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# Bubbly flow through fixed beds contactors. Experiments and modelling in the dense regime

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## Abstract

Multiphase reactors operated in fixed bed configuration are widely used in petrochemical industry but their hydrodynamic is still not well understood. In a previous paper (Bordas et al, 2006), a new one-dimensional model able to predict the pressure drop and the mean void fraction for bubbly flows in packed beds was proposed. The equations required closure laws accounting for the liquid-solid and the gas-liquid interactions. Compared to previous models, these laws had been completely revisited, in order to better account for the flow dynamics at the pore scale. In particular, it had been demonstrated that (i) in dilute conditions, the bubble size remains of the order of the pore (ii) and that the mean bubble dynamics is somewhat similar to that of a slug, with a relative velocity at meso-scale linearly increasing with the liquid superficial velocity. Besides, that relative velocity monotonically increases with the gas flow rate ratio, a behaviour that is tentatively attributed to the formation of preferential paths for the gas phase. (iii) Based on the motion of a bubble train in capillary tubes, the two-phase flow pressure drop  $f_{ls}$  scaled by its single-phase flow counterpart  $f_{ls\phi}$  at the same superficial liquid velocity is predicted to linearly increase with the void fraction, with a prefactor  $\lambda$  evolving with the Capillary number:  $\Psi = f_{ls} / f_{ls\phi} = 1 + \lambda(Ca)\alpha$ . Considering the capillary pressure contribution to the pressure drop, one expects  $\lambda \sim Ca^{-1/3}$  [Bretherton 1961]. These proposals were only partly validated and required further confirmation: Thus, new experiments have been performed at LEGI and at IFP in order to cover an enlarged range of flow conditions while gathering all the necessary information to test the model. Attention has been paid to the measuring techniques, both to understand the exact meaning of the measured quantities and to control the uncertainty. So far, the results confirm the postulated dynamic. The model presented assumes that all the bubbles contributing to the void fraction are mobile while in some circumstances, bubbles can be trapped within the bed. Fixed bubbles can also contribute to the pressure drop, and the closure proposed does not take such contributions into account. This question was addressed by way of a thorough data analysis. Data gathered on the void fraction and pressure loss shows that the proposed closure laws for the gas dynamic and two-phase flow pressure drop are still relevant for moderate pressure conditions (up to 10 bars) and gas volumetric ratio  $\beta$  ranging from 20% to 70%. In the range of operating conditions considered (capillary number, gas ratio  $\beta$ ), bubbles-bubbles interactions probably limit such blockage. Those interactions would explain also why the size distribution remains controlled by the pore characteristic dimension, through a process of continuous coalescence/breakup. Using these closures, the model sensitivity was studied and its ability to predict the void fraction and pressure loss addressed.

## Introduction

Dispersed two-phase flows in confined geometry such as packed beds are widely used in the chemical and the bio-chemical industry. Indeed, this type of reactor is often preferred to mobile beds because of its high conversion efficiency for rather low investments and maintenance costs. Yet, reliable tools are lacking for dimensioning and optimizing such systems. Many correlations are proposed in the literature aiming at predicting the pressure loss and the gas/liquid retention in packed beds, but they provide poorly reliable results when extrapolated to operating conditions outside the range for which they were established. Such drawbacks demonstrate that some hidden parameters accounting for the complexity of the phenomena occurring at the pore scale are missing. Another practical issue concerns the occurrence of improper functioning in these systems, which arise from maldistributions whose origin is still unclear.

To address these questions, an accurate modelling of the hydrodynamics of bubbly flows in packed beds is needed. Attempts have been made in that direction using mechanistic approaches. In particular, Attou et al. (1999)

were the first to propose a model inspired from an Eulerian two-fluid formulation. Yet, their model suffered from serious drawbacks, and we therefore proposed a new one dimensional two-fluid model that better accounts for phenomena arising at the pore scale (Bordas et al. 2006). The first comparisons with experiments proved encouraging. Yet, they were achieved over a limited range of flow conditions. The purpose of the present contribution is to test further the validity of our proposal using new data gathered in two different experimental facilities.

## Summary of the 1D model

The model itself was presented in Bordas et al (2006). It was grounded on local instantaneous Eulerian two-fluids balance equations averaged at a meso-scale scale. That meso-scale was selected so as to be intermediate between a pore characteristic size and the outer dimensions of the fixed bed. Accordingly, all the relevant variables were averaged at that meso-scale (this averaging is denoted by  $\langle \cdot \rangle$  in the sequel). The main features of this model are recalled here. Assuming quasi one-dimensional (along the z direction), and stationary motions at meso-scale for both phases, the continuity balances are automatically satisfied,

and the momentum balances for each phase writes:

$$-\frac{dp}{dz} = \rho_L g + f_{LS} - \left\langle \frac{M_z}{1-\alpha} \right\rangle \quad (1)$$

$$\left\langle \frac{F^*}{V_p} \right\rangle - \left\langle \frac{M_z}{1-\alpha} \right\rangle = -\rho_L g + \left\langle R_1 [\nabla p - \nabla \cdot \tau] \right\rangle \quad (2)$$

In the above equations,  $z$  is directed along the main motion so that  $-dp/dz$  and the liquid-solid force density  $f_{LS}$  are always positive quantities. Moreover,  $g$  should be understood as  $-|g| \cos(z,g)$  where  $(z,g)$  denotes the angle between the main motion direction and gravity. The average momentum exchange term  $\langle M_z/(1-\alpha) \rangle$  corresponds to the liquid-gas force density  $f_{LG}$ .  $F^*$  denotes the average resisting force acting on a test bubble of volume  $V_p$ . The remaining term  $\langle R_1 [\nabla p - \nabla \cdot \tau] \rangle$  represents an additional contribution of the unconditional continuous phase stress to the generalized Archimedian force acting on the bubble. Indeed, the operator  $R_1$  is of the order  $(a/L)^2$ , where  $a$  is the inclusion dimension and  $L$  the characteristic length scale of the unconditional continuous phase flow. In packed beds,  $a$  and  $L$  are of the same order since the bubble size happens to be comparable to that of the pore (at least, when the characteristic pore dimension is smaller than the capillary length - see Bordas et al. (2006), Jo et al. 2009). Therefore, the contribution  $\langle R_1 [\nabla p - \nabla \cdot \tau] \rangle$  cannot be neglected and the set of equations (1) and (2) must be kept as such. The drastic simplifications of the momentum equations that arise when  $a \ll L$  (for example for bubbly flows in large tubes or columns) are not feasible here. The set of momentum balances (1) and (2) would allow the determination of the pressure loss and of the mean void fraction  $\langle \alpha \rangle$  in a fixed bed if the closures for the four terms  $f_{LS}$ ,  $f_{LG}$ ,  $\langle F^*/V_p \rangle$  and  $\langle R_1 [\nabla p - \nabla \cdot \tau] \rangle$  are available. As shown in Bordas et al. (2006), the later three closures cannot, for the time being, be made explicit in terms of the considered meso-scale variables. To by-pass this limitation, the equation (2) which amounts for the gas phase dynamics, has been replaced by a kinematical relationship relating the average void fraction  $\langle \alpha \rangle$  with the volumetric gas ratio  $\beta$  and with the apparent relative velocity  $\langle U_r \rangle$ . Namely:

$$\frac{\beta}{1-\beta} - \frac{\langle \alpha \rangle}{1-\langle \alpha \rangle} = \frac{\langle \alpha \rangle \varepsilon \langle U_r \rangle}{V_{SL}} \quad (3)$$

Here,  $\varepsilon$  is the porosity,  $\beta$  is defined as  $V_{SG}/(V_{SG}+V_{SL})$ ,  $V_{SL}$  (respectively  $V_{SG}$ ) is the liquid (respectively the gas) superficial velocity (the superficial velocities are estimated as the volume flow rate divided by the entire cross section of the column). The apparent relative velocity  $\langle U_r \rangle$  involved in equation (3) is defined as the difference between the dispersed phase axial velocity averaged at meso-scale ( $\langle u \rangle$ ) and the continuous phase axial velocity averaged at meso-scale ( $\langle v \rangle$ ). In view of experimental evidences,  $\langle v \rangle$  was equated to the mean liquid velocity in a pore, i.e.  $V_{SL}/\varepsilon$  (Bordas et al. 2006). On another hand,  $\langle u \rangle$  is not readily accessible and the apparent relative velocity  $\langle U_r \rangle$  is the new unknown.

An additional simplification of eq.(1) can be done. Indeed, the average momentum exchange term  $\langle M_z/(1-\alpha) \rangle$  is

expected to be of order  $\langle \alpha(F^*/V_p) \rangle$  (Achard and Cartellier, 2000). Therefore, and at least for small to moderate void fractions, it can be assumed that its contribution to equation (1) can be neglected compared with  $f_{LS}$ . In these conditions, the model combining the equations (1) and (3) should be completed by the closures for the two quantities  $\varepsilon \langle U_r \rangle / V_{SL}$  and  $f_{LS}$ . The closures proposed in Bordas et al (2006) for these two quantities, which were based on the physical mechanisms occurring at the pore level, are recalled hereafter.

Concerning the apparent relative velocity, one expects that bubbles with a size comparable to that of the pore should behave similarly as slugs i.e. strongly confined gas inclusions, that is their relative velocity should be proportional to the continuous phase mean velocity. However, in single capillary ducts, the resistance to the slug motion is controlled by the liquid flow around the bubble, so that the ratio  $\langle U_r \rangle / V_{SL}$  is a Capillary ( $Ca = \mu_L (V_{SL}/\varepsilon) / \sigma$ ) dependent quantity. In fixed beds, the liquid can by-pass the bubble using nearby connected pores, and that feature leads to a ratio  $\langle U_r \rangle / V_{SL}$  that is insensitive to  $Ca$  as shown in Bordas et al. (2006) by the analysis of various data sets from the literature with the help of the kinematic relation (3). That analysis also shown that this ratio monotonously increases with the gas flow rate fraction  $\beta$ . For  $\beta$  below 0.6, the increase was nearly linear, and follows the trend:

$$\varepsilon \langle U_r \rangle / V_{SL} = f(\beta) \quad \text{with} \quad f(\beta) = 6.7\beta - 1.9 \quad (4)$$

where the coefficients have been identified from four data sets corresponding to various flow conditions (Bordas et al. 2006). This peculiar behaviour may be interpreted as an evolution of the bubble size with the gas content. Alternatively, and more probably, it could be the trace of a channeling effect. Whatever its origin, the combination of the kinematic law along with the above closure for  $\langle U_r \rangle$  allows to predict the mean void fraction  $\langle \alpha \rangle$  for known phasic superficial velocities  $V_{SL}$  and  $V_{SG}$ .

Concerning liquid-solid force density  $f_{LS}$ , and using an analogy between the bubble motion in a packed bed and bubble trains in capillaries, it was expected that the two-phase flow pressure drop  $f_{LS}$  scaled by its single-phase flow counterpart  $f_{LS1\phi}$  at the same superficial liquid velocity should linearly increase with the void fraction, namely:

$$\psi = f_{LS} / f_{LS1\phi} = 1 + \lambda \langle \alpha \rangle \quad (5)$$

Here  $f_{LS1\phi}$  is given by the Ergun law:

$$f_{LS1\phi} / (\mu_L V_{SL} / d_{beads}^2) = [(1-\varepsilon)^2 / \varepsilon^3] [A + B \cdot \text{Re}_p] \quad (6)$$

with  $A=180$  and  $B=1.8$  for a rhombohedral arrangement according to MacDonald et al (1979), and with a pore liquid Reynolds number defined as  $\text{Re}_p = \rho_L \cdot V_{SL} \cdot d_{beads} / [\mu_L (1-\varepsilon)]$ . Moreover,  $\lambda$  is expected to evolve with the Capillary number, according to a scaling  $\lambda \sim Ca^{-1/3}$  (Bretherton (1961). The analysis of available data has demonstrated the validity of the expected linear variation of  $\psi$  with the averaged void fraction  $\langle \alpha \rangle$ . In addition, the coefficient  $\lambda$  was found to

decrease with the Capillary number according to a power law, with an exponent close to the expected value of  $-1/3$  (Bordas et al. 2006).

Although encouraging, the underlying hypotheses need to be ascertained and the range of validity of the proposed closures deserves to be investigated. Among the main issues, let us recall that the proposed model is based on the assumption of a fixed "mean" bubble size which uniquely controlled by the packed bed geometry when the pore scale is much smaller than the capillary length scale. Such a behaviour has been demonstrated in dilute conditions (up to 0.02 in void fraction, Bordas et al. 2006) and recently confirmed by Jo and Revankar (2009) on a 2D experimental setup. However, the question remains open for dense regimes. In particular, the increase of the apparent relative velocity with  $\beta$  may be attributed either to a change in the bubble size or to an evolution of the bubble dynamics because of the formation of preferential paths. In the same perspective, some effects of the absolute pressure level on the flow dynamics have been reported in the literature. It is not clear whether bubbles expanding in a strong pressure gradient continuously break to keep their equilibrium size or whether their dynamics is actually modified. Concerning the limitations of the proposed closures, the proposed model predicts a void fraction less than the gas flow rate fraction for  $\beta$  above 10 to 20 %. In experiments, an opposite trend is systematically observed at lower gas flow rates that may be due to measurement uncertainties or that may correspond to a different gas dynamics, such as for example trapped bubbles. Indeed, the proposed model assumes that all the bubbles contributing to the void fraction were mobile (the void fraction involved in the kinematical relationship (3) corresponds to mobile bubbles only). Yet, it is known that bubbles can be trapped within the bed, and such bubbles may alter the void fraction - gas flow rate fraction relationship. They can also bring a contribution to the pressure drop, which is not accounted for by the closure proposed in equation (5).

To address these questions, new experiments have been performed in order to cover an enlarged range of flow conditions and flow parameters while gathering all the necessary information to test the model. In that scope, a great attention has been paid to the measuring techniques, both to understand the precise meaning of the measured quantities, and to control the uncertainty.

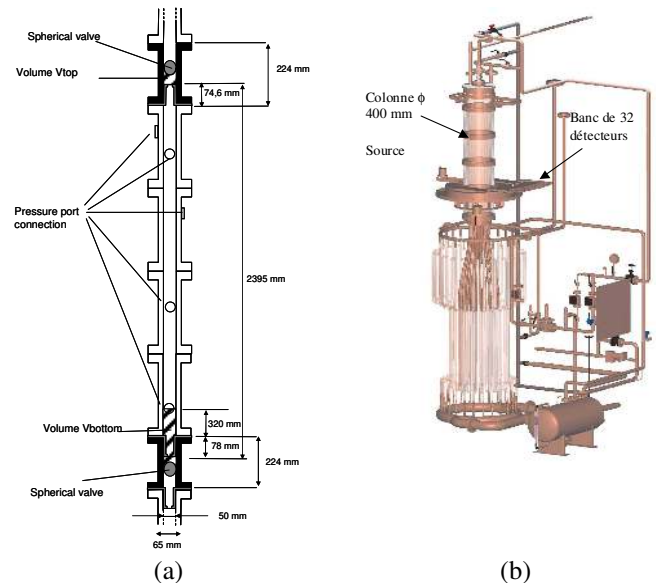
### Experimental Facility

To gather new sets of experimental results, experiments have been performed in parallel at LEGI and at IFP.

*LEGI experiment.* The experiments at LEGI (denoted Exp-LEGI in the sequel) have been performed in a 5 m high vertical cylindrical column with a 50 mm inner diameter. Air-water co-current, *upflow* conditions were considered. The column was made of plexiglass, except its lower part which was made of steel because of pressure constraints. The measurement section was 2.395 m long. It was completely filled with glass beads (diameter 2 mm) and this packed bed was tightly maintained between two fixed grids. The geometrical arrangement was close to be rhombohedric

and the porosity equal to  $\epsilon=0.34$ .

The gas was injected at the bottom of the column through a porous material, that allowed the formation of small bubbles, with a typical diameter about one millimeter. In order to ensure almost fully developed conditions from the very beginning of the fixed bed, the bubbles produced by this porous material were forced to break-up in a short (20 cm long, representing about 100 bead layers) packed bed socket inserted in the steel tube and located 15cm upstream the main bed entrance. Since the bead diameter and the arrangement were the same in the socket and in the test section, one expects that the bubbles have already reached their equilibrium size (as stated in Bordas et al. 2006) before entering the measuring section.



**Figure 1 : Experimental facilities (a)LEGI (b) IFP**

The measured quantities consisted of the pressure drop along the test section and of the mean void fraction. The absolute pressure level was recorded at the level of the gas injector. The mean void fraction was accessible by way of the gas volume fraction measurement. The later was achieved using two quick-closing ball valves located at each extremities of the measuring section, that is about 2 m and 4.5 m above the gas injector respectively. The simultaneous closure of the valves was ensured by a mechanical link driven by a pneumatic command. The closure time was sufficiently short (less than 0.1 s) for the flow conditions considered. The void fraction measurement requires the determination of the volume of liquid trapped inside the test section. Because of the column design, the liquid volume between the two valves could not be directly measured. Instead, the measuring section was drained using the lower pressure port. Sufficient time was allowed for the draining process to be completed. The volume comprised between that location and the upper valve was emptied and the corresponding liquid volume was measured by weighting. Knowing the available free volume within this test section, the mean void fraction could be estimated. The fact that all the measuring section is not drained is the main factor of uncertainties : indeed the gas trapped below the draining device can move in the measuring section. An upper and lower estimation of this uncertainty was computed assuming

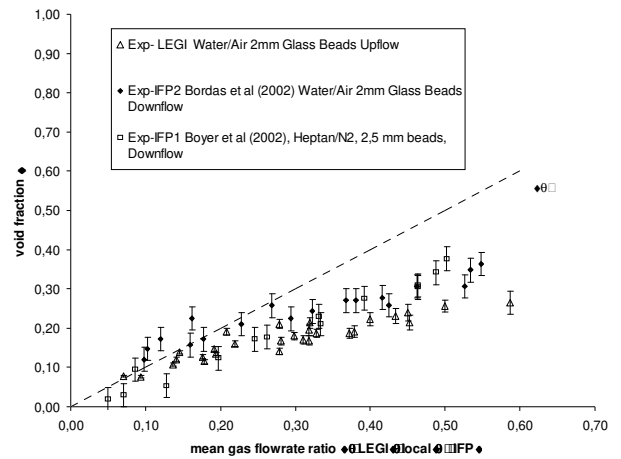
that all the gas moved in the measuring section or remained trapped outside the measuring section. Another source of uncertainties is the residual liquid which remains trapped within the test section when the column is drained. This source of uncertainty was estimated using results found in Ortiz-Arroyo et al (2003) : from the results found in this publication, the static liquid hold-up is estimated to be about 5% and the corresponding uncertainty on the void fraction was computed accordingly.

*IFP experiment.* The experiments at IFP were performed for co-current *downflow* conditions. The experimental facility was composed of a 400mm I.D. and 2.250 m long vertical column. The packed bed was installed over a height of 1.55 m. Experiments were performed either with heptane and nitrogen with 2.5 mm beads (refer to as Exp-IFP1 in the sequel, and with water and air with 2 mm beads (refer to as Exp-IFP2 in the sequel). The porosity was measured considering the weight of the beads filling the column. It was found equal to 0.348 for Exp-IFP1 and 0.35 for Exp-IFP2. The pressure drop between different sections was measured with differential pressure sensors (range 0-300 mb and 300-3000mb, relative uncertainty less than 2.5%) installed along the test section. The absolute pressure level was also recorded at the column inlet. The average void fraction was measured by  $\gamma$ -ray tomography at a location 80 cm downstream the injector. The  $\gamma$ -ray tomography, which was developed at IFP, provides a time-averaged value of the gas phase fraction in a cross section. The measurement principle is described in Boyer et al (2002) and a analysis of the uncertainties can be found in Boyer and Koudil (1999). The main source of error arise from statistical errors due to random  $\gamma$  photons emission, from the dynamic bias associated with the gas fraction fluctuations and from reconstruction errors. The intrinsic performance of the system with regards to these potential error sources was studied in Boyer et al. (2000) and Boyer et al. (2001). As shown in Boyer et al (2000), the maximum absolute error on the gas fraction measurement is 3%.

## Results

### Void fraction

Figure 2 shows the evolution of the void fraction  $\alpha$  with  $\beta$  for the three sets of experiments. For the IFP experiments, the void fraction in the ordinate represents the mean gas surface fraction in a cross section located 800 mm downstream the bed entrance. Accordingly, the  $\beta$  parameter has been computed in the same cross-section by accounting for the absolute pressure level. The uncertainties on  $\alpha$  have been reported in figure 1. For the LEGI experiments, the void fraction represents the gas volume fraction (relative to the total pore volume) between the two quick-closing valves. Clearly, the volumetric gas ratio  $\beta$  evolves along the column height because of the change in the absolute pressure. Since the flow conditions are diabatic, one can compute  $\beta(z)$  for known phasic mass flow rates and from the measurements of the pressure drop and the absolute pressure at the gas injection.



**Figure 2 : Evolution of the void fraction  $\alpha$  with  $\beta$**

For each operating conditions, the reference volumetric gas ratio  $\beta$  used in figure 1 was taken as its mean value between the bed inlet and outlet. For sake of clarity, the horizontal extend of the uncertainties on  $\beta$  are not drawn on figure 2. Lets stress that the variation on  $\beta$  between the inlet and outlet are such that in some configurations,  $\beta$  can be multiplied by a factor 3. The uncertainty on  $\alpha$  was computed according to the error sources identified in section *Legi-Experiment*. In both experiments, the flows are stationary and fully developed. Therefore, neglecting flow inhomogeneities that may arise close to external walls, the gas surface or volume fractions provide indeed a measure of the void fraction.

Whatever the experiment or the flow conditions considered, the void fraction  $\alpha$  is always significantly less than the gas flow rate fraction  $\beta$  provided that the later is not too small (say for  $\beta$  above about 0.15). This trend is the same as the one already noticed by Bordas et al (2006), one observes  $\alpha < \beta$ . In addition, both data series that correspond to upflow and downflow behave very similarly, indicating that gravity is not a key parameter governing the bubble dynamics.

### Relative velocity

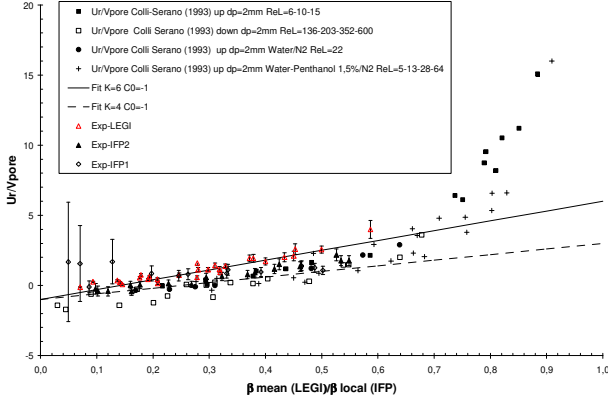
The kinematical relationship (3) was then used to derive the quantity  $U_r/V_{pore}$  where  $V_{pore}$  is the interstitial velocity defined as  $V_{SL}/\epsilon$ . The results are shown on figure 2 along literature data already presented in Bordas et al. (2006).

As stated in Bordas et al (2006), only data's in the range  $0.2 < \beta < 0.7$  can be interpreted with confidence in view of eq.(3). Below  $\beta=0.2$ , the uncertainty on the void fraction can be significant depending on the sensitivity of the measurement techniques. Moreover, as stated in Bordas et al (2006) and in the beginning of this paper, in this range, the proportion of blocked bubbles can be significant and add a supplementary source of error on the estimation of the void fraction associated to mobile bubbles. However, despite these uncertainties, in the range  $0.2 < \beta < 0.7$ , for which the kinematical relationship is assumed to be valid, in figure 3 the same trends as in Bordas et al, (2006) are observed and  $U_r/V_{pore}$  happens to a be a unique function of the gas flow rate fraction:



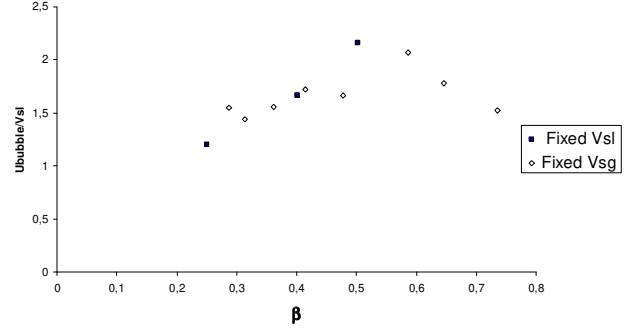
$$\frac{U_r}{V_{pore}} = f(\beta) = K\beta + C_0 \quad (7)$$

with the prefactor K independent on the superficial velocities.



**Figure 3 : Evolution of  $U_r/V_{pore}$  with  $\beta$**

This linear behaviour accounts for downflows as well as for upflows (see Coli-Serano data, along those taken in LEGI and IFP on figure 3). Therefore, at the first order,  $U_r/V_{pore}$  is not only a unique linear function of  $\beta$ , but the K coefficient seems not to depend on the flow direction. That gives weight to the assumption of a “slug-like” bubble dynamic. The linear fit presented in Bordas et al. (2006) gave  $K=6.7$  and  $C_0=-1.9$ . This fit was performed on all data including data point below  $\beta=0.2$ . As the bubble dynamic in a capillary can be expressed as  $U_{bubble}/V_{pore} = C$  or  $U_r/V_{pore}=C-1 \cdot V_{pore}$ , we get  $C=K \cdot \beta$  and so  $C_0=-1$ . Assuming that  $C_0=-1$ , the “best” fit corresponds to  $K=5$ . Two others values for K or  $C_0$  are also proposed on the figure: given the uncertainties on the measurements, they correspond to the probable domain of variation of K in the range  $0.2 < \beta < 0.7$ . The model sensitivity to these values of K will be analyzed further when it will be compared to Attou's model. Given the experimental results, the C coefficient depends on  $\beta$  ( $C=K \cdot \beta$ ) with value ranging from 1 to 3.5 ( $K=5$ ) in the range  $0.2 < \beta < 0.7$ . For low  $\beta$  these values correspond particularly well on the typical value of the C coefficient for an isolated slug in capillaries. Figure 3 presents results gained from a post-processing of data given in Jo and Revankar (2009). In this paper, the size and dynamic of bubbles moving in a 2D experimental setup was studied. Bubbles velocities were directly measured using a high speed camera. At a given liquid superficial velocity, it is found that the ratio  $U_{bubble}/V_{sl}$  ranges from 1.2 to 2.5 with increasing  $\beta$  ranging from 25% to 50%. At a given superficial gas-velocity, results are less monotonous (see figure 4), but the ratio  $U_{bubble}/V_{sl}$  globally increases with  $\beta$ , for  $\beta$  ranging 30 to 60%: this behaviour is in accordance with the present results.



**Figure 4 : evolution of bubble size with  $\beta$  in a 2D in packed bed made of cylinder (Jo and Revankar, 2009)**

Above that value, the bubble velocity behaviour experiences a drastic change but the tendency is opposite to the one observed on figure 3. In our case, this may be attributed to the fact that above 60-70%, we are no longer in bubbly configuration.

The bubble velocity increase with the volumetric gas ratio  $\beta$  seems then to be a dynamic specific to packed bed. The physical processes underlying this behaviour are however difficult to ascertain. The proposed closure deals with the mean behaviour of the bubble cloud and is based on average global parameters. Locally, the bubble dynamic is strongly influenced by the local fluid velocity which can experience strong fluctuations (fluid redistribution in adjacent interconnected pore, preferential path...): those fluctuations are strongly connected to the dispersed phase content.

#### Liquid-solid interaction and model closure

In this section, the objective is to test further the propositions made in Bordas et al. (2006) concerning the liquid-solid interaction term. From the total pressure loss and void fraction measurement, the two-phase pressure loss  $f_{ls}^*$  is expressed as:

$$\begin{aligned} \text{upflow} \quad f_{ls}^* &= |dp/dz|_{measured} - (1-\alpha) \rho_L \cdot g \\ \text{downflow} \quad f_{ls}^* &= -|dp/dz|_{measured} + (1-\alpha) \rho_L \cdot g \end{aligned} \quad (8)$$

In the above equation,  $f_{ls}^*$  holds for the quantity  $f_{ls} - \langle Mz / (1-\alpha) \rangle$  (see equation (1)). The ratio  $\psi$  between the two phase pressure drop  $f_{ls}^*$  and its counterpart in single phase flow  $f_{ls1\phi}$  at the same superficial liquid velocity was plotted as a function of  $\alpha$  for various pore Reynolds numbers  $Re_p$ . The results are presented in figure 5a and figure 5b.

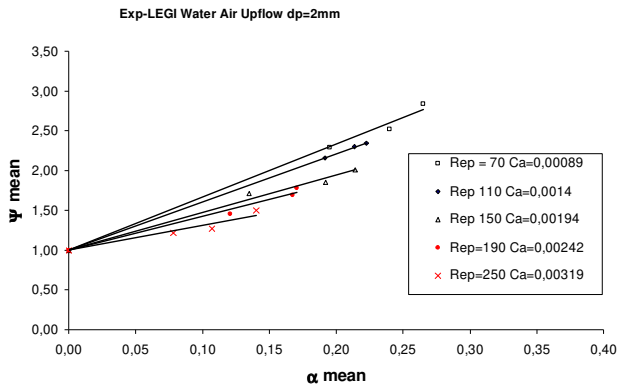


Figure 5a : evolution of  $\psi$  with  $\alpha$ . Upflow (Exp-LEGI)

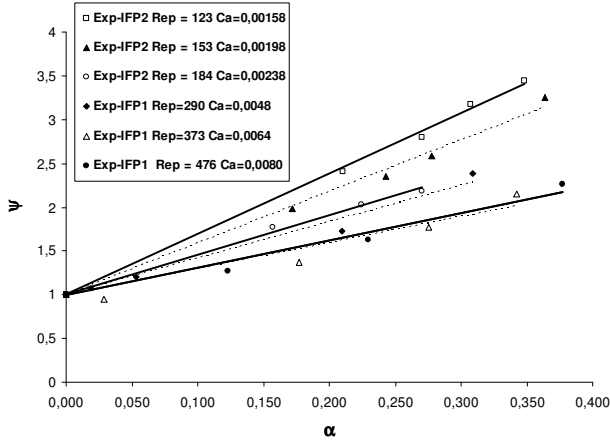


Figure 5b : evolution of  $\psi$  with  $\alpha$  Downflow. (Exp-IFP1 and ExpIFP2)

At first order, the linear evolution of  $\psi$  for  $\alpha < 0,40$  suggested in Bordas et al (2006) is confirmed whatever the flow direction:

$$\psi = 1 + \lambda(Rep).\alpha \quad (9)$$

This result gives weight to the physic postulated at the pore scale and is consistent with the assumption that the two phase pressure drop is mainly controlled by the liquid-solid friction term  $f_{ls}$  and that the gas-liquid interaction term  $\langle M_z / (1-\alpha) \rangle$  has a weak contribution. This behaviour is also encountered for two-phase pressure drop in duct (Rivière and Cartellier, 1999). In the sequel the term  $f_{ls}^*$  will be assimilated to  $f_{ls}$ .

The effect of the Reynolds number on  $\psi$  (or  $\lambda$ ) is also clearly apparent. But if we keep the analogy between bubbly flow in fixed bed and two-phase flow in capillaries made in Bordas et al (2006), this number may not be the correct scaling parameter and, following the analysis of Betherton (1961), the capillary number should be involved too. Figure 6 shows the evolution of  $\lambda$  with the capillary number  $Ca = (V_{SL} \cdot d_p) / (\epsilon \cdot \sigma)$  where  $d_p$  is the beads diameter,  $\epsilon$  the bed porosity,  $\sigma$  the surface tension and  $V_{SL}/\epsilon$  the pore velocity.

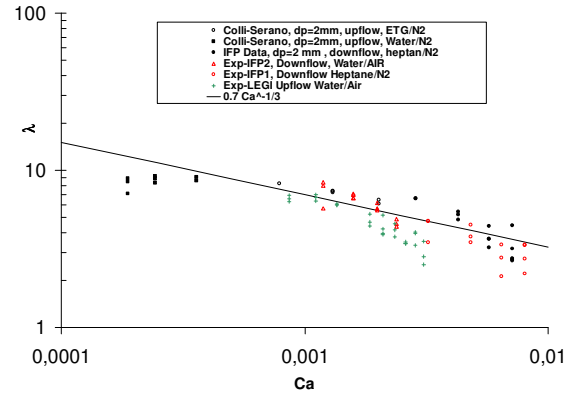


Figure 6 : evolution of  $\lambda$  with Ca.

On this figure, are plotted data from literature and the present data. For these data, at a given liquid flow rate, several gas operating conditions were tested. For Exp-LEGI data, the value of  $\alpha_{mean}$  was used in the calculation of  $\lambda$ . At a fixed capillary number, the discrepancy on  $\lambda$  value is not so high which seems to validate the  $\lambda$  dependency on Ca alone at the first order and the closure law structure presented above for fls. Let's notice also that as  $\lambda = (\psi-1) / \alpha$ , its value is very sensitive to the accuracy of the void fraction measurement. This explains the discrepancy of data when all the data are taken as a whole. However, the general trend corresponds to a global variation with  $Ca^{-1/3}$ .

Strictly speaking the pore velocity should be corrected by the space available for the fluid inside the pore after accounting for bubbles:  $Ca = (V_{SL} \cdot d_p) / (\epsilon \cdot \sigma \cdot (1-\alpha))$

Thus equation (9) should write

$$\psi = 1 + \lambda(Ca, \alpha) \cdot \alpha \quad (10)$$

and the model is not completely linear with the void fraction. Figure 7 shows the evolution of  $\lambda$  with the corrected Capillary number

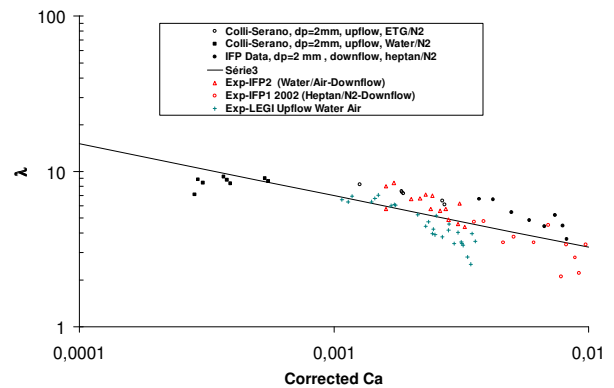


Figure 7 : evolution of  $\lambda$  with the corrected Ca

Given the uncertainties on the data and sensitivity of  $\lambda$  with  $\alpha$ , the trends already observed are not changed and if there are any non-linearity due to the void fraction, its effect seems weak and not easily noticeable from global measurements.

## Model testing and discussion

### Model consistency with the bubble dynamic in packed bed: the problem of the static void fraction

Although void fraction measurements were not performed with similar techniques at IFP and at LEGI, both methods give access to the total voidage in the packed bed. The total void fraction can be decomposed as a dynamic void fraction and a possible static void fraction due to bubbles blocked in the granular media. The model presented above considers only the dynamic void fraction. The relative importance of this static void fraction is fundamental to assess conclusions presented here and to estimate the limits of validity of the model presented above.

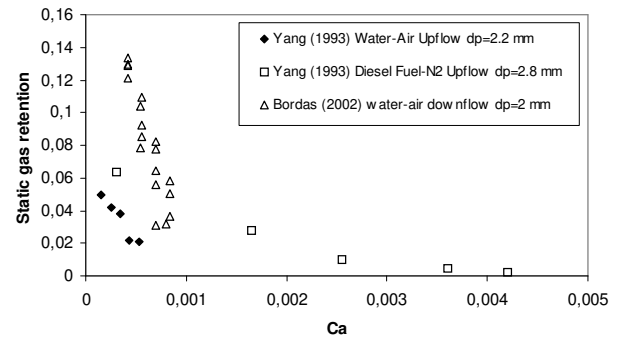
Liquid saturation measurements performed by Larachi et al. (1991) showed a different behavior depending on the liquid viscosity. For high viscous liquid, the liquid saturation goes toward 1 (i.e. the void fraction goes toward zero) when the gas flowrate goes toward zero. However, for low viscous liquid, the void fraction goes toward a non zero value when the gas flow rate decreases toward zero. This different behaviour is probably due to blocked bubbles. The shear rate is higher for viscous liquid, which favours the bubble detachment. To characterize this phenomenon, the capillary number seems to be the appropriate dimensionless number.

Measurement performed in LEGI on the index matching column presented in Bordas et al. (2006) showed that, below a given critical Reynolds number, a fraction of the bubble population was blocked in the porous media. This critical Reynolds number depends on the fluid used (Cyclooctene, Cargille code 5095). For experiments performed with cyclooctene (which has a low viscosity) and nitrogen, in the dilute regime, it was shown that the critical Reynolds number was  $Re_p=170$  (Bordas (2002)). This critical Reynolds number corresponds to a capillary number  $Ca=2.76e-03$ . In the case of the Cargille liquid, whose viscosity is ten times greater than the cyclooctene viscosity, no blockage was observed (Bordas (2002)). The operating conditions lied in the range of  $Re_p=8$  to 64 which corresponds to Capillary numbers  $Ca=0.03$  to 0.074. These values are above the critical Capillary number defined from the Cyclooctene experiments.

Concerning the experiments presented in this paper (non dilute regime), it was not possible to perform the estimation of the static void fraction in the case of the data acquired in LEGI (the measurement technique didn't allow a visual direct access in the column core). But some tests (for the water-air case performed by Bordas) were carried out at IFP with the tomographic technique for Exp-IFP1 and Exp-IFP2. The methodology was the same as the methodology followed in Yang et al (1993): the reactor was feeded with gas and liquid. After a given time, the gas feeding was stopped and the residual void fraction measured. This fraction can be then compared to the total void fraction in normal operating conditions.

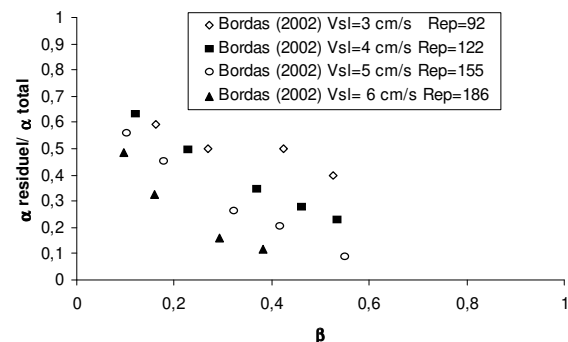
Given the operating conditions, the evolution of the static void fraction with the capillary number is shown on figure 6

along the data of Yang et al (1993).



**Figure 8 : Static gas retention EXP-IFP2 and Yang experiments.**

These later authors used several techniques to measure the liquid saturation : the liquid static retention was measured using gamma-ray adsorption. A careful analysis of their data led to the computation of the static void fraction presented on figure 8. This figure shows that the static void fraction decreases drastically with the capillary number. In downflow, even if this decrease is very steep, the amount of blocked bubbles can be quite significant. Unfortunately, the operating conditions in Yang et al (1993) (namely the gas flow rate) are not completely known to fully compare Yang data and Exp-IFP2 data. Figure 9 present the ratio between the static void fraction and the total void fraction versus the volumetric gas ratio  $\beta$  using Exp-IFP2 data.



**Figure 9 Residual void fraction. Exp-IFP2**

Results are consistent with figure 8. At a given  $\beta$ , the amount of residual gas decreases with the superficial velocity (and thus with the capillary number). This confirms results obtained by visualization on the index matching experiments presented in Bordas et al (2006). In Exp-IFP2, the capillary number lies between 0.001 and 0.0024 which is below the critical capillary number  $Ca=2.76e-03$ . That could explain the relatively high ratio "static void fraction/total void fraction". For Exp-LEG11, the value of the static void fraction is not given as this phenomena could not be quantified. Provided the experimental conditions, the Capillary number lies between, 0.003 and 0.008 which would correspond to no or a weak gas static retention.

The static void fraction decrease with  $\beta$ , for a given superficial velocity, is also consistent with the observations presented in Bordas et al (2002) for the dilute regime and



Larachi et al (1991) observations: the gas flow rate increase leads to more bubbles interactions which eventually leads to detachment. Those interactions can be direct or indirect, as the bubble motion induces pressure and velocity fluctuations in the adjacent pores. Lets point out that the methodology used by Yan et al (1993) (and applied for IFP experiments) leads certainly to an overestimation of the static void fraction presented on figure 8 and 9: measurements are performed when the gas flow is stopped which reduce the effect of bubbles/bubbles interactions.

To conclude, the following remarks can be made:

- whatever the measurement technique, it will be difficult to give a quantitative estimation of the static void fraction in our experiments: although this fraction could be estimated with the tomographic technique, it depends at least on two parameters : the volumetric gas-flow rate (which account for bubble interactions) and the capillary number—(which account for the force balance on the bubble). A specific study, beyond the scope of this paper, should be undertaken to fully understand the mechanism and the coupling leading to bubbles blockage.
- Qualitatively, it can be said that the bias on the data will be less important for the highest superficial velocity, high viscous fluid and above all, for highest volumetric gas ratio (typically > 20%, value corresponding to the lower limit of the model validity range presented in Bordas et al, (2006). For these values, bubbles interactions limit the static void fraction.

- In the worst case (water-air), the maximum ratio between static void fraction and the total void fraction in this  $\beta$  range is around 50%, which seems very high. But this value is probably greatly overestimated because of the measurement methodology, which doesn't account for bubbles interactions when the gas flow.

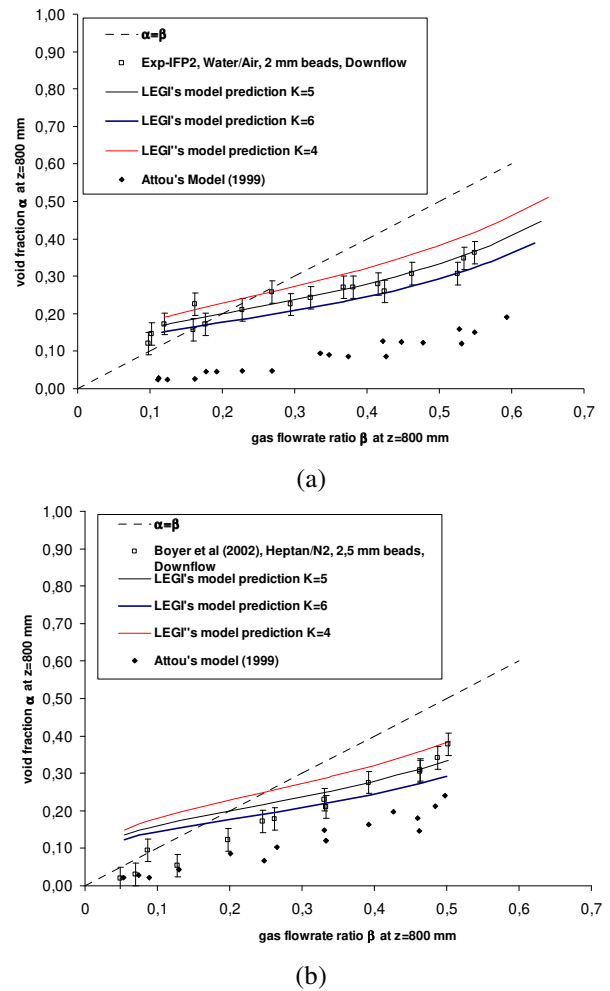
- Results presented in section **Results** are based on the total void fraction measured by both techniques (quick closing valve and tomographic technic). The results (for  $\beta > 20\%$ ) seems consistent with the presented model which doesn't include effect due to blocked bubbles which is coherent with the discussion above. For smaller  $\beta$  ( $\beta < 20\%$ ), some deviation from the model (for example the  $\alpha$  behaviour with  $\beta$ ) could be partly explained by a different dynamic, where the lack of interaction between bubbles could promote gas blockage within the porous matrix.

As a conclusion, the physics invoked in Bordas et al (2006) to express the closure law for the void fraction and the two-phase pressure drop is thus not invalidated by the present experimental data and that physics seems to hold for non dilute regime.

**Model sensitivity and model comparison with Attou's model**

In the section above, the structure of the closure law inferred in Bordas et al (2006) have been validated on experimental data. The model is compared here to another mechanistic model namely Attou's model, which was also based on a two-fluid formulation at the pore scale.

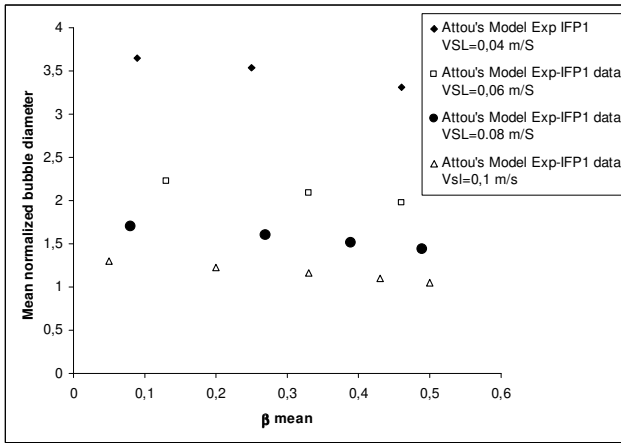
**Void fraction prediction**



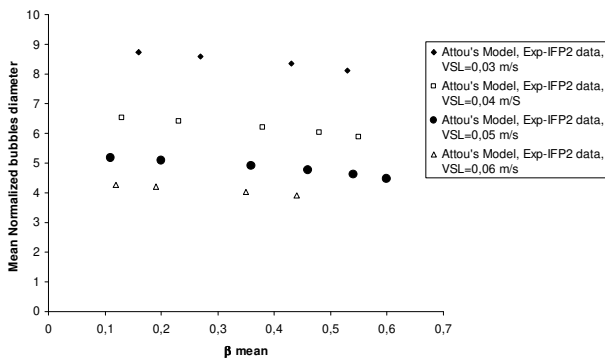
**Figure 10:  $\alpha$  prediction. Comparison with experimental data and Attou's model. (a) Exp-IFP2 (b) Exp-IFP1**

Data from Exp-IFP1 and Exp-IFP2 are plotted on Figure 10a and 10b along prediction given by the model. The aim is to check the model sensitivity to the parameter K. C0 was fixed to -1 for the reasons explained in the preceding paragraph. The model was run with the beads diameter and physical properties of the fluid corresponding to Exp-IFP1 and Exp-IFP2. As parameter K was fit with experimental values including Exp-IFP1 and Exp-IFP2 data, the values K=5 and C0=-1 (which correspond to the "best" fit on figure 9) gives obviously correct predictions. The two others curves (K=6 and K=4) are representative of the maximum error and incertitude on data, when K is fitted from the plot  $U_r/V_{pore}$  versus  $\beta$ . The figure shows that the predictions are sensitive to the parameter K. The bandwidth around the "mean" prediction (K=5) is such that  $\alpha \sim \alpha_{(K=5)} \pm 10\%$ . However, despite the model sensitivity to K, the predictions given for K=6 and K=4 follows the same tendency observed on the experimental data. Furthermore, the bandwidth on the void fraction lies in the range of incertitude of the measured value of  $\alpha$ . Finally, although the incertitude on the parameter K, the model is robust and its relevancy not invalidated in the frame of engineering applications. However, the correct derivation of K requires very accurate data in equation (3), that is to say very accurate measurement on  $\alpha$ . The model is also compared to Attou's model on the same figure. As we can see, the model fails to predict the correct void fraction

behaviour and underestimates the experimental value of both experiments. This difference is attributed to the bubble dynamic implemented in Attou's model which is not correct: the drag force which drive the bubble relative velocity is computed from a drag coefficient modified to take into account hindering effects. Furthermore the characteristic bubble diameter is obtained from consideration about the inclusion break-up by turbulence. Figure 11 shows the bubble diameter evolution with  $\beta$  for each operating conditions for both experiments. As for those operating conditions, the computed  $\beta$  and bubbles diameter doesn't evolve much between the inlet and outlet, results are presented in term of mean value between the inlet and outlet. The diameter is normalized by  $\delta$  the characteristic pore size defined in Bordas et al 2006. The predicted ratio  $db/\delta$  is far greater than 1 for most operating conditions which is not consistent with the hypothesis of hindering effects taken in the model. As well as the experimental results presented in Bordas et al 2006.



(a)



(b)

**Figure 11 : Normalized bubble diameter as predicted by Attou's Model (a) Exp-IFP1 (b) Exp-IFP2**

### $\Psi$ and pressure prediction

As in the case of EXP-IFP1 experiments, pressure profiles along the column were also available, the model was run and the computed pressure profiles were compared to the experimental profiles. Furthermore, compared to the initial proposal made in Bordas et al (2006), the correction of Ca with the void fraction  $\alpha$  introduce a non linearity in the

closure law for  $\Psi$ . As  $\beta$  evolves also with the pressure, the model consistency with quasi linear pressure profiles was checked back

The governing equations are recalled below:

- Kinematical relationship (model equation 1 : EqM1)

$$\frac{\beta(z)}{1-\beta(z)} - \frac{\alpha(z)}{1-\alpha(z)} = \alpha(z)[K \cdot \beta(z) + C_0]$$

with  $K=5$  and  $C_0=-1$  (best fit on figure 8 ).

- ratio between the two-phase pressure drop and the single flow pressure drop

$$\psi = f_{LS} / f_{LS1\phi} = 1 + \lambda(Ca, \alpha(z)) \cdot \alpha(z) \quad (\text{EqM2})$$

with  $\lambda = 0.7Ca(z)^{-1/3}$  (best fit on figure 12 )

$$\text{and } Ca = \frac{V_{SL} \cdot d_p}{\varepsilon \sigma (1 - \alpha(z))}$$

The single flow pressure drop is given by the Ergun law

$$f_{LS1\phi} / (\mu_L V_{SL} / d_{beads}^2) = [(1 - \varepsilon)^2 / \varepsilon^3] [A + B \cdot Re_p]$$

- relationship between the two phase liquid-solid friction and total pressure gradient

$$f_{ls} = -|dp/dz| + (1 - \alpha) \rho \cdot g \quad (\text{EqM3})$$

The effect of pressure on the gas density is taken into account using the perfect gas law. The inlet data were the liquid flowrate at the column injection, the mass gas flowrate at the inlet (or the volumetric gas flowrate given at the normal condition of pressure and  $T^\circ$ ) and the relative pressure at the injection. The corresponding volumetric gas ratio in the measured section at 800 mm from the injection will be recalled on the figure accounting for the results.

Knowing the value of  $\beta(z)$  in a given section, equation EqM1 allows the computation of the corresponding void fraction  $\alpha(z)$ .  $\beta(z)$  is computed knowing the mass gas flowrate at the inlet, the liquid flowrate and the gas density  $\rho(z)$  which depends on the pressure  $p(z)$ . We get:

$$Q_G(z) = \frac{P_{inlet}}{P(z)} \cdot Q_{Ginlet} \quad \text{and} \quad \beta(z) = \frac{Q_G(z)}{Q_L + Q_G(z)} \quad (\text{EqM4})$$

$\alpha(z)$  being known equation EqM2 with its closure allows the computation of  $f_{ls}$ . Then (EqM3) is integrated with space step  $dz$  using a simple Euler scheme to compute  $P(z+dz)$ .

First, the model was compared with Attou's model in term of  $\Psi$  (figure 12).

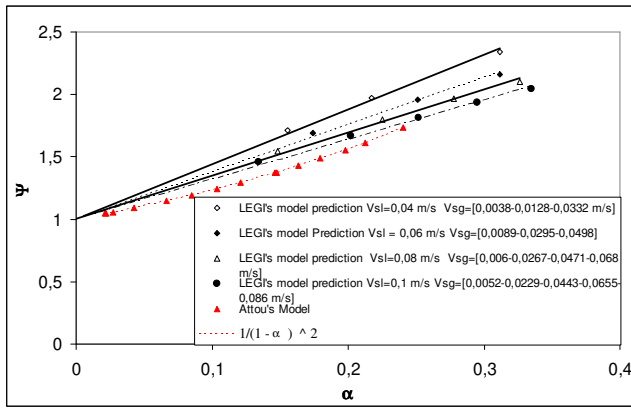
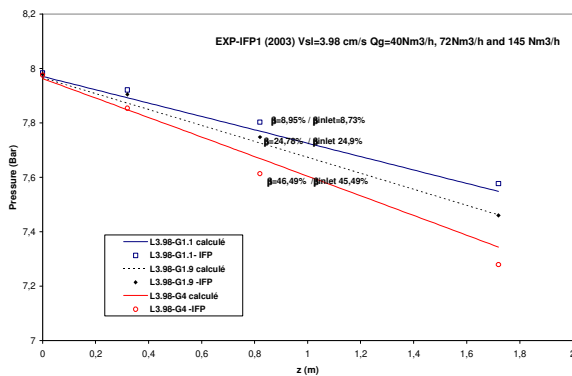


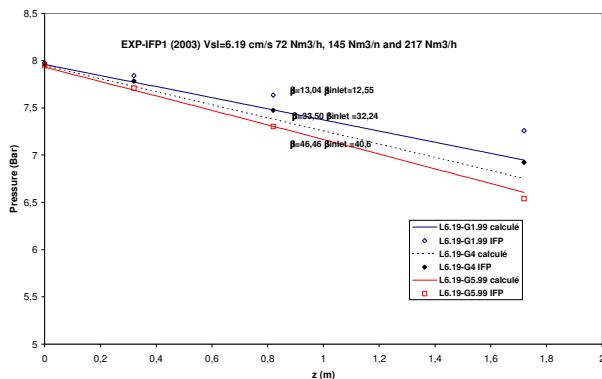
Figure 12 :  $\Psi$  prediction

Although the non-linearity introduced by the evolution of  $\beta$  with the column location and the dependency of the capillary number with  $\alpha$ , the linearity  $\Psi$  with  $\alpha$  and its dependency with the Reynolds number still holds. Prediction of Attou's model are also presented on the figure. Because of its structure, this model predict that  $\Psi$  evolves as  $1/(1-\alpha)^2$ . To distinguish between this behaviour and a linear behaviour can be difficult on experimental data because of measurements uncertainties. However, the Attou's model doesn't allow to recover the dependency of the two-phase pressure drop with the Reynolds number as observed on the experimental data.

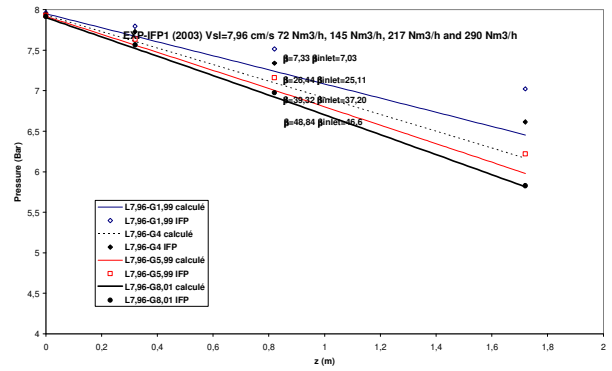
Results on the pressures profile, for the present model, are given on figure 13(a),(b),(c),(d) for each operating conditions.



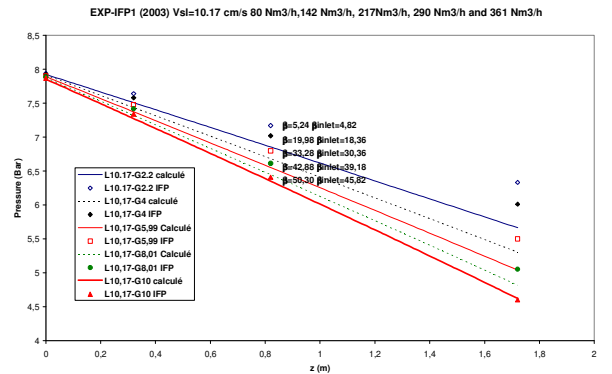
(a)



(b)



(c)



(d)

Figure 13 : Pressure profiles predictions

The results present interesting feature. The pressure profiles are reasonably well predict in the case of high  $\beta$  value whereas the difference between the predicted and measured profiles increases with decreasing  $\beta$ .

Below that value, because of the uncertainties on the measured value of  $\alpha$ , it could be argued that a different bubble dynamic is involved. In Bordas et al (2006) however it was shown that in the dilute regime, the ratio  $Ur/V_{pore}$  was of the order one for bubbles whose size was around the pore size  $\delta$ . The kinematic law should be still valid to low gas fraction ( $U_{bubble}/V_{pore} \rightarrow 1$  when  $\beta \rightarrow 0$  which is compatible with the motion of a bubble in confined capillary) : the model assumes that the momentum exchange and mean bubbles dynamic is controlled by the bubbles belonging to that size class.

In EXP-IFP experiments, the injection system is such (two phase jet) that maybe the equilibrium size distribution is not reached which can explain that the model can't account perfectly for the measured data. However we expect that this effect is more characteristic of high gas fraction that is to say relatively high  $\beta$  at the injection.

For low gas fraction, another phenomena which is probably important is the notion of static/dynamic void fraction evoked in the first paragraph. As said in this part, this static retention, although overestimated because of the measurement procedure, seems to be predominant for low  $\beta$ . It is difficult to estimate the effect of this static gas fraction on the pressure drop. One argumentation, given by Brenkrid (ref) would consist in considering a correction to

the porosity to compute  $f_{ls1\varphi}$ ,

$$f_{ls1\varphi}^{corrected} = f_{ls1\varphi} / (1 - \alpha_{static})^3 \quad (14)$$

However this correction does not take into account the change in the effective solid specific surface due to fluid redistribution in adjacent pore (because of the blocked bubbles). The physics in equation (14) is then too simple and probably doesn't account well for the pressure drop due to the blocked bubbles.

To summarize, even if the discrepancies at low gas fraction, correspond to phenomena not taken into account in the model, the maximum discrepancy between the computed pressure and measured pressure at the column bottom is 20% ( $V_{SL}=10,17$  cm/s  $\beta_{inlet}=4,82\%$ ).

## Conclusions

In Bordas et al (2006), a new one-dimensional model able to predict the pressure drop and the mean void fraction for bubbly flows in packed beds was proposed. The equations required closure laws accounting for the liquid-solid and the gas-liquid interactions. Those closures and some features of the model were deduced from experiments in the dilute regime or from the analysis of literature data. Therefore, the model needed to be validated on a larger set of experiments. In this paper, experiments performed in LEGI and IFP, in the dense regime, were thus presented.

The relationship between the void fraction and the volumetric gas ratio  $\beta$ , as well as the evolution of the apparent relative velocity, confirms a behaviour postulated from the analysis of previous literature data: the overall dynamic is mainly controlled by large bubbles who behave like "slug" in capillaries. Yet, and contrary to bubbles confined in a single duct, the relative velocity at mesoscale happens to be weakly dependent on the local flow organization, because the liquid can freely bypass the gas inclusion through neighbouring channels: such a flow organization almost eliminates any dependency of the relative velocity with the capillary number. However, the analysis of experiments indicates that the relative velocity at mesoscale monotonically increases with the gas flow rate fraction (between 1 up to 4 times the liquid superficial velocity), which seems to be a dynamic specific to packed beds. This phenomenon is tentatively attributed to the formation of preferential paths for the gas, but the physical processes underlying this behaviour are difficult to ascertain: the proposed closure deals with the mean behaviour of the bubble cloud and is based on average global parameters. Locally, the bubble dynamic is strongly influenced by the local fluid velocity which can experience strong fluctuations (fluid redistribution in adjacent interconnected pore, preferential path...). Those fluctuations are in return strongly connected to the dispersed phase content

Concerning the pressure drop in the liquid phase, the latter is mainly attributed to capillary excess pressure due to the presence of bubbles whose size scales as  $\delta$ . Consequently, the ratio of two-phase pressure drop to the one-phase pressure drop at the same superficial velocity is expected to linearly

increase with the void fraction. In addition, the proportionality coefficient should evolve with the capillary number. These anticipations are presently validated on both experiments in LEGI and IFP.

When tested on available pressure profile, the model reproduces relatively well the experimental data, provided that the volumetric gas ratio is sufficiently high, which corresponds to the range of validity of the closure laws when the data were analysed. The discrepancy at low  $\beta$  is attributed to the static gas fraction whose effect on the pressure drop is not taken into account in the model. It is postulated that, because of direct or indirect bubbles/bubbles interactions, this static fraction, in our case (Ca number greater than the critical capillary number), is negligible at higher  $\beta$ .

## Acknowledgements

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