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# Oxygen-limited aeration for relieving the impact of phenolic compounds in anaerobic treatment of coal gasification wastewater

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ABSTRACT

Anaerobic digestion is an efficient tool for improving aerobic biodegradability of coal gasification wastewater, but the shock loading usually results in a failed performance of the anaerobic process. The oxygen-limited aeration was conducted to handle the impact of coal gasification wastewater in an anaerobic expanded-bed GAC reactor. Results indicated that the oxygen-limited aeration of 0–0.5 $mgO_2$/L could shorten the recovery time from 23 to 11 days under natural condition, and the addition of 1 g/L PAC could reduce the recovery time to 14 days. The corresponding removal efficiencies of COD and phenolic compounds could increase to above 60 and 50%, respectively. The phenol utilization rates of sludge reached 83.9 ± 5.7 mg phenol/(g VSS d) after recovery of reactor performance by using oxygen-limited aeration, and only 70.4 ± 3.8 mg phenol/(g VSS d) by using natural recovery and 65.2 ± 5.9 mg phenol/(g VSS d) by using PAC addition. The recovery strategy of oxygen-limited aeration might be helpful to relieve the inhibitory effect of phenolic compounds and restore metabolic activity of anaerobic bacteria.



## 1. Introduction

At present, in China, coal syngas is produced in large quantities to meet the requirement of natural gas and to provide the raw materials in the manufacture of chemical products, such as methanol, synthetic ammonia and olefin (Chen and Xu, 2010; Yoon and Lee, 2012). However, the difficulties in treatment of coal gasification wastewater (CGW) are far beyond the imagination. The CGW contained phenolic compounds, heterocyclic and polycyclic aromatic hydrocarbons, long chain alkanes, ammonia, thiocyanate, and cyanide (Sharma et al., 2012). Among these compounds, phenolic compounds were the main organic pollutants and contributed about 30–60% of the COD content (Wang et al., 2010). Especially, the concentration of total phenols was still around 400 mg/L even though the pretreatment of solvent extraction (Gai et al., 2008; Yu et al., 2010). The toxicity and recalcitrance of phenolic compounds inhibited its biological degradation. Generally, the CGW was treated using anoxic–oxic process or conventional activated sludge process, because the most of phenolic compounds could be degraded readily under aerobic conditions (Li et al., 2011; Kim et al., 2011a,b). However, the aerobic biological treatment could not reduce the concentrations of COD and phenolic compounds in the CGW to meet the national emission standards.

Many attentions had paid to anaerobic biological technologies in the treatment of CGW because of their advantages, which included low energy consumption, detoxification and improvement of biodegradability (Fox et al., 1990; Nakhla et al., 1990; Wang et al., 2011). In the 1980s of last century, anaerobic activated carbon filter was used to treat the CGW, while excellent removal efficiency of organic matter was achieved by a combination of anaerobic fermentation and adsorption (Suidan et al., 1983). Operational techniques using expanded-bed or fluidized-bed anaerobic granular activated carbon (GAC) bioreactors were developed subsequently for reducing the cost in treating the CGW (Pfeffer and Suidan, 1989; Fox et al., 1990; Nakhla et al., 1990). Because of the toxicity of CGW, the full strength wastewater was not treatable at organic loading rates (OLR) above 10 kg COD/($m^3$ d) while the GAC reactors treating the diluted wastewater could achieve around 90% removal of COD and over 99.5% reduction of phenolic compounds at OLR range of 32–34 kg COD/($m^3$ d) in the synthetic CGW (Nakhla and Suidan, 1995). Furthermore, the periodic partial replacement of GAC was also an effective way to overcome the toxicity of the wastewater in the anaerobic GAC reactors (Pfeffer and Suidan, 1989; Nakhla et al., 1990). Except the anaerobic GAC technologies, anaerobic treatment of actual CGW was investigated in the conventional anaerobic processes, such as two-continuous UASB

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reactors and addition of PAC in the UASB reactor (Wang et al., 2011; Wang and Han, 2012). These results demonstrated that anaerobic digestion had a significant improvement effect on the biodegradability of CGW, but the COD removal exceeding 70% was difficult at OLR range of 2.5–3.0 kg COD/($m^3$ d) in the absence of activated carbon. Obviously, anaerobic expended-bed GAC (EB-GAC) reactor was more efficient than the conventional anaerobic processes for treating the actual CGW.

In fact, the composition of actual CGW was complicated and varied from one gasification plant to another, depending on the quality of raw coal, gasifier types, gasification temperature, and recovery methods of byproducts. So, the application of anaerobic processes in the treatment of CGW had been hampered by a lack of understanding of factors associated with process stability, such as shock loading and recovery methods. In the anaerobic GAC reactors, one method using the periodic partial replacement of GAC had been successfully applied for reducing the toxicity and enhancing the stability in the laboratory studies (Nakhla et al., 1990). However, on an industrial scale, the shock loading appeared frequently in the influent, and the periodic partial replacement of large volumes of GAC was unreasonable and very costly.

The aim of this study was to investigate the effect of oxygen-limited aeration and addition of powdered activated carbon (PAC) on the recovery process of anaerobic EB-GAC reactor treating CGW after shock loading. The start-up of the anaerobic EB-GAC reactor was recorded and the impact of shock loading was evaluated on the operation of the EB-GAC reactor. Comparing with the natural recovery, the recovery process and the mechanism of oxygen-limited aeration and addition of PAC were analyzed.

## 2. Material and methods

### 2.1. Experimental setup and inoculum

The EB-GAC reactor was constructed of cylindrical plexiglass column with an internal diameter of 5 cm and a working volume of 1 L. A supporting layer was placed at the bottom of the reactor to support the GAC and distribute influent wastewater and air. The temperature was maintained at 35 ± 2 °C by a constant temperature controller through electric heating. The reactor was initially charged with 100 g of 16 × 20 mesh GAC. An internal recycle wastewater of 0.2 L/min provided an initial expansion of GAC bed in the reactor. The seed sludge was obtained from a full-scale anaerobic reactor at a CGW treatment plant in the northeast region of China. The initial suspended solids (SS) and volatile suspended solids (VSS) added to the reactor were around 10.5 and 7.1 g/L, respectively.

### 2.2. Wastewater characteristics and GC-MS analysis

The organic composition of CGW after pretreatment by ammonia stripping and phenol extraction with methyl isobutyl ketone (MIBK) was presented in the previous study (Wang et al., 2011). The organic composition of GC-MS analysis is shown in Fig. 1. It was not difficult to find that phenolic compounds, such as phenol, *p*-cresol and 2-[(trimethylsily)oxy]-phenol, were the major organic constituents of the CGW. The actual CGW had a complicated composition and the wastewater characteristics with their constituent concentrations expressed in (mg/L) were as follows: COD (2023 ± 146), $BOD_5$ (685 ± 47), phenolic compounds (392 ± 35), volatile fatty acids (759 ± 84), ammonia nitrogen (129 ± 10), oil (45 ± 6), thiocyanate (65 ± 7) and pH (7.5 ± 0.3). The macro- and micro-nutrients were added to the anaerobic EB-GAC reactor as described in the early work (Wang et al., 2010).

### 2.3. Start-up and operation

The operational schedule of the EB-GAC reactor was divided into four phases: start-up, natural recovery, addition of PAC, and oxygen-limited aeration. Normal operation of the EB-GAC reactor was maintained at COD and phenolic concentrations of around 2000 and 400 mg/L at a HRT of 24 h. The first phase lasted for 156 days during the start-up period. For the second period extending to day 194, the organic loading on the EB-GAC reactor was maintained at 5.16 kg COD/($m^3$ d) with 24 h between days 157 and 158, while the concentrations of COD and phenolic compounds increased from 1859 and 354.5 mg/L to 5160 and 815 mg/L respectively in the influent. The anaerobic EB-GAC reactor was allowed to recover fully from the shock loading and the recovery time was defined as the time between the initiation of recovery action and the time when the removal efficiency of phenolic compounds achieved above 50%. After the natural recovery of reactor performance, the EB-GAC reactor was continuously operated at a pseudo-steady state for two weeks. Between days 195 and 196, the shock loading of 5.32 kg COD/($m^3$ d) was exerted on the EB-GAC reactor for one day time. 1 g

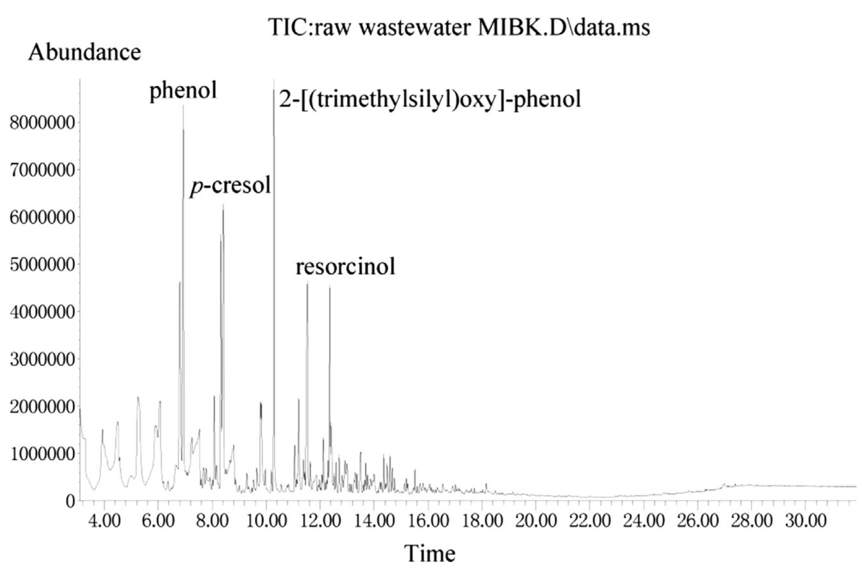


**Fig. 1.** Main organic composition in the influent by GC-MS analysis.