

## Two-stage air stripping combined with hydrolysis acidification process for coal gasification wastewater pretreatment

Xiurong Chen, Xiaoli Sun, Xiaoxiao Wang, Peng Xu, Chenchen Yang, Quanling Lu and Shanshan Wang

### ABSTRACT

Coal gasification wastewater is mainly from gas washing, condensation and purification processes in the gas furnace with high  $\text{NH}_3\text{-N}$  (nitrogen in water in the form of free ammonia ( $\text{NH}_3$ ) and ammonium ion ( $\text{NH}_4^+$ )), TN (total nitrogen) and refractory organics content, which will inhibit the subsequent biological treatment. The 'air stripping – hydrolysis acidification – air stripping' process was proposed as the pretreatment for coal gasification wastewater to improve the biodegradability and nitrogen removal, which could reduce the subsequent biological treatment load. The first-stage air stripping process before hydrolysis acidification could achieve a significant removal of  $\text{NH}_3\text{-N}$  (97.0%) and volatile phenol (70.0%), reducing the corresponding toxicity on hydrolysis acidification. The group with air stripping had more abundant microbial communities and a more effective organic degradation performance in hydrolysis acidification than that without air stripping. The second-stage air stripping removed  $\text{NH}_3\text{-N}$  released from hydrolysis acidification, and significantly reduced the TN concentration in effluent. The whole process achieved a TN removal from  $2,000 \pm 100$  mg/L to  $160 \pm 80$  mg/L, and a total phenols removal from  $700 \pm 50$  mg/L to  $80 \pm 20$  mg/L.

**Key words** | air stripping, ammonia removal, coal gasification, combination process, hydrolysis acidification

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### INTRODUCTION

In typical coal gasification wastewater, the concentration of chemical oxygen demand (COD),  $\text{NH}_3\text{-N}$  and total nitrogen (TN) can reach 22,000–35,000 mg/L, 3,000–5,000 mg/L and 6,000–8,000 mg/L, and the C/N (the ratio of the total amount of carbon in an organism to the total amount of nitrogen) only ranges between 3.5 and 4.4 (Feng *et al.* 2009; Yu *et al.* 2010; Wang *et al.* 2010; Li *et al.* 2011; Zhao *et al.* 2013). The relatively low C/N in coal gasification wastewater determines the insufficiency of the carbon source in subsequent biological nitrogen removal. Therefore, a desired pretreatment should be carried out to adjust C/N to a suitable range. It has been proved that the optimal C/N in wastewater for biological nitrogen removal is usually in the range of 5–10 (He *et al.* 2016; Krishna Mohan *et al.* 2016). Moreover, toxic and refractory organic compounds prevail in coal gasification wastewater, which will cause irreversible reduction of biological treatment performance (Li *et al.* 2011; Oladipo *et al.* 2017),

and a corresponding pretreatment is also needed before the aerobic biological treatment. The organic compounds in coal gasification wastewater mainly contain phenols (of which the concentration can reach more than 4,000 mg/L), polycyclic aromatic compounds, heterocyclic compounds, petroleum hydrocarbons, cyanide, sulfide, pyridine, carbazole, biphenyl, terphenyl and so on (Yu *et al.* 2010; Wang *et al.* 2014; Zhuang *et al.* 2014; Hou *et al.* 2015; Jia *et al.* 2015). In brief, coal gasification wastewater is a typical highly toxic, harmful and refractory wastewater.

Based on the characteristics of coal gasification wastewater, the phenol-ammonia recovery system is always adopted before biological treatment. The typical process of phenol-ammonia recovery is the combination of solvent extraction of phenols and steam stripping of ammonia. At present, the widely used efficient phenol extractant includes methyl, tert-butyl, ether (Zeng *et al.* 2011),

N-octanoylpyrrolidin (Li *et al.* 2014) and so on. Wang *et al.* (2011) used methyl isobutyl ketone as extractant combined with stripping of ammonia in treatment of coal gasification wastewater, and significant removals of various pollutants were obtained. The working efficiency of the ammonia removal tower is closely related to temperature, vacuum pressure, feeding rate and so on. In practical application, the Lurgi pressure gasification process was applied in China Coal Longhua Harbin Coal Chemical Industry Co. Ltd, and achieved ammonia removal from 2,300–8,500 mg/L to 100–300 mg/L (Feng *et al.* 2009). The cost of energy and reagents consumption in the above multi-stage extraction and steam stripping process is relatively high, and the concentration of phenols and ammonia in the effluent remains high at 500–1,000 mg/L and 100–300 mg/L respectively, which will still cause an inhibitory effect on microbial activity. In the view of the high energy consumption and high cost of reagent in a solvent extraction and steam stripping system, air stripping has been demonstrated for its rapid and stable removal of phenols and ammonia, which shows the advantages of low operation cost and stable efficiency.

At present, the effluent of the phenol-ammonia recovery system is usually treated by an anaerobic-aerobic combined biological process. For the anaerobic unit, a whole process of anaerobic methane production or hydrolysis acidification process can both be adopted. The hydrolysis acidification bacteria have a stronger tolerance for phenols and ammonia compared to the methanogenic bacteria, and hydrolysis acidification combined with aerobic biological treatment is widely applied (Jin *et al.* 2016). In the hydrolysis acidification process, the complex macromolecular organic compounds are transformed into small molecular compounds like volatile fatty acids (VFAs), and the biochemical oxygen demand (BOD), B/C (BOD<sub>5</sub>/COD) and BOD/TN of the effluent increase, which improves the biodegradability of wastewater. Although the hydrolysis acidification process has the better resistance to shock loads, it also has a certain tolerance to the influent concentration of ammonia and toxic organic loading (Ye *et al.* 2008). Therefore, proper pretreatment for removal of phenols and ammonia is necessary before the hydrolysis acidification process.

Based on the summary on pretreatment of coal gasification wastewater, this study proposed a combined process of air stripping and hydrolysis acidification to achieve the removal of phenols and ammonia with low cost and high efficiency. Air stripping has the removal effects of NH<sub>3</sub>-N and volatile phenols, and hydrolysis acidification can transform macromolecule organic compounds and rapidly release the NH<sub>3</sub>-N. When the relative content of organic-N

in wastewater is high, hydrolysis acidification is necessary before air stripping. However, when the raw coal gasification wastewater is directly treated by hydrolysis acidification, highly concentrated NH<sub>3</sub>-N and phenols will cause inhibition on the hydrolytic acidification bacteria. Therefore, in this study, different combinations of air stripping and hydrolysis acidification were studied and the removal effects were compared in order to optimize the pretreatment process of coal gasification wastewater.

## MATERIALS AND METHODS

### Wastewater

The test wastewater was taken from the influent of the aerobic sludge treatment in the coal gasification wastewater of a coal-gas company in Xinjiang province in China, and the water quality is shown in Table 1. Before air stripping and hydrolysis acidification, the coal gasification wastewater was pretreated through coagulation/flocculation to removal oil content and the results are also shown in Table 1. The 'raw wastewater' in the following paper all refers to the effluent of the coagulation process. The test activated sludge was taken from an aeration tank in the A<sub>2</sub>O system in a municipal wastewater treatment plant in Shanghai, China.

### Experimental design

#### Optimal experiment for air stripping

The influence of gas-liquid ratio and aeration time on the removal of NH<sub>3</sub>-N was investigated in air stripping process. The test was conducted in a 1,500 mL beaker, and each batch experiment used 1,000 mL coal gasification wastewater. The initial pH was 8.87 (the PH in experiment), and the initial concentration of NH<sub>3</sub>-N and total phenols

**Table 1** | Main characteristics of the coal gasification wastewater

Indices	Coal gasification wastewater	Effluent of coagulation
pH	8.5–9.0	8.5–9.5
COD (mg/L)	16,000 ± 1,000	6,000 ± 500
TN (mg/L)	4,000 ± 1,000	2,100 ± 150
Total phenols (mg/L)	2,500 ± 300	250 ± 40
Ammonia (mg/L)	1,500 ± 100	1,500 ± 100
Oil content (mg/L)	1,000 ± 100	90 ± 10

*Note:* The effluent of coagulation is the raw wastewater for this study.

was  $1,500 \pm 100$  mg/L and  $2,300 \pm 100$  mg/L respectively. In the optimal experiment, the gas-liquid ratio was set to 1,000, 1,500 and 2,000. During the experiment, the blown gas is directly injected into the absorbent for absorption. The schematic of experiment set up is shown in Figure 1.

### Hydrolysis acidification

The hydrolysis acidification process was conducted in a cylinder with 5 L effective volume. The sludge domestication adopted the batch operation with a 24 h cycle, and the influent for the sludge domestication was mainly the diluted coal gasification wastewater. The coal gasification (20% diluted) wastewater and effluent of air stripping (20% diluted) were adopted as the influent of sludge domestication during 0–30 d. Similarly, the wastewater (50% diluted) was adopted during 30–60 d, and wastewater (100% diluted) was adopted during 60–90 d.

### Water quality analysis

COD, TN, total phosphorus (TP),  $\text{NH}_3\text{-N}$  and total phenols were measured according to *Standard Methods (APHA-AWWA-WEF 2005)*. The VFA concentration was analyzed by gas chromatography (Shimadzu GC-2014). The molecular weight distribution of organic matter was measured by ultrafiltration. The organic matters were separated by the ultrafiltration membranes (80–90(diameter)/MSC300, Polysulfone resin, Shanghai Mosu Science Equipment Company) which block organic matter with different molecular weights, and the content of organic compounds in different molecular weight range was characterize by the results of TOC (Shimadzu TOC-V).

### High-throughput sequencing

In this study, the V3-V4 region of 16S rDNA was amplified by universal primers 341F (CCTACGGGNGGCWGCAG) and 805R (GACTACHVGGGTATCTAATCC) (Xie et al. 2016).

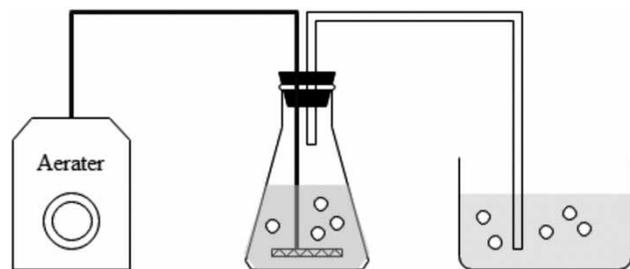


Figure 1 | The experimental equipment diagram of 'Optimal experiment for air stripping'.

The polymerase chain reaction (PCR) (a molecular biological technique for amplifying and amplifying specific segments of DNA) conditions included: 3 min pre-denaturation at  $95^\circ\text{C}$ , 30 s annealing at  $55^\circ\text{C}$ , 45 s extension at  $72^\circ\text{C}$  and 20 min extension at  $72^\circ\text{C}$ . PCR products were subjected to high-throughput sequencing on the Illumina MiSeq platform (Poszytek et al. 2017).

The Alpha indices of microbial diversity contained Chao1 index, Shannon index, Simpson index and coverage index which was calculated by Equations (1)–(4) where  $S_{\text{obs}}$  was the actual number of observed OTU,  $n_i$  was the number of OTU containing the 'i' sequences, N was the total number of sequences.

$$S_{\text{chao1}} = S_{\text{obs}} + \frac{n_1(n_1 - 1)}{2(n_2 + 1)} \quad (1)$$

$$H_{\text{shannon}} = - \sum_{i=1}^{S_{\text{obs}}} \frac{n_i}{N} \ln \frac{n_i}{N} \quad (2)$$

$$D_{\text{simpson}} = \frac{\sum_{i=1}^{S_{\text{obs}}} n_i(n_i - 1)}{N(N - 1)} \quad (3)$$

$$C = 1 - \frac{n_i}{N} \quad (4)$$

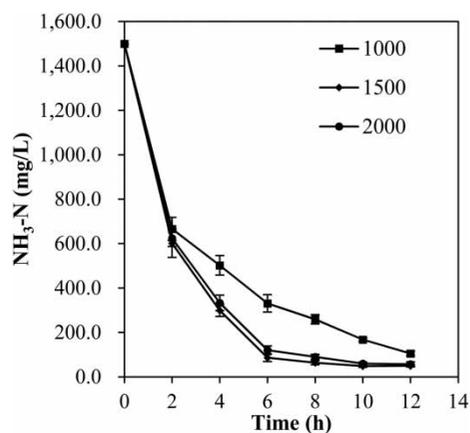
## RESULTS AND DISCUSSION

### The removal performance in air stripping process

#### Optimization of air stripping conditions

The influence of pH, gas-liquid ratio (G/L) and aeration time on the removal of  $\text{NH}_3\text{-N}$  was investigated in air stripping process. The experimental temperature was  $20^\circ\text{C}$  and the pressure was 101.3 kPa. It was proved that more than 90% of  $\text{NH}_3\text{-N}$  existed in the presence of free ammonia at  $\text{pH} > 11$ , and increasing the pH contributed little to the stripping efficiency (Guštin & Marinšek-Logar 2011). Therefore, in this experiment, the pH in stripping process was determined to be 11 to save the cost of alkali. The influence of gas-liquid ratio on removal of ammonia is shown in Figure 2.

As shown in Figure 2, the removal of ammonia varied under the different gas-liquid ratio, and the air volume was the key factor in air stripping process. In air stripping process, the ammonia was transferred from liquid phase to gas phase, and the driving force was from the difference of partial pressure between two phases. Under the G/L of 1,000, the residual concentration of ammonia in wastewater was significantly higher than that under the G/L of 1,500 or



**Figure 2** | The removal of ammonia in air stripping process under different gas-liquid ratio ( $n = 3$ ,  $\text{PH} = 11$ ).

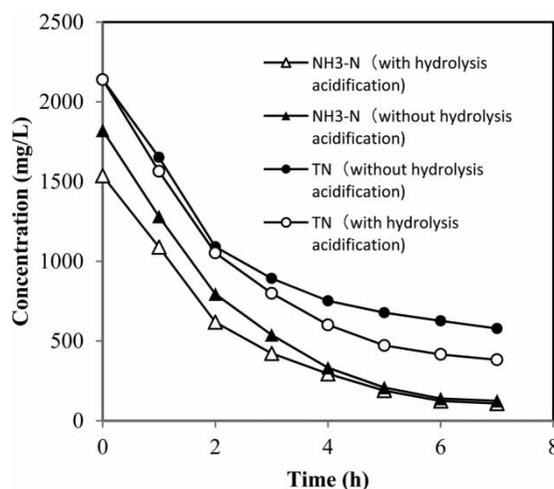
2,000. The residual concentrations of ammonia were quite similar under the G/L of 1,500 and 2,000. Considering the cost, the optimal G/L was chosen to be 1,500. In addition, a stable and great removal of ammonia can be achieved at the stripping time of 6 h at G/L = 1,500 where  $\text{NH}_3\text{-N}$  concentration dropped to  $86.2 \pm 17.0$  mg/L, and the removal efficiency reached 94.2%.

### Comparison of raw water and effluent of hydrolysis acidification in air stripping process

In this study, the raw wastewater and effluent of wastewater after hydrolysis acidification process were treated by air stripping respectively under the optimal conditions (as discussed in the previous section), and the removal of  $\text{NH}_3\text{-N}$  and TN was compared in Figure 3.

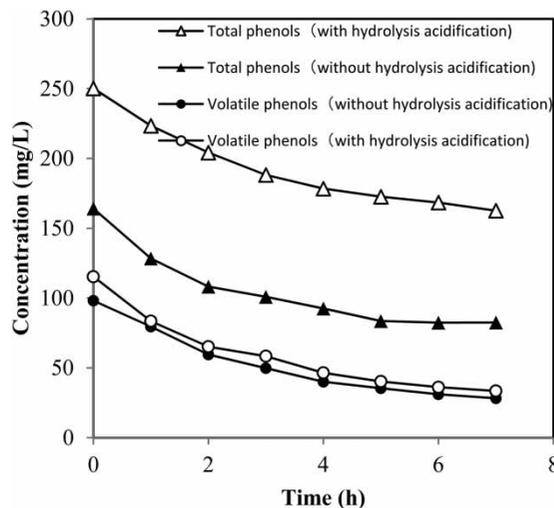
As shown in Figure 3, the  $\text{NH}_3\text{-N}$  concentration increased from  $1,500 \pm 50$  mg/L to  $1,820 \pm 70$  mg/L due to ammonification of organic-N after hydrolysis acidification. In air stripping process, the  $\text{NH}_3\text{-N}$  and TN concentration in raw wastewater dropped from  $1,500 \pm 50$  mg/L to  $150 \pm 10$  mg/L and  $2,100 \pm 100$  mg/L to  $570 \pm 30$  mg/L, and the  $\text{NH}_3\text{-N}$  and TN concentration in effluent of hydrolysis acidification dropped from  $1,820 \pm 70$  mg/L to  $120 \pm 10$  mg/L and  $2,100 \pm 100$  mg/L to  $380 \pm 40$  mg/L. When hydrolysis acidification was performed before air stripping, the TN removal increased from 73.0% to 82.1%.

The removal of TN was significantly improved when adding the hydrolysis acidification before the air stripping. The raw wastewater without hydrolysis acidification had the characteristics of high content of organic-N which was not converted to  $\text{NH}_3\text{-N}$  and was hard to be removed through air stripping; on the other hand, some macromolecular refractory organics were not hydrolyzed into small molecular



**Figure 3** | The variations of  $\text{NH}_3\text{-N}$  and TN concentration during air stripping process.

organic compounds. The hydrolysis acidification process could promote the rapid conversion from organic-N to  $\text{NH}_3\text{-N}$ . After the hydrolysis acidification treatment, the  $\text{NH}_3\text{-N}$  concentration of the coal gasification wastewater increased to  $1,820 \pm 70$  mg/L and TN was mainly composed of  $\text{NH}_3\text{-N}$  here. Wen *et al.* removed the ammonia nitrogen in the wastewater with high ammonia nitrogen by air stripping, and the removal rate was 95% under the optimal experimental conditions (Wen 2007). However, after 'two-stage air stripping combined with hydrolysis acidification process', the removal rate of ammonia nitrogen can reach 97%. In comparison, the hydrolysis acidification transformed organic-N into  $\text{NH}_3\text{-N}$  which significantly improved the removal of ammonia in air stripping process and achieved effective removal of TN reducing the TN load of subsequent biological treatment.



**Figure 4** | The variations of volatile phenols and total phenols concentration during air stripping process.

**Table 2** | Water quality index under different dilution ratio

Type of wastewater Period	Without air stripping			With air stripping		
	0–30 d	30–60 d	60–90 d	0–30 d	30–60 d	60–90 d
Dilution ratio	20%	50%	100%	20%	50%	100%
COD (mg/L)	1,000 ± 50	3,000 ± 150	6,000 ± 300	1,000 ± 50	2,500 ± 100	5,000 ± 200
Total phenols (mg/L)	150 ± 10	350 ± 30	700 ± 50	50 ± 10	125 ± 10	250 ± 10

The air stripping process also had the removal effects of some micromolecule and refractory volatile organic compounds, and the results were shown in Figure 4. In raw wastewater, the total phenols concentration dropped from  $250 \pm 20$  mg/L to  $165 \pm 15$  mg/L (34.0%), and the volatile phenols concentration dropped from  $100 \pm 5$  mg/L to  $30 \pm 2$  mg/L (70.0%) after 6 h air stripping. When the raw wastewater was treated by hydrolysis acidification, the total phenols concentration dropped from  $165 \pm 10$  mg/L to  $82 \pm 5$  mg/L (50.4%), and the volatile phenols concentration dropped from  $115 \pm 5$  mg/L to  $33 \pm 2$  mg/L (80%) after 6 h air stripping. During air stripping process, 6 h was the optimal operation time (comparative test results), and at this time, the residual phenols were mainly the macromolecule organics. Hydrolysis acidification process can transform and degrade macromolecular phenols, and after hydrolysis acidification, the total phenols in raw wastewater dropped from  $250 \pm 10$  mg/L to  $160 \pm 10$  mg/L while the volatile phenols concentration increased remarkably. Therefore, the hydrolysis acidification combined with air stripping had the great removal of volatile phenols.

### The removal performance in hydrolysis acidification process

#### Sludge domestication

The sludge domestication of hydrolysis acidification was conducted in a cylinder with 5 L effective volume under intermittent operation. The operation cycle was 24 h. The influent of sludge domestication was the diluted coal gasification with and without air stripping as shown in Table 2.

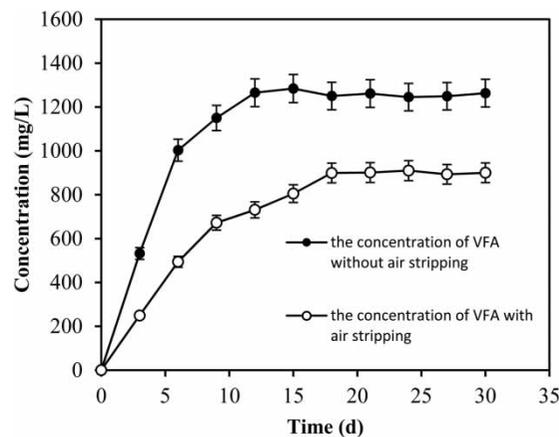
During sludge domestication process, the removal of COD and phenols in wastewater with and without air stripping were compared. In addition to the removal effects of COD and phenols in air stripping process, air stripping reduced the COD,  $\text{NH}_3\text{-N}$  and total phenols loads which weakened significantly inhibition on hydrolysis acidification (Ye et al. 2008). During the initial stage (0–30 d) of sludge domestication, there was no

remarkable difference between the experimental group with and without air stripping due to the high dilution ratio (20%) and low pollutant load. When entering into mid-late stage, the wastewater with air stripping had the distinct advantages on pollutants removal. The COD and total phenols concentration in wastewater without air stripping dropped from  $6,000 \pm 100$  mg/L to  $4,200 \pm 100$  and  $700 \pm 50$  mg/L to  $500 \pm 20$  mg/L in hydrolysis acidification process while COD and total phenols concentration in wastewater with air stripping dropped from  $5,000 \pm 200$  mg/L to  $3,000 \pm 100$  and  $250 \pm 10$  mg/L to  $150 \pm 5$  mg/L indicating the ability to remove high COD and phenol (Zhao et al. 2015).

### The comparison of release of VFAs

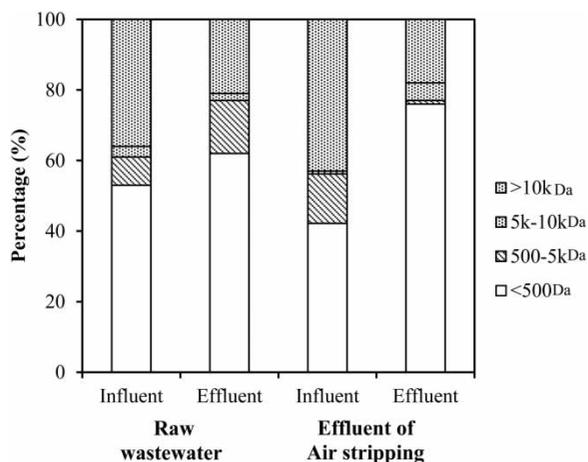
To investigate the effects of hydrolysis acidification on conversion from macromolecule organics to micromolecule organics, the raw wastewater and the effluent of air stripping were introduced to the hydrolysis acidification process. The comparison of VFA concentration is shown in Figure 5.

The VFAs is a key indicator of the degree of acid fermentation. During 0–15 d, a large amount of soluble organic compounds were converted to VFAs, and the VFA concentration rapidly increased up to  $1,300 \pm 100$  mg/L. During 15–30 d, the VFA concentration was stabilized at

**Figure 5** | The variation of VFA concentration during hydrolysis acidification process ( $n = 3$ ).

**Table 3** | The composition of VFA released in hydrolysis acidification process after air stripping (the proportion was measured by weight)

VFA	Acetic acid	Propionic acid	Isobutyric acid	Ethacetic acid	Isopentonic acid	Pentanoic acid
wt(%)	50	20.2	3.6	7.4	15.6	3.2

**Figure 6** | The changes of organic molecular weight distribution during hydrolysis acidification.**Table 4** | Analytic results of sludge sequencing sequence

Sample	Sequence numbers	Number of sequences after quality control	Mean sequence length
Effluent of air stripping	40,356	39,707	415.81
Raw wastewater	41,803	41,118	421.16

1,300 ± 100 mg/L, and the components of VFA in wastewater after air stripping are shown in Table 3. In addition, there are more refractory organics in the raw water and the concentration of ammonia nitrogen and total phenol is high, which inhibited the hydrolysis acidification process. Therefore, the rate of increase of VFA concentration in raw water is significantly lower than that of VFA in the pre-stripped wastewater.

As shown in Table 3, in the stage (15 d) with stable and exuberant production of VFAs, the acetic acid was the main component of VFAs, accounting for about 50%. In VFAs,

the acetic acid is not only the direct utilized substrate for methanogenesis, but also the carbon source which is easy to utilize in subsequent biological nitrogen removal.

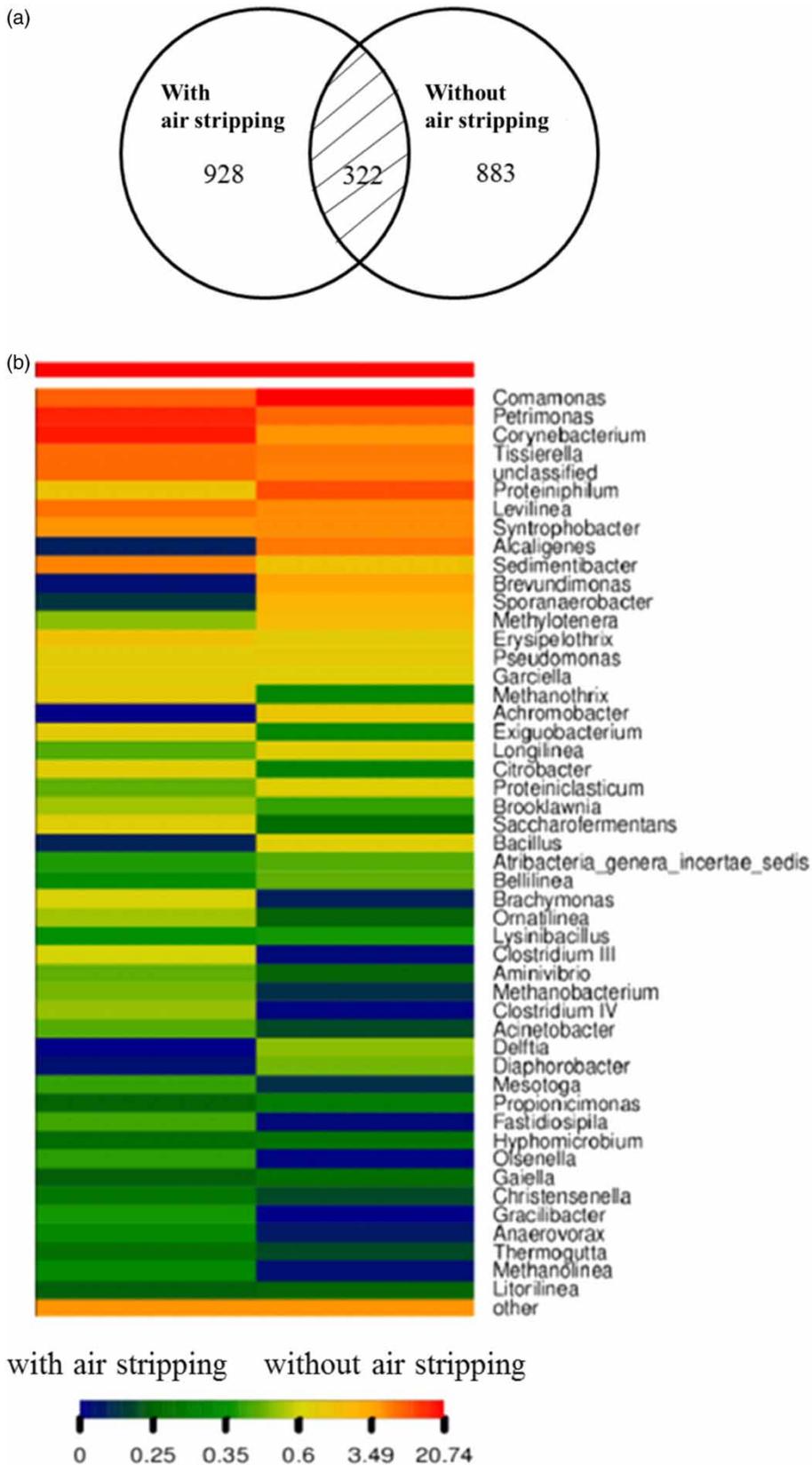
### Molecular weight distribution of effluent after hydrolysis acidification

To investigate degradation in the hydrolysis acidification process, the molecular weight distribution of organics in liquid phase characterized by the TOC concentration in the influent and effluent of hydrolysis acidification was measured, and the results are shown in Figure 6.

The TOC concentration at >10 k, 5 k–10 k, 500 k–5 k and 0–500 Da accounted for 36%, 3%, 8% and 53% in raw wastewater, and accounted for 21%, 2%, 15% and 62% in effluent of hydrolysis acidification. When treating the effluent of air stripping, the TOC concentration accounted for 43%, 0.8%, 14% and 42.2% in the influent and 18%, 5%, 1% and 76% in the effluent. It was obvious that the air stripping had the effect of removing small molecules, and there was significant reduction of the proportion of the molecule which was <500 Da after air stripping. In addition, the macromolecule organics were converted into VFAs with small molecule weight in the hydrolysis acidification process (Liu *et al.* 2016), leading to the reduction of the proportion of the organics at >10 k Da. In contrast, the proportion of molecules that were 0–500 k Da and 500–5 k Da in the raw wastewater increased and the proportion of molecules that were 5 K–10 K in the effluent of the stripping increased. This was mainly because some of the organic compounds that were not degraded were within the range of this molecular weight, and the microbial products within the range of this molecular weight were produced during microbial metabolism (Liu *et al.* 2016). However, due to the high concentration of COD, NH<sub>3</sub>-N and total phenols, the effect of hydrolysis acidification was still poor even after domestication for a period of time.

**Table 5** | Alpha diversity statistics of activated sludge microorganism

Sample	Number of sequences after quality control	OTU number	Shannon index	ACE index	Chao1 index	Coverage index
Effluent of air stripping	39,707	1,250	3.83	9,547.60	4,985.06	0.98
Raw wastewater	41,118	1,205	3.82	9,942.94	5,117.30	0.98



**Figure 7** | Comparison of microbial communities in different experimental groups.

## The comparison of microbial community in hydrolysis acidification

### Sample sequence

Phylum and genus were selected as classification units and species sequences were classified by using RDP classifier software, and the results are shown in Table 4.

From the comparison in Table 3 and 4, the microbial species in the effluent of air stripping and raw wastewater in the hydrolysis acidification process were sequenced, and the number of sequences was 39,707 and 41,118 respectively. The average lengths of these sequences were 415.81 bp and 421.16 bp, which were beneficial to diversity analysis.

### Microbial diversity analysis

The results of Alpha microbial diversity are shown in Table 5. The Shannon index of two samples was 3.83 and 3.82. The 39,707 and 41,118 sequences of the two samples were divided into 1,250 OTU and 1,205 OTU, of which the operation classification unit might be close to the genus.

### The analysis of microbial communities

The OTU number and richness thermography of genus species of two samples were compared, and the results are shown in Figure 7.

As shown in Figure 7(a), the shared OTU number between two samples was 322, while the particular OTU number of the sample with and without air stripping was 928 and 883. The toxicity and inhibition of coal gasification wastewater on sludge in hydrolysis acidification were reduced after air stripping, resulting in better degradation and transformation of pollutants, which was conducive to the formation of a wide range of microbial flora. Figure 7(b) shows the genus-level species richness thermal map. In the thermal map, the rows represent the community structure, and the color indicates the relative abundance of the species.

Several typical microbial flora shared in the two samples were compared and the results are shown in Table 6. In addition, we summarized and analyzed the functions of various microbial flora, as shown in Table 7.

To conclude, the activated sludge in hydrolysis acidification could be mainly divided into three categories according to the function of the flora: (1) hydrolytic fermentation flora: *Erysipelothrix*, *Sedimentibacter*, *Corynebacterium*; (2) syntrophic acetogenic flora: *Petrimonas*, *Proteiniphilum*, *Tissierella*, *Syntrophobacter*; (3) flora with tolerance of

Table 6 | Comparison of typical microbial flora

Genus	Sample with air stripping	Sample without air stripping
<i>Comamonas</i>	3,711	8,514
<i>Petrimonas</i>	6,387	3,657
<i>Corynebacterium</i>	6,770	1,528
<i>Tissierella</i>	3,522	2,863
<i>Proteiniphilum</i>	628	4,634
<i>Levilinea</i>	3,250	1,977
<i>Syntrophobacter</i>	1,680	2,196
<i>Sedimentibacter</i>	2,181	648
<i>Erysipelothrix</i>	657	387

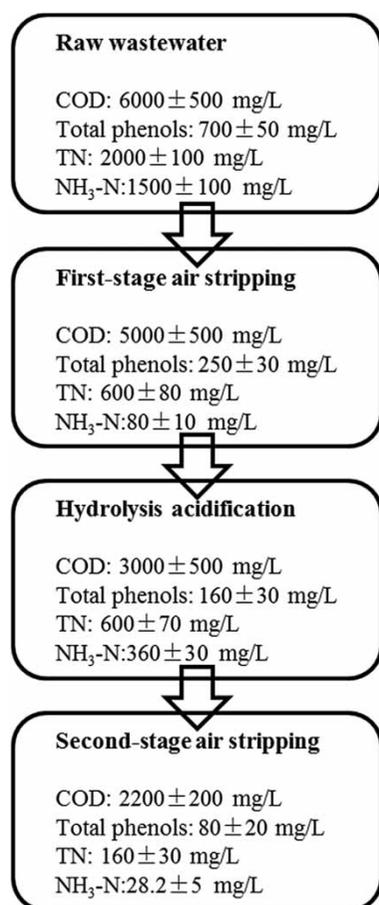
high organic load: *Comamonas* and *Levilinea*. By comparison, the sample with air stripping had more microbial species, and the species were more abundant. Therefore, air stripping can effectively reduce the toxicity and inhibition of coal gasification wastewater to the sludge in the hydrolysis acidification process and promote the formation of dominant flora corresponding to various pollutants.

Table 7 | The classification of the functions of various microbial flora

Genus	Function	References
<i>Comamonas</i>	Degrading chlorinated nitrobenzene compounds	Liu et al. (2016)
<i>Petrimonas</i>	The capacity of fermentative hydrogen production	Li et al. (2016)
<i>Corynebacterium</i>	A facultative anaerobe/one of the most important microorganisms in hydrolysis acidification process	Matsutani et al. (2017)
<i>Tissierella</i>	Degrading cellulose at hydrolysis stage/involved in the VFA fermentation at acidification stage	Zhao et al. (2016)
<i>Proteiniphilum</i>	Oxidizing various higher fatty acid and alcohols into acetic acid and H <sub>2</sub>	Huang et al. (2016)
<i>Levilinea</i>	Adapting to the complex environment of industrial wastewater	Zhang et al. (2017)
<i>Syntrophobacter</i>	Utilizing propionic acid to produce acetic acid and CO <sub>2</sub>	Pandey et al. (2011)
<i>Sedimentibacter</i>	A typical microorganism in the ethanol fermentation stage	Maspolim et al. (2015)
<i>Erysipelothrix</i>	Effectively decomposing lactic acid in the process of lactic acid metabolism	Zhang et al. (2015)

## Optimal treatment process for coal gasification wastewater

Based on the above discussion, the 'air stripping – hydrolysis acidification – air stripping' process was recommended as the pretreatment, as shown in Figure 8. The main reasons for this process were high TN concentration, low C/N and poor biodegradability. The first-stage air stripping played the role of removing highly concentrated  $\text{NH}_3\text{-N}$  and partial micromolecule phenols, which could reduce the toxicity and inhibition on microbial activities in the subsequent hydrolysis acidification process. The intermediate hydrolysis acidification stage transformed organic-N to  $\text{NH}_3\text{-N}$ , which could not be removed in the first-stage air stripping. The second-stage air stripping further removed the  $\text{NH}_3\text{-N}$  and micromolecule phenols produced in the hydrolysis acidification process, improved the biodegradability of the coal gasification wastewater, and reduced the subsequent pressure of biological nitrogen removal.



**Figure 8** | The removal performance in 'air stripping – hydrolysis acidification – air stripping' process.

## CONCLUSIONS

The optimal ammonia removal in the air stripping process reached 97%, which was achieved at G/L of 1,500 with a duration of 6 h. Air stripping could remove large amounts of  $\text{NH}_3\text{-N}$  and volatile phenols, which reduced the toxicity and inhibition of wastewater to microorganisms, and correspondingly the hydrolysis acidification had better performance in wastewater with air stripping before. Moreover, an additional stage of air stripping after the hydrolysis process could further remove  $\text{NH}_3\text{-N}$  and TN effectively. The main cost of the process is energy consumption, and the water treatment costs about \$3.50/ton. Compared with other pretreatment technologies (with the same removal effect), this process has a high removal rate of ammonia nitrogen and low cost. The 'air stripping – hydrolysis acidification – air stripping' process is recommended as the pretreatment for coal gasification wastewater to improve the biodegradability and to reduce subsequent pressure of biological nitrogen removal.

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## REFERENCES

- APHA-AWWA-WEF 2005 *Standard Methods for the Examination of Water and Wastewater*, 21st edn. American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC.
- Feng, D., Yu, Z., Chen, Y. & Qian, Y. 2009 Novel single stripper with side-draw to remove ammonia and sour gas simultaneously for coal-gasification wastewater treatment and the industrial implementation. *Ind. Eng. Chem. Res.* **48**, 5816–5823.
- Guštin, S. & Marinšek-Logar, R. 2011 Effect of pH, temperature and air flow rate on the continuous ammonia stripping of the anaerobic digestion effluent. *Process Safety and Environmental Protection* **89** (1), 61–66.
- He, Y., Wang, Y. & Song, X. 2016 High-effective denitrification of low C/N wastewater by combined constructed wetland and biofilm-electrode reactor (cw-ber). *Bioresour. Technol.* **203**, 245–251.
- Hou, B., Han, H., Zhuang, H., Xu, P., Jia, S. & Li, K. 2015 A novel integration of three-dimensional electro-Fenton and

- biological activated carbon and its application in the advanced treatment of biologically pretreated Lurgi coal gasification wastewater. *Bioresour. Technol.* **196**, 721–725.
- Huang, Y., Hou, X., Liu, S. & Ni, J. 2016 Correspondence analysis of bio-refractory compounds degradation and microbiological community distribution in anaerobic filter for coking wastewater treatment. *Chem. Eng. J.* **304**, 864–872.
- Jia, S., Han, H., Zhuang, H. & Hou, B. 2015 The pollutants removal and bacterial community dynamics relationship within a full-scale British gas/Lurgi coal gasification wastewater treatment using a novel system. *Bioresour. Technol.* **200**, 103–110.
- Jin, B., Wang, S., Xing, L., Li, B. & Peng, Y. 2016 Long term effect of alkali types on waste activated sludge hydrolytic acidification and microbial community at low temperature. *Bioresour. Technol.* **200**, 587–597.
- Krishna Mohan, T. V., Nancharaiah, Y. V., Venugopalan, V. P. & Satya Sai, P. M. 2016 Effect of C/N ratio on denitrification of high-strength nitrate wastewater in anoxic granular sludge sequencing batch reactors. *Ecological Engineering* **91**, 441–448.
- Li, H. Q., Han, H. J., Du, M. A. & Wang, W. 2011 Removal of phenols, thiocyanate and ammonium from coal gasification wastewater using moving bed biofilm reactor. *Bioresour. Technol.* **102**, 4667–4673.
- Li, Y., Han, F. & Xu, H. 2014 Potential lipid accumulation and growth characteristic of the green alga *Chlorella* with combination cultivation mode of nitrogen (N) and phosphorus (P). *Bioresour. Technol.* **174** (9), 24–32.
- Li, G., Zhang, P., Wang, Y., Sheng, Y., Lv, X. & Yin, J. 2016 Enhancing biological denitrification with adding sludge liquor of hydrolytic acidification pretreated by high-pressure homogenization. *Int. Biodeter. Biodegr.* **113**, 222–227.
- Liu, J., Yu, Y., Chang, Y., Li, B., Bian, D. & Yang, W. 2016 Enhancing quinoline and phenol removal by adding *Comamonas testosteroni*, bdq06 in treatment of an accidental dye wastewater. *Int. Biodeter. Biodegr.* **115**, 74–82.
- Maspolim, Y., Zhou, Y., Guo, C., Xiao, K. & Ng, W. J. 2015 The effect of pH on solubilization of organic matter and microbial community structures in sludge fermentation. *Bioresour. Technol.* **190**, 289–298.
- Matsutani, M., Nantapong, N., Murata, R., Paisrisan, P., Hirakawa, H. & Kataoka, N. 2017 Complete genome sequencing of newly isolated thermotolerant *Corynebacterium glutamicum*, N24 provides a new insights into its thermotolerant phenotype. *J. Biotechnol.* **247**, 29–33.
- Oladipo, A. A., Adeleye, O. J., Oladipo, A. S. & Aleshinloye, A. O. 2017 Bio-derived MgO nanopowders for BOD and COD reduction from tannery wastewater. *J. Water Process Eng.* **16**, 142–148.
- Pandey, P. K., Ndegwa, P. M., Soupir, M. L., Alldredge, J. R. & Pitts, M. J. 2011 Efficacies of inocula on the startup of anaerobic reactors treating dairy manure under stirred and unstirred conditions. *Biomass Bioenerg.* **35**, 2705–2720.
- Poszytek, K., Pyzik, A., Sobczak, A., Lipinski, L., Sklodowska, A. & Drewniak, L. 2017 The effect of the source of microorganisms on adaptation of hydrolytic consortia dedicated to anaerobic digestion of maize silage. *Anaerobe* **46**, 46–55.
- Wang, W., Han, H., Yuan, M. & Li, H. 2010 Enhanced anaerobic biodegradability of real coal gasification wastewater with methanol addition. *J. Environ. Sci.* **22**, 1868–1874.
- Wang, W., Han, H., Yuan, M., Li, H., Fang, F. & Wang, K. 2011 Treatment of coal gasification wastewater by a two-continuous UASB system with step-feed for COD and phenols removal. *Bioresour. Technol.* **102** (9), 5454–5460.
- Wang, W., Zhang, J., Wang, S., Shen, J. & Pan, S. L. 2014 Oxygen-limited aeration for relieving the impact of phenolic compounds in anaerobic treatment of coal gasification wastewater. *Int. Biodeter. Biodegr.* **95**, 110–116.
- Wen, Y. 2007 *Study on removal of ammonia nitrogen from coking wastewater by blowing and desorption*. MSc Thesis, Wuhan University of Science and Technology, pp. 24–30.
- Xie, X. H., Liu, N., Yang, B., Yu, C. Z., Zhang, Q. Y., Zheng, X. L., Xu, L. Y., Li, R. & Liu, J. S. 2016 Comparison of microbial community in hydrolysis acidification reactor depending on different structure dyes by Illumina MiSeq sequencing. *Int. Biodeter. Biodegr.* **111**, 14–21.
- Ye, S., Wu, H., Zhang, C. & Qin, X. 2008 Study of the hydrolytic acidification-SBR process in aquatic products processing wastewater treatment. *Desalination* **222**, 318–322.
- Yu, Z., Chen, Y., Feng, D. & Qian, Y. 2010 Process development, simulation, and industrial implementation of a new coal-gasification wastewater treatment installation for phenol and ammonia removal. *Ind. Eng. Chem. Res.* **49**, 2874–2881.
- Zeng, X., Yao, H., Ning, M., Fan, Y., Wang, C. & Shi, R. 2011 Synthesis, characterization and adsorption performance of a novel post-crosslinked adsorbent. *J. Colloid Interface Sci.* **354**, 353–358.
- Zhang, A., Xu, C., Wang, H., Lei, C., Liu, B. & Guan, Z. 2015 Presence and new genetic environment of pleuromutilin-lincosamide-streptogramin a resistance gene *Isa(E)* in *Erysipelothrix rhusiopathiae* of swine origin. *Vet. Microbiol.* **177**, 162–167.
- Zhang, J., Li, W., Lee, J., Loh, K. C., Dai, Y. & Tong, Y. W. 2017 Enhancement of biogas production in anaerobic co-digestion of food waste and waste activated sludge by biological co-pretreatment. *Energy* **137**, 479–486.
- Zhao, Q., Han, H., Xu, C., Zhuang, H., Fang, F. & Zhang, L. 2015 Effect of powdered activated carbon technology on short-cut nitrogen removal for coal gasification wastewater. *Bioresour. Technol.* **142**, 179–185.
- Zhao, J. G., Chen, X. R., Zhao, J., Lin, F. K., Bao, Z., He, Y. X., Wang, L. & Shi, Z. D. 2015 Toxicity in different molecular-weight fractions of sludge treating synthetic wastewater containing 4-chlorophenol. *Int. Biodeter. Biodegr.* **104** (10), 251–257.
- Zhao, B., Liu, J., Frear, C., Holtzapfel, M. & Chen, S. 2016 Consolidated bioprocessing of microalgal biomass to carboxylates by a mixed culture of cow rumen bacteria using anaerobic sequencing batch reactor (ASBR). *Bioresour. Technol.* **222**, 517–522.
- Zhuang, H., Han, H., Jia, S., Zhao, Q. & Hou, B. 2014 Advanced treatment of biologically pretreated coal gasification wastewater using a novel anoxic moving bed biofilm reactor (ANMBBR)–biological aerated filter (BAF) system. *Bioresour. Technol.* **157** (4), 223–230.