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An ultraviolet/biological (UV/B) reactor for the removal of nitrogenous compounds from the secondary effluent of wastewater treatment plants (WWTPs)

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Abstract: An ultraviolet/biological (UV/B) reactor was used to treat the secondary effluent of wastewater treatment plants (WWTPs). A continuous flow experiment proved that the UV/B reactor could significantly reduce nitrogenous compounds, and the UV/B reactor had a higher removal rate of total nitrogen (TN), nitrate nitrogen (NO_3^--N), ammonia nitrogen (NH_4^+-N), Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD) at an UV/biological degradation stage than a biological degradation stage. The effect of hydraulic retention time (HRT), water temperature, pH, and dissolved oxygen (DO) on NO_3^--N removal in the UV/B reactor was discussed, and it was found that the HRT and water temperature significantly influenced the NO_3^--N removal efficiency, but the effect of pH and DO on NO_3^--N removal was not significant.

Key words: secondary effluent; nitrogenous compound removal; ultraviolet/biological (UV/B) reactor; biological reactor; denitrification

1. Introduction

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The rapid development of industry and urbanization as well as population growth in China has resulted in increasingly serious water shortages and water pollution.[1, 2] Secondary treatment is a significant process undertaken in wastewater treatment plants (WWTPs) in China.[3] The concenpt of secondary treatment was that the wastewater was treated by an activated sludge process after some physical treatment processes (e.g. sediment, sand setting), the activated sludge process include A^2/O process, A^2/O^2 process, oxidation ditch and SBR. The secondary treatment process has a good removal efficiency for organic matter that is readily degraded and ammonia nitrogen (NH_4^+-N). Refractory organic matter is residualed and NH_4^+-N is converted into nitrate nitrogen (NO_3^--N) , resulting in the secondary effluents of WWTPs being characterized by high concentrations of NO_3^--N and total nitrogen (TN), as well as a low chemical oxygen demand (COD). [1, 4, 5] Moreover, some secondary effluent from WWTPs cannot meet the grade I (A) Discharge Standard of Pollutants for Municipal Wastewater Treatment Plant in China (GB 18918-2002) due to technical and management reasons (GB18918-2002 was showed in table 1). the secondary effluent was an effluent from secondary treatment system, e.g. the effluent from A^2/O, A^2/O^2, oxidation ditch and SBR, the A^2/O, A^2/O^2, oxidation ditch and SBR were presently main treatment processes of WWTPs in China. Such secondary effluents lead to eutrophication and water quality deterioration, as well as the ecological disturbance of receiving water bodies. In addition, secondary effluent from WWTPs contains materials that could be reused, and therefore the advanced treatment of the secondary effluent from WWTPs is urgent and necessary.

Table1 The highest discharged concentrations of relative basic control project (day mean value)(mg/L)

	Basic control project	Grade 1		Grade 2	Grade 3
		A	B		
1	Chemical oxygen demand（COD）	50	60	100	120
2	Biological oxygen demand（BOD_5）	10	20	30	60
3	Total nitrogen（TN）	15	20	-	-
4	Ammounia nitrogen（NH_4^+-N）①	5（8）	8（15）	25（30）	-

Note: The number outside of brackets is concentration control when water temperature is higher than 12 ^{o}C, the number inside of brackets is concentration control when water temperature is not higher than 12 ^{o}C

Many methods have been used to further treat nitrogenous compounds from the secondary effluents of WWTPs for the purpose of reuse or reduction of pollution. The most commonly used methods are based on physico-chemical techniques, such as advanced oxidation processes, adsorption, and filtration; [6, 7] however, nitrogenous compounds cannot be removed effectively from the secondary effluent of WWTPs by physico-chemical methods. The high energy and monetary costs, as well as low removal efficiency for nitrogenous compounds are serious drawbacks of the physical-chemical methods.

In recent years, some novel biological technologies have been developed for the removal of nitrogenous compounds from the secondary effluent of WWTPs. Biological methods for the removal of nitrogenous compounds should be able to overcome the deficiencies of the physico-chemical methods, because they can avoid the high energy costs. [2, 4] Zhao et al. developed a compound natural treatment system (Primary Subsurface Vertical Flow Wetland (PSVFW) +

Submerged Macrophyte Oxidation Ponds (SMOPs) + Secondary Subsurface Vertical Flow Wetland (SSVFW)) for nitrogen removal from WWTP secondary effluent that resulted in a TN reduction of 75.8%.[8] He and Xue applied an algal-based immobilization process to treat the effluent from a secondary WWTP and achieved a TN and COD average removal rate of 36% and 32%, respectively. [9] The above technologies also have shortcomings including a relatively low nitrogenous compound removal rate due to limited assimilation by macrophytes or microalgae, as well as large amounts of sunshine and an appropriate water temperature.[10]

The removal of nitrogenous compounds by microorganisms (nitrifying bacteria and denitrifying bacteria) via nitrification and de-nitrification processes is the most effective and economic biological treatment process. [11] However, the de-nitrification process requires a carbon source as an electron donor. The endogenous carbon (residual organic matter) in the secondary effluent of WWTPs is difficult to biologically degrade and the quantities of endogenous carbon are also very limited, resulting in insufficient levels of available carbon for de-nitrification. [4] However, there are disadvantages associated with liquor carbon sources such as methyl alcohol, [12] alcohol, [13] and acetic acid, [14] related to the need for sophisticated process control, which is necessary for the avoidance of overdosing risks, with a resulting deterioration in effluent water quality. [15] To solve the problem, many carbon based biodegradable polymers and biofilm carriers have been used to remove nitrate and TN from water, [1, 4, 16, 17] utilized biodegradable matter (polyhydroxyalkanoates (PHA) and wheat straw) as biofilm carriers and carbon sources to remove the TN from the secondary effluent of WWTPs. Cao used filamentous bamboo as a biofilm carrier and carbon source to remove the TN, [1] and although the TN removal rate was greatly improved, the bioavailability of the decomposed products of the biodegradable matter, especially the wheat straw and filamentous

bamboo, requires further study. In addition, it is feasible that the residual organic matter in the secondary effluent of WWTPs could act as a carbon source. However, residual organic matter is also difficult to biologically degrade using traditional biological treatment processes. Moreover, the discharge of residual organic matter has two shortcomings: (1) excessive changes in the COD or BOD_5 concentrations and (2) organic matter is not reused. The biodegradability of the residual organic matter in the secondary effluent of WWTPs must be enhanced to improve the sequential biochemical utilization. Consequently, to increase the treatment efficiency of the secondary effluents from WWTPs, it is important to enhance the bioavailability of residual organic matter and products containing bio-degradable matter (PHA, wheat straw, filamentous bamboo, and rice straw).

Many studies have used ultraviolet (UV) irradiation in photocatalysis, ultrasonic degradation, and ozone pre-oxidation technologies to convert complex chemical structures into simpler intermediates that are more bioavailable and can be biodegraded more readily by microorganisms. [18-21] UV irradiation technologies are the simplest and most traditional treatment processes available to engineers.

The focus of most previous studies has been the removal of one or more complex chemicals (e.g. pyridine, phenol, and 2,4,6-trichlorophenol) using an integrated ultraviolet/biological (UV/B) reactor, [18,20,22] but in this study we developed a UV/B bioreactor with filamentous bamboo as a biocarrier for treating secondary effluents. The objectives of this study are outlined below.

(1) To compare the removal efficiency of nitrogenous compounds (particularly NO_3^--N) and organic matter from secondary effluent from WWTPs in terms of efficacy and performance, when

using a UV/B reactor with filamentous bamboo to enhance removal, and to study the structural changes of filamentous bamboo during the experiment.

(2) To determine the characteristics of NO_3^--N removal with changes in water temperature, pH, dissolved oxygen (DO) and influent concentration.

2. Materials and methods

2.1 Filamentous bamboo

Samples of filamentous bamboo were obtained, by cutting 20 × 5 × 1 mm pieces. The measured physical characteristics of the filamentous bamboo were as follows: porosity, 80.4%; specific surface area, 118.1 m^2/m^3 (Autosorb IQ, USA); and bulk density, 1.1 kg/L.

2.2 Bioreactor and Biofilm formation

An internal-circulation baffled biofilm reactor (ICBBR) was used to treat secondary effluent from WWTPs, as shown in Figure 1. The reactor, which had a total liquid volume of 40 L, had the following components: (1) upflow and downflow sections (separated by a segregation board), both with a volume of 15 L, (2) top and bottom sections comprising an upper settling section with a volume of 7 L, and (3) a lower dilution section with a volume of 3 L for the inflow of air.

Figure 1. Schematic diagram of the internal circulation of a baffled biofilm reactor (ICBBR)

Filamentous bamboo was installed in both the upflow and downflow sections of the reactor. Raw water (untreated secondary effluent samples), obtained from the secondary effluent of a

WWTP in Xuzhou, China, was poured into a tank, from which it was pumped into the dilution section of the reactor. The flow rate was regulated using a peristaltic pump, and the column was operated in internal circulation mode. In addition, air was supplied to the bottom section of the reactor. The DO concentration in the top section of the ICBBR was 3.5-4.5 mg/L. The experiment was conducted at a water temperature of 13-28°C.

Seed sludge was collected from a WWTP (the secondary treatment process of this WWTP was oxidation ditch meathod) and fed into the reactor together with 2,552–3,621 mg/L of mixed liquor suspended solids (MLSS). This culture was maintained until a steady-state biomass loading on the filamentous bamboo was achieved. Microorganisms that did not adhere to filamentous bamboo were discarded from the valve at the bottom of the reactor.

To facilitate the photolysis of residual organic matter and products from the decomposition of bamboo, a UV light was located 10 cm above the water surface (wavelength: 253.7 nm (UV-C), power: 50 W, light intensity: 1.2 mW/cm^2).

2.3 Assessing the treatment of the effluents from an urban secondary wastewater treatment plant

The reactor, in which a steady-state biofilm was placed over the filamentous bamboo, was fed with secondary effluent from the WWTP with the following characteristics: COD, 85–145 mg/L; BOD, 17–42 mg/L; NH_4^+-N, 11.4–14.9 mg/L; NO_3^--N, 6.7–10.7 mg/L; nitrite nitrogen (NO_2^--N), 0.67–1.09 mg/L; TN, 22.4–26.8 mg/L; suspended solids (SS), 72.5–144.5 mg/L; pH, 6.8–8.3; and DO, 2.5-3.4 mg/L. The hydraulic retention time (HRT) of the continuous flow reactor was 5 h. The experiment was separated into two stages. (1) A biological degradation stage (B stage), in which the

main aim was to investigate the removal efficiency of nitrogenous compounds and organic matter using only a biofilm reactor, which utilized filamentous bamboo as a biocarrier. The experimental conditions were as follows: pH, 6.8-8.3; HRT, 5 h; and water temperature, 20-28□. (2) A UV/biological degradation stage (UV/B stage), in which the aim was to study the integrated UV irradiation/biological degradation of nitrogenous compounds and organic matter based on the experimental stage described above**.** The experimental conditions were as follows: pH, 7.0-8.2; HRT, 5 h, and water temperature, 13-22□. In addition, the effect of water temperature, DO, HRT, and pH on NO_3^--N removal characteristics was considered.

2.5 Analytical methods

Samples were collected at regular intervals and tested within 2 h of collection. All data generated in the study were obtained from three replicate trials. Samples were filtered through a 0.45-μm pore size membrane filter prior to analysis. The N content (including NH_4^+-N, NO_3^--N, and NO_2^--N) was determined using an ion chromatograph analyzer (model PIC-10A: Puren Instrument Co., Ltd., Qingdao, China). The TN and COD were assayed according to Chinese SEPA Standard Methods, [23] and the water temperature, pH, DO, and water temperature values were measured with a pH and oxygen meter (model, Oxi300i, WTW GmBH, Germany).

The carriers with biofilm were observed under a scanning electron microscope (model 6380LV: JEOL, Japan) at 20kV and the micro-structure of the filamentous bamboo were observed under a different scanning electron microscope (model, XL-30: ESEM, Holland) at 20kV. Changes in the surface structures of filamentous bamboo were observed under a stereomicroscope (model, K700: Motic, China). These samples were filtered through a 0.45 μm pore size membrane filter

before measurement.

2.6 Statistical analyses

Treatment methods were compared using one-way analysis of variance (ANOVA) and the least significant difference (LSD) procedure was used for the purpose of mean comparisons, using a significance level of $p = 0.05$. Statistical analyses were performed with SPSS Base 19.0 statistical software (SPSS Inc., Chicago, IL, USA).

3 Results

3.3 Removal efficacy of nitrogenous compounds

Figure 2 shows that the initial TN concentration was in the range of 22.4–26.8 mg/L. In the UV/B stage, the final TN concentration was in the range of 4.20-6.51 mg/L and the TN removal rates were 73.3%-83.2% with a mean value of 77.9%. In the B stage, the final TN concentration was in the range of 5.87-7.95 mg/L and the TN removal rates were 66.6%-75.2% with a mean value of 68.8%.

Figure 2. Removal efficiency of TN, NH_4^+-N, NO_3^--N, and NO_2^--N

The initial concentrations of NH_4^+-N, NO_3^--N, and NO_2^--N were in the range of 11.4–14.9, 7.1–10.7, and 0.67–1.09 mg/L, respectively. In the UV/B stage, the final concentrations of NH_4^+-N,

179 NO_3^--N, and NO_2^--N were in the range of 3.04-5.26, 0.76-1.78, and 0-0.20 mg/L, respectively, and
180 the NH_4^+-N, and NO_3^--N removal rates were 60.5-78.1% and 81.1-92.2%, with mean values of 68.0
181 and 86.5%, respectively. In the B stage, the final concentrations of NH_4^+-N, NO_3^--N, and NO_2^--N
182 were in the range of 4.23-5.91, 1.14-1.69, and 0.09-0.139 mg/L, respectively, and the NH_4^+-N and
183 NO_3^--N removal rates were 51.0-63.6%, 78.3-85.6% with mean values of 53.8 and 80.6%.
184 Compared to the UV/B stage, the average NO_2^--N accumulation was more than 0.04 mg/L in the B
185 stage. There were statistically significant differences of the TN, NH_4^+-N, NO_3^--N removal
186 efficiency between UV/B stage and B stage ($P < 0.05$).

187 In both the B and UV/B stages, the initial TN and NH_4^+-N concentrations did not meet the
188 Class-I (A) standard, in terms of the emission standards for pollutants from urban sewage treatment
189 plants (GB18918-2002), but the final TN and NH_4^+-N concentrations were much lower than those
190 of the maximum contaminant levels (15 and 8 mg/L, respectively) required to meet the Class-I (A)
191 emission standard. The final NO_3^--N concentration was much lower than the maximum allowable
192 contaminant content (10 mg/L) in the "Standards for Drinking Water (GB5749-2006)" of China.

193 A change in the filamentous bamboo before and after usage is indicated in Figure 3 and a
194 change in the surface micro-structures of bamboo is shown in Figure 3.

195

196

197 Figure 3 Changes in filamentous bamboo before (b, 20×) and after usage (a, 8×)

198 Figure 3 indicates that the surface of the filamentous bamboo was bio-eroded and contained
199 many irregular holes (maximum length of about 850 μm) after six months usage (Figure (a)). Most
200 holes ranged in size from 50-550 μm, but the surface of the raw filamentous bamboo was smooth

and dense (Figure (b)). The surface of the filamentous bamboo was decomposed by microorganisms, with the decomposed product of the filamentous bamboo becoming a carbon source. The holes on the surface of the filamentous bamboo also improved the specific surface area and increased the biomass of filamentous bamboo.

Figure 4 Changes in the surface micro-structures of bamboo

(Note: figure (a) shows the surface micro-structures of raw bamboo, figure (b) shows the surface micro-structures of bamboo used in the experiments)

Figure 4 shows that the fibers of the bamboo used in the experiment were already decomposed compared to the raw bamboo, which showed that the bamboo fiber can be decomposed by the microorganisms on the filamentous bamboo. The decomposed products of bamboo are chemical compounds with a complex structure, which can be carbon sources for denitrification after UV irradiation.

Higher removal rates of nitrogenous compounds were achieved during the UV/B stage than the B stage, which indicated that the quantity and quality of the carbon source was improved due to UV irradiation. The residual organic matter and decomposition products of bamboo can be sources of carbon and these materials were repeatedly UV irradiated and denatured due to the internal-circulation in the reactor. Both the residual organic matter and decomposition products of bamboo have a complex chemical structure and bio-refractory performance. The UV irradiation converts complex chemical compounds into much simpler intermediates or simple chemical compounds, [18, 22] which can then be biodegraded more readily by microorganisms, including denitrifying bacteria.

2 Removal efficacy of COD and BOD

The initial COD and BOD_5 were in the range of 85–145 and 17–42 mg/L, respectively. In the B stage, the corresponding COD and BOD_5 removal rates were in the range of 78.9–88.9% (mean value 84.2%) and 29.7-55.0% (mean value 43.7%), respectively. In the UV/B stage, the corresponding COD and BOD_5 removal rates were in the range of 80.0–91.3% (mean value 87.2%) and 40.9-81.0% (mean value 61.6%), respectively. The initial COD and BOD_5 concentrations did not comply with the emission standards for pollutants from an urban sewage treatment plant (GB18919-2002). However, the COD and BOD_5 concentrations in the effluent in the B and UV/B stages met the Class-I (A) emission standards for pollutants from an urban sewage treatment plant (GB18919-2002). There were statistically significant differences of the BOD_5 removal efficiency between UV/B stage and B stage ($P < 0.05$) but no significant difference of the COD removal efficiency($P > 0.05$).

Compared with the B stage, the UV/B stage achieved more than 3.0% removal of COD and 17.9% removal of BOD_5, which suggested that UV irradiation can enhance the removal of COD and BOD_5. The biofilm (Figure 5) adhered on the filamentous bamboo proliferated and degraded the organic matter. The residual organic matter and decomposition products from the bamboo transferred into the reactor, and were repeatedly irradiated because of the internal circulation in the reactor.

Figure 5. The biofilm adhered on the filamentous bamboos

3. The effect of operational conditions on the NO_3^--N removal efficiency

Many studies have shown that the denitrification process (NO_3^--N removal) is a limiting factor

247 during TN removal via nitrification and denitrification. The denitrification efficiency would
248 substantially influence the TN removal characteristics. The effect of HRT, water temperature, pH,
249 and DO on NO_3^--N removal in the UV/B reactor is shown in Figure 6.

250 The effect of HRT on NO_3^--N removal was obvious (Figure 6(a)). When the HRT was 1.5 and
251 3 h, the NO_3^--N removal rate was 45.4 and 52.2%, respectively, but when the HRT lasted for more
252 than 5 h, the NO_3^--N removal rate exceeded 85.2%, and the effluent NO_3^--N concentration was
253 lower than 1.4 mg/L. In addition, the effluent NO_2^--N concentration was constantly lower than 0.12
254 mg/L.

255 The effect of water temperature on NO_3^--N removal was also obvious (Figure 6(b)). When the
256 water temperature increased from 10 and 25□, the effluent NO_3^--N concentrations were
257 substantially decreased because the denitrifying bacteria were stimulated when the water
258 temperature increased. When the water temperature was more than 25□, the NO_3^--N removal rate
259 was reduced. In addition, the effluent NO_2^--N concentration was constantly lower than 0.12 mg/L.

260 The effect of both pH and DO on NO_3^--N removal was not significant, the effluent NO_3^--N
261 concentrations did not change significantly when the pH value ranged from 6.0-9.0 and the DO
262 ranged from 2.3-6.5 mg/L (Figures 6(c) and (d)).

263

264 Figure 6. The effect of pH, DO, water temperature, and influent NO_3^--N concentration on NO_3^--N
265 removal

266 **4. Discussion**

The key finding of this experiment was that the chemicals with a complex structure were continuously decomposed due to the internal circulation of the reactor, and both denitrification and bio-degradation of organic matter were increased due to UV irradiation, but the biofilm on the filamentous bamboo was reproduced at a relatively steady rate without being influenced by the UV radiation. The biofilm was maintained in a steady state and a carbon source was circulated in the UV/B reactor, with simple chemical compounds supplied continuously for de-nitrification and bio-degradation.

In addition, an UV/B bioreactor with filamentous bamboo as a biocarrier has an ampler biomass, cheaper biocarrier cost and less aeration as well as lower excess activated sludge based on the previous experiments results. Consequently, the UV/B bioreactor with filamentous bamboo as a biocarrier would be feasible economically in practice, and the power of UV lamp was 50W, the consume of electricity was also low.

Lastly, the proposed method has good environmental sustainability. Firstly, the proposed method is a biological treatment method, which has less secondary pollutants and less energy consume. Then, filamentous bamboo was natural and biodegradable material, its different processes including manufacturing process, utilized process and post-utilized process have not pollution compared to others biocarriers. Finally, the radiation pollution of UV lamp can be easily prevented by using partition.

4. Conclusions

A UV/B reactor was proposed to treat secondary effluent from a WWTP in this study, and the results obtained can be summarized as follows:

1. Compared with the B stage, the UV/B stage achieved a higher removal efficiency for TN, NO_3^--N, NH_4^+-N, COD, and BOD_5, and accumulated lower quantities of NO_2^--N. The carbon source was the decomposed products of bamboo and the residual organic matter of the secondary effluent.

2. The NO_3^--N removal efficiency was significantly influenced by HRT and water temperature, but changes in DO and pH did not have an obvious effect on NO_3^--N removal.

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342

343 **Captions of Figures**

344 Figure 1. Schematic diagram of the internal circulation of a baffled biofilm reactor (ICBBR)

345 Figure 2. Removal efficiency of TN, NH_4^+-N, NO_3^--N, NO_2^--N

346 Figure 3. Changes in filamentous bamboo before (b, 20×) and after usage (a, 8×)

347 Figure 4. Changes in the surface micro-structures of bamboo

348 Figure 5. The biofilm adhered on the filamentous bamboos

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350 removal

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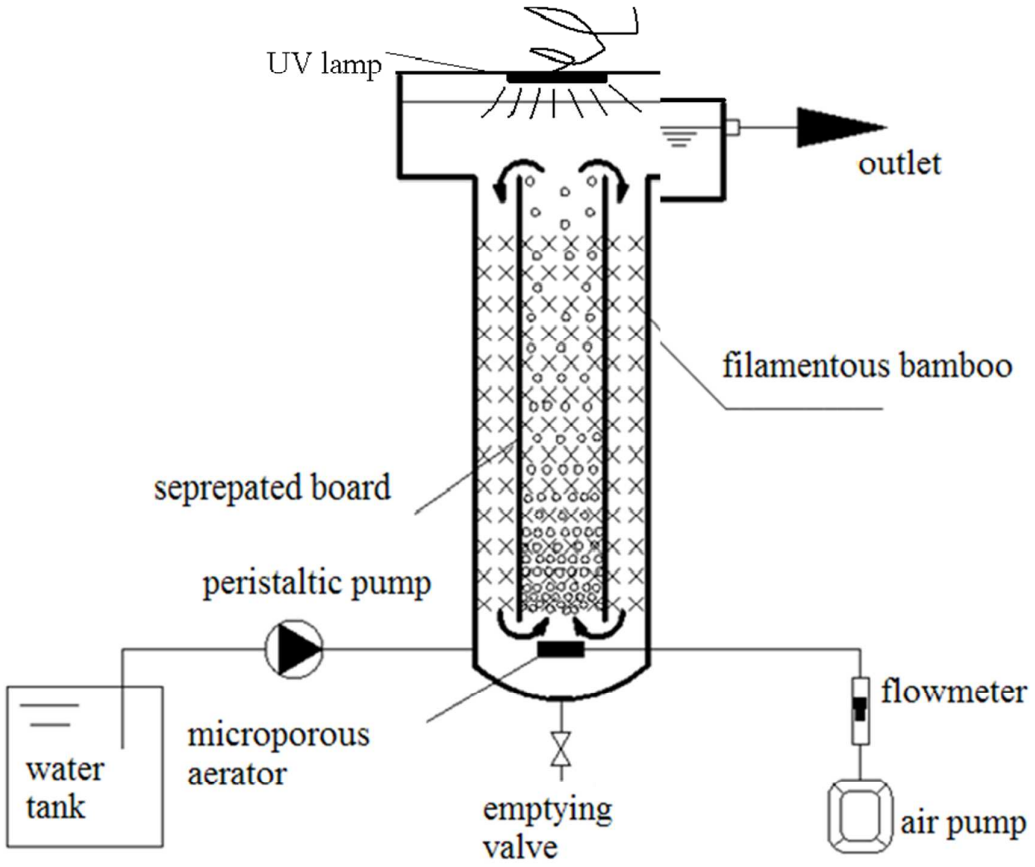
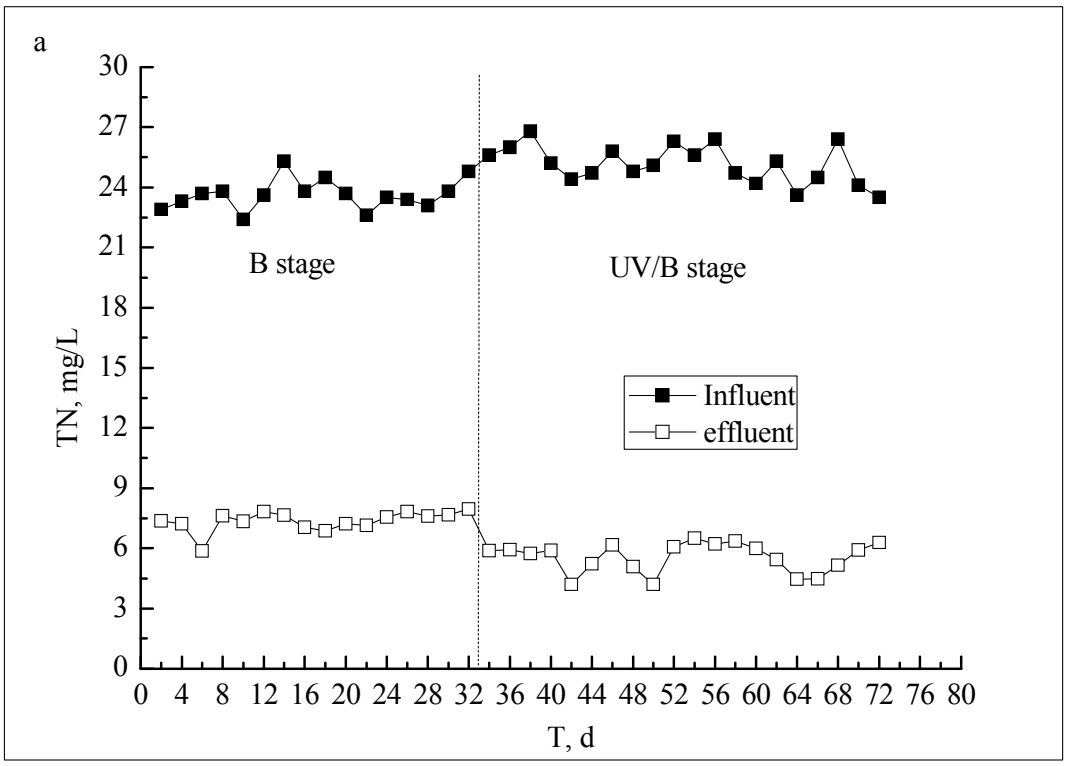


Figure 1. Schematic diagram of the internal circulation of a baffled biofilm reactor (ICBBR)


a
TN, mg/L
T, d
B stage
UV/B stage
Influent
effluent

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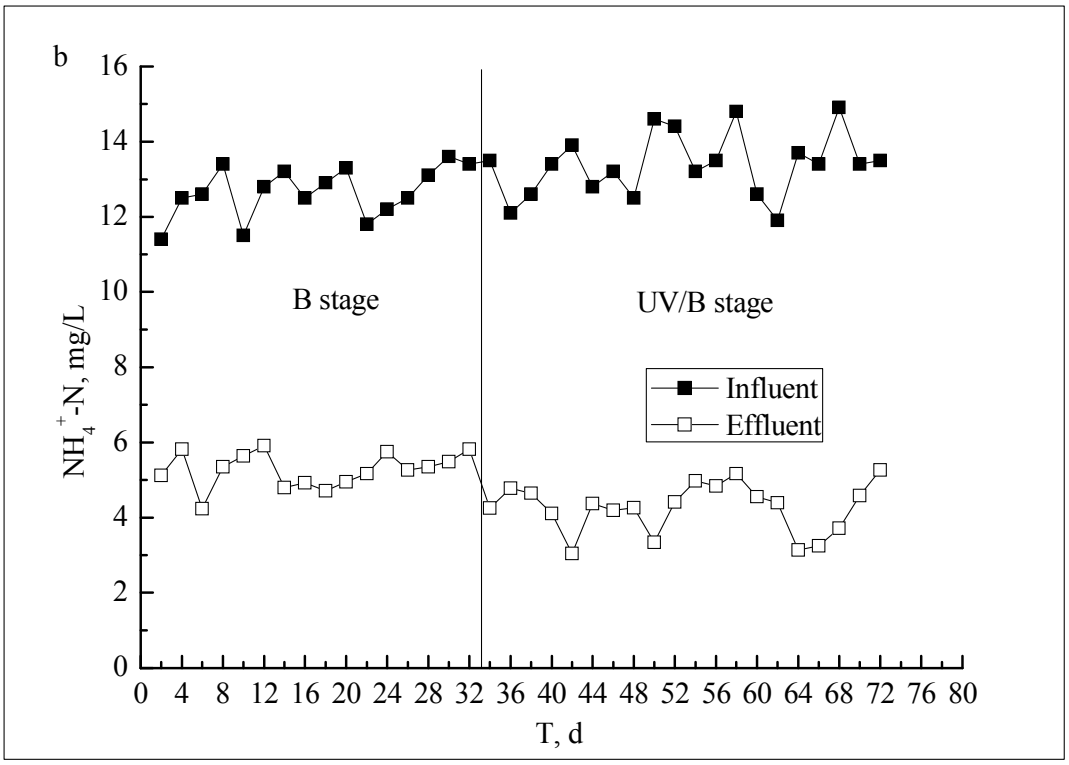
b
NH4+-N, mg/L
T, d
B stage
UV/B stage
Influent
Effluent

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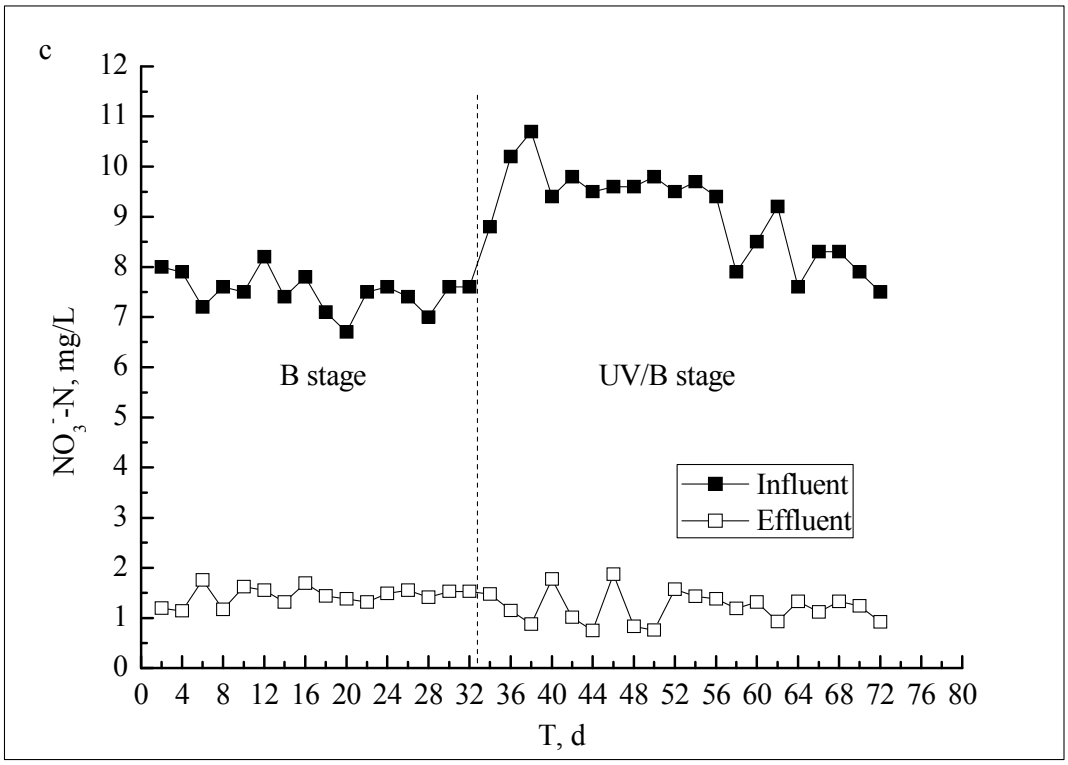


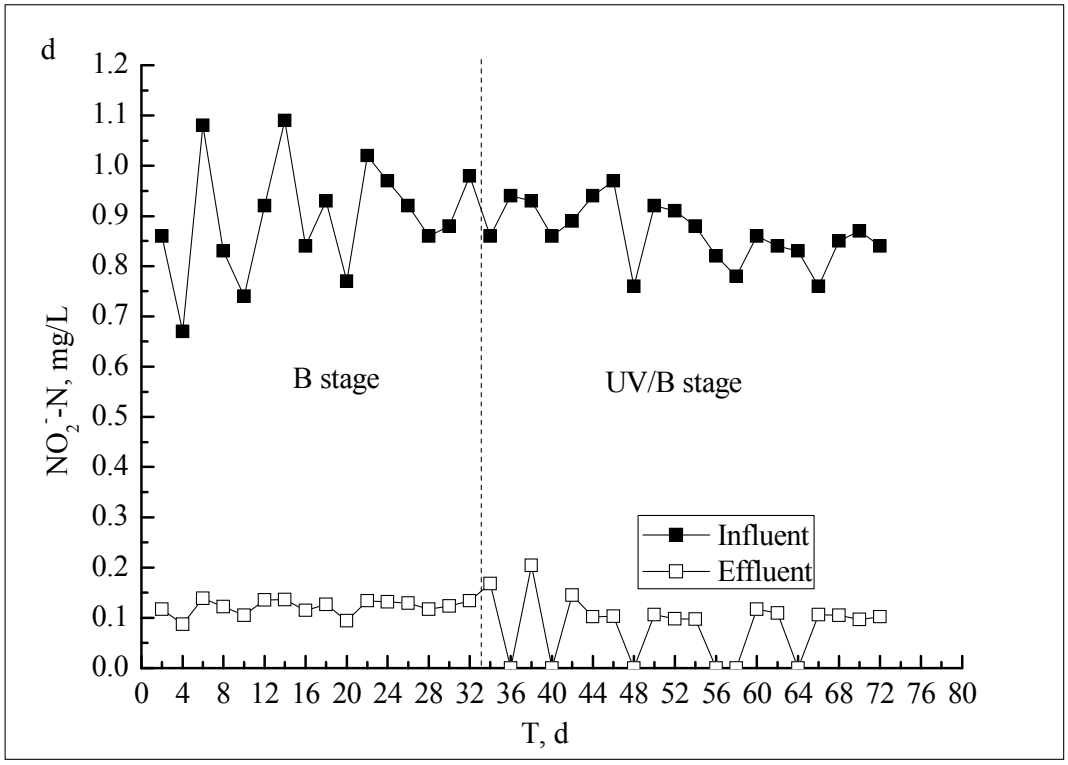


Figure 2. Removal efficiency of TN, NH_4^+-N, NO_3^--N, NO_2^--N

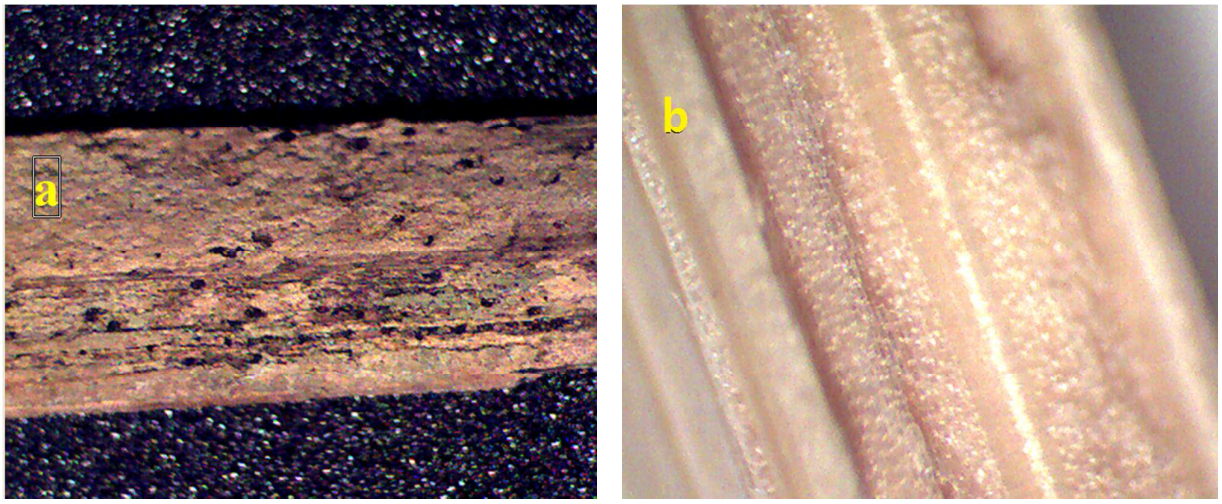


Figure 3 Changes in filamentous bamboo before (b, 20×) and after usage (a, 8×)

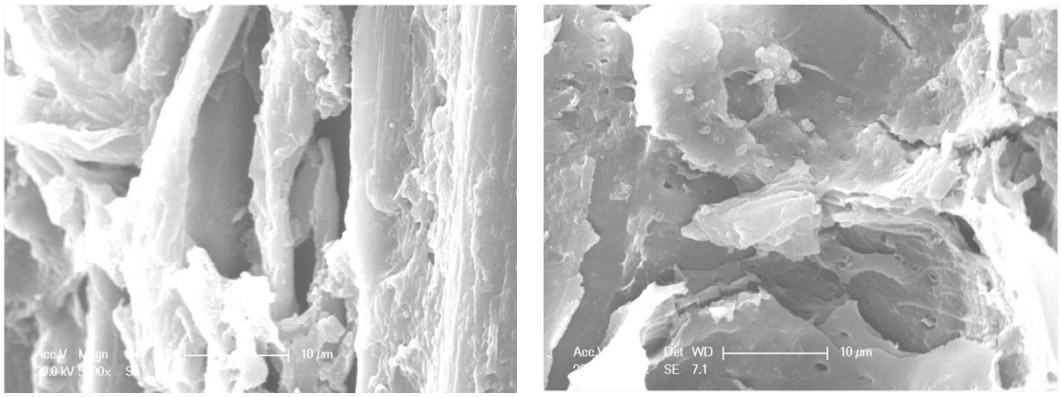


Figure 4 Changes in the surface micro-structures of bamboo

(Note: figure (a) shows the surface micro-structures of raw bamboo, figure (b) shows the surface micro-structures of bamboo used in the experiments)

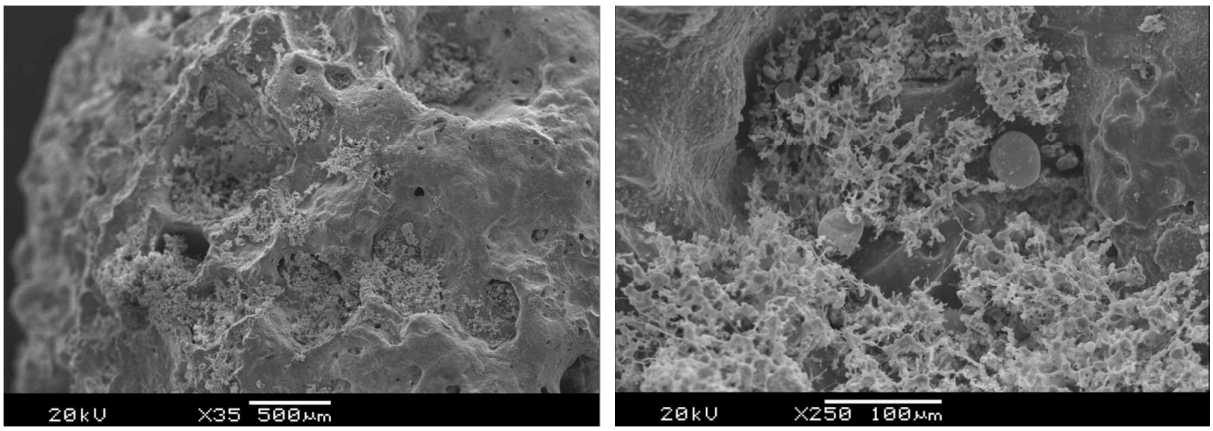
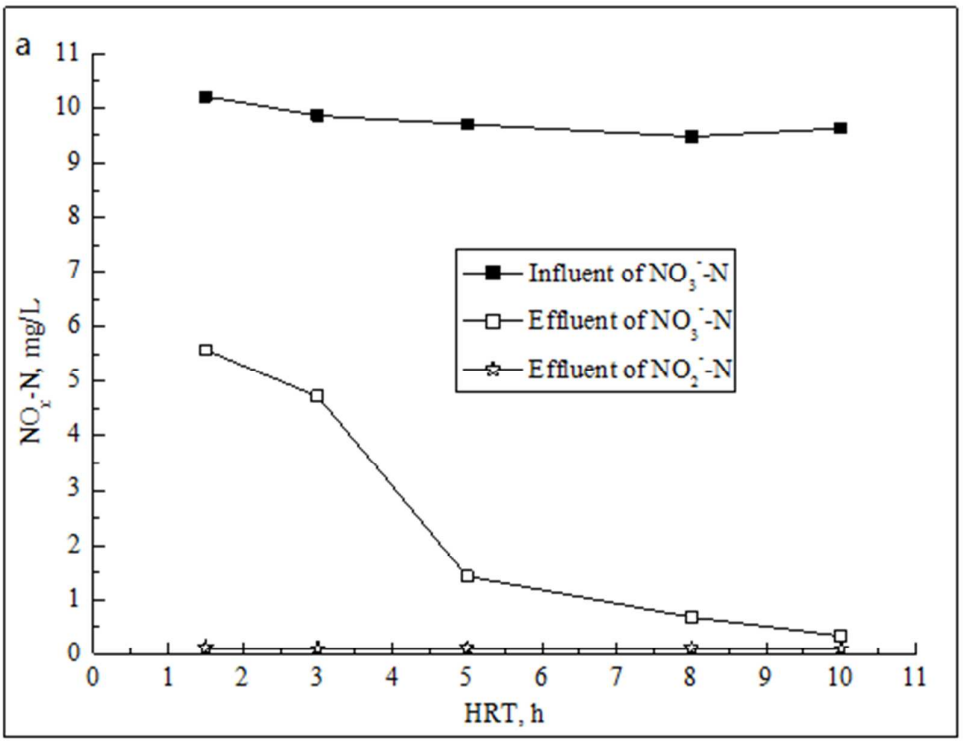
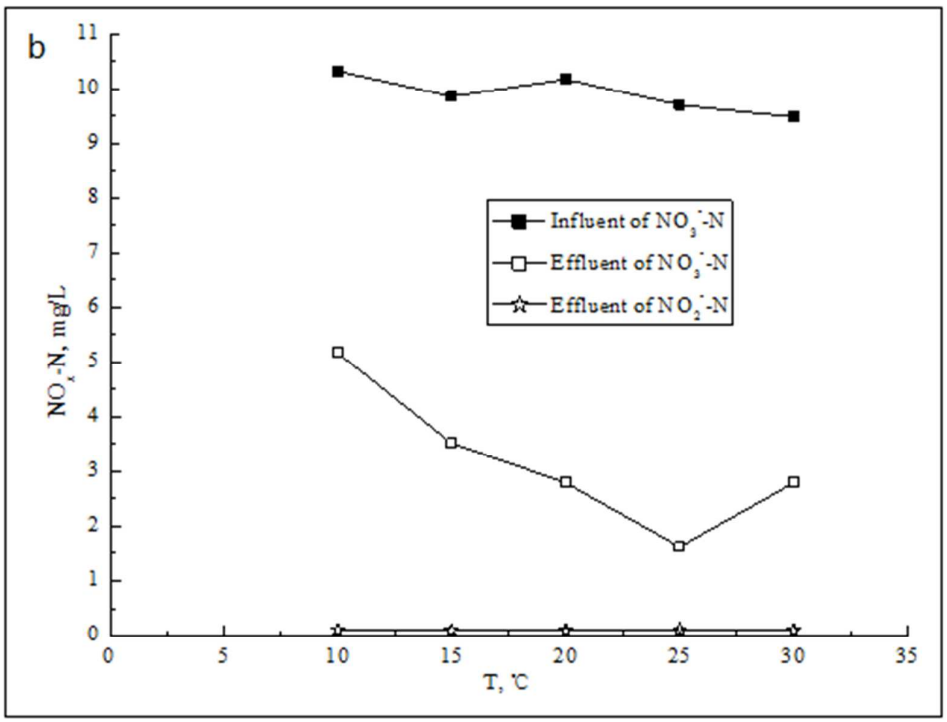


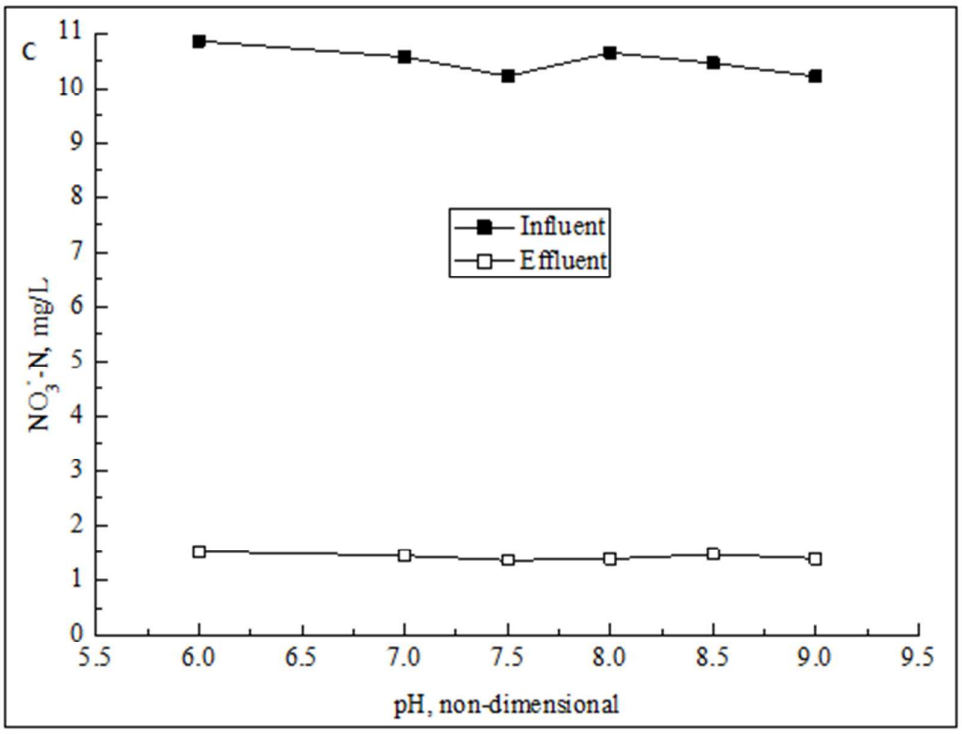
Figure 5. The biofilm adhered on the filamentous bamboos


a
Influent of NO_3^--N
Effluent of NO_3^--N
Effluent of NO_2^--N
NO_x-N, mg/L
HRT, h

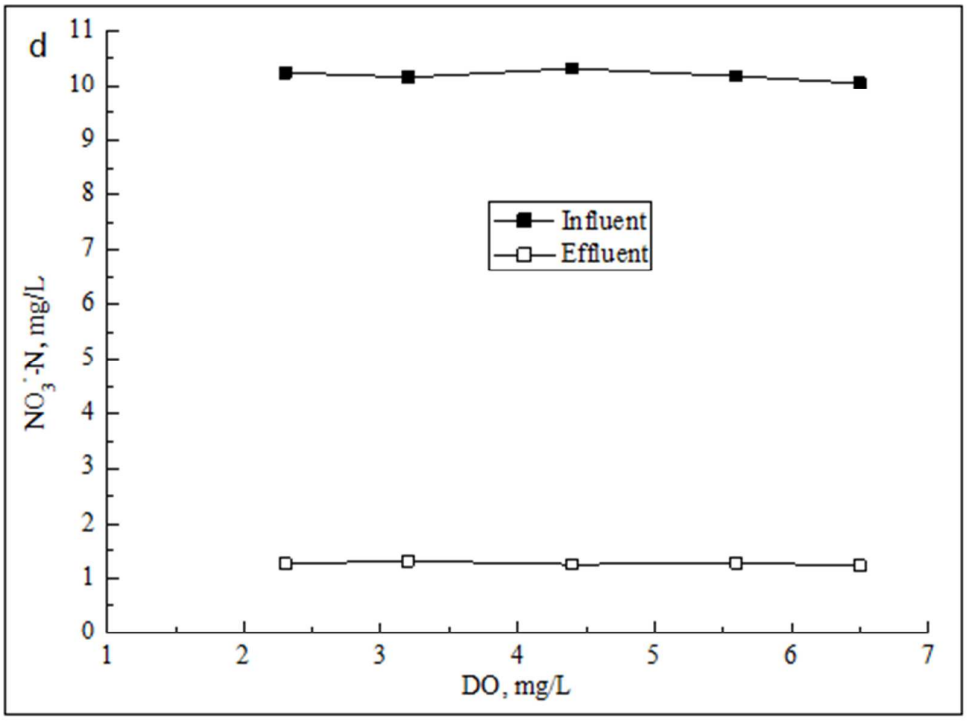
410


b
Influent of NO_3^--N
Effluent of NO_3^--N
Effluent of NO_2^--N
NO_x-N, mg/L
T, ℃

411



412



413

414 Figure 6. The effect of pH, DO, water temperature, and influent NO_3^--N concentration on NO_3^--N

415 removal