THE TAPERED FLUIDIZED-BED BIOREACTOR - AN IMPROVED DEVICE FOR CONTINUOUS CULTIVATION*

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INTRODUCTION

Although continuous culture processes have been in use for more than 50 years, full-scale production in continuous cultivation systems has been limited to a few processes, most notably the activated sludge process for treatment of sewage and industrial wastes. Since continuous reactors usually have a considerably higher productivity than batch reactors, there has been a renewed interest in developing continuous biocontactors or bioreactors for other biological processes, particularly the more efficient and controllable fixed-bed and fluidized-bed systems.

In comparing the latter configurations, both of which require a solid phase containing the active biological agent, certain advantages of the fluidized bed for use as a bioreactor become apparent:

1) minimization, if not elimination, of the plugging caused by biomass accumulation;

2) use of smaller particles with higher specific surface area, thereby allowing greater specific reaction rates at low pressure drop; and

3) removal and replacement of active bed during operations to maintain maximum reactivity.

On the other hand, equitable flow distribution throughout the reactor cross section, particularly near the feed entry point, can present a design problem in fluidized-bed systems, just as it does in fixed-bed systems.

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The usual fluidized bed also has a relatively narrow range of operating flow rates, thus making it rather difficult to maintain nonfluctuation operation. The tapered column design concept described here can be utilized to circumvent both of these problems.

The concept of the tapered fluidized bed bioreactor and some preliminary results have been presented earlier. This paper describes a mathematical model of the tapered fluidized bed and its use in conjunction with experimental results on small scale systems to evaluate this reactor concept for several important bioprocesses and provide information for scale-up.

**Reactor Concepts**

**Principles of operation**

The tapered fluidized bed differs from the more conventional fluidized bed by a configuration that resembles an inverted, truncated cone (with a small vertex angle) rather than a constant cross-sectional column. Thus, there is gradual expansion from a relatively small cross-sectional area of the entry to one that may be several times larger. The taper of the column allows a wide range of flow rates without loss of bed material since the fluidizing velocity decreases with reactor height. The gradual expansion of the column also allows for a stable feed introduction without gross eddies and significant backmixing. This is illustrated in Figure 1. As the flow rate is increased, the bed changes from fixed, through incipient fluidization, to fully expanded in the same fashion as a cylindrical fluid bed. However, in contrast at higher flow rates the tapered bed simply expands into a portion of the reactor with a larger cross-section rather than being swept from the reactor. At flow rates where the superficial velocity in the lower part of the reactor is greater than the Stokes settling velocity of the bed particles, the lower zone of the reactor will be free of the fluidized bed, but particles will remain in the upper zone.
Mathematical model

Although a spherical coordinate system is more natural for conic sections, the envisaged use of both cylindrical and tapered sections in full-scale bioreactors, suggested the use of cylindrical coordinates. Earlier an empirical mathematical model which predicted bed expansion, pressure drop, and chemical reactivity was developed by considering the reactor to be a series of discrete cylindrical sections, each subsequent section having a larger diameter. The differential model developed here begins with the continuity equation and simple chemical kinetics.

The continuity equation in cylindrical coordinates for a particular species of concentration, $C$, being consumed by an nth order chemical reaction, is:

$$\frac{\partial C}{\partial t} - D_a \frac{\partial^2 C}{\partial z^2} + \frac{U_z}{\varepsilon} \frac{\partial C}{\partial z} = \frac{-k C^n}{\varepsilon},$$

(1)

where

$D_a$ = axial dispersion coefficient,
$U_z$ = axial fluid velocity,
$\varepsilon$ = void fraction, and
$k$ = volumetric reaction rate constant.
For a tapered reactor, the radius is no longer fixed but is dependent on the height dimension, \( Z \), as can be easily visualized by Fig. 2.

Thus, the axial fluid velocity in a tapered reactor is not constant with a constant volumetric flow rate, \( Q \), but rather varies with axial position as

\[
u_Z = \frac{Q}{\pi R^2} = \frac{Q}{\pi R_0^2 (1 + \xi)^2} = \frac{u_{Z0}}{(1 + \xi)^2},
\]

where \( u_{Z0} \) is the axial velocity at the reactor inlet. The continuity equation for a tapered reactor can be obtained by modifying Eq. (1), using the taper coordinate as defined in Fig. 1 to give a dimensionless form:

\[
\frac{\partial F}{\partial \tau} - \frac{1}{Pe_a} \frac{\partial^2 F}{\partial \xi^2} + \frac{1}{\epsilon (1 + \xi)^2} \frac{\partial F}{\partial \xi} + \frac{F}{\epsilon} = 0,
\]

where the dimensionless variables are

\[
F = \frac{C}{\tau} ; \quad \tau = \frac{tu_{Z0}}{Q} ; \quad Pe_a = \frac{u_{Z0} Z_0}{D_a} ; \quad \xi = \frac{Z}{Z_0} ; \quad K = \frac{kC_0^{n-1}Z}{u_{Z0} \delta}.
\]

Eq. (3) has been solved for two sets of assumptions, and the results have been used to analyze experimental results obtained with tapered fluidized-bed bioreactors.

(A) Steady-state, first-order reaction with plug flow:

For this case, \( Pe_a = \infty \). Eq. (3) reduces to

\[
\frac{dF}{d\xi} + K(1 + \xi)^2 F = 0,
\]

with the boundary condition,

\[
\xi = 0, \quad F = 1.
\]
Thus, the concentration profile for a single reactor is:

$$ F = \frac{C}{C_0} = \exp\left\{ -K \xi \left(1 + \xi + 1/3 \xi^2\right) \right\}, \quad (6) $$

where $K = \frac{Z_0 u}{u_{Z_0}}$. By rearranging Eq. (6), the reaction rate coefficient becomes

$$ k = \frac{u_{Z_0}}{Z_0 \left(1 + \xi + 1/3 \xi^2\right) \xi \ln F}. \quad (7) $$

(B) Steady-state, nth-order reaction with plug flow:

For this case, Eq. (3) reduces to

$$ \frac{dF}{d\xi} + K(1 + \xi)^2 F^n = 0. \quad (8) $$

With the boundary condition given in Eq. (5), the concentration profile is given as

$$ F = \frac{C}{C_0} = \left[1 - (1 - n) K \xi \left(1 + \xi + 1/3 \xi^2\right)\right]^{\frac{1}{1-n}}, \quad (9) $$

where $K = \frac{kC_0^{n-1}Z_0}{u}$. By rearranging Eq. (9), the reaction rate coefficient becomes

$$ k = \frac{(C_0^{1-n} - C^{1-n})u_{Z_0}}{(1-n)Z_0 \xi (1 + \xi + 1/3 \xi^2)}. \quad (10) $$

EXPERIMENTAL

The tapered fluidized bed has been tested in several different configurations and with several different biological systems. The mathematical model was used to analyze experimental data obtained during biological conversion of dissolved nitrates to molecular nitrogen using *Pseudomonas* bacteria. Two tapered fluidized-bed reactors were arranged in series (Fig. 3) and initially
charged with approximately 1.2 l each of anthracite coal (150- to 180µm diam) on which Pseudomonas bacteria were allowed to grow. After a sufficient biomass was established in the reactors, the test solution with methanol as a carbon source (Table 1) was pumped through the reactors at a rate of 5 to 6 ml/s for several days. The effluent from the second reactor was retained in a cold room (4°C) and then used as a feed for the subsequent run. Nitrate concentrations in the effluent of each reactor were measured during each run using a specific ion electrode, and average values are shown in Table 2.

<table>
<thead>
<tr>
<th></th>
<th>Feed Composition for Denitrification Tests</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₄NO₃</td>
<td>5.7 g/l</td>
</tr>
<tr>
<td>KH₂PO₄</td>
<td>0.04 g/l</td>
</tr>
<tr>
<td>MgSO₄</td>
<td>0.18 g/l</td>
</tr>
<tr>
<td>FeCl₂</td>
<td>0.6 g/l</td>
</tr>
<tr>
<td>Na₂MoO₄·2H₂O</td>
<td>1.0 g/l</td>
</tr>
<tr>
<td>Methanol</td>
<td>13 ml/l</td>
</tr>
</tbody>
</table>

For the first-order reaction assumption, the experimental nitrate concentrations were used in Eq. (7) to calculate the reaction rate coefficient for each reactor in series (Table 2).

This rate coefficient was not constant but could be correlated in terms of initial concentration,

\[
k = 7.98 C₀^{-0.525} \text{ (min}^{-1})
\]

(11)

Using the rate coefficient calculated from Eq. (11) in Eq. (7), the nitrate concentrations exiting each reactor can be calculated. These are also shown in the fourth and fifth columns of Table 2. The agreement of experimental effluent concentrations and those calculated using Eq. (11) as the reaction rate coefficient is excellent.
Table 2. Comparison of experimental and calculated results

<table>
<thead>
<tr>
<th>Reactor</th>
<th>Experimental Nitrate concentration (mg/L)</th>
<th>Calculated reaction rate coefficient, k (min⁻¹)</th>
<th>Calculated effluent nitrate concentration (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Feed</td>
<td>Effluent</td>
<td>Using Eq. (7) with experimental results</td>
</tr>
<tr>
<td>1</td>
<td>2500</td>
<td>1900</td>
<td>0.1081</td>
</tr>
<tr>
<td>2</td>
<td>1900</td>
<td>800</td>
<td>0.1531</td>
</tr>
<tr>
<td>3</td>
<td>800</td>
<td>200</td>
<td>0.2453</td>
</tr>
<tr>
<td>4</td>
<td>200</td>
<td>13</td>
<td>0.4837</td>
</tr>
</tbody>
</table>
For the nth-order reaction assumption, trial-and-error calculations showed that the best fit to the experimental data is obtained with,

\[ n = 0.61, \]

\[ k = 2.426 \left( \frac{\text{mg}}{\text{l}} \right)^{0.39} \text{min}^{-1}. \]

The nitrate concentrations exiting each reactor were also calculated using Eqs. (9) and (12) and tabulated in the last column of Table 2. The comparison of this model with experimental results is not as good as the first-order reaction model with the reaction rate coefficient, depending on the initial concentration, but is still very reasonable.

Some interesting data on the hydraulic characteristics of a tapered fluidized bed were obtained with a variable angle test unit with movable side walls (Table 3). The tests were performed using 500 g of -25+40 mesh coal at flow rates from 0 to 1200 ml/min for each of five taper angles. The hydraulic pressure drop and expanded bed heights as a function of volumetric flow rate is from 0 to 1200 ml/min, are shown in Figures 4 and 5.

APPLICATIONS

The tapered fluidized-bed bioreactor has been used in a wide variety of applications, with immobilized enzymes or adhering microorganisms. Typical results obtained in some of these applications are summarized in Table 4. The results shown are not maximum values but rather typify readily attainable values.

Glucose was produced in a TFBBR by the hydrolysis of lactose using lactase enzyme immobilized on alumina (0.01 cm av diam) with an activity of 5.0 x 10^-5 moles of lactose per min per gram of solid. Lactose conversion rates greater than 90% were attained with residence times of 40 min in a TFBBR containing 710 g of bed.
Table 3. Variable angle test dimension

<table>
<thead>
<tr>
<th>Angle of divergence (degrees)</th>
<th>Flow Channel Dimensions</th>
<th>Top width (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Length = 114 cm</td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>Depth = 5.1 cm</td>
<td>3.8</td>
</tr>
<tr>
<td></td>
<td>Bottom width = 2.5 cm</td>
<td>5.4</td>
</tr>
<tr>
<td>0.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.6</td>
<td></td>
<td>8.1</td>
</tr>
<tr>
<td>1.4</td>
<td></td>
<td>10.2</td>
</tr>
<tr>
<td>2.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3.9</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 5. Tapered fluidized-bed bioreactor

<table>
<thead>
<tr>
<th>Application</th>
<th>Feed</th>
<th>Residence time (min)</th>
<th>Effluent concentration (mg/liter)</th>
<th>Reactor conversion rate (kg/day-m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glucose production</td>
<td>50 g/l lactose</td>
<td>6.7</td>
<td>$20 \times 10^2$</td>
<td>$\sim 5000$</td>
</tr>
<tr>
<td>Denitrification</td>
<td>1000 mg/l NO₃</td>
<td>17</td>
<td>242</td>
<td>97</td>
</tr>
<tr>
<td>Nitrification</td>
<td>630 mg/l NH₄ (97% recycle)</td>
<td>142</td>
<td>480</td>
<td>1.7</td>
</tr>
<tr>
<td>Phenol degradation</td>
<td>180 mg/l phenol</td>
<td>7.5</td>
<td>0.5</td>
<td>35</td>
</tr>
<tr>
<td>CCP waste treatment</td>
<td>31 mg/l phenol</td>
<td>7.6</td>
<td>0.15</td>
<td>6</td>
</tr>
<tr>
<td>Anaerobic sewage treatment</td>
<td>Synthetic sewage</td>
<td>9 mg/l thiocyanate</td>
<td>7.6</td>
<td>1.2</td>
</tr>
<tr>
<td></td>
<td>191 mg/l TOC</td>
<td>(97% recycle)</td>
<td>330</td>
<td>.8</td>
</tr>
</tbody>
</table>
In addition to the multiple reactor studies described earlier, biological denitrification of streams containing up to 7.5 g/liter NO₃ has been carried out in a 4.2 m tall TFBBR. Effluent nitrate concentrations less than 1 mg/liter can be obtained. The conversion of ammonia nitrogen to nitrate with subsequent denitrification has also been demonstrated.

Aqueous effluent waste streams from coking plants, coal conversion processes (CCP), and the petroleum industry often contain significant concentration of phenolic compounds. A TFBBR containing anthracite coal particles with adhering Pseudomonas bacteria and operated aerobically has been used to reduce the phenol content of an aqueous stream from 100 mg/liter to less than 25 µg/liter. Tests with an authentic CCP aqueous stream indicated that the phenol degradation rate was somewhat reduced by other contaminants, but phenol levels below 1 mg/liter could be attained. Simultaneous reduction of thiocyanate concentration was also achieved. In general, phenol degradation rates in the TFBBR were 10 to 50 times those reported for continuous stirred-tank reactors on a per-unit volume basis. The TFBBR has also been investigated as a bioreactor for anaerobic treatment of municipal sewage attaining results better than with a submerged filter.

CONCLUSIONS

The bioreactor concept utilizing a tapered fluidized bed has shown considerable promise for use in bioprocesses where the biological agents can be immobilized on a fluidizable solid phase, but the operating characteristics of such a reactor are not yet fully understood. A simple mathematical model (steady state, plug flow conditions) has been developed and tested with experimental data, and calculated values compare favorably with experimental values. A more complete and presumably more valid mathematical model incorporating void volume changes and particle size distribution is being developed.
TAPERED FLUIDIZED BED OPERATION

- FIXED BED
- INITIAL FLUIDIZATION
- INCREASED FLOW
- HIGHER FLOW
\[ R = (Z_0 + Z) \tan \theta, \]
\[ = R_0 + Z \tan \theta, \]
\[ = R_0 (1 + \xi). \]

where
\[ \xi = \frac{Z}{R_0} \tan \theta, \]
and
\[ R_0 = Z \tan \theta, \]
\[ \xi = \frac{Z}{R_0} \tan \theta = \frac{Z}{Z_0}. \]
ORNL DWG 76-932R2

TAPERED FLUID BED
42 in. long
1 in. diam bottom
3 in. diam top
2349 ml volume
1.36° included angle
BED EXPANSION IN WEDGE SHAPED BEDS

FLOW (ml/min)

BED HEIGHT (cm)

0°

0.6°

1.4°

2.8°

3.9°
FLOW RATE (ml/min)

PRESSURE DROP (kPa)

FLUIDIZATION CURVES FOR DIVERGING WEDGE BEDS

Figure 5
REFERENCES


FIGURE LIST

Fig. 1. Fluidization of a tapered bed.

Fig. 2. Coordinate system for the tapered fluidized bed.

Fig. 3. Experimental system with two TFBBRs in series.

Fig. 4. Bed expansion in wedge shaped fluidized beds.

Fig. 5. Fluidization curves for diverging wedge beds.