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USE OF MULTI-SCALE SIMULATIONS FOR LOWERING THE COST OF CARBON DIOXIDE CAPTURE

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Abstract
Carbon Capture and Storage is one important option for CO_2 mitigation[1]. Post-combustion capture processes using amines are considered as one of the preferred options for CCS. However, the cost of avoided CO_2 is very large and must be reduced. In the present article, it is shown from a macro-scale technico-economic analysis, that Capex represents about one third of the total CO_2 cost. A sensitivity analysis, via Aspen calculations performed at column scale, enables to identify key parameters that control column design. It is shown that the most important mass-transfer parameters is the interfacial area, the gas and liquid mass transfer coefficients having almost no influence. From CFD simulations performed at a finer scale, some insights are given in order to optimize column design.

Keywords: CO_2 capture, absorption, process evaluation, CFD, packing

1. Introduction

In the context of climate change and CO_2 mitigation, it is now well known that Carbon Capture and Storage, CCS, is one important solution to develop[1]. On the one hand, the development of this technology must go fast enough to meet environmental targets; on the other hand, the captured CO_2 will not have any direct added-value; it is then believed that only processes with lowest capture costs will be deployed. It is thus of high importance to identify the key parameters that affect the avoided CO_2 costs in order to be able to identify the subjects on which research and development must focus.

The Castor EU project has shown that post-combustion capture (PCC) processes based on the use of chemical solvents actually meet the requirements of 90% recovery of CO_2 from flue gas while delivering a high purity CO_2 for storage. However, the reference case, being the 30%wt. MEA process, is known to be energy consuming. Indeed, about 3.7 GJ/ton_CO_2 are required for solvent regeneration[2,3], which represents more than 50% of the operating costs. While lots of actions are now under way to find new original solvents requiring less energy consumption[e.g.2,4], much less works dealing with absorber design and linked investments are conducted. The impacts of design choices such as the pressure drop and mass transfer characteristics of the considered packing or the choice for the distributors on both investments and operational costs are indeed not precisely known.

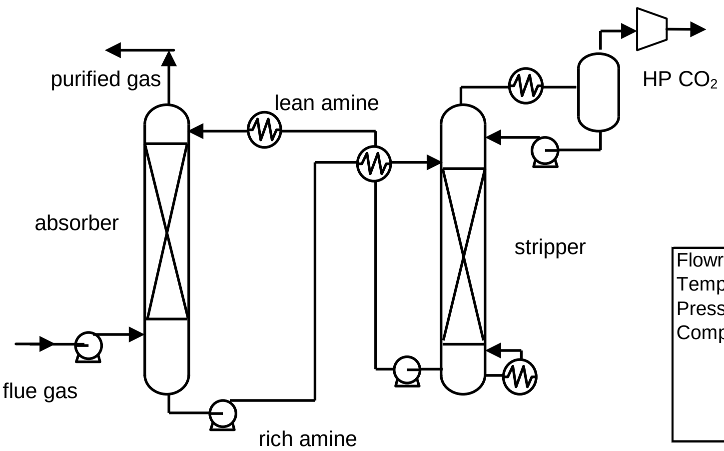
The main purpose of the present study is to show where R&D efforts should be put, according to where cost sensitivity is high. Particular attention is put on the design of the absorber. The second purpose is to show that proper process development must rely on different simulations tools used at different scales, one simulation at a given scale giving insights for the other. The followed approach is described in section 2, while section 3 is dedicated to the different results obtained at different scales.

2. Approach

The present approach involves three different types of simulations. First, a technico-economic simulation is performed at process global scale in order to determine the avoided CO_2 cost repartition. Second, process simulations are performed at reactor scale in order to study the main parameters that impact absorber design. Last, CFD simulations are used both to show how internals may impact the quality of the gas/liquid flow within the packed bed (meso-scale). All these three types of simulations are described in the following sub-chapters.

2.1 Process evaluation

To evaluate the impact of a design on the economics of a process, one has to consider the repartition of operating costs as well as the repartition of the investment costs. So far, most of the studies focus on the energy consumption and processes are compared between each other in terms of required GJ per ton of avoided CO_2[2]. The aim of the present process evaluation is to precisely determine the cost of the process in terms of both investment and operating costs. The result is expressed in €/t$_{CO_2}$. The process considered for analysis is the standard MEA 30%wt. process for CO_2 capture. The unit considered is applied to the treatment of 90%vol. of the CO_2 emitted by a 630 MWe CFB coal power plant. Flue gas characteristics and a simplified process flow diagram of the standard CO_2 capture plant are shown in Figure 1.



Flowrate (kmole/h)	78840
Temperature (°C)	45
Pressure (bar abs.)	1
Composition (%vol.)	
CO2	13
N2	75
O2	5
H20	7

Figure 1. Simplified process flow diagram of the MEA process and flue gas characteristics.

The approach followed in the present paper to evaluate the cost of a given process consists of the following steps :

Simulation of the process: The first step of the process evaluation consists of simulations of the absorption/desorption loop, using a commercial software (Aspenplus, see section 2.3). The boundary limit considered in the study includes flue gas at atmospheric pressure and delivery of CO2 at high pressure (110 bar abs.) The simulation obtained is used to determine the heat and material balance of the capture unit.

Process design of main equipments : Using the results of the simulation, the main equipments simulated are designed using an IFP in-house software. To obtain a quick and representative evaluation of CAPEX, only the main equipments are considered, all being shown in the process flow diagram of Figure 1 and given in Table 2.

Cost evaluation : The main equipments designed at the previous step are then cost evaluated using an IFP in-house software. The sum of CAPEX obtained is then used along with the OPEX obtained from the simulation in the economic evaluation. This light procedure allows to end-up with rough price of the process. Different modifications on the design are then quickly evaluated, allowing in particular a cost comparison of absorption tower design.

2.2 Economic evaluation

In Table 1 are described the main economical hypothesis considered to realise the cost analysis.

Table 1. Economical evaluation hypothesis

Reference year	2008
Capital allowances (yr)	25
Discount rate (%)	10
Owner cost (%)	10
Time of construction (month)	36
Interest rate (%)	8
Coal price (€/GJ)	2.3

2.3 Absorber design

The Castor pilot plant of Dong Energy is equipped with the Koch-Glitsch third generation random packing IMTP-50[2]. Since the design of the absorber is directly linked to the packing; it is of high interest to determine the sensitivity of the design towards it. The choice of the most adequate packing is linked to its performances in terms of pressure drop and mass transfer efficiencies, a compromise between capacity and efficiency being looked for. The capacity of the packing, which is further used to determine the diameter of the column, can be easily determined from packing vendors softwares. The efficiency, which is further used to determine the height of the column, is much more difficult to determine. The global mass transfer coefficient, $K_G a$, commonly linked to HETP or NUT values is given by the three parameters, the liquid side mass transfer coefficient, k_L, the gas side mass transfer coefficient, k_G, and the effective area, a_e, via the following relationship :

$$\frac{1}{K_G.a_e} = \left(\frac{1}{k_G} + \frac{He}{E.k_L}\right).\frac{1}{a_e} \tag{1}$$

where, *He (-)*, is the Henry coefficient and, *E (-)*, is the acceleration coefficient, both parameters being linked to the solvent characteristics in terms of thermodynamics and kinetics respectively. When comparing their simulations to the results of the Castor pilot plant, Dugas et al.[5] showed that the AspenTech Aspenplus 2006.5 software with Aspen RateSep module can accurately simulate the absorber performances provided that appropriate thermodynamics and kinetics corresponding to the 30%wt.MEA solvent are used. RateSep is a detailed model that takes into account mass and heat transfer transport equations in both gas and liquid phases, equipment hydrodynamics and chemical reaction mechanisms to predict column performance. With such a model the enhancement factor, *E* in Eq.1, is determined via the resolution of a transport equation solving species diffusion and reaction in the liquid diffusion film, the thickness of which being determined from the liquid mass transfer characteristics. The simulations performed in the present study are similar to those of Dugas et al.[5] in terms of thermodynamics and kinetics. But here, we considered the mass transfer parameters as variables. These parameters were modified in in-house Fortran model routines in order to characterize the different packings studied.

The work performed here has been done in two steps. First, a sensitivity analysis towards all three parameters, k_L, k_G, and a_e, has been conducted. In this case, each one of the three previous parameters is varied while all other parameters, flow conditions and design are being kept constant. The simulations give the respective CO_2 capture performances. Second, a comparison between IMTP packings (25, 40, 50 and 70) at two levels of flooding percentage, 50 and 80%, has been made. The determination of flooding percentage is done with the commercial software KG-Tower 4.0 for flow conditions at bottom of the absorber. The determination of the height is done using Aspen calculations with IFP in-house correlations for mass transfer parameters. These calculations are carried out for a constant CO_2 capture rate of 90%.

2.4 CFD simulations

CFD is more and more used to calculate flow characteristics in packed beds. Raynal and Royon-Lebeaud[6] have shown how simulations using different types of modelisation could complement each other in order to simulate gas/liquid flow in packed columns. Two main types of modelisation can be considered. The Volume Of Fluid (VOF) approach is used to simulate the gas/liquid flow at liquid film scale[7,8]. From such simulations one can determine local parameters such as the liquid film thickness, the liquid film velocity at interface, the wetting quality which are further used in determining parameters used in process simulations such as the liquid holdup, the liquid side mass transfer coefficient, via the Higbie theory or via direct simulations, and the effective area respectively for different types of packings[6,9]. Despite the interest of the latter approach, we focus here on the Euler/Euler approach to simulate the gas/liquid flow at column scale. The latter simulations, performed with the Fluent 6.0 commercial code with the standard *k-ε* turbulent model, enables to determine the quality of the distribution and the internals / packed bed interaction. In such an approach, some developments are needed for the gas/liquid interaction closure terms. The determination of such closure laws can either be done via appropriate measurements in perfectly controlled experiments or by CFD VOF calculations[6].

3. Results

3.1 Process simulations and economical evaluation

Here are summarised the results from the process simulations. The results given here below are an optimised configuration[7,10] of the process, the column design being optimized for IMTP50 packing.
- Process specific energy consumption (steam for reboiler) = 3.6 GJ/tco_2
- Solvent flowrate = 7100 m^3/h
- Absorber design (4 columns) - Diameter 8.8m, Height 40m, packing IMTP-50
- Regenerator design (2 columns) - Diameter 7.8m, Height 35m, packing IMTP-50
- Amine/amine heat exchanger (4 plates type) : 21 700m²
- Amine reboiler (16 Kettle type) : 25 MWth
- CO_2 compressor : 39 MWé

The corresponding cost analysis is given in Table 2. The CO_2 penalty obtained is 76 €/tCO_2 considering a non integrated plant (steam and electricity bought on market at 21.7€/t and 38.3€/MWh respective prices). By considering a non integrated power plant, there is no loss of efficiency of the plant, the penalty cost of CO_2 comes from the OPEX. One first observes that Opex represent more than 2/3 of the CO_2 capture cost, the energy penalty at reboiler being by far the most important cost. Second, one observes that Capex cannot be neglected since they correspond to 1/3 of the latter costs. Third, columns account for more than 50% of the Capex, when compression is excluded. Last, the energetic cost due to the use of a booster fan to overcome the pressure in the process corresponds to 2% of the operational costs. The choice of packing and corresponding designs of columns are thus of high importance if one expects a major CO_2 capture cost reduction.

Table 2. Cost repartition

	CAPEX (%)	OPEX (%)	CAPEX* (%)	OPEX* (%)	CAPEX (€/tCO2)	OPEX (€/tCO2)
Repartition	**32**	**68**				
Absorber	**28**		**40**		6.8	
Regenerator	**9**		**13**		2.2	
Flue gas blower	1	2	1	2.2	0.2	1
Amine rich pump	3	1	4	1.1	0.8	0.5
Amine/amine heat exchanger	10		14		2.4	
Reboiler	18	87	26	96.7	4.5	45.1
CO2 compressor	30	10			7.3	5.2
Sub-TOTAL			* compression excluded		**24.2**	**51.8**
TOTAL					**76**	

3.2 Absorber simulations

3.2.1 Sensitivity Analysis

Figure 2 shows the results concerning the sensitivity analysis toward mass transfer parameters. Figure 2.a shows the dry molar fraction of CO_2 in the gas stream along the absorber for various interfacial area values all other parameters being kept constant. The reference case corresponds to in-house values of a_e measured at IFP in good agreement with data deduced from the Intalox Packing brochure[11] or found in Billet[12]. One observes that slight changes of 20% (squares) or 40% (triangles) around the reference values have an important impact on the CO_2 profile further impacting the total performance of the absorber. A change of ±40% in the effective area induces changes from 1.5% to 3.2% of the CO_2 molar fraction at absorber outlet. Figure 2.b shows similar data but with changes in the gas side mass transfer coefficient. One observes that the sensitivity towards this parameter is much less than the one observed towards the effective area. A change of a factor of 25 on the gas side mass-transfer coefficient induces CO_2 molar fraction at absorber outlet values ranging from 2.5% to 2.2%. It can thus be concluded that, this process is not gas side controlled at all. Similar calculations have been performed to check the influence of the liquid side mass-transfer. The latter has been varied on a range from 1 to 7 around a reference value; the CO_2 molar fraction at absorber outlet happens to vary very little, from 2.2 to 2.3%.

The present sensitivity analysis clearly shows that, the performance of the absorber, or the height of the absorber for a given CO_2 capture rate, is mostly given by the effective area. There is almost no gas side resistance, and one can conclude that the column operates in the pseudo-first order regime in which the liquid side mass transfer coefficient has almost no impact, the enhancement factor being equal to the Hatta number. Prediction of mass transfer characteristics or packing efficiency is thus entirely given by the effective area.

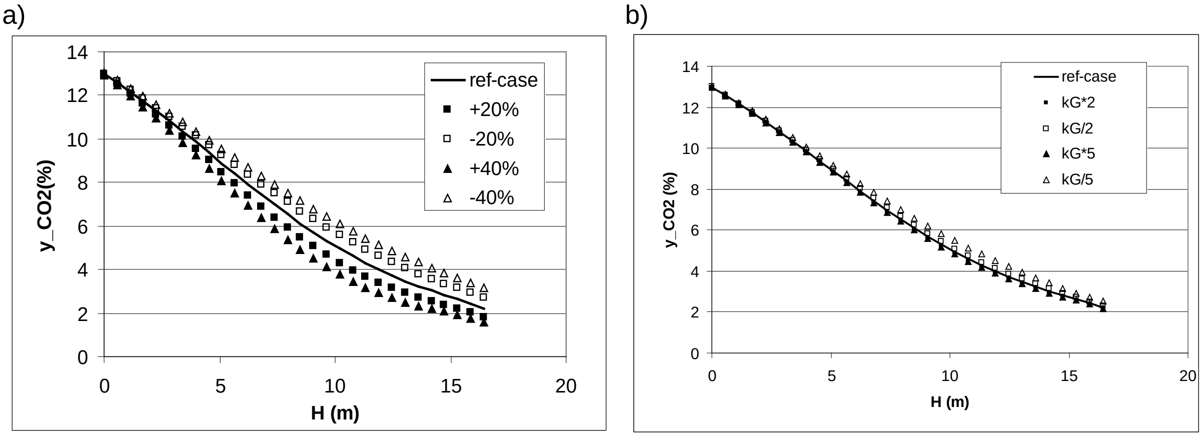


Figure 2. CO_2 gas concentration profiles along the absorber; a) influence of the effective area, a_e, b) influence of the gas side mass transfer parameter, k_G.

3.2.2 Packing influence

From previous results we know that the changes from one packing to another will impact on the height of the column via its interfacial area and on the diameter of the column via its capacity, the first parameter having a strong impact on the investments costs, the combination of the two having a moderate impact on the operational costs via the bed pressure drop. Figure 3 shows a comparison between the different packing of IMTP types for two different values of flooding percentage all other parameters being kept constants. One first observes that small size packings, IMTP 25 or 40, induce larger diameters than larger size packings, IMTP 50 or 70 but smaller height of packed beds. One also observes a relatively important impact of the flooding percentage on the diameter values while having little effect on the corresponding height. With such curves and with the economic methodology given in section 2, one can make the balance between capacity and efficiency transposing them into economic terms, that is cost of the column and power of the blower. From such an analysis, it is possible to properly choose the packing that corresponds to minimum costs.

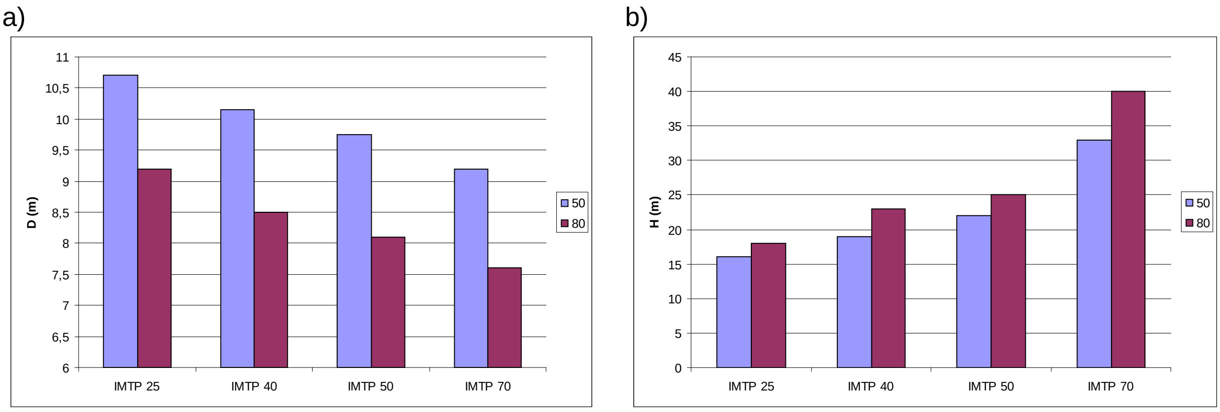


Figure 3. Design values needed respectively for the diameter (a) and the height (b) for two flooding percentage values, 50 and 80% for the different IMTP packings for achieving a CO_2 capture rate of 90%.

3.3 CFD simulations

In all previous simulations steps, one assumes perfectly distributed flow, which is quite easy to do at laboratory scales, but no longer obvious at large scale in particular when low gas pressure drop values are required and when dimensions are so huge. Indeed from Figure 4, one observes that the change from one distributor to another has no impact far from flooding, but may dramatically influences the gas/liquid flow at high gas flow (see dotted lines in Fig.4a). Such an impact can be explained via CFD simulations performed with identical conditions as experiments. Indeed, as shown from Fig.4b, the D1 distributor induces high velocities at the periphery of the column where one has higher local liquid retention thus inducing an "early flooding"; on the contrary, distributor D2, while still not perfect, induces high velocities at the core of the packed bed avoiding zones with high liquid concentrations.

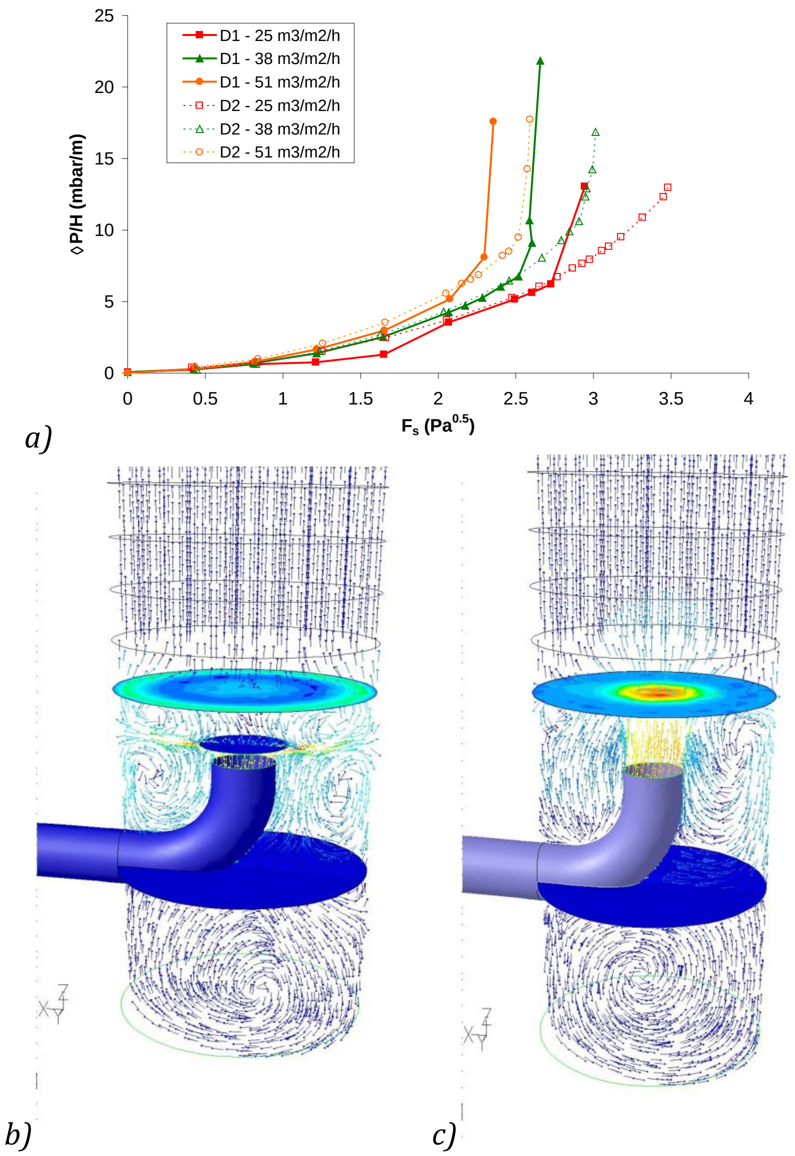


Figure 4. Influence of the gas distributor. a) experimental pressure drop curves versus gas load for three values of liquid load for two types of R&D gas distributor, column of 1 m in diameter at ambient pressure, gas is air liquid is water (distributor D1 : curved pipe with baffles – closed symbols with continuous lines; D2 curved pipe without baffles – open symbols with dotted lines); b-c) CFD pressure contours at bed inlet and velocity field in the y=0 plane for same gas and liquid flow conditions, only the gas distributor differs; b) D1 : curved pipe with baffles, c) D2 curved pipe without baffles.

4. Conclusions

It is shown here that a strategy implying different simulations tools at different scales enables to identify the key parameters that impact CO_2 capture cost. It is thus of high interest to combine economic estimation tools with process simulations and CFD simulations tools to be able to achieve the most optimized design in terms of choice of packing and adapted corresponding internals.

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