Trickle Flow
Hydrodynamic Multiplicity
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Werner van der Merwe

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“Hydrodynamics has little significance for the engineer because of the great mathematical knowledge required for an understanding of it and the negligible possibility of applying its results.”

Prandtl (1904)
ABSTRACT

Title: Trickle Flow Hydrodynamic Multiplicity
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Department: Chemical Engineering
Degree: PhD (Chemical Engineering)

Trickle flow is encountered in a variety of process engineering applications where gas and liquid flow through a packed bed of stationary solid. Owing to the complexities of three interacting phases, a fundamentally exhaustive description of trickle flow hydrodynamics has not been achieved. A complicating factor in describing the hydrodynamics is the fact that the hydrodynamic state is dependent not only on the present operating conditions but also on their entire history, including fluid flow rate changes and pre-wetting procedures. This phenomenon is termed hydrodynamic multiplicity and is the subject of this work. Hydrodynamic multiplicity greatly complicates both the experimental investigation into the behaviour of a trickle flow column and the theoretical modelling of the observed behaviour.

Broadly speaking, this study addresses hydrodynamic multiplicity on three levels. First, a conceptual framework is proposed that can be used to study hydrodynamic multiplicity with limited resources. It is based on the absolute limiting values that the hydrodynamic parameters can adopt for a certain set of conditions, and encompasses both flow rate hysteresis loops and pre-wetting procedures. There are 5 such hydrodynamic modes. When the existing literature is critically evaluated in light of this framework, it is established that the reported experimental studies have not addressed all the issues. Previous modelling
attempts are also shown to be unable to qualitative explain all the existing data. Moreover, authors have suggested different (and often contradictory) physical mechanisms responsible for hydrodynamic multiplicity.

Secondly, an experimental investigation intended to supplement the existing literature and illustrate the utility of the proposed framework is launched. This includes bed-scale measurements of liquid holdup, pressure drop and gas-liquid mass transfer for a variety of conditions including different flow rates, pressures, particle shapes, particle porosity and surface tension. The second part of the experimental effort uses radiography and tomography in new ways to visualise the temporal and spatial characteristics of the different hydrodynamic modes. The tomographic investigation incorporates advanced image processing techniques in order to culminate in a pore-level evaluation of the hydrodynamic modes that reveals additional features of hydrodynamic multiplicity.

Thirdly, the experimental insights are condensed into a set of characteristic trends that highlight the features of hydrodynamic multiplicity. A pore-level capillary mechanism is then introduced to qualitatively explain the observed behaviour. The mechanism shows how the differences in advancing and receding contact angles and the characteristics of the packed structure (or pore geometries) are ultimately responsible for the observed hydrodynamic multiplicity behaviour.

Lastly, the effect of hydrodynamic multiplicity on trickle bed reactor performance is discussed. It is established experimentally that depending on the reaction conditions, different modes yield optimal performance. The idea of optimizing the performance by manipulating the hydrodynamic state is introduced.

In totality, this work advances the understanding of trickle flow hydrodynamics in general and hydrodynamic multiplicity in particular.
Keywords: hydrodynamics, trickle flow, multiphase, hysteresis, pre-wetting, tomography, radiography, image processing, packed bed, multiplicity.
Many people have contributed to making this work possible. Thank you all.

My supervisor, Prof. Willie Nicol, for continued support and guidance, and most of all for supporting professional development opportunities.

Sasol Technology Research and Development for financial support, in particular the efforts of Arno de Klerk. The financial assistance of the National Research Foundation (NRF) towards this research is hereby acknowledged. Opinions expressed and conclusions arrived at, are those of the author and are not necessarily to be attributed to the NRF.

Prof. Muthanna Al-Dahhan and Prof. Mike Dudukovic at the Chemical Reaction Engineering Laboratory (CREL) at Washington University in St Louis, for hosting the high pressure and reaction work in sections 4.4 and 8.3. The assistance of all the other personnel at CREL is also appreciated.

Considerable assistance in generating some of the data came from fellow workers: Dylan Loudon and Ina van der Westhuisen for parts of Chapter 4, Zeljko Kuzeljevic for some of the high pressure data in section 4.3, Frikkie de Beer for technical assistance in the visualization studies of Chapters 5 and 6, and Arjan van Houwelingen, who collaborated on, amongst other things, the industrial data in section 8.1. Many thanks to Carl Sandrock, Renier Schwarzer, Arjan van Houwelingen, Dylan Loudon and Wouter de Vos for many hours of stimulating discussion on both academic and extra-curricular topics.

Finally, a debt of gratitude to Annel, who, more than anyone else, believed.

Werner van der Merwe, November 2007
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<td>$A$</td>
<td>Cross-sectional area (m$^2$)</td>
</tr>
<tr>
<td>$A$</td>
<td>Convolution target in Chapter 6</td>
</tr>
<tr>
<td>$A$</td>
<td>Ergun constant in Chapters 2 and 4</td>
</tr>
<tr>
<td>$a$</td>
<td>Hole area in Chapter 7 (m$^2$)</td>
</tr>
<tr>
<td>$a_1, a_2, a_3$</td>
<td>Size of $A$</td>
</tr>
<tr>
<td>$A_{\alpha S}$</td>
<td>Interfacial area between phase $\alpha$ and the solid (m$^2$)</td>
</tr>
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<td>$B$</td>
<td>Convolution kernel in Chapter 6</td>
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<tr>
<td>$B$</td>
<td>Second Ergun constant</td>
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<td>$b$</td>
<td>Stoichiometric coefficient</td>
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\( F \)  
Force (N)

\( f \)  
Wetting efficiency

\( f_s, f_v \)  
Shear and velocity slip factors

\( F_a \)  
F-function parameter

\( G \)  
Gas superficial flux (kg/m\(^2\)s)

\( g \)  
Gravity acceleration (m/s\(^2\))

\( Ga \)  
Galileo number

\( h \)  
Height in Chapter 7 (m)

\( h \)  
Hough-transform value in Chapter 6 (normalized)

\( H \)  
Packed height (cm)

\( I \)  
Image

\( k_{GLaGL} \)  
Volumetric gas-liquid mass transfer coefficient (1/s)

\( k_{LSaLS} \)  
Volumetric liquid-solid mass transfer coefficient (1/s)

\( k_\alpha, k_{ai} \)  
Relative permeabilities of phase \( \alpha \)

\( l \)  
Characteristic tube length (m)

\( L \)  
Liquid superficial flux (kg/m\(^2\)s)

\( M \)  
Maldistribuition factor

\( m \)  
Mass (kg)

\( n \)  
Normal vector

\( N \)  
Number of observations (Chapter 2)

\( N \)  
Number of particles in Chapter 6

\( P \)  
Pressure in Chapter 4 (Pa)

\( P \)  
Productivity in Chapter 8 (kmol/s)

\( Q \)  
Volumetric flow rate (m\(^3\)/s)

\( r \)  
Particle radius (m)

\( r_c \)  
Radial distance from central axis (pixels or m)

\( Re \)  
Reynolds number

\( S \)  
Solids image

\( T \)  
Ternary image
\[ t \quad \text{Threshold in Chapter 6} \]
\[ t \quad \text{Time (s)} \]
\[ u \quad \text{Direction normal to edge in Chapter 6} \]
\[ U \quad \text{Interstitial (or characteristic) velocity (m/s)} \]
\[ u \quad \text{Velocity (m/s or mm/s)} \]
\[ u \quad \text{Velocity vector (m/s)} \]
\[ u_{L\text{-pulse}} \quad \text{Pulsing velocity (m/s)} \]
\[ v \quad \text{Number of overlaps} \]
\[ V \quad \text{Volume (m}^3\text{)} \]
\[ W_{Pd} \quad \text{Mass palladium (kg)} \]
\[ X \quad \text{Conversion} \]
\[ x \quad \text{Generic hydrodynamic parameter in Chapter 2} \]
\[ x \quad \text{Thickness in Chapter 5 (m)} \]
\[ z \quad \text{Length (m)} \]

**Greek**

*Definition (units)*

\[ \Delta H \quad \text{Heat of reaction (kJ/kmol)} \]
\[ \Delta P/\Delta z \quad \text{Pressure drop (kPa/m)} \]
\[ \Delta s \quad \text{Image resolution (pixels or m)} \]
\[ \Omega \quad \text{Stresses} \]
\[ \Psi \quad \text{Momentum vector} \]
\[ \beta_{L/G} \quad \text{Liquid/Gas saturation} \]
\[ \delta \quad \text{Reduced liquid saturation in Chapter 2 and 4} \]
\[ \varepsilon \quad \text{Porosity} \]
\[ \varepsilon_L \quad \text{Liquid holdup} \]
\[ \phi \quad \text{Capillary correction factor for liquid in Chapter 7} \]
\[ \gamma \quad \text{Gas- or liquid limitation factor in Chapter 8} \]
\[ \lambda, \psi \quad \text{Parameters in Chapter 2} \]
\( \mu \) \hspace{0.5cm} \text{Viscosity (Pa.s)}

\( \theta, \phi \) \hspace{0.5cm} \text{Spherical voxel coordinates in Chapter 6}

\( \theta, \theta' \) \hspace{0.5cm} \text{Contact angle and bottom contact angle in Chapter 6}

\( \rho \) \hspace{0.5cm} \text{Density (kg/m}^3\text{)}

\( \rho_i \) \hspace{0.5cm} \text{Intensity density (1/m}^3\text{)}

\( \sigma \) \hspace{0.5cm} \text{Surface tension (N/m)}

\( \sigma_\alpha \) \hspace{0.5cm} \text{Tension vector (N/m)}

\( \psi \) \hspace{0.5cm} \text{Capillary correction factor for gas in Chapter 7}

\textbf{Subscripts}

0, o \hspace{0.5cm} \text{Reference}

A \hspace{0.5cm} \text{Gas phase reagent in Chapter 8}

B \hspace{0.5cm} \text{Liquid phase reagent in Chapter 8}

crit, rec \hspace{0.5cm} \text{Critical, receding (Chapter 7)}

G, L, g, l \hspace{0.5cm} \text{Gas, liquid}

i, j, k, m, n, p \hspace{0.5cm} \text{Indices in Chapter 6}

p \hspace{0.5cm} \text{Particle}

res \hspace{0.5cm} \text{Residual}

sub \hspace{0.5cm} \text{Substrate}

T \hspace{0.5cm} \text{Total}

v \hspace{0.5cm} \text{Volumetric}

\( \alpha \) \hspace{0.5cm} \text{Indicates phase } \alpha

\beta, \gamma \hspace{0.5cm} \text{Indicates liquid and gas phases in Chapter 2}