A NUMERICAL STUDY OF THE CARBONATOR DESIGN EFFECT ON CO_2 CAPTURE EFFICIENCY

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ABSTRACT

This work investigates the effect of the carbonator riser geometry on the gas-solid flow hydrodynamics and CO_2 capture efficiency, using a validated 3D CFD model. Five different design/operating concepts are tested and compared; an initial carbonator design (reference design) part of the 1 MW_{th} Dual Fluidized Bed (DFB) installation in TU Darmstadt (TUD) and four retrofitting concepts of the initial design. These concepts include two fat bottom design (FBD) test cases –the one with the same solids inventory as in the reference case, i.e. 282 kg, and the other with a higher solids inventory, i.e. 340 kg, and the same pressure drop as in the reference case- a sorbent staging design (SbS) and a sorbent in splash zone design (SZD). As concerns the applied numerical model, the TFM Eulerian approach coupled with the sub-grid EMMS drag scheme and an heterogeneous reaction rate obtained through the work of Hawthorne et al., is incorporated into the ANSYS FLUENT^{TM} commercial software. This model has been validated against experimental data for the reference design and the respective results and conclusions have been already presented in previous works. Likewise to the initial geometry numerical investigation, the numerical results of the retrofit cases are averaged over a time period of 65 seconds. The results of the CFD model indicate that a better CO_2 capture efficiency is achieved in all retrofit cases compared to the reference case. Specifically, a higher capture efficiency is achieved in the SbS case (89.94 %) than in the SD case (87.03 %), mainly because a better mixing is achieved in the lower part of the reactor. The efficiency in the SZD case is lower than the SbS concept (88.7 %) a fact that confirms the advantages of staging the sorbent injection since injecting all of the material either to the bottom or to the splash zone leads to lower capture efficiency. The best CO_2 capture is achieved in the FBD concept, i.e. 90.36 % and 91.91 %, for a solids inventory equal to 282 and 340 kg, respectively, values that are close enough to the CO_2 equilibrium capture efficiency (93.33 %). Amongst the cases examined, the SbS case is a very attractive solution since with a relatively simple design alternation, a significant improvement, in terms of capture efficiency, is achieved.

Keywords: Carbonator Design optimization, Fat bottom design, Sorbent staging, TFM-EMMS, CO_2 capture efficiency

INTRODUCTION

The CaL process is a post-combustion CO_2 capture technology that has been tested successfully up to present on a bench and pilot scale, ranging from 3 kWth up to 1.7 MWth [1-4]. Recently, attempts are being made towards its up-scaling to an industrial scale [5, 6]. For the implementation of this technology on a large scale, modeling work is essential to eliminate any financial risks that might arise during the construction of such units [7]. In this frame, computational fluid dynamic (CFD) models can be valuable cost-efficient tools for the optimum design and up-scaling of CaL systems.

In a calcium looping system the carbonator reactor is a critical component. Its optimal design and operation can increase considerably the CO_2 capture efficiency. Thus, a meticulous study of its performance under varying operating conditions and design parameters should be undertaken prior to constructing large-scale units. However, even though the study of the carbonator design effect on the CO_2 capture efficiency is an essential work, there are not any available published works in the recent literature; most of the research is focused on matters relative to the carbonation reaction itself, such as the sorbent maximum activity (X_{max}) [8], the effect of sulphur[9]/ hydration[10] on reaction kinetics etc. Some other available works only deal with the pilot plant operation under different carbonator operation regimes, but design effects have not been studied yet [11, 12]. However, the importance of a CFB reactor design on its performance is an undeniable fact that has been addressed in [13], as well.

In general, in reactor design the main target is the search of the appropriate size and operating method of a reactor for a specific process [14]. An optimum carbonator design for a CaL system should enable high solid particles residence times and good gas-solid mixing resulting, thus, in an efficient carbonation reaction. An idea, quite easy and flexible, is to provide the carbonator riser with more than one sorbent feeding lines, a concept that has been introduced in CFBC boilers [13] where solid fuels that are needed to react with gaseous species are injected in the boiler through various inlets. Another alternative is to modify the riser bottom bed design and introduce a varying riser diameter, rather than a constant value of this design parameter. Such concepts are quite innovative and have not been tested yet in a CaL system.

In the present study, five different carbonator design concepts are tested and compared, regarding static pressure profiles and CO_2 capture efficiency. These concepts include a reference carbonator design, part of the Dual Fluidized Bed facility of TUDA, with a riser height and diameter equal to 8.661 m and 590 mm, respectively, and four retrofitting concepts of the reference design, i.e. Fat Bottom Design (FBD) with two different solids inventories, Sorbent Staging Design (SbS) and Sorbent in Splash Zone Design (SZD). In the FBD case the riser diameter is modified and taken equal to 759 mm up to a height of 1 m from the distributor, whilst for a height range equal to [1, 8.661] m the riser diameter remains the same as in the initial geometry, i.e. 590 mm. In the SbS case, 10% of the regenerated material that comes from the calciner unit, is injected in the carbonator through a section (staging inlet) designed approximately 1 meter above the existing loop seal opening that connects the two reactors, whereas 90% of it keeps entering the unit from the existing loop seal section; in the reference design the regenerated sorbent enters the carbonator exclusively through the loop seal. Finally, in the sorbent in splash zone design (SZD) 100% of the regenerated material is injected from the staging inlet and 0% from the existing loop seal section. Three-dimensional CFD simulations of the CFB carbonator riser are carried out in each case in order to evaluate those scenarios effectiveness. The TFM Eulerian approach coupled with the sub-grid EMMS drag scheme is incorporated into the ANSYS FLUENT™ commercial software. The model applied has been already tested and validated for the reference carbonator design [15], using experimental data provided by TUDA.

NUMERICAL MODEL

In the present work a 3D numerical model based on the Eulerian approach is applied for the simulation and comparison of four different carbonator design/operation concepts. The riser of the carbonator unit, depicted in Figure 1a, is used as a reference design for the four retrofitting concepts, summarized in Table.1.

The first retrofitting concept includes the construction of the TUDA carbonator riser with a fat bottom design (FBD), Figure 1b. In the FBD concept the new riser diameter is equal to 759 mm, greater than that of the 590 mm of the reference design, and extends upwards, up to 1 m from the distributor section. For a height range equal to [1, 8.661] m the riser diameter remains equal to that of the initial geometry. The cyclone, the two loop-seals and the auxiliary burner section are not included in the simulation process likewise in the initial geometry. However, in their intersections with the riser proper boundary conditions are applied for the gas and solid phase, the same as the ones applied in the reference design. Loop Seal 1 is used for the recirculation of the solid particles inside the carbonator, whilst Loop Seal 2 performs the transportation of the regenerated limestone from the calciner to the carbonator unit. The solids inventory is set equal to 282 kg, as in the initial geometry. However, the new riser diameter taken for a height range equal to [0, 1] m, results in a lower pressure drop at this area compared to the reference design. In the second concept, the same fat bottom design is tested, without changing the boundary and operating conditions, except for the solids inventory, which is set equal to 340 kg. This value gives almost the same pressure drop with the one obtained for the reference design. It should be noted that from a designer point of view the approach of comparing cases characterized by the same pressure drop makes a technical sense, since the pressure drop and not the mass in the reactor affects the energy self-consumption of such a unit.

In the third concept, i.e. the sorbent staging design (SbS) concept, 10% of the regenerated sorbent, coming from the calciner unit, is injected from a higher height, approximately 1 meter from Loop Seal 2, whilst the rest of it keeps entering the reactor from Loop Seal 2. The geometrical configuration is the same as in the reference design, with the exception that an additional face is inserted, used as a mass flow inlet of the sorbent injection staging. The sorbent staging inlet has the same dimensions, with the intersection of the Loop Seal 2 section with the carbonator riser. In the fourth concept, i.e. the sorbent in splash zone design (SZD), 100% of the regenerated sorbent is injected exclusively from the sorbent staging inlet and a wall boundary condition is assigned to Loop Seal 2 face.

For the numerical simulations three-dimensional structured grids are constructed with ANSYS Meshing component fully respecting the modified geometries of the carbonator riser. The discretized domain consists of 35,870 hexahedral elements for the FBD case and of 31,346 hexahedral elements for the SbS and SZD cases. Both numerical grids correspond to a d_{cell} *to* d_p ratio equal to 465.64. A grid independency test has been already conducted for the reference geometry case [15].

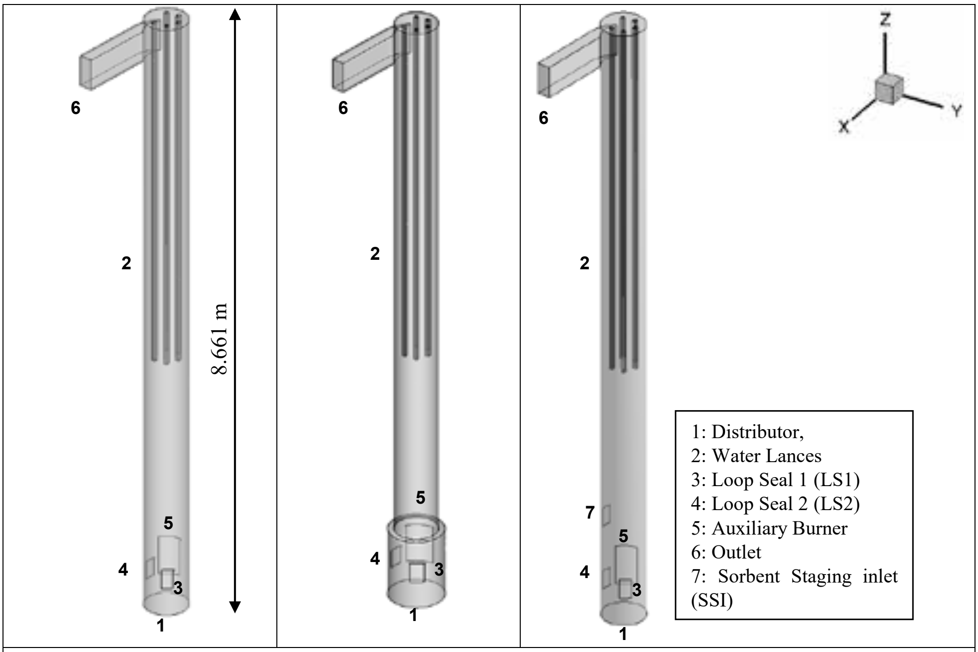

Fig. 1. Geometry layout of the a) Reference design, b) Fat bottom design (FBD) and c) Sorbent Staging (SbS)/Sorbent in Splash zone design (SZD).

The CFD model is incorporated into the ANSYS Fluent 15 platform. Transient calculations are performed, with a time step size equal to 10^{-4} s. Parallel computing is used, with 12 parallel cores, in order to enhance the CFD model computational efficiency. A time interval equal to 65 seconds is taken for the time averaging of the numerical results. It should be noted that 5 s after the initialization in all cases, an equilibrium state is almost reached as concerns the pressure profile. However, the solid species, i.e. CaO and $CaCO_3$, delay in reaching equilibrium requiring thus longer simulation times of almost 200 s in order to predict them correctly. The $CaCO_3$ mass fraction is initialized with a value of 0.16 close to the value of 0.1655 that corresponds to X_{max}=0.1, in order to speed-up the simulation. The phase-coupled SIMPLE scheme is applied for the pressure-velocity coupling. As concerns the spatial discretization of the main governing equations, i.e. momentum, volume fraction and species equations, the QUICK scheme is applied. The time discretization is conducted by a bounded second order implicit scheme [16]. A detailed description of the governing equations used can be found in the ANSYS Fluent Theory Guide [16]. For the drag force exerted on the solid particles the sub-grid EMMS scheme with a new cluster correlation is applied. This model has proven to give more accurate results than the homogeneous Gidaspow model. The limestone particles are of type Geldart A, with a density equal to 1650 kg/m^3 and a mean particle diameter 91.39 μm. The carbonation of CaO particles to $CaCO_3$ is taken into account by incorporating into the CFD model the reaction rate of Hawthorne et al. [17]. The solution of the energy equation is not included in the simulated case due to the fact that the CFB operates under almost isothermal conditions. Finally, the flow is simulated as laminar. Features of the CFD model that are not supported by the CFD platform, such as the EMMS model and the carbonation reaction rate are incorporated through custom-built UDFs. The boundary and operating conditions, except for some modifications analysed above, are presented in a previous work [15].

Table.1: Compared cases simulated.

Case Number	Test Case	Solids Inventory	Superficial gas velocity (U_g)
Case 1	Reference design (100%-0%)	282 kg	1.93 m·s^{-1}
Case 2	FBD-282	282 kg	1.162 m·s^{-1}
Case 3	FBD-340	340 kg	1.162 m·s^{-1}
Case 4	SbS (90% - 10%)	282 kg	1.93 m·s^{-1}
Case 5	SZD (0-100%)	282 kg	1.93 m·s^{-1}

RESULTS

Fig. 2 depicts the numerical results of the four proposed technical concepts tested, as regards the time-averaged static pressure profile, in comparison with the corresponding numerical results of the reference design case.

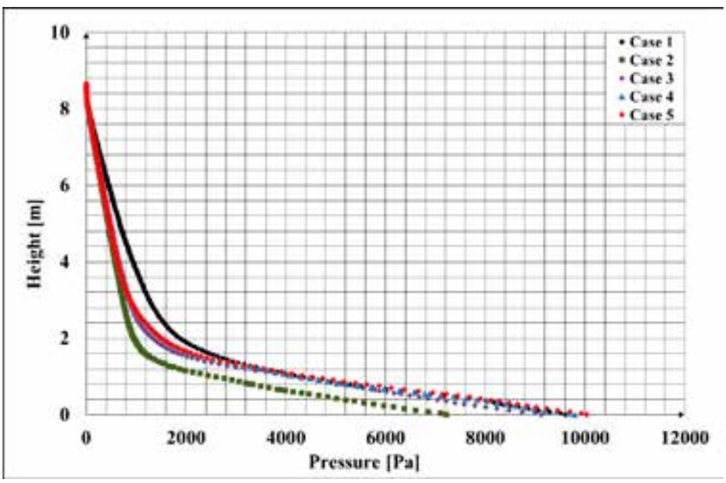

Fig. 2. Time-averaged mean static pressure along the riser axis for a time averaging equal to *t*=15 sec.

It is evident that near the dense bottom zone the predicted pressure in the FBD-282 test case is lower compared to one calculated for the reference design. In the second fat bottom design test case, i.e. FBD-340 kg, the static pressure profile is virtually the same as in the reference design. Additionally, concerning the SbS and SZD test cases compared to the reference design, it can be seen that all three static pressure-height curves almost coincide for a height range [0, 1] m. Above that height, a higher pressure drop is observed in the reference design than in the SbS and SZD test cases indicating the presence of higher amounts of solid particles in the freeboard.

This fact is also evident in Fig. 3, where the contours of the instantaneous solids volume fraction at a plane X=0 for all the compared cases is presented. As can be seen, in the reference design higher values of the solid volume fraction can be observed in the freeboard, when compared with the other cases. This is the reason why in the reference design case the mean static pressure drop is slightly higher compared to the other cases. Another important conclusion to be drawn is that the core-annulus flow pattern exhibited in CFB risers, is less intense in the FBD concept –especially in the FBD-282 case- than in the other design concepts. In this case, low values of the solid volume fraction are observed for heights greater than 2 meters.

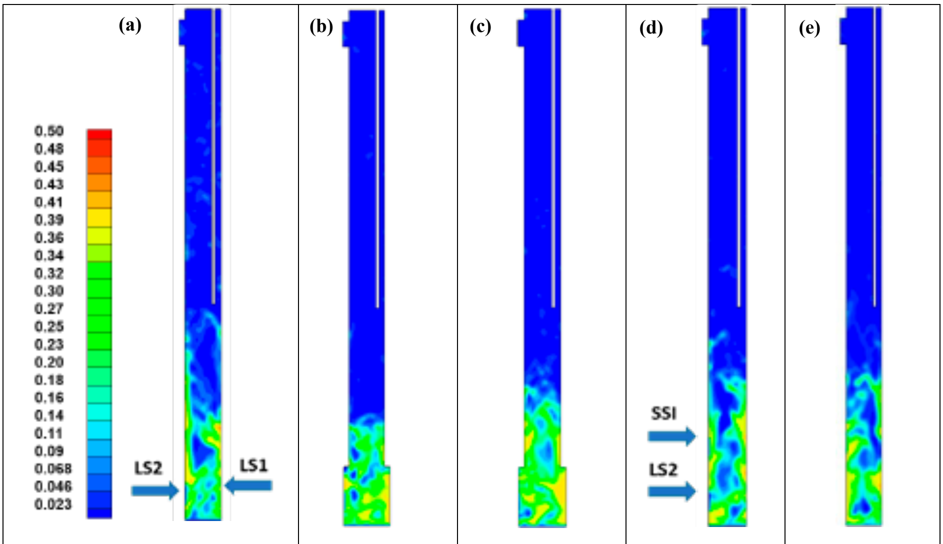

Fig. 3. Instantaneous solids volume fraction at X=0 (the medium X of the reactor) for a) the Reference design b) the FBD-282 kg, c) the FBD-340 kg, d) SbS and e) SZD.

In Fig. 4 the contours of the time-averaged mean solids volume fraction at a plane X=0 for the cases examined are presented. Generally, in the fat bottom design the solid volume fraction near the bottom zone is higher than the one observed in the reference design. Additionally, in the FBD-282 kg case, the flow tends to be homogeneous, for heights greater than 0.8 meters, resulting in a better gas-solid mixing. On the other hand in the FBD-340 kg test case, higher heterogeneity is observed, even if this case should be compared with the reference design, owing to the higher solids inventory. However, in the latter case a quite high CO_2 capture is achieved; the highest among all cases examined. This is attributed to the fact that high back flow mixing of the solid phase occur in the fat bottom bed, keeping thus more sorbent mass in this area. In the SbS and SZD concepts the gas-solid flow pattern is quite similar as in the reference design case.

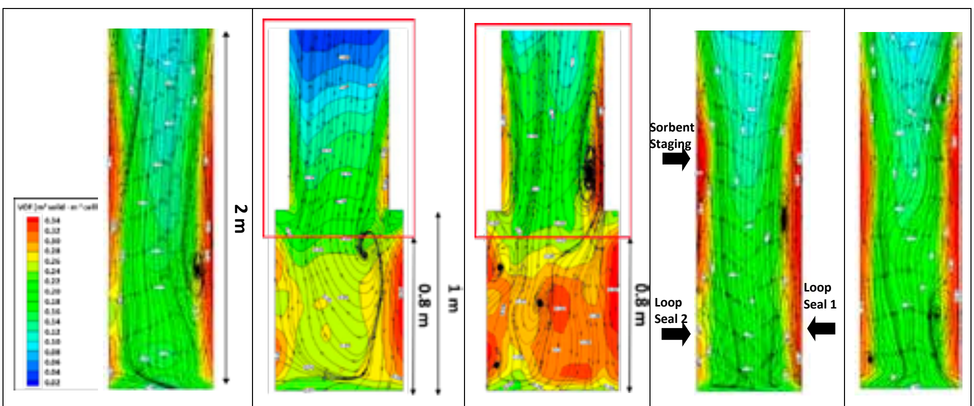

Fig. 4. Time-averaged mean solids volume fraction at X=0 for the a) Reference design, b) FBD-282 kg, c) FBD-340 kg, d) SbS and e) SZD concept.

Fig. 5 depicts the time-averaged spatial distribution of mass weighted CO_2 mean mass fraction at a plane X=0 for the cases tested. It is evident that in the reference design, SbS and SZD most of the CO_2 capture occurs near the center axis of the reactor and continues up to a height of approximately 1-2 meters. On the contrary, in the FBD, in both cases tested, most of the CO_2 capture occurs at the fat bottom area of the riser.

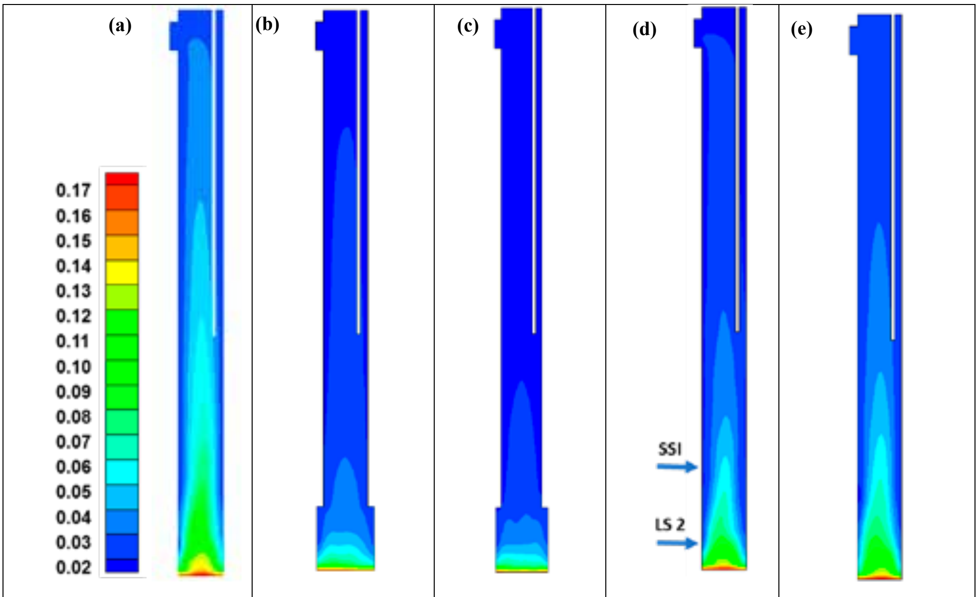

Fig. 5. Time-averaged mass weighted CO_2 mean mass fraction at plane X=0 for a) the Reference design b) the FBD-282 kg, c) the FBD-340 kg, d) SbS and e) SZD.

Finally, the numerical results indicate that a better CO_2 capture efficiency is achieved in all retrofit cases compared to the reference design. More specifically, the capture efficiency is equal to 90.36% and 91.91%, in the FBD-282 and FBD-340 concepts, respectively, as can be seen in Table.2; such values are close enough to the CO_2 equilibrium capture efficiency (93.33%). In the SbS design concept a CO_2 capture efficiency equal to 89.64% can be achieved, whilst the efficiency is lower (88.70%) when 100% of the sorbent phase is injected from the sorbent staging inlet. This confirms the advantages of staging the sorbent injection, since injecting all of the regenerated material to the bottom or to the splash zone leads to lower capture efficiency.

Table.2: Comparison of the capture efficiency achieved in the reference design and FBD concepts.

Case Studied	**Capture Efficiency**
Case 1	**87.03 %**
Case 2	**90.36 %**
Case 3	**91.91** %
Case 4	**89.64 %**
Case 5	**88.70 %**

Such fact is proven in **Błąd! Nie można odnaleźć źródła odwołania.**, as well, where numerical results of the CO_2 mean mass fraction at different riser heights of all the retrofit cases are presented. More specifically, it can be seen that higher capture efficiency can be achieved when part of the regenerated sorbent is split and introduced to the reactor at different heights than being introduced completely from the same height. A simple interpretation to such fact is that when the regenerated sorbent is introduced from the same section, the solids volume fraction and subsequently the flow heterogeneity increases locally, a fact that hinders the effective gas-solid mixing.

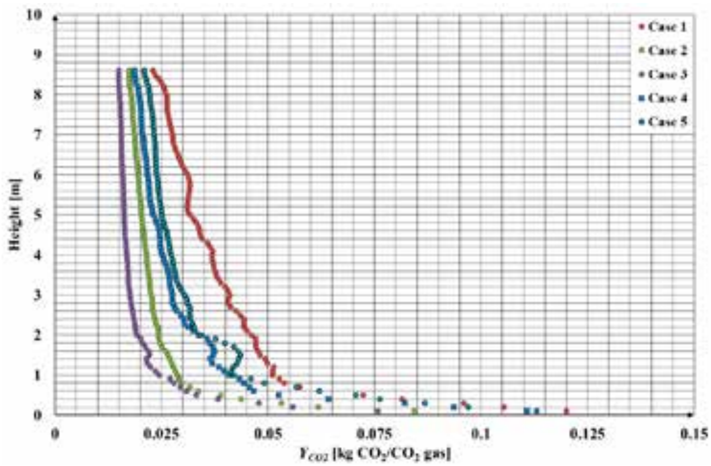

Fig. 6. Time-averaged mean Y_{CO2} along riser center line for the a) Reference design, b) FBD-282 kg, c) FBD-340 kg, d) SbS and e) SZD.

The advantages of the four retrofitting concepts over the reference design in terms of CO_2 capture efficiency are reflected, as well, by the values of the effective material to CO_2 ratio. The contours of this molar ratio at different horizontal slices near the bottom bed area are depicted in Fig. 7. In all retrofitting cases, better mixing is achieved and the regions of the reactor with low ratios are minimized. In such areas although there is CO_2 in the gaseous phase, the amount of the active CaO is not that high.

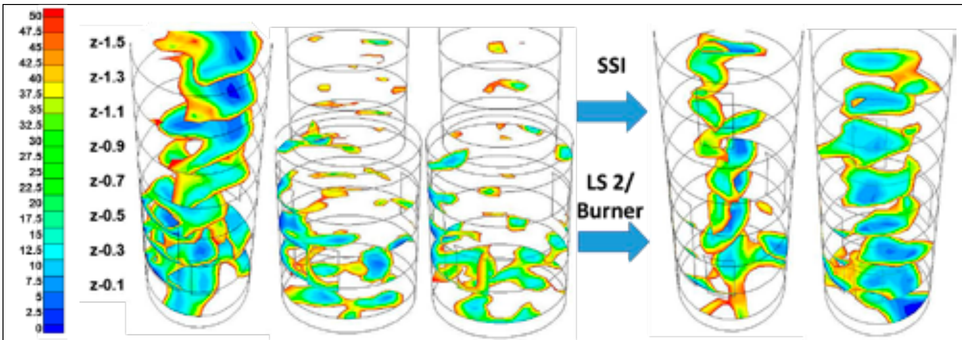

Fig. 7. Effective material to CO_2 ratio (kmol CaO (active) / kmol CO_2) for the a) Reference design, b) FBD-282 kg, c) FBD-340 kg, d) SbS and e) SZD. (Values greater than 50 have been cut-off)

It should be underlined that in terms of CO_2 capture efficiency the FBD concept seems the most promising among all, however the SbS design concept is the most preferable solution, because apart from the high capture efficiency, it can be more easily modified to the initial design concept by simply opening or closing the sorbent staging inlet.

CONCLUSIONS

The effect of the carbonator riser geometry on the gas-solid flow hydrodynamics and CO_2 capture efficiency is an important issue that is investigated in the present work. For this purpose, a validated 3D Eulerian CFD model is implemented for different design/operation concepts. All the test cases simulated are retrofitting concepts of a reference design. This design corresponds to the TU Darmstadt (TUD) carbonator, which is part of the 1 MW_{th} Dual Fluidized Bed (DFB) installation, and has been used in a previous research activity to validate the CFD model. The retrofitting concepts are a fat bottom design (FBD) with two different setups of the solids inventory, i.e. 282 kg and 340 kg, a sorbent staging design (SbS) and a sorbent in splash zone design (SZD). The drag force model implemented in all cases is the sub-grid EMMS scheme that takes into account the clustering formation inside such units. The numerical results indicate that a better CO_2 capture efficiency is achieved in all retrofit cases compared to the reference design. More specifically, the highest capture is achieved in the FBD concept for both solid inventories used; in the FBD-282 kg and FBD-340 kg cases the capture efficiency is equal to 90.36 % and 91.91 %, respectively, close enough to the CO_2 equilibrium capture efficiency (93.33 %). In the SbS and SZD cases the capture efficiencies are 89.94 % and 87.03 %, higher than the reference design, mainly because a better mixing is achieved in the lower part of the reactor, i.e. bottom and splash zone. As concerns the gas-solid flow patterns, in the FBD-282 kg case the flow tends to be homogeneous for heights greater than 0.8 m (the fat bottom extends up to a height equal to 1 meter). In the SbS and SZD cases the gas-solid flow pattern is quite similar to the reference design. Even if the FBD case seems the most promising among all, in terms of CO_2 capture efficiency, the SbS design is the most attractive solution, because apart from the high capture efficiency, it can be more easily incorporated into retrofit units. Among, the improvements investigated through the aforementioned numerical tools the sorbent staging idea could be implemented with not many technical complications in larger scale units to boost the efficiency of the carbonator.

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