Development of Carbonized Textile as Immobilizing Carrier for Wastewater Treatment

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Abstract: Carbonized material has a biological affinity and it is effective for the immobilization of microbes. In this study, in order to enhance the biological nitrogen removal in the wastewater treatment, the carbonized textile was applied as the immobilizing carrier of nitrifying bacteria.

Cotton and rayon were used as the sample textile and they were carbonized under various conditions. From the test of the mechanical strength of carbonized textiles, the rayon carbonized at the relatively low temperature was found to be good for the immobilizing carrier. The performances of microbe adhesion and nitrogen decomposition were examined by using the nitrifying bacteria which were acclimated with the model wastewater. From this examination, it was recognized that the carbonization of textile was effective for the immobilization of nitrifying bacteria. The wastewater treatment was performed by using a small reactor (200×100×350 mm). Ammonia-rich model wastewater was used and the limited aeration method (aerobic condition = 22 h, anaerobic condition = 2 h, mean retention time of wastewater = 3 d) was adopted. The ammonia removal performance was higher for the reactor installed with the carbonized textile than for that without it. From this result, it was found that the immobilizing carrier of carbonized textile was effective for the biological nitrogen removal.

For the continuous treatment of wastewater, the flow characteristics of a large reactor (rectangular airlift bubble column, 320×80×2000 mm) were measured. Based on the energy balance, the liquid circulation flow rate was analyzed, and the distribution of dissolved oxygen in the column was estimated. From these results, the optimum conditions for the reactor installed with the carbonized textile were examined.

Keywords: Carbonized Textile, Wastewater Treatment, Immobilizing Carrier

1. INTRODUCTION

Recently, the eutrophication in the closed-water system such as ponds and lakes has been a serious problem. Among various components in the wastewater, nitrogen is one of the main causes of the eutrophication. Accordingly, it is strongly desired that the development of the process for the high-grade treatment of wastewater including high-content nitrogen. As the nitrogen removal, the biological method, ion exchange, reverse osmosis and chemical method are used. The method of biological removal of nitrogen is proper for the organic wastewater from the food processing process etc.

The biological removal mainly consists of three steps such as ammonification, nitrification and denitrification. Two former steps proceed under the aerobic condition but the latter one dose under the anaerobic condition [1-4]. Among these steps, the nitrification step becomes a rate-determining one, since the nitrifying bacteria have a relatively low growth rate and are easily washed out from the reactor. So, the immobilization of the nitrifying bacteria in the reactor is considered to enhance the biological removal of nitrogen.

It has been reported that carbonized material has a biological affinity and is effective for the immobilization of microbes [5, 6]. Carbon fiber was well used in the laboratory experiment. However, since the production cost of carbon fiber is very high and the mechanical strength is very weak, the carbon fiber is not adopted in the actual treatment of wastewater.

In this study, it was considered to apply the carbonized textile as the immobilizing carrier of nitrifying bacteria. Cotton and rayon were used as the sample textile, and they were carbonized under various conditions. The performances of microbe adhesion and nitrogen decomposition were examined by using the nitrifying bacteria. In order to develop the compact reactor for the biological nitrogen removal, the rectangular airlift bubble reactor, which have been studied by the authors [7-10], was used, and the flow characteristics of the column were measured. Based on the energy balance, the liquid circulation flow rate was analyzed and the distribution of dissolved oxygen in the column was estimated. From these results, the optimum conditions for the reactor installed with the carbonized textile were examined.

2. EXPERIMENTAL

2.1. Performances of microbe adhesion and nitrification

Cotton and rayon were used as the sample textile. The textile was carbonized at an electric furnace. The carbonization temperature and the holding time at high temperature were combined. The size of carbonized textile was approximately 60×80 mm. The carbonized textile was immersed in a shaker (2 Hz) including activated sludge and the shaking time was changed. The weight of activated sludge adhered to the carbonized textile was measured.

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In the test of the nitrification performance of the carbonized textile, the activated sludge acclimated with ammonia-rich model wastewater (the composition is listed in Table 1) and a glass vessel (500 mL) installed with a gas sparger were used. The carbonized textile immobilized with sludge was immersed in the vessel and ammonia aqueous solution with the concentration of 90 mg/L was poured. The ammonia concentration and pH were measured during three days. In order to keep the aerobic condition, the liquid was aerated during 30 min every 12 h.

The wastewater treatment was performed by using two small reactors (200×100×350 mm). The carbonized textile was set in one reactor and another one was a control. The ammonia-rich model wastewater was used for the multiplication of nitrifying bacteria, and then it was changed to the normal model wastewater listed in Table 1. In this study, the enhance in nitrogen removal from the wastewater including high concentration nitrogen was aimed, and the nitrogen content of normal wastewater was prepared to be a little higher than the standard wastewater. The composition was as follows; BOD:TN:TP = 100:10:1. The limited aeration method (aerobic condition = 22 h, anaerobic condition = 2 h) was adopted. The mean retention time of wastewater was 3 d. The changes in ammonia removal and pH with time were measured during about 40 days.

### Table 1 Composition of model wastewater.

<table>
<thead>
<tr>
<th>Content</th>
<th>Ammonia-rich</th>
<th>Normal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glucose</td>
<td>—</td>
<td>800</td>
</tr>
<tr>
<td>NH₄Cl</td>
<td>800</td>
<td>500</td>
</tr>
<tr>
<td>K₂HPO₄</td>
<td>100</td>
<td>50</td>
</tr>
<tr>
<td>NaCl</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>KCl</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>CaCl₂·2H₂O</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>MgSO₄·7H₂O</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>NaHCO₃</td>
<td>300</td>
<td>210</td>
</tr>
</tbody>
</table>

2.2. Flow characteristics of rectangular airlift bubble column

Figure 1 shows the outline of experimental apparatus for the measurement of flow characteristics. The bubble column was made from transparent vinyl chloride resin, and the width, length and height were 80, 320 and 2000 mm, respectively. A partitioning plate of 1500 mm in height was vertically inserted into the column and the distance from the column bottom to the lower end of partitioning plate was fixed to be 100 mm. The width of riser was 80 mm. The gas sparger was a perforated plate with 4 holes of 2 mm in diameter and was placed at the bottom of riser. As shown in Fig. 1, the aluminum frames attached with carbonized textile were inserted in the column. Ten and five sheets of carbonized textile were attached to the frames in the riser and downcomer, respectively so that the carrier immobilized with nitrifying bacteria might be located in the aerobic region. In this type of airlift bubble column, both the riser and the upper half region of the downcomer are aerobic [7-9]. The numbers of frames in the riser and downcomer were changed till two and four, respectively.

As the gas and liquid phases, air and water were used for the measurement of the flow characteristics. The upper clearance length from the upper end of partitioning plate to the liquid surface at aeration and the superficial
gas velocity based on the cross-sectional area of column were changed. The gas holdup in the riser was obtained from the difference in static pressure. The liquid velocity was measured at the zone below the lower end of partitioning plate by using a three-dimensional velocity meter.

3. RESULTS AND DISCUSSION

3.1. Performances of microbe adhesion and nitrification

Cotton and rayon were carbonized at different temperatures. As the carbonation temperature was higher, the mechanical strength became weaker. The holding time had no significant influence on the property of carbonized textile. Accordingly, in this study, the textile carbonized at 350°C was used.

Figure 2 shows the change in weight of sludge adhered to the carbonized textile with the shaking time for different immobilizing carriers. The ordinate is the sludge weight per unit area of the carbonized textile. Except the cotton not carbonized the weight of adhered sludge increases with the shaking time and becomes almost constant beyond 3 h. For cotton, the carbonation is found to be effective for the immobilization, because of the reduction in hydrophobic property. Although the carbonized cotton increased the adhered amount of sludge, the mechanical strength was much weakened. So, in the wastewater treatment experiment, the carbonized rayon was used.

Figure 3 shows the change in ammonia removal and pH with time for the small reactor. Till 11th day, the ammonia-rich model wastewater was used for the multiplication of nitrifying bacteria. For both the reactors, the ammonia removal decreases until 6th day but it increases from 7th day. The pH decreases largely until 6th day and gradually from 7th day. From these, the multiplication of nitrifying bacteria is recognized. On 11th day, the model wastewater was changed from ammonia-rich one to the normal one. The ammonia removal increases but pH decreases till 20th day. This is because the multiplication of nitrifying bacteria continued. Beyond 20th day, the ammonia removal increases but pH decreases. Beyond 25th day, both the values become almost constant. Over a whole experiment, for the reactor with immobilizing carrier, the ammonia removal is a little higher and pH is lower than for the reactor without immobilizing carrier. From this, the immobilizing carrier is found to be effective for the enhancement in nitrification. The small difference with and without immobilizing carrier is considered due to the masking effect of free bacteria in the reactor. In addition, it is another reason that the contact between wastewater and immobilizing carrier was not sufficient in the small reactor. However, in the large reactor utilizing the liquid circulation flow by an airlift, the immobilizing carrier is expected to enhance the nitrification.

3.2. Flow characteristics of rectangular airlift bubble column

The large scale rectangular airlift bubble column [7-10] was used and the flow characteristics were measured. The data were analyzed on the basis of the energy balance. By using these characteristics, the distribution of dissolved oxygen and the fraction of anaerobic region in the column were estimated to design the reactor installed with the carbonized textile.
Figure 5 shows the effect of gas velocity on the gas holdup and liquid velocity in the riser. The parameter is the numbers of frames for the immobilizing carrier in the riser and downcomer, and the gas velocity is based on the cross-sectional area of column. Both the gas holdup and liquid velocity in the riser become higher with increasing gas velocity. As the number of frame increases, the liquid velocity becomes lower because the flow resistance is larger. As a result, the drift force of liquid becomes smaller and the gas holdup becomes higher.

Figure 6 shows the effect of upper clearance on the gas holdup and liquid velocity in the riser. The upper clearance is the length from the upper end of partitioning plate to the liquid surface at aeration. With increasing upper clearance the liquid velocity increases and becomes unchanged beyond the upper clearance of 0.05 m. This is because the flow resistance decreases with increasing upper clearance and it becomes constant at the enough long clearance. The gas holdup shows the opposite tendency, because of the decrement in drift force of liquid.

In Figure 7, the true gas velocity is plotted against the sum of superficial gas and liquid velocities in the riser [11]. All the data roughly ride on a line, in spite of the frame numbers. From this plot, the following equation for the gas holdup in the riser was obtained;

\[ \frac{u_G}{\varepsilon_R} = 0.3 + 55 \left( u_{GR} + u_{LR} \right) \]  

(1)

In the right hand of Eq. (1), the first term corresponds to the rising velocity of bubble swarm and the value (0.3 m/s) is reasonable. However, the coefficient of the second term meaning the drift flux of gas and liquid flows (55) is extremely large. This is considered because the liquid flow in the rectangular column was much fluctuated.

The liquid flow in the column was analyzed on the basis of the following energy balance;

\[ g L_P \varepsilon_R = n_R K_R + n_D K_D + \left( \frac{S_R}{S_D} \right)^2 u_{LR}^2 / 2 \]  

(2)

where \( K_T \) and \( K_R \) are the loss coefficients at the upper and lower clearances of the column, and \( K_R \) and \( K_D \) are the loss coefficients of the frames attached with carbonized textile in the riser and downcomer. In Eq. (2), the friction losses in the riser and downcomer were neglected, and the gas holdup in the downcomer was assumed to be zero.
The sum of all loss coefficients was calculated from Eq. (2). The results are shown in Figure 8. From this, it is found that the upper clearance is a controlling factor to the liquid flow in this column [10]. The loss coefficients are expressed as follows;

\[ K_T = 9000 \exp(-26.4 L_T) \]

\[ K_B = 500 \]

\[ K_R = 80 \]

\[ K_D = 50 \]

By using Eqs. (1) – (3), the gas holdup and liquid velocity in the riser were calculated. Figures 9 and 10 show the comparisons of gas holdup and liquid velocity in the riser between the measured data and calculated results.

The calculated results agree with the measured data within the errors of ±20 %, and the validity of above analysis is recognized.

To make the aerobic and anaerobic regions in the column, the distribution of dissolved oxygen in the column should be designed by controlling the liquid flow [10]. The dissolved oxygen distribution in the column can be estimated on the basis of the dissolved oxygen balance by using the calculated liquid velocity. Here, the followings were assumed.
The balances of dissolved oxygen in three parts are expressed as follows:

\[ u_{lR} \frac{dC}{dz} = k_{lA} (C^* - C) - \gamma X \]

\[ u_{lD} \frac{dC}{dz} = -\gamma X \]  

where \( C \) and \( C^* \) are the dissolved oxygen concentrations in the column and at the saturated state. \( z \) is the distance from the riser bottom along the liquid flow and \( \tau_T \) is the residence time in the upper part. \( k_{lA} \) is the liquid-phase volumetric mass transfer coefficient, and the following empirical equation [8] was used.

\[ k_{lA} = 2.5 \sigma_r \]  

\( \gamma \) is the specific oxygen consumption rate of microbe and was constant to be 0.035 kg-Oxygen/(kg-MLSS•s). \( X \) is MLSS in the column (2 kg/m³). When the immobilizing carrier was inserted, MLSS adhered to the carbonized textile was 0.016 kg-MLSS/m² (see Figure 2).

Figure 12 shows the distribution of dissolved oxygen in the column. The abscissa is the fraction of volume at \( z \) in the total volume. The concentration of dissolved oxygen increases in the riser and upper part. In the downcomer, it decreases linearly and becomes zero. From the position where the dissolved oxygen becomes zero to the bottom of downcomer, the anaerobic region exists.

When the frames of immobilizing carrier are inserted in the column, it is found that the dissolved oxygen is reduced in a whole column and the anaerobic region is larger.

Table 2 shows the volume fraction of anaerobic region for different numbers of frames for the immobilizing carrier. With increasing number of frame, the anaerobic region becomes larger. For the biological nitrogen removal, it is very important to make the volume fraction of anaerobic region about 50% [7-9]. Even when several frames of immobilizing carrier were inserted in the riser and downcomer, the anaerobic region fraction is expected to be about 50%.

For the rectangular bubble column used in this study, the main parameters are the gas velocity, upper clearance and number of frame for the immobilizing carrier. Under the various combinations of these parameters, the distribution of dissolved oxygen and the fraction of anaerobic region in the column are estimated by above the energy and dissolved oxygen balances, and the optimum condition can be designed.

4. CONCLUSIONS
Carbonized textile was applied as the immobilizing carrier for the biological nitrogen removal and the performances of microbe adhesion and nitrification were examined. The frames attached with the carbonized textile were inserted into the rectangular airlift bubble column, and the flow characteristics were analyzed. The following facts were clarified;

1) The rayon carbonized at the relatively low temperature is good for the immobilization of nitrifying bacteria, and enhances the ammonia removal.
2) With increasing number of the frames attached with the carbonized textile, the liquid velocity decreases but the gas holdup in riser increases. These flow characteristics are reproduced by using the empirical equations and the energy balance.
3) From the dissolved oxygen balance, the distribution of dissolved oxygen and the volume fraction of anaerobic region in the column are estimated. When several frames of immobilizing carrier were inserted in the riser and downcomer, about 50% of anaerobic region fraction is expected.

5. NOMENCLATURE
\( A \) = surface area of textile [m²]
\( C \) = concentration of dissolved oxygen [kg/m³]
\( C_{NH4-N} \) = ammonia concentration [mg/L]
\[ k_{L}a = \text{volumetric mass transfer coefficient} \quad [1/s] \]
\[ K_b = \text{loss coefficient at lower clearance} \quad [-] \]
\[ K_s = \text{loss coefficient of frame in downcomer} \quad [-] \]
\[ K_r = \text{loss coefficient of frame in riser} \quad [-] \]
\[ K_t = \text{loss coefficient at upper clearance} \quad [-] \]
\[ L_p = \text{length of partitioning plate} \quad [m] \]
\[ L_t = \text{length of upper clearance} \quad [m] \]
\[ MLSS = \text{mixed liquor suspended solid} \quad [kg/m}^3] \]
\[ n_D = \text{number of frame in downcomer} \quad [-] \]
\[ n_R = \text{number of frame in riser} \quad [-] \]
\[ S_D = \text{cross-sectional area of downcomer} \quad [m}^2] \]
\[ S_R = \text{cross-sectional area of riser} \quad [m}^2] \]
\[ t = \text{time} \quad [d] \]
\[ u_G = \text{superficial gas velocity based on cross-sectional area of column} \quad [m/s] \]
\[ u_{GR} = \text{superficial gas velocity based on cross-sectional area of riser} \quad [m/s] \]
\[ u_{LD} = \text{superficial liquid velocity based on cross-sectional area of downcomer} \quad [m/s] \]
\[ u_{LR} = \text{superficial liquid velocity based on cross-sectional area of riser} \quad [m/s] \]
\[ V = \text{total volume of column} \quad [m}^3] \]
\[ V_{AN} = \text{volume of anaerobic region} \quad [m}^3] \]
\[ V_z = \text{volume of region from riser bottom to } z \quad [m}^3] \]
\[ X = \text{concentration of microbe} \quad [kg/m}^3] \]
\[ \Delta W = \text{adhered weight of sludge} \quad [g] \]
\[ z = \text{distance from riser bottom along liquid flow} \quad [m] \]
\[ \varepsilon_R = \text{gas holdup in riser} \quad [-] \]
\[ \gamma = \text{specific oxygen consumption rate of microbe} \quad [\text{kg-Oxygen/(kg-MLSS•s)}] \]
\[ \eta_{NH_4-N} = \text{ammonia removal} \quad [%] \]
\[ \theta = \text{shaking time} \quad [h] \]
\[ \tau_T = \text{residence time in upper part} \quad [h] \]

6. REFERENCES