

## MSG Wastewater Treatment with UASB-AOMBR Coupling process

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**Abstract**—Monosodium glutamate (MSG) wastewater is a very refractory case of high strength organic wastewater. To investigate the effect of operating variables on treatment of MSG wastewater, a laboratory scale study was conducted. The treatment of MSG wastewater was studied by UASB-A/OMBR process. The effect of operating variables such as organic volumetric load rate (OLR), pH, volatile fatty acids (VFA), ammonia ( $\text{NH}_3\text{-N}$ ) and alkalinity on efficiency of treatment and recovery of biogas was investigated. The results showed that, in the UASB stage, the gas production rate was  $3.02 - 8.83 \text{ kg COD} / \text{m}^3 \cdot \text{d}$  volume load, the gas production rate and COD load volume was positively related; as influent COD in range of  $11878 - 16922 \text{ mg} \cdot \text{l}^{-1}$ , COD removal rate of effluent was in range of  $57.00 - 79.63 \%$ . The removal of COD by MBR can reach 90%, even to 96%, and the removal of ammonia is  $95\% - 100\%$ . UASB--A/O MBR has a good ability of preventing fluctuation of load and can remove COD and ammonia effectively.

### INTRODUCTION

Monosodium glutamate (MSG) factories, both at medium and large scale, suffer from inadequate treatment and disposal problems due to high concentration of COD,  $\text{Cl}^-$ ,  $\text{NH}_3\text{-N}$ ,  $\text{SO}_4^{2-}$  contents present in the wastewater<sup>[1, 2]</sup>. In order to insure the sustainable development of the MSG industry, more efforts have been directed toward the acquisition of feasible, efficient and economical methods for treating MSG wastewater<sup>[3]</sup>. With the increase of MSG production, efficient treatment of MSG wastewater had been considered as an emergency problem to be solved<sup>[4]</sup>.

Many of these studies and nearly all applications have incorporated anaerobic pretreatment with aerobic biological treatment as polishing stage. Anaerobic treatment, while removing a high percentage of many contaminants, some of which are quite recalcitrant in aerobic treatment, produces effluent that is rarely suitable for direct discharge to a receiving water. Subsequently, much attention has been focused on anaerobic-aerobic treatment sequences to biologically treated wastewaters containing a wide variety of novel, often toxic, xenobiotic compounds.

The upflow anaerobic sludge blanket (UASB) has been successfully implemented for the treatment of high strength industrial effluents. Researchers have conducted many studies. The characteristics of anaerobic microbial granules grown in an UASB reactor treating catechol bearing synthetic wastewater (SWW) were studied by Revanuru Subramanyam (2008)<sup>[5]</sup>, the specific methanogenic activity of the sludge showed an increase in trend with an increase in the organic loading rate and the catechol concentration in the SWW. The substrate composition and organic loading rate on the process performance during start-up and steady state were studied by N. Musee (2007)<sup>[6]</sup>, Kaan Yetilmezsoy (2009)<sup>[7]</sup> Matsumoto and Noike (1991)<sup>[8]</sup>. The optimum values of operating variables for treating hog wastewater were reported (Chen et al., 1997)<sup>[9]</sup> for the anaerobic fluidized bed treatment of hog wastewater. The feasibility of treatment of monosodium glutamate fermentation wastewater was evaluated (Tseng and Lin, 1990)<sup>[10]</sup> in terms of removal efficiency and methane content in the biogas. A BOD removal efficiency of 90% was attained with a methane content of 80.8% and OLR of  $10.1 - 31.1 \text{ kg COD m}^{-3} \cdot \text{day}^{-1}$ .

In the past three decades, the membrane bioreactor (MBR) has been paid high attention in wastewater treatment and reuse as a promising technology<sup>[11,12]</sup>. In contrast to the traditional technologies (i.e. conventional activated sludge, stabilization ponds, SBR, etc.), Replacing secondary clarifiers in the conventional activated sludge process with membrane units, MBR have better effluent quality, smaller footprint and much less waste sludge owing to its operation at relatively long sludge age and low sludge loading rate (Brindle and Stephenson, 1996; Rosenberger et al., 2002).

In order to investigate the effect of operating variables on treatment of MSG wastewater, a laboratory scale study was conducted. Undoubtedly, this work would be very significant to clean production of the MSG Industry, to efficient treatment and safe discharge of MSG wastewater<sup>[13, 14, 15]</sup>.

## MATERIAL AND METHODS

### Experimental set-up

A schematic diagram of the experimental set up is shown in Figure 1. Anaerobic pre-treatment (Upflow Anaerobic Sludge Blanket, UASB) followed by aerobic post-treatment (Membrane Bioreactor, MBR) was chosen for the present study.

An UASB reactor consisted of a perspex column made by joining two sections of different lengths of 90 mm internal diameter measuring to a height of 1,200 mm with a constant temperature plexiglass casing. Over this, an upper section of 150 mm diameter and 300 mm length was mounted to prevent carry over of suspended particles into the effluent and also to serve as a gas holder, named three-phase separator. The working volume of the reactor was worked out to be 9.5 L. The feed pumping system consisted of two peristaltic pumps operated alternatively at specified time interval by means of a digital timer system to ensure 24 hours continuous operation. The UASB reactor was operated with a temperature of  $38 \pm 1$  °C. The UASB effluent was connected to the MBR inlet.

The A/OMBR unit consists of two perspex columns with a diameter of 90 mm and a height of 1,500 mm. The membrane module was a microfiltration hollow fiber made of Polyvinylidene fluoride (PVDF). The membrane module was fully submerged into the medium in the column. The bottom part incorporated inlets for the influent and air, the top part incorporated outlets for the effluent and air. Bioreactor was operated at room temperature (20-25°C), controlled by a programmable logic controller (PLC). Liquid level probes (including a high level detector and a low one) were placed in the reactor. Pumps and automated valves were interconnected via the PLC with the liquid level probes. The total volume of the reactor is 11.8 l and the hydraulic residence time (HRT) was 24 h. The valves for sludge discharge and the air inlet were set at the bottom of the reactor.

### Industrial wastewater and seed

The industrial wastewater (MSG) was supplied by a MSG factory in North China, and the characteristics of MSG wastewater are presented in Table 1. The seeding material required for the start-up of the reactor was procured from a municipal wastewater plant.

TABLE I. CHARACTERISTICS OF MSG WASTEWATER

| Parameter                     | Unit               | Value        |
|-------------------------------|--------------------|--------------|
| COD <sub>cr</sub>             | mg•l <sup>-1</sup> | 5405 - 17000 |
| BOD <sub>5</sub>              | mg•l <sup>-1</sup> | 3000 - 9000  |
| Cl <sup>-</sup>               | mg•l <sup>-1</sup> | 8000 -10000  |
| SO <sub>4</sub> <sup>2-</sup> | mg•l <sup>-1</sup> | 3500 - 6000  |
| pH                            | -                  | 3.0 - 3.2    |
| NH <sub>3</sub> -N            | mg•l <sup>-1</sup> | 800-2100     |

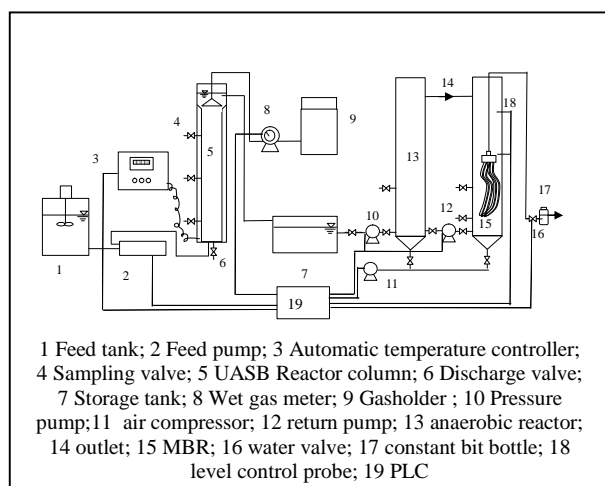


Figure 1. Schematic diagram of the UASB-Reactor and MBR-System

## Chemical analyses

The Chemical Oxygen Demand (COD), Volatile Solids (VS), ammonia ( $\text{NH}_3\text{-N}$ ), pH, and Volatile Suspended Solids (VSS) of samples are analyzed as per the procedure given in the Standard Methods (APHA, 2005)<sup>[16]</sup>. Volatile fatty acids (VFA) determination was performed by HPLC, with a Jasco unit.

## RESULTS AND DISCUSSION

### UASB stage

After 2 months cultivate, UASB reactor deal with a high strength MSG wastewater successful implementation of the sludge particles and maintain the reactor higher sludge concentration, and ensuring the stable operation of high load. The operating experiment was divided into five phases, the operating parameters of each operating phase were showed in Table 2. Weekly analysis of the sludge collected from the UASB reactor indicate that the concentration of the sludge ranged from  $7.49 \text{ g VSS} \cdot \text{l}^{-1}$  ( $\text{VSS/TSS} = 0.22$ ) to  $13.27 \text{ g VSS} \cdot \text{l}^{-1}$  ( $\text{VSS/TSS} = 0.47$ ), with an average value of  $11 \text{ g VSS} \cdot \text{l}^{-1}$ .

The characteristics of raw wastewater and UASB-effluent are recorded in Table II and illustrated graphically in Figs 2-6.

TABLE II. EFFICIENCY OF THE UASB REACTOR FOR THE TREATMENT OF MSG WASTEWATER OF EACH OPERATING PHASE

| Phases | Time of each phase / d | Influent COD / $\text{mg}\cdot\text{l}^{-1}$ | effluent COD / $\text{mg}\cdot\text{l}^{-1}$ | Removal of COD / % | effluent $\text{NH}_3\text{-N}$ / $\text{mg}\cdot\text{l}^{-1}$ | HR T / d | Gas yield Ratio / $\text{l}\cdot\text{kg}^{-1}\text{COD}$ | OLR / $\text{kg}\text{COD}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ |
|--------|------------------------|--|--|--------------------|---|----------|---|---|
| 1      | 14                     | 11986  | 10196  | 16.02              | -   | 1.901    | 0.474   | 5.82  |
| 2      | 8                      | 6000   | 4251   | 32.43              | 953   | 1.967    | 0.615   | 3.02  |
| 3      | 9                      | 7934   | 3226   | 59.36              | 1188  | 2.147    | 0.509   | 3.29  |
| 4      | 16                     | 14149  | 3928   | 69.83              | 1777  | 1.910    | 0.461   | 6.08  |
| 5      | 13                     | 14667  | 5060   | 65.44              | 2338  | 1.731    | 0.58  | 8.83  |

The quality of the reactor effluent confirms the effectiveness of the UASB as a pre-treatment technology for MSG wastewater. Total COD removal values were 48.6%. The corresponding residual value was  $5332 \text{ mgO}_2\cdot\text{l}^{-1}$  in average. The biogas production rate in this experiment ranged between  $0.461 \text{ l}\cdot\text{kg}^{-1}\text{COD}$  and  $0.615 \text{ l}\cdot\text{kg}^{-1}\text{COD}$ .

#### a. Effect of influent COD in UASB

Initially the influent COD was kept within the range  $10476 - 13333 \text{ mg}\cdot\text{l}^{-1}$  (the top concentration of influent COD at the end of start-up phase), but the results showed that the influent COD was too high that methanogenic bacteria in the reactor could not fit it. This caused acid accumulation, pH dropped to 6.5, and gas yield was declined. Thus the influent COD was decreased to  $5405 - 7372 \text{ mg}\cdot\text{l}^{-1}$  in Phase 2. It was observed from Figure 2 that the effluent COD was decreased, and the COD removal percentage was increased from 16 to 70 % with operation time in Phase 2. An efficiency of 76.0 % was obtained at a maximum OLR of  $8.83 \text{ kg COD}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ .

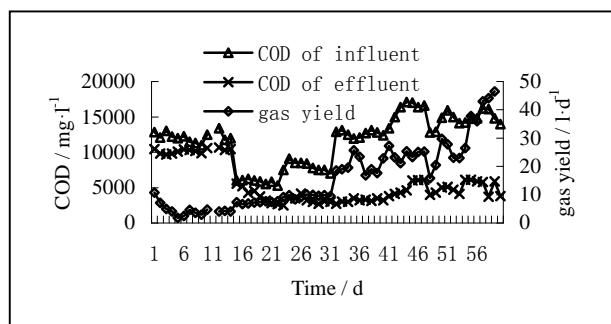


Figure 2. Variation of COD and gas yield in UASB

#### b. OLR and COD removal rate

By the OLR and the COD removal rate curve (Figure 3), the OLR and removal rate showed approximate linear relationship in low-load operation phase, COD removal rate was increased with

the OLR increasing. With the sludge acclimation, the COD removal rate increased rapidly. And when to high-load stage, COD removal rate gradually stabilized and declined slightly.

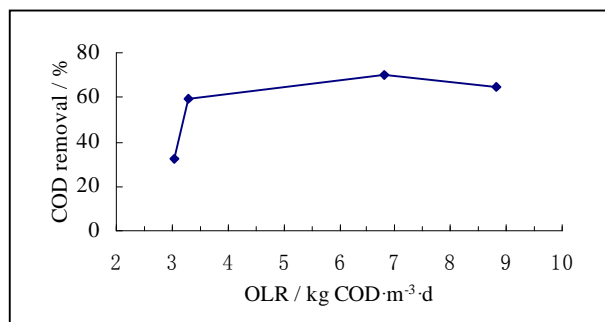


Figure 3. Variation of COD removal with OLR in UASB

### c. Biogas Recovery in USAB

The variation of biogas with time of operation for each phase was plotted in Figure 2. Because of methanogenic bacteria failed to adapt to the influent COD concentration at Phase 1, gas yield was declined from  $10 \text{ l} \cdot \text{d}^{-1}$  to  $2 \text{ l} \cdot \text{d}^{-1}$ . However, after more than 10 days of operation, the micro-organisms adapt quickly to the influent concentration, gas production has gradually picked up. It was observed that the biogas yield was increased with increase of OLR. The maximum biogas yield was observed to be  $46.5 \text{ l} \cdot \text{d}^{-1}$ . The composition of the biogas was analyzed using a high performance gas chromatograph and the consistent methane percentage was found to be 53 – 62 %. Figure 4 showed that the volumetric gas yield and OLR was positive correlation.

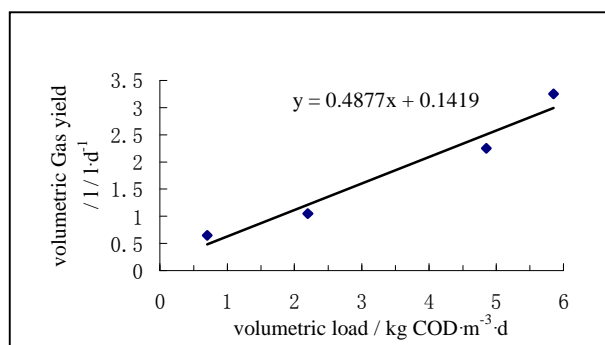


Figure 4. Variation of gas yield with OLR in UASB

### d. VFA and pH in UASB

The variation of the effluent VFA with the pH for each phase was plotted in Figure 5. The results showed that: at the phase 1, too higher influent COD caused VFA accumulation in the reactor, effluent pH declined significantly too; because of the buffer capacity for pH in the reactor was low at the initial stage. With the buffer capacity increasing, although the fluctuation of influent VFA was violently, pH and VFA of effluent were stable, even influent pH declined to 3.2, at the end of Phase 5. This means the UASB with good buffer capacity for pH could ensure the acidogenic bacteria and methanogenic bacteria balance and Methanogens high activity, and could deal with the acidic organic wastewater (pH<3.5) without adjusting pH by dozing alkali salts.

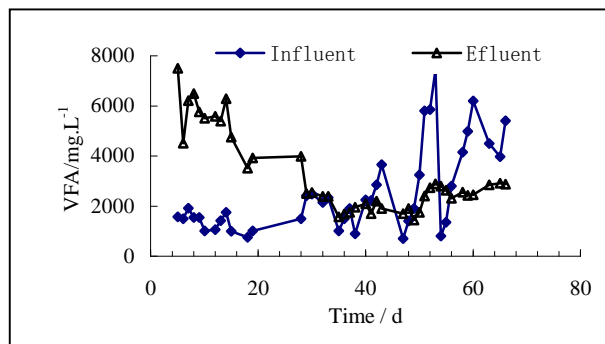


Figure 5. Variation of VFA with time in UASB

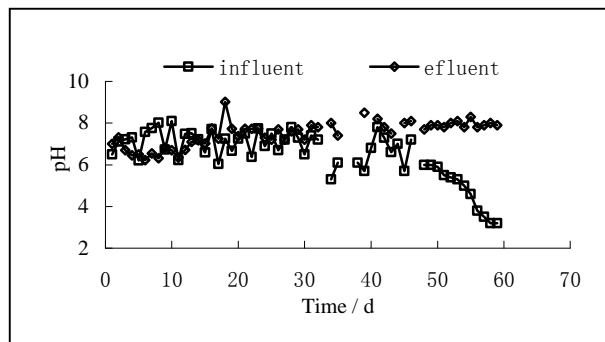


Figure 6. Variation of pH with time in UASB

### MBR stage

The results recorded in Table II indicate that the quality of the UASB-effluent narrowly complied with the Standards regulating discharge of wastewaters into the sewerage network. Therefore, proper post-treatment is required. MBR was used for that purpose. The results obtained indicate very good carbonaceous matter and ammonia elimination as reacted in the removal of COD (97%), and  $\text{NH}_3\text{-N}$  (96%). Residual COD values varied from  $62 \text{ mg}\cdot\text{l}^{-1}$  to  $242 \text{ mg}\cdot\text{l}^{-1}$ , with an average value of  $101 \text{ mg}\cdot\text{l}^{-1}$ . Corresponding  $\text{NH}_3\text{-N}$  values ranged from  $21 \text{ mg}\cdot\text{l}^{-1}$  to  $96 \text{ mg}\cdot\text{l}^{-1}$ , with an average value of  $51 \text{ mg}\cdot\text{l}^{-1}$ , respectively.

TABLE III. EFFICIENCY OF THE MBR REACTOR FOR THE TREATMENT OF MSG WASTEWATER OF EACH OPERATING PHASE

| Phases | Time of each phase / d | Influent COD / $\text{mg}\cdot\text{l}^{-1}$ | Effluent COD / $\text{mg}\cdot\text{l}^{-1}$ | Removal of COD / % | Influent $\text{NH}_3\text{-N}$ / $\text{mg}\cdot\text{l}^{-1}$ | Effluent $\text{NH}_3\text{-N}$ / $\text{mg}\cdot\text{l}^{-1}$ | Removal of $\text{NH}_3\text{-N}$ / % | pH  |
|--------|------------------------|--|--|--------------------|---|---|---------------------------------------|-----|
| 1      | 14                     | -  | -  | -                  | -   | -   | -                                     |     |
| 2      | 8                      | 4251   | 143  | 96.6               | 953   | 27  | 97.2                                  | 6.7 |
| 3      | 9                      | 3226   | 90   | 97.2               | 1188  | 33  | 97.2                                  | 7.7 |
| 4      | 16                     | 3928   | 90   | 97.7               | 1931  | 74  | 96.2                                  | 7.5 |
| 5*     | 13                     | 2552   | 84   | 96.7               | 1392  | 68  | 95.1                                  | 7.9 |

\*In the phase 5, the effluent was returned back to MBR with returned ratio of 1:1

#### a. Effect of influent COD in MBR

COD removal in MBR was shown in Figure 7, influent COD fluctuation range of  $2552 \sim 4251 \text{ mg}\cdot\text{l}^{-1}$ , effluent COD was under  $150 \text{ mg}\cdot\text{l}^{-1}$  stably and removal rate of COD up to about 97%.

When the COD value is transferred to  $6100 \text{ mg} \cdot \text{l}^{-1}$ , COD values could meet the  $170 \text{ mg} \cdot \text{l}^{-1}$ , COD that the system has strong resistance to impact load.

### Ammonia nitrogen removal

Ammonia nitrogen removal in MBR was shown in Figure 8, ammonia nitrogen removal rate in the each phases of the experiment reached above 95%, but as the ammonia nitrogen of influent above  $2300 \text{ mg} \cdot \text{l}^{-1}$ , the ammonia nitrogen of effluent increased to  $96 \text{ mg} \cdot \text{l}^{-1}$ . In the phase 5, the effluent was returned back to MBR with returned ratio of 1:1, and the ammonia nitrogen of effluent decreased back to  $48 \text{ mg} \cdot \text{l}^{-1}$ . In the process of ammonia removal, ammonia oxidation per gram of nitrate need to consume  $7.14 \text{ g}$  alkalinity<sup>[17]</sup>. Raw water alkalinity in  $1500 \sim 2400 \text{ mg} \cdot \text{l}^{-1}$  between the fluctuations, can not fully provide the necessary alkalinity of ammonia removal, so the addition of  $\text{Na}_2\text{CO}_3$  was required to the system to adjust the system pH value and alkalinity.

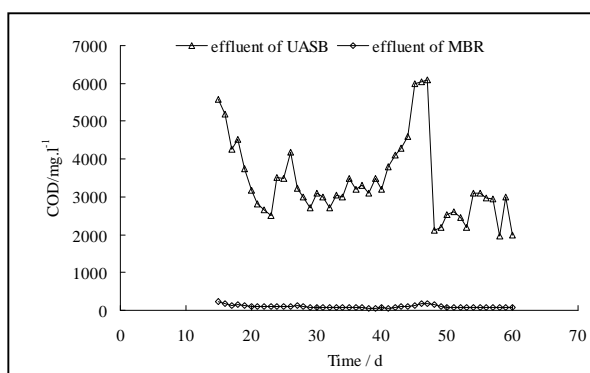


Figure 7. COD removal effect by MBR

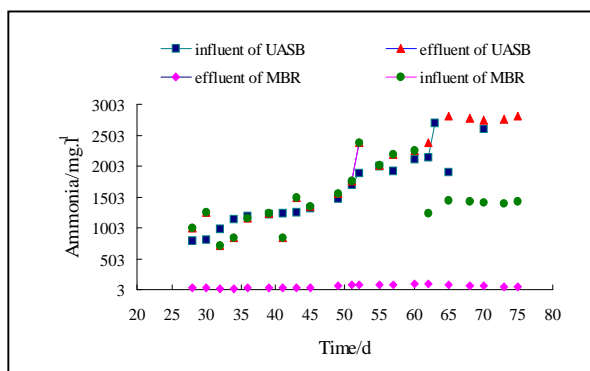


Figure 8. Ammonia removal effect by MBR

### c. pH value change

The pH value changes within the system was shown in Figure 9. The nitrification could cause the accumulation of  $\text{H}^+$  and the decreasing of pH. Nitrifying bacteria had the highest activity under the pH value of 7.0 to 8.1. Nitrifying bacteria sensitive to changes in pH value in the biological treatment system, the denitrification optimum pH range of 7.0 to 7.5, if the pH value dropped to 5.0 to 5.5, the denitrification reaction almost stopped. With adding soda ash, pH remained 7.5, so the nitrification reaction was accomplished well.

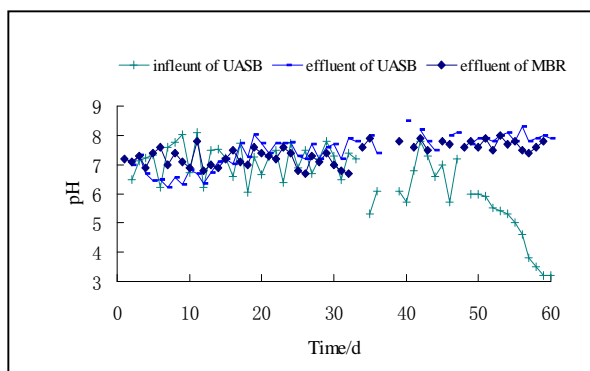


Figure 9. Variation of pH in MBR

## CONCLUSION

- The treatment of high COD and high ammonia monosodium glutamate wastewater by A/O MBR is studied. UASB reactor under high concentrations of MSG wastewater treatment, there is a higher gas production rate, at  $3.02 - 8.83 \text{ kg COD} / \text{m}^3 \cdot \text{d}$  volume load, the average gas production rate of  $0.599 \text{ m}^3 / \text{kg COD}$ , High Load under the gas production rate and COD load volume was positively related to each other.
- In the stable operating phases, as influent COD in range of  $11878 - 16922 \text{ mg} \cdot \text{l}^{-1}$ , COD removal rate of effluent was in range of  $57.00 - 79.63 \%$ , average COD concentration of effluent was  $5446 \text{ mg} \cdot \text{l}^{-1}$ ; the average gas production rate was  $0.5085 \text{ m}^3 \cdot \text{kg}^{-1} \text{COD}$ , gas production rate and the organic load was positive correlation.
- UASB with high buffering capacity of the stability in case of high load of acid-base environment, could deal with the acidic organic wastewater ( $\text{pH} < 3.5$ ) without adjusting pH by dozing alkali salts.
- The removal of COD by MBR can reach 96% even to 98% and the removal of ammonia is 95% -99%. The experimental results show that A/O MBR has a good ability of preventing fluctuation of load and can remove COD and ammonia effectively.

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