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MASS TRANSFER AND PRESSURE DROP OF A NOVEL STRUCTURED PACKING FOR CO_2 POST-COMBUSTION CAPTURE

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Abstract

A novel packing named Sepcarb® 4D had been developed and characterized within 150 mm internal diameter columns in terms of pressure drop, interfacial area (a_e) and gas side mass transfer coefficient (k_G). The 4D packing consists in a complex arrangement of hollow carbon tubes. Its geometry is given by some geometric parameters, one of which results in the opening fraction, Ω (from 30% to 50% for the present study). Pressure drop curves show that the capacity of the present packing is considerably increased when Ω increases. Flooding limits become higher than those expected for recent Mellapak 500.Y and 750.Y. The geometric area of the 4D packing, a_g, is inversely proportional to Ω. However the effective area, a_e is not reduced when Ω is increased from 30 to 50% which indicates that a_e can considerably exceed a_g for high Ω. It apperas that a_e is very sensitive to the gas flowrate, and much higher than those measured for Mellapak 500.Y, 252.Y and 250.Y. Such results can be explained by a non negligible amount of droplets, and are confirmed by k_G*a_e measurements. The latter are based on water evaporation and require a very sophisticated experimental set-up. In conclusion, the novel 4D packing seems to be very adapted to CO_2 capture.

Keywords: 4D structured packing, capacity, mass transfer, CO_2 capture

1. Introduction

To reduce greenhouse gases emissions, the GASCOGNE project target is to enable the capture of the CO_2 emitted by power plants by proposing a novel packing to allow for important avoided CO_2 cost reduction. In the case of post-combustion capture by an amine plant, typically MEA 30% wt, huge gas flow rates must be treated which lead to very large capture plants. The optimisation of such high volume reactor design is thus of great importance to minimize investments. Since capture process operates downstream the power plant, it also requires low pressure drop. This calls for the development of reliable models for pressure drop and mass transfer characteristics determination of existing packing but also calls for new high efficiency, high capacity packings. A novel carbon-carbon structured packings, the Specarb® 4D packing initially developed by SPS, had been adapted to capture process. The aim of the present paper is to show the results of characterization experiments with this new packing.

2. Packing structure

The new packing is made of hollow carbon tubes. These tubes are formed with carbon fabrics which are woven on a mandrel according to a particular braid angle (Fig. 1a). The braid angle (θ) corresponds to the angle formed between a braid thread and the braid axis (Fig. 1b). The distance between two fibre crossings (Fig. 1c) along the circumference can be adjusted in order to generate some openings in the structure (Fig. 1d). The value of the braid angle determines the tube opening sizes, and the lower the braid angle the bigger the openings.

It has to be noticed that the surface tube opening fraction, Ω, ranges from 0% to 85% which correspond to a unit opening surface area of 0 and 735 mm² respectively. The tubes are arranged according to the four diagonals of a cube (Fig. 2a), which demonstrates why this packing is called "Sepcarb® 4D packing". The layout is repeated in the three spatial directions (Fig. 2b) to obtain the complete structure (Fig. 2c). This structure is very flexible since many parameters can be modified in order to adapt the packing structure (tube diameter, opening fraction and sizes) to the process requirements (capacity, effective area, etc.). In parallel, this packing possesses other interesting properties such as a small tube thickness (0.2 mm), a very low weight of approximately 40 kg per cubic meter, and a good structural cohesion (mechanical strength).

For the present study, two 4D packing geometries have been manufactured and characterized with 150 mm inner diameter columns. Both geometries use 10 mm diameter carbon tubes, and the braid angle is adjusted to obtain an opening fraction, Ω, equal to 30% and 50% respectively. The latter corresponds to a maximum value which should maximize the packing capacity (at a fixed tube diameter). Such geometries will be called 4D-30% and 4D-50% respectively. The corresponding specific geometric area, a_g, equals 420 $m^2.m^{-3}$ and 327 $m^2.m^{-3}$. It has to be noticed that the packed bed is compounded with 50 mm height blocs which are turned at 45° from each other (Fig. 2d).

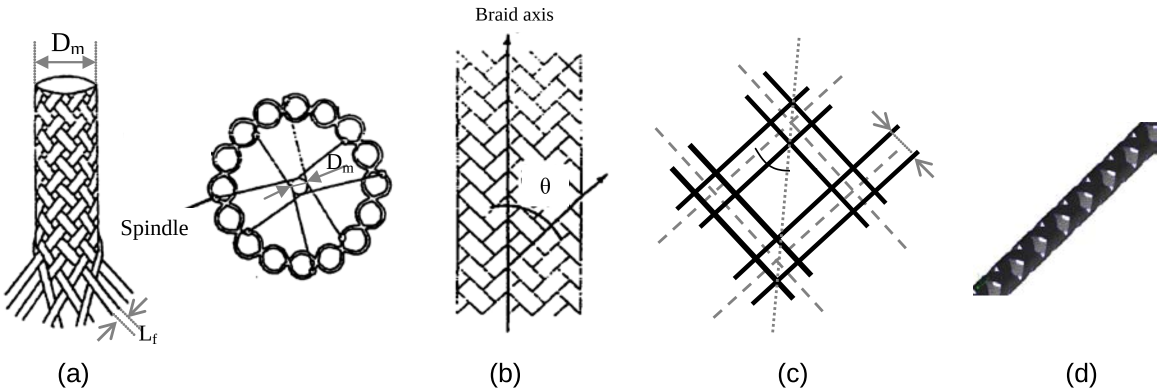

Figure 1. Carbon fabrics woven on a mandrel (a), braid angle (b), crossing of fibres (c), tube with openings (c)

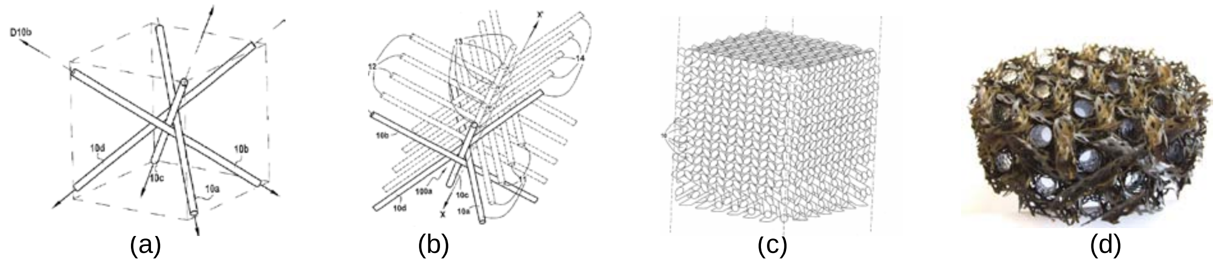

Figure 2. Tubes fitting according to the four diagonals of a cube (a) and reproduction of the layout in one direction (b) for the final structure (c), picture of Sepcarb® 4D packing (d)

3. Experimental set up and methods

3.1. Pressure drop

Eighteen 50 mm height blocs were installed in a 150 mm inner diameter glass column. Then, the packed bed height equals 900 mm. Counter current operation with an air-water system has been used and all studies were carried out at room temperature and under standard atmospheric pressure. The packing was only set in the upper part of 1m high columns; an other 1m part was used to reduce the influence of the perturbations caused by the liquid level at the bottom of the column. The liquid is supplied at the top of the column via a plate distributor. The drip point density of this liquid distributor, equals 283 holes per square meter. This ensures that there is no liquid distributor effect on the results. For the present study, the liquid load, Q_L, ranges from 1 to 60 $m^3.m^{-2}.h^{-1}$. The gas flow is supplied at the bottom of the column and was measured by two different flowmeters. The pressure drop was measured by an inclined U-tube filled with water, which yielded pressure measurements with an experimental error of ± 0.05 mbar. Before each test, the packed bed is pre-wetted by the use of a high liquid load during 30 minutes. This ensures a fully wet of the packing and avoids dry zones.

3.2 effective area, a_e

The air/NaOH (0.1N) system had been selected to measure a_e [1,2]. For this system it is assumed that the measured absorbed rate is directly linked to a_e. CO_2 gas concentration is measured at column inlet and outlet by an infrared system, and a_e is adjusted to fit CO_2 contents. Eight (resp. four) 4D-30% (resp. 4D-50%) packing blocs had been installed in a 150 mm diameter stainless steel column. This corresponds to a total packed bed height of 400 mm (resp. 200 mm). It has to be noticed that, in the case of the 4D-50% packing, there is a non negligible empty height which could increase the experimental error.

3.3 Gas side mass transfer coefficient

Generally used methods of $k_g.a_e$ determination can be classified into three main categories:

- the absorption of a gas/vapour soluble in liquid-phase,
- the evaporation of a pure liquid by an inert gas,
- the absorption with chemical reaction.

Using the method of evaporation of pure liquid, we ensure that the transfer resistance is totally limited to gas-phase. This method is applied here to water as the pure liquid and to air as the inert gas; despite the fact that the method is less used nowadays, it presents however some advantages. The operating mode is to obtain adiabatic conditions of saturation by taking into contact the air with water at counter-current under steady-state conditions. The air humidification as cooling procedure, leads the system towards a stable operating conditions in terms of water temperature at adiabatic conditions of saturation (T_{AS}). The gathered experimental data's at stable condition of functioning permit the determination of k_ga_e, using mass balance for gas-phase[3] :

$$\mathrm{Ln}\left[(Y_{AS} - Y_E) / (Y_{AS} - Y_S) \right] = k_g a\, Z / G_0 \qquad (1)$$

With Y_E et Y_S as absolute inlet and outlet humidities of air in column, G_0 as air flow, Z as column's height and Y_{AS} as absolute humidity at adiabatic temperature of saturation. In order to overwhelm the impact of extremity effects, authors recommend realising the same measurements with at least two different heights of packing. So, our pilot contains a column of 150mm diameter, with water and air at counter-current. Dry air flow is measured; then it is humidified and heated (by an evaporator feeded by a measuring micro-pump) in order to obtain desired conditions of bottom-column. Temperature and dew point are measured at inlet and outlet sections by a hygrometer (hygrometer with cooled mirror permitting an accuracy of ±0,1°C of the dew point). Water circulates in closed loop through storage, up-column, liquid distributor, gas distributor to come back into storage. Flows, inlet and outlet temperatures of column, as well as storage temperature have to be measured. The liquid storage has to be so limited in terms of volume (20 litres) in order to reduce the time of stabilisation. The operating conditions are predicted to obtain an adiabatic temperature of saturation close to the surrounding temperature. For the present study six 4D-30% and 4D-50% blocs had been tested.

4. Results

4.1. Pressure drop

Figure 3 gives the wetted pressure drop as a function of F_s for the 4D-30% and 4D-50% packings. The liquid load is fixed at 23 $m^3.m^{-2}.h^{-1}$ which is a typical value for CO_2 absorbers. Present results are compared with calculated pressure drop (manufacturer software Sulcol 2.0) for different structured packings commercialized by Sulzer Chemtech. It has to be noticed that the specific geometric areas of Mellapak 500Y and 750Y (a_g=500 and 750 $m^2.m^{-3}$ respectively) are closed to the 4D ones (a_g=420 $m^2.m^{-3}$ with Ω=30%, down to 327 $m^2.m^{-3}$ with Ω=50%). For this study, the MellapakPlus 252Y (M252Y, a_g=250 $m^2.m^{-3}$) is the reference case.

First, one can observe that the 4D-30% pressure drop is a little bit higher than the one generated by Mellapak 750Y (M750Y). Its capacity (flooding limit) is comparable to the M750Y and M500Y ones. However, the M252Y capacity is much higher (factor 2). When Ω increases from 30 to 50%, it reduces the generated pressure drop significantly since 4D-50% generates less pressure drop than M750Y. In parallel, the capacity becomes higher than the M500Y one. However the M252Y capacity is still higher (+ 40%).

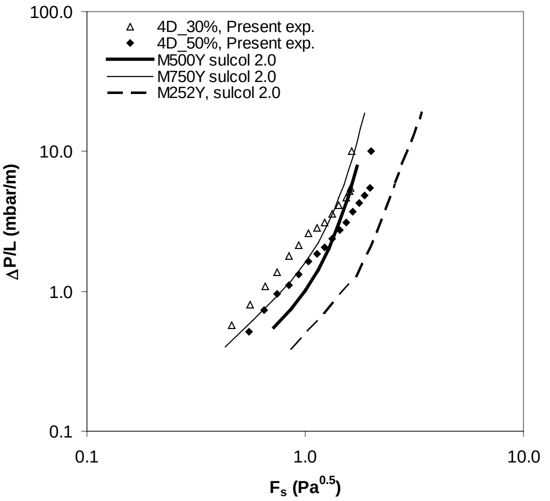

Figure 3. Wetted pressure drop for 4D-30% and 4D-50% as a function of F_s. Q_L=23 $m^3.m^{-2}.h^{-1}$. Comparison with Sulcol 2.0 calculations for M750Y, M500Y and M252Y.

4.2 Mass transfer : effective area

Figure 4 gives the 4D-30% and 4D-50% effective area, a_e, as a function of Q_L with a large range of F_s. This effective area is compared to those measured for M500Yand Mellapak 250Y (M250Y, a_g=250 $m^2.m^{-1}$). First, it has to be noticed that M500Y and M250Y effective areas had been measured with the same chemical system and a 430 mm inner diameter column[4]. Second, M252Y effective area had been measured with the same 150 mm inner diameter column, however the chemical system was different[5]. M250Y and M252Y effective areas are comparable to a_g, not very sensitive to F_s and similar. The latter was expected since both geometries are very similar, and it tends to indicate that scale and chemical system effects are moderate for this packing. M500Y effective area is much higher than the M252Y and M250Y ones (+ 75%) in spite of the fact that a_e is much lower than a_g (a_e/a_g < 0.7), and a_e is not very sensitive to F_s.

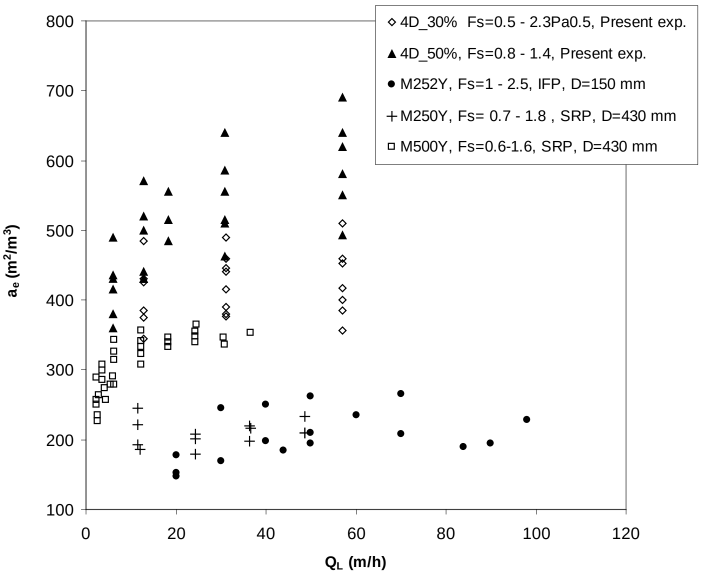

Figure 4. Effective area for 4D-30% and 4D-50% as a function of QL. Comparison with effective area measured with commercial structured packings M250Y[4], M252Y[5] and M500Y[4].

4D-30% and 4D-50% effective areas are somewhat higher than the M500Y one, and very sensitive to F_s which explain why experimental points are more dispersed. The ratio a_e/a_g could be higher than 1. The strong impact of F_s and the high a_e/a_g ratios could be explained by the fact that large openings generate a non negligible amount of droplets. 4D structures seem to be very interesting since they are much more efficient that commercialized packings. For example a_e is at least doubled comparatively to M252Y. 4D-50% effective area appears to be higher than the 4D-30% one, this leads to higher a_e/a_g ratios with Ω=50%. However, these very good results could be a consequence of the higher experimental error (cf. 3.2). To illustrate this, k_G*a_e measurements show an opposite trend (cf. 4.3). Then, the main conclusion is that a_e should not be significantly decreased when Ω ranges from 30 to 50%. The 4D-50% structure could be more adapted to CO_2 capture than 4D-30% one since the efficiency is not really affected by Ω increased, while capacity is significantly increased. However result obtained with 4D-50% should be further validated with a higher packed bed height in order to reduce void zone effects.

4.3 Mass transfer : k_G*a_e

$k_g.a_e$ measurements are realised with 3 different heights of packing in order to deduce the extremity effects. First, the packing 4D-50 % had been tested for a liquid load scale of 1,5-23 $m^3.m^{-2}.h^{-1}$, and for a gas flow rate scale of 3500-7200 $kg.m^{-2}.h^{-1}$. The values of $k_g.a_e$ are presented in logarithmic scale as a function gas flow rate's logarithm in figure 5. Regarding to this figure, two straight lines (dotted) are added which correspond at + 10% and -10% of the average value, and points are included between these two boundaries.

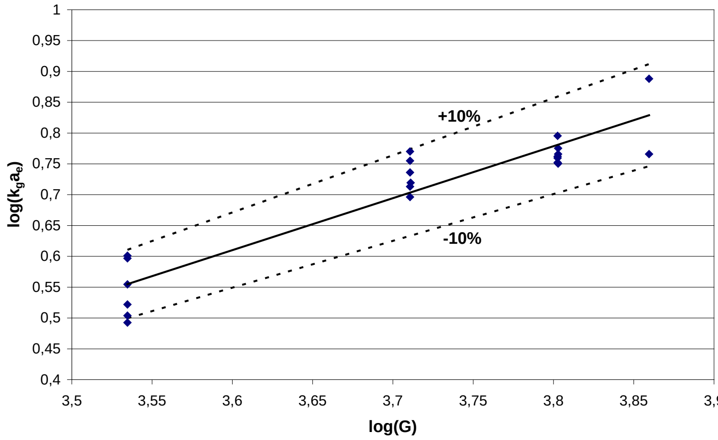

Figure 5. Dependency of $\log(k_g a_e)$ as a function of log(G) for Sepcarb® 4D 50%

Therefore, under functional conditions of this packing, the results present a quasi-independency on liquid flow rate, and an important dependency on gas flow rate. The following relationship is then suggested:

$$k_g*a_e = 3{,}7488.10^{-3} * G^{0{,}843} \quad (2)$$

with G in $kg.h^{-1}.m^{-2}$ and k_g*a_e in s^{-1}. The power of G is around 0,8 even if it is not traditional, it has been already observed for some other internals of column. However, it is possible that this value is linked to the fact that a_e is very sensitive to the gas flowrate (cf. 4.2). Regarding to its particular structure, this packing generates:

- liquid droplets through large openings,
- liquid film which follows the assembly angle of packing tubes and therefore an increase in movement with respect to a vertical flow.
- the possibility of creating the equivalent of a bubbling in the middle of tubes of packing structure at high gas flow rates, with maybe a possible liquid upstream in the channels formed by tubes.

Second, the 4D-30% packing is tested with a liquid load equals 16 $m^3.m^{-2}.h^{-1}$ and the previous range of gas flow rate. The results are illustrated in logarithmic scale in figure 6. One can observe a high dependency on LOG(G) (1,54 power of G), as for the 4D-50%. This could be also explained by the fact that a_e is very sensitive to the gas flowrate.

$$k_g*a_e = 1{,}4612.10^{-5} * G^{1{,}54} \quad (3)$$

with G in $kg.h^{-1}.m^{-2}$ and k_g*a_e in s^{-1}.

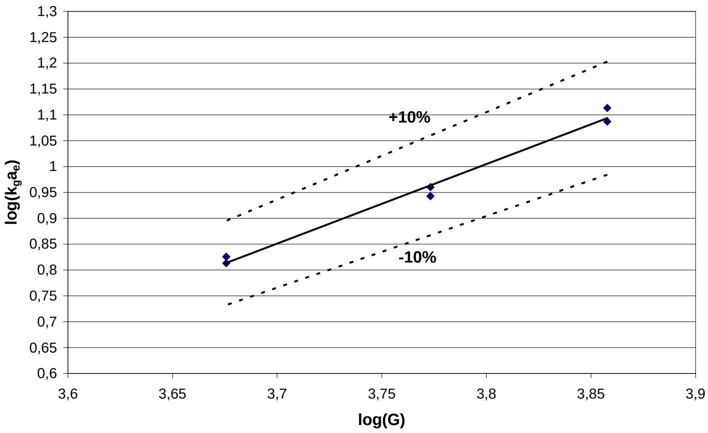

Figure 6. Dependency of log(k_ga_e) on log(G) for the Sepcarb® 4D 30%

5. Conclusions / perspectives

A novel packing named Sepcarb® 4D had been developed for CO_2 capture and characterized in terms of pressure drop and mass transfer (*a_e, k_g*). For an opening fraction comprised between 30% and 50%, 4D capacity is similar or higher than Mellapak 500Y and 750Y ones. However the capacity of the MellapakPlus 252Y is at least 40% higher. 4D packings effective areas are much higher than those measured with Mellapak 500Y, 250Y and 252Y. The opening fraction increase do not seem to affect the interfacial area for the tested range, and *a_e* is very sensitive to the gas flowrate. The latter is confirmed by k_g*a_e measurements. The 4D packing seems to be very interesting for CO_2 capture since its capacity is acceptable (acceptable absorber diameter) while the effective area is very high (low absorber height).

Effective area measurements will be further conducted with 4D-50% structure to valid present results by minimizing the void zone impact. Then, a 400 mm prototype will be realized and characterized. This will allow to check scale effects in order to extrapolate present results to industrial plants. In parallel, process calculations will be run to valid the interest of the 4D packings by including all parameters (packed bed volume calculations via mass transfer parameters, costs, etc.).

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Nomenclature

a_e : interfacial area ($m^2.m^{-3}$)
a_g : geometric area ($m^2.m^{-3}$)
D_m : diameter of the mandrel (m)
F_s : F-factor = $G_0.\sqrt{\rho_g}$ ($Pa^{1/2}$)
G : Gas flow rate ($kg.h^{-1}.m^{-2}$)
G_0 : air flow ($m.s^{-1}$)
k_g : gas side mass transfer coefficient ($m.s^{-1}$)
L_f : width of carbon fabric (m)
Q_L : the liquid load ($m.h^{-1}$)
T_{AS} : temperature at adiabatic conditions of saturation (°C)
Y_{AS} : absolute humidity at adiabatic temperature of saturation
Y_E,Y_S : absolute inlet and outlet humidities of air in column
Z : column’s height (m)
θ : braid angle (°)
ρ_G : gaz density ($kg.m^{-3}$)
Ω : the opening fraction (%)

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