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Horizontal gas mixing in the distributor region of a fluidised bed reactor

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Abstract

The gas lateral mixing in the distributor region of a fluidised bed is studied by means of injecting a tracer axially in the cold model of an oxychlorination reactor. The horizontal spread of the tracer concentration is also related to the fluidisation regime, which is characterised by pressure fluctuation measurements.

The tracer concentration at the bed wall increases with the fluidising velocity and reaches its maximum when *turbulent fluidisation* is approached. The experimental observations are well explained by the model proposed by Rowe [1] to characterise gas flow in the *entry region* of a fluidised bed chemical reactor. ©2000 Elsevier Science S.A. All rights reserved.

Keywords: Fluidised bed reactor; Gas lateral mixing; Turbulent fluidisation

1. Introduction

The combination of chemical kinetics with effective ways of bringing reactant elements into contact with one-other lies at the core of chemical reaction engineering. The understanding of these processes and the elaboration of physical models that agree reasonably well with the actual behaviour is particularly crucial in the *entry* region of a gas fluidised bed chemical reactor, where the overall gas flow rate is split into bubbles, almost empty of catalyst particles, and the interstitial region of the so-called *emulsion* or *dense* phase.

It has been demonstrated [2] that in the shallow zone at the bottom of a fluidised bed reactor the chemical conversion is much more rapid than in the rest of the bed: in some cases, 50% or more conversion takes place in a layer of the order of 10 cm deep.

Several models have been proposed, combining kinetic and fluid dynamic aspects, with the object of predicting accurately the particular features of fluidised bed reactors [3–5]. These make use of different assumptions, and therefore may be appropriate in different cases. However, some model parameters are difficult to predict or observe.

1.1. The Rowe model for gas flow in the distributor region

A very simple approach has been suggested by Rowe [1] for describing the gas behaviour in the distributor region,

based on the experimental evidence gathered by means of X-ray ciné photography over a wide range of operating conditions [6].

A considerable gas leakage towards the emulsion phase is postulated to explain the mechanism of bubble formation and growth. Close to the distributor, as gas flows past the particles, it exerts a drag force on them, moving them far from the orifice thereby creating empty space for the growing bubble; the total amount of gas percolating into the dense phase of the bed is estimated as about two-thirds of the overall incoming gas flow rate. This is a particular feature of the bottom zone of the bed, typically of the order of 10-20 cm deep; when the bubbles detach from the orifices and start rising through the suspension, the excess gas that has been responsible for the bubble formation and growth, and is now in the emulsion phase, goes back into the bubbles, so that higher up in the bed the interstitial flow becomes close to that needed just to fluidise the particles — as is assumed in the well known two-phase theory of fluidisation [7].

This treatment is able to explain successfully the high reaction rate that may occur in the entry region, especially with fast reactions.

1.2. The gas lateral mixing

Less attention has been paid in the literature to the gas lateral mixing in the distributor region of a gas fluidised bed, and to its effect on the reactor performance. This point is of

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Fig. 1. The overall experimental rig. PT 1–4: pressure transducers; FI: flow indicator; PIC: pressure indicator-controller.

great importance when two or more reactants are fed to the reactor without any previous mixing, by means of separate orifices or nozzles located on the bottom plate of the bed. This paper describes an experimental investigation into this phenomenon.

The work has been carried out by using a tracer gas, injected in a cold model of a catalytic fluidised bed reactor for oxychlorination processes. The mixing data have been related to instantaneous pressure fluctuation measurements; in this way the extent of lateral mixing in the bottom region of the bed has been related to the overall fluidisation quality within the bed itself. The experimental data show a considerable degree of gas horizontal mixing at the bottom of a fluidised bed reactor operating in the bubbling regime, which can be accounted for in terms of the model proposed by Rowe to describe the gas behaviour in the entry region of the bed [1]. In addition, this study indicates that conditions close to perfect lateral mixing are reached in that region as the gas superficial velocity is increased to approach the transition to turbulent fluidisation.

2. Experimental

The experimental apparatus, schematically shown in Fig. 1, consists essentially of a Plexiglas column containing the fluidised bed, the gas feeding system, measurement devices and ancillaries. As has been anticipated in the introduction, the whole system and its operating conditions have been



Fig. 2. A schematic view of the bottom part of the fluidising column.

chosen to represent the cold model of a oxychlorination fluidised bed reactor, with the aim of studying its fluid dynamic behaviour. The similarity rules applied to the design of the experimental rig are those originally proposed by Glicksman [8], and more recently reviewed in [9]. The vessel has an internal diameter of 376 mm and contains a bundle of vertical tubes positioned 500 mm above the bottom plate which simulate the heat exchange system of the reactor. Air is used as the fluidising gas, and the operating pressure at the top of the bed is 250 kPa. The gas distributor consists of seven nozzles, equally and symmetrically spaced on the bottom plate; the flow rate through the central nozzle can be fixed independently, whereas the overall gas stream fed to the lateral nozzles is controlled and measured before being divided among them.

Operating such a large experimental rig in an industrial research centre certainly imposes intrinsic limitations to the extension of the experimental programme; on the other hand, the type of tests performed in this study would have negligible significance for industrial processes when carried out at university laboratory scale [4,10].

The mixing tests have been performed, at different superficial velocities of the fluidising gas, by injecting a tracer gas, Helium, continuously through the central nozzle, at a constant concentration. Under steady state conditions, the Helium concentration has been determined in gas samples taken from the column by means of two horizontal probes, both located 258 mm above the bottom plate. The effective vertical distance between the sampling points and the gas feeding orifices is less than 200 mm, because the gas inlet nozzles are placed above the bottom plate; therefore the tracer concentration measurements can be considered representative of the gas mixing achieved in the distributor region. The tip of the probes is protected with a wire screen to prevent entry of, or blockage by, particles. The first probe was positioned axially inside the bed, the latter drew gas from close to the bed wall. A scheme of the bottom part of the fluidising column is reported in Fig. 2.

The gas sample was spilled from the pressurised vessel through the probe by opening a valve connecting it to a 250 cm^3 sampling bottle, initially filled with water to prevent gas contamination. It took about 15-20 s. to fill the

Table 1 Main properties of the bed inventory^a

1 1 2	
d _p (μm)	52
$\rho_{\rm p} ~({\rm kg/m^3})$	1470
$\hat{U}_{\rm mf}$ (m/s)	0.018
U _{mb} (m/s)	0.05
$\varepsilon_{ m mf}$	0.4
€ _{mb}	0.5

^aThe values for $U_{\rm mf}, U_{\rm mb}, \varepsilon_{\rm mf}$ and $\varepsilon_{\rm mb}$ have been measured at ambient conditions.

bottle. The Helium molar fraction was then obtained by gas-chromatography, with Argon as the carrier gas, utilising 1 cm^3 of the gas stored in the bottle. The accuracy of the analysis was verified by repeating it at least twice, and the error was always less than 2%. On the other hand, with different gas samples taken at the same operating conditions the variation in the tracer concentration measurements did never exceed 6%.

For comparison, the average value of the Helium concentration over the whole section of the bed was detected by the same technique, utilising a probe located axially 4258 mm above the gas distributor, well inside the free-board over the bed surface where complete horizontal mixing can be assumed.

Measurements of the pressure fluctuations were also performed over the same range of superficial gas fluidising velocities and utilising the same gas sampling ports. A piezo-electric pressure transducer was used to convert the instantaneous pressure values to a voltage signal, which was stored in a computer and later analysed to obtain the standard deviation and, by means of fast Fourier transform (FFT), the dominant frequencies characterising the random fluctuations. As is well known, the fluctuation of pressure around its average value at a given point inside a fluidised suspension is a function of the overall bubbling behaviour [11]. More specifically, the standard deviation of the pressure signal can be related to the average bubble size, whereas the frequency values, associated with the most powerful components of the same signal, are linked to the processes of bubble formation at the distributor plate and bubble bursting at the surface of the bed, as well as to particular characteristics of the fluidised bed itself [12].

The initial bed height was 2360 mm. The main physical properties of the bed inventory are listed in Table 1, and its particle size distribution is shown in Fig. 3; on the Geldart diagram [13], this is a typical *Group A* powder: the experimental value of the minimum bubbling voidage, measured at ambient pressure and temperature, is greater than the void fraction at minimum fluidisation conditions. A uniform bed expansion range was also observed over the operating conditions used for the cold model.

In the experimental tests, the ratio of the volumetric gas flow through the central nozzle to the total feed rate was fixed at 18.5%; this value is dictated by the reactor operating conditions. For each choice of fluidising velocity, the pressure drop through the gas feeding system was also checked to verify an even gas distribution.

3. Results and discussion

Measurements have been taken at three different gas superficial velocities; the number of tests has been limited by the consumption of Helium involved by these experiments, mainly related to the size of the experimental rig and the necessity to reach steady state conditions. For each experimental condition, the concentrations of the tracer gas, detected at the locations described in the previous chapter, are reported in Table 2; the values are averaged over several gas analyses. The data show that at low gas velocity the tracer gas concentration at the wall is about 55% of that measured in the middle of the column. This ratio increases to 81% with increasing gas superficial velocity, almost reaching a plateau: a further increase in gas velocity does not influence appreciably the horizontal spread of the tracer gas.

This result can be interpreted with the help of the instantaneous pressure measurements. The standard deviations of pressure fluctuations, detected in the bottom region of the bed from the axial and lateral sampling points, are shown in Fig. 4 as functions of the gas superficial velocity. No appreciable differences are induced by the location of the pressure tap because the experimental technique adopted in this study is sensitive to the overall bubbling behaviour, rather than to local perturbations caused by the passage of a single bubble. This latter event becomes the more relevant when two pressure sensors are used with probes located a few centimetres apart inside the bed so that the instantaneous pressure difference between the two is measured [14].

Fig. 4, in addition to the pressure fluctuation standard deviations, also reports the bed wall/axis tracer concentration ratios (b/a of Table 2); the length of the vertical bars indicates the error estimated on the base of the experimental results. The latter points provide a measure of the gas lateral mixing in the bed entry zone. It will be seen that both sets of data show the same qualitative trend. With fine powders, when the pressure fluctuation amplitudes tend to level off, there is the indication of bubbles reaching their maximum size, due to conditions of more intense interaction among them (splitting and coalescing), which finally lead to the transition from the bubbling to the so-called *turbulent* regime [15]. Undoubtedly, these conditions should have a beneficial effect on a horizontal mixing of the inlet gas: the schematic picture of a fluidised bed with individual bubbles detaching periodically from each nozzle and rising vertically through the particle suspension, with limited interactions with neighbouring bubbles, becomes progressively less realistic when the fluidising velocity is increased. Under turbulent fluidisation conditions, on the other hand, bubbles loose their individual *life* and become progressively closer to gas pockets, which appear and disappear rapidly at different places in the emulsion phase provoking intense gas interchange.



Fig. 3. Particle size distribution of the bed inventory.

Table 2 Tracer gas concentrations detected at different sampling points in the fluidising column, and for different gas superficial velocities

U _o (m/s)	<i>F</i> at central nozzle (m ³ /h)	<i>F</i> at lateral nozzles (m ³ /h)	Tracer concentration (mol%)			b/a
			Entry region (axial position), a	Entry region (wall position), b	Freeboard (axial position)	
0.062	4.55	20.04	5.8	3.2	4.0	0.55
0.123	9.09	40.08	6.9	5.5	5.6	0.80
0.185	13.64	60.12	5.8	4.7	4.8	0.81



Fig. 4. Standard deviation of the pressure fluctuations and normalised tracer concentrations: Central (\bigcirc) and lateral (\triangle) sampling points. Normalised tracer concentrations (\bullet) (*b/a* of Table 2).

According to this interpretation, the phenomenon of lateral gas dispersion can be considered in terms of two different mechanisms. The first, prevailing at relatively low gas velocities may be termed *emulsion phase dispersion*: this is mainly governed by dispersion within the dense phase, which may be considered well mixed in the bubbling regime; however a substantial part of the gas injected through the nozzles is segregated inside the bubbles and does not migrate horizontally, at least in the distributor region. When the fluidising velocity is increased enough to approach *turbu*-



Fig. 5. The frequency spectrum of the pressure fluctuations.

lent fluidisation conditions, the whole gas flow rate injected through the central nozzle is available for horizontal mixing, and a *turbulent dispersion* process takes place in the bottom region of the bed.

The *emulsion phase dispersion* process described above corresponds to a considerable extent to the mechanism proposed by Rowe [1] to justify the high degree of conversion in the entry region of a fluidised bed reactor in the bubbling regime. The model is able to explain how tracer gas can be both entrained in the emulsion phase and rapidly dispersed over the bed cross section.

A further point of agreement with the Rowe model, and with the experimental, X-ray ciné photography of bubbling fluidised beds presented in support of it, is provided by the actual values of the tracer gas concentration reported in Table 2. At the lowest fluidising velocity, the ratio between the Helium concentration close to the column wall and that at the centre of the column (b/a) is about two-thirds of the corresponding value obtained when the bed is approaching the transition to the turbulent fluidisation regime. Since the ratio of the overall gas flow through the central nozzle to the lateral feed rate was fixed, this suggests that the amount of gas involved in the mixing process is different in the bubbling and turbulent regimes; the emulsion phase dispersion process should involve more or less two-thirds of the overall gas flow responsible for the mixing process in turbulent conditions. As a result, an estimate is obtained of the gas segregated in the bubbles at low fluidising velocity, which does not percolate into the dense phase of the bed, and therefore does not take part in the emulsion phase dispersion process. Such an estimate is in complete agreement with the experimental observation reported by Rowe [1]; close to the distributor bubbles were found to occupy about a third of the expected volume, and gas leakage through the bubble boundary is at a rate of two-thirds of that through the orifice.

When the frequency spectrum of the pressure fluctuations is considered (Fig. 5), a point of disagreement with the Rowe model is found. No significant components of the signal are present above 5 Hz, whereas the X-ray observations reported by Rowe show an average frequency of bubble formation at each orifice of about eight bubbles per second. This discrepancy could be ascribed to the almost immediate coalescence at the point of detachment of strings of bubbles originated from the same nozzle, in the range of gas flow rates used in this work. In this way a reduction takes place of the dominant frequency of the phenomena characterising the distributor region.

It should be pointed out that the experimental data reported in this study were obtained just above the so-called *entry region* of the fluidised bed; the design of the distributor may be crucial to the promptness and effectiveness of lateral mixing in that region. This aspect has not been considered here.

4. Conclusion

Experimental data have been presented to quantify the phenomenon of horizontal gas mixing at the bottom of a fluidised bed reactor, which is of great industrial relevance for cases where the reactants are injected separately through different nozzles.

Measurements taken at the upper boundary of the distributor region show that the concentration near the wall of a cylindrical column of a tracer gas fed axially to the bed, increases with the fluidising velocity, and a fairly complete lateral mixing is reached. This condition has been associated with the approach to the *turbulent fluidisation* regime, by means of simultaneous records of instantaneous pressure fluctuations within the bed that provide a measure of *fluidisation quality*.

The experimental observations are shown to relate to the model proposed by Rowe [1] to characterise gas flow in the *entry region* of a fluidised bed chemical reactor.

5. Notation

- $d_{\rm p}$ average particle size
- *F* volumetric gas flow rate (at the operating conditions)

- U_o gas fluidising velocity
- $U_{\rm mb}$ minimum bubbling velocity
- $U_{\rm mf}$ minimum fluidisation velocity
- $W_{\rm s}$ power spectral density function, divided by the signal variance
- $\varepsilon_{\rm mb}$ minimum bubbling voidage
- ε_{mf} minimum fluidisation voidage
- $\rho_{\rm p}$ particle density

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