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# Inhibitory effect of high phenol concentration in treating coal gasification wastewater in anaerobic biofilter

*Yajie Li*[1,2], *Salma Tabassum*[2], *Chunfeng Chu*[2], *Zhenjia Zhang*[2,*]

1. *School of Environmental Science and Engineering, Suzhou University of Science and Technology, Suzhou 215009, China*
2. *School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China*

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ABSTRACT

In this paper, the inhibition of methanogens by phenol in coal gasification wastewater (CGW) was investigated by both anaerobic toxicity tests and a lab-scale anaerobic biofilter reactor (AF). The anaerobic toxicity tests indicated that keeping the phenol concentration in the influent under 280 mg/L could maintain the methanogenic activity. In the AF treating CGW, the result showed that adding glucose solution as co-substrate could be beneficial for the quick start-up of the reactor. The effluent COD and total phenol reached 1200 and 100 mg/L, respectively, and the methane production rate was 175 mL $CH_4$/g COD/day. However, if the concentration of phenol was increased, the inhibition of anaerobic micro-organisms was irreversible. The threshold of total phenol for AF operation was 200–250 mg/L. The extracellular polymeric substances (EPS) and particle size distribution of anaerobic granular sludge in the different stages were also examined, and the results indicated that the influence of toxicity in the system was more serious than its effect on flocculation of EPS. Moreover, the proportion of small size anaerobic granular sludge gradually increased from 10.2% to 34.6%. The results of high through-put sequencing indicated that the abundance of the *Chloroflexi* and *Planctomycetes* was inhibited by the toxicity of the CGW, and some shifts in the microbial community were observed at different stages.




## Introduction

Nowadays, coal gasification technology is gaining more and more attention because of the current increase in the consumption of natural gas. However, a large quantity of coal gasification wastewater (CGW) is generated during the coal gasification processes, which contains a lot of toxic and refractory compounds such as phenolic compounds, cyanide, pyridine, and long-chain alkanes and so on (Cui et al., 2017; Xu et al., 2017). Although pretreatment by ammonia-stripping and solvent extraction is an effective method for cutting down ammonia and phenolic compounds (Li et al., 2016a), the residual refractory organic compounds in CGW remain high, of which phenolic compounds account for 40%–50% (Xu et al., 2014). Hence, it is necessary to explore an effective treatment method for the removal of phenols from CGW. The absorption of concentrated phenol solutions may cause severe pain, renal irritation, and shock in humans, and fatalities have been reported. A total dose of 1.5 g may be fatal. Phenols at high concentration do harm to most microorganisms and are also resistant to biodegradation (Hussain and Dubey, 2014).

Anaerobic treatment is by far the most widely applied and cost-effective process for wastewater treatment, improving the degradation rate for refractory organic compounds and enhancing the wastewater biodegradability (Li et al., 2016b; Park et al., 2008). Tests increasing phenol loads (from 100 to

* *Corresponding author*. E-mail: zjzhang@sjtu.edu.cn (Zhenjia Zhang).

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5000 mg/L) as the sole carbon source in a semi-continuous mesophilic anaerobic adaptation experiment were carried out, using an unadapted microbial community from a standard biogas plant. Phenol was completely reduced at starting concentrations of up to 2000 mg/L (Wirth et al., 2015). However, the large amount of granular activated carbon required as an absorber would be costly on an industrial scale (Ahmaruzzaman, 2008; Lin and Juang, 2009). Hence, there is a need for an advanced anaerobic reactor that is more practical in terms of investment costs and operability for improving the anaerobic biodegradation of CGW. The anaerobic biofilter (AF) is a kind of anaerobic reactor filled with fillers. Enriched functional bacteria grow on the surface of fillers and help in the degradation of toxic pollutants in CGW (Wang et al., 2013). Moreover, the mixed liquor suspended solids (MLSS) of an AF can reach to 30 g VSS/L (Wang et al., 2014a). The spatial distribution of microorganisms in the AF is conducive to reducing the effects of toxic pollutants on vulnerable bacteria. However, the start-up of anaerobic reactors always requires a long time, especially in the treatment of toxic and refractory wastewater. Nowadays, in order to reduce the startup time in anaerobic systems, anaerobic co-digestion has been employed for the disposal of refractory or toxic wastewater. Moreover, the anaerobic co-digestion of organic waste has the potential to make a significant contribution to the generation of renewable energy (Larsen et al., 2013).

Directly utilizing refractory pollutants as a carbon or energy source is difficult for anaerobic micro-organisms, but when another easily utilized carbon or energy source is added in the influent, the refractory substances can be degraded efficiently (Li et al., 2016a). Co-digestion using methanol addition to improve the biodegradation of CGW has previously been investigated. Q5 When a methanol concentration of 500 mg COD/L was added (organic loading rate 3.5 kg COD/m$^3$/day and phenol loading rate 0.6 kg/m$^3$/day), the corresponding maximum COD and phenol removal rates were 71% and 75%, respectively (Wang et al., 2010). An advanced anaerobic expanded granular sludge bed (AnaEG) was also developed recently by our research group for the treatment of CGW that adopted glucose as the substrate; it took about 87 days to start up the reactor (Li et al., 2014), which is faster than in previous reports.

In this study, anaerobic toxicity and recovery tests were performed, and then a laboratory-scale AF reactor was adopted to treat CGW. The aim was to explore the quick start-up of the reactor with the aid of co-digestion, and to determine the inhibitory effect of high phenol concentrations as well as the inhibition threshold of phenol. Finally, high-throughput sequencing was used to characterize the microbial communities during different stages of the AF operation. The abundance of microorganisms in the sludge in relation to the different stages of the operation was discussed.

## 1. Materials and methods

### 1.1. Anaerobic biofilter reactor

The AF was made of cylindrical plexiglass and filled with soft fillers. The influent was pumped into the bottom of the reactor and the effluent flowed out from the top. The AF was operated at 35°C with the effective volume of 13.4 L. The hydraulic retention time (HRT) was controlled at 96 hr and the methane content was analyzed by passing the gas emitted through a 3 mol/L NaOH solution followed by collection in a gas collection bag. The volume of methane ($CH_4$) was monitored using a gas flow meter.

### 1.2. Inoculated sludge

The inoculated anaerobic activated sludge was taken from an expanded granular sludge bed (EGSB) treating starch wastewater. The inoculation volume was 20% of the effective volume of the reactor. The suspended solids (SS) and volatile suspended solids (VSS) in the reactor were 8.3 g/L and 4.8 g/L, respectively.

### 1.3. Coal gasification wastewater

Coal gasification wastewater was obtained from the Harbin Coal Chemical Industry Co. Ltd., Harbin, China. It was pretreated by phenol extraction and ammonia stripping. Some available substances can be recovered by pretreatment, and the toxicity of the CGW can be decreased for the subsequent bio-treatment. The characteristics of the CGW used in our study are shown in Table 1. (See Table 2.) Q6

### 1.4. Methanogenic toxicity batch assays Q7

Methanogenic toxicity batch tests were conducted in 250 mL sealed bottles, seeded with anaerobic sludge. Five sealed bottles, namely B1, B2, B3, B4 and B5, were prepared and some seeded sludge was added. A glucose solution (3 g COD/L) was used as the medium and $NaCO_3$ was used to adjust the pH to 6.8–7.2. The phenol concentrations were controlled at 0 (control), 280, 400, 550, and 800 mg/L, respectively, for the five samples. Moreover, a blank sample with only pure water and anaerobic granular sludge without the addition of glucose solution was also prepared. Methane production was measured by a gas displacement device filled with 3 mol/L NaOH solution. Nitrogen gas was bubbled up into each bottle to remove air and the assays were performed at 35°C in an oscillating shaker at 120 r/min. All batch tests were repeated three times.

The relative activity (RA) was adopted to determine the degree of inhibition by phenol. RA can be calculated as follows:

$$RA = \frac{V_t - V_{blank}}{V_{control} - V_{blank}} \times 100\% \quad (1)$$

where, $V_t$ is the cumulative methane production of the sample at a certain time; $V_{blank}$ is the cumulative methane production of the blank sample at a certain time; $V_{control}$ is the cumulative methane production of the control sample at a certain time. RA of 75%–95% indicated slight inhibition; RA of 40%–75% indicated moderate inhibition; and RA < 40% indicated severe inhibition (Driessen et al., 1994).

### 1.5. The effect of phenol concentration on the treatment performance of the AF operation

The effect of phenol concentration on the treatment performance of the AF was investigated. The HRT was controlled at



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