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CONSTRUCTION OF NITRIFICATION MODEL WITH NITRIFYING COAL ASH IN AEROBIC TREATMENT OF HIGH STRENGTH WASTEWATER

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Highlights

- Monod model was applied in nitrifying coal ash system successfully.
- Factors of pH, temperature and DO concentration were introduced in Monod model.
- Only 0.08% inconsistency between experiment and simulated data occurred in treating inorganic wastewater with high-strength ammonia.

Abstract. Nitrifying carriers can provide good settle ability and stable removal efficiency for nitrogen. Models for ammonia removal rate for nitrifying carriers will improve its engineering application. This study was conducted in nitrifying coal ash system with Monod model. Results indicated the maximum NH_4^+-N removal rate and half-saturation constant of NH_4^+-N in Monod model were 110.48 mg/L and 59.19 mg/L, respectively. Introduction of the correction coefficients, including pH, temperature and dissolved oxygen (DO) concentration, decreased the average gap between experiment data and simulated data from 6.48 to 2.74 mg N/(L·h). And improved accuracy of the Monod model by 5.11%. The differences between experiment and simulated NH_4^+-N removal rate ranged from 0.08 mg N/(L·h) to 8.34 mg N/(L·h) when the influent concentration of NH_4^+-N increased from 443.18 to 1121.29 mg N/L and without organic. Only 0.08% inconsistency between experiment and simulated data occurred in treating wastewater with high-strength ammonia. However, NH_4^+-N removal rate of the nitrifying coal ash was inhibited about 40% when influent with averaged 173.19 mg COD/L and 37.20 mg N/L, therefore, other factors, the content of nitrifying bacteria for example, need to be introduced into the Monod model when treating organic wastewater.

Keywords: nitrifying coal ash, Monod model, correction coefficients, NH_4^+-N removal rate, high-strength ammonia.

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Introduction

Nitrifying bacteria, such as ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB), play a major role in nitrification (Liu et al., 2017b). However, slow growth and low survival rate usually cause its low abundance in systems, then significantly restrict nitrification (Wang et al., 2016). There are many solutions for that, membrane bio-reactor (MBR) and aerobic granule processes for example (Dong et al., 2021). However, many problems exist in among of them, such as the difficulty in operating due to the membrane fouling of MBR and production of aerobic granule (Meng et al., 2010; Wang et al., 2018). Nitrifying carrier is a kind of carriers with abundant of nitrifiers which are formed by the microorganisms immobilization method, which act as an alternative way to maintain the nitrifiers (Liu et al., 2021). Compared with free nitrifiers, it has the advantages of good settle ability, high biomass retention and ability to remove nitrogen (Shin et al., 2019). Due to these advantages, nitrifying

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carrier has attracted much attention in recent years, and most of them were focused on the enrichment and application. Recent carriers used for enrichment have included activated carbon, ceramic, composites, etc. (Chaali et al., 2021; Parker et al., 2002; Li et al., 2013; Han & Park, 2012). After enrichment, the nitrifying carrier were also widely used for improving sustainable nitrification in lab-scale and full-scale (Wang et al., 2013; Liu et al., 2018). However, the nutrients utilization mechanism of nitrifying carrier should be focused, and a better understanding of nutrients availability is essential to control aquatic environment.

Mathematical models have been proposed to study the nutrients utilization by microbes, which were capable of modeling and control of biological treatment process (Wu et al., 2017). Nitrifiers is the body part of nitrifying carrier and carrier provides support for cellular adhesion when treating wastewater. Hence, activated sludge models (ASMs) could equally well apply to nitrifying carrier, such as Monod function (Wu et al., 2017). Monod-type equations, introduced by Monod, is a function used for describing the relationship of the specific growth rate of phytoplankton, microbes with external nutrients and the degradation rate of substrate (Monod, 1942; Fernandez-Fontaina et al., 2014). However, Monod function has some limitations in its wide application (Huang et al., 2019). For effective application of it, the model parameters need to be calibrated and validated (Fernandez-Fontaina et al., 2014; Saeed & Sun, 2011), especially with different methods for calibrating.

Nitrifiers are known to be sensitive to environmental parameters, such as DO and pH, which influence the activity of nitrifiers dramatically (Dong et al., 2011). In addition, a certain amount of organic may inhibit the growth and activity of nitrifiers, while that is benefit for heterotrophic bacteria (Qiao et al., 2008). That indicates nitrifiers are more suitable for treating wastewater with inorganic carbon or low COD/N ratio, especially with high ammonia-loading. It was already known that coal ash has good compatibility with nitrifying bacteria compared with other common carriers (Liu et al., 2017a). And nitrifying coal ash was stable and efficient for treating wastewater with low C/N ratio and high ammonia in our previous studies (Liu et al., 2021). However, its nitrogen utilization metabolism is still unclear. This work intends to describe the metabolic process of high-concentration nitrogen wastewater treatment by nitrifying coal ash using Monod function. Amendments to Monod function mainly include:

(1) Adopt simultaneous storage and growth for the utilization of the substrates.
(2) Work out the basic parameters of Monod function.
(3) Take environment parameters into account, including pH, DO and temperature (T).
(4) Validate the calibrated Monod model.

We shall demonstrate the effectiveness of the proposed modified Monod function by simulations for the metabolic process of nitrifying coal ash when treating high strength inorganic wastewater with different concentration of nitrogen and actual domestic sewage. It is expected to provide theoretical basis for the application and optimization in the application of nitrifying coal ash.

1. Materials and methods

1.1. Laboratory experiment setup

Sequencing batch reactor (SBR) with working volume of 2 L (48 cm high, 9 cm internal diameter) was used for model assessment, and was seeded in 20% (v/v) nitrifying coal ash. The seeding nitrifying coal ash was cultivated in our laboratory (Liu et al., 2021), and maintained at high activity before this experiment. Reactors were operated in a sequencing batch mode of 12 h cycle, with 11 h for aeration. Ammonium chloride and sodium bicarbonate acted as nitrogen and carbon substrate in the synthetic wastewater, respectively. Other details of synthetic referred to Liu et al. (Liu et al., 2021). Sampling ports for both liquid and sludge were located in depth of 5 cm as measured from the top of the reactors, the diagram was showed in Supporting Information (S1). Aeration system was set in the bottom of the reactor, and the DO concentration at startup period was stabled at 4.0–4.5 mg/L.

After the system having stable operation, dynamic test was applied. To solve the basic parameters of Monod model, the influent NH_4^+-N concentration changed from 50 mg/L to 1200 mg/L. On the other hand, to modify the Monod model with environmental parameters, batch experiments were conducted at pH, DO and T setting as 6.5–9.5, less than 0.5 to 6.5 mg/L and 10–35 °C, respectively. Concentration of NH_4^+-N and pH controlled by changing the amount of ammonium chloride in influent and sodium bicarbonate-buffer.

Then, the modified Monod model was evaluated by treating synthetic inorganic wastewater (operated for 25 days) and actual domestic sewage (operated for 57 days). The other components of the synthetic inorganic wastewater are consistent with the setup period, and the concentration of NH_4^+-N ranged from 400 to 1200 mg N/L. Domestic sewage was taken from a full-scale municipal wastewater treatment plant (WWTP) in Shenyang in China, in which the influent of Chemical oxygen demand (COD), NH_4^+-N concentrations and pH was around 110–250 mg COD/L, 28–50 mg N/L and 7, respectively. The room temperature was maintained at 20±2 °C in validate periods.

1.2. Determination of model parameters and simulation methods

In this study, Monod function is the basis for modeling nitrification efficiency of nitrifying coal ash, which can be simply represented as Eq. (1):

$$r = r_{\max} S/(K_s + S), \tag{1}$$

where r and $r_{\max}$ are the reaction rate and maximum reaction rate of the NH_4^+-N or substrate (mg/(L·h)), S is the

NH_4^+-N or the limiting nutrient concentration (mg/L), K_s is the half-saturation constant of NH_4^+-N or substrate (mg/L). And there are two fundamental assumptions: a) each cell component growth by an equal proportion, and b) endogenous respiration rate $K_d = 0$.

The basic parameters in Monod function was simulated by using the First Optimization (1stOpt) software (7D-Soft High Technology Inc., China). In order to make the model succinct to the focus of this study, only essential environmental parameters were included in the modified model, naming *f* (pH, DO, T). DO concentration, pH and T parameters were obtained through calibration with the soft 1stOpt according to the batch experimental data.

1.3. Sampling and chemical analysis

Sampling was done to measure the concentrations of COD, NH_4^+-N at a certain time interval. All measurements were conducted when the system reached a steady state, and each reactor was collected twice every day. Average value and standard deviation was used for further analysis. The water samples were filtered through 0.45 μm filter to remove suspended solids for chemical analysis. NH_4^+-N, NO_2^--N and NO_3^--N concentrations were measured in accordance with Standard Methods (APHA, 2005) using spectrophotometer (SHIMADZU, Japan). COD were determined with Hach photometer (DR 5000). The DO and T of the reactors were measured by portable DO meter (WTW, Germany), pH value was monitored and controlled by automatic measuring equipment.

1.4. Statistical analysis

The paired-sample T test in the SPSS 19.0 software (Chicago, USA) was used to analyze the statistical significance in experimented and simulated data. All values were pointed out as means and standard deviation.

2. Results and discussion

2.1. Model descriptions and parameter computation

In this study, the Monod model was modified to simulate the biological treatment processes of treating wastewater with high nitrogen concentration and to make a comparison with experimental data under the same conditions. In the modified model, the nitrifying coal ash used *f* (pH, DO, T) for the correction coefficients involved in environmental parameters. Autotrophic nitrification bacteria were the dominant in the nitrifying coal ash, which was the major contributor to nitrification in this study. Thus, heterotrophic nitrification bacteria were not considered in this model to reduce the ammonia.

The results of batch experiment for NH_4^+-N degradation under different loads are shown in Figure 1. According to Figure 1a, nitrifying coal ash could quickly utilize ammonia (S) and the NH_4^+-N concentration didn't restrict its activity at all. Growing in high-concentration nitrogen for long term caused community shift, which make nitrifying bacteria was less sensitive to high NH_4^+-N and free ammonia (FA) concentration (Posmanik et al., 2014). At 10 h, the NH_4^+-N removal efficiency was more than 94.80% under influent concentration of 1185 mg N/L. It was confirmed that the NH_4^+-N was gradually oxidized to NO_3^--N and no NO_2^--N detected (data not shown). The ammonia degradation process met with the zero-order kinetics by fitting analysis, with R square bigger than 0.99 through a linear fitting. Then, the NH_4^+-N removal rate (mg N/(L·h)) was calculated by concentration of influent and effluent, showing in Figure 1b. The growth of NH_4^+-N removal rate has started to flatten out with the substrate increases, and reached 113.25 mg N/(L·h) at 1185 mg N/L. Parameters of r_{max} and K_s in basic Monod model were solved with nonlinear fitting according to the variety ammonia degradation rate, with values of 110.48 mg N/L and 59.19 mg N/L, respectively. It is reported that r-strategists nitrifiers grow quickly at high substrate concentration (Yu et al., 2020), which promote a high maximum reaction rate (r_{max}, mg N/(L·h)) and high substrate half-saturation value (K_s, mg N/L). In this study, the dominant nitrifiers was *Nitrosomonas* and *Nitrobacter* (Liu et al., 2021), which belongs to *r*-strategists AOB and NOB respectively.

As shown in the Figure 1b, a small portion of simulated NH_4^+-N removal rate snugly into experiment data,

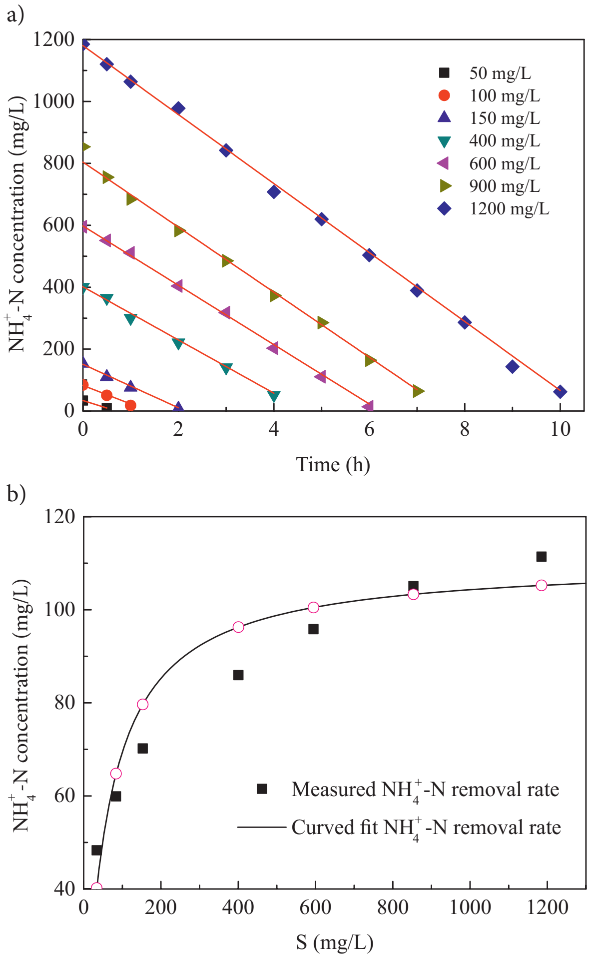

Figure 1. NH_4^+-N concentrations every hour (a) and basic Monod model for NH_4^+-N removal rate (b)

however, most simulated data were behind or ahead the experiment data. And the average gap between them was 6.48 mg N/(L·h). The basic Monod model takes no account of environmental factors when simulate the NH_4^+-N degradation process, however, it was usually influenced by pH, DO concentration, temperature, etc. (Dong et al., 2011). Therefore, the influence of pH, DO concentration and T on nitrifying coal ash were considered for improving the accuracy of the basic Monod in this study.

2.2. Parameter calibration for Monod model

The pH, DO concentration and T were ranged under routine conditions in this study, and NH_4^+-N removal rate under different conditions were shown in Figure 2. As illustrated in Figure 2a, NH_4^+-N removal rate increased with pH and then decreased, bordering by pH of 8.2 at 120.55 mg N/(L·h). That was sharply limited when the pH exceeded 9.0, and barely stayed at 23.00 mg N/(L·h) under pH of 9.5. High or low pH condition not only reduce the enzyme activity of microbe, but also can affect the concentrations of FA and free nitrous acid (FNA) in system (Park et al., 2010). Both of them suppressed the nitrification activity of nitrifying coal ash. However, the different inhibition mechanism for neutral and highly alkaline condition were lacking of sufficient alkalinity and ultra-high FA concentration, respectively (Liu et al., 2021). That leaded to a lower nitrification efficiency at neutral condition and a sharp decrease of NH_4^+-N removal rate when the pH increased slightly from 9.0 to 9.5.

Figure 2b showed the NH_4^+-N removal rate changed with DO concentration from less than 0.5 to 6.5 mg/L. DO as an important factor in nitrification will directly affects the oxygen penetration depth (OPD) and the degree of nitrification (Yuan & Gao, 2010). During the anoxic stage (DO < 0.5 mg/L), the substrate was not be used by the nitrifying coal ash because of the very limited OPD at relatively low DO concentrations, only with 4.41 mg N/(L·h) removal rate at DO of 0.2 mg/L. The NH_4^+-N removal rate increased with the increased DO concentration and peaked at about 120 mg N/(L·h) when the DO concentration was around 6.0 mg/L. That was higher than other systems (Zhu & Chen, 2003), which resulted by the abundant nitrifiers and strong flow turbulence through air diffusion.

Curved fit NH_4^+-N removal rate with T was shown in Figure 2c. The trend of increase and then decrease with increasing T was just like other nitrifiers, which suitable T for nitrification occurred at approximately 30 °C (Cho et al., 2014). As shown in Figure 2c, the optimal T for NH_4^+-N removal rate (between 23 °C and 28 °C) were slightly lower than values previously reported. The growth and bioactivity of nitrifiers was suppressed seriously at low T (Young et al., 2017), and NH_4^+-N removal rate was about 28.16 mg N/(L·h) at 9 °C in this study. That means the nitrifying coal ash possess some nitrification ability in winter.

Model equations developed according to the batch experimental results, showing as in Supporting Information (S2), with R-square more than 0.99. However, the model equations showed the relationship between NH_4^+-N removal rate and single parameter of pH, DO and T. Correction coefficients involved them simultaneously was tested by orthogonal experiment basing on the results of single factor experiment, and the analysis of variance and surface response graph was shown in Supporting Information (S3 and S4).

For example, the NH_4^+-N removal rate reached at 133.69 mg N/(L·h) under pH, DO and T was 8, 4.5 mg/L and 25 °C and that was 30.89 mg N/(L·h) with pH of 7, DO of 3 mg/L and T of 25 °C, as shown in Supporting Information (S5). According to analysis of variance in S2, A, B and A^2 effect of NH_4^+-N removal rate were very significant. And pH was the remarkable factor influencing the NH_4^+-N removal rate among all of the parameters judging by the F-value, followed by DO and T. Surface response graph shows interactions between factors; and the steeper is the surface, the greater is the factor influence on NH_4^+-N removal rate. The curve of pH values changed steeply, which was consistent with the variance analysis. The optimal conditions of pH, DO and T were 8.20, 5.12 mg/L and 25.33 °C, respectively, under the trial conditions.

Then, f (pH, DO, T) was simulated by 1stOpt combining the above experiment results with R-square of 0.999, as shown in Eq. (2). Once again, the mathematical equation verified that pH was the remarkable factor influencing the NH_4^+-N removal rate.

$$f\,(\text{pH, DO, T}) = 7.08 \times 10^{-7} \times y_1^{1.14} \times y_2 \times y_3^{0.80}, \quad (2)$$

where y_1, y_2 and y_3 represent for the NH_4^+-N removal rate decided by pH, DO and temperature, respectively.

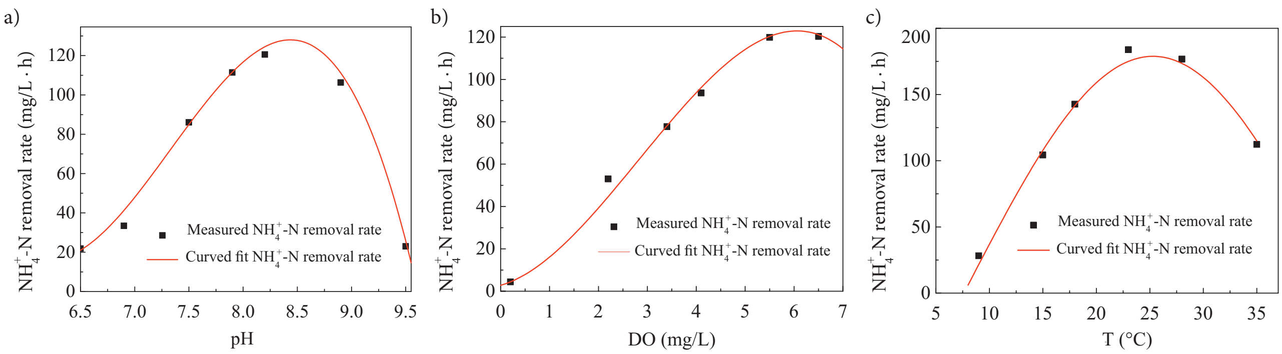

Figure 2. NH_4^+-N removal rate under different pH (a), DO (b) and T (c) conditions

In conclusion, the modified Monod model combining the basic Monod model and the correction factor was shown as Eq. (3):

$$r = \frac{110.48 \times s}{59.19 + s} \times (7.08 \times 10^{-7} \times y_1^{1.14} \times y_2 \times y_3^{0.80}). \quad (3)$$

Bringing these parameters of pH, DO and T into Eq. (3), then worked out the simulated NH_4^+-N removal rate, as shown in Supporting Information (S6). The average gap between experiment data and simulated data was 6.48 mg N/(L·h), while that decreased to 2.74 mg N/(L·h) after introducing the correction coefficients. And the conjoint ratio increased about 5.11% calculating by the ratio of the gap and the experiment data.

2.3. Application of modified Monod

2.3.1. Removal rate of NH_4^+-N with inorganic wastewater

The difference of NH_4^+-N removal rate between the modified Monod model and experiment data response to different NH_4^+-N concentrations was showed in Figure 3. In theory, the increased influent NH_4^+-N concentration from 400 mg/L to 1200 mg/L will led to an increased inhibition on nitrification because of the added FA and FNA (Aktan et al., 2012). However, NH_4^+-N removal rate obtained by experiment just decreased from 99.78 mg N/(L·h) under low concentration to 93.63 mg N/(L·h) under high concentration, showing a slightly downward trend with the rising influent NH_4^+-N. Nitrifiers used was taken from long running reactors feeding with high concentration of ammonia (Liu et al., 2021), which significantly lessened the negative effects of the inhibition of FA and FNA. As showed in Figure 3, the simulated data obtain a more stable removal rate than the actual data, increasing from 90.73 to 97.64 mg N/(L·h). The difference in stability is usually expected between simulated and actual data under unstable water quality, which can be ascribed to the fact the simulated data normally neglect the negative impact of substrate concentration fluctuation on microorganisms and compute based on the average concentration (Linkès et al., 2012). The fluctuation of substrate concentration in a shorter period caused a slow decline of actual data, while the simulated data showed slow growth with the substrate concentration according to Eq. (3). That eventually led to the NH_4^+-N removal rate of actual revealed a relatively high trend than simulated at influent concentration of NH_4^+-N less than 579.48 mg N/L, then, showed an obviously opposite trend at higher concentration as indicating in the graph. According to ANOVA analysis, NH_4^+-N removal rate of actual and simulated showed significant differences. The maximum and minimum differences between them is 8.34 mg N/(L·h) at 443.18 mg N/L and 0.08 mg N/(L·h) at 816.45 mg N/L, occupying about 8.42% and 0.08% of experiment data. These very close data meant the modified Monod had high accuracy when treating inorganic wastewater especially with high-ammonia nitrogen.

Compared to the other previously investigated cases (Valentukevičienė et al., 2018), the modified Monod model in this study only considered the influence of environment parameters on nitrifying coal ash except wastewater parameters, which made it seems simpler. However, it didn't affect the accuracy of the prediction for the nitrifying coal ash mainly aiming at inorganic wastewater with high ammonia.

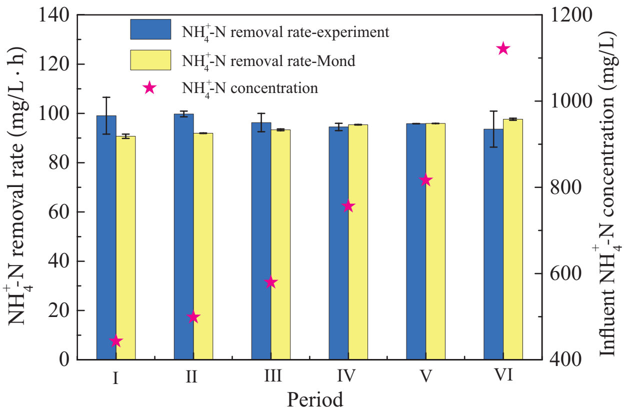

Figure 3. NH_4^+-N removal rate of Monod model at different NH_4^+-N concentration in comparison with the lab observation

2.3.2. Removal rate of NH_4^+-N with sewage

Wastewater is always composed of nitrogen and organic substances. The difference between experimented and simulated removal rate with actual sewage are conducted under pH, DO and temperature of 7.0, 4.0 and 20 °C conditions, respectively, which involved ammonia and organic. As shown in Figure 4, influent COD concentration was ranged from 113.52 to 231.89 mg COD /L and effluent averaged of 33.46 mg COD/L. Compared with the influent concentrations of COD, the effluent were much more stable. It was reported that a high abundance levels of heterotrophic bacteria was existed in inorganic system (Rejish Kumar et al., 2013), which was survived by soluble microbial products (SMPs) mainly included extracellular polymers and dead autotrophic bacteria (Shi et al., 2010). The amount of heterotrophic bacteria ensured the stable effluent concentration and high removal rate of COD (Figure 4a).

However, the presence of COD in system would hinder the nitrification of nitrifiers theoretically (Sharma & Gupta, 2004), and this was verified by the significant difference between the simulated and experimental data (Figure 4b). Divided these data into two phases, phase I and II, according to the effluent concentrations of NH_4^+-N. Influent concentrations of NH_4^+-N fluctuated between 28.50 and 45.12 mg N/L, however, the NH_4^+-N removal rate was low and constantly on the rise in phase I, increasing from 1.03 mg N/(L·h). The adjustment of nitrifiers to organic wastewater would lead to this trend.

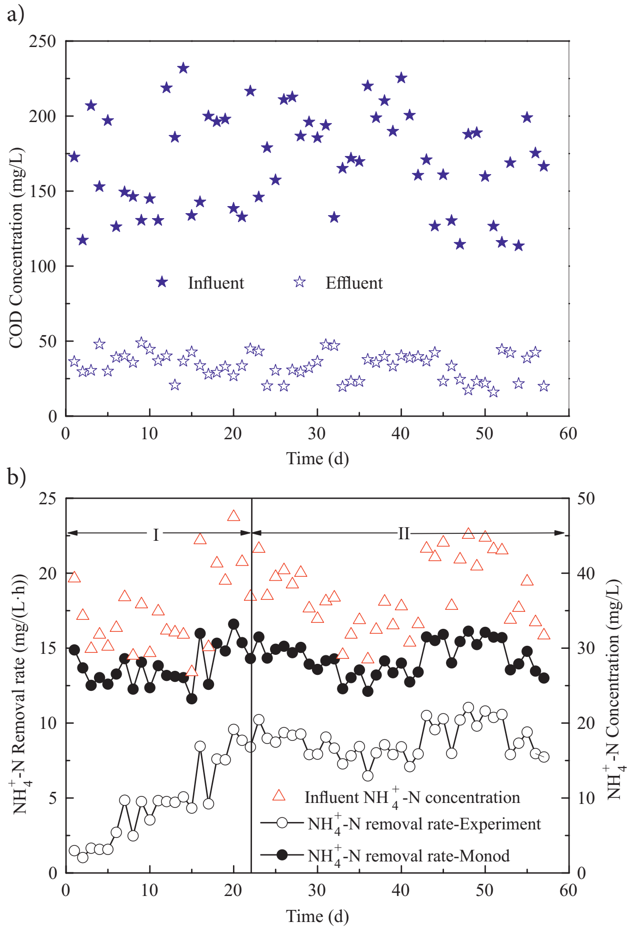

Figure 4. Concentrations of COD (a) and NH_4^+-N (b) simulation result of model in comparison with the lab observation in treating municipal wastewater

In phase II, the NH_4^+-N removal rates of experiment were stable, and the average for that was 8.85 mg N/(L·h) at the stable stage. The trend of experiment and simulated removal rate kept highly consistency in the stable phase, however, the simulated data was apparently higher than the experiment. And the gap between them gradually narrowed to 5.09 mg N/(L·h) from 13.39 mg N/(L·h). Exist of organic in influent not only act as a disincentive to nitrification of nitrifiers but also change the microbial community structure in system (Li et al., 2020). Both of them weaken the NH_4^+-N removal rates of experiment. Combing with the experiment and simulated data, about 40% of NH_4^+-N removal rate was cut down because of the existence of organic in influent with averaged 173.19 mg COD/L. Accordingly, the effect of organic on the stability of nitrifying coal ash, the content of nitrifying bacteria for example, needs to be considered when treating organic wastewater.

Conclusions

A modified Monod model was developed for simulating the nitrification process by using nitrifying coal ash, including pH, DO and T correction coefficients. When treating inorganic wastewater, the results showed the modified Monod model was capable for effectively simulating the NH_4^+-N removal rate especially treating high-ammonia nitrogen, only 0.08% inconsistency with experiment data. In treating organic wastewater, NH_4^+-N removal rate of the nitrifying coal ash was inhibited about 40% with averaged 173.19 mg COD/L.

Acknowledgements

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