Hydrodynamic study of a Two-Section Two-Zone Fluidized Bed Reactor with an immersed tube bank via PIV/DIA

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HIGHLIGHTS

Coupled PIV/DIA allows the hydrodynamic study of a pseudo-2D TS-TZFBR with internals.
DIA has been optimized to accomplish global bed mass balances.
The effect of diverse tube bank alignments on the reactor dynamics has been determined.
High agreement between experimental trends for solids and bubble dynamics.
Bubble size decreases by 30% while only slightly affecting the solids circulation time.

GRAPHICAL ABSTRACT

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ABSTRACT

The hydrodynamic behaviour of a pseudo-2D Two-Section Two-Zone Fluidized Bed Reactor (TS-TZFBR) with an immersed tube bundle in its lower zone has been studied using non-invasive Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA). Coupled porosity distribution maps from DIA post-processing and solids velocity vector fields from PIV analysis allowed the reconstruction of the transient and time-averaged solid fluxes along the bed. Six different tube bank configurations at several different superficial gas velocities have been tested to evaluate the hydrodynamic behaviour within the lower zone of the TS-TZFBR. The solids axial mixing along the vertical bed position has been quantified for this novel reactor configuration. Besides, the effect of the internals on the gas-solid mass transfer has been estimated by means of bubble size. A five-staggered tube bundle configuration was able to diminish the average bubble size by 30% within the lower bed zone at usual TS-TZFBR gas flow rates, $u_{\text{gas}}/u_{\text{mf}} = 3.0$, without increasing the solids circulation time dramatically.

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1. Introduction

The Two-Zone Fluidized Bed Reactor (TZFBR) allows performing heterogeneous gas-solid catalytic reactions with in situ catalyst regeneration (Herguido et al., 2005). The reactor design has two separated gas inlets to the fluidized bed: an immersed gas inlet for the gaseous reactant and a gas distributor at the bottom of the bed for the oxidizing agent. Heterogeneous catalytic reactions take place in the upper region of the TZFBR whereas catalyst regeneration takes place below the reactant injection point. This process integration is possible owing to the continuous catalyst axial circulation through the bed typical of fluidization processes. This
reactor concept becomes especially interesting for the conversion of gaseous hydrocarbons, where the catalyst suffers from deactivation by carbon (coke) deposition on its active surface. Under certain process conditions, the continuous coke burning within the lower bed zone in the presence of an oxidizer, i.e. diluted oxygen, may result in a long term steady state reactant conversion without net catalytic deactivation. The performance of this reactor has been successfully tested for alkane dehydrogenation (Medrano et al., 2013a, 2013b) as well as for ethanol reforming (Pérez-Moreno et al., 2012) and methane aromatization (Gimeno et al., 2010). The achievement of a steady state operation in a TZFBR depends on several process variables such as operating temperature, catalyst performance or reaction-to-regeneration kinetics, but also on the bed hydrodynamics and the gas-solid mass transfer rates. Indeed, a proper axial solids circulation between the lower and the upper bed zones would enhance the process integration. Analogously, control of the fluidization regime within the bed would help to decrease mass transfer limitations.

Recently, novel TZFBR configurations that incorporate two zones with different cross sectional areas, i.e. Two-Section TZFBR (TS-TZFBR), have been proposed (Julián et al., 2013, 2014a, 2012). These improved reactor configurations allow the use of low regenerative-to-reactive gas flow rates if required. Their main feature is the use of a tapered transition region between the two bed zones. Julián et al. (2012) studied the formation of defluidized bed regions above the section change under certain fluidization conditions, both experimentally and by Computational Fluid Dynamics (CFD) simulations (Julián et al., 2014b). It was suggested to adopt a conservative tapered section angle (α) limit to avoid defluidization effects: α ≥ 80°, with respect to the horizontal position. Moreover, the most suitable axial location of the immersed gas distributor to attain a proper solids circulation pattern has been identified and suggested. However, as a result of using narrow bed sections in the lower zone of the TS-TZFBR, new fluid dynamic issues arose. Under certain fluidization gas flow conditions, slugging regimes tend to appear leading to short-circuited solids recirculation within the lower bed region (Julián et al., 2013). Furthermore, these regimes decrease the gas-solid mass transfer rates and may lead to high concentrations of oxidizing agent in the reactive zone, if the contact between the oxygen and the deactivated catalyst surface becomes poor.

The novel reactor configuration that is proposed in this work consists of a TS-TZFBR (α = 80°) with a tube bundle in its lower zone to allow bubble breakage and improve gas-solid contact while also possibly feeding the oxidizing agent through the tubes. It is widely accepted that the use of tube banks normally decreases axial solids mixing (e.g. (Hull et al., 2000; Sitnai, 1981)). However, if the tube bundle is attached to a column in which slugging regimes normally take place the circulation time could be significantly decreased.

The purpose of this work is, thus, to evaluate the effect of different configurations of internals, as well as some other operational variables (gas velocity and immersed distributor location) on the fluid dynamic behaviour of a TS-TZFBR scaled-up with respect to that used by Julián et al. (2012). The determination of the axial solids mixing and bubble properties has been carried out using a combination of non-invasive Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA) in pseudo-2D reactors. Although the quantitative extrapolation of the phenomena observed in the 2D beds to real 3D fluidized beds is in general not trivial, the obtained results should help to gain a better understanding on the effect of immersed tube banks on these fluid dynamic behaviors; as well as to validate numerical models with which 3D beds can be investigated.

2. Experimental set-up

The experimental set up consists of a Perspex® pseudo-2D Two-Section Two-Zone Fluidized Bed Reactor, an ultra-fast high-resolution camera, a software for image post-processing, two led lamps and two mass flow controllers for gas supply. A detailed scheme of the experimental set up is presented in Fig. 1.

The dimensions of the reactor used in this work are 65 × 8 × 0.8 cm (height × max. width × depth). The maximum width corresponds to the upper bed zone width. The straight lower zone is 4 cm wide and 12 cm high. The tapered region between bed zones has an angle, α, of 80° with respect to the horizontal position and connects the two straight bed zones. The tapered section angle has been defined elsewhere (Julián et al., 2012, 2013, 2014b) to be suitable to avoid defluidized bed regions in a TS-TZFBR. The reactor had an immersed T-shaped orifice gas distributor (d₉₀₉₅ = 3 mm). The external diameter of the distributor was 4 mm and it could be shifted along the vertical bed position. The bottom inlet consists of a glass porous plate with a small pore size, d₉₅, of 40–100 μm to obtain uniform gas distribution. Fluidization gas was pressurized air and bed particles were non-spherical SrAl₂O₄ based phosphorescent solids (Geldart-type B), used as optical tracers in previous studies (Julián et al., 2013). Their size distribution, d₉₅, was in the range 200–320 μm with a bulk density, ρₑ, of 1.43 g/cm³ and a fixed bed porosity, Eₚ₉₅, of 0.58. The minimum fluidization velocity of these particles, uₑ₉₅ = 10.1 cm/s, was determined by measuring the bed pressure drop at decreasing gas velocities in a straight vessel.

Compressed air was fed through two mass flow controllers (Brooks® 20 NL/min). Air was first humidified to minimize electrostatic effects between the walls and the solids. A black sheet
was placed behind the rear reactor wall to increase the image contrast between the dense and dilute phases, i.e. emulsion and gas bubbles, within the fluidized bed.

Fluoridization recordings were performed with a LaVision® Imager Pro high speed camera and analyzed in DaVis 8.0.3, a commercial software for image processing. The recording frequency was adjusted to 750 Hz to avoid image blurriness related to the fast solids movement and a time delay, $\Delta t$, of 0.3 ms between two consecutive frames was selected for PIV processing. For each 12 s video, a total amount of 9000 double-frame images were recorded. Some of them were discarded to get, finally, 900 images (1800 frames) at 1/75 s rate. For each experimental condition, three different recordings were performed. Therefore, experimental results shown in this study have been averaged out from 2700 pairs of frames and 36 s of fluidization. Previous studies on similar reactors with the same particles and gas flow rates indicate that both this recording time and the number of images are suitable to get representative results in terms of solids mixing and bubble properties (Julián et al., 2012, 2014b). The image resolution has been adjusted to 90 pixels per cm or, analogously, between 2 and 3 pixels per particle depending on the particle size, as some authors recommend (de Jong et al., 2012; Westerveel, 2000) to get proper PIV analysis performance.

Two dimmable led lamps have been used to get homogeneous illumination on the front reactor wall. The intensity has been adjusted together with the shutter speed of the camera to avoid overexposure or low illumination of the bed. A homogeneous and proper light intensity for the raw images is a must, since bed porosity results depend directly on the image intensity map. Although some image filters can be applied in the DIA post-processing to improve image contrast or homogenize light intensity, illumination settings have been kept constant throughout the experimental series.

Essentially, the effect of three operational parameters on the TS-TZFB fluid dynamics has been tested: the fluidization regime, the relative axial location of the immersed orifice gas distributor ($x_{or}$) with respect to the bottom end of the tapered bed section ($x_2$) and the presence of internal elements, i.e. tube bundle (horizontal cylinders, $d_{tube} = 4$ mm) in the lower bed zone. Three different relative gas velocities, $u_{mf}/u_{mf}$ has been tested ranging from 2 to 3 times $u_{mf}$. Besides, two axial distributor locations, $x_{or} - x_2 = [0–1.5]$ cm, and six different configurations of the tube bank have been evaluated. The number of internal elements located within the narrow bed section of the TS-TZFB varied among the experiments between 3 and 8. The tube bundle configurations were either staggered or hybrid (staggered + in-line), as illustrated in Fig. 2. A total of 36 experimental conditions have been performed to study the coupled effect of these operational variables. Nevertheless, experimental results shown throughout this paper will only be referred to 3, 5 and 8–tube configurations in order not to crowd graphs and figures. The other tube bundle arrangements tested do not lead to significant differences in solids motion and bubbling behaviour in comparison to results from the three shown configurations.

3. Particle Image Velocimetry

Particle Image Velocimetry (PIV) is a widely used experimental technique for the measurement of solids motion in pseudo 2D fluidized beds. PIV/DIA has been selected for this experimental study due to its simplicity, its non-intrusiveness and the detailed whole-field information on both gas and emulsion phase with high spatial and temporal resolution that this technique can give (Laverman et al. 2008; Westerveel, 1997). Further available experimental techniques for fluid dynamic studies on fluidized beds are: Electrical Capacitance Tomography, X-ray Tomography, Magnetic Resonance Imaging or Positron Emission Particle Tracking (van Ommen and Mudde, 2008). Although PIV was originally developed to visualize fluid flow patterns using disperse tracer particles, several authors (de Jong et al., 2012; Laverman et al., 2008; Link et al., 2004; van Buijtenen et al., 2011; Westerveel, 1997) successfully extended the technique to dense gas–solids systems. The non-intrusiveness becomes here essential, since the tested TS-TZFB configurations are small and in case immersed probes were necessary they could dramatically change the fluid dynamics of the bed. The main disadvantage of the technique is the requirement of visual access. That means, only pseudo-2D reactor configurations can be effectively studied.

The way to determine the experimental solids velocity field is to compare two consecutive instantaneous images separated by a short time delay, $\Delta t$, and apply a cross-correlation analysis to the interrogation areas in which every image is divided. Fig. 3a shows an example of raw image divided by interrogation areas (32 $\times$ 32 px) from which the PIV analysis is carried out. The most likely volume-average displacement of the particles in the selected interrogation area divided by the time delay between frames gives the solids spatial velocity in each region of the bed.

The PIV post-processing of the recorded pairs of images was carried out using the commercial software package DaVis (LaVision). A multi-pass algorithm was employed using, initially, 32 $\times$ 32 px interrogation areas (with 50% overlap) and, afterwards, two steps of 16 $\times$ 16 px (50% overlap) to reconstruct the corresponding vector images. Interrogation areas need to be small enough to represent regions with nearly uniform motion but not too small to miss particles displacement between consecutive frames if it exceeds the length of the interrogation area. The use of a multi-pass algorithm allows the reduction of image noise and increases the spatial resolution with respect to the standard cross-correlation algorithm. After applying PIV, the solids velocity field is obtained for every pair of images (Fig. 3b). However, resulting velocity vectors cannot be directly transposed into solids flux profiles since the PIV software does not discriminate between dense and dilute phases. Therefore, a method to account for the solids fraction in every interrogation zone is required to determine the local solids fluxes. The information about the bed porosity distribution can be extracted from the Digital Image Analysis post-processing. The DIA method used in this work will be described in detail in Section 4. The solids flux provides the physically most important information on the solids motion in gas-solid fluidized beds. Therefore, the coupled use of PIV and DIA becomes essential to reconstruct both the solids velocity and porosity and get detailed information on the solids fluxes throughout the TS-TZFB.

241

To determine the solids flux profiles from the solids velocity profiles, a reliable correlation between the 2D and 3D solids fraction (or porosity) is required.

4. Digital Image Analysis technique

Digital Image Analysis (DIA) is a technique that allows the extraction of useful information from 2D digital images. Applied to fluidized beds, DIA represents the use of algorithms to find out relevant aspects of the bed such as porosity distribution and gas bubble statistics by means of local pixel intensity. DIA was first applied in bubbling fluidized beds by Agarwal et al. (1997) to detect bubbles. Goldschmidt et al. (2004) used this technique to detect spout fluidized beds. The conventional DIA algorithm applied to bubbly fluidized beds discriminates between the bubble and solids phases on the basis of the pixel intensity, employing a prescribed threshold value. The algorithm corrects the velocity vector maps generated by PIV filtering out high negative velocity vectors that frequently occur within big gas bubbles as a result of particle rain. Therefore, the conventional DIA binarizes the solids fraction into bubbles, i.e. regions free of solids, and emulsion, having this last phase a constant porosity which is usually referred to the minimum fluidization porosity (Hernández-Jiménez et al., 2011; Laverman et al., 2008). Using pixel intensities of the acquired images as a measurement of the local solids fraction may introduce an error, because the pixel intensity does not contain any depth information of the solids hold-up, since this is not visible from the frontal projection (de Jong et al., 2012). Moreover, the conventional DIA algorithm does not account for local variations in the solids fractions. These shortcomings led to the development of a new DIA algorithm with improved accuracy and wider applicability.

To improve accuracy and overcome conventional DIA limitations, van Buijtenen et al. (2011) and de Jong et al. (2012) developed a correlation (Eq. (1)) between the image 2D bed intensity and the solids 3D volume fraction based on reconstructed artificial images from Discrete Particle Modeling (DPM) simulations. When using a “linear” intensity decay along the bed depth to generate the DPM artificial images, these authors found that the “true” 3D solids fraction from simulations displayed a linear profile with a small slope for low image intensities (or analogously “2D solid fractions”). Besides, the 3D solids fraction for high intensities showed a steep asymptotic increase until the maximum 3D packing fraction, i.e. minimum fluidization porosity. Two fitting parameters (A and B) were used to take into account the effect of the bed depth to particle size ratio, \( \Delta z/d_p \).

\[
\varepsilon_{3D} = \begin{cases} \frac{\varepsilon_{2D}}{1 + A \frac{\varepsilon_{2D}}{\varepsilon_{3D} \max}}, & \text{for } \varepsilon_{3D} < \varepsilon_{3D} \max \\ \varepsilon_{3D} \max, & \text{for } \varepsilon_{3D} \geq \varepsilon_{3D} \max \end{cases}
\]

(1)

Artificial images from DPM obtained by these authors were expected to mimic experimental light intensity variation of the acquired images.

In the present work, however, the map of pixel intensities rarely became completely homogeneous in experimental video recordings due to inhomogeneous illumination issues. Therefore, the 2D to 3D volume fraction correlation (Eq. (1)) could not be directly applied to the recorded raw images. The map of intensities of experimental fluidization images (Fig. 4a) needed certain improvements before the correlation could be used. The first step was to subtract a “background” intensity map (Fig. 4b) to the raw images in order to minimize inhomogeneous illumination effects along the bed (Fig. 4c). The “background” image was here determined as the local averaged bed intensity along the recording time once outliers, i.e. overexposed pixels, and gas bubbles were filtered out from the raw images. In practice, local intensities under 80% of the maximum image intensity were not considered in the calculation of the local average bed intensity. Secondly, a median filter was applied in order to smooth the local variation of intensities within the emulsion phase due to image noise (Fig. 4d). Next, corrected image intensities were normalized to 1 dividing by the maximum corrected intensity of the emulsion phase (Fig. 4e). This step allowed the comparison between our experimental values and the normalized intensities obtained from DPM artificial images by de Jong et al. (2012). The histogram of the normalized images was, then, used to get the 3D volume fraction based on the correlation developed by de Jong et al., 2012, Eq. (1) (Fig. 4f). Fitting parameters A and B have been selected in this work minimizing errors in the total bed mass. Their value differ slightly from these obtained by de Jong et al. These authors considered spherical particles with mono-modal distribution and, thus, with a constant bed depth-to-particle size ratio (\( \Delta z/d_p \)). This \( \Delta z/d_p \) mainly influences the \( \varepsilon_{2D} \rightarrow \varepsilon_{3D} \) curve shape. The correlation curve obtained in this work suggests that the experimental light intensity transition between low and high bed porosities is more gradual than in artificial images from DPM simulations. This will be discussed in detail below. Once the \( \varepsilon_{2D} \rightarrow \varepsilon_{3D} \) correlation was applied, the transient volumetric bed porosity maps (Fig. 4g) were determined. Although it seems that the majority of the emulsion phase has the same porosity, the improvement compared with the binary approach suggested by some authors (Hernández-Jiménez et al., 2011; Laverman et al., 2008) lays in the high precision for the determination of bubble wake and cloud porosities. Solids motion in bubble surroundings is normally high. Therefore, this DIA improvement may have a critical impact on the proper determination of time-averaged solid fluxes, since 3D porosity data are then coupled with transient PIV results to quantify axial mass fluxes within the bed.

As already discussed, experimental histograms do not fit completely with reconstructed artificial images from DPM within the emulsion phase. In artificial images, a quite narrow range of intensities represents a wide interval of volumetric solid fractions...
(see 2D–3D curve in Fig. 4f). However, experimental histograms suggest that the porosity of the emulsion phase corresponds to a wide range of pixel intensities (Fig. 4d). Therefore, some assumptions need to be made.

As can be observed in the histogram of Fig. 4d, the distribution of intensities within the emulsion phase for a filtered image (corresponding to a grayscale value between 0.8 and 1, in this case) is still wide. Due to the asymptotic shape of the 2D-to-3D correlation at high image intensities, the cut-off intensity value must be selected carefully in order not to underestimate the solids fraction of the low-intensity regions of the emulsion phase. A number of criteria could be applied to get the most suitable cut-off value regarding either the maximum pixel intensity, the average pixel intensity within the emulsion phase (range 0.8–1) or the minimum intensity in the range of the emulsion phase (the whole emulsion may have uniform maximum porosity). In this work, the second criterion has been applied. With this criterion, the most representative intensity within the emulsion phase, i.e. intensity mode, is selected as cut-off intensity, \( I_{\text{cut-off}} \) (Fig. 4d). Bed intensities over \( I_{\text{cut-off}} \) are then assigned to \( I_{\text{cut-off}} \) and intensities under it are normalized against \( I_{\text{cut-off}} \).

The determination of the fitting parameters \( A \) and \( B \) in the 2D–3D porosity correlation (Eq. (1)) was carried out by minimizing errors in the total bed mass for every fluidization experiment. The optimal values of \( A \) and \( B \) for each experimental condition differ from each other and need to be determined individually. Therefore, a multivariable root-seeking algorithm has been applied to minimize the target function, which is here defined as the bed mass relative error, i.e. \( E_m = (m_{\text{real}} - m_{\text{calc}})/m_{\text{real}} \). The real bed mass \( m_{\text{real}} \) is well known and \( m_{\text{calc}} \) represents the estimated bed mass from DIA 3D porosity maps. As an example, Fig. 5a shows the evolution of the errors in the bed mass estimation for different pairs of tested parameter values \( A \) and \( B \) (from Eq. (1)) for a 0-internals configuration at \( \dot{u}_{\text{gas}}/\dot{u}_{\text{mf}} = 3.0 \). In this case, it can be observed that parameter values in the range \( A = [0.02 - 0.1] \) and \( B = [0.8 - 1.0] \) may lead to a suitable estimation of bed mass and, thus, bed porosity. Within the ‘valley’ of different pairs of values of \( A-B \) that lead to low errors in the estimation of the real bed mass (Fig. 5a), it has been found that the optimal window of values that minimize errors in this test was: \( A = [0.02 - 0.03] \) and \( B = [0.94 - 0.96] \) (Fig. 5b). In particular, the optimal values of \( A \) and \( B \) for some tests performed at \( \dot{u}_{i} = 3.0 \) in different reactor configurations are: \([A, B]_{1} = [0.023, 0.948], [A, B]_{2} = [0.023, 0.942], [A, B]_{3} = [0.023, 0.935] \) or \([A, B]_{4} = [0.024, 0.917] \). These values are in agreement with those obtained by de Jong et al. (2012), based on DPM simulations.

With such optimal parameter values, the bed mass constraint (estimation error \( \leq 0.5\% \)) was accomplished for every test. Regarding
the mass flux conservation it was found that the global unbalance of axial solids fluxes, i.e. sum of positive and negative local mass fluxes along the bed height, was very low ($< 5$ kg/m$^2$s in the worst scenario).

In order to check the solids mass flux conservation as a function of $A$ and $B$, an additional target function was tested. The alternative function was defined as the product of the bed mass estimation error by the global axial mass flux unbalance. For both target functions same optimal $A$ and $B$ values were found, illustrating that the own minimization of the bed mass estimation error enhances mass flux conservation. Therefore, the use of the suggested $E_m$-method to estimate the correlation parameters $A$ and $B$ allows a proper determination of the 3D solids hold up and local axial mass fluxes. Nevertheless, the use of this method is restricted to recordings in which the entire bed is captured. Otherwise, further assumptions need to be made.

Concerning the different sources of errors that may affect the proper PIV/DIA analysis, non-uniform illumination issues have been solved by subtracting a background image to the raw frames, as already discussed. In addition, the bed discretization into $16 \times 16$ interrogation windows ($1.8 \times 1.8$ mm bed regions at a resolution of 90 pixels/cm) allowed enough spatial resolution for the proper determination of the porosity gradient within bubble clouds. Lastly, cross-correlation errors from PIV analysis were minimized applying a multi-pass algorithm with window size reduction, interrogation areas overlapping and removal of eventual outliers. Therefore, velocity vector maps should not represent a relevant source of errors to be considered in the bed mass and mass conservation balances.

5. Results and discussion

This section illustrates the main experimental findings on the TS-TZFBR hydrodynamics from coupled PIV–DIA analysis in terms of solids circulation patterns, quantitative axial solids mass fluxes, particle mixing between bed zones, bubbling regimes at different gas velocities and effect of different tube bank configurations on the particle and bubble dynamics.

5.1. Interpretation of transient PIV results

Axial solids velocity vector maps provide useful information about particles motion around gas bubbles. Indeed, bubble wake contours and even clouds of single bubbles may be estimated from transient PIV velocity maps. As Fig. 6 illustrates, bubbles usually appear as regions with high solids velocity in the downward direction as a result of particle raining. Immediately under bubbles, solids upward velocity regions are found. These regions are normally well enclosed and represent bubble wakes. Therefore, PIV results would allow estimating bubble wake fractions as a function of bubble size and fluidization regime if required. Moreover, transient solids velocity maps illustrate solids circulation paths in the presence of internal elements, as shown in Fig. 6. Analogously, it can be observed that bed regions which are far enough from a gas bubble show velocities close to zero. This is in agreement with the fact that only the presence of gas bubbles promotes solids motion in a fluidized bed. Bubble clouds can also be distinguished in Fig. 6 as the regions with low solids downward velocity in the bubble surroundings. Particles in the bottom-end of bubble clouds flow towards bubble wakes. Lastly, the transient raw image of Fig. 6 illustrates the effect of internal elements on the formation of ‘gas pockets’ under the tubes. It has been observed experimentally that the fluidization gas gets partially trapped under the internals, whereas the dense phase moves axially on both sides of the tube. As a result, particle-free regions grow at the bottom side of the tubes from a fluid dynamic point of view, these regions should not be strictly considered as gas bubbles but as particle-free gaps or ‘gas pockets’. Therefore, these low porosity regions which would be detected as gas bubbles by the DIA algorithm were filtered out from bubble size distribution results presented in Section 5.5. A more detailed investigation of the occurrence of gas pockets and their
effect on heat and mass transfer rates will be presented in a future work.

5.2. Solids circulation patterns in a TS-TZFBR

Solids circulation provides particles mixing between the two bed zones in a TS-TZFBR and allows integration of reaction and catalyst regeneration. In Fig. 7, the time-averaged radial distribution of axial solids fluxes at different vertical bed positions is presented. The radial distribution gives an idea on how solids circulate along the bed. Particles preferentially flow upwards through the bed center and downwards close to the lateral walls. Average solids flux profiles are smoother in case of the lower reactor zone whereas they become sharper above the secondary gas injection. Just above this injection point, gas flow is twice higher while the cross-section remains almost constant. This is the reason why the positive and negative solids fluxes become locally higher at this stage. The morphology of the immersed distributor plays also a role in the particles circulation within the tapered bed region. The use of a T-morphology of the immersed distributor plays also a role in the positive and negative solids circulation paths in the upward and downward directions, respectively. Therefore, the solids flux diagram presented in Fig. 8 does not give detailed information on the axial mixing rate but on the circulation patterns.

In order to evaluate solids axial mixing, the average solids mass flux in the upward (Fig. 9a) and downward directions (Fig. 9b) have been analyzed separately. Figs. 9a and b illustrate the average positive (towards the bed top) and negative (towards the bottom) solids fluxes, respectively, for the three experiments presented in Fig. 8. The calculation of positive and negative mass fluxes has been carried out considering the average positive and negative value of the transient solids mass flux in each bed interrogation area. If there is no particles movement in a certain interrogation area at a certain fluidization time, i.e. mass flux=0, this value is zero.

5.3. Solids flux profiles along the bed height

As previously discussed, solids fluxes provide the physically most important information on the solids motion in gas-solid fluidized beds. Fig. 8 shows the average radial distribution of axial solids fluxes for 3 different reactor configurations at the different vertical bed positions at which the tube bank are located. The reference case, i.e. TS-TZFBR without internals, is compared to two different configurations with 5 staggered and 8 hybrid (staggered and in-line) internals. The same gas velocity ($u_{gax}/u_{mf}=3.0$) has been used in every experiment.

A first comparison between the radial distributions of solids fluxes for the three cases suggests that the axial circulation in the case without internals is substantially lower than in the other configurations, since the axial profile of fluxes becomes smoother in this case. This assertion is not necessarily true; it only means that the circulation patterns are clearer in the case in which internals are used, since they act as draft tubes providing preferential solids circulation paths in the upward and downward direction, respectively. Therefore, the solids flux diagram presented in Fig. 8 does not give detailed information on the axial mixing rate but on the circulation patterns.

![Fig. 8. Average axial solid fluxes at $u_{gax}/u_{mf}=3.0$ for three different configurations of internals at tube array vertical locations ($x_{int}=6.5, 8.5$ and 10.5 cm).](image)

![Fig. 7. Average axial solids flux map (a) and corresponding average axial solids fluxes at different vertical bed positions (b) for a reactor configuration with 3 staggered internals, $3i$, at $u_{gax}/u_{mf}=3.0$.](image)
not considered when averaging out either positive or negative mass fluxes.

As can be observed in both Figs. 9a and b, the radial profile of average solids flux in the upward (+) and downward (−) direction is quantitatively similar either in experiments without or with internals. In the experiment without tube bank (0i − u_l = 3.0), it can be seen that both the positive and negative average mass fluxes follow a turbulent flow profile along the radial position, i.e. almost constant solids flux within the bed center with a sharp mass flux gradient close to the reactor walls where no net particles movement is detected. In case internals are used, positive and negative radial profiles are affected by the location of the internals. Local positive (upward) and negative (downward) fluxes in interrogation areas far from the region of influence of an internal differ in less than 3% with respect to those obtained for the base case without internals. However, particles movement in the surroundings of an internal is strongly decreased, as can be observed for both positive and negative mass fluxes in experiments with 5 and 8 internals (5i and 8i, respectively).

Regarding the effect of different configurations on the +/− mass fluxes, results show that the highest number of internals (8i) leads to the lowest solids hold-up and, thus, to a lower axial mixing with respect to the 0i case. Besides, it can be observed that 5i and 8i configurations mimic their first row of internals (z = 6.5 cm) and their mass flux profile at that height becomes similar regardless the different configurations of the second and third rows at z = 8.5 and 10.5 cm, respectively. It suggests that, in this case, the arrangement of the upper tubes does not affect the solids motion through the lowest row of the tube bank.

Fig. 10 represents the axial profile of the average radial solids flux in the upward and downward direction for the 0i, 5i and 8i configurations. The general trend for a 0i configuration shows that both the positive and negative average solids mass fluxes increase from the very bottom of the bed along the vertical position within the lower straight section. Mass flux peaks observed at z = 12 cm above the porous plate are related to the immersed secondary gas inlet. The secondary gas feed leads to high local gas velocities, i.e. fast solids transport, in this region of the bed where the reactor width is still narrow (bottom-end of the tapered bed section). Furthermore, the average solids mass flux increases substantially along the tapered region and finally drops around the freeboard. The axial profiles of mass flux for different configurations of internals follow a similar trend. However, a local solids flux decrease is found at the bed height at which the tube bank is located. For instance, the axial mass flux in the upper region of the lower bed zone (immediately under the secondary gas injection) is around 30% lower in case of 8i than without internals at u_l = 3.0. Therefore, the use of internals may have an important role on the solids axial mixing or, analogously, on the TS-TZFB reactor performance.

For every experiment, the accomplishment of global (whole bed) and local mass balances was checked. Of course, the direct summation of axial positive and negative average mass fluxes for an experiment is not representative of its mass balance, since the frequency of positive and negative transient mass flux occurrence is not taken into account. For instance, considering the 0i case of Fig. 10, the rate of solids flowing in the upward direction against those flowing down within the lower bed zone is around 0.9 whereas upward-to-downward ratios within the tapered section are between 0.6 and 0.8. Actually, it has been found that the upward-to-downward ratio increases with the gas flow rate in the tapered region over the immersed gas distributor location and decreases with u_gfluent/u_l in the lower bed zone. Moreover, the upward-to-downward solids flux frequency varies locally in the presence of internal elements. If the positive and negative axial mass flux profiles as these presented in Fig. 10 are multiplied by the frequency of positive and negative events, i.e. mass fluxes, along the fluidization time then the so-called ‘cross-sectional mass flux profiles’ (F_c) in the upward and downward directions can be obtained. Physically, F_c would represent the average positive and negative mass fluxes if these are referred to the entire bed cross-section at each vertical position. The symmetry between positive
and negative cross-sectional mass flux profiles indicates the qualitative mass balance accomplishment along the vertical bed position and, thus, gives an idea on the validity of the method to obtain porosity maps from bed intensities and the 2D–3D correlation described in Eq. (1). In this regard, Fig. 11a shows the vertical profiles of positive and negative $F_c$ for the experiments of Fig. 10 and Fig. 11b illustrates the time average errors in the $F_c$ solids flux accomplishment at every vertical bed position. It can be observed that local errors are low in comparison to the average positive and negative mass fluxes. Besides, the global mass balance to the whole bed is accomplished for all tests. This illustrates the validity of the method used to obtain the map of porosities through the minimization of the error in the estimation of the real bed mass.

5.4. Particle mixing between lower and upper bed zones. Solids circulation time

According to previous works (Sánchez-Delgado et al., 2013), the solids circulation time within a fluidized bed can be measured from PIV/DIA experiments once the average axial solids mass flux has been determined. The theoretical circulation time of a group of particles is described elsewhere (Rowe, 1973) as the mean time required by the particles to reach the freeboard and return to their original position. The mean circulation time, $T_c$, can be expressed as shown in Eq. (2), being $h_{\text{min}}$ the average fluidized bed height (or, analogously, the height at which the freeboard starts), $h_{\text{min}}$, the minimum height at which bubbles promote solids axial movement and $v_u$ and $v_d$ the mean solids velocity in the upward ($u$) and downward ($d$) direction.

$$T_c = \int_{h_{\text{min}}}^{h_{\text{fb}}} \frac{dz}{v_u(z)} + \int_{h_{\text{fb}}}^{h_{\text{min}}} \frac{dz}{v_d(z)}$$

Both the minimum height, at which solids movement is promoted, and the average fluidized bed height depend on the gas velocity. High gas velocities increase $h_{\text{min}}$ due to an enhanced bed expansion and decrease $h_{\text{min}}$ due to an increased gas excess over the minimum fluidization with a subsequent bubble size and solids drag enhancement. The mean solids velocity in the upward and downward direction has been determined taking into account the axial solids mass flow profiles, the bulk density of the bed ($\rho_b$) and its average porosity ($\epsilon(z)$) from the DIA post-processing. The effect of gas velocity and internals configuration on the circulation time is presented in Table 1. As already discussed $h_{\text{min}}$ and $h_{\text{fb}}$ are not affected by the presence of internals in the lower bed zone but by the inlet gas velocity. The circulation time slightly increases with the number of internals and increases dramatically at gas flow rates close to the minimum fluidization velocity conditions. The rough estimation of the circulation time may be helpful to improve the TS-TZFBR reactor design.

5.5. Bubble properties

Solids motion is promoted by gas bubbles, which drag upward certain amount of solids on their wakes. It is well known that the wake fraction of a bubble remains almost constant for a wide range of fluidized particle sizes (Rowe and Partridge, 1965). Therefore, the study of the average bubble size evolution along the vertical bed position may help to understand and validate experimental PIV results on axial solids fluxes. DIA post-processing allowed the study of bubble hydrodynamics as presented in the following sub-sections. Firstly, the bubble size distribution (BSD) along the bed vertical position for a 0i TS-TZFBR configuration will be discussed and further compared to that obtained in similar reactor configurations at smaller scale, using the same gas velocities and bed particles. Next, the effect of the tube bundle configuration on the bubble size along the lower TS-TZFBR bed zone will be analyzed. Finally, the effect of the relative gas velocity on the bubble growth will be presented.

<table>
<thead>
<tr>
<th>$u_i$ ($-$)</th>
<th>No. internals</th>
<th>$h_{\text{min}}$ (cm)</th>
<th>$h_{\text{fb}}$ (cm)</th>
<th>$t_{c,u}$ (s)</th>
<th>$t_{c,d}$ (s)</th>
<th>$t_c$ (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0</td>
<td>0</td>
<td>0.9</td>
<td>20.4</td>
<td>358.4</td>
<td>319.9</td>
<td>11.3</td>
</tr>
<tr>
<td>2.5</td>
<td>0</td>
<td>0.6</td>
<td>22.1</td>
<td>223.0</td>
<td>215.9</td>
<td>7.3</td>
</tr>
<tr>
<td>3.0</td>
<td>0</td>
<td>0.5</td>
<td>22.9</td>
<td>172.5</td>
<td>187.6</td>
<td>6.0</td>
</tr>
<tr>
<td>3.0</td>
<td>5</td>
<td>0.5</td>
<td>22.9</td>
<td>195.4</td>
<td>211.3</td>
<td>6.7</td>
</tr>
<tr>
<td>3.0</td>
<td>8</td>
<td>0.5</td>
<td>22.9</td>
<td>199.2</td>
<td>211.7</td>
<td>6.8</td>
</tr>
</tbody>
</table>

Table 1: Effect of gas velocity and configuration of internals on the solids circulation time.

Fig. 11. (a) Positive and negative cross-sectional average solids flux $(F_c)$ along the vertical bed position for 0, 5 and 8 internals configurations (0i, 5i and 8i) at $u_{\text{gas/lum}}=3.0$. (b) Average errors in the $F_c$ solids flux accomplishment along the vertical bed position for 0i, 5i and 8i configurations at $u_{\text{gas/lum}}=3.0$. 
5.5.1. Bubble size distribution in TS-TZFBR

Fig. 12 illustrates typical bubble size profiles along the vertical bed location for different TS-TZFBR sizes without tube bundle. Experimental DIA results obtained for the reactor configuration described in Section 2 are compared to bubble size profiles from TS-TZFBR reactors used by Julián et al. (2012) in previous experimental works. The reactors used in these studies were twice smaller in terms of height and width but preserved the same depth, particle size and density. The section change height of the reactors used in previous studies was $z_{sc,1x} = 8\text{ cm}$ (in this work: $z_{sc,2x} = 12\text{ cm}$). In both cases, an almost linear bubble growing trend has been found along the narrow zone of the bed whereas a sharp bubble size decrease was found within the tapered bed section, where an additional gas injection takes place. The equivalent bubble diameter increases, analogously, along the wide reactor section towards the bed freeboard in both configurations. The TS-TZFBR scaling seems to affect significantly neither the bubble growing trends nor the bubble size shrinkage in the surroundings of the bed section enlargement. In a previous study (Julián et al., 2014a), we described a model to predict the average axial bubble size profile in TS-TZFBR reactors based on the Mori–Wen correlation, incorporating some equations to take into account the coupled effect of section enlargement and additional gas feed on the bubble size. The so called JHM model was able to fit experimental measurements at small bed scale (‘1x’). However, same JHM model formulation has been applied to model DIA results in a twice bigger (‘2x’) TS-TZFBR leading to a poor estimation of both the maximal bubble size under the bed section change (up to 19% deviation) and its evolution through the tapered region. Nevertheless, the model results are still satisfactory in terms of qualitative prediction of growing trends and bubble shrinking effects in the same range of operational conditions.

5.5.2. Effect of tube bundle configuration on BSD

Fig. 13a shows the effect of different configurations of internals (3i, 5i and 8i) on the bubble size reduction within the upper region of the narrow reactor zone fluidized at $3 \cdot u_{mf}$ gas velocity. Error bars have been added to the case without internals to illustrate that the bubble shrinkage in the presence of internals is, at least for the 5i and 8i configurations, representative. These refer to the standard deviation of the average $d_0(z)$ among the three recordings performed at each experimental condition. Error bars for the bubble size profiles obtained with different tube bundle configurations are of the same order but have been removed from the figure for clarity. Experimental results suggest that the different rows of internals play different roles on the average shrinkage of the bubble diameter, i.e. bubble breaking, with respect to the 0i configuration case. The BSD obtained for the 3i configuration illustrates that the single-centered internal of the second row does not lead to substantial bubble breaking. The average bubble diameter at this vertical position is around 1.7 cm, which is lower than both the distance between the internal and the bed side walls and the distance between the rows. Therefore, bubbles tend to round the tube on both sides without decreasing their size. The third row of the 3i configuration consists of two staggered internals. Since the emulsion phase tends to flow downwards close to the reactor walls, bubbles are forced to flow through the narrow gap between these tubes, thus, leading to bubble shrinkage and breaking effects.

The staggered 5i configuration leads to an effective bubble size reduction. The two tubes located at the first row height have a clear shrinking effect. Small bubbles coming up from that first row round easily the single tube located at the bed center without increasing their size. The two staggered tubes on the third row lead, then, to bubble diameter reduction as in the 3i configuration case. Bubbles are, at this height, big enough to be affected by internals.

The hybrid 8i configuration leads to the highest bubble size reduction, as expected. Its first row of internals forces bubbles to shrink passing through the gap between tubes. The next row consists of 3 internals through which bubbles break. Resulting bubbles tend to break again in the presence of the 3 internals of the third row, thus, leading to a sharp reduction of the average bubble diameter with respect to experiments without internals. Nevertheless, the final bubble size reduction obtained with 8i and 5i configurations is very similar. This suggests that the 5i staggered configuration is effective enough for bubble breaking. An increase in the number of internals would not enhance bubble size reduction. Moreover, it would lead to an almost 2% circulation time increase, as described in Section 5.4.

Regarding the faster bubble growing trend above the immersed distributor for the configurations with internals compared to that without tubes, this effect may be related to the average bubble size just under the distributor. It has been experimentally observed that ‘slugs’, i.e. bubbles that tend to occupy the whole bed section ($d_0 > D_{bed}/2$), reduce their size drastically when reaching the tapered region. Therefore, the average bubble size at that height is mainly related to the bubble diameter of gas bubbles coming from the immersed distributor. On the other hand, the shrinkage of the medium size bubbles ($d_0 < D_{bed}/2$) due to bed section enlargement is not as sharp. As a consequence, the use of a tube bank to break ‘slugs’ in the vicinity of the section enlargement may result in an increased average bubble size upwards.

5.5.3. Effect of fluidization regime on BSD

Fig. 13b shows the effect of the relative gas velocity ($u_r$) on the average bubble diameter within the lower bed zone for the TS-TZFBR configuration without internals. As can be observed, the lowest gas velocities tested, 2. $u_{mf}$, lead to quite small bubble sizes. Under these fluidization conditions, the effect of the tube bank on the bubble size reduction is marginal. It has been experimentally observed that such small bubbles flow up rounded internals without breaking. It is remarkable that, in this fluidization regime, no shrinking effect in the bubble size is observed through the transition region, i.e. tapered bed section with additional gas.
This suggests that the biggest bubbles coming from the lower bed zone are almost of the same size as these new bubbles coming from the secondary gas distributor. An increase from $u_r = 2.0$ to $3.0$ in the gas flow rate makes gas bubbles to be around two times bigger along the whole narrow bed section. The relation between bubble size and relative gas velocity ($u_{gas}/u_{mf}$) or, analogously, gas excess over the minimum fluidization ($u_{gas} - u_{mf}$) in fluidized beds has been extensively reported in literature, leading to a number of well-known correlations that are listed and evaluated in a recent review (Karimipour and Pugsley, 2011).

5.6. Bubble hydrodynamics as promoter of solids motion

In this section, axial average bubble size ($d_b$) and bubble velocity ($u_b$) profiles obtained from DIA are compared to experimental PIV/DIA solids flux profiles. Fig. 14 shows the evolution of $d_b$ and $u_b$ along the vertical bed position, together with axial profile of the solids flux in the upward direction for a 0i configuration at $u_r = 3.0$. It can be observed that average bubble size and velocity are intimately related to each other: bigger bubbles are faster than smaller ones, leading to similar size growth and velocity profiles in agreement with classical hydrodynamic correlations (Davidson and Harrison, 1966). It is well known that solids motion in the upward direction is promoted by ascending gas bubbles that transport certain amount of particles in their wake. Assuming an almost constant volumetric fraction of solids in the bubble wake (Rowe and Partridge, 1965), regardless of the bubble size and velocity, the experimental axial solids upward flow would follow a similar trend as the $d_b(z)$ and $u_b(z)$ profiles. Indeed, taking into account that wake and emulsion porosities are similar, the evolution of solids mass flux along the bed height agrees with that of the bubbles. The only region in which DIA bubbling results do not match the experimental solids flux profile is the surrounding area of the immersed gas distributor. We accept that local solids velocity enhancement takes place as an effect of the secondary gas injection. However, we have not observed a similar change in bubble velocity in that region. This may be related either to the cut-off maximal bubble velocity imposed in the DIA algorithm to track bubble centroids between subsequent frames or to the fact that solids may have been sprayed up by the gas jet as a local pseudo-spouted regime without gas bubble formation within that region.

6. Conclusions

Coupled PIV/DIA techniques allowed the hydrodynamic study of a pseudo-2D Two-Section Two-Zone Fluidized Bed Reactor using internals in its lower zone. The correlation developed by de Jong et al. (2012) to determine the local bed voidage from
The determination of bubble and solids hydrodynamic trends has been performed by a range of fluidization conditions. A staggered configuration of 5 internals was able to reduce the average bubble size around 30% within the upper region of the lower reactor zone, avoiding slugging regimes at usual TS-TZFBR fluidization flow rates. The use of internals slightly increased the solids circulation time. Nevertheless, the axial solids mixing did not change substantially within the tube bank region. The radial profiles showed that internals play a role in the solids circulation patterns, acting as draft tubes that re-direct the particle flow towards the center of the bed.

A DIA procedure was developed to quantify solid fluxes in upward and downward directions, respectively. This allowed the determination of average wake and emulsion solids fluxes. The high agreement between experimental trends for solids and bubble dynamics suggests that the PIV/DIA procedure was well implemented.

The scaled reactor dimensions used in this work, with respect to the TS-TZFBR geometries used in previous studies, provided satisfactory results in terms of repeatability of experimental bubble and solids hydrodynamic trends.

### Nomenclature

#### Acronyms

- BSD: Bubble size distribution
- DIA: Digital Image Analysis Technique
- PIV: Particle Image Velocimetry Technique
- TS-TZFBR: Two-Section Two-Zone Fluidized Bed Reactor
- TZFBR: Two-Zone Fluidized Bed Reactor

#### Greek symbols

- $\alpha$: Tapered section angle with respect to horizontal position, (°)
- $\varepsilon_{\text{packed}}$: Packed bed solids volume fraction, (-)
- $\varepsilon_{\text{s,2D}}$: Superficial solids volume fraction, (-)
- $\varepsilon_{\text{s,3D}}$: Solids volume fraction, (-)
- $\varepsilon_{\text{max}}$: Maximum solids volume fraction, i.e. packed bed solids volume fraction, (-)
- $\rho_{\text{b}}$: Bulk solids density, (g/cm$^3$)

#### Symbols

- $A$: Fitting parameter in the $\varepsilon_{\text{s,2D}} - \varepsilon_{\text{s,3D}}$ correlation, (-)
- $B$: Fitting parameter in the $\varepsilon_{\text{s,2D}} - \varepsilon_{\text{s,3D}}$ correlation, (-)
- $D_{\text{bed}}$: Bed diameter, (cm)
- $d_{0}$: Equivalent bubble diameter, (cm)
- $d_{\text{orif}}$: Diameter of the immersed gas distributor orifices, (cm)
- $d_{\text{pore}}$: Pore size of the porous glass plate distributor, (µm)
- $d_{p}$: Particle diameter, (µm)
- $d_{\text{tube}}$: Diameter of internal tubes, (cm)
- $\Delta z$: Bed depth, (cm)
- $\Delta t$: Time delay between subsequent fluidization frames, (ms)
- $E_{\text{r}}$: Absolute error in the mass conservation equation, (kg/m$^2$s$^2$)
- $E_{\text{F}}$: Relative error in the bed mass determination, (-)
- $E_{\text{C}}$: Coupled bed mass and mass conservation error in PIV/DIA results, (a.u.)
- $F_{\text{c}}$: Cross-sectional solids mass flux, (kg/m$^2$s)
- $F_{\text{e}}$: Solids mass flux, (kg/m$^2$s$^2$)
- $F_{\text{up}}$: Solids mass flux in the upward direction, (kg/m$^2$s$^2$)
- $F_{\text{down}}$: Solids mass flux in the downward direction, (kg/m$^2$s$^2$)
- $h_{\text{tb}}$: Average bed height or height above which the freeboard starts, (cm)
- $h_{\text{min}}$: Minimum bed height at which bubbles promote solids motion, (cm)
- $I_{\text{cut-off}}$: Pixel intensity mode within the emulsion phase in fluidization frames, (-)
- $m_{\text{real}}$: Real bed mass, (g)
- $m_{\text{calc}}$: Calculated bed mass via DIA porosity maps, (g)
- $u_{b}$: Bubble velocity, (cm/s)
- $u_{\text{gas}}$: Gas velocity, (cm$^2$/cm$^2$s$^{-1}$)
- $u_{\text{inf}}$: Minimum fluidization velocity, (cm/s)
- $u_{g}$: Relative gas velocity, $u_{\text{gas}}/u_{\text{inf}}$, (-)
- $\tau_{c}$: Particles axial circulation time, (min)
- $\bar{v}_{\text{up}}$: Average particles velocity in the upward direction, (cm/min)
- $\bar{v}_{\text{down}}$: Average particles velocity in the downward direction, (cm/min)
- $z$: Vertical bed axis, (cm)
- $z_{\text{dis}}$: Bed height at which the immersed distributor is located, (cm)
- $z_{\text{sc,1x}}$: Bed height at which the section change is located, (cm)
- $z_{\text{sc,2x}}$: Section change location used in previous studies [6–9], (cm)
- $z_{\text{sc,2x}}$: Section change location in the present reactor configuration, (cm)

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