Chemical Engineering Journal 82 (2001) 259–266

Chemical Engineering Journal

www.elsevier.com/locate/cej

The staging in fluidised bed reactors: from CSTR to plug-flow

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Received 30 May 2000; accepted 25 October 2000

Abstract

Widespread application of bubbling fluidised bed reactors to chemical processes has been hindered by some inherent drawbacks, like the high degree of internal mixing and the low contact efficiency between gas and solid phases. The staging of fluidised beds through the insertion of horizontal baffles may overcome these drawbacks, approaching plug-flow and limiting bubble size. The authors, based on theory and experiments, have developed a methodology to properly design staged fluid bed catalytic reactors, considering gas and solid backmixing, bubble size, flooding and dynamic forces, which is described in the paper.

Keywords: Plug-flow; Fluidised bed reactors; Baffles

1. Introduction

The phenomenon of fluidisation, i.e. the capability of fine powders to be suspended by an upward flowing fluid has been widely applied to the commercial exploitation of catalytic reactions.

The first applications of fluidised bed technology to catalytic reactions took advantage of the chance to continuously transfer large fluxes of aerated solids between two adjacent vessels in cases where periodical regeneration of the catalyst was needed. This is the typical case of fluid catalytic cracking (FCC) technology in which coke, formed as a by-product in the reactor, is burnt in the regenerator.

Another advantage of fluidised bed reactors is the high bed-to-wall heat transfer coefficient allowing safe and efficient exploitation of very exothermic reactions (e.g. synthesis of acrylonitrile). Circulating catalyst may also be used as heat carrier for endothermic reactions (e.g. FBD technology for dehydrogenation of light paraffins or FCC itself), transferring hot catalyst from the regenerator exploiting the good gas–solid heat transfer coefficient of fluidised beds.

However, in spite of these advantages, a number of drawbacks compared to fixed bed reactors still hinder the development of economically viable fluid bed technologies.

High degree of internal mixing is the main disadvantage, causing fluid bed reactors to approach a completely mixed reactor (CSTR) rather than a plug-flow. This is particularly important for the exploitation of equilibrium-limited reactions or when a narrow distribution of gas phase residence times is needed to minimise the formation of by-products. This phenomenon is worsened by the multiphase nature of bubbling fluidised beds where most of the gas flows in the form of "bubbles" or "channels" and only a very low amount is contacting catalyst particles in the dense phase (emulsion) in each moment. Consequently, the performances of a fluidised bed reactor may be much worse than a CSTR of the same volume.

This drawback was partly by-passed by the development of circulating fluid bed technologies in which high gas superficial velocities necessary to achieve pneumatic transport of the catalytic particles result in a close approach to gas phase plug-flow and in a single phase system in which solid particles are uniformly dispersed in the up-flowing gas.

However, circulating fluid bed technologies based on riser reactor may be implemented only when very active catalysts are available (gas residence time may not be higher than a few seconds) and when adiabatic ΔT of the reaction is not so high to bring the exit temperature outside the range of technical feasibility.

In bubbling fluidised bed reactors plug-flow of the gas phase may be approached by the introduction of internals limiting the length of the vertical path for axial mixing reducing available cross-section for the passage of gas and solid at various levels.

Next chapters will deal with proper design of these internals to control gas and solid backmixing and bubble size, also taking into account the dynamic forces exerted on them by the fluidised bed itself.

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1385-8947/01/$ – see front matter
PII: S1385-8947(00)00341-7

Nomenclature

A	area of element immersed in a fluidised bed (m^2)
A_{el}	projected area of element immersed in the bed in a direction perpendicular to gas flux (m^2)
A_{tot}	vessel cross-sectional area (m^2)
B_f	frequency of bubbles impinging upon an element immersed in the bed (s^{-1})
D_b	bubble diameter (m)
F	vertical force acting upon an element immersed in a fluidised bed (N)
g	gravitational constant: 9.81 m/s^2
G_{n-1}, G_n	upward gas flowrate leaving ($n-1$)th and nth stage (kg/s)
$G_{b,n+1}$, $G_{b,n}$	gas backmixing flowrate leaving ($n+1$)th and nth stage (kg/s)
J	solid mixing flux (kg/m^{2}s)
k, K	proportionality constants
K_f	frequency constant ($m^{0.5}$/s)
L	height of a fluidised bed volume element (m)
N_b	number of bubbles in a given cross-section of the fluidised bed
N_{bt}	number of bubbles impinging on A_{el} in unit time (s^{-1})
N_{bv}	number of bubbles in a given volume of fluidised bed (m^{-3})
$S_{b,n-1}$, $S_{b,n}$	solid backmixing flowrate leaving ($n-1$)th and nth stage (kg/s)
S_{n+1}, S_n	downward solid flowrate leaving ($n+1$)th and nth stage (kg/s)
U	gas superficial velocity (m/s)
U_b	bubble rising velocity (m/s)
U_{mf}	minimum fluidisation velocity (m/s)
Y	ratio between theoretical and actual volumetric flowrate of bubbles

Greek letters

α	surface shape factor
β_d, β_w	fraction of solids carried up by a bubble within, respectively, its wake and drift
ρ	fluidised bed density (kg/m^3)
ρ_D	density of dense phase (emulsion) (kg/m^3)
ρ_{mf}	minimum fluidisation density (kg/m^3)

2. The staging of fluidised beds

In analogy with gas–liquid separation systems (e.g. distillation columns), the staging in fluidised beds could be achieved by the insertion of sieve or valve trays with downcomers discharging the solid phase to the tray below. However, the dense emulsion phase is inherently unstable, since the gas tends to flow upward leaving a de-fluidised solid phase. This could cause local de-fluidisation in the downcomer zone and poor contact between the phases, also considering that radial mixing is much slower than axial mixing [1].

A simpler option, providing no preferential passages either for lean or dense phase is represented by grid trays realised with tube or chevron elements equally spaced across the whole reactor cross-section [2]. A statistically uniform distribution of gas and solid flows is assured if the gas phase is properly distributed in the bottom of the bed and the solid phase is well distributed in the top of the bed. Moreover, the width of the holes or slots allowing gas and solid passage between two adjacent stages should be small enough compared to the total open area of the tray to avoid by-pass or channelling phenomena.

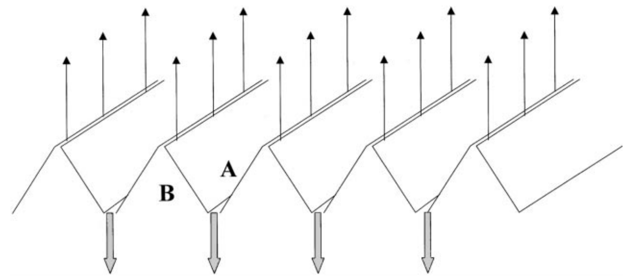

Fig. 1. Section of a baffle with separate passages for gas and solid.

A step forward in the development of baffle systems for the staging of fluidised beds is the introduction of separate passages for gas and solid flow [3]. These passages must be very short to avoid deaeration of the dense phase. An arrangement of a chevron grid with separate passages for gas and solid is shown in Fig. 1. The gas phase flows preferentially upward through the holes on top of the chevron, while solid particles flow downward through the slots at the base of the chevron, as demonstrated by large scale cold model experiments. In fact, a denser phase tends to build-up in zone A in Fig. 1, increasing the pressure drop of the upward gas phase, while zone B tends to be void of solids. The vertical zone in which the two fluxes are separated is equal to the height of the baffle.

3. Effects of staging

Different approaches have been proposed to describe the behaviour of bubbling fluidised beds. Two-phase models consider the presence of a leaner dispersed phase (bubble) and a denser continuous phase (emulsion), mainly differing for the shape of the bubbles, the distribution of gas and solid between the phases and the type of flow (plug-flow, well mixed or intermediate for each phase). Three-phase models consider the presence of a third-phase (wake or cloud), with the bubbles having a concave base below which the

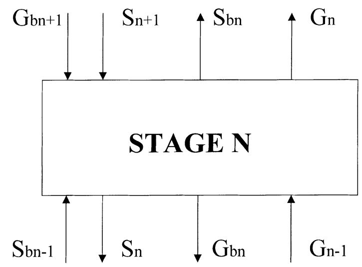

Fig. 2. Material balance around a stage in the reactor.

third-phase (of intermediate density between bubble and emulsion) is mainly located.

However, it is known from the literature [2] that in a freely bubbling bed the experimental gas residence time distribution approaches quite closely the theoretical curve calculated considering a single stage in which mass transfer takes place between two well mixed phases (bubble and emulsion), independently from gas superficial velocity. Based on the above considerations and for the sake of simplicity, a two-phase approach with spherical bubbles was considered suitable to simulate the behaviour of a freely bubbling bed by known mathematical techniques.

Considering now a staged fluidised bed reactor with countercurrent flow of gas and solid, the overall weight material balance relative to a single nth stage (i.e. the volume between two consecutive baffles n and $n+1$) may be expressed, with reference to Fig. 2, as follows.

$$\text{Gas phase } G_{n-1} + G_{\mathrm{b},n+1} = G_n + G_{\mathrm{b},n} \tag{1}$$

where G_{n-1} and G_n represent the upward gas flowrates, respectively, entering and leaving nth stage, while $G_{\mathrm{b},n+1}$ and $G_{\mathrm{b},n}$ represent the downward gas flowrates caused by backmixing phenomena, respectively, entering and leaving nth stage.

$$\text{Solid phase } S_{n+1} + S_{\mathrm{b},n-1} = S_n + S_{\mathrm{b},n} \tag{2}$$

where S_{n+1} and S_n represent the downward solid flowrates, respectively, entering and leaving nth stage, while $S_{\mathrm{b},n-1}$ and $S_{\mathrm{b},n}$ represent the upward solid flowrates caused by backmixing phenomena, respectively, entering and leaving nth stage.

Consequently, for proper design and optimisation of a staged fluidised bed reactor, the following variables need to be defined:

- The extent of gas backmixing ($G_{\mathrm{b},n}$ and $G_{\mathrm{b},n+1}$ in Eq. (1) and in Fig. 2), i.e. the flowrates of gas flowing downward with the solid between the stages.
- The extent of solid backmixing ($S_{\mathrm{b},n}$ and $S_{\mathrm{b},n-1}$ in Eq. (2) and in Fig. 2), i.e. the flowrates of solid entrained upward between two stages.

Additional information needed to design a staged fluidised bed reactor regards the ability of baffles to control the size of gas bubbles and the approach to flooding in case of countercurrent flow of the two phases.

3.1. Gas backmixing

The determination of gas backmixing is of utmost importance to allow prediction of chemical reaction performances. Gas backmixed flowrate may represent a significant deviation from the overall plug-flow of the gas phase (achieved through a series of mixed stages), affecting both reactant conversion and selectivities to the main product and by-products. In principle gas backmixing should be minimised, especially when dealing with equilibrium-limited reactions.

Published data [2] show that open area (defined as the ratio between cross-sectional area free for the passage of fluids and total cross-sectional area) and slot width affect the capability of a baffle to limit gas backmixing. Constriction velocity (i.e. gas velocity through the openings of a baffle) seem to play a role only beyond a certain limit, presumably when flooding conditions are closely approached.

A series of experiments was performed by the authors to assess the influence of the above cited features and other operating parameters on gas backmixed flowrate, using a large scale acrylic cold model (rectangular cross-section 140 cm × 30 cm, total bed height of about 300 cm with four stages created by inserting three horizontal baffles) with air feeding at the bottom and controlled catalyst circulation flowrate (a group A powder according to the classification by Geldart [4]). Once the bed was steadily fluidised, helium was injected at a certain level, and gas samples were collected in each stage. The percentage of helium contained in each sample allowed the determination of the amount of gas backmixing for each experiment.

The main experimental results are represented in Figs. 3–5 in which the gas backmixing flowrate is reported in the ordinate axis in arbitrary units, maintaining the correct proportionality between the various points. Each figure shows the effect of a single variable, all of the others being kept constant. Particularly gas superficial velocity was maintained constant for all data reported in a single figure. In the investigated field, the baffle open area percentage was found to have the strongest effect on gas backmixing, as reported in Fig. 3. Increasing baffle open area, gas backmixing tends to increase exponentially, so that a limiting open area may be defined, below which staging is so effective that chemical performances are not affected. Of course, for further increase in baffle open area, gas backmixing is expected to increase less, because the limiting situation (absence of baffles) is approached. Slot width is also affecting gas backmixing as shown in Fig. 4, where arbitrary lengths are reported in the abscissa (actual values are in the order of the tens of millimetres): smaller slots or holes, maintaining the same open area percentage decrease backmixing. Another variable affecting gas backmixing is the percentage of fine particles in the catalyst (diameter $<45\,\mu\text{m}$) (Fig. 5).

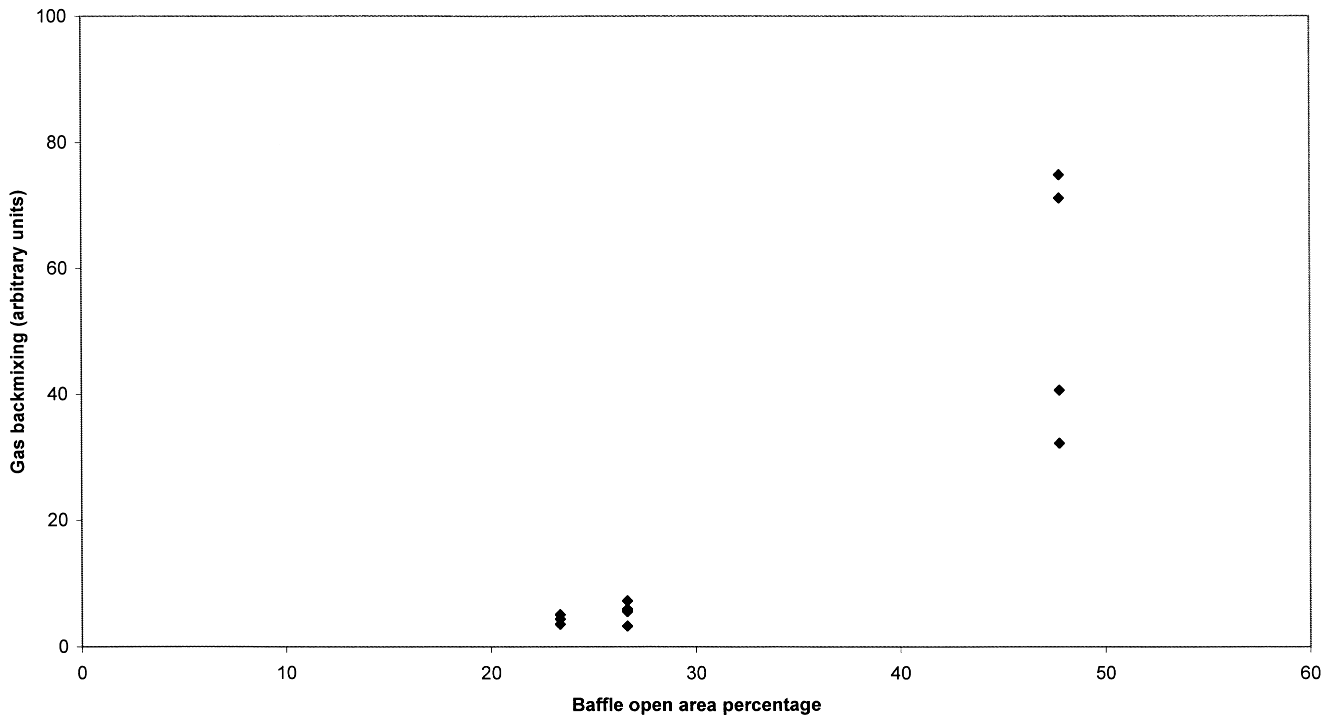

Fig. 3. Gas backmixed flowrate vs. baffle open area.

An increased percentage of fines causes an increased backmixing. This may be related to the ability of fine particles to retain more gas in the dense phase compared to larger particles.

Experiments performed varying either feed gas flowrate or solid circulation flowrate did not reveal any influence of these parameters in the investigated range.

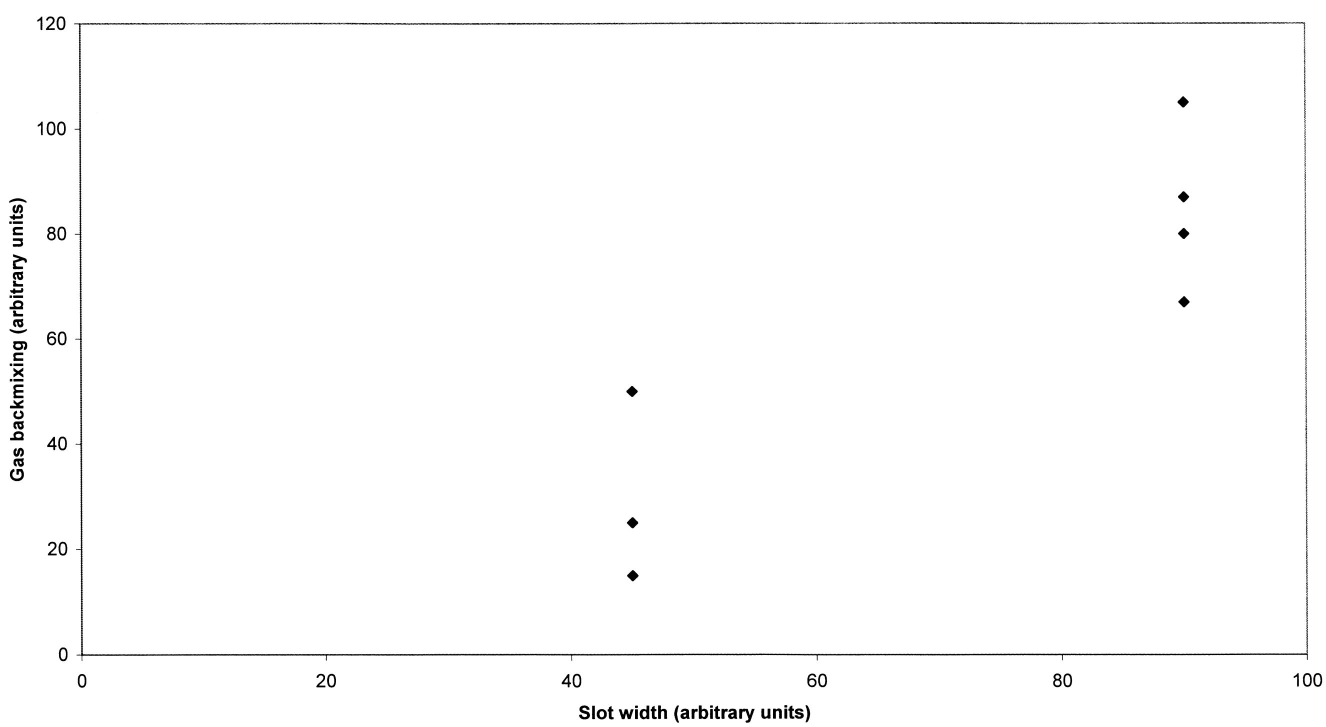

Fig. 4. Gas backmixed flowrate vs. baffle slot width.

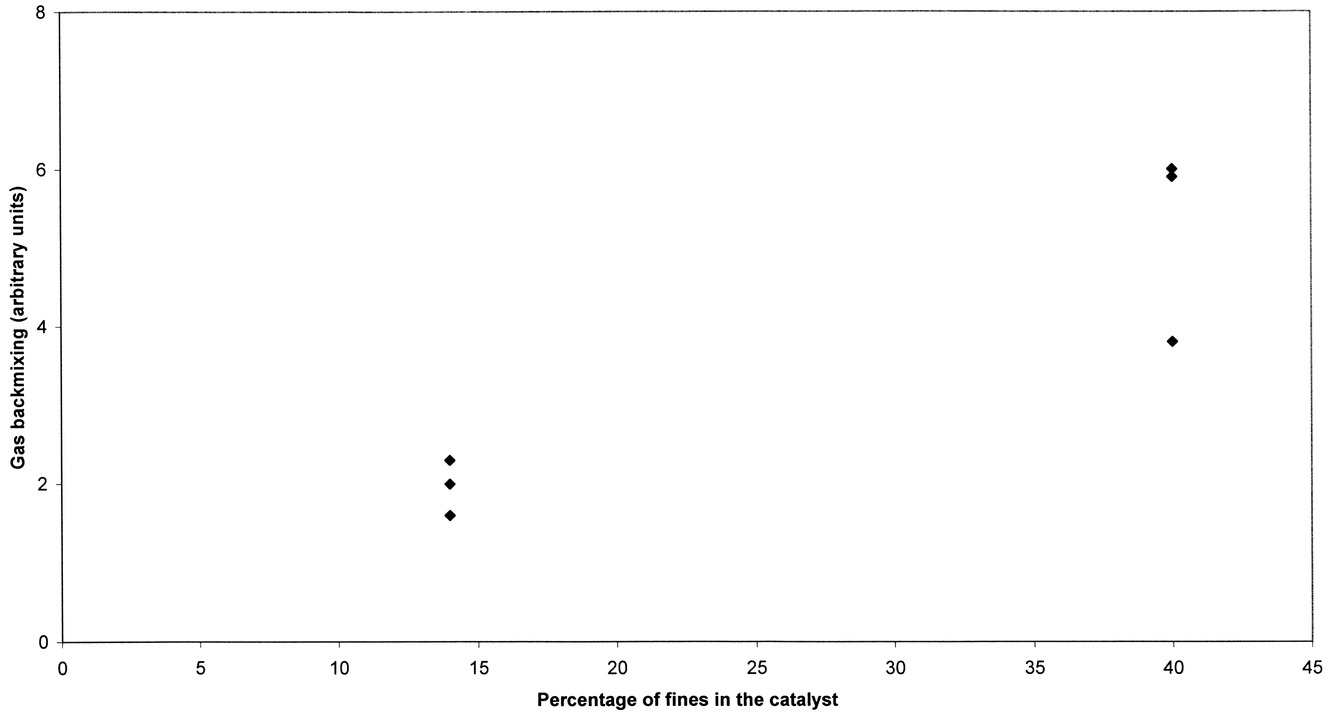

Fig. 5. Gas backmixed flowrate vs. fines content in the solid phase (<45 μm).

3.2. Backmixing of solids

Calculation of the solid flux entrained to the upper stage is important to define the reactor axial temperature profile, particularly in the case of endothermic reactions where the regenerated catalyst also acts as heat carrier to the reaction itself. In this case the achievable axial temperature difference along the reactor may be directly related to the degree of staging achieved by the introduction of baffles. Baffles are less effective in limiting solid backmixing than gas backmixing [2].

According to [5] the solids mixing flux in a freely bubbling bed is proportional to gas superficial velocity and solid density.

$$\text{Mixing flux } J = \rho_D Y (U - U_{mf})(\beta_w + 0.38\beta_D) \quad (3)$$

where U is the superficial gas velocity, U_{mf} the minimum fluidisation velocity, ρ_D the bulk density of the dense phase, β_w and β_D are wake and drift fractions, respectively, and Y is the correction factor for the two-phase theory (ratio between theoretical and actual gas flowrate in bubbles).

The above correlation was modified to take into account the limitation to mixing given by the presence of a horizontal baffle, including baffle open area and slot width. Finally a multiplying efficiency factor was added so that experimental temperature profiles measured in pilot and commercial units could be well simulated.

3.3. Flooding

In analogy with gas–liquid systems, in case of countercurrent flow in fluidised beds there is a maximum flux achievable for one of the phases once the flux of the other phase has been fixed. This phenomenon must be carefully considered when baffles are inserted in a fluidised bed reactor. A lower baffle open area reduces all backmixing phenomena as shown above, but leads to a closer approach to flooding. A correlation to determine flooding conditions in fluidised beds was developed by Zenz and co-workers [6] and criteria for its application to fluidised beds were published later [7].

The correlation was validated by experimental tests performed on large scale acrylic cold model in which the achievement of flooding conditions could be easily visualised.

3.4. Effects of baffles on bubble size

Bubble size is a key parameter in the design of a fluidised bed catalytic reactor. In fact catalytic reaction takes place in the dense emulsion phase where all or most of the solid phase is present, while most of the gas remains inside the bubbles in each moment. Interphase mass transfer coefficient of the gas phase between bubble and emulsion is roughly inversely proportional to the bubble diameter [8]. Moreover, bubble rise velocity is proportional to the square root of bubble diameter. Consequently, larger bubbles have less efficient mass transfer and escape more quickly, also affecting bed density. It is therefore, desirable to maintain bubble size as small as possible. In a freely bubbling bed bubbles grow until they reach the top or their maximum stable size.

The presence of baffles, forcing bubbles to pass through small holes or slots, breaks or slows down bubbles, improving gas–solid contact. If the openings in the baffle are in the form of holes the bubble is broken at the outlet of each stage and a new bubble is formed in the upper stage, whose initial size and growth rate may be calculated by available literature correlations, e.g. [5]. When the holes are in the form of slots, the effect of bubble breaking is more uncertain, since bubbles may "slide" through the slots modifying their shape. In any case, the presence of the obstacles represented by the baffles slows down the bubbles increasing gas residence time in the bed, as proven by calculations based on experimental density profiles in pilot and commercial reactors.

4. Evaluation of dynamic forces in a fluidised bed

Every fluid bed internal is subjected to dynamic forces exerted by the catalyst displaced by the rising bubbles. A calculation of these forces is necessary to properly design the internals avoiding damages that may also affect reactor performances.

The vertical forces transmitted by a fluid with ρ density and v velocity onto a surface of A area are equal to

$$F = \alpha\rho v^2 A \tag{4}$$

where α is a coefficient related to the shape of the surface.

Referring to a fluidised bed, the density of the fluid may be considered equal to the emulsion density (i.e. the minimum fluidisation density, as widely reported in [9]). If the fluid bed under examination is shaken by a gas bubble, in principle it can be written that:

$$F = \rho_{\mathrm{mf}} U_{\mathrm{b}}^2 A\alpha = \rho_{\mathrm{mf}} K g D_{\mathrm{b}} A\alpha \tag{5}$$

where D_{b} is the bubble diameter and the bubble rising velocity may be expressed as

$$U_{\mathrm{b}} = k(gD_{\mathrm{b}})^{0.5} \tag{6}$$

The surface area A is defined as the projected area of the internal element under consideration (A_{el}) when the impinging bubble is larger than the element. In the most common case in which bubbles are smaller than the element

$$A = \frac{\pi}{4(1.5D_{\mathrm{b}})^2 N_{\mathrm{b}}(A_{\mathrm{el}})} \tag{7}$$

where $N_{\mathrm{b}}(A_{\mathrm{el}})$ is the number of bubbles which may at any time strike the element, assuming homogeneous dispersion of the bubbles in the bed that will be examined later on. The multiplying coefficient 1.5 derives from the inclusion of the "cloud" observed by Botterill and other authors [10]. $N_{\mathrm{b}}(A_{\mathrm{el}})$ can be assessed by the following volume balance, considering that all the gas is rising in the form of bubbles:

$$A_{\mathrm{tot}} U = \frac{N_{\mathrm{b}}\pi}{4D_{\mathrm{b}}^2 U_{\mathrm{b}}} \tag{8}$$

where A_{tot} is the cross-sectional area and U the superficial gas velocity. Hence, combining with Eq. (6) the bubbles per m^2 can be calculated as

$$\frac{N_{\mathrm{b}}}{A_{\mathrm{tot}}} = \frac{4U}{\pi D_{\mathrm{b}}^2 U_{\mathrm{b}}} = \frac{KU}{D_{\mathrm{b}}^{2.5}} \tag{9}$$

Therefore, the number of bubbles impinging at the same time on the A_{el} area element will be

$$\frac{N_{\mathrm{b}} A_{\mathrm{el}}}{A_{\mathrm{tot}}} = \frac{KUA_{\mathrm{el}}}{D_{\mathrm{b}}^{2.5}} \tag{10}$$

The above can be further developed in order to assess the characteristic frequency to be linked to the force calculated earlier. The assumptions are the same as specified above.

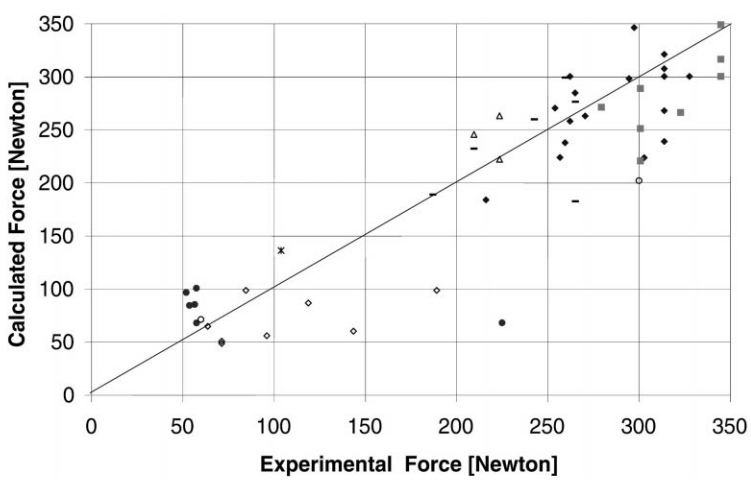

Fig. 6. Calculated vs. experimental forces (N) measured in cold model tests performed by the authors.

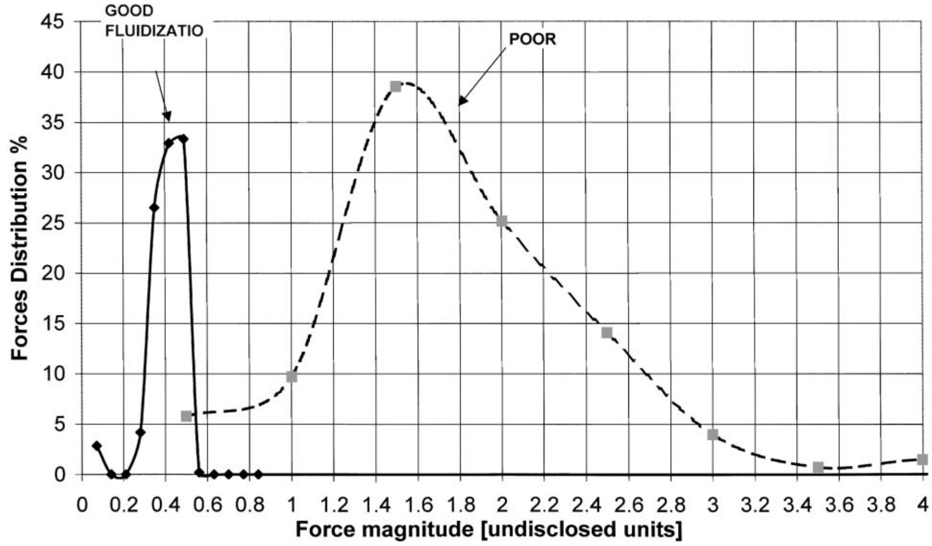

Fig. 7. Distribution of forces calculated from pressure fluctuation data in a large size vessel for two cases: good gas distribution and poor gas distribution.

- Bubbles homogeneously distributed.
- All bubbles have D_b size.
- Bubble velocity equal to Eq. (6).

Considering now a volume element of height L in the fluidised bed, the gas volumetric flowrate through that element will be equal to the ratio between the volume and the residence time of bubbles in the element. Consequently, N_{bv}, i.e. the number of bubbles present in the volume at a certain moment is equal to

$$N_{bv} = \frac{A_{tot} UL}{\pi/6 D_b^3 U_b} \tag{11}$$

Consequently, the number of bubbles impinging on the internal element of area A_{el} in the unit time will be

$$N_{bt} = N_{bv} \frac{A_{el}}{A_{tot}} \frac{L}{U_b} \tag{12}$$

The frequency B_f at which the bubbles impinge on the element is equal to the ratio N_{bt}/N_b, from Eq. (12) and Eq. (9) under the hypothesis that $N_b > 1$, results in the following equation

$$B_f = \frac{K_f}{D_b^{0.5}} \tag{13}$$

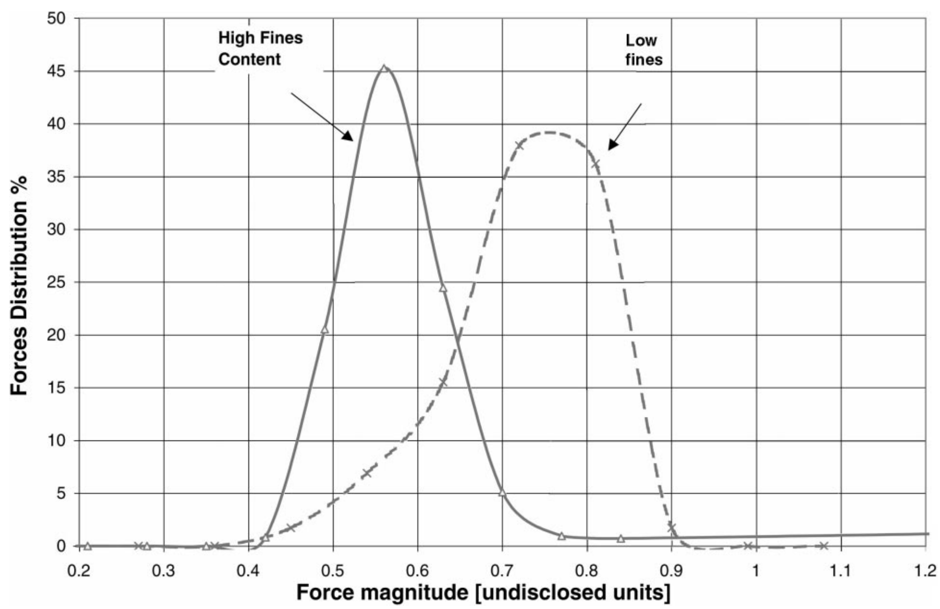

Fig. 8. Distribution of forces calculated from pressure fluctuation data in a large size vessel for two cases: high content of fine particles and low content of fine particles.

Forces and frequencies calculated with the method described above are obtained from the assumption of homogeneous distribution of equally sized bubbles. The inevitable deviation from the assumed hypotheses will lead the system to show a spectrum of force/frequency couples, as already illustrated in the literature [11].

It is also necessary to point out that the frequency of the impulses can be equal to double the bubble frequency, because a bubble seems to be able to cause two impulses when it passes through the baffle. This seems to be confirmed both from the theoretical flow fields and those experimentally observed by some authors [12]. Both impulses seem to have an upward direction, although, according to the tests conducted by some researchers [13] some forces have a downward direction after the passage of the bubble. Application of the above explained method could fairly fit cold model data produced by different authors. Calculated versus experimental data produced by the authors of this paper using air as fluidising medium at ambient conditions while varying gas superficial velocity, baffles geometric features and solid physical properties, are represented in Fig. 6. Fig. 7 shows the effects of the fluidisation quality (i.e. the effectiveness of gas distribution) on the dynamic forces in a large size vessel: a good gas distribution results in forces of much lower magnitude and spread. Fig. 8 shows the effect of fines content in the catalyst. The effect is similar to the one described above: a higher content of fines reduces both the magnitude and the spread of the forces acting on the internals.

5. Conclusions

The staging of bubbling fluidised bed reactors through the insertion of horizontal baffles may overcome some drawbacks inherent in fluidised bed technologies and currently limiting their fields of application. Particularly, the mixing of gas and solid may be limited to the extent that a plug-flow is closely approached, and the size of gas bubbles may be controlled to maximise the efficiency of gas–solid contact and reduce the dynamic forces acting on the internals.

Through the theoretical correlations and regression of cold model, pilot and commercial reactor data above cited, a method has been developed to design the system of baffles in a fluidised bed reactor, defining their type, number, open area percentage, slot or holes width, etc. to maximise achievable performances, while taking into account flooding phenomena and the effect of dynamic forces.

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