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ON THE USE OF GAS RECIRCULATION TO INCREASE THE ADSORPTION CAPACITY IN THE NATURAL GAS STORAGE SYSTEMS

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Abstract. *The use of adsorption to increase the storage capacity of gas tanks has become a promising area in engineering. In the process to store natural gas (CH₄), the main challenge is the management of the heat transfer in the filling process. This happens once the adsorption is an exothermic process and the adsorbent materials have low thermal conductivities, which leads to the high temperatures and reduction of the adsorption capacity. This work proposes a numerical study of a new solution to improve the thermal control of the process using a gas recirculation during the filling process, when part of the heated gas inside the tank is extracted, cooled and returned to the inlet. Moreover, the main goal of this work is investigated ratios of recirculation that increased the gas capacity stored. The study was based on the natural gas adsorbed by active carbon, and the numerical simulations were performed using a code programed in the open source FreeFem++, considering a cylindrical tank. The results showed the potential of the recirculation analysis demanding a complete sensitivity analysis to find the best configurations of the ANG filling process.*

Keywords: *Adsorption, Natural Gas, Porous Media*

1. INTRODUCTION

In the past few years, countries around the world are focused on changing its energetic matrices. In this scenario, the use of natural gas is increasing in the last years once it is seen as a transition fuel to reduce the use of fossil fuels. The use of NG is also encouraged due to the possibility of gas reform to produce hydrogen and carbon capture. Usually, the consumption of the gas does not occur where it is produced, hence it is necessary to transport the gas through different modes and, to optimize this distribution it is important to store as much gas as possible. The two more conventional ways to do so are the compressed natural gas (CNG) and liquefied natural gas (LNG). In the first one, the gas is stored inside a tank that receives NG through a compressor. The pressure range is from 10 bar up to 200 bar. In the second, the NG is liquefied by means of a cryogenic process.

Alternatively, to these two modes, one can consider the Adsorbed Natural Gas (ANG). It has been identified as the most promising low-pressure alternative for storing natural gas. Although the ANG technology does not attain the store capacity achieved in LNG, it provides a method of storing gas at substantially higher concentration than can be achieved with CNG and dismiss the use of a cryogenic process (Judd, R.W.; Gladding, R. et. Al, 1998).

Regarding the ANG technology, it is important to improve the adsorption (filling) process. The best way to achieve this goal is increase the relation between the volume of stored gas at ambient conditions and the volume of the tank (known as V/V). One of the first relevant works about ANG systems are done by Matranga and Myers, where they performed molecular simulations using Monte Carlo Method to predict the adsorption capacity of Methane in a simple slit-model carbon. The simulations compare the capacity of ANG and CNG with results that show an advantage of the first. For 3,4 MPa in an ANG system, the V/V in the filling process achieved 209 (Matranga, K. R.; Myers, A. et Al, 1992).

A two-dimensional model was developed by Mota and others which describes the hydrodynamics, heat transfer and adsorption phenomena (Mota J.; Saadjian E., et Al.,1995). The authors pointed out two important problems in the ANG systems: the first is related to the shape of adsorption isotherm, which prevents the system from responding linearly to pressure. In other words, the pressure drop required to remove the first 10% of the fuel store is not the same as to remove the last 10%. The second problem is about the heat of adsorption. For methane adsorption in activated carbons, the values

varying from 10 to 18kJ/mole. The heat is responsible for increasing the temperature inside the tank during the filling process, thus reducing the storage capacity.

A recent work, done by Sahoo and John, provided a numerical and experimental analysis of different ANG cylindrical tanks and propose solutions to the heat management using forced convection (Sahoo, P.; John, M. et. al, 2011). All the tests using 300K pure methane as inlet gas and the maximum temperatures after the process of adsorption achieved close to 360K. The results showed significant reductions (from 352K to 334 K) of the volume average temperature as the ratio between the length and the tank diameter (L/D) increases. Also, the use of the forced convection reduces the temperature close to the tank walls, where the most part of the cylinder volume is located. However, the forced convection has a limit range and the maximum temperature in the middle-center of the tanks stayed high.

Then, to perform an active thermal control of the filling process in different ways such as using heat exchange's devices inside the tank, this research group use the same geometry of the Sahoo's work, did numerical simulations of the proposal and published in COBEM (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017), where one of the results is presented in the figure 1:

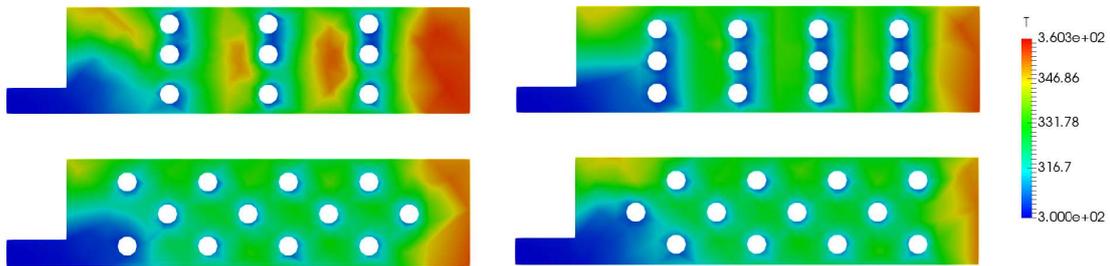


Figure 1. Temperature distributions of numerical 2D-axisymmetrical simulations using different internal heat exchangers disposals (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017).

The work showed that the tubes that are arranged in tandem showed best results of V/V in comparison to aligned tubes, where the capacity of the tank increased more than 20% as compared to baseline test, without the heat exchangers. Although the results were satisfactory, the trade-off between the quantity of tubes and the available volume inside the tank is an important parameter to find the best configurations.

Advancing in the research, the group focused on to study the sensitivities of the parameters that influence the capacity of stored gas. Using the solver developed in the previous work (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017), a sensitivity analysis using the so-called Adjoint Method was developed, where the influence of the forced convection, inlet gas temperature and filling flow curves were evaluated, considering as objective functions the volume average pressure and volume average temperature (Chierigatti, B.G; Brasil Lima, J.S. et al, 2018) and the quantity of adsorbed gas stored (Brasil Lima, J.S; Chierigatti, B.G. et al, 2018).

Assembling all works developed, a complete optimization loop algorithm was developed (Chierigatti, B.G; Brasil Lima, J.S. et al, 2021) and systematic studies of the filling flow curve were performed and published. The results showed a non-intuitive curve that proposes a rapid increase of the flow during the first 25% of the process time and a smooth decrease of the flow in the remaining time. The figure 2 shows one of the cases published (Brasil Lima, J.S; Chierigatti, B.G. et al, 2020):

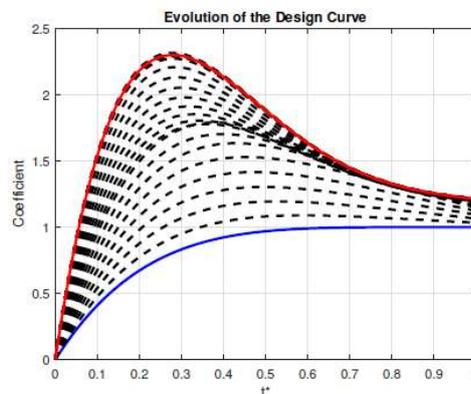


Figure 2. Filling flow design optimization (Brasil Lima, J.S; Chierigatti, B.G. et al, 2020)

As the research group realized a potential in the control of the flow curve, this work starts an exploratory work, aiming to verify the potential of using gas recirculation in the filling process, in order to be able to apply the same sensitivity concepts in the future and consequently perform design optimizations.

2. MATERIALS AND METHODS

This work is based on the numerical simulations, that results will be used to the future experimental validations.

2.1 Mathematical model

The governing equations which modeled a flow through a porous media with adsorption were developed in a previous work published in this congress (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017) and presented below:

$$\epsilon_t \frac{\partial \rho_g}{\partial t} + \rho_b \frac{\partial q}{\partial t} + \nabla \cdot \vec{G} = 0 \quad (1)$$

$$\rho_g \nabla p + \frac{\mu}{K} \vec{G} = 0 \quad (2)$$

$$C_{eff} \frac{\partial T}{\partial t} - \epsilon_t \frac{\partial p}{\partial t} + \nabla \cdot (C_{pg} \vec{G} T) - \lambda_{eff} \nabla^2 T - \frac{\Delta H}{M_g} \rho_b \frac{\partial q}{\partial t} = 0 \quad (3)$$

$$q = \rho_{ads} \cdot W_0 \cdot \exp \left[- \left(\frac{A}{\beta E_0} \right)^n \right] \quad (4)$$

The eq. (1) is the continuity equation where:

- ϵ_t is the total porosity of adsorbent bed (no-dimensional);
- ρ_g is the free gas density (located in the void and pores);
- ρ_b is the density of adsorbent bed;
- q is the density of adsorption, determined by the eq. (4)
- \vec{G} is the mass flux vector.

The eq. (2) are the momentum equations (2D axi-symmetric) with Darcy law simplifications, where:

- ∇p is the pressure gradient;
- μ is the viscosity of the gas
- K is the permeability of the adsorbent bed (m²).

The eq. (3) is the Energy equation, where:

- $C_{eff} = (\epsilon_t \rho_g + \rho_b q) C_{pg} + \rho_b C_{ps}$, which C_{pg} and C_{ps} represents the specifics heat of gas and adsorbent respectively.
- T represents the temperature
- $\lambda_{eff} = \epsilon_t \cdot \lambda_g + (1 - \epsilon_t) \lambda_s$ is the effective thermal conductivity in terms of porosity and thermal conductivity of gas (λ_g) and adsorbent (λ_s)
- ΔH is the heat of adsorption
- M_g is the molar mass of the gas.

The eq. (4) is the Dubinin-Astakov (D-A) adsorption model (Sahoo, P.; John, M, 2011), where:

- ρ_{ads} is the adsorbed gas density, defined by: $\rho_{ads} = \frac{\overline{\rho_{ads}}}{\exp [\alpha_e (T - T_b)]}$, where $\overline{\rho_{ads}}$ is the density of liquid phase of the adsorbed fluid in the saturation region (T_b)
- α_e is the mean value of the thermal expansion of liquefied gas.
- W_0 is the microporous volume per unit mass of adsorbent
- β is the affinity coefficient related to the adsorbate-adsorbent interaction
- E_0 is the characteristic energy of adsorption
- n is the DA exponent which is related to the pore size dispersion (Sahoo, P.; John, M, 2011).
- The parameter A is the so-called as Polany adsorption potential and it is defined by: $A = RT \ln \left(\frac{P_s}{P} \right)$, where $P_s = P_{cr} \left(\frac{T}{T_{cr}} \right)^2$.

The evaluation of the time derivative $\partial q / \partial t$ w which appears in eqs. (1) and (3) is based on the model called linear driving force (Xiao, J; Peng, R et al.,2012):

$$\frac{\partial q}{\partial t} = k \cdot (q^* - q) \quad (5)$$

Where q^* is the adsorbed gas in the equilibrium with saturated gas phase, which is calculated using the eq. (4) and k is the mass transfer coefficient at an aggregated level. In the numerical simulations, q is calculated explicitly using the previous time step.

2.2 Solver Implementation

These equations are carried out by using a finite-element platform FreeFEM++. This platform is a high-level integrated development environment (IDE) for numerically solving partial differential equations (PDE) in 2 and 3 dimensions. It is an ideal tool for studying the finite element method, but it is also very useful for research, to quickly test new ideas or multi-physics and complex applications (Hecht, F., 2012).

The software has a simple mesh generator, suitable for the 2D-axi symmetric simulations. The mesh convergence was explored in the previous work and we use the same discretization (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017). The only difference is the placing of the outlet section, opposite to the inlet section, as can be seen in the figure 3:

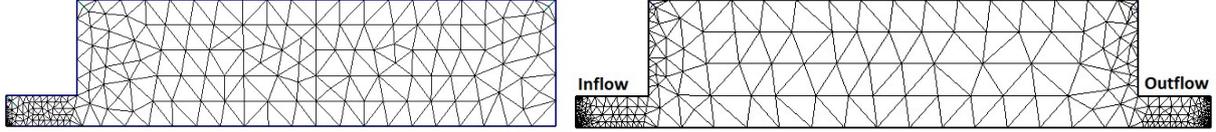


Figure 3. 2D-axisymmetric mesh geometries. Left: Previous work with only inflow section (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017). Right: New geometry with the outflow section.

The baseline test was also the same of the last works, to compare the different solutions with the same reference. The reference inlet flow was 15 LPM ($G = 5.5615 \text{ kg/m}^2/\text{s}$) and the other parameters is presented in the table 1:

Table 1 – Data and Initial Conditions – Same of presented in previous work (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017)

	Value		Value		Value
ρ_0	0.7049 kg/m^3	T_∞	300 K	α	$2.5 \cdot 10^{-3} \text{ K}^{-1}$
C_{pg}	2450 J/kg.K	C_{ps}	650 J/kg.K	E_0	$25.04 \cdot 10^3 \text{ J/mol}$
μ_g	$1.25 \cdot 10^{-5} \text{ Pa.s}$	λ_g	0.0343 W/m.K	P_{cr}	$45.96 \cdot 10^5 \text{ Pa}$
ϵ_t	0.65	ϵ_b	0.30	$\overline{\rho_{ads}}$	422.62 kg/m^3
λ_s	0.54 W/m.K	ρ_b	500 kg/m^3	n	1.8
ΔH	12000 J/mol	h	$5 \text{ W/m}^2\text{K}$ (natural)	β	0.35
M_g	0.016 kg/mol		$700 \text{ W/m}^2\text{K}$ (forced)	W_0	$3.3 \cdot 10^{-4} \text{ m}^3/\text{kg}$
		K	$3.7 \cdot 10^{-10} \text{ m}^2$	T_{cr}	191 K
* STP Conditions		P_i	20000 Pa	T_b	111.2 K
		T_i	303 K	k	3.2 s^{-1}
		q_i	$q(P_i, T_i)$		

The active thermal management may have a most significant impact on the filling process of ANG systems. Hence the boundary conditions that affect those process were the focus of a relevant publication of the authors (Chierigatti, B.G; Brasil Lima, J.S. et al, 2021). To insert these conditions in this particular test, it is necessary to define the recirculation mass flux that it be imposed in the outflow condition. The table 2 present the boundary conditions:

Table 2: Boundary Conditions with the recirculation mass flux. Adapted from (Chierigatti, B.G; Brasil Lima, J.S. et al, 2021).

Boundary Type	Conditions
Inflow	$\vec{G} \cdot \vec{n} = (G_{ref} + G_{rec})$ $-\lambda_{ref} \cdot (\vec{n} \cdot \nabla T) = (G_{ref} + G_{rec}) \cdot C_{pg} \cdot (T_{in} - T)$
Outflow	$\vec{G} \cdot \vec{n} = G_{rec}$ $-\lambda_{ref} \cdot (\vec{n} \cdot \nabla T) = G_{rec} \cdot C_{pg} \cdot (T_\infty - T)$
Wall	$\vec{G} \cdot \vec{n} = 0$ $-\lambda_{ref} \cdot (\vec{n} \cdot \nabla T) = h \cdot (T - T_\infty)$
Symmetry	$\vec{G} \cdot \vec{n} = 0$ $(\vec{n} \cdot \nabla T) = 0$

Where G_{ref} is the reference mass flux ($5.5615 \text{ kg/m}^2/\text{s}$) and G_{rec} is the recirculation mass flux, which is in terms of percentage of G_{ref} . All the simulations considered a transition time of 30s to archive the reference mass flux. In the other words, the inlet mass flux started at zero in $t = 0$ and grow linearly until G_{ref} during the first 30 seconds. After this time, the mass flux was kept constant at value of G_{ref} . For the recirculation, it was defined the time when it starts and a transition

time to archive the value of G_{rec} (in this case, we use 30s for all the tests). During the recirculation, the instant value of G_{rec} is added to the inflow, as presented in the table 2. The objective of this addition is for the value of the liquid flow in the tank equals to G_{ref} during the recirculation.

3. RESULTS AND DISCUSSION

3.1 Baseline Test

The baseline test was solved without the recirculation. The objective is creating a reference result and define the time of the whole filling flow process. Based in the previous works, the simulations were set to stop when the pressure archive 35 bar (3.5 MPa) and this was occurred close to 410s. To isolate the effect of the recirculation, the convection in the tank walls was considered natural. The figure 4 shows the evolution of the temperature distribution during the filling time:

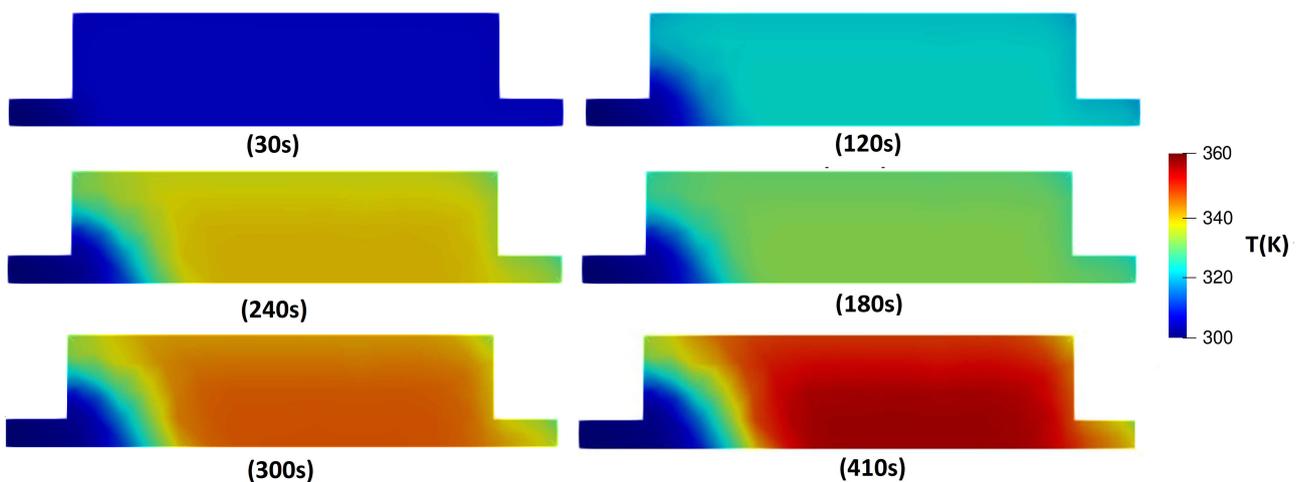


Figure 4. Evolution of the temperature distribution during the filling time. Baseline test without the recirculation

At 410s, the volume average pressure inside the tank was 33.65 bar, the volume average temperature was 347.4 K and the maximum temperature 357.8 K. The volume average density of adsorption was 5.82 % and the V/V was 57.66. It is important to point that the objective of this research is the study of the influence of the recirculation in the fields, with an intention of growing the areas with low temperatures (blue, green and yellow areas), which increase the local adsorption and the quantity of gas stored consequently.

3.2 Influence of the recirculation ratio

The first test varying the recirculation ratios (50, 100, 200, 300, 500% of G_{ref}), keeping the same initial time and the same duration. In the other words and showing the values, the filling flow starts at $t = 0$, growing linearly until $t = 30$ s, the recirculation starts at 120s and stops in 410s. The figures 5-7 show the results:

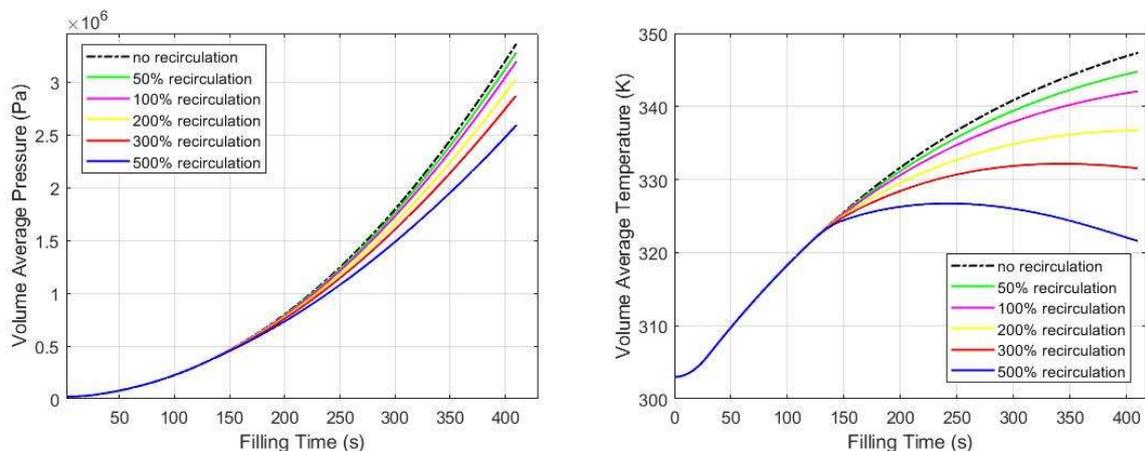


Figure 5. Left: Evolution of the volume average pressure. Right: Evolution of the volume average temperature. Recirculation started at 120s.

The results showed a negative sensitivity between the recirculation ratio, volume average pressure and temperature. Figure 5 left shows the pressure reduced from 33.65 bar without recirculation at 410s to 32.83 bar (-2,4%) with 50% recirculation and 31.98 bar (-5,0%), 30.34 bar (-9,8%), 28.78 bar (-14,5%) and 25.94 bar (-22,9%), with 100, 200,300 and 500% recirculation respectively. About the temperature, the values reduced from 347.4 K to 344.7 K (-0,8%), 342.0 K (-1,6%), 336.7 (-3,1%), 331.5 K (-4,6%), 321.5 K (-7,5%) at 410 for 50 ,100, 200, 300 and 500% recirculation respectively.

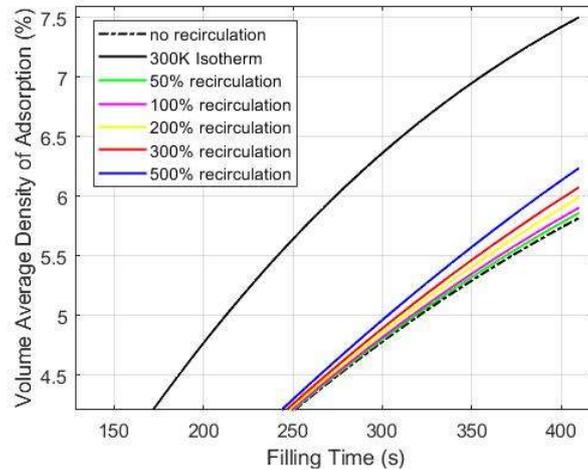


Figure 6. Detail of evolution of the volume average density of adsorption. Recirculation started at 120s.

The consequence of the results presented in the figures 4-5 was the positive sensitivity with the recirculation ratio and the density of adsorption. Figure 6 shows at 410s a growing from 5.82% without recirculation to 5.86, 5.90, 5.99, 6.07 and 6.24% to 50, 100, 200, 300 and 500% respectively. Moreover, the figure 6 presents the isotherm curve in black continuous line. This curve represents the maximum capacity of adsorption in the ideal process which value at 410s is 7.5%.

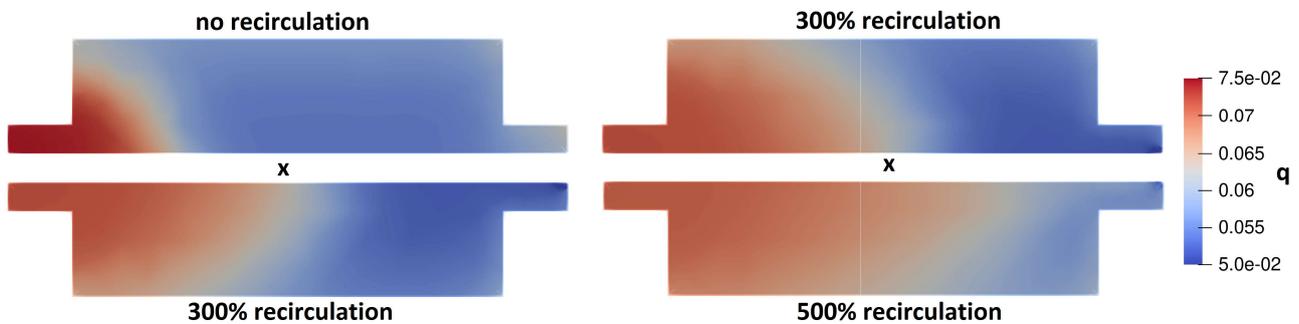


Figure 7. Density of Adsorption distribution comparison at 410s. Left, no recirculation x 300% recirculation. Right, 300% recirculation x 500% recirculation

The figure 7 presents the advance of the region with more gas adsorbed. It is important to note that the liquid flow of gas entering in the tanks was kept the same during the simulations to make a fair comparison. The reduction of pressure, temperature and the growing of the density of the adsorption means more gas adsorbed and the potential to continue the filling process until the tanks with recirculation achieve 35 bar. When it occurs, the mass inside the tanks will be higher in comparison between the baseline test.

3.3 Influence of the recirculation init

The other analysis of this research was the influence of the time that the recirculation begins. The recirculation ratios were kept the same that were presented in the sec. 3.1 and the new simulations varying the initial time of the recirculation (60, 180 and 240s). The next figures show some of these results, focused on the ratios of 300% and 500%.

300% recirculation – The figures 8-10 present the behavior of the volume average pressure, temperature and density of adsorption during the filling time:

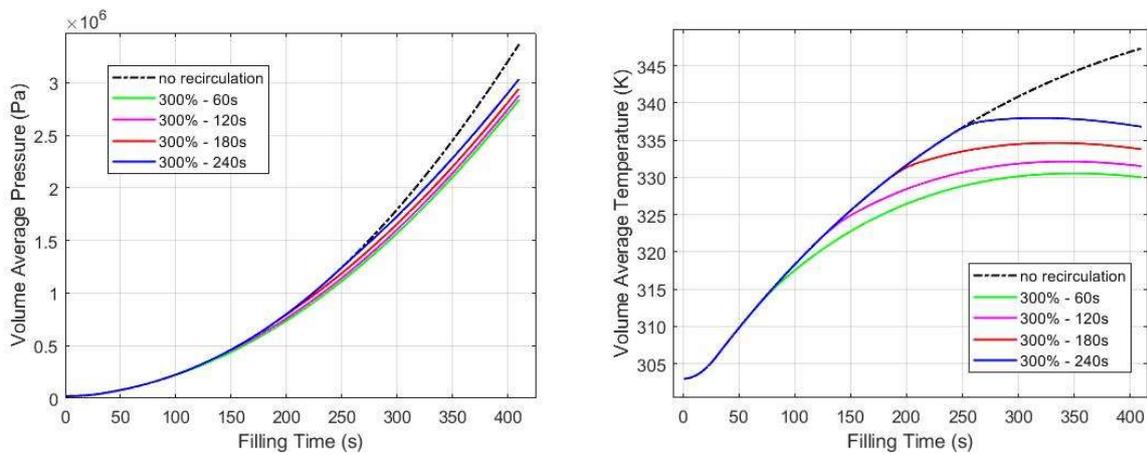


Figure 8. Left: Evolution of the volume average pressure. Right: Evolution of the volume average temperature. Recirculation ratio fixed in 300%.

The behavior of the sensitivity was opposite that it was seen in recirculation ratio. For a fixed ratio, when the starting time grows, the volume average pressure and temperature grows and density of adsorption decreases consequently. The results show the same comparison between the baseline test, with a pressure values of 28.36 bar (-15,7%), 28.78 bar (-14,5%), 29.45 bar (-9,8%) and 30.35 bar (-9,8%) for recirculation starts in 60, 120, 180 and 240s respectively. For the temperature, the values grown to 330.4 K (-4,9%), 331.5 K (-4,6%), 333.8 K (-3,9%) and 336.8 K (-3,1%) for the same recirculation starts.

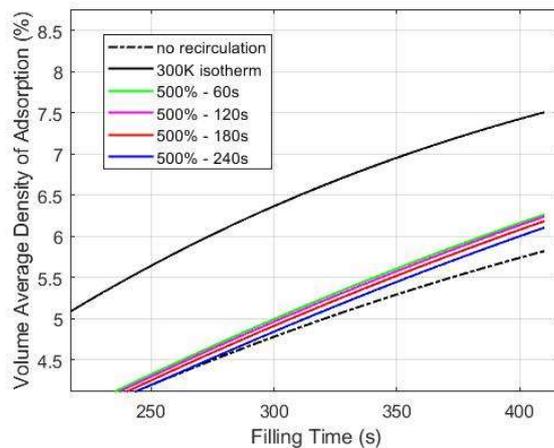


Figure 9. Detail of evolution of the volume average density of adsorption. Recirculation ratio fixed in 300%.

The figure 9 shows the decrease of the average density of adsorption in tanks with recirculation. For the recirculation starts in 60, 120, 180 and 240s, the values were 6.10, 6.07, 6.04 and 5.09% at 410s respectively. The consequence of the reduction is the contraction of the regions with more density of adsorption, as it can be seen in the figure 10:

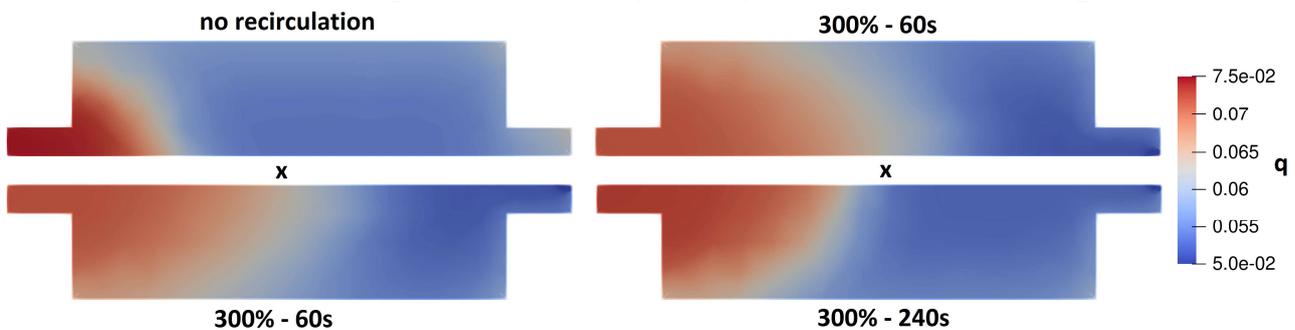


Figure 10. Density of Adsorption distribution comparison at 410s with recirculation ratio in 300%. Left, no recirculation x 60s recirculation start. Right, 60s recirculation start x 240s recirculation start

The results showed an important trade-off between the recirculation ratio and the time that it starts. There is a possibility to have an optimal point with these parameters and a complete analysis of the values of the sensitivities will be necessary.

500% recirculation – The figures 11-13 present the behavior of the volume average pressure, temperature and density of adsorption during the filling time:

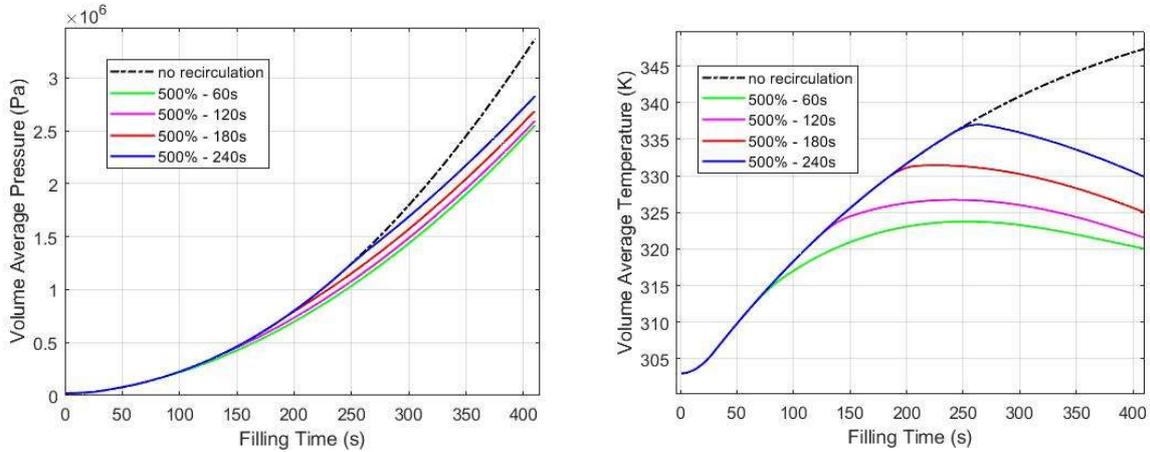


Figure 11. Left: Evolution of the volume average pressure. Right: Evolution of the volume average temperature. Recirculation ratio fixed in 500%.

The results show the same comparison between the baseline test without the recirculation, with a pressure values of 25.52 bar (-24,2%), 25.94 bar (-22,9%), 26.89 bar (-20,1%) and 28.26 bar (-16,0%) for recirculation starts in 60, 120, 180 and 240s respectively. For the temperature, the values grown to 320.0 K (-7,9%), 321.5 K (-7,5%), 325.0 K (-6,4%) and 329.8 K (-5,1%) for the same recirculation starts.

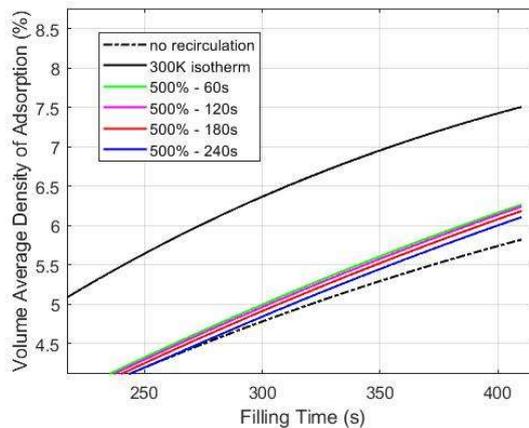


Figure 12. Detail of evolution of the volume average density of adsorption. Recirculation ratio fixed in 500%.

For the density of adsorption, the results showed values above 6,0%, with the best value of the 6,26% to 60s recirculation start. In 120s, the value was 6,24%, followed by 180 and 240s with 6,18 and 6,10% respectively. The figure 13 presents the reduction of red adsorption areas as the time of recirculation start grows:

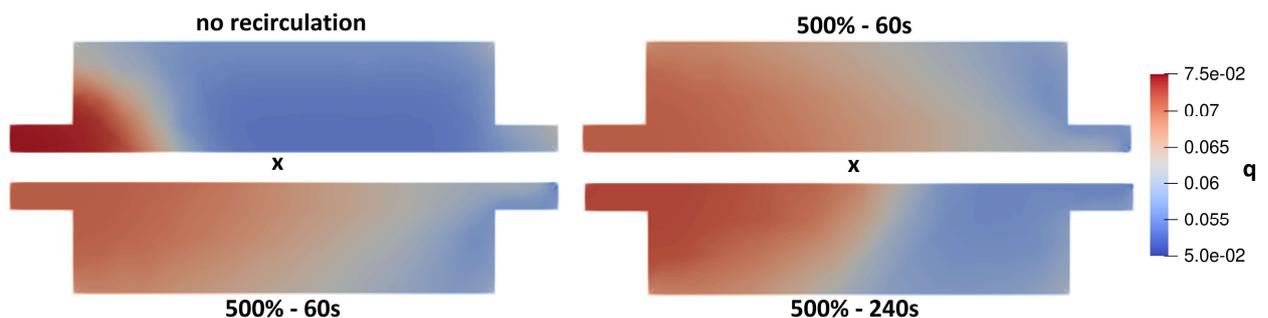


Figure 13. Density of Adsorption distribution comparison at 410s with recirculation ratio in 500%. Left, no recirculation x 240s recirculation start. Right, 240s recirculation start x 60s recirculation start

Also, it was possible to see a reduction of the best adsorption areas, which means a trade-off between the time when the recirculation starts and the recirculation ratio. Moreover, this analysis focused on these parameters, but the study can expand to other control parameters process, as forced convection.

4. CONCLUSIONS

This research archives the objective of explore the potential of the recirculation during the filling of the Adsorbed Natural Gas (ANG) storage tanks. Numerical simulations were performed with a comparison between the same baseline test and showed a possibility to increase the distribution of the density of adsorption not only in tank center but also close to the tank walls, where the most part of the geometric volume in cylindrical tanks was concentrated. One of the possibilities to improve the results is combine the recirculation with a forced external convection in the tank walls and it will be study in the future.

The values found in the density of adsorption were close to the results using the internal heat exchangers published recently (Chierigatti, B.G; Brasil Lima, J.S. et al, 2017). The great news was the recirculation do not need a reduction of internal volume, where in some cases is about 10% due presence of the internal tubes. Then, if a comparison based on the V/V or the mass inside the tank, the recirculation has a significant advantage.

The trade-off between the time when the recirculation starts and the capacity of the storage tank open a possibility to a detail study about the sensitivities and the research team already have experience in this approach using the so-called Adjoint Method (Chierigatti, B.G; Brasil Lima, J.S. et al, 2021). With an optimization algorithm developed, the gradient expressions will be deducted and the authors expected to advance the studies for publish the results in the future articles. The expectation is to develop a complete control during the filling process, allying the inlet filling flow curve, recirculation and external convection, to find the best configuration that get closer to isotherm process.

Also, the research team started a project at Maua Institute of Technology to study CCS (Carbon Capture Storage) and the expectation is the use of the expertise acquired in natural gas to study the possibilities to store carbon dioxide with adsorption in activate carbon or in ZIF-8, material that shows with great affinity with CO_2 . The first simulations shows a large quantity of heat produced by the adsorption and the recirculation can be one of the solutions to maximize the capacity stored.

5. ACKNOWLEDGMENTS

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