Experimental study on the intensive treatment of coal mine domestic sewage by biofilm A/O

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Abstract. In recent years, China attached much significance to the reuse and high standard discharge of domestic sewage. Coal mines in China are mainly distributed in north and northwest areas with shortage of water. To improve the treatment effects and reuse standard of coal mine domestic sewage, the biofilm A/O reactor with one anoxic zone and two aerobic zones was used to carry out an experiment. The pollutants removal effects and influencing factors were analyzed. The results indicate: Under the conditions of HRT=5.92 h, ρ(DO)A = 0.5±0.2 mg/L, ρ(DO)O1 = 2±0.2 mg/L, ρ(DO)O2 = 2.5±0.2 mg/L, and R (reflux ratio) =100%, the maximal concentration of NH4+-N, COD, TN and TP in the last effluent were respectively 0.52mg/L, 15.3 mg/L, 13.1 mg/L and 0.78mg/L. Under the conditions of HRT=4.74 h and R=50%, the reactor achieved optimal performance. Moreover, the effluent concentration of NH4+-N, COD, TN, TP met The reuse of urban recycling water - Water quality standard for industrial uses (GB/T 19923-2005).

1. Introduction
In recent years, our country and locality attached much significance to the reuse and high standard discharge of domestic sewage. Coal mines in China are mainly distributed in north and northwest areas with shortage of water. The research on coal mine domestic sewage treatment focuses on pollution abatement but short of reuse. The utilization coefficient and utilization level should be increased rapidly [1]. In fact, emission requirements are stricter than reuse requirements in some areas. Under these circumstances, the reuse projects of coal mine domestic sewage are developed fast. At present, domestic sewage is partly reused in some coal mines, but the reuse ranges are popularly limited in low level, such as green water, dustproof sprinkling water and flushing water [2]. The requirements for high-level reuse of domestic sewage are urgent, but the corresponding research is not enough [3]. For further improvement of utilization coefficient and utilization level, the technological development of the coal mine domestic sewage treatment should be intended for industrial uses. NH4+-N, COD, TP are highly restricted in The reuse of urban recycling water - Water quality standard for industrial uses (GB/T 19923-2005). The NH4+-N of make-up water is even restricted at 1mg/L when the heat exchanger for open circulating cooling water system is copper. Biological nitrogen removal capability and impact-resistant capacity of the biological treatment process should be powerful. Biological treatment process is the core of domestic sewage treatment plants. Multistage A/O, A2/O, step-feed inlet technology, SBR and MBR were popularly researched to remove nitrogen and phosphorus [4-8]. Organics and phosphorus can be effectively further removed by advanced
treatment, but nitrogen removal is mainly dependent on biological treatment. To meet the high demand for coal mine domestic sewage treatment and reuse, an efficient nitrogen and phosphorus removal technique under the high load condition was discussed by integration of superiorities biological multiplication and A/O in this research.

2. Materials and methods

2.1. Test device

As shown in Figure 1, the biofilm A/O reactor is composed of anoxic zone (A), 1st aerobic zone (O1) and 2nd aerobic zone (O2) three-stage reaction chambers connected in series. The effective volumes of A, O1 and O2 are respectively 0.832 m$^3$, 0.768 m$^3$ and 0.768 m$^3$, and the total effective volume is 2.368 m$^3$. A, O1 and O2 are equipped with porous carriers and sludge discharge tubes. A is configured with a rabbler. O1 and O2 are equipped with aeration pipes. The sewage is introduced into the upper part of A by the influent pump. The effluent of A flows into O1 through the higher water flow channels. The effluent of O1 flows into O2 through the lower water flow channels. The discharging tube and back-flow pipe are located in the upside of O2. Different from traditional A/O reactor, secondary settling tank and sludge reflux system are not necessary in this biofilm A/O reactor.

![Fig.1 Schematic diagram of biofilm A/O reactor](image)

2.2. Test method

The test water was domestic sewage from a coal mine. The raw water quality during the test period: $\text{NH}_4^+-\text{N}$ 22.6~28.5 mg/L, TN 23.2~29.8 mg/L, TP 1.69~2.37 mg/L, COD 68.5~89.7 mg/L, pH 7.46~7.72. In start-up phase of the reactor, after inoculation with activated sludge from secondary settling tank in this coal mine sewage plant, the aeration began. After the biofilm cultured in the carrier, the inlet water flow was gradually increased according to the actual treatment effect of the reactor. In the stable operation stage, the influent flow rate was 0.4 m$^3$/h (HRT=5.92h), reflux ratio (R) was 100%, and the DO concentration of A, O1 and O2 are respectively controlled to 0.5±0.2 mg/L, 2±0.2 mg/L and 2.5±0.2 mg/L. The removal effect of the reactor on the main pollutants was analyzed. In the parameter optimization stage, the treatment effect was further optimized by adjusting HRT and reflux ratio.

Analysis and test methods: $\text{NH}_4^+-\text{N}$, TN, COD and TP were determined by Nessler reagent colorimetry, TNT persulfate digestion method, rapid digestion spectrophotometry and ammonium molybdate spectrophotometric method.
3. Results and discussion

3.1. Analysis of pollutant removal effect

Under the conditions of HRT=5.92h, ρ(DO)₁=0.5±0.2 mg/L, ρ(DO)₂=2±0.2 mg/L, R=100%, the removal effect of NH₄⁺-N, COD, TN and TP is shown in Fig.2~ Fig.5.

In the case of influent ρ(NH₄⁺-N)=24.9~28.5mg/L, the effluent concentration and removal rate of NH₄⁺-N are respectively 0.11~0.52mg/L and 97.9%~99.6%. The effluent concentration of NH₄⁺-N reaches the make-up water for open circulating cooling water system with copper heat exchanger in urban recycling water of GB/T 19923-2005 (ρ(NH₄⁺-N) ≤1mg/L). The scanning electron microscopy and microphysical properties indicate that AOB and NOB adhere to the carriers in O₁ and O₂. NH₄⁺-N is consequently and efficiently oxidized under the aerobic nitrification. Compared with routine municipal sewage, the COD concentration of coal mine sewage is lower. In the case of influent ρ(COD)=68.5~85.3mg/L, the effluent concentration and removal rate of COD are respectively 7.93~15.3mg/L and 81.2%~88.5%. COD can be removed in A, O₁ and O₂ of the biofilm A/O reactor, and three-level reaction guarantee the steady removal rate. Although 60mg/L COD meet the demand of GB/T 19923-2005, the effluent concentration of COD is under the GB3838-2002 class III standard limit.

In the case of influent ρ(TN)=25.9~29.8mg/L, the effluent concentration and removal rate of TN are respectively 10.3~13.1mg/L and 54.5%~63.6%. The effluent concentration of TN meets the class A discharge standard of pollutants for municipal wastewater treatment plant (GB18918-2002). Lower carbon-nitrogen ratio is not propitious to TN removal, but the denitrification can be improved by pre-stage denitrification of the biofilm A/O reactor. Meanwhile, the finite organic carbon source is efficiently used. In the case of influent ρ(TP)=1.79~2.25mg/L, the effluent concentration and removal rate of TP are respectively 0.55~0.78 mg/L and 63.9%~71.9%. The effluent concentration of TP meets the demand of GB/T 19923-2005. The anaerobic microenvironment and polyp bacteria concentration are optimized by the introduction of carrier, and the action of anaerobic phosphorus release and aerobic phosphorus absorption are better than biological contact oxidation and activated sludge A/O.
According to the analysis of water quality along reactor, NH$_4^+$-N, COD, TN and TP can be synchronously removed in O1 and O2. NH$_4^+$-N and COD can be deeply removed with the impact-resistant capacity biofilm A/O reactor. The removal effect of TN and TP can be improved by simultaneous denitrification and phosphorus removal after anoxic zone. Sludge concentration is raised by biofilm A/O reactor, and the sludge bulking problems of activated sludge method are solved [9]. Because of no secondary settling tank and sludge reflux system, biofilm A/O reactor is better than A/O, A$^2$/O and biological contact oxidation.

3.2. Effect of HRT on treatment efficiency

Under the conditions of ρ(DO)$_A$= 0.5±0.2 mg/L, ρ(DO)$_{O1}$=2±0.2 mg/L, ρ(DO)$_{O2}$=2.5±0.2 mg/L, R=100%, the effect of HRT on treatment efficiency is analyzed through the adjustment of the influent flow rate, the results are shown in Fig.6.

In the case of 2.96h≤HRT≤4.74h, prolonging HRT can significantly reduce the effluent NH$_4^+$-N concentration. When HRT>4.74h, the effect of continuing to extend HRT on NH$_4^+$-N treatment efficiency is not obvious, and NH$_4^+$-N concentration of the effluent reaches the make-up water for open circulating cooling water system with copper heat exchanger in urban recycling water of GB/T 19923-2005 with the ρ(NH$_4^+$-N)=0.61mg/L. When 2.96h≤HRT≤7.89h, the effluent COD can meet the requirements of GB/T 19923-2005. In the case of 2.96h≤HRT≤5.92h, prolonging HRT can significantly reduce the effluent concentration of COD and TN. When HRT>5.92h, the effect of continuing to extend HRT on treatment efficiency of COD and TN is not obvious. In the case of 2.96h≤HRT≤4.74h, prolonging HRT can significantly reduce the effluent TP concentration. When HRT>4.74h, the effect of continuing to extend HRT on TP treatment efficiency is not obvious. When HRT≥3.95h, the effluent TP can meet the requirements of GB/T 19923-2005. According to the effect of HRT on treatment efficiency, prolonging HRT can improve the effluent quality to a certain extent, but too long HRT may reduce treatment efficiency. In the case of 4.74h≤HRT≤7.89h, the concentration of COD, NH$_4^+$-N, TP can meet the requirements of GB/T 19923-2005 and ρ(TN)=12.1mg/L. Because the HRT of traditional A/O should be controlled above 8h [10], the advantage of biofilm A/O reactor is obvious. The HRT of biofilm A/O reactor should be controlled at 4.74h in consideration of treatment efficiency and effluent effect.
3.3. Effect of reflux ratio on treatment efficiency
Reflux ratio is a key parameter to influence the nitrogen removal of A/O. Under the conditions of HRT = 5.92h, $\rho(\text{DO})_A = 0.5 \pm 0.2$ mg/L, $\rho(\text{DO})_{O1} = 2 \pm 0.2$ mg/L, $\rho(\text{DO})_{O2} = 2.5 \pm 0.2$ mg/L, the effect of reflux ratio on treatment efficiency are shown in Fig.7. In the case of $R \leq 150\%$, the effect of different reflux ratio on treatment efficiency of NH$_4^+$-N and COD is not obvious. The TN and TP of effluent are lowest with the reflux ratio of 100%. When $R = 200\%$, NH$_4^+$-N, COD, TN and TP of effluent raised by different degrees. The suspended solid concentration raises with the biofilm loss during the test. In conclusion, nitration liquid reflux and internal recycle flow are both increased with the higher reflux ratio. Without secondary settling tank and sludge reflux system, the microbial concentration is reduced by the loss of biofilm in large flow. Under the conditions of simultaneous denitrification and phosphorus removal in O1 and O2, reflux ratio can be reduced in biofilm A/O reactor. The reflux ratio of biofilm A/O reactor should be controlled at 50% in consideration of treatment efficiency and effluent effect.

4. Conclusion
Without secondary settling tank and sludge reflux system, biofilm A/O reactor can treat coal mine domestic sewage efficiently. Under the conditions of HRT = 5.92h, $\rho(\text{DO})_A = 0.5 \pm 0.2$ mg/L, $\rho(\text{DO})_{O1} = 2 \pm 0.2$ mg/L, $\rho(\text{DO})_{O2} = 2.5 \pm 0.2$ mg/L, and $R$ (reflux ratio) = 100%, the maximal concentration of NH$_4^+$-N, COD, TN and TP in the last effluent are 0.52 mg/L, 15.3 mg/L, 13.1 mg/L and 0.78 mg/L. The effluent concentration of NH$_4^+$-N, COD, TN, TP meet the GB/T 19923-2005. The HRT and reflux ratio of biofilm A/O reactor should be respectively controlled at 4.74h and 50% in consideration of treatment efficiency and effluent effect.

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