UNIFORM BUBBLE GROWTH IN FLUIDIZED BED AND ITS STABILITY ANALYSIS
Chenxi ZHANG*, Weizhong QIAN, Fei WEI
Department of Chemical Engineering, Tsinghua University,
Beijing, China
*Email: zhangcx13@mails.tsinghua.edu.cn

Abstract – The uniform bubble distribution is ensured to gain sufficient performance of dense fluidized bed reactors. Although distributor plays an important role in obtaining homogenous and small bubbles, the highly inelastic energy dissipation from solids bed triggers non-uniform bubbles growth. Hence, the total solids pressure loading ($\Phi_T$) is introduced here to present the interaction between contribution from solids bed and distributor. The boundary between uniform and non-uniform bubble growth can be detected by criterion based on Prigogine’s minimum of the entropy production principle. Furthermore, an industrial case is studied further based on stability analysis and new operational conditions are provided to improve the performance through eliminating non-uniform bubble growth.

INTRODUCTION
Consider gas solids fluidization that begins in an initial state of uniform density where gas is evenly introduced into the base of the bed via distributor. The solids then “cools” as the collisions dissipate the kinetic energy of the initial condition, and the cooling solids do not remain homogenous, i.e. the state of uniform density is unstable so that the system totally exhibits the coexistence of two phases: clusters and bubbles. Thus, uniform gas solids distribution, resulting from the distributor, will be damaged by solids beds itself and this process is self-aggregating until the size of large bubbles reaches the bed diameter1-4. Recent studies have shown that non-uniform bubble growth even slugging may occur in a sufficiently deep bed, as exemplified in Fig. 1. The result of this phenomenon will resist mass transfer and bypass reactant in the form of large bubbles or slugs, leading to poor conversion and selectivity in reactors. Therefore, a deep fluidized bed is a fundamental model in uniformity breaking mechanics and should be investigated carefully to ensure the performance of fluidized bed reactors.

STABILITITY ANALYSIS
The distribution of bubbles is distributed uniformly through the distributor, while the uniformity is broken due to inelastic energy dissipation from solids beds. Therefore, in order to describe the multi-phase interaction in dense fluidization, the dimensionless number total solids pressure ratio ($\Phi_T$), i.e. the pressure ratio between solids bed and distributor, is introduced here:

$$\Phi_T = \frac{\Delta P_s}{\Delta P_d}$$

(1)

Apparently, deep fluidized bed or low gas velocity have large value of solids pressure loading while $\Phi_T$ is small in the case of shallow bed or high gas velocity. The regime transitions can be attributed from pressure ratio because it is a pseudo-stable intermediate balance between particle collision and uniform gas
suspension. After this point, the gas-solid interaction in beds dominates the system. The critical total solids pressure loading \((\Phi_{T_c})\) is the solution of the following non-linear equation:

\[
\det(H_\gamma) = \frac{\partial^2 \Delta P}{\partial \Phi_\gamma} = 0
\]  

(2)

The result of the Equation (2) is \(\Phi_{T_c} = 6.1\) where \(\gamma_1 = \gamma_2\) and the phase diagram of stability of uniformity is shown in Fig. 2. When \(\Phi_T < \Phi_{T_c} = 6.1\), the contour of multivariate surface is concave, indicating the surface is convex. Thus, uniform bubble growth is stable. While the solutions of uniformity is stable when \(\Phi_T > \Phi_{T_c} = 6.1\), where the contour of multivariate surface is convex meaning the surface is concave. The contour becomes straight when \(\Phi_T = \Phi_{T_c} = 6.1\), the point of inflection from concave to convex surface, namely the turning point from uniformity to mal-distribution.

![Fig. 2. (a) The global total pressure drop surface; (b) The contour of the pressure drop surface.](image)

**CASE STUDY**

In this study, the reaction of preparing \(p\)-chlorobenzonitrile (PCBN) by ammonoxidation of \(p\)-chlorotoluene (PCT) is used as the case study. The VPO/SiO\(_2\) catalyst is prepared by impregnation method. The industrial unit is carried out in a flow apparatus equipped with a fluidized bed reactor (i.d.: 1.2 m, length: 20 m), feeding section, cooling and product separation units, as shown in Figure 10. The gaseous stream is preheated and then introduced to the reactor. The product stream from the catalytic reactor consists of unconverted feed components (i.e. PCT, oxygen and ammonia) and the reaction products and by-products (e.g. \(\text{NH}_4\text{Cl}\) formed by PCT total oxidation).

\[
\text{ClPhCH}_3+\text{O}_2+2\text{NH}_3 \rightarrow \text{PhCN}+2\text{H}_2\text{O}+\text{NH}_4\text{Cl}
\]  

(3)

![Fig. 3. (a) Pressure fluctuation profile and (b) temperature profile along the industrial fluidized bed; the operational conditions are listed in Table 1.](image)
The variation of bed pressure drop and temperature profiles with time, as shown in Fig. 3, is employed to establish the analysis of quality of fluidization. Fig. 3(a) shows the different type of pressure fluctuation in Case A and Case B. Apparently, the pressure profile in Case A, in which the bed depth is beyond the critical point, is low frequency and large amplitude due to large bubbles formation. On the contrary, high frequency and small amplitude of pressure fluctuation in Case B indicates better quality of fluidization where numerous small bubbles exist. Fig. 3(b) depicted the temperature profile alone the bed. In the reaction zone, the temperature is set to 410°C. Interestingly, the temperature distribution in Case A is much more uniform than that in Case B. Generally, more uniformity of temperature results from better gas solids interaction. However, because mass transfer resistance increases with the increasing bubble size and the residence time is not enough due to high velocity of large bubbles in deep fluidized bed, the heat production in Case A is dramatically limited. On the contrary, the fluidized bed gains super heat and mass transfer, the non-uniform temperature distribution is reasonable due to fully heat production in Case B.

Table 1. Operational Conditions for Case A and Case B

<table>
<thead>
<tr>
<th>Units</th>
<th>Case A</th>
<th>Case B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed</td>
<td>PCT: NH₃: Air</td>
<td>PCT: NH₃: Air</td>
</tr>
<tr>
<td>Diameter of reactor, D</td>
<td>m</td>
<td>1.2</td>
</tr>
<tr>
<td>Total mass of catalyst</td>
<td>kg</td>
<td>7000</td>
</tr>
<tr>
<td>Static depth of bed, H</td>
<td>m</td>
<td>7.29</td>
</tr>
<tr>
<td>Height-to-Diameter ratio, H/D</td>
<td>~6</td>
<td>~4</td>
</tr>
<tr>
<td>Feeding temperature</td>
<td>K</td>
<td>483</td>
</tr>
<tr>
<td>Operating pressure</td>
<td>kPa</td>
<td>101.3</td>
</tr>
<tr>
<td>Superficial gas velocity, U</td>
<td>m/s</td>
<td>0.3</td>
</tr>
<tr>
<td>Total solids pressure loading, Φ_T</td>
<td>8.6</td>
<td>5.3</td>
</tr>
</tbody>
</table>

Table 1. Operational Conditions for Case A and Case B

The values of conversion and selectivity are illustrated in the Fig. 4. The conversion in Case A is much lower than that in Case B. The main reason for this poorer performance is found in the presence of non-uniform bubble growth. Large bubbles form an additional resistance to mass transfer between reactant gas and catalyst particles. On the other hand, large bubbles usually have a high rise velocity, thus little time is left for mass transfer. Both effects, which increase with increasing bubble size, lead to unwanted bypass of reactants in Case A. In addition, selectivity of complex catalytic reactions usually depends on the mass transfer properties of the reactor in which the reaction takes place. Consider the consecutive reaction (Cl-Ph-CH₃ to Cl-Ph-CN to NH₄Cl) and suppose mass transfer to and from the catalyst particles is slow; Conversion will generally be low and selectivity will be in favor of the unwanted product NH₄Cl. Therefore, the selectivity of complex reactions in gas solids fluidized bed reactors depends not only on catalyst properties, but to a large extent on the mass transfer properties, i.e. bubble size of the fluidized bed as well. In conclusion, by preventing uniform bubble growth in dense fluidization, both conversion and selectivity of mass transfer limited catalytic reaction in dense gas solids fluidized reactors can be improved.

Fig. 4. The performance of ammoxidation in fluidized beds for Case A and Case B; The operational condition is listed in Table 1.
The fluidization quality of a bed is highly dependent on the distribution of bubbles. Ideally, for there to be good quality fluidization, the population of bubbles in the bed should be large, but the bubbles should be small in size, and homogeneously occupy the location of the bed. Many models have been developed for residence time distribution (RTD) fluidized bed applications involving heterogeneous catalytic reactions. The reaction, in this case on contact with the solid, takes place primarily in the dense phase. Furthermore, because of the intensive motion of particles and the effective gas solids interaction, the residence time across bed may reasonably be assumed proportional with the bed depth. However, in the case of non-uniform fluidized beds, this assumption is no longer justified because the amounts of reactants escape through bypassing channel. Accordingly, the residence time may drop down when the bed depth is beyond the critical point. Bubble induces heterogeneity and the gas within them rises through the bed without much interaction with solids phase. As deep bed generates non-uniform bubbles where bypassing of reactants is obtained, process efficiency and heat and mass transfer can be severely affected.

**NOTATION**

- $U_g =$ superficial gas velocity, m/s
- $\Delta p =$ pressure drop, N/m$^2$
- $\gamma_i =$ mass fraction of gas flowing through orifice $i$

**REFERENCES**