EFFECTS OF GEOMETRICAL VARIABLES OF HELICAL RIBBON IMPELLERS ON MIXING OF HIGHLY VISCOUS NEWTONIAN LIQUIDS

Koji TAKAHASHI, Minoru SASAKI, Kunio ARAI and Shozaburo SAITO
Department of Chemical Engineering, Tohoku University, Sendai 980

Mixing patterns in agitated vessels equipped with various types of helical ribbon impellers are observed by using capsules of liquid crystal as a tracer. As a result, the optimum geometrical variables for the mixing process are provided and the results explained by considering the relation between the mixing pattern and impeller geometry. A mathematical model is developed and applied in order to correlate the measure mixing times.

Introduction

A helical ribbon impeller is one of the most common types of close-clearance impellers used for the mixing of highly viscous liquids. In a helical ribbon agitator, mixing proceeds mainly in the clearance between blades and wall where the liquid is subjected to very high shearing, but the overall mixing appears to be controlled by the flow pattern which permits the renewal of the liquid in the clearance and distributes the liquid flowing out of the clearance region to the low-shear region of the vessel. The flow pattern may be easily affected by small changes of impeller geometrical variables. Several investigators have reported the effects of geometrical variables on mixing time, but the relation between mixing pattern and geometrical variables has not been thoroughly established because conventional methods for the observation of mixing patterns are not suitable for the detection of a poorly mixed zone.

In this work, providing a suitable method for observation of mixing patterns with the use of capsules of liquid crystal as a tracer, observation of mixing patterns and measurement of mixing times for helical ribbon impellers of different geometrical variables were carried out for Newtonian liquids under laminar-flow conditions. The optimum geometrical conditions for the mixing process have been established. On the basis of the observation of mixing patterns, the mixing time has been correlated with geometrical and operational variables.

1. Experimental

In the literature, the coloration method and the decoloration method are commonly used for observation of mixing patterns. However, these methods are unsuitable for observation of a three-dimensional mixing process in an agitated vessel, because of the overlap of the colored liquids in the vessel.

In this work, mixing patterns were observed by the method using a tracer liquid containing small capsules of liquid crystal described in a previous work. An aqueous solution of corn syrup having a viscosity of about 10 poise and density of 1.37 g/cc was used as a test liquid. Corn syrup of the same viscosity and density as the test liquid containing capsules of liquid crystal was used as a tracer liquid.

The experimental apparatus shown in Fig. 1 is the same as in the previous paper. The geometrical configuration of the helical ribbon impeller is shown in Fig. 2 and details of impeller geometries are summarized in Table 1. The vessel was a transparent glass cylinder with a flat bottom and a flat lid. The impeller blades were made of transparent acrylic resin so that observation of the mixing pattern might not be impaired.

The agitated vessel was filled with the test liquid. A small amount of tracer liquid was put into the top of the vessel near the shaft. The mixing of tracer with mother liquid was visualized in the vertical plane.
2. Effects of Impeller Geometries on Mixing Patterns

Figure 3 shows a cross-section view of the primary circulation flow in the illuminated vertical plane. Figures 4 and 6 to 8 show photographs of mixing patterns in this cross section in agitators with different geometrical variables. In each figure, \( N_r \) represents the number of impeller revolutions counted from the start of mixing. The patterns of the primary circulation flows are almost the same as mentioned by previous investigators\(^1\) and the mixing patterns are approximately similar in spite of the different impeller geometries. Namely, as observed by Carreau \textit{et al.}\(^2\), shear mixing proceeds mainly in the clearance between blades and vessel wall, and the well-mixed liquid flowing out of the clearance is easily distributed to the circulation path between the blades by the primary flow. This well-mixed zone is denoted by \( A \) in Fig. 3. The tracer in this region, however, can permeate only gradually into the inner circulation flow situated in the region inside the inner edge of the blade, denoted by \( B \) in Fig. 3, and the exchange flow between regions \( A \) and \( B \) is commonly observed to be a controlling step in the mixing process.

The effect of each geometrical variable of a helical ribbon impeller on mixing time is discussed in detail in the following sections.

2.1 Effect of clearance between blades and vessel wall

Figure 4 shows the mixing patterns in agitators with different clearances between blades and wall. It can be seen from the photographs at \( N_r = 42 \), that the agitator with medium-clearance impeller is superior to the others for mixing. With the impeller of narrowest clearance, an unmixed zone can still be observed in region \( B \). On the other hand, with the impeller of widest clearance, stagnant zones are detected at both top and bottom corners in region \( A \).

These phenomena may be explained by the difference in the effect of secondary flow on primary flow. As shown in Fig. 5, the same kind of secondary flow as observed by Peters and Smith in an anchor agitator\(^18\),\(^19\), must also be created in a helical ribbon agitator, due to the change of centrifugal force in the axial direction. This secondary flow will deform the primary flow to enhance the exchange flow between
regions A and B. On the other hand, the secondary flow will be depressed by the side wall, an effect which decreases as the clearance becomes wider. With the impeller of narrowest clearance, the flow pattern is not so much affected by the secondary flow and thus the liquid in region A exchanges only gradually with the liquid in region B. With the medium-clearance impeller, moderate deformation of the flow stream in both the top and the bottom regions seems to accelerate the exchange flow between regions A and B. With the impeller of widest clearance, stable secondary flow dominates in the top and bottom corners and closed loops develop.

2.2 Effect of impeller pitch

Photographs of mixing patterns in agitators of different pitches are shown in Fig. 6. As shown in the photographs at \( N_r = 42 \), mixing time for the impeller of \( s/D = 0.90 \) is shortest. The photographs at \( N_r = 9 \) show that the axial velocity becomes greater as the \( s/D \) ratio increases in this experimental range, as expected from the circulation flow models proposed by Bourne and Butler\(^1\), and Carreau et al.\(^2\). The photographs at \( N_r = 14 \) show that the deformation of the tracer liquid at the clearance for small \( s/D \) is greater than that for large \( s/D \). Namely, the shear rate, which affects the mixing in the clearance, becomes smaller and the circulation flow rate, which permits the renewal of the liquid in the clearance, greater with an increase of \( s/D \) in this experimental range. These two mutually competing effects on mixing lead to an optimum in pitch size.

2.3 Effect of blade width

Photographs of mixing patterns in agitators of different blade width are shown in Fig. 7. The photographs at \( N_r = 42 \) show that the impeller of medium blade width is superior to the others. As shown in the photographs at \( N_r = 9 \), the circulation flow becomes stronger as the blade widens. But if blade width exceeds a certain limit, the circulation flow rate decreases because the area of the upward flow becomes too great compared with that of the downward flow. Then the circulation flow rate, which permits the distribution of liquid in the clear-
2.4 Effect of number of blades

As shown in Fig. 8, even after 500 impeller revolutions, a poorly mixed zone was observed inside the blade for all single helical ribbon impellers. In single helical ribbon agitators, the secondary flow is weak, due to the long periodic passage of the blade, and thus the overall flow is controlled by the primary flow. Therefore, the double helical ribbon impeller is more suitable than the single one for mixing.

3. Relation between Mixing Time and Impeller Geometries

It is suggested that the relation between the mixing time, $t_m$, and the rotational speed of the impeller, $N$, can be written as Eq. (1) in the laminar flow region.

$$Nt_m = C_1$$

where $C_1$ is a geometrical constant.

Mixing times measured by the liquid crystal method for 14 different impellers are summarized in Table 2. The relation between $C_1$ and each of three geometrical ratios, $c/D$, $s/D$ and $w/D$, is shown in Figs. 9, 10 and 11 respectively. $C_1$ may vary according to measuring method and definition of mixing time chosen. To compare the sensitivities of the methods for measuring mixing time, the results of the decoloration method are also given in Figs. 9 and 10. The shaded area represents the range where unmixed zones were observed even after 500 impeller revolutions. Figures 9 and 10 show that the sensitivity of the liquid crystal method is superior to that of the decoloration method.

As mentioned in section 2, there are optimum values for the three geometrical ratios in the mixing process. The optimum values of $c/D$, $s/D$ and $w/D$ obtained in this work are 0.06, 0.90 and 0.15 respectively. These values differ from those suggested by Nagata et al. by the decoloration method, i.e. 0.025, 0.95 and 0.10.

3.1 Development of a model

Shear mixing of the liquid proceeds mainly in the clearance between blades and vessel wall where the shear rate is high. But the renewal flow rate of the liquid in the clearance, which is nearly equal to the circulation flow rate, and the exchange flow rate between regions $A$ and $B$, due to the combined effect of the primary and secondary flows, also play important roles in the uniformity of concentration in a vessel, as suggested in section 2. On the basis of this consideration, mixing time is expressed as a function of magnitude of shear rate in the clearance, $M$, circulation flow rate, $Q_c$, and exchange flow rate, $Q_e$. That is,
### Table 2  Correlations of circulation flow rate and mixing time

<table>
<thead>
<tr>
<th>Geometry No.</th>
<th>((Q_i/Ne)^{exp.})</th>
<th>((Q_i/Ne)^{cal.})</th>
<th>((N_{m2})^{exp.})</th>
<th>((N_{m2})^{cal.})</th>
<th>((N_{pRe})^{exp.})</th>
</tr>
</thead>
<tbody>
<tr>
<td>SH1</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>203.1</td>
</tr>
<tr>
<td>SH2</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>141.2</td>
</tr>
<tr>
<td>SH3</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>126.7</td>
</tr>
<tr>
<td>DH1</td>
<td>0.108</td>
<td>0.110</td>
<td>239</td>
<td>236</td>
<td>406.2</td>
</tr>
<tr>
<td>DH2</td>
<td>0.112</td>
<td>0.119</td>
<td>53.8</td>
<td>53.1</td>
<td>288.9</td>
</tr>
<tr>
<td>DH3</td>
<td>0.123</td>
<td>0.117</td>
<td>—</td>
<td>592</td>
<td>256.6</td>
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<tr>
<td>DH4</td>
<td>0.0732</td>
<td>0.0867</td>
<td>56.4</td>
<td>60.0</td>
<td>352.6</td>
</tr>
<tr>
<td>DH5</td>
<td>0.0708</td>
<td>0.0678</td>
<td>68.5</td>
<td>79.0</td>
<td>392.7</td>
</tr>
<tr>
<td>DH6</td>
<td>0.113</td>
<td>0.136</td>
<td>245</td>
<td>69.4</td>
<td>259.4</td>
</tr>
<tr>
<td>DH7</td>
<td>0.120</td>
<td>0.132</td>
<td>—</td>
<td>82.4</td>
<td>222.6</td>
</tr>
<tr>
<td>DH8</td>
<td>0.104</td>
<td>0.0795</td>
<td>106</td>
<td>122</td>
<td>273.6</td>
</tr>
<tr>
<td>DH9</td>
<td>0.150</td>
<td>0.143</td>
<td>42.3</td>
<td>43.5</td>
<td>315.7</td>
</tr>
<tr>
<td>DH10</td>
<td>0.185</td>
<td>0.155</td>
<td>33.0</td>
<td>33.9</td>
<td>334.6</td>
</tr>
<tr>
<td>DH11</td>
<td>0.137</td>
<td>0.160</td>
<td>56.2</td>
<td>33.4</td>
<td>365.6</td>
</tr>
</tbody>
</table>

\[ \frac{1}{N_{m2}} = f(M, Q_i, N_e) \quad (2) \]

1) Magnitude of shear rate in the clearance Since power is consumed mainly in the clearance between blades and vessel wall, magnitude of shear rate in this region is assumed to be represented by the following equation.

\[ M = \frac{1}{N} \sqrt{\frac{P_{2}}{\mu}} = \sqrt{N_{pRe}} \left( \frac{d}{D} \right)^{4/3} \pi \quad (3) \]

\( N_{pRe} \) can be calculated from the power correlation derived by the authors. The calculated values are shown in Table 2.

2) Circulation flow rate In this work, the time required for the tracer liquid to complete one circuit, \( t_i \), was measured. Since \( t_i \) is approximately proportional to the average circulation time, the circulation flow rate is represented by:

\[ Q_i = V/t_i \quad (4) \]

where \( V \) is the total liquid volume in a vessel. The values of circulation number, \( Q_i/(Ne^2) \), are summarized in Table 2.

To correlate the circulation flow rate, a velocity profile model was proposed. In the vessel, the pumping action of the blade creates a drag flow as shown in Fig. 12(a). This flow is restricted at the top and bottom walls of the vessel and an axial pressure gradient builds up. The effect of this gradient is introduced as a pressure flow, as shown in Fig. 12(b). The volume rate of these two flows should be the same while the direction of the two flows are opposite. The resultant of these two flows is observed. In this paper, the velocity profile of the drag flow is designated \( v_{zd} \), and that of the pressure flow \( v_{zp} \). Thus, the velocity profile of the resultant flow, \( v_z \), shown in Fig. 12(c) is expressed as follows:

\[ v_z = v_{zd} - v_{zp} \quad (5) \]

The circulation flow rate, \( Q_z \), is given in the form of the following integral:

\[ Q_z = \int_{2\pi r_w}^{2\pi r} 2\pi r v_z dr \quad (6) \]

where \( r_w \) is the radius of the vessel and \( \lambda r_w \) is the radial position where the sign of \( v_z \) changes from negative to positive, as seen in Fig. 12(c).

The velocity profile of the drag flow was defined as follows. By a modification to Bourne's model, the velocity profile is given as a function of the radial position, \( r \), at the blade \((r_{b1} \leq r \leq r_{b2})\). In the regions between shaft and inner edge of blade \((r_{s} \leq r < r_{b1})\), and between outer edge of blade and wall \((r_{b2} < r \leq r_{w})\), the velocity profile is obtained by assuming that these drag flows are flows through an annulus, where one cylinder is moving with the axial velocity of liquid at the blade, and the other cylinder is stationary.
Fig. 13  Correlation of circulation flow rate

\[ r_s \leq r < r_b \]
\[ v_{ex} = 2\pi \frac{N r_{bo}}{r^2} \frac{2\pi r_{so}}{r + (2\pi r_{so})^2} \ln \left( \frac{r}{r_s} \right) \]
\[ r_b \leq r \leq r_{bo} \]
\[ v_{ex} = 2\pi Nr^2 \frac{2\pi r_{bo}^2}{r^2 + (2\pi r_{bo})^2} \ln \left( \frac{r_{bo}}{r_s} \right) \]
\[ r_{bo} < r \leq r_w \]
\[ v_{ex} = 2\pi Nr^2 \frac{2\pi r_{bo}^2}{r^2 + (2\pi r_{bo})^2} \ln \left( \frac{r_w}{r} \right) \]
\[ r_w < r \leq r_o \]
\[ v_{ex} = 2\pi Nr^2 \frac{2\pi r_{bo}^2}{r^2 + (2\pi r_{bo})^2} \ln \left( \frac{r_o}{r_{bo}} \right) \]

On the other hand, \( v_{zp} \) is assumed to be the velocity profile through an annulus. In the calculation of \( v_{zp} \), the pressure gradient was obtained from the fact that the volume rate of the pressure flow is equal to that of the drag flow obtained from the integration of Eq. (7).

Thus, the circulation flow rate could be calculated, but the proportionality of this estimation with the experimental values of \( Q \pm \) was not satisfactory. Based on the velocity profile model proposed, a circulation flow rate was correlated with a multiple non-linear regression of the experimental data.

\[ Q_{z} = \left( \frac{c + 0.179}{D + 0.179} \right)^2 - 295 \left( \frac{w}{D - 0.188} \right)^2 + 6.27 \]

The calculated values from Eq. (8) are summarized in Table 2. The comparison between Eq. (8) and the experimental data is shown in Fig. 13.

3) Exchange flow rate As mentioned in section 2.1, it is found that the exchange flow between regions A and B is determined by the combined effect of the primary and secondary flows. The primary flow rate is nearly equal to the circulation flow rate. Since the volume rate of secondary flow cannot be measured, it must be estimated, even approximately, according to a mathematical model.

In this paper, the secondary flow rate is calculated assuming that a helical ribbon impeller is a vertical cylinder, whose axis is the same as that of the agitated vessel. The outer cylinder of this imaginary agitator has the same diameter as that of the actual vessel. The diameter of the inner cylinder is given by assuming that the shear rate at the surface of the inner cylinder is equal to the average shear rate in the clearance between blades and wall, as given by Chavan and Ulbrecht, and the area of the inner cylinder is supposed to be the same as that of one side of the blade surface.

\[ \kappa = \frac{1}{2} \ln \left[ \frac{2w/D}{\left( (D/d) - (1 - 2w/d)/(D/d - 1) \right]} \right] \]
\[ a = \frac{h}{d} \sqrt{\frac{\pi^2}{(s/d)^2} \frac{Q_s}{N d^3}} \]

The pressure difference between the two cylinder surfaces is given by Eq. (11), when the inner one is rotating with an angular velocity, \( \Omega_i \), and the outer one is stationary.

\[ \Delta P = a^2 \left( \frac{D^2}{\pi d} \frac{v_{ex}^2}{r} \right) \left( \frac{\Omega_i D}{4} \right)^2 \times \left( \frac{1}{2\kappa^2} - \frac{2}{2\kappa^2} \ln \kappa \right) \]

The volume rate of the secondary flow in the actual vessel is assumed to be equal to that of the flow produced by this pressure gradient through the annulus in the imaginary vessel.

\[ Q_s = a^2 \frac{\pi^2}{512} \left( \frac{D^2}{\pi d} \right) \left( \frac{\Omega_i D}{4} \right)^2 \times \left( \frac{1}{2\kappa^2} - \frac{2}{2\kappa^2} \ln \kappa \right) \left( 1 - \frac{1 - \kappa^2 \ln (1/\kappa)}{1 - \kappa^2} \right) \]

Neglecting the effect of Reynolds number in Eq. (12) by taking account of the fact that the experimental values of \( C_i \) are independent of Reynolds number:

\[ Q_s = a^2 \frac{\pi^2}{256} \left( \frac{D^2}{\pi d} \right) \left( \frac{\kappa}{\kappa - 1/\kappa} \right) \left( \frac{1}{2\kappa^2} - \frac{2}{2\kappa^2} \ln \kappa \right) \times \left( 1 - \frac{1 - \kappa^2 \ln (1/\kappa)}{1 - \kappa^2} \right) \]

The volume rate of the exchange flow should have a maximum value when the ratio of \( Q_t \) and \( Q_s \) has a certain value. As a first approximation, we defined this exchange flow rate as a quadratic equation:

\[ Q_e = a^2 \frac{\pi^2}{128} \left( \frac{D^2}{\pi d} \right) \left( \frac{\kappa}{\kappa - 1/\kappa} \right) \left( \frac{1}{2\kappa^2} - \frac{2}{2\kappa^2} \ln \kappa \right) \times \left( 1 - \frac{1 - \kappa^2 \ln (1/\kappa)}{1 - \kappa^2} \right) \]

3.2 Correlation of mixing time

Equation (2) is assumed to be given by the following equation with the use of Eqs. (3), (8), and (14).

\[ \frac{1}{N_t} = \sqrt{\frac{N_r R e (D)^3}{\pi^4 \frac{Q_t}{N d^3} \frac{Q_s}{N d^3}}} \]

The three constants \( \alpha, \beta, \) and \( \gamma \) in \( Q_e (N d^3) \) were
determined by a multiple non-linear regression using the experimental data. The values of $\alpha$, $\beta$ and $\gamma$ are $-12800$, $0.000909$ and $0.0114$ respectively.

The calculated values of $N_{tm}$ are summarized in Table 2. Figure 14 shows the comparison of calculated and experimental values. The agreement is satisfactory except for DH6 and DH11 impellers. Therefore, this correlation is applicable to the following ranges of each geometrical variable of $n_p = 2$, $0.019 \leq c/D \leq 0.060$, $0.45 \leq s/D \leq 1.0$ and $0.075 \leq w/D \leq 0.16$ in the laminar flow region $Re = 0.1$ to 10.

4. Mixing Performance

To compare the mixing performance of the agitators, several criteria have been proposed. However, the results, listed in Table 2, indicate that a small variation of impeller dimensions has a great effect on mixing time, but a small effect on power consumption. Thus, mixing time mainly governs the selection of the geometrical configuration of the impeller.

Conclusion

The liquid crystal method is suitable for investigation of the mixing process and for observation of poorly mixed or stagnant zones. Using this method, observation of the mixing processes and measurement of mixing times were carried out for helical ribbon impellers of different geometries. It was found that the optimum values of $c/D$, $s/D$ and $w/D$ for the mixing process are about $0.06$, $0.90$ and $0.15$ respectively. These experimental results are correlated on the basis of a model considering the relation between flow pattern and impeller geometry.

Acknowledgment

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Nomenclature

\begin{align*}
\alpha &= \text{dimensionless area defined by Eq. (10)} \quad [-] \\
C_1 &= \text{geometrical constant in Eq. (1)} \quad [-] \\
c &= \text{clearance between blades and wall} \quad [m] \\
D &= \text{diameter of vessel} \quad [m] \\
d &= \text{diameter of impeller} \quad [m] \\
d_i &= \text{diameter of shaft} \quad [m] \\
H &= \text{height of vessel} \quad [m] \\
h &= \text{height of impeller} \quad [m] \\
L &= \text{length of blade} \quad [m] \\
M &= \text{magnitude of shear rate} \quad [-] \\
N &= \text{rotational speed of impeller} \quad [-] \\
N_p &= \text{power number} \quad [-] \\
N_r &= \text{number of impeller revolutions} \quad [-] \\
n_o &= \text{number of arms on each blade} \quad [-] \\
n_p &= \text{number of blades} \quad [-] \\
P &= \text{power consumption of impeller} \quad [W] \\
\Delta P &= \text{pressure gradient} \quad [Pa]
\end{align*}

Fig. 14 Correlation of mixing time

\begin{align*}
P_v &= \text{power consumption per unit volume} \quad [W \cdot m^{-3}] \\
Q_1 &= \text{circulation flow rate} \quad [m^3 \cdot s^{-1}] \\
Q_2 &= \text{secondary flow rate} \quad [m^3 \cdot s^{-1}] \\
Q_3 &= \text{exchange flow rate} \quad [m^3 \cdot s^{-1}] \\
Q &= \text{circulation flow rate defined by Eq. (6)} \quad [m^3 \cdot s^{-1}] \\
Re &= \text{Reynolds number} \quad [-] \\
r &= \text{radial position} \quad [m] \\
r_{2i} &= \text{radial position of inner edge of blade} \quad [m] \\
r_{2e} &= \text{radial position of outer edge of blade} \quad [m] \\
r_s &= \text{radial position of shaft} \quad [m] \\
r_v &= \text{radial position of vessel} \quad [m] \\
s &= \text{impeller pitch} \quad [m] \\
t &= \text{time required for tracer liquid to complete one circuit} \quad [s] \\
t_m &= \text{mixing time} \quad [s] \\
V &= \text{total volume in a vessel} \quad [m^3] \\
v_z &= \text{velocity profile of resultant flow} \quad [m \cdot s^{-1}] \\
v_{dr} &= \text{velocity profile of drag flow} \quad [m \cdot s^{-1}] \\
v_{pp} &= \text{velocity profile of pressure flow} \quad [m \cdot s^{-1}] \\
v_{tg} &= \text{tangential velocity} \quad [m \cdot s^{-1}] \\
w &= \text{blade width} \quad [m] \\
\sigma, \beta, \gamma &= \text{constants in Eq. (14)} \quad [-] \\
\theta_n &= \text{blade angle} \quad [-] \\
\kappa &= \text{dimensionless diameter of inner cylinder} \quad [-] \\
\lambda &= \text{dimensionless radial position where } v_z \text{ is equal to zero} \quad [-] \\
\mu &= \text{viscosity} \quad [Pa \cdot s] \\
\rho &= \text{density} \quad [kg \cdot m^{-3}] \\
\Omega &= \text{angular velocity} \quad [s^{-1}]
\end{align*}

Literature Cited

EXPERIMENTAL INVESTIGATION OF CONTINUOUS NAD RECYCLING BY CONJUGATED ENZYMES IMMOLIZED IN ULTRAFILTRATION HOLLOW FIBER

Osato Miyawaki, Kozo Nakamura and Toshimasa Yano
Department of Agricultural Chemistry, University of Tokyo, Tokyo 113

Dynamic NAD recycling, in which native NAD, without immobilization, was dynamically recycled in continuous operation by immobilized conjugated enzymes (alcohol dehydrogenase and lactate dehydrogenase), was investigated experimentally. The results were compared with the theoretical model calculations in the preceding paper. Although there were some quantitative differences, the theoretical model could explain the experimental results at various operating conditions. Under a properly selected operating condition, a high NAD recycle number (6180) as well as a satisfactory conversion (34.6%) of the limiting substrate was obtained experimentally. The operational stability of the present system was fairly good, retaining 70% of its original activity after one month's continuous operation. Compared with the immobilized-NAD method, the dynamic NAD recycling method is simple and is free from the problem of coenzyme inactivation during continuous operation. The experimental results obtained here showed the practicability of dynamic NAD recycling.

Introduction

The establishment of an effective method of cofactor recycling is essential for the utilization of immobilized enzyme reactions requiring such free organic cofactors as NAD, ATP etc. because these cofactors are generally very expensive and often cost-limiting. As for NAD, the immobilized-NAD method has been studied by several investigators. The present authors, however, theoretically studied another method, dynamic NAD recycling, as described in the preceding paper. Free NAD, without immobilization, was dynamically recycled between its oxidized and reduced state by the action of immobilized conjugated enzymes. With properly selected conditions of reactor and operation, this method was proved to give very high NAD recycle number with satisfactory conversion of limiting substrate.

The situation assumed in the theoretical model is as follows. The conjugated enzymes (alcohol dehydrogenase=ADH and lactate dehydrogenase=LDH) are immobilized in an ultrafiltration hollow fiber tube. The conjugated substrates (ethanol and pyruvate) and NAD are fed to the shell-side of the hollow fiber reactor, permeate into the tube and participate in the following enzyme reactions.

\[ \text{ADH} \quad \text{Ethanol} + \text{NAD} \rightarrow \text{Acetaldehyde} + \text{NADH} \]  
\[ \text{LDH} \quad \text{Pyruvate} + \text{NADH} \rightarrow \text{L-Lactate} + \text{NAD} \]

Reactor performance at steady state was analyzed.