18th Brazilian Congress of Thermal Sciences and Engineering
November 16-20, 2020 (Online)

ENC-2020-0468
CONSTRUCTAL DESIGN OF A MINERAL CARBONATION SYSTEM FOR POST-COMBUSTION CARBON CAPTURE

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Abstract. *Carbon capture (CC) has been presented as a crucial way of mitigating the effect of greenhouse gas emissions. Mineral carbonation (MC) processes are among the safest and most promising alternatives for CC due to product stability. Nevertheless there are technical challenges that must be overcome in order to scale-up the technology. Devices must be designed for energy efficiency and sufficiently fast kinetics. This paper presents the first steps in designing a MC reactor for* CO_2 *capture by way of constructal theory. This will be achieved by applying the constructal method to the process' degrees of freedom such as characteristic time of reaction, and geometric scales for the elemental volume and first construct, and studying the effect on merit figures such as rate of* CO_2 *captured and pressure losses under global restrictions such as a fixed reactor volume, using analytical and numerical models of the process. Results show good agreement between numerical and analytical results and allowed to obtain dimensioning guidelines for time of residence in the porous bed in order to obtain a desired* CO_2 *abatement, as well as the evolutionary path of geometry scales for the first construct which matches* CO_2 *conversion rate with the input of exhaust gases, as well as minimize pressure losses.*

Keywords: *carbon capture, greenhouse gases, constructal design, mineral carbonation*

1. INTRODUCTION

The concern with global warning has motivated research work regarding the control and reduction of anthropogenic emissions of greenhouse gases (CECCHI et al., 2017; LEUNG et al., 2014), especially of carbon dioxide (CO_2), which is considered the main responsible gas for the phenomenon (MAZZELLA et al., 2016; SIRIRUANG et al., 2016).

Among the options for mitigation, post-combustion carbon capture has been considered crucial due to the great amount of point emissions of CO_2 that occurs in the energy generation chain (UKWATTAGE et al., 2015). The processes of Mineral Carbonation (MC), in which carbon in gaseous form (CO_2) is converted to stable solid carbonates (UKWATTAGE et al., 2017) has shown to be an interesting solution for the disposal of CO_2 in the long term, due to the stability of the produced products (MURIITHI et al., 2013), thus being the safest way of storing CO_2 (CECCHI et al., 2017).

Despite its advantages, there are technical challenges that need to be overcome in order to scale-up the technology, such as energy efficiency and achieving sufficiently fast kinetics. Design of devices need to address these challenges to make equipments compatible with flue gas emissions flow rate. Better designs can be achieved by the constructal approach, which guides the designer to the evolutionary path of configurations that lead to increase in the flow access of the system's currents. A better reactor design is one which shows maximum mass transfer over time into a fixed reactor space with minimum energy expenditure.

This paper aims to present the first steps in designing a MC reactor for CO_2 capture by way of constructal theory. This will be achieved by applying the constructal method to the process' degrees of freedom such as characteristic time of reaction, and geometric scales for the elemental volume and first construct. This is achieved by studying their effect on merit figures such as amount of CO_2 captured averaged over time and pressure losses, under global restrictions such as fixed reactor volume, using analytical and numerical models of the process. This analysis continues from previous results of temporal scales of the process (GASPAROVIC et al., 2019) and is based on Rivera-Alvarez and Bejan (2006) and Malley-Ernewein and Lorente (2019).

2. MATERIALS AND METHODS

2.1 Mathematical modelling and process description

In order to analyze the process, a simple model that retains the main features is considered, as illustrated in Figure 1.

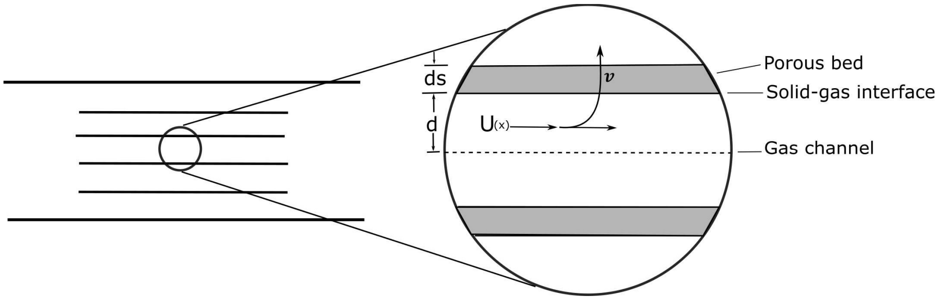


Figure 1. Model of the process

In the process, CO_2 from a gas feeding stream permeates a porous capturing media partially saturated with water, dissolves in the liquid phase and reacts with the alkali species dissolved from the substract via a series of reactions of the carbonate system. The capturing media considered is a commercial product which consists mostly of $Ca(OH)_2$ (75% w/w).

A mathematical model for the process was built based on the work of Rivera-Alvarez and Bejan (2003) as well as Ishida and Maekawa (2000). The model considers molar balances, equilibrium in the gas-liquid interface via Henry's Law, chemical equilibrium of the carbonate system, chemical kinetics for the carbonation reaction, mass transport in the porous medium and fluid flow in free and porous media (Navier-Stokes and Darcy equations, respectively). The assumptions considered in the model are presented in Table 1.

Table 1. Model assumptions

There are only two components in the gas;	Stationary state
Isothermal system;	Side reactions are not considered;
The gas flow rate is high enough that CO_2 concentration in the gas channel is constant;	There is mass transfer in the gas-liquid interface of the bed for only one of the components (CO_2);
The pore distribution in the capturing medium is random and homogeneous;	Only the calcium species react with CO_2;
	Laminar gas flow;
The equilibrium in the gas-saturated solid interface is dictated by Henry's Law;	The level of saturation in the porous media is low and its presence does not affect flow of gas through the bed.

The problem was modeled firstly in a 1D approach, in which the concentration profile along the width of the porous bed was solved in software MATLAB v.2015a, and then the 2D first construct was studied numerically (velocity, pressure and concentration fields) in the FEM software COMSOL Multiphysics® (the width scale being omitted in the analysis). In both models, stationary state is considered, based on the assumption that the operating time before substitution of substract is not enough for the transient phenomena involved (consumption of calcium species, evolution of porosity due to product precipitation) to occur significatively. Results were compared with analytical values obtained for velocity in each medium as well as pressure losses in the reactor.

2.1.1 Model of the physical-chemical equilibrium and carbonation kinetics

The chemical and kinetical model is the same for the 1D and 2D models. Carbon dioxide equilibrium in the gas-liquid interface is given by Henry's Law $[CO_{2d}] = HP_{co_{2g}}$ in which H is Henry's constant, equal to 3,3 x10^{-4} mol/m^3.Pa (SANDER, 2015). The chemical equilibrium of species in the solution was modeled according to Ishida and Maekawa (2000) and Snoeyink e Jenkins (1980), and basically refers to the equilibrium equations of the carbonate system, in which the dissolution of CO_2 in water depends on pH, due to the dissociation of carbonic acid in ($H_2CO_3{}^*$) in carbonate (CO_3^{-2}) and bicarbonate (HCO_3^-) ions (PAN et al., 2012). Thus, the concentration of each species is given by chemical equilibrium, so that the dominant species vary according to the pH of the solution according to the equilibrium equations. The pH considered was that of equilibrium $Ca(OH)_2$, equal to 12.37.

The carbonation kinetics in aqueous route was also modeled based on Ishida and Maekawa (2000). The authors proposed that the formation of calcium carbonate should be a first order reaction with respect to Ca^{2+} and CO_3^{2-} concentrations:

$$CO_3^{2-} + Ca^{2+} \rightarrow CaCO_3 \tag{1}$$

$$r_i = \frac{d}{dt}[CaCO_3] = k[Ca^{2+}][CO_3^{2-}] \tag{2}$$

In which [.] are the species concentrations (mol.m^{-3}) and k is the kinetic constant of the reaction, equal to 2,08 L.mol^{-1}.s^{-1}. We consider that since the capturing substract is in a condition of excess, that the concentration of calcium species in the pore water is constant and approximated to the solubility limit of Ca^{2+} in water.

2.1.2 Molar balance of CO_2 in the 1D Model

In the 1D model we consider the concentration profile of CO_2 along the width dimension of the porous bed, as illustrated in Figure.

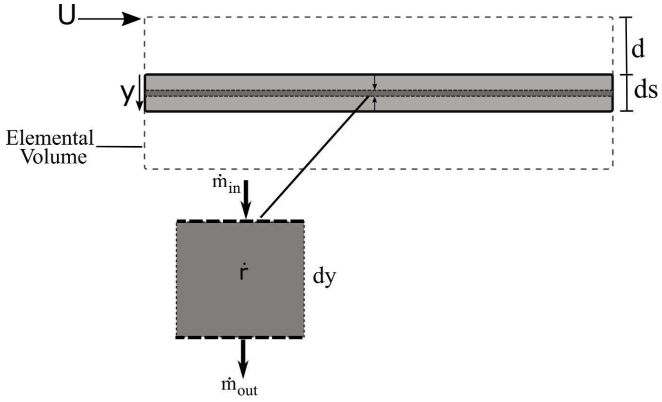


Figure 2. 1D Model of the process

The molar balance of carbon dioxide in the width dimension of porous bed considers advection and reaction terms. We consider, based on simulations for the first construct, that the pressure curves in the bed occur in the direction of Darcy flow, and so, that diffusion of CO_2 in the direction of flow is negligible in comparison to convection, as well as that Darcy velocity in the x direction is negligible. The model is presented in Eq. (3):

$$v\frac{d[CO_2]}{dy} + \dot{r} = 0 \tag{3}$$

In which v is the Darcy velocity, $[CO_2]$ is the concentration (mol.m^{-3}) in each differential volume, and $\dot{r}$ is the reaction rate, given by expression (2).

2.1.3 Molar balance of CO_2 in the 2D Model

The 2D model for the first construct includes the diffusive transport of carbon dioxide, and considers Fick's second law. The equations for transport in the porous media are presented in Eq. (4) and (5), respectively:

$$\frac{\partial[CO_2]}{\partial t} = \nabla\cdot(\boldsymbol{v}[CO_2]) + \nabla\cdot(\mathrm{D}\nabla[CO_2]) + R \tag{4}$$

$$\frac{\partial[CO_2]}{\partial t} = \frac{D_m}{\varepsilon}\nabla^2[CO_2] \tag{5}$$

In which Darcy $\boldsymbol{v}$ is velocity in the porous media, $[CO_2]$ the CO_2 concentration in gas, D and D_m are the carbon dioxide diffusivity coeficients in aqueous and porous media, in which D is equal to 1,97×10-9 (m^2/s) (FRANK; KUIPERS; VAN SWAAIJ, 1996) and D_m is given by (LI et al., 2017):

$$D_m = \frac{\varepsilon}{\tau} D_f \tag{6}$$

In which D_f is the diffusive coefficient in the fluid and τ the tortuosity, given by $\varepsilon^{1/3}$ according to (MILLIGNTON; QUIRK, 1959).

Fluid flow in porous media was modeled with Darcy equation, which for isotropic media can be written (NIELD; BEJAN, 2017):

$$\boldsymbol{v} = \frac{-K}{\mu} \nabla P \tag{7}$$

In which $\boldsymbol{v}$ is the Darcy velocity, K the permeability, μ the dynamic viscosity, and ∇P the pressure gradient in the flow direction.

For free media such as gas channel, fluid flow is described by Navier-Stokes equations, considering incompressible and constant viscosity flow (NIELD; BEJAN, 2017):

$$\rho \frac{d\boldsymbol{U}}{dt} + \rho(\boldsymbol{U} \cdot \nabla)\boldsymbol{U} = -\nabla p + \mu \nabla^2 \boldsymbol{U} \tag{8}$$

In which $\boldsymbol{U}$ is the fluid velocity in the gas channel, and ρ is fluid density.

2.2 Constructal design of the process

The geometry of the device was studied under the constructal method, firstly by identifying the smallest finite-size unit that is able to carry out the process of CO_2 capture through carbonation with fly ash. This “elemental volume” is illustrated in Fig. 2.

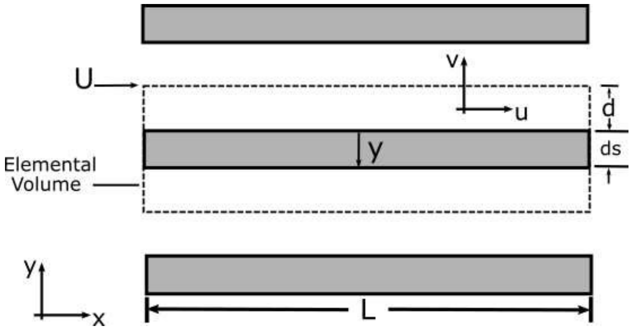


Figure 3. Elemental volume with longitudinal gas flow and transversal diffusion length scales.

The elemental volume is composed of substract bed (of width ds) and gas channel (of width d), in a proportion defined by an apparent porosity $\phi = 1 - (\frac{ds}{d})$.

According to the Constructal Method, the fundamental parts of a flow system are first identified: what flows, what are the degrees of freedom for design, and what are the restrictions the system is subject to. We then vary each of the degrees of freedom, study the effects it has in the system, select the best performing result, and repeat the process considering the previous results. Here, we wish to provide better access to the capturing bed to the reacting flow containing CO_2. Having established the degrees of freedom, restrictions, and goal, we then identify the compromises that present opportunities for design. The degrees of freedom relate to the temporal and geometry scales of the elemental volume and will be further explained.

2.2.1 Elemental volume with 1D Model

We start with the elemental volume, where we look at the degree of freedom that relates to temporal scales of the process, that is, time of residence in the porous bed and in the channels.

One expects the best use of the capturing bed will occur when the time of the reaction matches the time that the component (carbon dioxide) takes to penetrate the transversal length scale (ds). This length scale in the porous media is given in an order of magnitude sense by the Darcy equation, as described in the scale analysis:

$$ds \sim (\frac{K}{\mu}\Delta P_s t_0)^{1/2} \tag{9}$$

In which K is the permeability of the porous media for gas flow, μ the gas viscosity, ΔP_s the pressure loss in the bed, and t_0 the time of residence in the bed, which is a degree of freedom of the process to be further analyzed, and ~ stands for "equal in order of magnitude".

The characteristic time of reaction, according to the model used, is given by the reaction rate (mol.m^{-3}.s^{-1}):

$$r_i = \frac{d}{dt}[CaCO_3] = k[L_{Ca^{2+}}][CO_3^{2-}] \tag{10}$$

Where $L_{Ca^{2+}}$ is the solubility limit of Ca^{2+} in water. Whereas the reaction rate for a control volume depends on the concentration of carbonates, we can define a constant parameter k_c (s) that defines the reaction rate as a function of carbonate concentration:

$$k_c = k[L_{Ca^{2+}}] \tag{11}$$

Since k_c is given in [s^{-1}], it can be understood as the inverse of the characteristic time of the reaction.

We identify a compromise relating to this parameter and extent of reaction. For a given volumetric flow rate and CO_2 concentration, we can imagine a scenario of a high t_0 in which the all CO_2 is captured early and there is waste of bed space. We can also imagine a scenario of low t_0 such that, even though the whole bed is being used, the amount captured is insufficient.

Given that the amount of CO_2 captured depends on the reaction rate, the question that emerges is: is there an optimal ratio β [dimensionless] of t_0 and characteristic time of reaction that maximizes the amount of CO_2 removed within the imposed restrictions, such that:

$$t_0 = \frac{\beta}{k_c} \tag{12}$$

To solve the problem, it was considered a fixed volumetric flow rate, Darcy velocity, CO_2 concentration and dimensions W and L, and we model the concentration profile in the bed, in the stationary state.

We solve the constructal problem by varying t_0, and consequently, width (ds) of the bed. We calculate ds by the approximation $ds \sim ut_0$.

We then calculate the ratio of CO_2 concentration at exit and enter points in the bed. Results of dimensionless CO_2 concentration abatement ($1-C_s/C_0$) as a function of β to are shown in Figure 4.

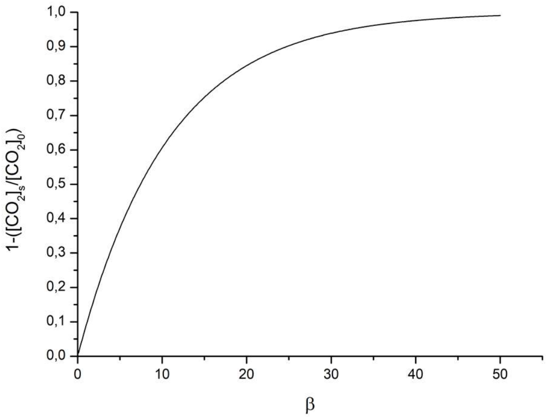


Figure 4. Dimensionless concentration abatement as a function of β

Based on these results, we may determine β and, thus, t_0, so that we operate in the range for which the reaction rate is highest. Here we opt, for dimensioning reasons, to work with a removal of 10%, which attends that criteria, and thus, work with $\beta = 1.2$.

2.2.2 First Construct

Consider now the first construct of the system shown in Fig. 4, where, in accordance to the constructal method, n_1 elemental volumes are stacked and fed the fixed total gas mixture stream $\dot{m}_1$, where the stream passing in each channel is $\dot{m}_0 = \frac{2\dot{m}_1}{(n+1)}$, and the stream passing in each bed is $\dot{m}_s = \dot{m}_0/2$. The exhaust ducts have the same geometry as the feeding ducts.

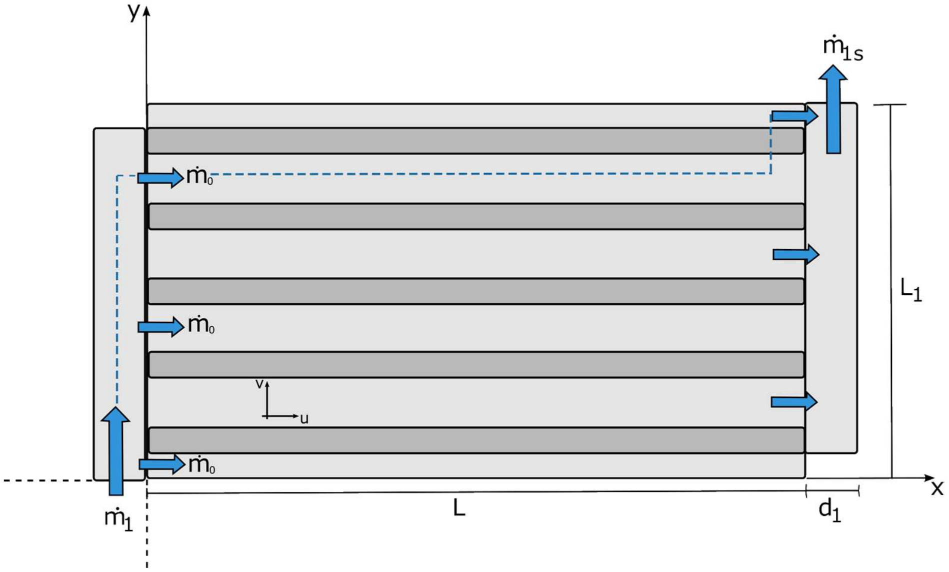


Figure 5. First construct of the process

The global size of the first construct (A_1) is fixed, $V_1 \sim (L + 2d_1)WL_1$ where L_1 is the total thickness of the stack, which is of order $L_1 \sim n_1 d$, and so that n_1 and L may vary. The width d_1 is here defined as a fixed ratio ϕ of dimension L, so that $d_1 = \phi L$. Dimensions W and L are also considered fixed in this work.

In the first construct, the time the gas takes to reach the bed must match the same t_0, in order for a continuous supply of reactant to occur. To guarantee this happens, we consider the situation in which the path of the gas to reach the bed in the longest, which is indicated in the Figure in red. This means that:

$$t_0 = \frac{L}{U_0} + \frac{L_1}{U_1} \tag{13}$$

However, the velocities in both channels depend on the volumetric flow rate and dimensions. By substituting the expressions for velocities in each of the ducts, we arrive at a relation between dimensions d and ds that guarantees the expression above holds. This relation is given by:

$$\frac{ds}{d} = \frac{(2 + 4\varphi)}{(1 - 2\varphi)} \tag{14}$$

Dimension L_1 is calculated for a given mass flow rate of CO_2, based on the value for t_0 found for the chosen parameter β, and the relation above. For this condition, we have fixed restrictions of total volume as well as capturing media volume ($V_s \sim ndsWL$) (Vs = ndsWL). We are then left with one degree of freedom for the elemental volume: the number of elemental volumes in the first construct, which will allow to determine d and ds with the equation above.

To increase flow access of the first construct means to minimize the pumping power required to drive $\dot{m}_1$ through the construct, which is the same as minimizing the overall pressure drop ΔP between the inlet and outlet of the stream. ΔP has three components: the pressure drops in the L direction of the elemental volume (ΔP_0), the pressure drops in the L_1 direction (ΔP_1), and the pressure drops in the porous bed (ΔP_s).

Each term of the pressure drops can be estimated analytically as (BEJAN; LORENTE, 2008):

$$\Delta P_0 \sim \frac{48\mu U_0}{(D_{h0})^2} \; (15) \qquad \Delta P_1 \sim \frac{48\mu U_1}{(D_{h1})^2} \; (16) \qquad \Delta P_s \sim \frac{\mu ds^2}{Kt_s} \; (17)$$

Thus we then address the following constructal problem: we want to minimize total pressure drop ΔP, subject to the restrictions of fixed V, Vs and a given $\dot{m}_1$, by varying the number of elemental volumes in the first assembly. For each configuration, both the mass flow rate as well as the available substract volume Vs will be divided in *n* elemental volumes.

The parameters and dimensions for the first construct are presented in the Table 2.

Table 2. Parameters and dimensions for the first construct

Total volume (L)	13.58
Substract volume (L)	8.15
Length, L (m)	0.3
Width, W (m)	0.1
Height, H (m)	0.45
Total bed height, ds	0.27
Volumetric flow rate (L/min)	10
Inflow CO_2 concentration (mol/m³)	9.94
Temperature (°C)	80
Bed porosity, ε	0.47
Saturation	0.2
Range for n	1-15

We solve this problem first by calculating the analytical solution for pressure drops for each value of n, and then compare these results with the numerical solution obtained by applying the 2D Model in COMSOL Multiphysics® v. 5.3a.

2.3 Numerical solution

The procedure for numerical solution in software COMSOL Multiphysics® v. 5.3a consisted in solving mass transport equations in the free and porous medium, Navier-stokes equations for free flow in the gas channel, and Darcy's law for flor in the porous media.The interfaces used ere Free and Porous Media Flow and Transport of Diluted Species in Porous Media, and the boundary conditions of an inlet velocity, outlet atmospheric pressure, no flux and no-slip at the internal walls, inflow CO_2 concentration, reaction in the porous bed and the transport properties.

The mesh employed in the runs was built with the built-in configurations of the software ("physics-controlled"), in the configuration "Finer". To select this configuration, a mesh convergence study was performed with the progressively refined built-in configuration, and the results were compared for a "Cut line" data set crossing the reactor in the transversal direction. Since no difference in results was perceived, the mesh was adopted according to quality plots provided by the software, in order to avoid elements of bad quality in the mesh. The mesh adopted was the less refined one in which the improvement on minimum element quality was less than 5%. The adopted mesh and its quality for n=6 are illustrated in Figure. It should be noted that results for pressure drop with the chosen mesh converge to the analytical solutions.

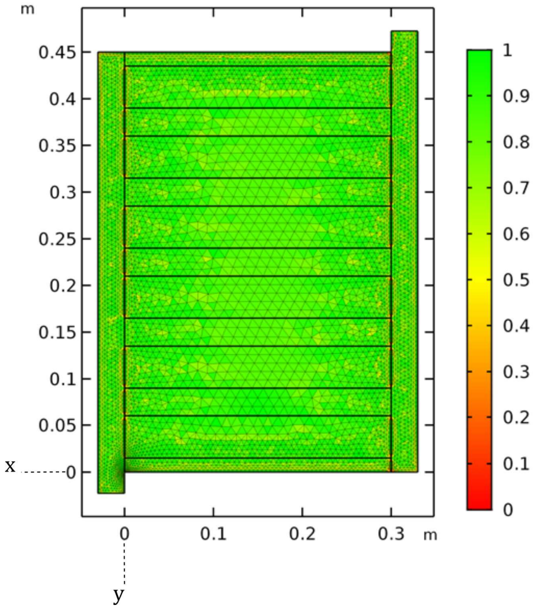


Figure 6. Quality of mesh adopted for simulations

The parameters of the selected mesh are presented in Table 3.

Table 3. Mesh Parameters

Maximum elemento size (m)	0.0126	Curvature factor	0.3
Minimum elemento size(m)	3.6e-4	Resolution of narrow regions	1
Maximum growth rate	1.13	Number of elements	11735

3. RESULTS

Results for the pressure, velocity and concentration profiles show a convergence to analytical results. For n=6, analytical and average numerical results for parameters of velocity and exit concentration of the reactor are shown in Table.

Table 4. Comparison of Analytical and Numerical Results

Parameter	Analytical	Numerical (average)
Velocity in feeding channel, U_1 (m/s)	0.0278	0.0295
Velocity in elemental channel, U_0 (m/s)	0.0092	0.0093
Darcy velocity in porous bed, u (m/s)	9.259e-4	9.212e-4
Ratio Outflow and Inflow concentration, C_s/C_0	0.8941	0.912

Results also show that the dimensioning guidelines (parameters β and ds/d) used for, respectively, obtaining the adequate bed height for the desired removal and guaranteeing that the times of residence are adequate, hold on the numerical simulations.

Figure shows the velocity, dimensionless pressure (in relation to total pressure drop in the bed) and dimensionless concentration profiles in the first construct for n=6.

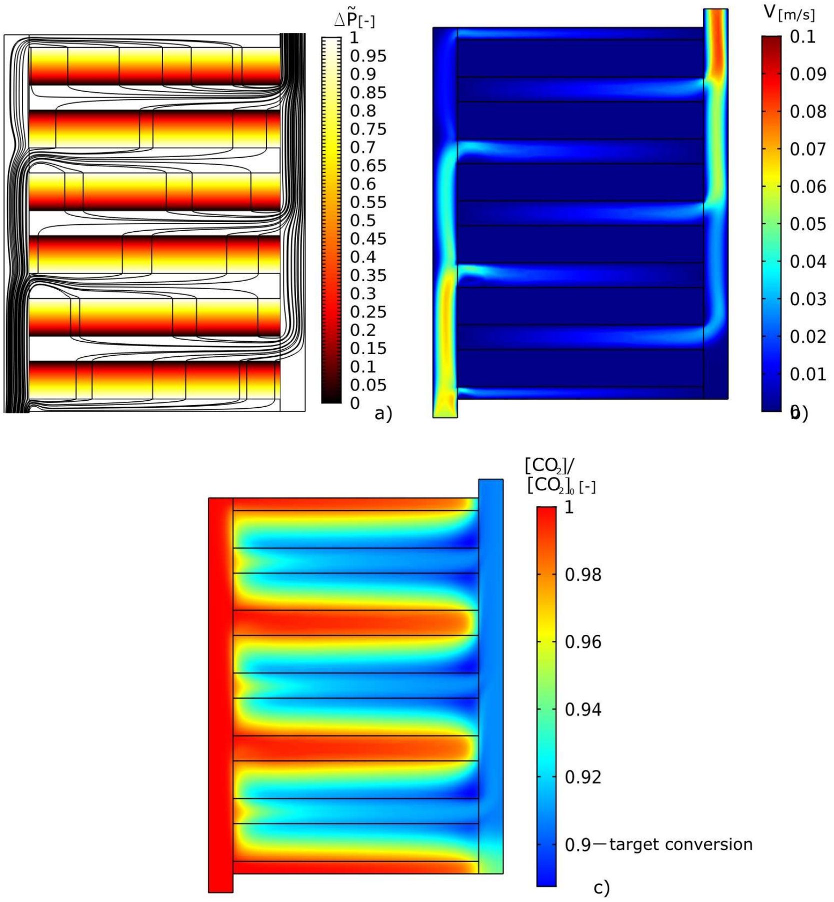
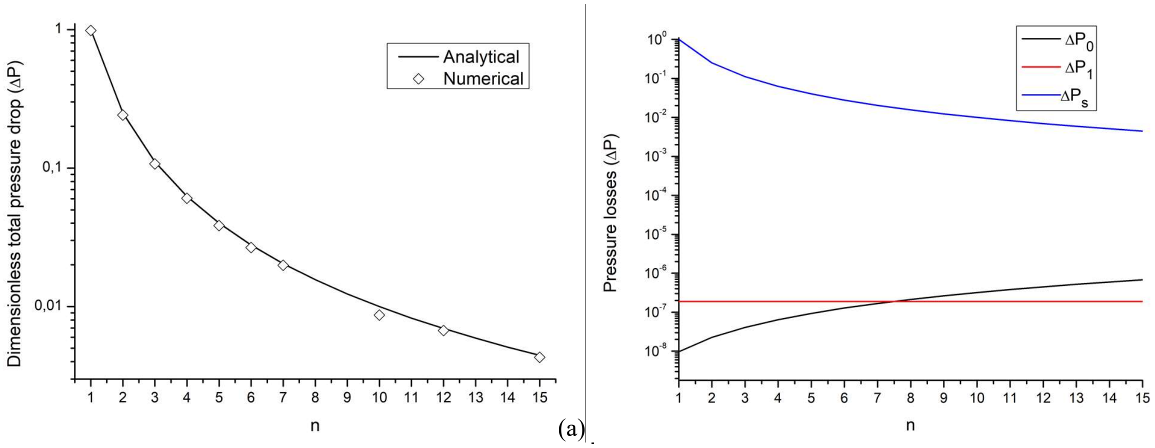


Figure 7. Numerical results for the first construct: a) Dimensionless pressure; b) Velocity (m/s); c) Dimensionless concentration profile

Results show how the concentration abatement corresponded within a reasonable margin with the target removal of 10%. Also, since the pressure drop in each bed is predominantly in the transversal direction of the bed, numerical results also prove that the assumptions made in the 1D Model (negligible diffusion in the y direction of bed and negligible Darcy velocity in the x direction of bed) was adequate.

Figure 8(a) shows the corresponding values found for total pressure drop in the first construct obtained in the numerical 2D Model and the analytical solution. Figure 8(b) shows each term of the pressure drops calculated analytically. Results shown are dimensionless, in relation to the analytical results for n=1 (configuration with the maximum pressure drop).



(a) (b)

Figure 8. Dimensionless pressure drops for number of elemental volumes in the first construct: (a) analytical and numerical results for total pressure; (b) analytical results for each term of pressure loss

Results show that the minimum thickness of the capturing medium that yields the targeted conversion leads to the pressure losses on the porous bed being the predominant term, as predicted by the unidimensional model, as well as that the evolutionary path of configurations for less pressure losses is found by going in the direction of increasing number of elemental volumes in the bed.

4. CONCLUSIONS

The application of the Constructal method to a process of carbon dioxide capture with mineral carbonation made possible to obtain dimensioning guidelines for time of residence in the porous bed in order to obtain a desired CO_2 abatement, as well as the evolutionary path of geometry scales for the first construct of the process which matches the CO2 conversion rate with its input of exhaust gases from the given point source, as well as minimize pressure losses.

Numerical results of removal in the porous bed showed good agreement between 1D and 2D Models for the elementary volume and first construct, respectively, and results of the numerical 2D Model for pressure losses converge to the analytical results. The next steps in the research will continue to address the geometry problem for the device, searching optimal spacing distribution of higher constructs, aiming to maximize carbonation for a fixed reactor volume.

5. ACKNOWLEDGEMENTS

Financial assistance by CAPES and ANEEL/COPEL PD 2866-0470-2017 are gratefully acknowledged.
MRE work was partially supported by grant CNPq 312.615/2018-3 (Brazilian Federal Government).

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