Multiscale hydrodynamic simulations of chemical looping combustion using a CuO/Al_2O_3 oxygen carrier

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Abstract:

Chemical looping combustion (CLC) is considered one of the most promising pathways for achieving low-emission energy conversion with inherent CO_2 separation. The hydrodynamics of gas–solid flow in the fuel reactor plays a crucial role in the stable operation of CLC systems and in the effective utilization of oxygen carrier particles. This study focuses on the hydrodynamic behaviour of a multifunctional CuO/Al_2O_3 composite oxygen carrier within an innovative fuel-reactor concept equipped with an integrated inertial solids separator. The reactor configuration is designed to promote stable fluidization, controlled solids circulation and reduced losses of oxygen carrier particles.

The work is conducted within the 3rd Polish-Chinese Joint Research Programme "Multi-scale investigation of chemical looping combustion of biomass pellets towards negative CO_2 emission" (MsLimitCO2). A combined numerical methodology is applied. Comprehensive Simulator of Fluidized and Moving Bed equipment (CeSFaMB) is used to analyse the global gas–solid hydrodynamics of the CLC process, while SolidWorks is employed to perform dedicated simulations of the internal separator within the fuel reactor. This integrated, multi-scale approach enables a comprehensive assessment of flow structure, solids distribution and the influence of key geometric features on hydrodynamic performance.

The hydrodynamic insights obtained from the numerical simulations will support further development and experimental validation of the proposed reactor configuration. Ultimately, this study contributes to advancing robust fluidized-bed designs for chemical looping applications and to improving the reliability of future low-carbon energy systems.

Keywords:

Chemical Looping Combustion; CuO/Al_2O_3 Oxygen Carrier; Fluidized Bed; Hydrodynamics; Inertial Solids Separator; Multiscale Simulations.

1. Introduction

Growing concerns about climate change and the need to reduce anthropogenic CO_2 emissions have driven intensive research into advanced energy conversion technologies with integrated carbon capture [1]. Among these, chemical looping combustion (CLC) has emerged as a promising option, as it enables inherent CO_2 separation during combustion and eliminates the need for energy-intensive gas separation processes [2].

For industrial deployment, CLC requires reactor configurations that ensure stable solids circulation and high CO_2 purity at the fuel reactor outlet. In CLC systems, oxygen is transferred from air to fuel using a circulating solid oxygen carrier, which prevents direct contact between air and fuel gases and enables inherent separation of combustion products [3,4]. This concept has been validated in pilot installations ranging from laboratory to megawatt scale [5,6]. However, reactor performance remains strongly dependent on multiphase hydrodynamics rather than solely on intrinsic redox kinetics [7].

Experience from dual circulating fluidized bed (DCFB) units shows that overall fuel conversion and CO_2 purity at the fuel reactor outlet are governed by the solids circulation rate, gas–solid contact quality, residence time distribution, and inter-reactor mass transfer [3,8,9]. Even when highly reactive oxygen carriers are employed, non-uniform solids distribution, carbon slip, and oxygen carrier entrainment may significantly reduce system performance and long-term operational stability [10,11]. These limitations are inherently hydrodynamic and cannot be resolved by global mass and energy balance considerations alone [12].
The complexity further increases when solid fuels are used. The coexistence of fuel particles, ash, and oxygen carrier material leads to intensive solids mixing, abrasion, and potential loss of active material. In circulating systems, the entrainment rate from the fuel reactor determines the loading of downstream separation units and directly affects the stability of the solids inventory. Excessive entrainment may result in oxygen carrier losses and circulation instability, whereas insufficient transport can limit oxygen availability within the fuel reactor. Therefore, a consistent analysis of the hydrodynamic regime and separation performance is essential [9].
Copper-based oxygen carriers are frequently investigated due to their high reactivity and ability to operate in chemical looping with oxygen uncoupling (CLOU) mode [13–15]. CuO-based materials supported on Al_2O_3 exhibit favorable redox characteristics within the typical CLC temperature range [16]. However, even for highly reactive carriers, the effective oxygen transport capacity at reactor scale depends strongly on superficial gas velocity, solids mixing intensity, and circulation stability. Variations in gas flow may induce transitions between bubbling and fast fluidization regimes, which significantly modify axial concentration profiles and particle entrainment rates [17].
Advanced numerical tools are increasingly applied to investigate gas–solid behavior in CLC systems. Process-scale simulators and CFD-based approaches enable the prediction of concentration fields, temperature distributions, and solids fluxes under various operating conditions [18]. These methods support the interpretation of pilot-scale experiments and contribute to scale-up strategies [19]. Nevertheless, most studies focus either on local flow structures within a single reactor or on global reactor performance, without consistently linking fuel reactor hydrodynamics to downstream solids separation behavior [20,21].
In circulating systems, the interaction between entrainment and separator efficiency is critical. The solids flux leaving the fuel reactor determines the performance of cyclones or inertial separators and directly influences oxygen carrier retention. Reliable prediction of CO_2 capture efficiency therefore requires linking operating conditions, reactor hydrodynamics, and solids circulation. A multiscale perspective is thus needed. At the reactor scale, axial concentration and temperature profiles must be resolved to quantify fuel conversion and redox activity, while at the system scale, solids transport and separation efficiency must be evaluated to ensure stable circulation and long-term operability [9,22].
This study employs multiscale hydrodynamic simulations of a dual fluidized-bed CLC system with a CuO/Al_2O_3 oxygen carrier. The analysis examines how gas flow rate below the distributor affects superficial gas velocity in the fuel reactor and the resulting entrained solids flux. Simulations performed in the CeSFaMB environment quantify the mass flow of solids under varying operating conditions. The predicted entrained solids flux is then used as an input for separator simulations in SolidWorks. This approach enables evaluation of particle trajectories, recirculation efficiency, and potential oxygen carrier losses, including the fraction of solids returned by the separator and the fraction escaping the system. It also provides a consistent multiscale framework for evaluating circulation stability under varying gas velocity conditions, with particular attention to the relationship between distributor gas flow, superficial velocity, hydrodynamic regime, entrained solids flux, and separator efficiency.

2. Methodology

The present study extends previous work on a cold-flow chemical looping combustion (CLC) unit by introducing a multiscale modelling approach. In the earlier study, the reactor was analysed under cold-flow conditions using reactor-scale simulations supported by local analysis of the separator. The focus was on the influence of separator geometry, in particular the length and inclination angles of internal baffles, on particle recirculation [23].
In the present work, initial simulations are performed for hot operating conditions, moving towards a more realistic representation of the process. The methodology combines reactor-scale modelling with detailed CFD analysis of the separator.
The reactor-scale model, implemented in CeSFaMB, is used to describe the overall hydrodynamics of the system, including solids circulation and pressure distribution. The local CFD model, developed in SolidWorks, is applied to analyse flow structures in the separator, with particular attention to velocity fields and recirculation zones resulting from the internal geometry. CeSFaMB is a 1.5D modelling tool dedicated to bubbling and circulating fluidized bed systems and has previously been applied and validated for chemical looping combustion simulations and oxygen carrier transport analyses [9,24]. Due to the simplified reactor-scale character of the CeSFaMB model, detailed local flow phenomena inside the separator cannot be directly reproduced. Therefore, additional CFD simulations were performed in SolidWorks Flow Simulation to analyse

particle trajectories, local recirculation behaviour, and gas flow structures in the upper section of the fuel reactor.
The models are linked by transferring boundary conditions from the reactor-scale simulations to the CFD model. This ensures consistency between the two approaches and allows the influence of local flow behaviour on the overall reactor performance to be assessed.

2.1. Reactor configuration and system description

The analysed system is based on a dual fluidized bed chemical looping combustion unit for solid fuels (FB-CLC-SF), consisting of an air reactor and a fuel reactor connected by a circulating solids loop. A schematic of the hot FB-CLC-SF system is shown in Fig. 1 [9].

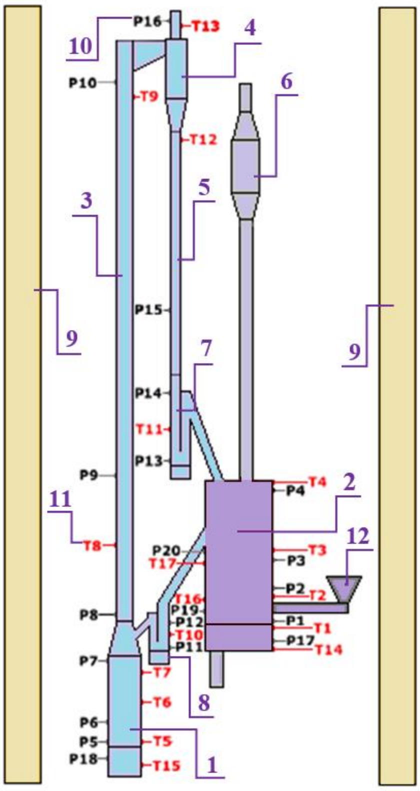

Figure 1. *Schematic of the hot FB-CLC-SF system.*

The air reactor (1) is the oxidation zone of the oxygen carrier. The oxidized solids are transported upward through the riser pipe (3) to the cyclone (4), where gas–solid separation takes place. The separated solids are then directed through the downcomer (5) to the upper loop seal (7) and subsequently transferred to the fuel reactor (2). In the fuel reactor, the oxygen carrier releases oxygen. Solids leaving the fuel reactor are captured in the particle collector (6). After oxygen release, the oxygen carrier is directed to the lower loop seal (8) and then returned to the air reactor, closing the circulation loop. Both reactors are externally heated using electric heaters (9), allowing operation under hot-flow conditions. The installation is equipped with pressure sensors P1–P20 (10) and temperature sensors T1–T16 (11), enabling monitoring of the hydrodynamic and thermal behaviour of the system. Fuel is supplied to the fuel reactor through the fuel feeder (12).
The analysed configuration follows previous studies [9,24]. In the present work, the same reactor concept is used as a basis for simulations under hot operating conditions. The main modification concerns the fuel reactor, where a separator has been introduced in the upper section. This modification is shown in Fig. 2a.
In the upper part of the reactor, within the separator, the lengths and inclination angles of the internal baffles are indicated in red. The lengths of the two individual baffles were set to 38 mm, while the angles of the inclined elements were defined as 60° and 55°, as shown in Fig. 2b. These parameters were determined in previous studies conducted for the cold-flow model, where they were identified as the most effective configuration. Therefore, the same values were adopted in the present study.

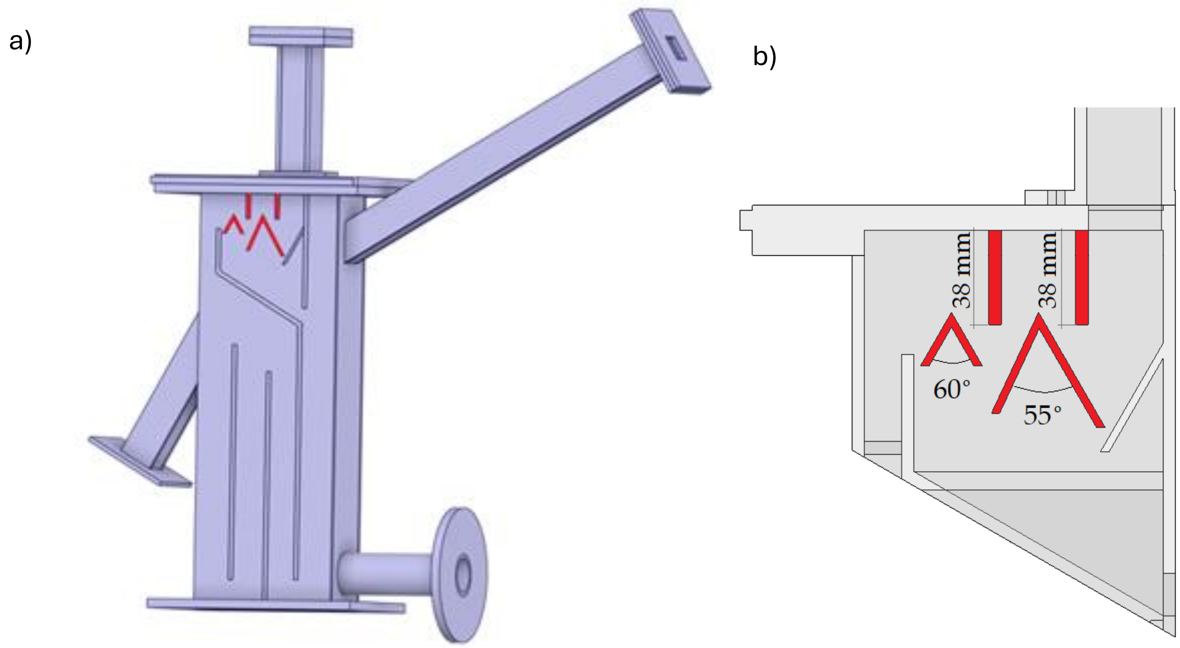

Figure 2. *Fuel reactor with an integrated separator: a) overall geometry, b) definition of baffle lengths and inclination angles.*

2.2. Multiscale modelling framework

The analysis was carried out using a multiscale modelling approach that combines reactor-scale simulations with local CFD analysis of the separator. This approach follows the modelling strategy adopted in previous studies, where global simulations were used as a basis for more detailed local analysis. The reactor-scale model, implemented in CeSFaMB, is used to describe the overall hydrodynamic behaviour of the system, including solids circulation and pressure distribution in both reactors. Due to its simplified 1.5D formulation, the model does not resolve local flow structures in the upper part of the fuel reactor. To address this limitation, a separate CFD model of the separator was developed in SolidWorks. This model enables detailed analysis of velocity fields and flow structures in the region where the internal geometry exerts the strongest influence on the flow.

The modelling procedure is sequential. First, simulations are performed at the reactor scale using CeSFaMB. The obtained results are then used to define the boundary conditions for the CFD model of the separator. The methodology follows the framework developed in the authors' previous cold-flow studies [23], extended here to hot operating conditions. This ensures consistency between the global and local descriptions of the system and enables assessment of the impact of local flow structures on the overall reactor behaviour.

2.3. Reactor-scale modelling using CeSFaMB

Reactor-scale simulations were performed using the CeSFaMB simulator, which is dedicated to modelling circulating and bubbling fluidized bed systems. The model was applied to describe the overall hydrodynamic behaviour of the FB-CLC-SF unit, including pressure distribution and solids transport in both the air and fuel reactors.

CeSFaMB is based on a 1.5D formulation, enabling efficient simulation of large-scale fluidized systems at relatively low computational cost. The model has previously been validated for CLC applications and provides reliable predictions of key hydrodynamic parameters, including pressure distribution, solids circulation, and entrainment flux [9,24].

In the present study, the simulations were carried out using the properties of a CuO/Al_2O_3 oxygen carrier intended for application in the CLC process. The material consisted of 60–70 wt.% Al_2O_3 and 30–40 wt.% CuO. The particle sphericity was assumed as 0.86, corresponding to round sand. The solids density was 3860 kg/m³, while the bulk density was 2020 kg/m³. The Sauter mean diameter was equal to 632 μm, with a particle size range of 400–1000 μm. A representative view of the oxygen carrier particles is shown in Fig. 3.

The selected properties reflect realistic operating conditions and allow for representation of the system under hot-flow conditions. The model was used to determine key parameters required for further analysis, including pressure distribution, gas flow characteristics, and entrained solids flux in the upper part of the fuel reactor. The results obtained from the reactor-scale simulations were subsequently used as boundary conditions and input data for the local CFD model of the separator.

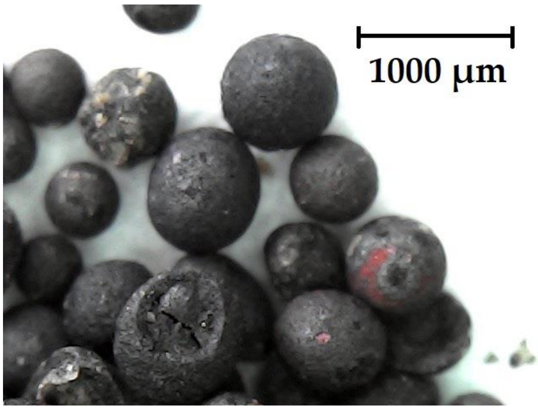

Figure 3. *CuO/Al_2O_3 oxygen carrier particles used in the study.*

2.4. Local CFD modelling of the separator using SolidWorks

Local CFD simulations of the separator were performed using SolidWorks Flow Simulation. Due to the local and geometrically complex character of the separator region, the CFD model was used to analyse velocity fields and flow structures in the upper part of the fuel reactor, with particular focus on recirculation zones and the influence of the internal geometry.

The geometry of the separator was consistent with the configuration shown in Fig. 2, including the defined lengths and inclination angles of the internal baffles. Boundary conditions for the CFD model were based on the results obtained from the reactor-scale simulations. In particular, gas flow parameters and entrained solids flux at the outlet of the fuel reactor were used as input data, ensuring consistency between the two modelling approaches.

The analysis focused on the identification of flow structures, velocity distribution and recirculation regions, which are critical for the performance of the separator and for limiting the escape of oxygen carrier particles from the system.

3. Results and discussion

The results of the multiscale simulations are presented and discussed in this section. The analysis focuses on the effect of gas mass flow rate on the hydrodynamic behaviour of the fuel reactor and the resulting particle entrainment.

First, the reactor-scale results obtained using the CeSFaMB model are analysed, including superficial gas velocity, bed structure and entrained solids flux. These parameters define the operating conditions and determine the separator's loading. Next, the results of CFD simulations performed in SolidWorks are discussed, with emphasis on particle trajectories, recirculation and the ability of the integrated separator to limit oxygen carrier losses.

This combined analysis makes it possible to link reactor hydrodynamics with separator performance and to identify operating conditions that ensure stable solids circulation.

3.1. Reactor hydrodynamics and particle entrainment

The hydrodynamic behaviour of the fuel reactor was analysed to assess how gas flow conditions influence particle transport towards the separator. The analysis focuses on the relationship between gas mass flow rate, flow structure in the reactor and particle entrainment. Reactor-scale simulations in CeSFaMB were performed for a range of gas mass flow rates from 0.001 to 0.008 kg/s. However, particles started to reach the upper part of the reactor and enter the separator only for gas flow rates above 0.005 kg/s, indicating the onset of significant entrainment. Below this value, particle transport to the separator was negligible. For this reason, the analysis of the separator in SolidWorks was limited to the range of 0.005–0.008 kg/s. Within this range, a further increase in gas flow rate results in a higher amount of solids reaching the separator and increased loading of this region. This behaviour directly affects the stability of solids circulation in the system.

As shown in Fig. 4, the superficial gas velocity in the fuel reactor increases with gas mass ($\dot{m}_g$) flow rate, both in the lower ($V_{FR(L)}$) and upper ($V_{FR(U)}$) sections of the reactor. The relationship is close to linear in the analysed range. A clear difference can be observed between the lower and upper sections of the reactor, with significantly higher velocities in the upper region.

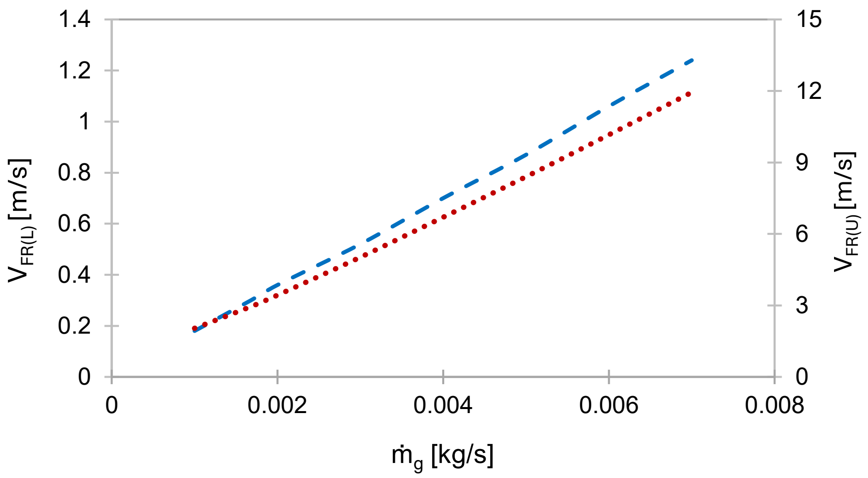

Figure 4. *Superficial gas velocity in the fuel reactor as a function of gas mass flow rate.*

The dependence of superficial gas velocity on gas mass flow rate can be approximated using the following linear correlations:

$$V_{FR(L)} = 175.81\dot{m}_g + 0.0015, \qquad (1)$$

$$V_{FR(U)} = 1662.6\dot{m}_g + 0.1619, \qquad (2)$$

The higher slope of the correlation for the upper part of the reactor indicates a stronger sensitivity to changes in gas flow rate. This is related to the lower solids concentration and more dilute flow conditions in this region, which allow for a faster increase in gas velocity.

As shown in Fig. 5, the void fraction in the reactor changes with gas mass flow rate, reflecting modifications in the bed structure. An increase in gas flow rate leads to a higher void fraction in the dense bed region (ε_B), while the opposite trend is observed in the freeboard (ε_F).

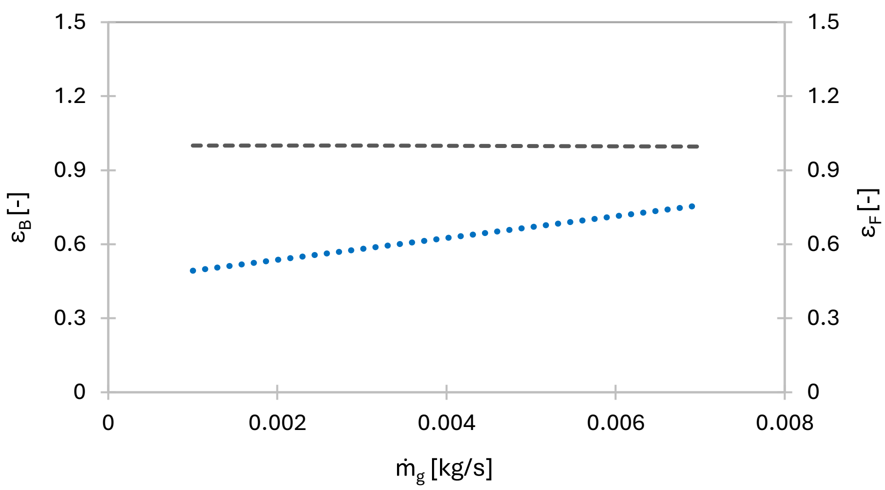

Figure 5. *Void fraction in the bed and freeboard as a function of gas mass flow rate.*

The dependence of void fraction on gas mass flow rate can be approximated using the following linear correlations:

$$\varepsilon_B = 44.164\dot{m}_g + 0.4487, \qquad (3)$$

$$\varepsilon_F = -0.7001\dot{m}_g + 1.0013, \qquad (4)$$

The increase in void fraction in the bed indicates bed expansion and a reduction in solids concentration as the gas flow rate increases. At the same time, the decrease in void fraction in the freeboard suggests a higher presence of solids in this region, which is consistent with intensified particle transport towards the upper part of the reactor.

The mass flux of entrained particles ($\dot{m}_p$) in the upper part of the fuel reactor increases with gas mass flow rate, as presented in Fig. 6. The relationship is nonlinear, with a more pronounced increase observed at higher values of gas flow rate, indicating intensified particle transport towards the separator.

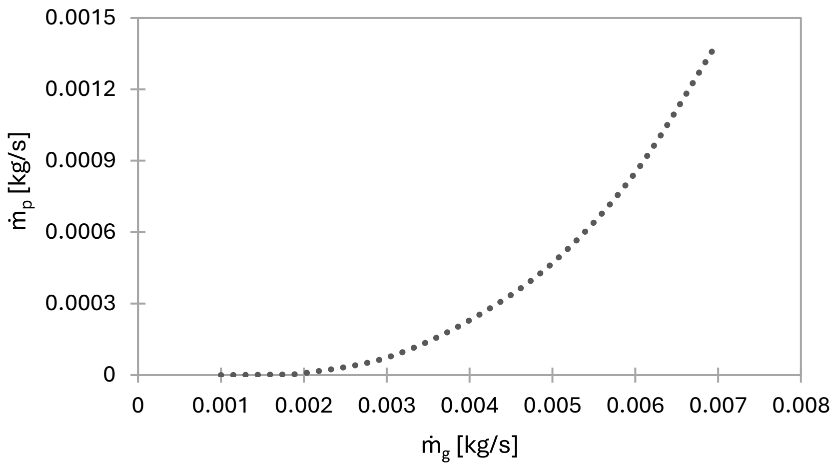

Figure 6. *Entrained solids flux in the upper part of the fuel reactor as a function of gas mass flow rate.*

The dependence of entrained solids flux on gas mass flow rate can be approximated using the following correlation:

$$\dot{m}_p = 53.188m_g^2 - 0.2022m_g + 0.0002, \qquad (5)$$

The nonlinear character of this relationship suggests that particle entrainment becomes significantly more intensive beyond a certain gas flow rate. This behaviour is consistent with the transition from denser to more dilute flow conditions in the upper part of the reactor, which promotes particle transport into the separator region.

A similar trend is observed for the number of particles (N) entering the separator, as presented in Fig. 7.

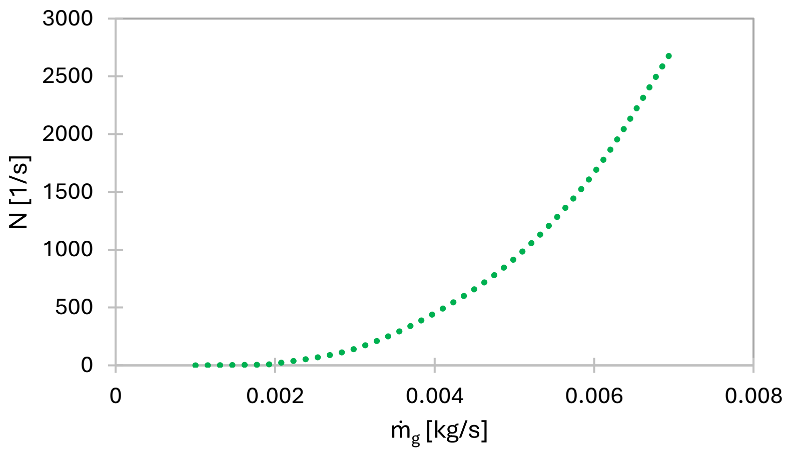

Figure 7. *Number of particles entering the separator as a function of gas mass flow rate.*

As shown in Fig. 7, with increasing gas mass flow rate, the number of transported particles increases, indicating a more intensive transfer of solids from the reactor to the separator region.

The relationship between the number of particles and gas mass flow rate is described by a quadratic function (Eq. 6):

$$N = 1 \cdot 10^8 m_g^2 - 396442 m_g + 352.35, \qquad (6)$$

The nonlinear increase in particle number confirms that even a relatively small change in gas flow rate can lead to a significant increase in separator loading. This effect is particularly important from the operational point of view, as it directly influences particle recirculation and potential losses of oxygen carrier material.

3.2. Separator performance and particle recirculation

To better understand the results, it is useful to consider the separator geometry and the inlet location, as shown in Fig. 8. The separator inlet is relatively small compared to the freeboard volume, which limits direct particle access to this region.
As a result, although particle entrainment occurs already at lower gas flow rates, particles do not enter the separator until a certain threshold is reached. In the present case, this threshold corresponds to a gas mass flow rate of approximately 0.005 kg/s. Below this value, particles remain in the freeboard and do not reach the separator inlet.
This behaviour explains the difference between the CeSFaMB and SolidWorks simulations. While reactor-scale simulations capture particle entrainment in the freeboard over a wide range of operating conditions, CFD analysis of the separator becomes meaningful only when particles effectively reach the inlet. For this reason, the SolidWorks simulations were performed for gas mass flow rates in the range of 0.005–0.008 kg/s.

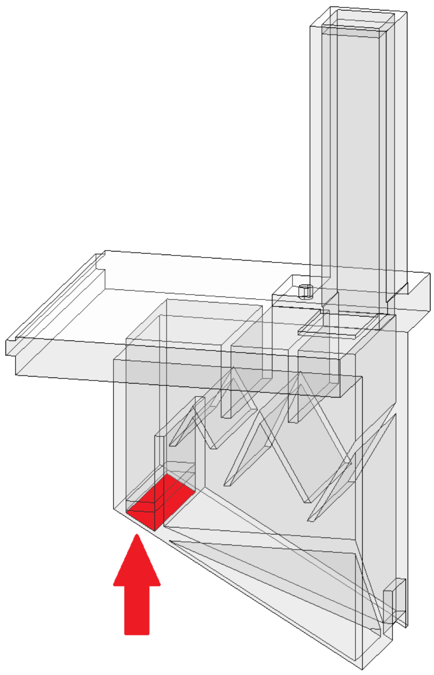

Figure 8. *Geometry of the separator and location of the inlet in the upper part of the fuel reactor.*

The CFD simulations were performed to analyse particle behaviour within the separator under different operating conditions. Particular attention was given to particle trajectories and their interaction with the internal geometry.

The results allow for the assessment of how particles entering the separator are redistributed within this region and whether they are effectively redirected back towards the reactor. This is essential for evaluating the separator's ability to retain the oxygen carrier and limit particle losses.
The particle distribution obtained for the analysed gas mass flow rates is presented in Fig. 9.

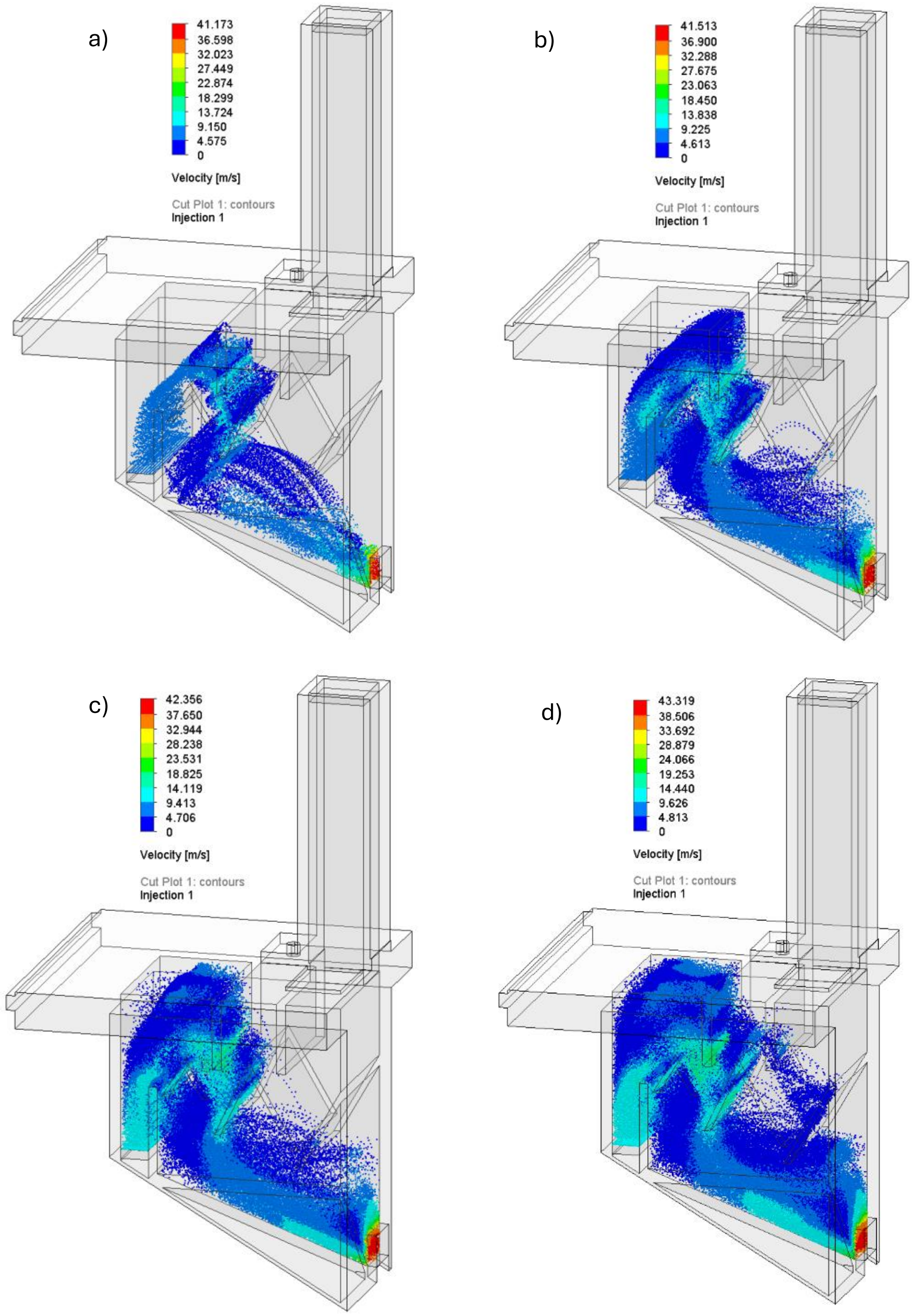

Figure 9*. Particle distribution in the separator for different gas mass flow rates: a) 0.005 kg/s, b) 0.006 kg/s, c) 0.007 kg/s, d) 0.008 kg/s.*

The results show that particles entering the separator are effectively redirected towards the reactor for all analysed cases. The observed particle distribution indicates the formation of recirculation zones within the separator, which promote particle return to the lower part of the reactor. This behaviour is closely related to the internal geometry of the separator. Particles entering the separator interact with the inclined baffles, where their trajectories are deflected and redirected downward. As a result of these interactions, particles lose part of their upward momentum and are prevented from further transport towards the outlet.

The flow structure inside the separator is characterized by regions of downward and recirculating motion, which enhance particle retention. The presence of the baffles promotes repeated interactions between particles and the separator walls, increasing the probability of their return to the reactor. This mechanism is particularly important under conditions of increased particle loading.

With increasing gas mass flow rate, particle motion becomes more dynamic and the number of particles entering the separator increases, which is consistent with the trends observed in the reactor-scale simulations.

Despite the higher particle loading, the separator maintains its effectiveness, and no significant particle escapes from the separator was observed. This confirms that the separator geometry is effective in maintaining solids within the system under the analysed operating conditions.

4. Conclusions

This study presents a multiscale analysis of gas–solid hydrodynamics in a dual fluidized-bed CLC system with a CuO/Al_2O_3 oxygen carrier, combining reactor-scale simulations with local CFD modelling of the separator.
The results confirm that gas mass flow rate is a key parameter controlling reactor hydrodynamics, including superficial gas velocity, bed structure and particle entrainment. Increasing the gas flow rate intensifies particle transport towards the upper part of the reactor and increases the loading of the separator.
The CFD analysis shows that the applied separator geometry effectively promotes particle recirculation. Interactions with the inclined baffles modify particle trajectories and reduce their upward momentum, resulting in efficient return of particles to the reactor.
No significant particle escape from the separator was observed under the analysed operating conditions, indicating high efficiency of the proposed design in retaining the oxygen carrier.
The combined use of CeSFaMB and SolidWorks provides a consistent multiscale framework for linking reactor hydrodynamics with separator performance. The proposed approach can support the design and optimisation of CLC systems with improved solids circulation and reduced material losses.

Nomenclature

$\dot{m}_g$ gas mass flow rate [kg/s]
($V_{FR(L)}$) superficial gas velocity in the lower sections of the reactor [m/s]
($V_{FR(U)}$) superficial gas velocity in the upper sections of the reactor [m/s]
(ε_B) void fraction in the bed [-]
(ε_F) void fraction in the freeboard [-]
$\dot{m}_p$ mass flux of entrained particles [kg/s]
N Number of particles entering the separator [1/s]

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