FLUIDIZATION IN SUPERCRITICAL CARBON DIOXIDE

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ABSTRACT

An experimental study was carried out to determine the behaviour of group A glass bead beds under supercritical conditions. During experiments the fluidization state was followed by pressure drop measurements. The particle fluidization was obtained with carbon dioxide conditions far from the critical point whereas near this point a gas channelling seems to occur. The increase in interparticle forces in these conditions leads to the particle aggregation.

INTRODUCTION

Gas-solid fluidized beds are widely used in process industries to carry out reactions, combustion, drying, granulation and coating. The particle behaviour in a gas-solid fluidized bed at ambient conditions mainly depends on the particle diameter and density. Thus, in 1973, Geldart classified particles into four groups, A, B, C and D, according to their behaviour [1]. The group A powders are increasingly used in the industry. Geldart A particles are typically within the size range 20-100 µm and are defined as "aeratable" particles. Indeed, during the fluidization, group A particle beds considerably expand and their expansion is homogeneous between the minimum fluidization velocity u_{mf} and the velocity u_{mb} at which the first bubbles appear.

With particle sizes lower than 100 µm, cohesive forces are noteworthy in fluidized beds at ambient conditions. The main adhesion forces, which can be disadvantageous for fluidised bed, are the capillary force, the electrostatic force and the Van der Waals force. When these forces become dominant, they bring about particle clustering and gas channelling.

However, studies of the fluidized bed behaviour at high pressures reported that the pressure or gas density improve the bed expansion [2] . Therefore, fluidization with supercritical carbon dioxide (SC CO_2) can be shifted continuously from aggregative to particulate [3]. Likewise Vogt and al. have shown that glass beads can be fluidized with supercritical carbon dioxide [4]. Indeed, over the critical point (P_c=7.38 MPa and T_c=31°C) CO_2 is located in a domain where its properties, like density and viscosity, are intermediate between those of liquid and gas. Moreover, in the supercritical domain, CO_2 density and viscosity can be tuned over a wide range by screening pressure or temperature.

In this study, the influence of SC CO_2 conditions (P, T) on the fluidization quality has been assessed.

MATERIALS

Fluidization experiments were carried out in a laboratory set up. A schematic view of this device is shown on Figure 1. The main component is a stainless steel high pressure autoclave which is made for maximum operating conditions of 28 MPa and 100 °C. In this autoclave, the fluidized bed column is inserted. With an inner diameter of 30 mm and an height of 370 mm, up to 200 grams of solid particles can be processed. A 8 µm porous plate is used as fluid distributor.

The carbon dioxide is supplied from a CO_2 tank at approximately 6 MPa. Before being compressed, the carbon dioxide is cooled down to 5°C. A maximum carbon dioxide flow of 3 $kg.h^{-1}$ can be achieved using a membrane pump. The CO_2 flow is monitored by a Coriolis mass flowmeter. Then, the carbon dioxide is heated at the process temperature thanks to an electric heat exchanger. The pressure inside the fluidized bed is adjusted using a back pressure regulator. The fluidization is controlled thanks to the pressure drop measurement by a 0-373.10^2 Pa pressure differential transducer (Rosemount, model 1151 HP) which is heated at the process temperature. With this device the pressure drop created by the fluidized bed column and the particles is measured. Hence, the distributor pressure drop is figured out with a specific run without any particles. A pressure transmitter, set above the fluidized bed, and two thermocouples, one below and another above the reactor, monitor the CO_2 overall temperature and pressure in order to determine its density and viscosity. Entrained particles are retained thanks to a filter placed at the fluidised bed exit. Thus the carbon dioxide is recycled.

In this study, glass beads (Potters, Spheriglass) were chosen as model particles. The bead density ranges from 2460 to 2490 $kg.m^{-3}$ (supplier data). Their mean diameter distribution was checked with a particle size analyser (Malvern Instruments).

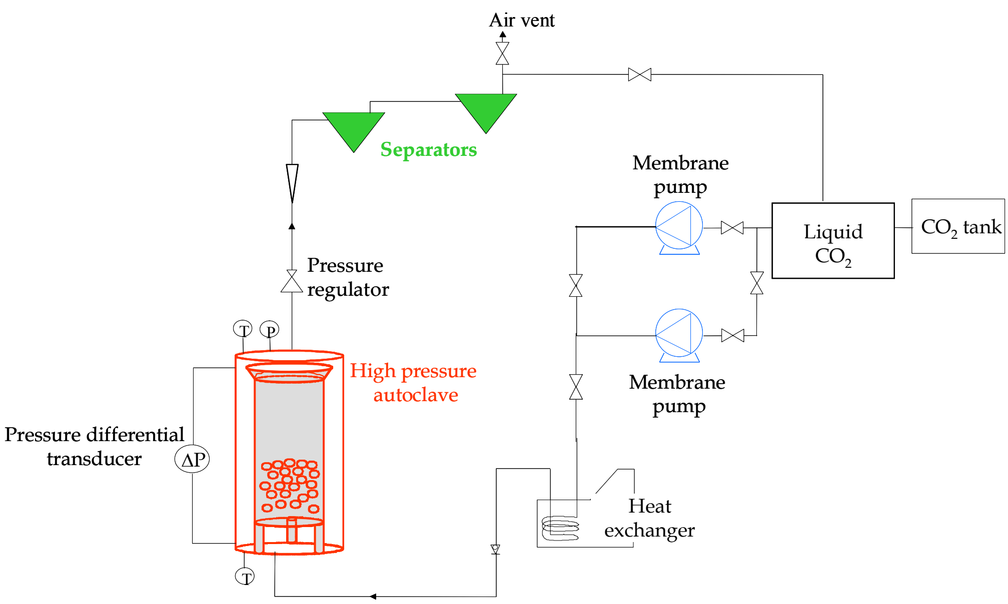


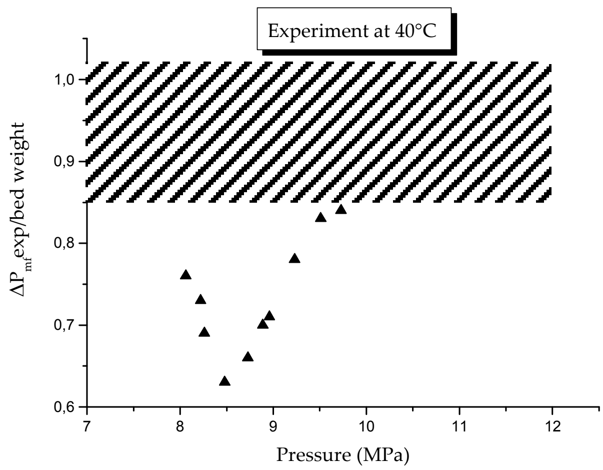
Figure 1 : Schematic view of the set up.

RESULTS

71 µm glass beads fluidization was tested in CO_2 under a pressure of 7.5 up to 15 MPa and a temperature of 33 to 50°C. Two different behaviours, fluidization and no fluidization, were observed during these experiments. Indeed, in some conditions (for example 8.1 MPa, 41°C) it was observed that the measured pressure drop stabilizes at a value lower than the bed weight [defined by $m_p\, g\, (\rho_S - \rho)/(S\, \rho_S)$] to 30 – 40 per cent. The same observation is made when a gas channelling phenomenon occur.

To understand this phenomenon, experiments were carried out to measure the experimental pressure drop $\Delta P_{mf\ exp}$ at various pressures at $u > u_{mf}$. This pressure drop was compared to the apparent bed weight. The figures 2 and 3 represent the ratio between the experimental pressure drop and the apparent bed weight ($\Delta P_{mf\ exp}$/bed weight) as a function of the pressure. If particles are fluidized this ratio must be equal to 1. Given that the pressure differential induces an error of 200-300 Pa, it was considered that fluidization occurred with ratio values between 0.85 and 1.15 (hatched zone). However at 40°C the ratio falls between 8 and 10 MPa whereas at 50°C the drop occurs at about 8.5 to 12 MPa. The phenomenon is repeatable (assay 1 and assay 2) as shown on the figure 6. Furthermore it is observed also on pressurization or depressurization of the bed (assay 2, arrows).

So, at specific conditions, particles seem to be not fluidized. They should rather aggregate owing to interparticle forces and SC CO_2 flows preferentially along micro channels. However, this phenomenon occurs only for some pressures.



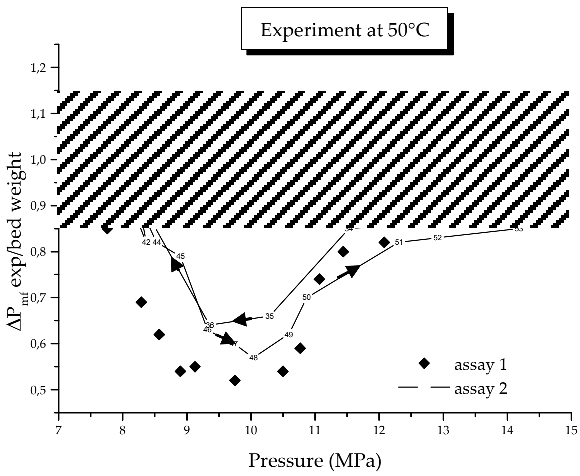


Figure 2 : Ratio $\Delta P_{mf\ exp}$/bed weight versus the pressure at 40°C
Figure 3 : Ratio $\Delta P_{mf\ exp}$/bed weight versus the pressure at 50°C

This phenomenon appears close to the critical point conditions where supercritical fluids are widely known for their molecular distribution inhomogeneity and their density fluctuation. In a phase diagram, the density fluctuation forms a ridge when the contour map of its value is drawn [5]. According to Nishikawa and Morita, the ridge is the boundary which separates the supercritical domain into two regions. The upper one is rather a liquid-like region whereas the other is a more gas-like region [6].

All the experiments are represented on this graph (figure 4). The crosses fit to the conditions where the fluidization is proved. The circles are the experimental results of a ratio lower than 0.85. These points are plotted in the liquid-like region between the ridge and the contour of the density fluctuation for the value of 2 [7]. On this plot a region where the fluidization quality may be more efficient can be defined.

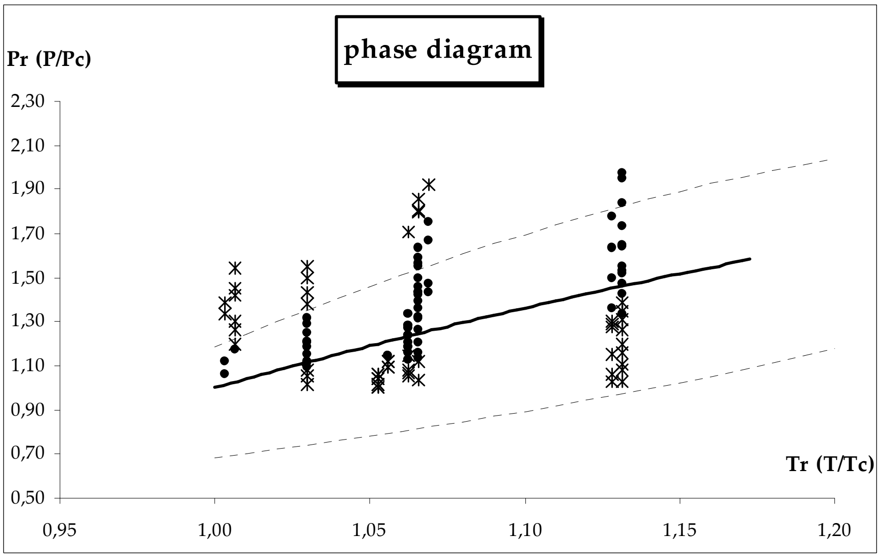


Figure 4 : Phase diagram of the supercritical carbon dioxide. Ridge (solid curve) and the contour of density fluctuation for the value of 2 (dotted curve). (o) are the experimental results with a ratio lower than 0.85, (*) fluidization.

CONCLUSION

Fluidization experiments in supercritical carbon dioxide with group A glass beads were tested. In an area located between the ridge and the contour of the density fluctuation for the value 2, particle fluidization can not be proven. The glass beads should aggregate and a gas channelling should occur. However, outside this area, particles are fluidized. Therefore, it was obvious that SC CO_2 properties have a great effect on the fluidization quality.

To confirm this hypothesis, some visualisation experiments were carried out with a quartz plane reactor. The first observations lead to observe the 71 µm particle fluidization at 11.2 MPa; 39°C while some CO_2 channels have been observed near the critical point (8 MPa; 34°C). However, other experiments are necessary to go into more detail.

Notation:

g	gravitational acceleration ($m.s^{-2}$)
m_p	solid weight (kg)
S	area (m^2)
u	fluid velocity ($m.s^{-1}$)
ΔP	pressure drop (Pa)
ρ	fluid density ($kg.m^{-3}$)
ρ_s	solid density ($kg.m^{-3}$)

Subscript:

mf	at the fluidization beginning
exp	experimental

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