Research on the Mechanism of Simultaneous and Efficient Removal of Ammonia, NO₃⁻-N and TN in the Coking Wastewater

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Abstract. Through the sequential-recirculation and cross-recirculation ways, the two-stage micro-aerobic EGSB reactor system was operated to treat the actual coking wastewater. And the removal efficiencies of SCN⁻-N, CN⁻-N, NH₃-N, NO₃⁻-N and TN in coking wastewater were investigated, and then the mechanism of simultaneous and efficient removal of NH₃-N, NO₃⁻-N and TN in coking wastewater was also analyzed. The results showed that, using the two-stage micro-aerobic EGSB reactor to treating actual coking wastewater, for the sequential recirculation way and the cross-recirculation way, the removal efficiencies of SCN⁻-N, CN⁻-N, NH₃-N, NO₃⁻-N, TN were 98.0%, 91.5%, 81.3%, 30.4%, 24.5% and 97.3%, 97.0%, 92.7%, 92.0%, 70.9%, respectively. And the two-stage micro-aerobic EGSB reactor system could achieve simultaneous and efficient removal of ammonia and TN in the coking wastewater through the cross-recirculation way. For the sequential recirculation two-stage micro-aerobic EGSB reactor system, high SCN⁻-N and NO₃⁻-N removal of 94.2% and 90.8% was accomplished in the EGSBI, and high NH₃-N removal of 89.8% could attain in the EGSBII. However, high NO₃⁻-N accumulation in the EGSBII also caused very low TN removal of 24.5% for the two-stage micro-aerobic EGSB reactor system. High SCN⁻-N removal in the EGSBI could ensure high NH₃-N removal in the EGSBII. More importantly, the EGSBBhad a very strong ability to support SCN⁻-N shock. For the cross-recirculation two-stage micro-aerobic EGSB reactor system, the EGSBICould keep simultaneous and efficient removal of SCN⁻-N, CN⁻-N, NH₃-N, NO₃⁻-N and TN (93.5%, 92.4%, 84.1%, 92.0% and 73.7%). When using sequential recirculation way, thiocyanate-degrading bacteria could form competitive inhibition to ammonia oxidizing bacteria in the EGSBI, and thus, simultaneous and efficient removal of SCN⁻-N and NH₃-N could not accomplish in the EGSBI. High concentration NO₃⁻-N in the EGSBII was removed mainly through denitrification. However, through the cross-recirculation way for the two-stage micro-aerobic EGSB reactor system, the effluent recirculation of the EGSBI and/or the EGSBII were changed from simple high NH₃-N to mixed recirculation of high NH₃-N+high NO₃⁻-N and/or from high NO₃⁻-N to high NH₃-N+high NO₃⁻-N, respectively. Consequently, thiocyanate-degrading bacteria, denitrifying bacteria and anammox bacteria could keep high activity at the same time and also the effective coupling of denitrification and anammox bacteria was realized in the EGSBIII and EGSBII, which ensured the simultaneous and efficient removal of SCN⁻-N, NH₃-N and NO₃⁻-N in the EGSBII and EGSBII, and finally realized the simultaneous and efficient removal of NH₃-N, NO₃⁻-N and TN in the two-stage micro-aerobic EGSB reactor system.
1. Introduction
The components of coking wastewater are complex and contain dozens of toxic pollutants, such as cyanide, thiocyanate and phenols, etc. Meanwhile, there are large amount of ammonia in coking wastewater [1]. Thus, coking wastewater treatment not only involved the simultaneous and efficient removal of COD and NH$_3$-N, but also the efficient removal of TN. For the traditional coking wastewater treatment process, A$^2$/O process, even if A and/or O section was strengthened by bio-film, the effluent COD and NH$_3$-N were still difficult to reach the discharge standard [2,3]. At present, more attention should be paid to the paineful and stable operation of A and/or O section.

The appearance of high-speed reactor and granular sludge provided a new idea for the efficient treatment of coking wastewater. UASB reactor had been widely used in the pretreatment of coking wastewater, Pharmaceutical Wastewater, dye wastewater and other wastewater containing toxic and refractory substances due to its advantages of forming granular sludge, but the treatment effect was limited[4]. The dilution and high efficiency mass transfer produced by effluent reflux in EGSB reactor provided the possibility for high efficient treatment of refractory pollutants. Furthermore, under micro-aerobic conditions, the con-existence of anaerobic and aerobic microenvironment in the EGSB reactor provided the possibility for the simultaneous removal of COD and NH$_3$-N[5]. However, the previous researches showed that although single-stage micro-aerobic EGSB reactor system could attain high efficient removal of COD, phenols, SCN$^-$ and CN$^-$, the removal efficiency of NH$_3$-N was always negative. Furthermore, the two-stage micro-aerobic EGSB reactor system was employed to treat coking wastewater (the primary EGSB reactor used for the removal of toxic refractory pollutions, and the secondary EGSB reactor used for the removal of NH$_3$-N), but the removal efficiency of NH$_3$-N was still very limited (only with NH$_3$-N average removal of 26.5%). A coupled two-stage micro-aerobic EGSB system with diatomite could attain high efficient removal of COD, phenol, SCN$^-$, CN$^-$, and NH$_3$-N. However, the accumulation of NO$_3^-$ in the secondary EGSB reactor made it difficult to achieve the simultaneous and efficient removal of NH$_3$-N and TN [6].

When treating the actual coking wastewater, the two-stage micro-aerobic EGSB reactor system encountered the problem of product accumulation (high NH$_3$-N accumulation in the primary EGSB reactor and high NO$_3^-$-N in the secondary EGSB reactor). In the present study, the two-stage micro-aerobic EGSB reactor system was used to treat actual coking wastewater, and simultaneously two kinds of effluent recirculation ways (the sequential-recirculation and cross-recirculation ways) were supplied in the two-stage micro-aerobic EGSB reactor system. The main objective was to evaluate whether changing the effluent recirculation way (from sequential-recirculation to cross-recirculation) of the two-stage micro-aerobic EGSB reactor system could eliminate the accumulation of NH$_3$-N and NO$_3^-$-N and TN. The second objective was to analyze the mechanism of simultaneous and efficient removal of NH$_3$-N, NO$_3^-$-N and TN and determine the feasibility of cross-recirculation two-stage micro-aerobic EGSB reactor system for highly efficient treatment of coking wastewater.

2. Materials and Methods

2.1. Reactor Set-up
The research was carried out in two lab-scale micro-aerobic EGSB reactors (EGSB I and EGSB II) . A schematic diagram of the micro-aerobic EGSB reactor (EGSB Ior EGSB II) used is presented in Fig.1. Both of the EGSB I and EGSB II reactor were 2.3m height acrylic column with a conical-shaped bottom, a working volume of 12L, an internal diameter of 10cm. And the EGSB I and EGSB II reactor were connected to aeration column I and aeration column II with a liquid volume of 2.5L (50cm$^3$×10.0cm i.d., respectively). For the purpose of mixing and oxygen transfer to the sludge bed, liquor was recycled from the top of the reactors to the aeration columns and then back to the base of the reactors by means of a peristaltic pump. By controlling the oxygenation rate in the aeration column, different concentrations of dissolved O$_2$ could be generated in the recirculated fluid which supplied dissolved O$_2$ to the granule sludge bed in the EGSB reactor and to generate microaerophilic environments.
2.2. Wastewater
The wastewater used as influent to the MAEGSB reactor was the actual coking wastewater collected from the buffer tank of the second coking plant of Taiyuan Coal Gas and Chemical Stock Co., LTD located in Shanxi Province, North China with 651-2163mg.L⁻¹COD, 35.4~102.7mg.L⁻¹NH₃-N, 5.37-352.5 mg.L⁻¹phenol, 0.03-5.93mg.L⁻¹CN, 205.5-539.9mg.L⁻¹SCN and 8.86-9.71pH.

![Fig.1 Schematic diagram of the micro-oxygenic EGSB reactor](image)

2.3. Seed sludge
The seed sludge used in this experiment was obtained from a pilot-plant anaerobic EGSB reactor treating actual coking wastewater. The EGSB I and EGSB II kept 29.8g.L⁻¹MLSS, 0.54 MLVSS/MLSS and 26.7g.L⁻¹MLSS, 0.51MLVSS/MLSS, respectively.

2.4. Analytical methods
Effluent was collected and centrifuged at 4000rpm for 5-6min with a centrifuge (80-2B, ANTING). And then the supernatant was used for further analysis. COD, NH₃-N, NO₂⁻-N, NO₃⁻-N, TN, phenol and CN were measured according to the Chinese Standard Methods for Water and Wastewater Monitoring and analytical methods [7]. SCN measurement was performed with the American Standard Methods [8]. And simultaneously the pH was rapidly monitored using Lei-Ci PHS-3C pH meter.

2.5. Operating strategy
The two-stage micro-aerobic EGSB reactor system (EGSBI+EGSBII) was used to treat actual coking wastewater with 1.0L.h⁻¹ influent flow and 12h+12h HRT, and the oxygenation flow rates (air flow rates) in the aeration column for the EGSBI and EGSBII were 10000mL.min⁻¹ and 8000 mL.min⁻¹, respectively. In order to achieve the simultaneous and efficient removal of all kinds of pollutants in coking wastewater, two kinds of effluent recirculation ways, sequential recirculation cross-recirculation, were used. and the performance process of the two-stage micro-aerobic EGSB reactor system treating actual coking wastewater based on the removal of various pollutants (especially NH₃-N, NO₃⁻-N and TN) was investigated by following two operation stages:

Stage I (from days 1 to days 198) : with the same recirculation flow of 20L.h⁻¹, the effluent from EGSB I flow back to EGSB I and the effluent from EGSB II flow back to EGSB II. From days 1 to days 150d, the influent SCN⁻ concentration fluctuated greatly (350mg.L⁻¹→400mg.L⁻¹→500mg.L⁻¹→400mg.L⁻¹→350mg.L⁻¹), the ability of supporting SCN⁻ shock was analysed. And from days 151 to days 199 (stable operation stage), the removal of various pollutants in coking wastewater by the two-stage micro-aerobic EGSB reactor system was investigated.
Stage II (from days 199 to days 281) : with the same recirculation flow of 11L·h⁻¹ and 9 L·h⁻¹ to itself and the other reactor, respectively, the effluent from EGSB I flow back to EGSB I and EGSB II, and the effluent from EGSB II flow back to EGSB II and EGSB I. The removal of SCN⁻-N, NH₃-N, NO₃⁻-N and TN from coking wastewater was mainly investigated.

3. Results and discussions

3.1. Operation characteristic at stage I

NH₃-N, NO₃⁻-N, SCN⁻-N, CN⁻-N removal: With 24h HRT(12h EGSB I +12h EGSB II), the two-stage micro-aerobic EGSB reactor system treating actual coking wastewater could had stable COD removal efficiency and phenol removal efficiency of nearly 80% almost 100%. However, the removal efficiencies of N pollutants (NH₃-N, NO₃⁻-N, SCN⁻-N, CN⁻-N) were very low, and the variations of the NH₃-N, NO₃⁻-N, SCN⁻-N, CN⁻-N removal efficiencies for the two-stage micro-aerobic EGSB reactor system were showed in Fig.2.

![Fig.2 NH₃-N, NO₃⁻-N, SCN⁻-N and CN⁻-N removal for the two-stage micro-aerobic EGSB reactor system](image)

The two-stage micro-aerobic EGSB reactor system could effectively remove SCN⁻-N and CN⁻-N from the actual coking wastewater, and in the stable operation stage, the average removal efficiencies of SCN⁻-N and CN⁻-N were up to 98.0% and 91.5%, respectively. However, the removal effect of the two-stage micro-aerobic EGSB reactor system on the NH₃-N and NO₃⁻-N was not ideal. For the initial 151 days, the removal efficiency of NH₃-N was negative (the average removal efficiency was -35.4%), and however, the average removal efficiency of NO₃⁻-N was up to 91.7%. From days 152, the removal efficiency of NH₃-N became positive and increased rapidly. At days 154, days 157 and days 160, the NH₃-N removal efficiencies were 12.0%, 70.6% and 84.3%, respectively. And subsequently, the two-stage micro-aerobic EGSB reactor system could have stable and efficient NH₃-N removal (81.3% average removal efficiency) (fluctuating between 70.6% and 84.8%). However, the NO₃⁻-N removal efficiency began to decrease continuously (95.2%, 88.6% and 52.0% at days 154, days 160 and days 166) and then stabilized at low level NO₃⁻-N average removal efficiency of 30.4% (fluctuating
between 24.4% and 37.4%). Therefore, the two-stage micro-aerobic EGSB reactor system had high NH$_3$-N removal by conversion to NO$_2$--N and/or NO$_3$--N, and the accumulation of NO$_2$--N and NO$_3$--N occurred successively in the EGSB II reactor. Thus, the TN removal of two-stage micro-aerobic EGSB reactor system was not high, only 24.5%.

**Influence of SCN--N on the removal of NH$_3$-N:** Table 1 showed the changes of SCN$^-$ and NH$_3$-N removal efficiencies for the two-stage micro-aerobic EGSB system.

| Pollutions | EGSBI Removal (%) | EGSBII Removal (%) | Total Removal (%) |
|------------|-------------------|---------------------|-------------------|
| SCN$^-$    |                   |                     |                   |
| 1-116d     | 32.2%             | 79.3%               | 85.0%             |
| 117-157d   | 90.2%             | 65.2%               | 97.0%             |
| 158-196d   | 94.2%             | 62.7%               | 98.0%             |
| NH$_3$     |                   |                     |                   |
| 1-116d     | -10.1%            | -22.7%              | -34.6%            |
| 117-157d   | -69.9%            | 31.6%               | 14.7%             |
| 158-196d   | -82.6%            | 89.8%               | 81.3%             |

The toxic compounds in coking wastewater include phenols, SCN$^-$ and CN$^-$, among which phenols are easy to be removed and nearly 100% phenols removal were attained in the whole operation stage. Although it has been reported that CN$^-$ has a strong toxic effect on NH$_3$ oxidizing bacteria, however, because of the low influent CN$^-$ concentration of 0.1-1.97mg·L$^{-1}$ and high removal efficiency of 78.2%-91.5% for the two-stage micro-aerobic EGSB reactor system, the toxic effect of CN$^-$ on NH$_3$ oxidizing bacteria was not obvious.

Relatively high SCN$^-$ concentration (283.1-539.9 mg·L$^{-1}$) in coking wastewater had a significant effect on the NH$_3$-N removal. From day 1 to days 116, the EGSBI only had 32.2% SCN$^-$ removal efficiency, and thus, the high average influent SCN$^-$ concentrations of 413.0mg·L$^{-1}$ and 278.3mg·L$^{-1}$ for the EGSBI and EGSBII resulted in negative removal of NH$_3$-N in the EGSBI and EGSBII. It can be seen that SCN$^-$ has a strong toxic inhibitory effect on NH$_3$ oxidizing bacteria. From days 117 to days 157, the average removal of SCN$^-$ was high to 90.2%, which made the influent SCN$^-$ average concentration of 43.6mg·L$^{-1}$. Simultaneously, the NH$_3$-N removal became positive and steadily increased from 2.4% to 86.4%. From days 158 to days 196, because the EGSBII could keep high SCN$^-$ removal of 94.2%, the EGSBII could keep stable high removal of NH$_3$-N, with an average removal of 89.8%. In a word, the removal effect of SCN$^-$ in the EGSBII directly affected the removal effect of NH$_3$-N of the two-stage micro-aerobic EGSB reactor system.

**Supporting SCN$^-$ shock ability for the EGSBI:** Table 2 analyzed the change of the SCN$^-$ removal efficiency in the EGSBI when influent SCN$^-$ concentration greatly fluctuating.

| Operation stage | SCN$^-$ (mg·L$^{-1}$) | SCN$^-$ removal (%) |
|-----------------|------------------------|---------------------|
| 0-35d           | 350                    | 26.1                |
| 36-44d          | 400                    | 52.3                |
| **45-67d; 68-73d, 113-116d;117-131d** | 500 | **21.3;55.8;85.6** |
|                 |                        | **73.4;76.9;79.3**  |
|                 |                        | **78.8;88.7;97.5**  |
| 132-139d        | 400                    | 92.2                |
| 140-148d        | 350                    | 92.2                |
| 149-196d        | 300                    | 94.1                |

From day 1 to days 35, with 350mg·L$^{-1}$ influent SCN$^-$ concentration, the EGSBI only had 26.1% SCN$^-$ removal. However, with 400mg·L$^{-1}$ influent SCN$^-$, the EGSBI had a greatly improved SCN$^-$
-N removal of 52.3%. Subsequently, only after 8 days, the influent SCN--N increased to 500mg.L⁻¹, the removal efficiency of SCN--N in the EGSBI suddenly decreased to 21.3%. But only after 22 days (from days 45 to days 67), the SCN--N removal efficiency could rapidly increase to 55.8% in the EGSBI. And then only after 4 days, the EGSBI had a high SCN--N of 85.6%, which indicated that the EGSBI had very strong ability of supporting SCN--N shock. The EGSBI could attain high SCN--N removal of about 90% with high influent SCN--N concentration of 350-500mg.L⁻¹, and which ensuring the high NH₃-N removal in the EGSBI.

3.2. operation characteristic at stage II
The removal efficiencies of various pollutions in the two-stage micro-aerobic EGSB reactor system treating actual coking wastewater for the sequential recirculation way and the cross-recirculation way were presented in Table 3.

Table 3 Removal efficiencies of various pollutions for sequential recirculation and the cross-recirculation

| Influent (mg·L⁻¹) | Removal (%) |
|------------------|-------------|
|                  | Sequential-recirculation | Cross-recirculation | Sequential-recirculation | Cross-recirculation |
|                  | EGSBI | EGSBII | EGSBI | EGSBII |
| COD              | 824-2163 | 651-970 | 65.8 | 29.1 | 75.4 | 72.1 | 13.1 | 75.8 |
| NH₃-N            | 37.6-102.4 | 35.4-48.8 | -82.6 | 89.8 | 81.3 | 84.1 | 58.3 | 92.7 |
| NO₃--N           | 97.2-198.4 | 62.6-97.6 | 90.8 | -586.4 | 39.9 | 92.0 | -3.4 | 92.0 |
| TN               | 141.2-167.2 | 90.3-174.1 | 31.4 | -10.0 | 24.5 | 73.7 | -30.5 | 70.9 |
| Phenol           | 5.37-352.5 | 6.1-104.4 | 96.3 | 96.4 | 99.9 | 99.1 | 100.0 | 100.0 |
| SCN⁻             | 283.1-539.9 | 205.5-303.0 | 94.2 | 62.7 | 98.0 | 93.5 | 57.3 | 97.3 |
| CN⁻              | 0.10-1.97 | 0.87-5.93 | 79.5 | 55.1 | 91.5 | 92.4 | 62.5 | 97.0 |

Compared with the sequential recirculation way, the removal efficiencies of COD, phenol, and SCN⁻ in the two-stage micro-aerobic EGSB reactor system with cross-recirculation way did not changed significantly, which were 75.8%, 100.0% and 97.3%, respectively (75.4%, 99.9% and 98.0% for the sequential recirculation way). However, the removal efficiencies of NH₃-N, NO₃--N, TN and CN⁻ improved greatly and were 92.7%, 92.0%, 70.9% and 97.0%, respectively (only 81.3%, 39.9%, 24.5% and 91.5% for the sequential recirculation way).

The cross-recirculation way could enhance the removal effect of NH₃-N, TN and CN⁻ in the EGSBI. The removal of NH₃-N in the EGSBI was negative (-82.6%) under the condition of sequential recirculation, and positive (84.1%) under the condition of cross-recirculation. And simultaneously the removal efficiencies of TN and CN⁻ was low, only 31.4% and 79.5%, under the condition of sequential recirculation, and it increased to 73.7% and 92.4% after using cross-recirculation way, respectively.

The cross-recirculation way could solve the problem of high NO₃--N accumulation in the EGSBI. The removal efficiency of NO₃--N in the EGSBI was -586.4% for the sequential recirculation way and only -3.4% for the cross-recirculation way (almost no NO₃--N accumulation).

Through the cross-recirculation way, the two-stage micro-aerobic EGSB reactor system could effectively treat actual coking wastewater, and the removal efficiencies of COD, phenol, SCN⁻, CN⁻, NH₃-N, NO₃--N and TN were high to 75.8%, 100.0%, 97.3%, 97.0%, 92.7%, 92.0% and 70.9%, respectively.

4. Conclusions
The effect of effluent recirculation way on the performance process of two-stage micro-aerobic reactor system treating actual coking wastewater based on the removal of COD, phenol, SCN⁻, CN⁻, NH₃-N, NO₃--N,
NH$_3$-N, NO$_3$-N and TN were investigated. The two-stage micro-aerobic EGSB reactor system was performed with the sequential recirculation way and the cross-recirculation way, respectively. According to the findings of this study, the following conclusions can be drawn:

With the sequential recirculation way, the removal efficiencies of SCN$^-$-N, CN$^-$-N, NH$_3$-N, NO$_3$-N and TN in the two-stage micro-aerobic EGSB reactor system were 98.0%, 91.5%, 81.3%, 30.4% and 24.5%, respectively. The high removal efficiencies of SCN$^-$-N and NO$_3$-N (94.2% and 90.8%) could attain in the EGSBⅠ, and the high removal efficiency of NH$_3$-N (89.8%) could accomplish in the EGSBⅡ. Furthermore, high removal of SCN$^-$-N in the EGSBⅠ could ensure the high removal of NH$_3$-N in the EGSBⅡ. The EGSBⅠ had very strong ability of supporting SCN$^-$-N shock.

With the cross-recirculation way, the removal efficiencies of SCN$^-$-N, CN$^-$-N, NH$_3$-N, NO$_3$-N and TN in the two-stage micro-aerobic EGSB reactor system were 97.3%, 97.0%, 92.7%, 92.0% and 70.9%, respectively.

With the cross-recirculation way for the two-stage micro-aerobic EGSB reactor system, the effluent recirculation of the EGSBⅠ and/or the EGSBⅡ were changed from simple high NH$_3$-N to mixed recirculation of high NH$_3$-N+high NO$_3$-N and/or from simple high NO$_3$-N to high NH$_3$-N+high NO$_3$-N, respectively. Consequently, the thiocyanate-degrading bacteria, the denitrifying bacteria and the anammox bacteria could keep high activity at the same time and also the effective coupling of the denitrification bacteria and the anammox bacteria could realize in the EGSBⅠand the EGSBⅡ, which ensured the simultaneous and efficient removal of SCN$^-$-N、NH$_3$-N and NO$_3$-N in the EGSBⅠand the EGSBⅡ. And finally could realize the simultaneous and efficient removal of NH$_3$-N, NO$_3$-N and TN in the two-stage micro-aerobic EGSB reactor system.

Acknowledgements

This research was supported by grants from the Shan-Xi Province Key Technologies R & D Program of Shan Xi (NO.20120313008 and NO. 20150313002-1).

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