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Inhibition of ammonia on anaerobic digestion of synthetic coal gasification wastewater and recovery using struvite precipitation

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HIGHLIGHTS

- Struvite precipitation was provided for recovering ammonia from the CGW.
- SUR_{phenol} and SMA of sludge were decreased by 89% and 67% at 5 g TAN L^{-1}.
- SUR_{phenol} and SMA of sludge were decreased by 94% and 100% at 10 g TAN L^{-1}.
- The optimum struvite crystallization conditions were obtained in the synthetic CGW.
- Performance failure of UASB was recovered by pretreatment of struvite precipitation.

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ABSTRACT

Coal gasification wastewater (CGW) contains very high concentrations of phenols and ammonia. However, the potential impact of ammonia on the anaerobic digestion of phenols remained unclear. Firstly, the methanogens and phenols degraders had a good tolerance up to 1 g L^{-1} of total ammonia nitrogen (TAN), but the substrate utilization rate for phenol, and specific methanogenic activity of sludge were decreased by 89% and 67% at 5 g TAN L^{-1}, and 94% and 100% at 10 g TAN L^{-1}, respectively. Secondly, the optimum struvite crystallization conditions (pH = 8.5, 10 g TAN L^{-1}, n(Mg^{2+}):n(TAN):n(PO_4^{3-}) = 1:1:1) were obtained by the orthogonal tests. Thirdly, the removal efficiencies of chemical oxygen demand (COD) and phenols were recovered to around 82% and 66%, respectively in the upflow anaerobic sludge blanket reactor using the pretreatment of struvite precipitation at 10 g TAN L^{-1} and 1 g phenols L^{-1}. Therefore, anaerobic digestion coupled with struvite precipitation was considered as an alternative way for CGW treatment.

1. Introduction

Coal gasification wastewater (CGW) has attracted a lot of attentions due to high concentration of toxic pollutants and very complex composition [1]. Ammonia and phenolic compounds are the major pollutants of CGW, and their concentrations are in the range of 3-9 and 4.5–7.5 g L^{-1}, respectively [2]. However, the conventional ammonia stripping and phenols extraction process still faced a great challenge on the maintenance and operating costs. Especially, the ammonia stripping process consumed a lot of alkali and stream, and regularly exhibited unstable performance [3,4]. Therefore, an alternative way should be considered for recovering ammonia from the CGW.

Recently, some state-of-art technologies for recovery of ammonia and reduction of ammonia inhibition on anaerobic digestion have been reported. Zhang et al. developed a new bioelectrochemical method termed bipolar bioelectrodialysis to recover ammonia and sulfate thereby offsetting their toxicity on anaerobic digestion [5]. It was reported that a submersible microbial desalination cell could decrease the ammonia concentration in a continuous stirred tank reactor by in situ ammonia recovery and electricity production [6,7]. Furthermore, the hydrogen enriched biogas upgrading process could promote the tolerance on ammonia toxicity during anaerobic digestion [8]. Struvite ($MgNH_4PO_4{\cdot}6H_2O$) crystallization is one of the most widely recommended technologies for treating ammonia and phosphorus containing wastewater [9]. Struvite crystallization only occurs when the concentrations of Mg^{2+}, NH_4^+, and PO_4^{3-} exceed the struvite solubility product (K_{sp}) [10]. Approximately 88% removal of ammonia nitrogen was achieved by using the struvite crystallization combined with microwave radiation technology from coking wastewater at pH 8.5

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and P:N molar ratio of 1:1 for 50 min [11]. In addition, the influence of organic matter on struvite precipitation was positive on the size of the struvite crystals but passive on the reaction kinetics in acidified swine wastewater [12]. The increase of ion concentration could stoichiometrically improve crystal yield, and the purity of the precipitate depended on the concentration of ammonia nitrogen [13]. Apparently, the struvite crystallization efficiency was related to a variety of environmental and operational conditions. The ionic strength of solution, pH, temperature, stirring rate, and proportion of materials affected nucleation and growth mechanisms strongly [14]. Although the recovery of ammonia from the CGW by struvite precipitation was technically feasible, the struvite crystallization should consider a various influencing factors, such as pH, organic composition, and potential impact on the biological process. To the best of our knowledge, there was no report dealing with the struvite crystallization in the CGW containing extremely high concentration of ammonia and phenols.

Previously, anaerobic digestion has become a competent technology for CGW treatment [15]. Nevertheless, the elimination of ammonia is very necessary for anaerobic digestion due to its toxicity on the microorganism [2]. The solution total ammonia nitrogen (TAN) is present in two forms, ionized ammonium nitrogen (NH_4^+) and free ammonia nitrogen (FAN) [16]. pH and temperature directly affect the form of TAN [17]. The removal efficiency of chemical oxygen demand (COD) decreased from 96% to 85% when the influent TAN concentration was increased from 5 to 6 g L^{-1} in the anaerobic bioreactor [17]. Sung and Liu found that the concentrations of 4.92 and 5.77 g TAN L^{-1} caused a drop in COD removal efficiency as much as 17.4% and 31.8% in the anaerobic process [18]. However, the potential impact of ammonia on the anaerobic digestion of phenols remained unclear. The aims of this study were to clarify the inhibition of ammonia on the anaerobic digestion of phenols and reveal the mechanism of struvite precipitation coupled with anaerobic treatment of synthetic CGW.

2. Materials and methods

2.1. Experimental setup

The upflow anaerobic sludge blanket (UASB) reactor was made up of a plexiglass cylinder with a diameter of 7 cm and a working volume of 3.5 L, and was maintained at the temperature of 35 ± 1 °C by a jacket of warm water. Solution pH in the reactor was around 8.0. The hydraulic retention time (HRT) and the liquid up-flow velocity were controlled at 48 h and 1 m h^{-1}, respectively in the reactor. The biogas volume was measured by using liquid replacement method [19].

2.2. Inoculum and synthetic wastewater

The UASB reactor was operated for treating phenolic wastewater for a period of 280 days. The sludge concentrations were 45.2 g mixed liquor suspended solids (MLSS) L^{-1} and 30.3 g mixed liquor volatile suspended solids (MLVSS) L^{-1} in the UASB reactor at the beginning of this study. Real coal gasification wastewater contained high concentration of phenol, dihydric phenols, ammonia, and fatty acids, etc. [2]. The concentration of total phenols and fatty acids in the real coal gasification wastewater were around 6000 and 3000 mg L^{-1}, respectively. After the pretreatment of real wastewater by phenols solvent extraction process, the concentration of total phenols was in a range of 400–1000 mg L^{-1}. Therefore, the synthetic CGW consisted of sodium acetate (3.85 g L^{-1}), phenol (625 mg L^{-1}), hydroquinone (125 mg L^{-1}), catechol (125 mg L^{-1}), and resorcinol (125 mg L^{-1}), macronutrients (4 mL L^{-1}), and micronutrients (0.4 mL L^{-1}). The composition of macronutrients and micronutrients were described as previous study [19]. Ammonium chloride was used as total ammonia nitrogen source. During the whole period, the influent concentrations of COD and total phenols were maintained at 5200 and 1000 mg L^{-1}, respectively with an organic loading rate of 2.52 g COD L^{-1} d^{-1} and phenols loading rate of 0.49 g phenols L^{-1} d^{-1}.

2.3. Experimental design and operational procedure

2.3.1. The ammonia inhibition tests

The substrate utilization rates (SUR) of phenols and specific methanogenic activity (SMA) of sludge were employed to evaluate the inhibition of ammonia on the activity of methanogens and phenols degraders. The concentration of TAN was controlled at 0, 0.23, 1, 5, 10 g L^{-1}, respectively in the batch tests. The SMA and SUR tests were carried out in a series of 300 mL serum bottles and the inoculum was obtained from the UASB reactor. In the SUR tests, the mass ratio of sludge and phenols was 40:1 and the concentration of phenol, hydroquinone, resorcinol and catechol was controlled at 20 mg L^{-1}. In the SMA tests, 2 g COD L^{-1} of sodium acetate was employed as the substrate with sludge mass (g VSS): substrate COD (g) ratio of 1:1. All batch tests were firstly flushed with high purity nitrogen (99.99%) for about 1–2 min and immediately plugged. Both SUR and SMA tests were performed in triplicate and incubated at 35 °C and 140 rpm. The details of SMA and SUR tests were operated as previous study [19].

2.3.2. The orthogonal tests for struvite precipitation

Three factors including initial pH, TAN concentration, and proportion of materials n(Mg^{2+}):n(TAN):n(PO_4^{3-}) were selected in the orthogonal tests for struvite precipitation. Each factor had five levels. The levels of initial pH were 7.0, 8.0, 8.5, 9, and 10, respectively. The levels of TAN concentration were 0.23, 1, 3, 5, and 10 g L^{-1}, respectively. The levels of proportion of materials n(Mg^{2+}):n(TAN):n(PO_4^{3-}) were 1:1:1, 1:1.6:1, 1:1.9:1, 1:1:0.9, and 1:1:0.6, respectively. The tests were carried out in the beakers in triplicate. The reaction and precipitation time were set at 15 and 20 min, respectively. The results of the orthogonal tests by running the SPSS software are shown in Table S1.

2.3.3. The operational procedure of UASB reactor

In order to clarify the inhibition of ammonia on anaerobic digestion of phenolic compounds in the UASB reactor, the influent concentration of TAN was gradually increased from 0.23 to 1, 3, 5, and 10 g L^{-1}. After the occurrence of the performance failure in the reactor, the anaerobic digestion coupled with struvite precipitation was carried out to investigate the potential impact on the removals of COD and phenols. There were six phases during the operational procedure of the UASB reactor. At the phase I (1-47 days), the influent concentration of TAN was 230 mg L^{-1}. At the phase II (48-68 days), the concentration of TAN was increased to 1000 mg L^{-1}. At the phase III (69-78 days), the UASB reactor was fed with the concentration of 3000 mg TAN L^{-1} in the influent. Furthermore, at the phase IV (79-122 days), the concentration of TAN was adjusted to 5 g L^{-1} in the influent. Finally, the concentration of TAN was 10 g L^{-1} at the phase V (123-142 days) and the phase VI (143-207 days), but the struvite precipitation was used to remove ammonia from the influent at the phase VI with the reaction condition of initial pH of 8.5 and n(Mg^{2+}):n(TAN):n(PO_4^{3-}) of 1:1:1.