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Bioactivity kinetics of organic matter biodegradation and nitrification

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Abstract−Biodegradation of organic matter and nitrification of ammonia nitrogen was studied by measuring the electron transport system (ETS) activity in activated sludge. The feasibility of characterizing the bioactivity of activated sludge based on the ETS was discussed. Then, bioactivity kinetics for the biodegradation and nitrification of organic matter was analyzed using the Michaelis–Menten equation. The results indicated that the ETS activity of activated sludge reflects the progression of organic matter biodegradation and nitrification of ammonia nitrogen; moreover, ETS activity is sensitive to the loading of organic matter and ammonia nitrogen and also to changes in alkalinity during the reaction. Therefore, it is feasible to characterize the bioactivity of an activated sludge system with ETS activity. The Michaelis constant for organic matter biodegradation was K^T_s=368.9 mg/L; U^T_m=90.9 mgTF/(gTss·h); K^I_s=88.42 mg/L; and U^I_m=277.8 mgINTF/(gTss·h); for the nitrification of ammonia nitrogen, the Michaelis constant was K^T_s=16.89 mg/L; U^T_m=34.6 mgTF/(gTss·h); K^I_s=6.0 mg/L; and U^I_m=196.08 mgINTF/(gTss·h). Additional analyses of bioactivity kinetics confirmed that the organic matter oxidation rate of heterotrophic bacteria was higher than that of autotrophic nitrifying bacteria.

Keywords: Organic Matter Biodegradation, Nitrification, ETS Activity, Activated Sludge, Kinetics

INTRODUCTION

Activated sludge activity is an important operational indicator of biological sewage treatment systems. It reflects the microbial degradation capacity of organic matter in terms of molecular biology, as well as the degradation rate and general performance of sewage treatment systems [1,2]. Microbial activity can be evaluated using online monitoring of the sewage treatment effect based on activated sludge activity. Generally, operational parameters can be adjusted automatically or manually, so the sewage treatment system can maintain optimal performance, and the effluent quality can meet standards [3-7]. Moreover, the increasing number of studies on activated sludge activity as well as the application of their findings are of high significance for promoting biological sewage treatment technology and improving the operational control of sewage treatment systems [8-11].

Dehydrogenation of the organic substrate is one of the most important steps in biological degradation to remove pollutants from biological sewage treatment systems. The free electrons produced as a result of microbial aerobic respiration can be transported to natural electron acceptors via electron carriers that are the underlying mechanism for the biodegradation of organic pollutants [9,12-14]. Electron transport system (ETS) activity is an extensively studied indicator, while electron transport rate is considered an indirect indicator of microbial respiration activity, based on which activated sludge activity is quantified. There is no limit on the number of samples detected simultaneously for ETS activity; only conventional laboratory equipment is needed for onsite applications, and the detection procedures are simple [15,16]. ETS activity, which is a measure of microbial degradation capacity of organic matter in terms of molecular biology, also reflects the degradation rate and general performance of sewage treatment systems. It is therefore used extensively for analyzing, evaluating, and predicting biological activity in sewage treatment plants [17].

At present, the most commonly used artificial electron acceptors for detecting the activity of ETS in activated sludge system are 2,3,5-triphenyltetrazoxazole (TTC, 2,3,5-triphenyltetrazolium Chloride) and iodo-nitro-tetrazole (INT, 2-(p-iodophenyl)-3-(p-nitrophenyl)-5-phenyl Tetrazolium Chloride). These two kinds of tetrazolium salts have different characteristics in the detection of sludge ETS activity due to their different functional groups. TTC, with a redox potential of +460 mV, in the determination of TTC-ETS activity replaces the natural H^+/e^{-1} receptor in the respiratory chain of activated sludge microorganism. The respiration of microorganisms reduces the colorless, water-soluble TTC to TF (Triphenyl For-

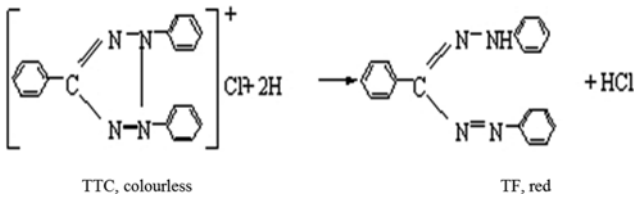

Fig. 1. Reaction of TTC reduction.

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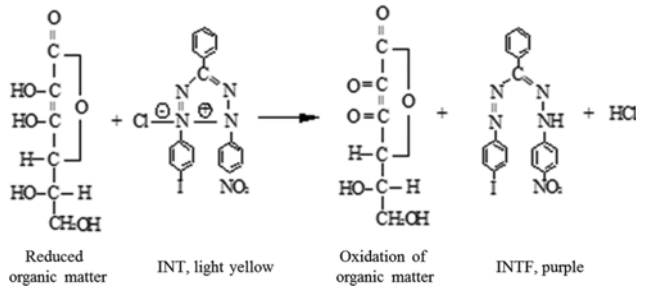

Fig. 2. Reaction of INT reduction.

mazan), a red, water-insoluble substance, TF [18,19]. The activity of TTC-ETS can be quantified by measuring the light density produced by TF with a spectrophotometer, as soon in Fig. 1.

The oxidation-reduction potential of INT is +90 mV; it is a pale yellow crystalline powder dissolved in water. After receiving H+/e, it will produce red violet and water-insoluble INTF (Iodonitrotetrazolium-Formazan). The reaction is shown in Fig. 2 [20-22].

Biodegradation of organic matter and nitrification of ammonia nitrogen are very dominant biochemical reactions in sewage treatment plants that use aerobic microbes. Organic carbon oxidizing microbes and nitrifying microbes are different in terms of nutrition since they are chemoheterotrophic and chemoautotrophic, respectively [23]. Both reactions are associated with electron transport. Organic matter serves as the electron donor, and O_2 is the electron acceptor for the oxidation of organic matter. During nitrification, the electron donor is NH_3 or NO_2^-, while O_2 acts as the electron acceptor [24-27]. In this study, changes in ETS activity during the oxidation and nitrification of organic matter were examined in a sewage treatment plant to discuss the feasibility using ETS activity to characterize activated sludge activity. The relationship as to how the biochemical reaction rate is related to substrate load was investigated using the Michaelis-Menten equation. Furthermore, differences in activation kinetics in organic matter oxidation and nitrification were characterized to promote the biodegradation of pollutants.

MATERIALS AND METHODS

1. Sewage Treatment Plant

The sewage treatment plant used in the experiment, shown in Fig. 3, was made in a cylindrical shape with organic glass and had a total working volume of 5 L. A fine bubble aerator was installed at the bottom of the reactor, and the mixture was agitated by blowing air through it. Compressed air was diffused into the reactor through a sand-core aeration head with the aid of an air pump. Air supply was controlled by a rotameter. An outlet was made at the bottom of the reactor for the discharge of sludge. A temperature sensor and a magnetic stirrer placed at the bottom of the reactor were used to control temperature. When the reactor was inadequately aerated, the sludge was mixed with the substrate to ensure full contact with the help of a stirrer. The reactor was also installed with dissolved oxygen (DO), oxidation-reduction potential (ORP), and pH sensors to monitor dynamic changes. The operating temperature of the reactor was maintained at 25±1 °C.

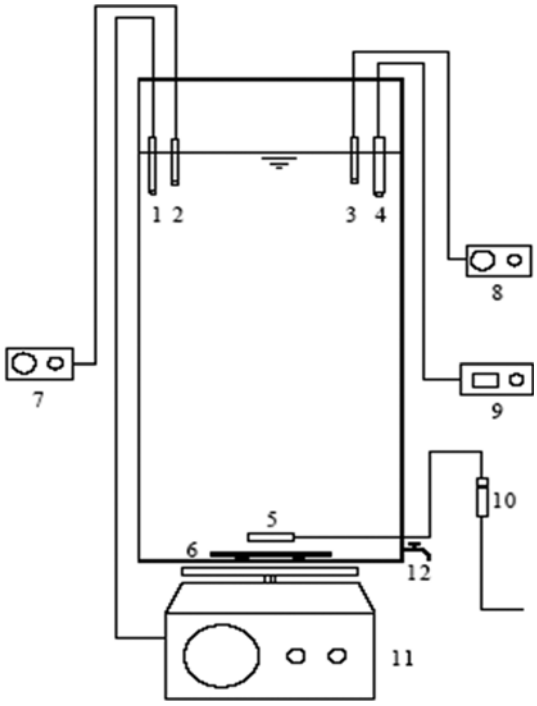

Fig. 3. Schematic diagram of reactor.

1. Temperature sensor
2. pH sensor
3. ORP sensor
4. DO sensor
5. Fine bubble aerator
6. Magnetic follower
7. Acidimetre
8. ORP analyzer
9. DO analyzer
10. Rotameter
11. Magnetic stirrer
12. Sludge outlet

2. Analytical Techniques

2-1. ETS Activity

2-1-1. TTC-ETS Activity

0.8 mL of sludge (mixed liquor), 0.5 mL of 0.36% Na_2SO_3 solution, and 0.3 mL of 0.4% TTC solution were prepared and pour into a 10-mL centrifuge tube. After the mixture was cultured in a 37±1 °C shaking water bath for 30 min, the enzymatic reaction was terminated by adding 1 mL of 37% formaldehyde. All above-mentioned procedures were performed in the dark. The solvent-sludge mixture was centrifuged at 4,000 r/min for 5 min, and the supernatant was discarded. Afterwards, 5 mL of acetone was added to the mixture to properly mix up, and placed in the dark at 37±1 °C for 10 min for shake extraction. The tube was then centrifuged at 4,000 r/min for 5 min, and the supernatant was separated from the precipitated sludge. The absorbance of the extract was measured using a spectrophotometer at a 485-nm wavelength. The precipitated sludge was dried for 1 h at 105±1 °C to measure its dry weight.

2-1-2. INT-ETS Activity

0.5 mL of sludge (mixed liquor) and 0.1 mL of 0.2% INT solution were added in a 10-mL centrifuge tube. The enzymatic reaction was conducted in a shaking water bath at 37±1 °C for 30 min and was terminated by adding 1 mL of 37% formaldehyde all in the dark. The solvent-sludge mixture was centrifuged at 4,000 r/min for 5 min, after which its supernatant was discarded. Then, 5 mL of methanol was added, after which extraction was done while stirring for 10 min at 37±1 °C. The tube was further centrifuged at

4,000 r/min for 5 min, and the supernatant was separated from the precipitated sludge. The absorbance of the extract was measured using a spectrophotometer at a 485-nm wavelength. The precipitated sludge was dried at 105±1 °C for 1 h to measure the dry weight.

ETS activity was calculated using Eq. (1) as follows:

$$U = \frac{D_{485}V}{K_T W t} \quad (1)$$

where U is the ETS activity expressed as ([mgTF]/[gTSS·h]) or ([mgINTF]/[gTSS·h]); D_{485} is the absorbance of the supernatant at a 485-nm wavelength; V is the volume of the extracting agent (mL); K_T is the slope of the standard curve; W is the dry weight (g) of the sludge; and t is the culture duration (h).

2-2. Other Routine Indicators

COD was measured with a 5B-1 rapid COD analyzer; NO_3^--N concentration was measured using thymol spectrophotometry; NO_2^--N concentration was measured with spectrophotometry using N-(1-naphthyl) ethylenediamine dihydrochloride, and NH_4^+-N concentration was measured using Nessler's reagent colourimetric method. Preparation, pre-treatment and procedure were done as previously stated in Guo et al. [28].

EXPERIMENT

1. Organic Matter Biodegradation Experiment

The sludge sample initially obtained from Changchun Xijiao Sewage Treatment Plant, Changchun-China was inoculated to the activated sludge system running in the laboratory. At each inoculation, 1 L of sludge was introduced into the system. The experimental reactor, as shown in Fig. 3, was aerated for 1 h to ensure that all organic substrate was depleted and that all ammonia nitrogen was completely nitrified. Then, 4 L of water containing only organic carbon sources, without nitrogen or phosphorus, was poured into the reactor, after which the mixture was stirred for 30 min to ensure complete denitrification of nitrate nitrogen or nitrite nitrogen in the sludge. After stirring, the system was aerated and sludge was sampled to measure initial TTC- and INT-ETS activity as well as COD. For the subsequent aeration processes, COD value was measured every 10 min together with the measurement of TTC- and INT-ETS activity of sludge. Each experimental cycle was repeated five times after which the average value was calculated as the final value to increase precision. Each cycle was repeated under the same experimental conditions, and the probability of the absolute difference between the test results was not less than 0.95.

2. Nitrification Experiment

Same sludge parameters used during the organic matter biodegradation process were used for the nitrification stage. Before nitrification, the remaining activated sludge was first aerated in the reactor for 1 h to deplete all organic substrate. At this stage, the 4-L substrate contained only ammonia nitrogen. After the aeration of the system, the initial TTC and INT-ETS activity, ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentration were measured. Subsequently, ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentrations were measured every 10 min; the sampling interval for the measurement of TTC- and INT-ETS activity depended on the duration of the experiment.

RESULT AND DISCUSSION

1. Changes in TTC- and INT-ETS Activity During Organic Matter Biodegradation

1-1. One-time Substrate Dosing

The value of DO, ORP and pH as process control parameters for the biological treatment of sewage has received much attention worldwide, making them effective control parameters for the biological treatment of sewage. To further verify the feasibility of characterization of organic matter degradation by TTC and INT-ETS activity of sludge, DO, ORP and pH values in the system were also monitored in real time by detecting sludge TTC and INT-ETS activity and COD. Results are shown in Fig. 4(a).

In Fig. 4(a), the DO was suddenly lowered at the beginning of the aeration system and then maintained unchanged at about 0.2 to 0.5 mg/L forming a platform phase which lasted until the 80th minute. DO began to rise after 80 minutes, indicating COD degradation, exactly the time of low sludge TTC and INT-ETS active platform phase in the reaction system. ORP decreased rapidly during the first 20 min of aeration. As the aeration continued, the descending speed slowed down until the degradation of organic matter was over after 80 minutes and then suddenly increased. The time of occurrence of this characteristic point is the same as the time of transfer of sludge TTC and INT-ETS activity to low active platform phase. pH increased continuously with the aeration, but when the

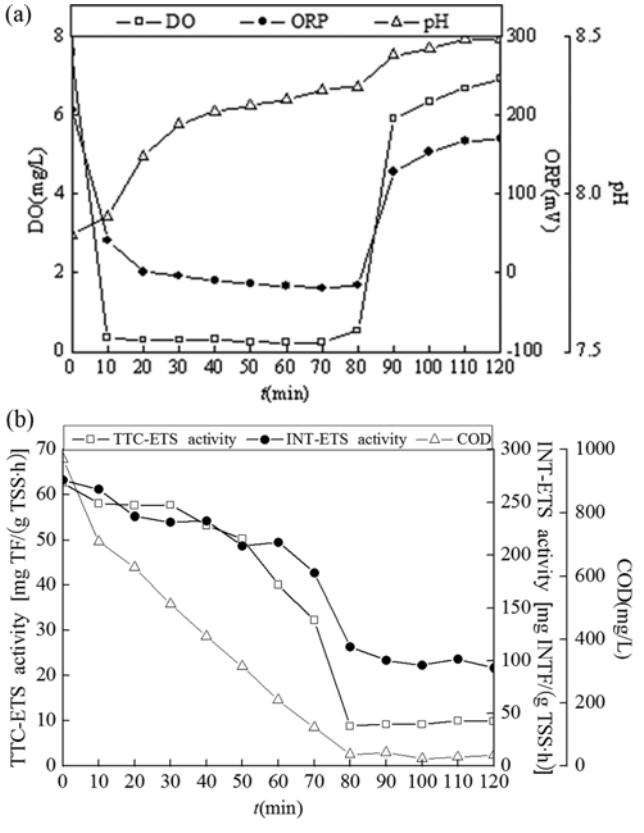

Fig. 4. (a) DO, ORP and pH as a function of time; (b) changes in TTC- and INT-ETS activity and COD concentration over time.

organic matter was degraded, the pH value underwent a significant jump, which was the turning point of the activity of TTC and INT-ETS in the sludge. The results showed that the DO, ORP and pH values in the system were correlated with sludge TTC and INT-ETS activities during the degradation process of organic matter.

One dose of the organic substrate and sodium bicarbonate was added during this stage. In the reactor, the initial COD_{Cr} was 970.75 mg/L; pH value was 7.87, and sludge concentration was about 5,000 mg/L. Fig. 4(b) shows changes in COD, TTC-, and INT-ETS activity for the sewage over time.

It can be seen from Fig. 4(b) that as the aeration duration increased, the COD gradually decreased, and the TTC- and INT-ETS activity decreased as well. At process duration of 80 min, when COD was completely removed, the TTC- and INT-ETS activity sharply decreased until they were below the lower activity threshold, beyond which TTC- and INT-ETS activity stabilized. The results indicated that the TTC-ETS activity had a good correlation with the progression of organic matter biodegradation. Thus, TTC- and INT-ETS sludge activity in the reactor are indirect indicators of COD degradation.

1-2. Intermittent Dosing of Organic Substrate

The feasibility of using TTC- and INT-ETS activity to characterize the progression of organic matter biodegradation was further analyzed. The response of TTC- and INT-ETS activity to changes in organic loading was investigated using intermittent dosing during aeration. An organic substrate was added intermittently three times as the aeration process proceeded. For the first addition, the organic substrate was initially dissolved in 4 L of water before mixing with the activated sludge in the reactor. For the latter two additions, the concentration of required organic substrate was computed and directly poured into the reactor. Fig. 5 shows changes in TTC- and INT-ETS activity and COD over time.

From Fig. 5, TTC- and INT-ETS activity was not only highly responsive to organic loading in the reactor, but also demonstrated a good correlation with changes in COD at each stage. That means TTC- and INT-ETS activity can characterize the progression of organic matter biodegradation. Each stage of organic matter biodegradation can be assessed and tracked based on TTC- and INT-ETS activity.

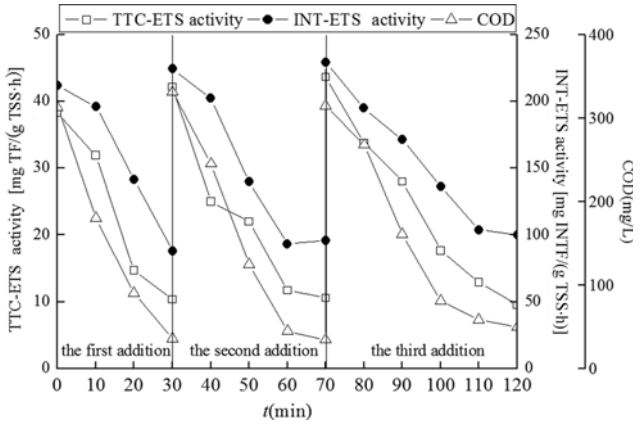

Fig. 5. Changes in TTC- and INT-ETS activity and COD over time with intermittent dosing of organic substrate.

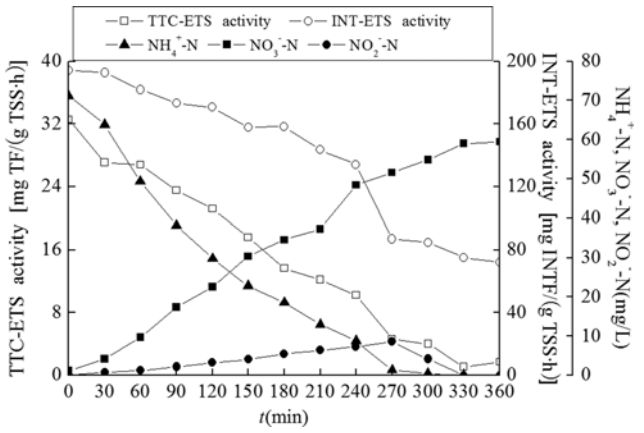

Fig. 6. Changes in TTC- and INT-ETS activity and ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentration over time with 1 dose of ammonia nitrogen.

2. Changes in TTC- and INT-ETS Activity in a Nitrification Reaction

2-1. One-time Ammonia Nitrogen Dosing

As a nitrogen source, ammonium sulfate along with an appropriate amount of sodium bicarbonate was simultaneously added to the mixture in the reactor. The initial NH_4^+-N concentration in the reactor was 71.25 mg/L; pH value was 8.22, and sludge concentration was about 5,300 mg/L. Fig. 6 shows changes in TTC- and INT-ETS activity and ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentration in the reactor as aeration proceeded.

In Fig. 6, TTC- and INT-ETS sludge activity decreased as ammonia nitrogen was consumed. The low-activity stage was reached when the ammonia nitrogen was completely converted (at 270 min). Another obvious decline in TTC- and INT-ETS activity occurred at 330 min, indicating that all nitrite nitrogen in the reactor had been converted into nitrate nitrogen. TTC- and INT-ETS activity experiences two major declines during aeration that correspond to the complete conversion of ammonia nitrogen into nitrate nitro-

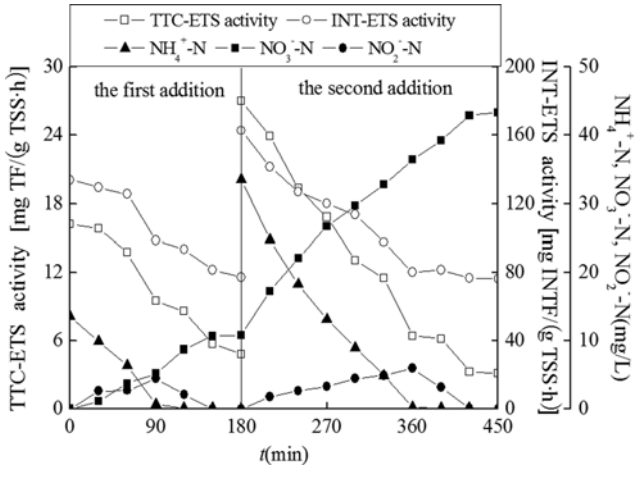

Fig. 7. Changes in TTC- and INT-ETS activity and ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentration over time with intermittent dosing of ammonia nitrogen.

gen and nitrite nitrogen in the sludge, as well as the complete conversion of nitrite nitrogen into nitrate nitrogen. It has been suggested that TTC- and INT-ETS activity is an eligible indicator of the activity of nitrifying bacteria, based on which the nitrification progress can be monitored.

2-2. Intermittent Dosing of Ammonia Nitrogen

Ammonia nitrogen was intermittently added twice to study the response of the TTC- and INT-ETS activity to ammonia nitrogen loading. Fig. 7 shows changes in TTC- and INT-ETS activity over time. For nitrifying bacteria, it was apparent that the TTC- and INT-ETS activity was sensitive to ammonia nitrogen loading. It characterized the progression of nitrification at different stages.

2-3. Intermittent Dosing of Bicarbonates

Nitrifying bacteria are sensitive to changes in pH values in the system, and they generally prosper in a neutral to an alkaline environment. Nitrification of ammonia nitrogen can occur at a fast rate in a proper pH environment, and the maximum specific growth rate of the nitrifying bacteria can also reach its highest possible level [29,30]. A small amount of bicarbonate insufficient for complete nitrification in the reactor was first added. When the nitrification speed slowed down, a second dose of sodium bicarbonate was added to examine how the TTC- and INT-ETS activity responded to a sudden change in alkalinity. The initial ammonia nitrogen concentration in the reactor was 36.19 mg/L; pH value was 7.59, and sludge concentration was about 3,200 mg/L. Fig. 8 shows changes in TTC- and INT-ETS activity and ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentration in the reactor with intermittent bicarbonate dosing.

In Fig. 8, after the first addition of bicarbonate, the pH value of the system and TTC- and INT-ETS activity decreased as nitrification proceeded. After reaching a certain pH threshold (at 120 min), the nitrification speed slowed, and TTC- and INT-ETS activity decreased as well. The second addition of bicarbonate caused an abrupt increase in the pH of the system, and the TTC- and INT-ETS activity returned with an increase in nitrification speed. The pH of the system decreased again as nitrification proceeded, and the TTC- and INT-ETS activity decreased correspondingly. However, the pH value of the system became a limiting factor for nitrification at 270 min; the TTC- and INT-ETS activity abruptly decreased with a small amount of ammonia nitrogen left in the reactor. After nitrification proceeded at a low speed for about 90 min, nearly all the ammonia nitrogen was converted, which caused another downturn in TTC- and INT-ETS activity; this characteristic decline indicated that all the ammonia nitrogen was converted, which was accompanied by a concave turn in the pH curve, followed by an increase. All nitrite nitrogen was converted to nitrate nitrogen at 330 min, leading to pH stabilization in the reactor. At this moment, the TTC- and INT-ETS activity demonstrated another obvious decline, after which stabilization occurred. The variational pattern for the pH value during nitrification was consistent with that of other literature reports [31,32]. Combining these results with detection, we concluded that TTC- and INT-ETS activity could effectively characterize changes in the activity of nitrifying bacteria; they can be used to monitor the progression of nitrification.

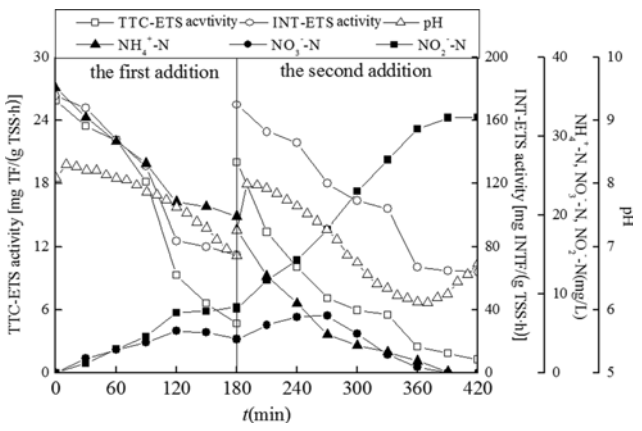

Fig. 8. Changes in TTC- and INT-ETS activity and ammonia nitrogen, nitrite nitrogen, and nitrate nitrogen concentration over time with intermittent bicarbonate dosing.

3. Activity Kinetics Analysis

Monod model and Haldane model are based on the total biomass and it is difficult to distinguish living bacteria from dead bacteria in the wastewater treatment process. This study was based on Michaelis-Menten model upon which electron transfer system (ETS) activity was introduced as its basics. The activity of ETS can be used as a model to analyze the relationship between biochemical reaction rate and substrate load. It can effectively characterize microbial activity and degradation efficiency of pollutants in the system, as well as the biological activity kinetics of biodegradation of pollutants by microorganisms.

TTC- and INT-ETS activity exhibits a linear relationship with substrate concentration during organic matter biodegradation and nitrification that can be expressed using the Michaelis-Menten equation. The relationship of TTC-ETS activity (U^T) or INT-ETS activity (U^I) with maximum TTC-ETS activity (U^T_m) or maximum INT-ETS activity (U^I_m), respectively, resembles that of the specific growth rate of the microbes (μ) with the maximum specific growth rate of the microbes (μ_m). This relationship can be expressed by the following formulas (2)-(3) [31,32]:

$$U^T = \frac{U^T_m S}{K^T_S + S} \quad (2)$$

where U^T is TTC-ETS activity ([mg TF]/[g TSS·h]); U^T_m is the maximum TTC-ETS activity ([mg TF]/[g TSS·h]); K^T_S is the Michaelis constant for TTC-ETS activity (mg/L); and S is the concentration of organic substrate or ammonia nitrogen (mg/L).

$$U^I = \frac{U^I_m S}{K^I_S + S} \quad (3)$$

where U^I is INT-ETS activity ([mg INTF]/[g TSS·h]); U^I_m is the maximum INT-ETS activity ([mg INTF]/[g TSS·h]); K^I_S is the Michaelis constant for INT-ETS (mg/L); and S is the concentration of organic substrate or ammonia nitrogen (mg/L).

The experimental data were inputted into formulas (2) and (3). The kinetic constants U^T, U^T_m, K^T_S, and K^I_S were calculated for the organic matter biodegradation and ammonia nitrogen nitrification stages using a double-reciprocal plot, as shown in Figs. 9-12. As indicated by the calculated kinetic constants, U^T and U^T_m were higher for COD removal than for nitrification. This is a convincing evi-

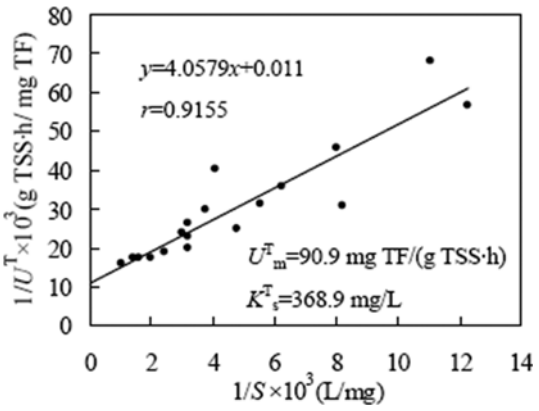

Fig. 9. $1/U^T-1/S$ relationship in COD removal.

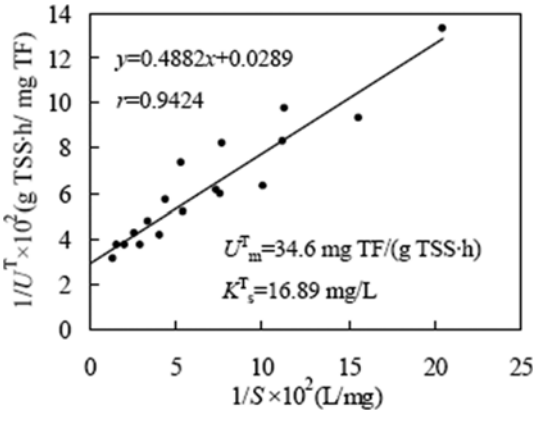

Fig. 10. $1/U^T-1/S$ relationship in ammonia nitrogen nitrification.

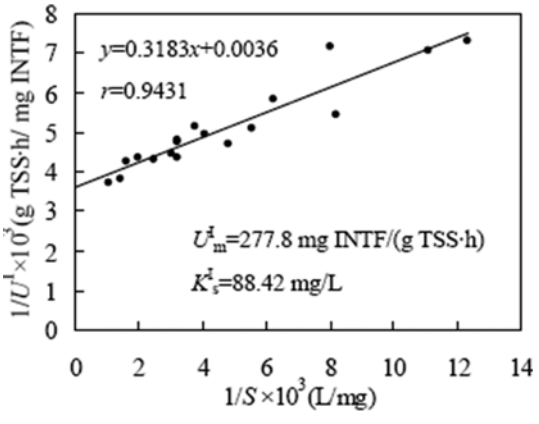

Fig. 11. $1/U^I-1/S$ relationship in COD removal.

dence from the perspective of sludge activity kinetics as the organic matter oxidation rate is higher for heterotrophic bacteria than for nitrifying bacteria.

CONCLUSIONS

The experiment demonstrated that TTC- and INT-ETS activity could characterize the progression of organic matter biodegradation using a single organic matter. TTC- and INT-ETS activity can also reflect the response of microbial activity to organic matter loading. Thus, it is feasible to characterize the bioactivity of an activated sludge system with TTC- and INT-ETS activity.

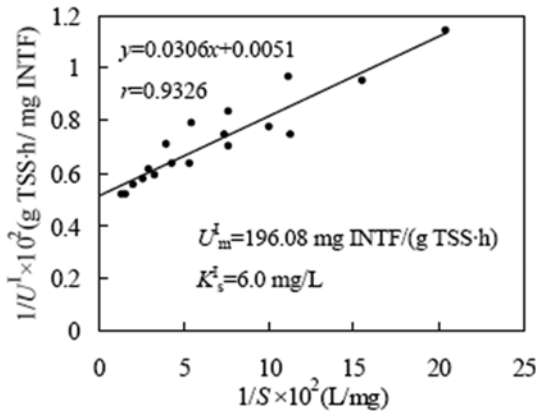

Fig. 12. $1/U^I-1/S$ relationship in ammonia nitrogen nitrification.

For nitrifying bacteria, both TTC- and INT-ETS activity demonstrated characteristic declines at the end of nitrification and the complete conversion of nitrite nitrogen to nitrate nitrogen. This implies that TTC- and INT-ETS activity is a qualified indicator of the progression of nitrification; they can reflect the dynamic response of nitrification speed to ammonia nitrogen loading or to sudden changes in alkalinity.

Activity kinetics analysis of the activated sludge demonstrated that the organic matter oxidation rate was higher for heterotrophic bacteria than for autotrophic nitrifying bacteria.

ACKNOWLEDGEMENT

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NOMENCLATURE

U : ETS activity [mgTF/(gTSS·h), or mgINTF/(gTSS·h)]
D_{485} : absorbance of the supernatant at a 485-nm wavelength
V : volume of the extracting agent [mL]
K_T : slope of the standard curve
W : dry weight of the sludge [g]
t : culture duration [h]
U^T : TTC-ETS activity [mgTF/(gTSS·h)]
U^T_m : the maximum TTC-ETS activity [mg TF/(g TSS·h)]
K^T_s : michaelis constant for TTC-ETS activity [mg/L]
U^I : INT-ETS activity [mgINTF/(gTSS·h)]
U^I_m : the maximum INT-ETS activity [mg INTF/(g TSS·h)]
K^I_s : michaelis constant for INT-ETS [mg/L]
S : concentration of organic substrate or ammonia nitrogen [mg/L]

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