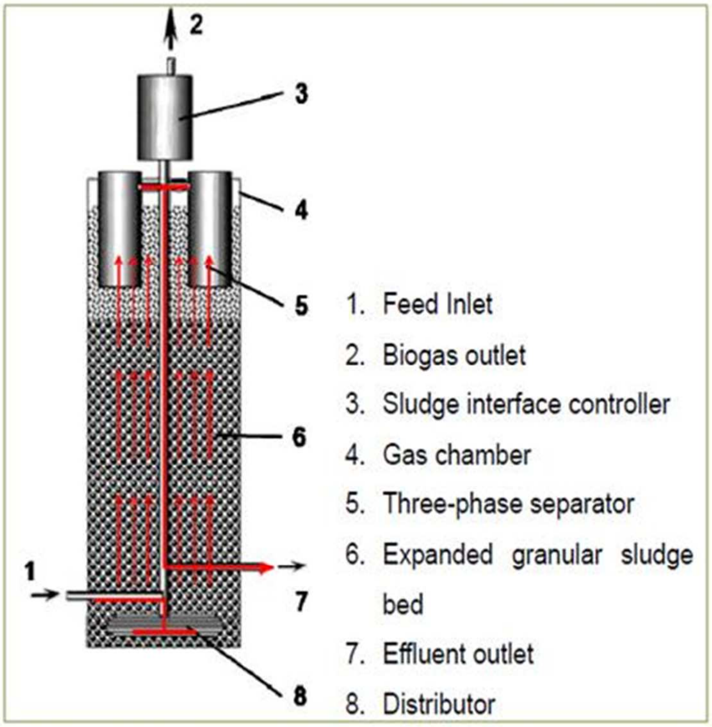
Table 1

	AnaEG ^{TM}	**EGSB**
Power consumption	The grade of sludge expansion depends upon the influent and the biogas production, and automatically adjusted by loading rate. Circulating pump is not required for maintaining the expansion purpose. Therefore, power consumption is smaller.	The reactor requires a recirculation pump to adjust and maintain the expansion.
Removal efficiency & Adaptability of shock-load	AnaEG reactor is a plug-flow reactor, so its adaptability of shock-load is stronger, organic matter removal rate is comparatively higher (generally is above 90%)	It is a complete mixed reactor, its adaptability of shock-load is lower, organic matter removal rate is comparatively lower (generally is below 70%~75%
Organic loading	AnaEG reactor can pick up the load to 50,000mg/L COD	It generally, cannot pick up more than 10,000mg/L COD.

Table 2

Anaerobic reactor operating conditions

Stage	First Start-up I phase	II phase	III phase	IV phase	V phase	Increasing system loading and stable running			Second start-up and stable running	
Time (days)	1-34	35-54	55-65	66-76	77-87	88-117	118-142	143-200	201-222	223-333
HRT (hr)	96	96	96	96	96	72	48	48	48	48
Amount of glucose added (mg COD/L)	1000	800	500	300	0	0	0	0	0	0
COD conc. (mg/L)	2340-2500	2180-2240	1940-2010	1710-1760	1350-1440	1400-1410	1270-1420	1360-1370	1210-1330	1000-1210
Total phenolic conc. (mg/L）	290-320	310-330	290-320	310-330	300-310	320-330	270-310	300-310	240-250	230-250
Volatile phenol conc. (mg/L）	150-160	140-150	130-140	130-140	140-150	130-140	140-150	140-150	120-140	110-130



Graphical Abstract
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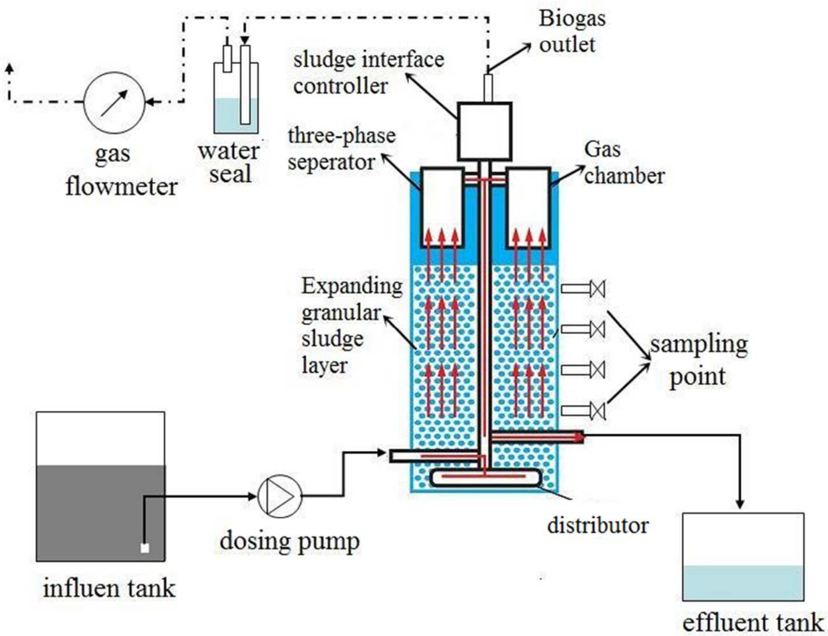

Figure 1. Schematic diagram of experimental apparatus of anaerobic reactor (AnaEG)
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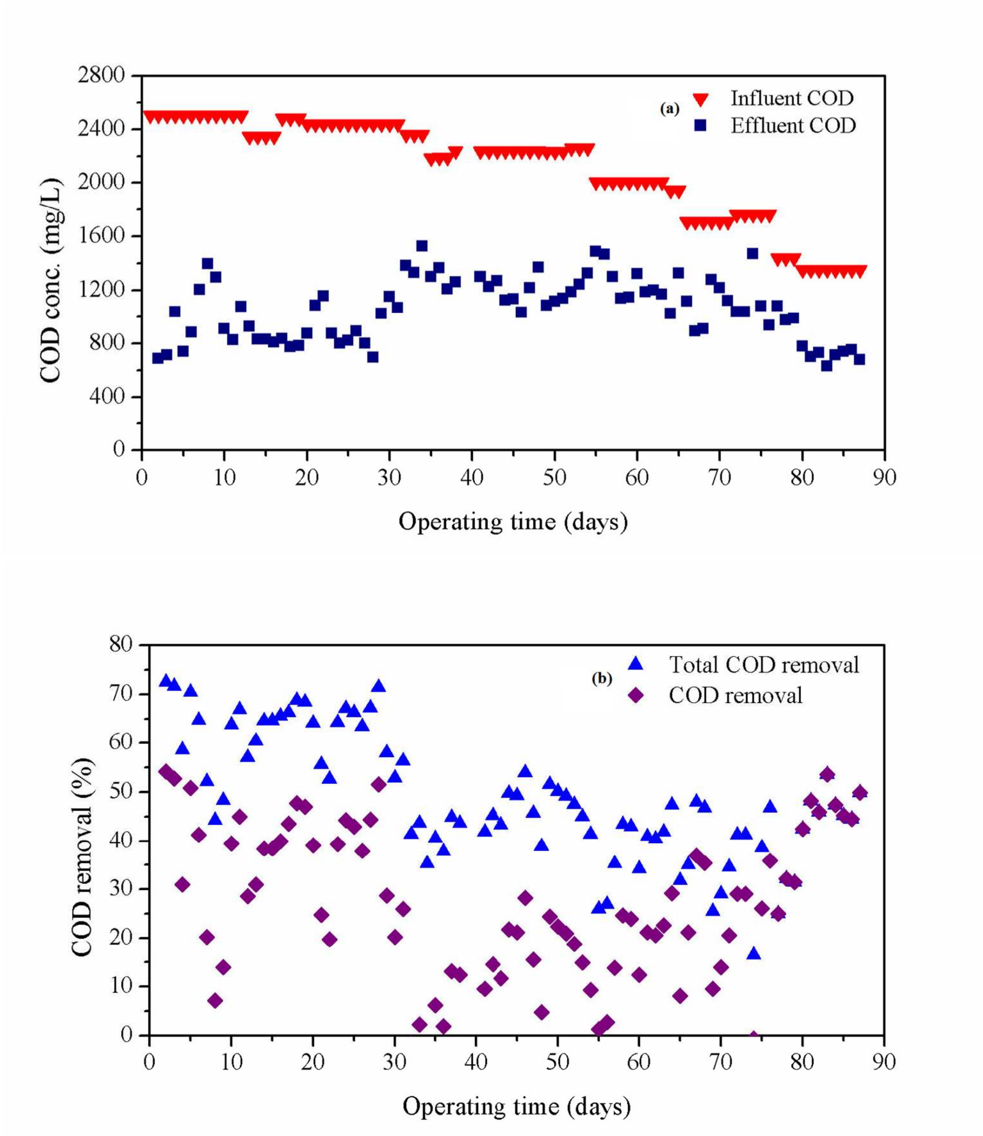

Figure 2. Anaerobic reactor initialization phase degradation trends (a) COD degradation trends (b) COD removal efficiency (c) Total phenol (d) Volatile phenol (e) pH variations.
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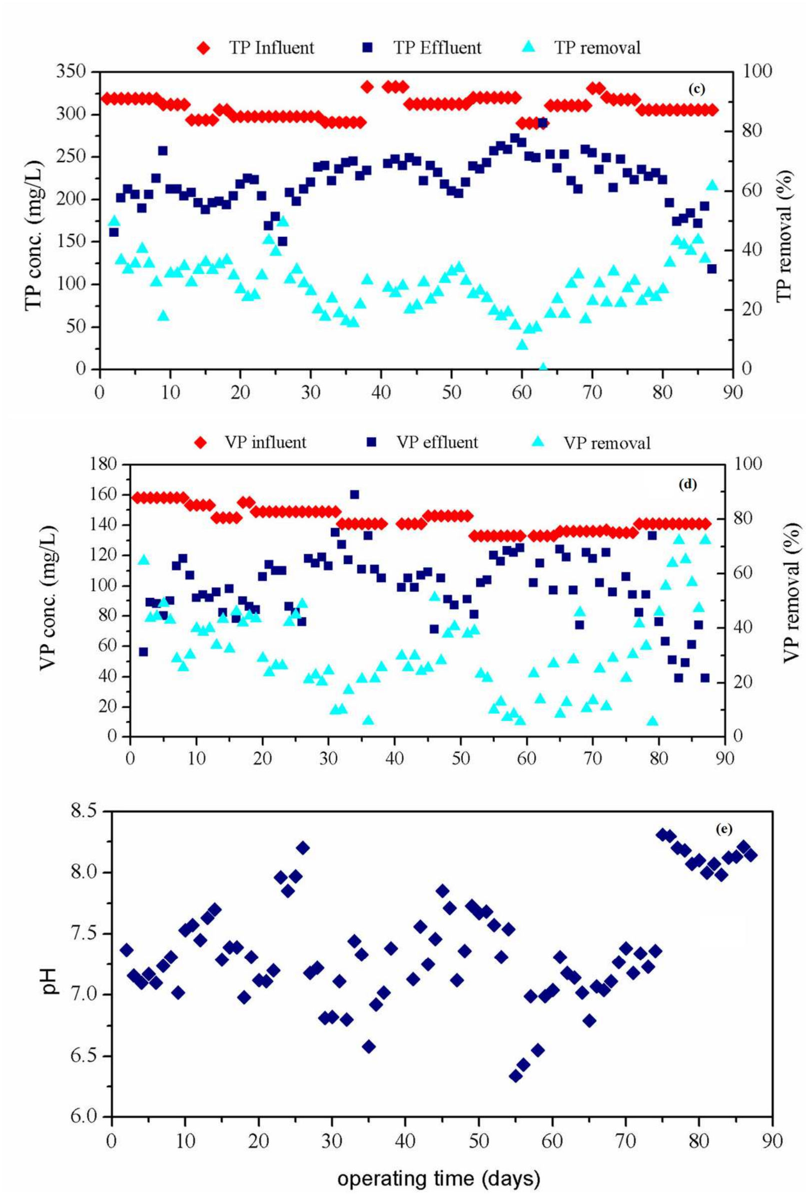

Figure 2. Anaerobic reactor initialization phase degradation trends (a) COD degradation trends (b) COD removal efficiency (c) Total phenol (d) Volatile phenol (e) pH variations.
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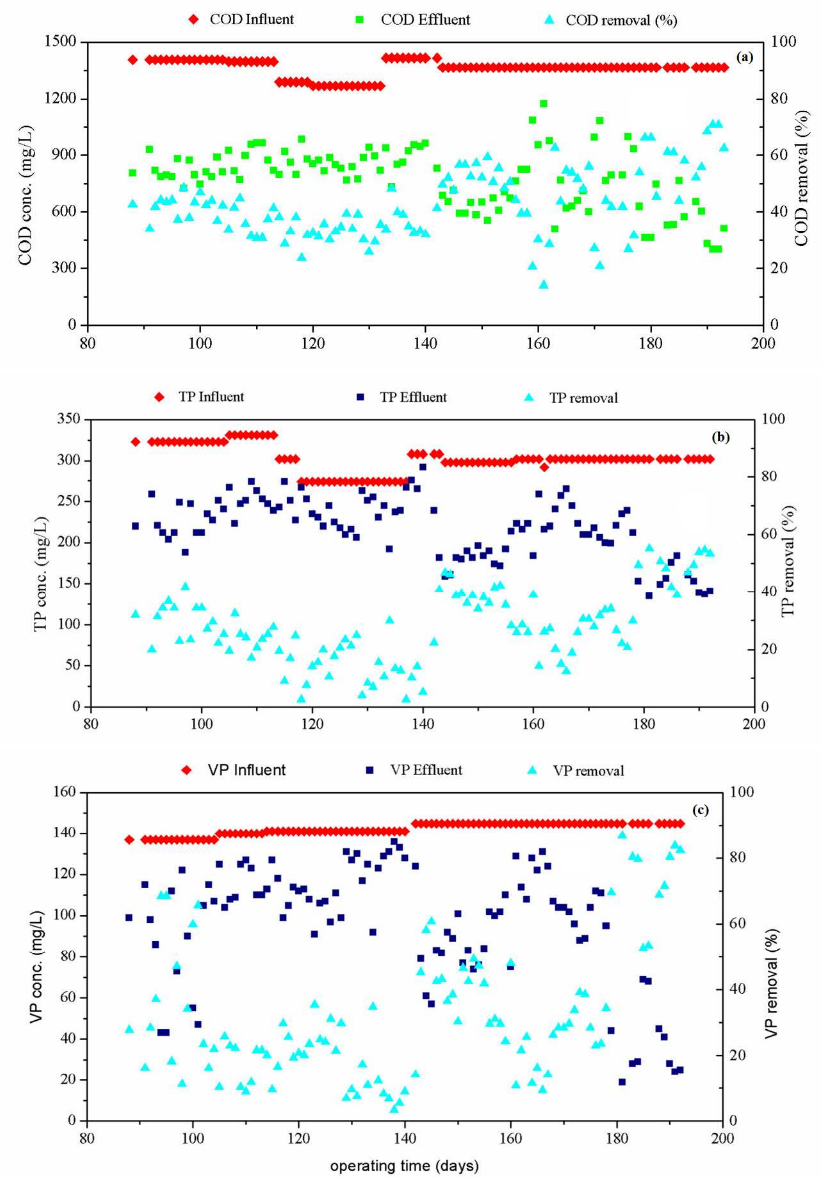

Figure 3. Anaerobic system loading trends during stable running phase (a) COD degradation (b) Total phenol degradation (c) Volatile phenol degradation
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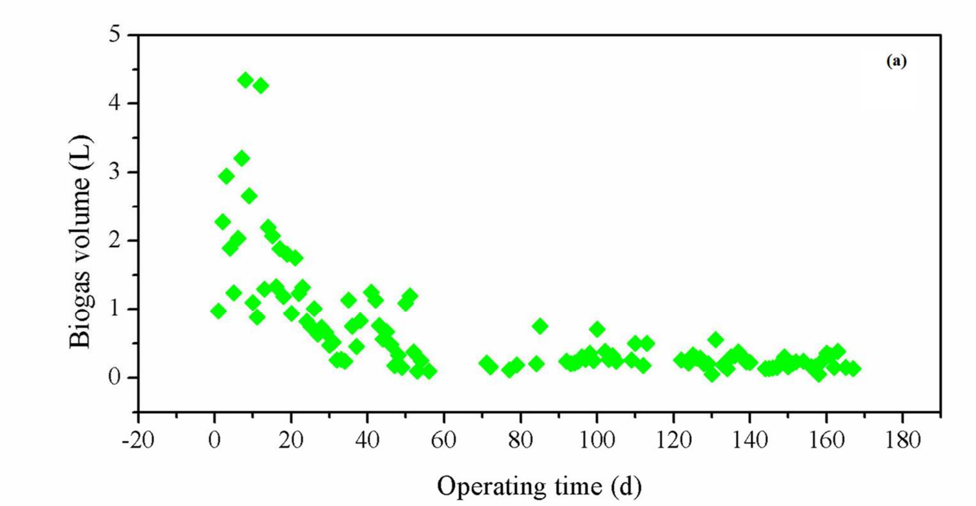

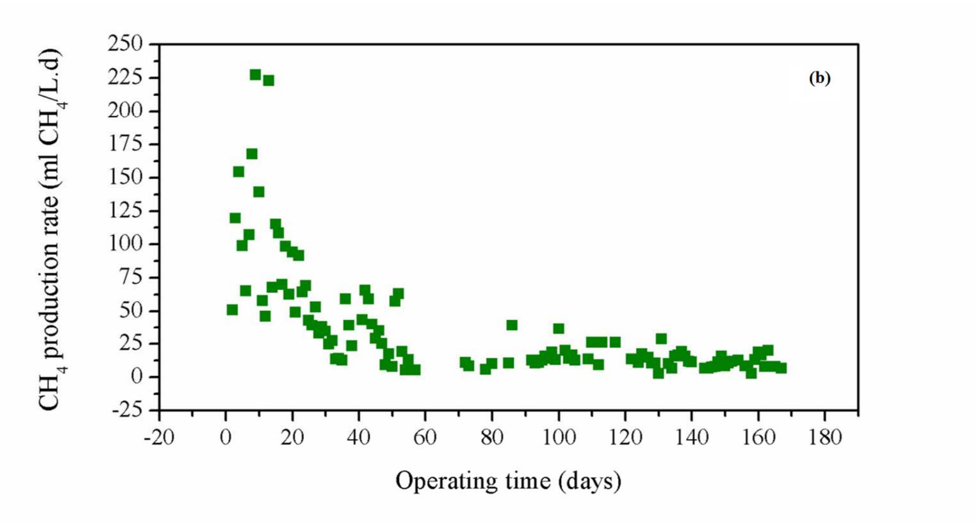

Figure 4. (a) Biogas production (b) Methane production
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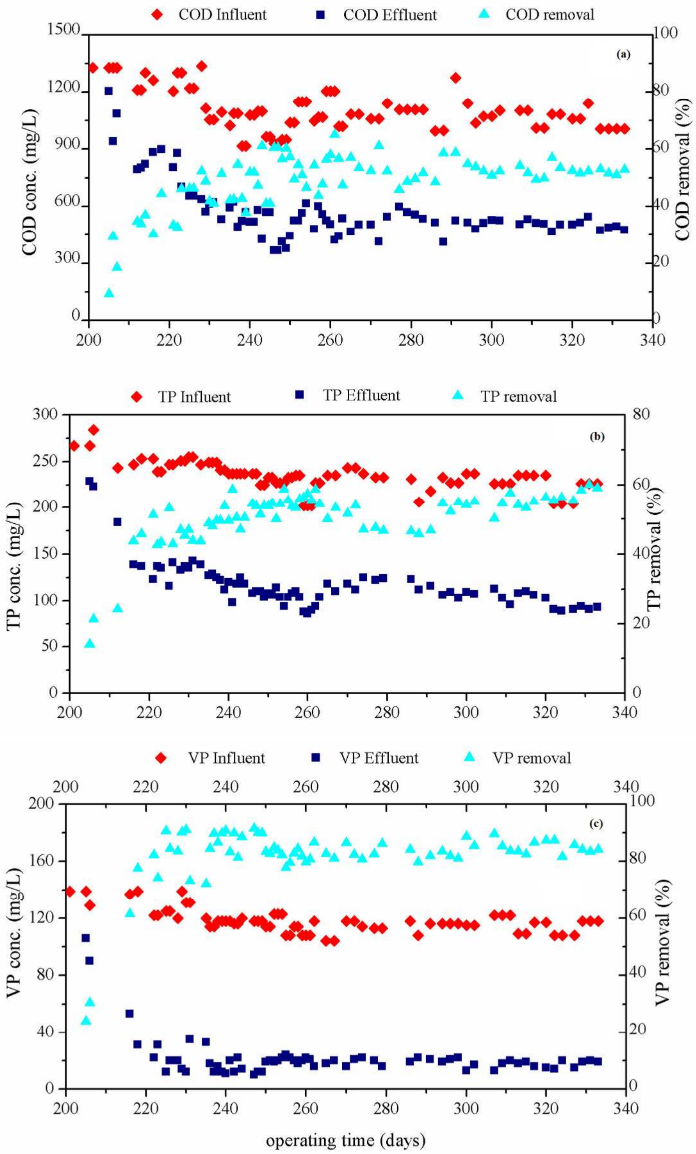

Figure 5. Anaerobic reactor second start-up and stable running (a) COD degradation trends (b) Total phenol degradation trends (c) Volatile phenol degradation trends
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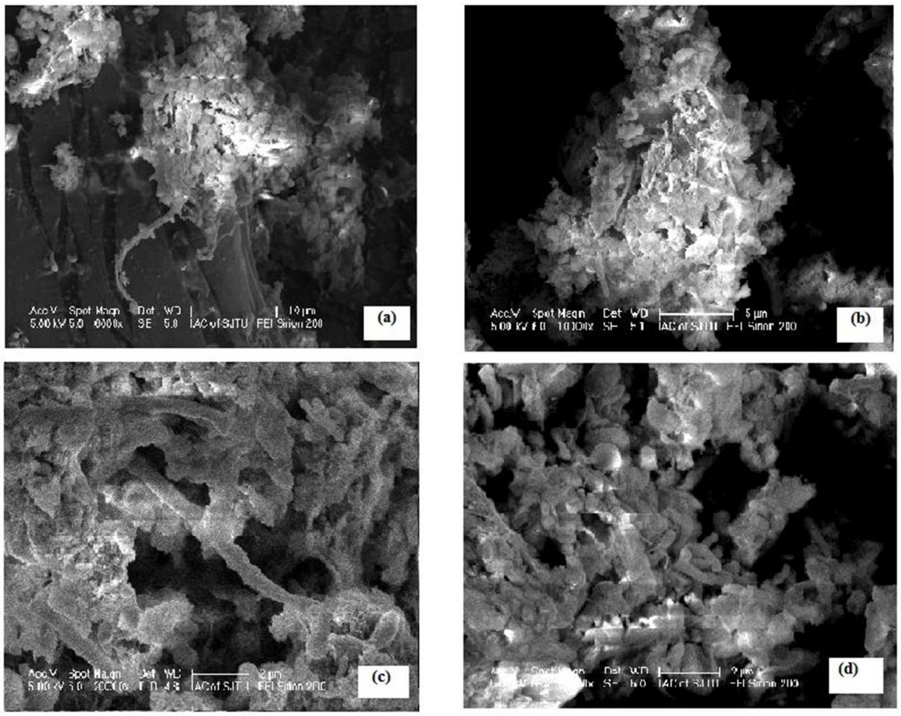

Figure 6. Anaerobic sludge scanning electron micrograph (a), (b), (c) and (d) filamentous bacteria and bacillus intertwined together.
233x184mm (96 x 96 DPI)