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Effect of alkalinity on nitrite accumulation in treatment of coal chemical industry wastewater using moving bed biofilm reactor

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ABSTRACT

Nitrogen removal via nitrite (the nitrite pathway) is more suitable for carbon-limited industrial wastewater. Partial nitrification to nitrite is the primary step to achieve nitrogen removal via nitrite. The effect of alkalinity on nitrite accumulation in a continuous process was investigated by progressively increasing the alkalinity dosage ratio (amount of alkalinity to ammonia ratio, mol/mol). There is a close relationship among alkalinity, pH and the state of matter present in aqueous solution. When alkalinity was insufficient (compared to the theoretical alkalinity amount), ammonia removal efficiency increased first and then decreased at each alkalinity dosage ratio, with an abrupt removal efficiency peak. Generally, ammonia removal efficiency rose with increasing alkalinity dosage ratio. Ammonia removal efficiency reached to 88% from 23% when alkalinity addition was sufficient. Nitrite accumulation could be achieved by inhibiting nitrite oxidizing bacteria (NOB) by free ammonia (FA) in the early period and free nitrous acid in the later period of nitrification when alkalinity was not adequate. Only FA worked to inhibit the activity of NOB when alkalinity addition was sufficient.

Introduction

With human progress and the improvement of living standards, an increasing amount of contaminants containing nitrogen have been discharged into the environment. The large amount of nitrogen discharged into water bodies has undermined the nitrogen cycle in nature, causing world-wide eutrophication that occurs repeatedly. Nitrogen removal has been a hot and difficult research issue in the environmental protection area. Numerous new theories and technology have been developed on the basis of traditional treatment theories and processes. Several typical new theories and processes have emerged such as: nitrogen removal via nitrite pathway (Hellinga et al., 1998; Van Hulle et al., 2007), anaerobic ammonium oxidation (Mulder et al., 1995; Strous et al., 1998), the combination of partial nitrification to nitrite and anaerobic ammonium oxidation (CANON) (Strous et al., 1999; Third et al., 2001) and the enhanced biological nitrification bacteria process (Salem et al., 2002), drawing a considerable amount of attention of scholars. New nitrogen removal theories and processes have shown great advantages compared to traditional theories and technology, especially nitrogen removal via the nitrite pathway and CANON, which are both based on partial nitrification to nitrite. The nitrite pathway is a more practical process among these new processes. Theoretically, nitrogen removal via nitrite yields a 25% reduction in oxygen demand and 40% reduction in carbon source requirement for denitrification.

Coal chemical industry wastewater is discharged in the processes of coal gasification and coal chemical production (Yang et al., 2006; Wang et al., 2010), the composition of which is very complex, containing various toxic compounds and a large number of refractory organic and inorganic contaminants, with poor biodegradability (Marañón et al., 2008). The wastewater characteristics vary significantly according to the coal quality used in the production. More than 244 kinds of organic com-

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pounds have been detected in the wastewater. Phenolic compounds are the main pollutants. The wastewater also contains polycyclic aromatic hydrocarbons, heterocyclic compounds, long-chain hydrocarbons, ammonia, cyanide and thiocyanate (Nakhla et al., 1990; Gai et al., 2008; Felföldi et al., 2010; Yu et al., 2010). Although the wastewater is pretreated via ammonia-stripping, it also contains a high concentration of ammonia. Nitrogen removal was unsatisfactory in the suspended activated sludge process due to the presence of toxic and inhibitory matter and limited available carbon sources (Kumar et al., 2000). Under high concentrations of phenolic compounds and inhibitors, nitrifying bacteria were out-competed in the suspended activated sludge system by the fast growth of heterotrophic microorganisms (Kim et al., 2007). However, the moving bed biofilm reactor has proved to be an effective process to remove both organic contaminants and ammonia in the treatment of coal chemical industry wastewater (Li et al., 2011). Considering the limited carbon source characteristic and complex composition of coal chemical industry wastewater, nitrogen removal via a nitrite pathway that requires less carbon source is a desirable method for the treatment of coal chemical industry wastewater. Alkalinity plays a vital role in nitrification, especially carbonate alkalinity. Alkalinity is not only the inorganic carbon source of heterotrophic nitrifying bacteria, but also balances the acid-base level of the mixture, affecting the state of matter present in aqueous solution. In this context, a moving bed biofilm reactor (MBBR) was adopted to treat coal chemical industry wastewater to investigate the effects of variations of nitrogen and alkalinity on the treatment process. The study involved five operation phases with different alkalinity dosage ratios. The effect of alkalinity on nitrification to nitrite and nitrite accumulation were the key issues to be investigated. The evolution of NH_4^+-N removal and nitrite accumulation were studied by raising the alkalinity dosage ratio stepwise.

1 Materials and methods

1.1 Experimental apparatus

The MBBR was a cylindrical Plexiglas reactor with an internal diameter of 120 mm and height of 450 mm. The effective volume of the MBBR was 4.85 L, followed by a 0.5 L settling tank. The suspended carriers used in the MBBR were circular polyethylene flakes, with a diameter of 22 mm and thickness of 1.5 mm. The density of the carriers was about 0.86 g/cm^3, lower than that of water. The density of carriers with attached wet biofilm was 1.11 g/cm^3. The filling ratio (volumetric filling in empty reactor) was 35%.

1.2 Inoculum and wastewater characteristics

Seed was collected from a full-scale coal chemical industry wastewater treatment plant in Harbin, China. The sludge was gray-black and the settling characteristic was good with a sludge volume index of 83.

Real coal chemical industry wastewater used in this study was obtained from the full-scale wastewater treatment facility of a coal chemical plant in Harbin, China. The characteristics of the wastewater as following: COD 895–1109 mg/L with mean value 1065 mg/L, total phenol 198–249 with mean 226 mg/L, NH_4^+-N 92–118 mg/L with mean 108 mg/L, and pH 6.5–7.5 with mean 7.16. Considering the fluctuations in the parameters of the real wastewater, the main parameters in the influent were controlled by adding tap water into the real wastewater. The concentration of ammonia was adjusted by adding ammonia chloride. Sodium bicarbonate was added to control alkalinity.

1.3 Experiment operation

Initially, the reactor ran for 30 days as a batch system after being inoculated with the seed sludge obtained from the full-scale facility, followed by a continuous flow process. A stable biofilm was formed on the carriers, with the biomass of 0.28 g VSS/g. During batch culture, the reactor was fed with the real wastewater diluted by adding tap water, with the COD concentration increasing stepwise from 500 to 1000 mg/L in three steps. The hydraulic retention time was 36 hr in the continuous flow process during the experimental period. The motion of carriers was driven by aeration introduced at the bottom of the reactor and the dissolved oxygen (DO) concentration was kept around 1.8 mg/L. The experiment operation was divided into 5 phases (**Table 1**) and the temperature was controlled at (25 ± 2)°C throughout the experiment.

Table 1 Operational conditions

Phase	Time (days)	COD (mg/L)	Total phenols (mg/L)	$NaHCO_3$ (dosage ratio)*
I	1–15	1002.65 ± 11.72	208.99 ± 5.24	–
II	16–30	501.95 ± 7.37	114.26 ± 5.38	–
III	31–45	504.28 ± 9.44	115.50 ± 5.66	2:1
IV	45–60	1000.46 ± 9.51	210.66 ± 6.89	2:1
V	61–75	998.78 ± 8.45	209.72 ± 5.83	0.5:1
	76–90			1:1
	91–105			1.5:1
	106–120			2:1
	121–135			2.5:1

*Dosage ratio: defined as the molar ratio of sodium bicarbonate to ammonia