Measurement of Wetting Efficiency in a Trickle-Bed Reactor

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Abstract

Wetting efficiency in trickle-bed reactor (TBR) is defined as the fraction of outer surface area wetted by liquid phase. In this study, a model is established based on the non-steady state adsorption of extremely diluted phenol solution by highly porous activated carbon to estimate the liquid to solid mass transfer coefficient and the wetting efficiency in accordance with its original definition. Both the measured mass transfer coefficient and wetting efficiency were found in good agreement with other relevant studies.

Keywords: wetting efficiency; trickle bed reactor; adsorption; mass transfer; modelling; non-linear dynamics

Introduction

In trickle-bed reactor (TBR), the incomplete wetting normally occurs in low interaction regime where the flow rates of both gas and liquid phase are considerably reduced. Studies on incomplete wetting in TBR are of great significance since it will affect the contact area of liquid to solid phase and liquid-solid mass transfer rate.

As most of the solid phase in TBR is highly porous, wetting efficiency can be classified into internal wetting efficiency and external wetting efficiency. Internal wetting efficiency refers to the portion of internal area of the catalyst filled by liquid due to capillarity. While external wetting efficiency refers to the fraction of external area of the catalyst wetted by liquid.

Except for the highly exothermic reactions and volatile liquid reactants, the internal wetting efficiency can be considered unity for most cases as a result of capillarity. Thus, wetting efficiency in TBR is usually referred to as what ‘external wetting efficiency’ is defined:

\[ \eta = \frac{a_S}{a_P} \]  \hspace{1cm} (1)

Tracer technique is the most commonly adopted approach in measuring wetting efficiency (Al-Dahan & Dudukovic, 1995; King & Misset, 1989, 1991; Mills & Dudukovic, 1981; Tsamatsoulis & Papayannakos, 1996), where a step or impulse concentration of tracer is injected into the liquid at the inlet of TBR and the distribution of the concentration of the tracer with time is monitored at the outlet of the reactor. The effective diffusivity can be calculated through the analysis on this response curve. Therefore the wetting efficiency is then defined by

\[ \eta = \sqrt{D_T / D_F} \]  \hspace{1cm} (2)

or, \[ \eta = D_T / D_F \]  \hspace{1cm} (3)

Although this technique is widely adopted, there are always some uncertainties that which of the two aforementioned expressions should be more appropriate. Neither sufficient discussions nor conclusions have been mentioned in the relevant reports.

On basis of the measurement of volumetric liquid-solid mass transfer coefficients \( k_a a_b \) by the dissolution technique, Lakota & Levec (1990) once defined the wetting efficiency as the ratio of mass transfer coefficients under two-phase-flow to single-phase-flow:

\[ \eta = \frac{(k_a a_b)_{two-phase-flow}}{(k_a a_b)_{single-phase-flow}} \]  \hspace{1cm} (4)

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Likewise, Pironti et al. (1999) defined the wetting efficiency as the ratio of liquid-solid shear stress between two-phase flow and liquid-full operation at the same intrinsic liquid and gas velocities:

\[
\eta = \frac{\tau_s a_L}{\tau_s a_L + \tau_{L \text{ - } \text{solid Right)}}} \quad (5)
\]

Both of these two definitions [eqs (4) and (5)] imply that the presence of gas phase accounts for partial wetting phenomenon in TBR.

Chemical reaction is another frequently mentioned method (Saroha & Nigam, 1996; Llano et al., 1997) employing conversion rate data of a chemical reaction under different wetting situations to estimate wetting efficiency.

In this study, we established a non-steady adsorption model that endeavored to measure the liquid-solid contact area at incomplete wetting region. The model indicates that the wetting efficiency is obtainable directly from its original definition that it is the ratio of liquid-solid contact area to the total surface area of the solid phase.

Model

According to ‘film theory’, the whole liquid to solid mass transfer process in a TBR can be further divided into two sub-steps. First the solute transfers from the bulk liquid to the interface of liquid-solid (sub-step 1) and subsequently from the interface to the solid (sub-step 2). Usually it requires fairly complex analysis on sub-step 2 which involves both inter- and intra-particle diffusions. Therefore, we focused our investigation on sub-step 1 and established a model by assuming that:

(1) The liquid phase is in ‘plug flow’ pattern, i.e., no axial diffusion exists;
(2) The concentration of solute (A) is so diluted that the flux of solvent (B) is constant and no convective mass transfers across the liquid-solid interface; and
(3) The size of solid particle is rather small so that concentration of A surrounding any given particle is essentially constant.

Thus for a bed layer with thickness of dz, the mass transfer between the bulk liquid and liquid-solid interface can be described by eq. (6):

\[
\varepsilon dz \frac{\partial c_A}{\partial t} = -F_B dz \frac{\partial c_A}{\partial z} - Sdz(k_a a_l)(c_A - c_{Al}) \quad (6)
\]

Eq. (6) can be simplified by introducing a modified time variable

\[
t' = t - \frac{z}{F_B} \quad (7)
\]

Here t' can be interpreted as the time it requires for the liquid ‘front’ to arrive at a certain length of z in the bed.

With assistance of t' we may rewrite eq. (6) into

\[
\left( \frac{\partial c_A}{\partial t} \right)_t = \frac{-S(k_a a_l)}{F_B}(c_A - c_{Al}) \quad (8)
\]

with initial condition:

at \( t' = 0, \ c_{A0} = 0 \) \quad (9)

and boundary condition:

at \( z = 0, \ c_A = c_{Al} \) \quad (10)

The partial differential eq. (8) has a specific solution at \( t' = 0 \) and \( z = 1 \), as shown in eq. (11):

\[
c_{A0} = c_{Al} \exp \left( -\frac{k_a a_l}{u_B} \right) \quad (11)
\]

Eq. (11) indicates that once \( c_{A0} \) has been determined at certain liquid superficial velocity \( u_B \), the volumetric liquid-solid mass transfer coefficient \( k_a a_l \) can be calculated. It has to be noted that eq. (11) is invalid for estimating \( k_a a_l \) unless the bed volume is very small to make sure that the effluent concentration at \( t' = 0 \) \( (c_{A0}) \) is greater than 0. This equation is identical to what Tan and Smith (1982) derived in measuring \( k_a \).

Moreover, eq. (11) gives rise to the estimation of wetting efficiency. The specific interfacial area \( a_l \) can be estimated as below:
\[ a_{st} = a_e \eta = (1 - \varepsilon) a_p \eta \]  \hspace{1cm} (12)

It has been summarized from the survey on a number of literatures that wetting efficiency is usually in the range of 0.6 to 1.0. Besides, when liquid flow rate has been increased to be higher than 4.0 kg/m\(^2\)/s, the incomplete wetting phenomenon vanishes (Lakota & Levec, 1990; Al-Dahhan & Dudukovic, 1995; Mille & Dudukovic, 1981).

Based on this information, we believe that the specific contact area \( a_e \) can be further written into:

Completely Wetted Region: \( a_{st} = (1 - \varepsilon) a_p \) when \( L_B > 4.0 \) kg/(m\(^2\)/s); \hspace{1cm} (13)

Partially Wetted Region: \( a_{st} = \eta (1 - \varepsilon) a_p \) when \( L_B < 4.0 \) kg/(m\(^2\)/s). \hspace{1cm} (14)

Since in the completely wetted region \( a_e \) is only subject to the physical properties of the bed, we can calculate mass transfer coefficient \( k_{st} \) through extracting \( a_e \) from \( k_{st} \). Once \( k_{st} \) has been rationally extrapolated from completely wetted region to partially wetted region, \( a_e \) under partially wetted region is attainable. Accordingly the wetting efficiency can be obtained according to its original definition in eq. (1). This point comprises the foundation of this study.

**Experimental**

**Bed**

In our tests, a small steel tube (inner diameter = 1.0 cm) was used to simulate the reactor column. Activated carbon (AC) purchased from American Norit Co. served as the solid phase. To avoid possible wall flow, the cylindrical AC extrudates were crushed and sieved into several categories. The particles sizing from 125 to 250 \( \mu \)m were selected and loaded into the reactor. Due to the large particle diameter ratio of reactor-particle (>40), the wall flow problem can be adequately prevented. In addition, such fine particles were able to meet the requirement set in assumption (3). We assumed the particles were in spherical shape and expressed the solid contact area \( a_e \) as:

\[ a_e = 6/d_p \]  \hspace{1cm} (15)

To ensure the validity of using eq. (11), each test the AC loading was 0.1 g which formed a layer of around 2.7 mm in thickness. Within such thin layer the axial diffusion may be ignored, as mentioned in assumption (1).

The bed porosity was experimentally determined by eq. (16) to be 0.14, according to the bulk density of AC particles.

\[ \varepsilon = 1 - \left( \frac{m_{AC}}{\rho} \right)/V \]  \hspace{1cm} (16)

In order to improve the uniformity of liquid distribution within the reactor, in addition to the AC layer, we loaded a layer of inert polymer cylindrical extrudes (3.5 mm x 1.5 mm). Normally the polymer layer had a thickness of 1.9 cm or so.

**Liquid**

Aqueous phenol was adopted as the liquid phase. The initial concentration was set to 5 mg L\(^{-1}\) and it was found experimentally that after passing through the bed the reduction in liquid flow rate was negligible. With regards to these facts, the requirement of assumption (2) can be met. The liquid flow rate varied from 2 to 60 mL min\(^{-1}\), corresponding to trickle flow pattern. A detailed description of both the solid and liquid properties is listed in Table 1.

The effect of gas flow on liquid-solid mass transfer at dispersed flow regime, as reviewed by most of the researchers, is much less significant particularly in trickle flow region. Lakota & Levec (1990) found that when the gas flow rate was increased by nearly 100 times (from 0.003 to 0.294 kg/(m\(^2\)/s)), in the trickle flow region the increase of mass transfer coefficient was a round 30%. Therefore, gas flow effect was not taken into consideration in this work.

**Experimental Procedure**

For each run, Fresh AC and inert polymer particles were loaded into the reactor. Then the prepared phenol solution was pumped into the column through a measuring pump at a chosen flow rate, as shown in Fig. 1. The liquid was sampled at the bottom of the tube immediately after the liquid had flown through the fixed bed.

**Analysis**

The sampled aqueous phenol first reacted with 4-amionantipyrine solution at certain prepared buffer solution. The product then was colored with the addition of K\(_2\)Fe(CN)\(_6\). A Shimadzu UV-visible
spectrophotometer was used to determine the phenol concentration by measuring the light intensity of the colored solution at a wavelength of 510 nm.

**Results and Discussions**

**Reliability on the Measurement of \((c_0/d)\)**

Eq. (11) also implies that the measurement of \(c_{A0}\) is particularly important in estimating \(k_a\) and \(\eta\). As there is no on-line reading device connected to the reactor with the UV spectrometer in our study, for each run we first sampled approximately 20mL of solution and then transferred it to UV spectrometer for analysis. Consequently each time it took us 20-600 seconds to get one sample, depending on the liquid flow rates. It may be questionable whether or not such way of sampling could provide satisfactory accuracy in determining \(c_{A0}\). Thus, in test No.19 we took four samples consecutively with each sampling time of 5 seconds. The readings of these samples on UV spectrometer are shown in Table 2.

The insignificant discrepancy among the readings indicates that the 'batch-wise' sampling method is able to accurately estimate \(x_{A0}\).

**Volumetric Mass Transfer Coefficient \((k_a \alpha)\)**

The volumetric mass transfer coefficient \((k_a \alpha)\) estimated according to eq. (11) is plotted in Fig. 2. In this study, the completely wetted region was compulsorily defined where the superficial liquid velocity \(u_b\) is above 0.004 m s\(^{-1}\) (or \(L_b\) is higher than 4.0 kg (m\(^2\))\(^{-1}\) correspondingly).

As mentioned previously, in the completely wetted region \(\alpha_b\) is a fixed parameter \((=\frac{6(1-\eta)}{d_p})\), here \(d_p\) refers to the algebraic average of the AC particle size range). Thus \(k_b\) can be extracted from \(k_a \alpha_b\) and correlated by

\[
Sh = \frac{k_a \alpha_b d_p}{D} = 3.37 \times 10^3 \text{Sc}^{0.80} \text{Re}^{0.80} \text{Sc}^{1/3} \quad (17)
\]

To compare our result with those acquired by other researchers, Fig. 3 displays the various relationship of Sherwood Number with Reynolds Number as reported by other relevant studies (Correlation 1 by Hirose, Mori & Sato, 1976; Correlation 2 by Specchi, et al., 1978; Correlation 3 by Tan & Smith, 1982; Correlation 4 by Latifi, et al., 1988; Correlation 5 by Burghardt, et al., 1995).

Fig. 3 indicates that the correlation of this work is within the range scattered by other available expressions and quite close to that obtained by Tan and Smith (1982). The deviations arising among these various expressions may be ascribed to the variations in the bed characters, i.e. physical properties of the fixed and mobile phases. The computation method also accounts for the disagreements.

The reproducibility of the tests was verified by observing nearly constant \(k_a \alpha_b\) at various AC loading from 0.1 to 0.2 g, as shown in Table 3.

**Wetting Efficiency**

By means of eq. (17) \(\alpha_b\) at partially wetted region can be obtained by extracting \(\alpha_b\) from \(k_a \alpha_b\) and therefore the wetting efficiency is obtainable according to:

\[
\eta = \frac{\alpha_b \text{ partially-wetted region}}{\alpha_b \text{ completely-wetted region}} \quad (18)
\]

The resultant wetting efficiency at various liquid velocities is measured and roughly correlated, as displayed in Fig. 4.

It can be seen from Fig. 4 that generally the particle wetting situation can be improved by increasing liquid velocity. The wetting efficiency increased from around 0.5 to at \(u_b\) of \(4.25 \times 10^4\) m s\(^{-1}\) to nearly unity at \(u_b\) of \(2.12 \times 10^5\) m s\(^{-1}\) and can be approximately correlated by

\[
\eta = 4.45u^{0.27} \quad (19)
\]
This is in agreement with what the other researchers observed (Mills & Dudukvoic, 1981; Llano et al., 1997; Ring & Missen, 1991; Lakota & Levec, 1990; Lakota & Levec, 1990) but differs with Tsamatsoulis & Papayannakos’s report (1996), where the wetting efficiency was found weakly dependent on liquid flow rate (3 - 10 × 10^{-4} m/s).

Conclusions

In this study we adopted a non-steady state adsorption method to investigate the liquid-solid mass transfer-related parameters in trickle bed reactor. A model based on mass transfer from bulk liquid to liquid-solid interface was established that is found capable of calculating volumetric mass transfer coefficient ($k_0a_0$). The whole region was divided into completely wetted region and partially wetted region, on basis of the summary of relevant literatures that the solid phase can be deemed completely wetted when liquid flow rate is higher than 4.0 kg (m^2 s)^{-1}. In completely wetted region the contact area ($a_0$) is determined by the physical properties of the bed and mass transfer coefficient ($k_0$) was measured and correlated by $\displaystyle Sh = \frac{k_0d_p\rho}{D} = 3.37Re^{0.40}Sc^{1/3}$. By means of this correlation, the contact area ($a_0$) in partial wetting region was obtained to derive the wetting efficiency. All the results were in good consistency with other reports.

Acknowledgments

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Notation

- $a_b =$ contact surface area of liquid-solid per unit volume of solid, m\(^{-1}\)
- $a_r =$ total surface area of particle, m\(^2\) m\(^{-3}\) of empty bed
- $a_p =$ total surface area of particle, m\(^2\) m\(^{-3}\) of particle
- $a_w =$ wetted surface area of particle, m\(^2\) m\(^{-3}\) of particle
- $c_A =$ concentration of solute (A) in bulk liquid, kg m\(^{-3}\)
- $c_{Al} =$ concentration of solute (A) at liquid-solid interface, kg m\(^{-3}\)
- $c_{AI} =$ concentration of solute (A) in bulk liquid at $t' = 0$ and $z = l$, kg m\(^{-3}\)
- $c_{AO} =$ initial concentration of solute (A) in bulk liquid, kg m\(^{-3}\)
- $F_B =$ flow rate of solvent (B), m\(^3\) s\(^{-1}\)
- $k_b =$ liquid-solid mass transfer coefficient, m s\(^{-1}\)
- $l =$ whole bed depth, m
- $L_B =$ liquid flow rate, kg (m\(^2\) s\(^{-1}\))
- $m_{AC} =$ the mass of activated carbon;
- $S =$ cross-sectional area of (empty) bed, m\(^2\);
- $Sh =$ Sherwood number
- $Sc =$ Schmidt number
- $t =$ time, s
- $t' =$ modified time, s
- $Re =$ liquid Reynolds number
- $u_0 =$ superficial velocity of water, m s\(^{-1}\)
- $z =$ bed length, m

Greek Letters

- $\varepsilon =$ volume fraction of column occupied by liquid (bed porosity)
- $\eta =$ partial wetting efficiency
- $\mu =$ liquid viscosity, kg (m s\(^{-1}\))
- $\rho =$ bulk density of activated carbon, 0.5956 g mL\(^{-1}\)

References


Figures

Fig. 1 Schematic Diagram of Apparatus
Fig. 2 Dependence of Volumetric Mass Transfer Coefficient ($k_{lb}a_b$) on Liquid Superficial Velocity $u_b$

Fig. 3 Various Correlation for $Sh/Sc^{1/3}$ (●: Experimental Data of this Work; Lines from top to bottom: Correlation 3, Correlation of this Work, Correlation 4, Correlation 5, Correlation 3 and Correlation 2)
Fig. 4 Dependence of Wetting Efficiency $\eta$ on Liquid Superficial Velocity $u_B$

Tables

Table 1 Physical Properties of Liquid

<table>
<thead>
<tr>
<th>Solid</th>
<th></th>
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</thead>
<tbody>
<tr>
<td>AC Bulk Density, g mL$^{-1}$</td>
<td>0.5956</td>
</tr>
<tr>
<td>AC Bed Porosity</td>
<td>0.14</td>
</tr>
<tr>
<td>AC Bed Thickness, mm</td>
<td>2.8 – 4.2</td>
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<tr>
<td>AC Particle Size, $\mu$m</td>
<td>125 - 250</td>
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<tr>
<td>Inert Bed Thickness, mm</td>
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<table>
<thead>
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<th>Liquid</th>
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</tr>
</thead>
<tbody>
<tr>
<td>Flow Rate, mL min$^{-1}$</td>
<td>2 - 60</td>
</tr>
<tr>
<td>Density, kg m$^{-3}$</td>
<td>$1.0 \times 10^3$</td>
</tr>
<tr>
<td>Viscosity, kg (m-s)$^{-1}$</td>
<td>$1.004 \times 10^{-3}$</td>
</tr>
<tr>
<td>Diffusivity of Aqueous Phenol in Water, m$^2$ s$^{-1}$</td>
<td>$6.23 \times 10^{-10}$</td>
</tr>
</tbody>
</table>

*: Estimated according to Wilke-Chang Method.

Table 2 UV Spectrometer Readings for four Consecutively Taken Samples ($F_B = 59$ mL min$^{-1}$)

<table>
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<tr>
<th>Time, second</th>
<th>0–5</th>
<th>5–10</th>
<th>10–15</th>
<th>15–20</th>
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</thead>
<tbody>
<tr>
<td>UV Spectrometer Readings</td>
<td>0.056</td>
<td>0.051</td>
<td>0.056</td>
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Table 3 Measured $k_{aB}$ at Various AC Loading ($F_B = 50$ mL min$^{-1}$)

<table>
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<th>AC Weight, g</th>
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<th>0.15</th>
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<tr>
<td>$k_{aB}$, s$^{-1}$</td>
<td>3.226</td>
<td>3.226</td>
<td>3.446</td>
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</tbody>
</table>