This is a postprint version of the following published document:

Hernández-Jiménez, F., García-Gutierrez, L., Acosta-Iborra, A. & Soria-Verdugo, A. (2019). Numerical study of the effect of pressure and temperature on the fluidization of solids with air and (supercritical) CO₂. *The Journal of Supercritical Fluids*, vol. 147, pp. 271–283.

DOI: [10.1016/j.supflu.2018.11.008](https://doi.org/10.1016/j.supflu.2018.11.008)

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Numerical study of the effect of pressure and temperature on the fluidization of solids with air and (supercritical) CO₂

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Abstract

This work performs numerical simulations of fluidized beds under different conditions of pressure and temperature and using air and CO₂ as fluidizing agents. The conditions of high temperature and pressure tested turn the CO₂ into supercritical conditions, so the differences when the fluidizing agent is at supercritical conditions are also tested. The results show that when pressure and temperature are increased, fluidization with air or CO₂ shifts from the typical bubbling fluidization characteristic of ambient conditions, to a more homogeneous fluidization where not only bubbles and dense phase are present in the bed, but also a dilute phase of moderate solids concentration. The main consequence is an increase of the lateral motion of gas and solids at high pressure and temperature. A deviation from the classical Two-phase theory occurs because the gas velocity through the dense phase at high pressure and temperature is higher than the corresponding minimum fluidization velocity.

Keywords: Supercritical fluid; fluidized bed; two-fluid model.

1. Introduction

Fluidized beds have several applications in chemical and process industries, such as fluid catalytic cracking (FCC), gasification, combustion of solid fuels, Fischer–Tropsch synthesis, drying and coating. Reported experimental works concerning fluidized bed systems are typically performed in lab units operated at ambient conditions. However, most processes in fluidized bed systems occur at conditions of high temperature and/or high pressure and can use either reactant gases or inert gases as fluidizing agents [1]. The conditions of high temperature and pressure can entail some difficulties which make its experimentation complex. Furthermore, if pressure and temperature are sufficiently high, the gaseous fluidizing agent can reach supercritical conditions.

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The isolated effect of pressure on the behaviour of fluidized beds has been widely addressed in the literature. Olowson and Almstedt [2] and Wiman and Almstedt [3] experimentally analysed the effect of pressure on bubbling fluidized beds. Olowson and Almstedt [2] observed that the mean bubble frequency, the mean bubble rise velocity, the average bubble volume fraction and visible bubble flow rate increase with increasing both pressure and excess gas velocity. Wiman and Almstedt [3] developed a dimensionless drag force that allowed the results for the bed expansion, bubble rise velocity, bubble volume fraction and visible bubble flow to collapse in a single curve for different pressure conditions.

More recently, Sidorenko and Rhodes [4] found a decrease of the minimum fluidization velocity with pressure for Geldart B particles. Willem et al. [5] used numerical simulations based on a discrete particle model to analyse fluidized beds at elevated pressure. They stated that the application of Two-phase theory [1] becomes less apparent at elevated pressures, as a consequence of the bubble size reduction found for increasing pressures. Godlieb et al. [6] experimentally found, in a large scale unit, that the emulsion phase becomes less dense and more bubbles and an intermediate phase appear. Brouwer et al. [7] also measured experimentally, using X-ray tomography, that bubble size was significantly reduced at higher pressures for similar gas flows at ambient pressure. Verma et al. [8] employed two-fluid model numerical simulations to analyse the effect of pressure on fluidization in a 3D fluidized bed. They also found a slight decrease of bubble diameter and a change in the solids circulation vortex when pressure was increased. Yates [9] reviewed experimental and theoretical studies of gas-solid fluidization at elevated temperatures and pressures, with special emphasis in circulating fluidized beds, combustion and scaling. Yates [9] concluded that the behaviour of beds operating at high temperatures and pressures is different from that of laboratory scale beds and more research was required to assess these differences, specially for high temperature as the number of works available is lower than for high pressure.

Very few attempts have been done to use fluids in supercritical conditions as fluidizing agents. These attempts concern basically water and CO₂. The applications of supercritical water as gas fluidizing agent include the gasification of wet biomass, coal and waste disposal [10], whereas CO₂ in supercritical state is employed in the coating of particles type C of Geldart classification, mainly in pharmaceutical applications [11]. One of the problems in the fluidization of these kind of particles is the appearance of channelling and the agglomerates formation due to their cohesive character. Thus, this kind of fluidization employing supercritical fluids improves the fluidization of cohesive particles [12]. Furthermore, Vogt et al. [11] proposed several applications for the supercritical CO₂ fluidization such as high temperature hydrogenation reactions or extraction of natural products by studying a bed fluidized with supercritical CO₂ at $p = 80 - 300$ bar

and temperatures near to $T = 55\text{ }^{\circ}\text{C}$.

Some authors have observed that the solids behaviour using supercritical fluids differs from the classical gas-particulate fluidization. Poletto et al. [13] studied the fluidization of solid particles with CO_2 in several conditions to analyse the differences between gas-solid fluidization and fluidization with CO_2 under close to supercritical conditions, which resulted in fluidization patterns more similar to the liquid–solid fluidization. Using water in supercritical conditions instead of CO_2 , Wei and Lu [14] and Lu et al. [15] analysed the fluidization behaviour of a bed fluidized with supercritical CO_2 based on pressure fluctuations and heat transfer between bed and wall, respectively. Wei and Lu [16] showed the existence of an intermediate state between incipient fluidization, without bubbles, and the bubbling fluidization regime. This state consists in a homogeneous bed expansion with more and smaller bubbles compared to the classical bubbling fluidization. Similar behaviour was observed experimentally by Vogt et al. [11] and Marzocchella and Salatino [17] employing supercritical CO_2 as fluidizing agent.

Numerical simulations of fluidized bed systems constitute a complementary tool to experiments, either to analyse variables difficult to measure in experimental units or to test extreme experimental conditions. The most common numerical techniques to simulate fluidized beds are Eulerian–Eulerian Two–Fluid Models (TFM) [18, 19, 20], Eulerian–Lagrangian approaches such as Discrete Element Models (DEM) [21, 22], or a combination of both strategies (coupled TFM–DEM) [23, 24]. The use of these techniques combined with experiments can be very effective to achieve a detailed analysis of the fluid–dynamics and heat and mass transfer of complex gas–solids systems [25, 26]. Rodríguez-Rojo and Cocero [27] made a first attempt to simulate the fluidization of solid particles using CO_2 in supercritical conditions using a TFM. They found a homogeneous fluidization without the presence of bubbles and simplified the model by considering an axisymmetric flow.

The present work performs TFM simulations of fluidized bed systems working under different conditions of temperature and pressure using air and CO_2 as fluidizing agents. The main objective is to analyse the differences in the fluidization behaviour among these conditions. In particular, supercritical conditions are achieved by the CO_2 when temperature and pressure increase enough. First, the numerical prediction of the minimum fluidization velocity under such conditions is compared to classical correlations found in literature. Most of the experimental works using CO_2 in supercritical conditions as fluidizing agent were aimed at obtaining the minimum fluidization velocity and found a good agreement with correlations from the literature. Therefore, the numerical analysis of the minimum fluidization velocity can serve as a practical validation of the simulation results. Then, fluidization characteristics, such as bed expansion,

bubble characteristics and, as a novelty, velocities of gas and solids are analysed. This allows a comparison between the classical gas–solid fluidization under ambient conditions and the fluidization under high pressure and temperature, including the case with a fluidizing agent in supercritical conditions.

2. Simulated systems

The open–source MFIx–TFM code, developed at the Energy Technology Laboratory of the US Department of Energy, was used to conduct the numerical simulations of a two–dimensional (2D) bubbling fluidized bed. In MFIx–TFM, an Eulerian–Eulerian approach is employed, considering the gas and the solid phases as Eulerian continuous phases. The continuum description of the gas and dense phases, i.e., Two–Fluid Model, is based on the equations of mass and momentum conservation and the balance of granular temperature [28, 29]. The governing equations for the Two–Fluid Model are summarized in the following lines. The subscript s is referred to the solid phase and g to the gas phase.

Mass conservation of the gas and solid, continuum phases:

$$\frac{\partial}{\partial t}(\alpha_g \rho_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g) = 0 \quad (1)$$

$$\frac{\partial}{\partial t}(\alpha_s \rho_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s) = 0 \quad (2)$$

Momentum conservation of the gas and solid phases:

$$\frac{\partial}{\partial t}(\alpha_g \rho_g \vec{v}_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g \vec{v}_g) = -\alpha_g \nabla p_g + \nabla \cdot \overline{\overline{\tau}}_g + \alpha_g \rho_g \vec{g} + K_{gs}(\vec{v}_s - \vec{v}_g) \quad (3)$$

$$\frac{\partial}{\partial t}(\alpha_s \rho_s \vec{v}_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s \vec{v}_s) = -\alpha_s \nabla p_g - \nabla p_s + \nabla \cdot \overline{\overline{\tau}}_s + \alpha_s \rho_s \vec{g} + K_{gs}(\vec{v}_g - \vec{v}_s) \quad (4)$$

where p_i is the pressure and $\overline{\overline{\tau}}_i$ is the stress tensor for phase i .

The balance for the granular temperature, Θ , is:

$$\frac{3}{2} \left(\frac{\partial}{\partial t}(\rho_s \alpha_s \Theta) + \nabla \cdot (\rho_s \alpha_s \vec{v}_s \Theta) \right) = (-p_s \overline{\overline{I}} + \overline{\overline{\tau}}_s) : \nabla \vec{v}_s + \nabla \cdot (k_\Theta \nabla \Theta) - \gamma_\Theta - 3K_{gs}\Theta \quad (5)$$

where $(-p_s \overline{\overline{I}} + \overline{\overline{\tau}}_s) : \nabla \vec{v}_s$ is the generation of Θ by the solids stresses, $k_\Theta \nabla \Theta$ is the diffusion of Θ , γ_Θ is the collisional dissipation of Θ and $3K_{gs}\Theta$ is the transfer of random kinetic energy between the solids and the gas. In Equations 3, 4 and 5, K_{gs} is the drag force between the gas and the solid phase. The drag force correlation for the gas–solid interaction proposed by Beetstra et al. [30] was used in this work.

Table 1: Simulation properties.

Bed height, H [m]	0.7
Bed width, W [m]	0.3
Static bed height, h_0 [m]	0.3
Bed material density, ρ_s [kg/m ³]	3300
Bed material diameter, d_s [μ m]	280
Coefficient of restitution, e_s [–]	0.9

Table 2: Cases simulated and fluid properties.

	Conditions	Density ρ (kg/m ³)	Viscosity, μ_g (Pa s)
Air	$T = 293$ K, $p = 1$ bar	1.16	18.47e-6
	$T = 973$ K, $p = 1$ bar	0.35	42.65e-6
	$T = 633$ K, $p = 100$ bar	53.18	32.63e-6
	$T = 973$ K, $p = 200$ bar	66.82	43.58e-6
	$T = 293$ K, $p = 200$ bar	232.18	23.88e-6
CO ₂	$T = 293$ K, $p = 1$ bar	1.78	14.92e-6
	$T = 973$ K, $p = 1$ bar	0.54	40.47e-6
	$T = 633$ K, $p = 100$ bar	85.31	30.04e-6
	$T = 973$ K, $p = 200$ bar	104.22	41.62e-6
	$T = 293$ K, $p = 200$ bar	937.84	104.82e-6

A 2D bed of 0.3 m width, 0.7 m height and 0.3 m of static bed height was used in all the cases simulated. Air and CO₂ were employed as fluidizing agents, covering a wide range of operating conditions varying from ambient conditions to conditions of high temperature ($T = 973$ K) and pressure ($p = 200$ bar). Five cases have been simulated for each fluidizing agent. Tables 1 and 2 show the bed configuration and the operating conditions employed as well as the density and viscosity of air and CO₂ at the selected conditions. Note that the critical points are 7.38 MPa and 31.1 °C for CO₂ and 3.7 MPa and -141 °C for air. The interest of using CO₂ in supercritical conditions is due to the relatively wide range of densities of CO₂ when it is at ambient temperature and high pressure, which is not observed for air. Gas properties were calculated based on the database of Burcat et al. [31], which depend on temperature and pressure. Each of the simulations were carried out at constant temperature to capture only the fluid–dynamic effects, so the bed is assumed well isolated and, therefore, the walls are considered adiabatic. The solids employed as bed material belong to the B classification of Geldart [32].

The 2D computational domain was meshed with square cells of 2.5 mm size and a second order accurate scheme was selected to discretise the convective derivatives of the governing equations. This mesh size has been proved to reproduce satisfactory the fluidization behaviour under conditions of high pressure and temperature [33]. The gas flow is fed uniformly through the bottom of the bed using a velocity inlet

boundary condition. The gas flow leaves the system through the top boundary at a constant pressure. A no-slip boundary condition is used for the gas and continuum solid phases at the side walls of the bed. Previous works have demonstrated that the effect of the lateral boundary condition on this kind of simulations is relatively small [34, 35]. To increase the accuracy of the simulations, the balance for the granular temperature is solved using a partial differential equation formulation instead of a simplified algebraic equation. The physical time simulated was 20 s in all cases, however, the first 5 s were not used in the analysis to eliminate the start-up effect.

3. Results & Discussion

3.1. Validation through the estimation of the minimum fluidization velocity

This section focuses on the estimation of the minimum fluidization velocity, U_{mf} . The fluidizing agents studied are air and CO₂. The estimations obtained by the simulations are validated by comparison with well-known, empirical or semi-empirical correlations from the literature. Most of the experimental works using fluids in supercritical conditions as fluidizing agent were aimed to report the minimum fluidization velocity and found a good agreement with the correlations from the literature [17, 11, 36]. The effects of pressure and temperature are considered in these correlations through the variation of density and viscosity of the fluidizing agent. The correlations used for the present validation are those proposed by Ergun [37], Wen & Yu (W-Y) [38], Carman-Kozeny (C-K) [39] and Grace [40]. The method of the mean pressure was employed to estimate U_{mf} from the results of the MFIX-TFM simulations performed using 5 different superficial gas velocities in the fixed bed regime and 5 gas velocities in the fluidized bed regime.

Figure 1: Minimum fluidization velocity estimation, U_{mf} , of different correlations and predicted by the TFM simulations, a) Air and b) CO₂. □ represents ambient conditions, + represents $T = 973$ K and $p = 1$ bar, and ○ represents $T = 973$ K and $p = 200$ bar.

Figure 1 shows the minimum fluidization velocity, U_{mf} , predicted by different correlations and obtained from the simulations for different conditions: ambient conditions, $T = 293$ K and $p = 1$ bar, high temperature conditions, $T = 973$ K and $p = 1$ bar, and conditions of high temperature and pressure, $T = 973$ K and $p = 200$ bar. Relatively small deviations among the different correlations can be observed and the determined U_{mf} from the TFM simulations is within these deviations. Differences between ambient conditions and high pressure and temperature conditions can also be observed. According to the results, the variation of density and viscosity when increasing both temperature and pressure promotes a reduction of the apparent minimum fluidization velocity. Analysing the correlations employed to estimate U_{mf} , the main affecting parameter is the viscosity of the gas, which is basically a function of temperature, T , so the greatest variation occurs when passing from ambient conditions to $T = 973$ K, as can be seen in the numerical results. Then, a lower variation of U_{mf} is found when increasing pressure at high temperature since the viscosity is less affected by the increase of pressure. Both for air and CO_2 , the differences between the simulation estimation and correlations of U_{mf} are greater at ambient conditions whereas these differences are reduced at high temperature and pressure. Subramani et al. [41] and Sánchez-Prieto et al. [42] experimentally measured U_{mf} for different temperatures and also found a good agreement with Ergun, Wen & Yu and Carman–Kozeny correlations, similar to what is observed in Figure 1. In particular, Subramani et al. [41] tested quartz magnetite particles with a size and density close to the particles employed in this work. Therefore, the simulation seems to reproduce reasonably well the minimum fluidization velocity. Thus, the value U_{mf} obtained from the simulations will be used as a reference parameter for the cases analysed.

3.2. General bed behaviour

The changes in the fluidization general behaviour when increasing T and p from those of typical fluidization under ambient conditions are analysed in this section. To do that, air and CO_2 are used as fluidizing agents at ambient conditions, increasing temperature to $T = 973$ K and increasing both temperature and pressure to $T = 973$ K and $p = 200$ bar.

The comparison is based on the bed fluid–dynamics, which is basically affected by the bubbles created due to the excess gas velocity above the minimum fluidization velocity. Therefore, for a consistent comparison of the results, the excess gas velocity, defined as $U_0 - U_{mf}$, is kept constant in all the cases. The value of the excess gas velocity is selected from $2.5U_{mf}$ in the case with the lowest minimum fluidization velocity, which corresponds to CO_2 at $T = 973$ K and $p = 200$ bar and results in a value of $U_0 - U_{mf} = 4$ cm/s. Thus, the superficial gas velocity, U_0 , for each case is extracted from this excess gas velocity and the U_{mf} obtained previously. The highest gas velocity is found then for the case of CO_2 at ambient conditions and,

Table 3: Gas velocities for the simulated cases.

$U_0 - U_{mf} = 4 \text{ cm/s}$	Air		CO ₂	
	$U_{mf} \text{ (cm/s)}$	$U_0 \text{ (cm/s)}$	$U_{mf} \text{ (cm/s)}$	$U_0 \text{ (cm/s)}$
$T = 293 \text{ K}, p = 1 \text{ bar}$	7.9	11.9	8.5	12.5
$T = 973 \text{ K}, p = 1 \text{ bar}$	3.6	7.6	3.5	7.5
$T = 633 \text{ K}, p = 100 \text{ bar}$	4.0	8.0	3.5	7.5
$T = 973 \text{ K}, p = 200 \text{ bar}$	2.9	6.9	2.7	6.7
$T = 293 \text{ K}, p = 200 \text{ bar}$	3.0	7.0	0.8	4.8

the resulting gas velocity corresponds to $1.5U_{mf}$. Table 3 summarises the gas velocities for the simulation cases.

Figures 2 and 3 show some example snapshots of the solids concentration, α_s , for all the cases simulated. Bubbles are clearly distinguishable for both fluidizing agents at ambient and high temperature conditions. However, when pressure increases, either using air or CO₂, only some irregular voids, far from the shape of a typical bubble, are found in the bed. In bubbling gas–solid fluidized beds at ambient conditions, two main phases are distinguished, bubbles and dense phase, which is the so–called Two–phase theory. However, in the cases of intermediate and high pressure not only voids and dense phase are appreciated, but also regions of dilute solids can be observed. These regions present a concentration slightly lower than the maximum concentration of solids. This pattern is mainly caused by the increase of density of the gas due to the pressure increase. Density of air and CO₂ increases when pressure augments, which implies that the weight of the gas in the bed is non–negligible and the interaction with the solids is completely different to that with lower values of gas density. In fluidization under ambient condition, bubbles mainly ascend due to buoyancy, the vertical component of the bubble velocity being higher than the horizontal component. At high temperature and pressure conditions, the irregular voids found instead of bubbles show a different motion, with a much more important horizontal component than in the cases at ambient conditions. In particular, the case of CO₂ at $T = 293 \text{ K}$ and $p = 200 \text{ bar}$ shows a remarkable different behaviour from the other cases. In this case, the density of the CO₂ is comparable to that of a common liquid showing a fluidization behaviour pretty similar to liquid–solid systems.

To further analyse the patterns of solids in the bed under the different configurations considered, the concentration histogram can be studied. This histogram is calculated using every cell up to a height of 0.5 m and all the simulated time steps analysed. This is done for the cases at ambient conditions, $T = 293 \text{ K}$ and $p = 1 \text{ bar}$, high temperature conditions, $T = 973 \text{ K}$ and $p = 1 \text{ bar}$, and conditions of high temperature and pressure, $T = 973 \text{ K}$ and $p = 200 \text{ bar}$. Not all the cases are included in the following subsections to avoid repetitiveness. The most important results are gathered and compared for all the cases

Figure 2: Instantaneous snapshots of α_s for air: (from left to right) ambient conditions, $T = 973$ K and $p = 1$ bar, $T = 633$ K and $p = 100$ bar, $T = 973$ K and $p = 200$ bar and $T = 293$ K and $p = 200$ bar. The colorbar represents α_s .

Figure 3: Instantaneous snapshots of α_s for CO_2 : (from left to right) ambient conditions, $T = 973$ K and $p = 1$ bar, $T = 633$ K and $p = 100$ bar, $T = 973$ K and $p = 200$ bar and $T = 293$ K and $p = 200$ bar. The colorbar represents α_s .

in the last subsection to evaluate the effects of pressure and temperature on the fluidization behaviour. Each histogram is normalised, so the sum of the relative frequency in all the intervals is equal to 1. These results are presented in Figures 4, 5 and 6 for both fluidizing agents, air and CO_2 . At ambient conditions only two phases are distinguished, gas ($\alpha_s < 0.1$) and dense phase ($\alpha_s > 0.5$), whereas the region in the middle, which corresponds to solids concentration, α_s , from 0.1 to 0.5, is negligible. The minimum value in the histogram appears at $\alpha_s = 0.3$, which will be the threshold value further selected to distinguish bubbles from dense phase. When temperature increases, the dilute phase of solids slightly starts to appear. Still, the presence

of bubbles and dense phase separately is much more important than this dilute phase.

Figure 4: Histogram of the solids concentration, α_s , for a) air and b) CO_2 at ambient conditions.

Figure 5: Histogram of the solids concentration, α_s , for a) air and b) CO_2 at $T = 973$ K and $p = 1$ bar.

The greatest differences appear in the cases of high T and p , where the presence of the dilute phase cannot be neglected any more. The presence of bubbles and dense phase is considerably reduced from the previous cases in favour of the dilute phase, which shows a growing tendency towards higher values of α_s . For these high T and p conditions, there are almost no differences between the fluidizing agents employed in terms of solids concentration.

Figure 7 shows the solids hold-up as a function of the vertical coordinate, y . The solids hold-up has been calculated using the time averaged solids volume fraction, α_s , also spatially averaged over the width

Figure 6: Histogram of the solids concentration, α_s , for a) air and b) CO_2 at $T = 973 \text{ K}$ and $p = 200 \text{ bar}$.

of the bed. Higher bed expansion, detected as high values of α_s for high y , occurs when the bed is fluidized under high T and p . Considering that the excess gas velocity, $U_0 - U_{mf}$, is the same for all the cases, the main differences between simulated cases are due to the distribution of this excess of gas in the bed, which indicates a less packed dense bed, slightly decreasing also the mean porosity of the solids. This means that in the cases of ambient conditions and high T , all the excess gas over minimum fluidization contributes mainly to bubble formation and throughflow, but in the case of high T and p the distribution of the excess gas is different, contributing also to form the dilute phase found in the histogram of α_s , Figure 6.

Figure 7: Solids hold-up, α_s , as a function of bed height, y a) air and b) CO_2 .

3.3. Bubble behaviour

Bubbling fluidized beds are typically analysed by means of bubbles. In numerical simulations, a threshold for the solids volume fraction contours is required to distinguish bubbles or voids from the dense phase. A value of 0.3 for this threshold, coinciding with the minimum value in the histogram for air at ambient conditions (Figure 4), is commonly accepted as a reasonable value to capture the contour of such bubbles [43].

The variation of bubble diameter with the distance to the distributor was obtained for all cases. The results are depicted in Figures 8, 9 and 10. The bubble diameter is calculated as the equivalent diameter of a circle that has the same area of the bubble. The data are represented in the form of boxplots for intervals of height of 4.7 cm to show the statistical distribution of bubble diameters instead of only average values. On each boxplot, the central mark indicates the median, and the bottom and top edges of the box indicate the lower and upper quartiles corresponding to percentiles of 25% and 75%, respectively. The whiskers extend to the most extreme data points located at a distance of 1.5 times the inter-quartile distance from the quartiles, and the outliers are plotted individually using the '+' symbol. The typical bubble growth with the vertical coordinate is observed for the case of air and CO₂ at ambient conditions (Figure 8) which is mainly attributed to bubble coalescence in gas–solid fluidization. The boxplot at the highest y shows a slight decrease of D_b since this height interval captures the bubble eruption at the surface of the bed.

Figure 8: Bubble diameter D_b versus bed height y for a) air and b) CO₂ at ambient conditions.

When temperature alone is increased (Figure 9), the bubble sizes obtained are similar to that of ambient conditions although the size distribution differs. The greatest bubbles are less prone to be found at every height interval as the shape of the boxplot for the high temperature case indicates. Significant differences

appear in the height range 0.15–0.3 m (upper part of the bed), where the number of large bubbles is reduced for high temperature (Figure 9) compared to the case at ambient conditions (Figure 8). Besides, the decrease of D_b at the surface of the bed is less pronounced than in the case at ambient conditions, which can be attributed to the slightly higher bed expansion in the case analysed in Figure 9.

Figure 9: Bubble diameter D_b versus bed height y for a) air and b) CO_2 at $T = 973 \text{ K}$ and $p = 1 \text{ bar}$.

The cases at conditions of high T and p show a more flat profile of the bubble diameter, indicating that there is a uniform distribution of the bubble sizes along the bed height. The bubble size distribution is remarkably different with regards to the previous cases, obtaining significantly smaller bubbles when fluidizing the bed at high T and p . This different bubble size distribution can be attributed to two main causes. The first cause is that indeed the bubbles in the case at high T and p are smaller because the pressurised fluid is enhancing the bed expansion (see Figure 7) instead of the formation of bubbles. The second cause is that no regular bubbles are generated, but blurry voids merely as an attempt of bubbles (see Figures 2 and 3). Furthermore, the bubble diameter distribution in this case means that the presence of large bubbles in the bed operated at high T and p is probable but less likely than at ambient conditions.

Figure 11 shows the time evolution of bubble area, δ_b , in the whole bed. δ_b represents the total frontal area occupied by bubbles at each time instant in the 2D bed. A higher visible area in the case under high T and p is found whereas very similar values are found for the cases at ambient conditions and at high T . These results, combined to what was extracted from the analysis of the bubble size distribution (Figure 10), might indicate that a higher number of smaller bubbles is needed to reach such value of δ_b when the bed is fluidized at high T and p . It is also noticeable that in all the cases, CO_2 presents higher values of δ_b than air. This can be attributed to the different properties of the CO_2 compared to air, mainly because of the

Figure 10: Bubble diameter D_b versus bed height y for a) air and b) CO_2 at $T = 973 \text{ K}$ and $p = 200 \text{ bar}$.

slightly higher density for CO_2 in the ranges studied. Typical correlations used to estimate bubble growth do not consider variations of fluid properties, however, in view of the present results, fluid properties seem to affect bubble growth.

Figure 11: Time evolution of δ_b , a) air and b) CO_2 .

3.4. Gas and solids velocities

To further study bed dynamics, gas and solids velocities are analysed. Figures 12 and 13 show the velocity components of the gas and the solid phases for the different conditions studied. The horizontal and vertical components of the velocity, u and v respectively, have been extracted from the simulation for each phase, gas (g) and solids (s). The velocity is also represented in the form of boxplots to analyse its

magnitude and distribution. No outliers are included in the representation to enhance visualisation. Each boxplot contains the information of every time instant and every cell up to a height of 0.5 m. For the case at ambient conditions, the solids vertical velocity is substantially higher than that of the horizontal component, both of which have a median value of 0. In this kind of fluidization, bubbles tend to ascend due to buoyancy effects which promote the vertical motion. Concerning the gas velocity, higher values are also obtained for the vertical component, in consonance with the solids velocities. The median value of the gas vertical component is displaced towards positive values, as expected, due to the vertical velocity inlet through the distributor of the bed.

Figure 12: Boxplot of the gas and solids velocities for air at ambient conditions: $T = 973$ K and $p = 1$ bar; and at $T = 973$ K and $p = 200$ bar.

When the gas temperature is increased, the horizontal component, both for the gas and solid phases, becomes more important than at ambient conditions but it is still comparatively lower than the vertical component. In this case, U_{mf} is lower than at ambient conditions and, according to the cases simulated (Table 3), a smaller superficial gas velocity is employed. Therefore, this effect is appreciated in the values obtained for the vertical component of the gas, which are smaller than in the case at ambient conditions.

When both temperature and pressure are increased, the horizontal component becomes as important as the vertical component for the gas and solid phase velocities. The distribution of velocities is almost the same for the two components with the unique exception of the displacement towards positive values of the vertical component of the gas due to the net ascend of the gas flow from the inlet boundary conditions.

Besides, the differences between air and CO_2 for the gas and solid velocities can be considered negligible (Figures 12 and 13). When temperature alone is increased in the case of CO_2 , the resulting values of the solids and gas velocities are slightly higher than those of the case of air. However, when both pressure and

temperature are increased, which turns the CO_2 into supercritical conditions, almost the same results as the air are obtained. This means, that the selection of the gas, either air or CO_2 , in the cases analysed in this work, does not affect the fluidization behaviour of the bed. In contrast, as commented before, the main reason affecting the bubbling fluidization behaviour seems to be the density variation of the gas under high temperature and pressure. Gas density is halved from the case at ambient conditions to the case of high T but is increased around 2 orders of magnitude in the case at high T and p . The results show that the greatest variations in the fluidization occur when increasing both T and p . Nevertheless, the transition from a common gas to a supercritical fluid does not seem to have a strong effect on it.

Figure 13: Boxplot of the gas and solids velocities for CO_2 at ambient conditions: $T = 973$ K and $p = 1$ bar; and at $T = 973$ K and $p = 200$ bar.

3.5. Analysis of the effects of pressure and temperature

This subsection comprises the comparison of all the cases simulated (Table 2). Figure 14 shows the relative frequencies of $\alpha_s = 0$ and $\alpha_s = 0.6$ extracted from the histograms of the solids concentration (similar to those of Figures 4–6) and the bed expansion for the cases simulated (similar to Figure 7). In Figure 14 and in the subsequent figures, the horizontal axis represents the case studied: *Amb* is used for the case at ambient conditions, T_h for 1 bar and 973 K, $(T, p)_{int}$ for 100 bar and 633 K, $(T, p)_h$ for 200 bar and 973 K and p_h for 200 bar and 293 K. Regarding the relative frequency, its value decreases as T and p increases. In particular, the decrease rate for $\alpha_s = 0.6$ is higher than for $\alpha_s = 0$. This indicates the appearance of the new dilute phase in contrast to the existence of bubbles and dense phase that can be appreciated in the classical gas–solid fluidization. The intermediate pressure and temperature case shows also an intermediate behaviour among the cases analysed. In Figure 14a, the high pressure at ambient

temperature case has relative frequencies of α_s much smaller than those of the ambient pressure at high temperature, which indicates that the effect of pressure is stronger than the effect of temperature. In particular, for the case of CO_2 at $p = 200$ bar and $T = 293$ K, the relative frequency of $\alpha_s = 0$ and $\alpha_s = 0.6$ is reduced from 0.35 to nearly 0.2 ($\alpha_s = 0$) and 0 ($\alpha_s = 0.6$), indicating that the dilute region comprising intermediate values of solids concentration has become strongly important in this case. As a consequence, bubbles need to be discriminated with a lower value of α_s , which has been selected as 0.15 for this case.

In Figure 14b, the bed expansion has been extracted as the value of y in Figure 7 where $\alpha_s = \alpha_{s,max}/2$. An increase of the bed expansion among the cases analysed can be observed from left to right of Figure 14b. As long as temperature and pressure increase, U_{mf} decreases (see Table 3), so the fluidization number (number of times U_{mf}) is higher provided that the excess gas velocity is kept constant for all the cases. Therefore, a higher bed expansion is obtained for higher values of both temperature and pressure. Specifically, the case of CO_2 at $T = 293$ K and $p = 200$ bar shows a remarkable higher bed expansion, as it was also illustrated in Figure 3, due to the density reached by the CO_2 at such conditions.

Figure 14: Relative frequency of $\alpha_s = 0$ and $\alpha_s = 0.6$ (a) and bed height (b) for the cases simulated.

Figure 15 shows the median value of the bubble diameter distributions of Figures 8-10 for all the cases simulated. The typical bubble growth of gas fluidization along the bed height can be seen for the case at ambient conditions. This growth is slightly reduced for the case at high temperature. However, when pressure increases no matter the temperature value, the bubble diameter distribution shows a roughly constant profile along the bed height. This indicates that when pressure increases sufficiently, smaller bubbles are created and are uniformly distributed along the bed height. Similar results are obtained for the three cases at elevated pressure, confirming the stronger effect of pressure on the fluidization behaviour compared to temperature.

Figure 15: Median of the bubble diameter distribution versus bed height for air (a) and CO₂ (b).

Figure 16 shows the interquartile distance of the velocity distributions previously illustrated in Figures 12 and 13 for the solids and the fluidizing agent. The interquartile distance has been selected as this parameter can describe faithfully the differences in the distribution of the velocities. It is important to note here the similitude of the interquartile distance of the horizontal and vertical components of the velocities as long as pressure and temperature increases. This indicates an increase of the horizontal component as a consequence of the higher fluid density when pressure increases. This density change promotes that buoyancy is not the main effect acting on the bubbles to ascend and modifies the interaction between bubbles and dense phase.

Figure 16: Interquartile distance of the velocity distribution for the cases simulated, (a) air and (b) CO₂.

3.6. Deviation from the Two-phase theory

The results described above are certainly important to be considered for the performance of fluidized bed reactors. A main research line in fluidized bed units during the last decades has been the characterisation of the mixing of solids and dispersion of fuel particles. Due to the vertical motion of bubbles, beds are typically characterised by a certainly good axial mixing and dispersion compared to the lateral or radial component. Therefore, special effort has been paid in studying and improving the lateral mixing and dispersion of fluidized bed units [44, 45, 46, 47, 48, 49, 50]. The fact that under high pressure and temperature conditions the horizontal component of the gas and solids velocities becomes comparable to the vertical component, indicates that lateral mixing and dispersion is increased compared to the fluidization at ambient conditions.

The results shown have proved the changes in the fluidization behaviour when pressure and temperature increase. The main difference is the appearance of a dilute phase characterised by solids concentration, α_s , in the range 0.1–0.5. This constitutes a great difference compared to the classical gas–solid fluidization which is identified by two phases that can be perfectly distinguished, bubbles and solids, i.e., the well-known Two-phase theory. The presence of this dilute phase at moderate excess gas velocities, where a bubbling regime is expected, might indicate a slight deviation from the classical Two-phase theory for beds operated at high T and p . To characterise this deviation the following procedure is followed.

The Two-phase theory assumes that the inlet velocity is employed in the fluidization of the solid particles, which becomes the dense phase, in the formation of bubbles and in the gas bypassing bubbles, typically known as throughflow [51, 52]:

$$U_0 = U_{dp} + U_{vis} + U_{th} \quad (6)$$

where U_0 is the superficial inlet velocity, U_{dp} is the superficial gas velocity through the interstices of the dense phase, U_{vis} is the superficial velocity concerning the visible bubble flow and U_{th} is the contribution of throughflow to the superficial velocity. The above balance can be written as a function of U_{mf} .

$$U_0 = U_{mf} + \psi(U_0 - U_{mf}) + (1 - \psi)(U_0 - U_{mf}) \quad (7)$$

where ψ is the visible bubble flow as a fraction of the excess gas velocity. In the above balance (Equation 7), according to the Two-phase theory, the regions outside bubbles, i.e., the commonly named dense phase,

is assumed to be at minimum fluidization conditions ($U_{dp} = U_{mf}$). Therefore, the vertical component of the gas velocity can be extracted from the simulations in the cells of the regions outside bubbles, for every time instant, and the time averaged can be performed. This velocity can be compared to the assumption of the Two-phase theory, in which the superficial gas velocity is at minimum fluidization velocity, U_{mf} . Then a relative variation from the Two-phase theory can be defined as follows:

$$\Delta U = \frac{U_{TMF} - U_{mf}}{U_{mf}} \quad (8)$$

In Equation 8, U_{TMF} is the averaged gas velocity extracted from the simulations in every time instant and cell of the region outside bubbles. The results of ΔU are plotted in Figure 17 for all the cases analysed. This figure clearly shows that the simulations at ambient conditions satisfy the Two-phase theory whereas deviations start to appear when the pressure and temperature are increased. These deviations are almost negligible for the case when temperature is increased alone but becomes certainly important when pressure is also increased. In particular, Figure 17b shows this deviation versus the density of the fluidizing agent. It is important to note that the density of CO_2 at high pressure and ambient temperature is 937.84 kg/m^3 , which is comparable to that of a conventional liquid, e.g. water, whereas the air at such conditions has a much smaller density. This case shows the greatest deviation from the Two-phase theory among all the cases studied. Figure 17b also seems to indicate a roughly linear deviation from the Two-phase theory with the density of the fluidizing agent.

This result is in agreement with the findings of previous sections that show a clear deviation of the bed dynamics from the classical gas–solid fluidization. The irregular shape of bubbles, the presence of the dilute phase with intermediate values of solids concentration and the bubble size distribution in the bed are in concordance with the fact that the regions outside bubbles are no longer at minimum fluidization conditions when the gas is at high pressure and temperature. In these conditions, a fraction of the inlet gas passes through the dense phase at a velocity higher than the expected of U_{mf} , which generates a dilute third phase whose presence cannot be neglected. According to the cases simulated, the deviation from the Two-phase theory seems a direct consequence of the density change of the fluidizing agent (Table 2). The variation of the fluid viscosity with temperature and pressure may have an indirect effect on the fluidization behaviour as the viscosity affects the minimum fluidization velocity. However, the increase of the fluid density makes buoyancy not to be the main effect acting on the bubbles and a more complex interaction between bubbles

and dense phase occurs.

Figure 17: Deviation from the Two-phase theory, $\square$ represents air and $+$ CO_2 .

4. Conclusions

The present work has performed numerical simulations of fluidized beds under different conditions of pressure, from 1 bar to 200 bar, and temperature, from 293 K to 973 K, using air and CO_2 as fluidizing agents. In particular, the conditions of high temperature and pressure tested make the CO_2 to turn into supercritical conditions. Firstly, the numerical results showed that minimum fluidization velocity is mainly affected by the viscosity of the fluid, which is a strong function of the temperature. These results are in agreement with classical correlations to calculate the minimum fluidization velocity, which serves as a practical validation of the numerical results. Furthermore, when both pressure and temperature are increased, fluidization shifts from the typical bubbling fluidization to a more homogeneous fluidization. In this state, not only bubbles and dense phase are present in the bed but also a dilute phase of moderate solids concentration is encountered. Bubbles found at high pressure combined or not with high temperature are smaller, with a relatively similar size all over the bed height, and more uniformly distributed compared to the case at ambient conditions and the case at high temperature and ambient pressure. The main consequence is that the horizontal component of the solid and gas phases is greater at high pressure than at ambient conditions for which bubbles mainly ascend vertically. Also, according to the simulation results, the bed at high T and p , and specially and high p and ambient T , presents an increase of the interstitial gas velocity in the region outside bubbles that deviates from the expected U_{mf} predicted by the classical Two-phase theory. The results indicate that the pressure of the fluidizing agent has a stronger effect on the bed dynamics than the

fluidizing agent temperature. This seems to be caused by the fact that the density of the fluidizing agents varies more for the pressure range than for the temperature range analysed in this work. Finally, at high T and p almost no differences are encountered between the solids fluidization behaviour by air and that produced by CO_2 , even though the CO_2 is in supercritical conditions. However, the main difference between both fluidizing agents was at $p = 200$ bar and $T = 293$ K, for which the CO_2 density becomes similar to that of a common liquid.

Nomenclature

D_b	Bubble diameter [m]
d_s	Dense phase particle diameter [m]
e_s	Coefficient of restitution [–]
$\vec{g}$	Gravity [m^2/s]
H	Bed height [m]
h_0	Static bed height [m]
K_{gs}	Drag force between gas and solids [$\text{kg}/\text{m}^3\text{s}$]
k_Θ	Diffusion coefficient for granular energy [kg/ms]
p	Pressure [Pa]
p_g	Gas pressure [Pa]
p_s	Solids pressure [Pa]
T	Temperature [K]
U/U_{mf}	Dimensionless gas superficial velocity [–]
U_0	Superficial gas velocity [m/s]
U_{TFM}	Gas velocity obtained from the TFM simulations [m/s]
U_b	Bubble velocity [m/s]

U_{dp}	Gas velocity in the dense phase [m/s]
U_{mf}	Minimum fluidization velocity [m/s]
U_{th}	Throughflow velocity [m/s]
U_{vis}	Visible bubble velocity [m/s]
u_g	Horizontal gas velocity [m/s]
u_s	Horizontal solids velocity [m/s]
$\vec{v}_g$	Gas velocity [m/s]
$\vec{v}_s$	Solids velocity [m/s]
v_g	Vertical gas velocity [m/s]
v_s	Vertical solids velocity [m/s]
W	Bed width [m]
x	Horizontal coordinate [m]
y	Vertical coordinate [m]

Greek letters

α_g	Gas volume fraction [–]
α_s	Solids volume fraction [–]
ΔU	Relative deviation from the Two–phase theory [–]
δ_b	Bubble frontal area in the bed [m ²]
γ_Θ	Collisional dissipation of Θ [m ² /s ²]
μ_g	Gas viscosity [Pa s]
μ_s	Solids viscosity [Pa s]
$\overline{\overline{\tau}}_g$	Gas stress tensor [Pa]
$\overline{\overline{\tau}}_s$	Solids stress tensor [Pa]

ψ	Visible bubble flow fraction [–]
ρ_g	Gas density [kg/m ³]
ρ_s	Dense phase particle density [kg/m ³]
Θ	Granular temperature [m ² /s ²]

Acknowledgments

This work has been financially supported by the Spanish government under the project ENE2015-69486-R (MINECO/FEDER, UE).

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