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EVALUATION OF THE PRESENCE OF CARBON MONOXIDE IMPACT ON CO_2-RICH MIXTURE DENSITY AND PHASE BEHAVIOR AT CCS PROCESS CONDITIONS

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Abstract. *Carbon Capture and Storage (CCS) technology is one of the main alternatives to help reduce CO_2 emissions into the atmosphere and thus contribute to the UN's Sustainable Development Goals. However, impurities in the captured CO_2 play a crucial role in safely and effectively transporting and storing CO_2 in the CCS chain, interfering with the cost and posing challenges to these processes. This article focuses on investigating the impact of carbon monoxide in CO_2-rich mixtures over the operational range applied to CCS processes concerning thermophysical properties and phase changes. The CO_2 - CO density was modeled using the equations of state of PR78, GERG-2008, and EOS-CG through the software Multiflash. The density simulations were performed at temperatures from 283 to 373 K, pressures from 1.9 to 48.65 MPa, and CO mole fractions from 0.05 to 0.51. The results were compared to experimental data from the literature to calculate the EoSs' accuracy. These were also compared to the pure CO_2 density from the Span and Wagner model to measure the impact of the CO. The EOS-CG performed better than the other evaluated models, with a MAPE of 1.76%. The presence of CO has an inversely proportional effect on the CO_2 density.*

Keywords: *CO_2, Impurities, Phase Behavior, Equations of State, CCS*

1. INTRODUCTION

Carbon Capture and Storage (CCS) arose as a sustainable solution for those who want to compensate for their CO_2 emissions. Considering the high investment cost involved in the whole process, energy companies are the main financiers of these technologies. On the other hand, CCS presents high aggregated value for all economies enrolled, and this gives a positive balance on the closing counts not only for the companies but also for the committed countries.

The segment of decarbonization worldwide is growing fast and fomenting eco-development, which means the improvement by self-potentiality of the countries (Araújo et al., 2017). Geopolitics is highly influenced by the development of offshore wind, electric cars, carbon capture, utilization, and storage, to name some technologies in development that focus on reducing carbon footprint worldwide and entering the carbon credit market.

Carbon Capture and Storage appears to be the scenario with more significant challenges for working with a composition that presents a very low critical point compared to other gases (304.13 K and 7.38 MPa). The carbon dioxide flow conditions (temperature and pressure) are above this critical point, which means the gas will be in a supercritical state that behaves non-linearly (Mills et al., 2022). Its viscosity performs as gas while its density as liquid, which makes the pressure drop and thermic losses to be high, affecting the thermophysical properties of the fluid and bringing challenges for the flow assurance segment in lines of design, safety, integrity, and health (Vitali et al., 2022).

During capture, carbon dioxide is initially present in a low percentage and is transported to storage after being conditioned to become a CO_2-rich mixture. Pre-combustion, post-combustion, and oxyfuel are the main processes that process the fluid. However, those still leave traces of impurities (from 0.01% to more than 20%), such as carbon monoxide (CO), water, nitrogen, oxygen, and argon. Each method results in a different associated percentage of impurities. There are methods to reduce these fractions even more, but this would increase the costs of the capturing project (Nazare et al., 2021).

The water, for example, induces corrosion from forming carbonic acid and hydrate formations when it is present on the supercritical flow. Although, there are some impurities, such as carbon monoxide, which the behavior influence is not known (Nazare et al., 2021). The presence of CO in a CO_2-rich mixture makes the flow assurance even more puzzling the mix of thermophysical properties and phase behavior, although not broadly studied (Westman et al., 2019). Evaluation of phase behavior and thermophysical properties of impurities on the CO_2 supercritical flow is critical for optimizing

CCS's projects. This leads to better prediction of design, time and integrity of equipment, health and security of the location, and others (Nazare et al., 2021).

As many impurities are present in CO_2-rich mixture flow, not only one equation of state (EoS) can predict its thermophysical properties. One of them is Peng-Robinson (PR78), which, according to Cipollina et al. (2007), is widely used to describe the phase behavior of fluids in a supercritical state. The EoS GERG-2008 was built to calculate the thermophysical properties of natural gas mixtures and has limitations related to the boundaries of the vapor-liquid phase of CO_2-rich mixtures. Another equation of state is the EOS-CG, which was developed specifically for CO_2-rich mixture.

There is still no broad evaluation of carbon monoxide fractions impacts on CCS operations, which are the finest Equations of State to describe the behavior of CO_2 -CO mixtures and its effects on the fluid and further impacts during CCS/CCUS ongoing processes.

This paper aims to investigate the impact of CO on the density and phase behavior of CO_2-rich mixture in conditions similar to CCS processes, as well as define which EoS best describes the $CO_2 + CO$ system.

2. THEORETICAL BACKGROUND

The phase mass density is defined as:

$$\rho_m = \frac{p \cdot M_w}{ZRT} \tag{1}$$

in which p is the pressure, M_w is the phase's molecular weight, T is the temperature, R is the universal gas constant, and Z is the isothermal compressibility factor that may be obtained from EoS. This paper considers the EoSs of PR78, GERG-2008, and EOS-CG for the density calculation.

2.1 Peng-Robinson 78 (PR78)

The EoS PR78 considers the critical properties of the substance, such as critical temperature (T_c), critical pressure (P_c), and acentric factor (ω) to calculate vapor-liquid equilibrium, phase behavior of mixtures, compressibility factor, and fugacity coefficient, which is written as (Peng and Robinson, 1976; Robinson and Peng, 1978):

$$p = \frac{RT}{v-b} - \frac{a\alpha}{v(v+b)+b(v-b)} \tag{2}$$

in which a is the temperature-dependent attractive parameter and b is the co-volume parameter, while α is a temperature-dependent parameter,

$$\alpha = \left[1 + k\left(1 - \sqrt{\frac{T}{T_c}}\right)\right]^2 \tag{3}$$

here, k is a constant referred to as the acentric factor.

Specifically, the PR78 selected on Multiflash for this study was the PR78A, which, besides Eq. (*1)*, employs the Peneloux density correction (Péneloux, 1982) and the classical van der Waals mixing rule.

2.2 GERG-2008

The GERG-2008 equation is based on 21 natural gas components: methane, nitrogen, carbon dioxide, ethane, propane, n-butane, isobutane, n-pentane, isopentane, n-hexane, n-heptane, n-octane, n-nonane, n-decane, hydrogen, oxygen, carbon monoxide, water, hydrogen sulfide, helium, and argon. Over the entire composition range, GERG-2008 covers the gas phase, liquid phase, supercritical region, and vapor-liquid equilibrium states for mixtures of these components. It is considered a standard reference equation suitable for natural gas applications, requiring highly accurate thermodynamic properties.

The EoS GERG-2008 is explicit in the reduced Helmholtz energy,

$$\alpha = \frac{a}{RT} \tag{4}$$

with the independent mixture variables molar density ρ, temperature T, and the vector of the molar composition x. The function $\alpha(\rho, T, \mathrm{x})$ comprises an ideal gas part α_0, which represents the properties of ideal-gas mixtures at given ρ, T, x, α_r, which takes into account the residual mixture behavior:

$$\alpha(\delta,\tau,\mathrm{x}) = \alpha_0(\rho,T,\mathrm{x}) + \alpha_r(\delta,\tau,\mathrm{x}) \quad (5)$$

in which δ is the reduced molar density and τ is the inverse reduced temperature. It is:

$$\delta = \frac{\rho}{\rho_r(\mathrm{x})} \quad (6)$$

$$\tau = \frac{T_r(\mathrm{x})}{T} \quad (7)$$

with ρ_r and T_r being the composition-dependent reducing functions for the mixture density and temperature. The details of the complete formulation are shown and discussed in Kunz and Wagner (2012).

The range of these EOS can be separated into three parts: standard, extended, and extrapolation for temperature and pressure. Each field presents an associated uncertainty. The first one considers $90\,K \le T \le 450\,K$ and $p \le 35\,MPa$ for pressure and the extended $60\,K \le T \le 700\,K$ and $p \le 70\,MPa$.

2.3 EOS-CG

The EOS-CG was developed to present substances related to CCS and humid gas mixtures. It focuses on improving the description of phase boundaries and the solubility of gases in water with a wider temperature and pressure range.

It is similar to the GERG-2008 model described earlier and is presented in detail by Gernet and Span (2016). At Multiflash, EOS-CG includes high-accuracy reference EoSs for 14 components: methane, nitrogen, carbon dioxide, hydrogen, oxygen, carbon monoxide, water, hydrogen sulfide, argon, hydrogen chloride, diethanolamine, monoethanolamine, and chlorine. Furthermore, the EOS-CG's temperature and pressure range considers $62\,K \le T \le 2000\,K$ and $p \le 800\,MPa$. An essential factor to highlight is that the CO_2 + CO combination in the EOS-CG had its parameters adjusted ($\beta_{T,ij} = 0.993245, \gamma_{T,ij} = 1.068392, \beta_{v,ij} = 1.030855, \gamma_{v,ij} = 1.245499\ and\ F_{ij} = 0$), while in the GERG-2008 it did not.

3. METHODOLOGY

3.1 Simulations procedure

For the development of this paper, a literature review was conducted seeking experimental data that will serve as a basis for carrying out the analyses and comparisons presented here. All the surveyed studies were compiled in a databank, but this paper does not cover this step. Hence, we chose to work with the data from Souza et al. (2019). Density values were found by the author for CO compositions ranging from 5.03, 10.11, 25.25, and 50.18 % mol, pressure between 1.90 and 48.65 MPa, and for temperatures 283.15, 293.15, 303.15, 313.15, 333.15, 353.15 and 373.15 K. Overall, 720 points of data were used.

From the data selection, for each of the situations, simulations of the density values were performed for three different equations of state (EoS): PR78, GERG-2008, and EOS-CG, to understand which best describes the behavior of the CO_2 + CO system. The software used to carry out this simulation was Multiflash through the "Properties Table" tool, setting pressure values to each fixed temperature value. In Microsoft Excel, these results were compared using the concepts of MAPE, RMSE, and ARE, and the model with the slightest error will be considered more adjusted to the data set.

This same process in Multiflash was repeated, but this time considering a pure CO_2 fluid. The simulation considered the pressures and temperatures presented by Souza et al. (2019) so that it was possible to identify the impact of CO in the mixture concerning the density of the pure compound. For the simulation of pure CO_2, the Spam and Wagner model was considered. Multiflash also generated the pure CO_2 phase envelope in this step.

Having defined the best EoS for predicting the density of CO_2 + CO mixture and the density values for pure CO_2, analyses could be performed to understand how CO impacts this property and how it behaves with pressure and temperature variations.

Returning to the procedures in Multiflash, as well as constructed for pure CO_2, the phase envelopes for each CO concentration were established and compiled into a single diagram, allowing us to understand the impurity influence on the phase behavior of the mixture.

3.2 Result analysis

As mentioned, the simulated density values for each EoS were compared with the experimental values provided in the literature to identify which best fits the system. For this identification, the concepts of Root Mean Square Error (RMSE), Mean Absolute Percent Error (MAPE), and Average Relative Error (ARE) were used and are defined, respectively, by Eq. (8), Eq. (9) and Eq. (10).

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(y_i - \hat{y}_i)^2} \quad (8)$$

$$MAPE = \frac{100}{n}\sum_{i=1}^{n}\left(\left|\frac{y_i - \hat{y}_i}{y_i}\right|\right) \quad (9)$$

$$ARE = \left|\frac{y_i - y_i}{y_i}\right| \quad (10)$$

in which the term n is the number of analyzed data for the same situation, i is the Nth density value. The y_i refers to the real data (experimental densities) and the $\hat{y}_i$ is the values produced by the models (simulated densities).

At this point, it is important to highlight that both indicators are widely used for this purpose (evaluate the accuracy of prediction models) and indicate how far the simulated data are from the experimental data, RMSE expressed in the same unit of the studied property, MAPE and ARE in percentage. Both statistical methods are applied because they make interpreting the results more assertive.

4. RESULTS AND DISCUSSIONS

4.1 Comparison between model

The first analysis was to identify which of the equations of state best describes the phase behavior and density of CO_2-rich mixtures with the presence of CO. To compare the models, three factors were considered: RMSE at kg/m^3, MAPE, and ARE at %. These values were found by comparing the experimental data published by Souza (2019) and the simulated values for each model (PR78, GERG-2008, and EOS-CG) without adjustment to experimental data. The results are reported in Table 1 and Figure 1. It is worth mentioning that all the simulations were carried out with a temperature range between 283.15K - 373.15K. The general result line in Table 1 was calculated through the weighted average of experimental data.

Table 1. MAPE and RMSE for Peng-Robinson 78, GERG – 2008, and EOS – CG Equation of State.

General Data			**PR78**		**GERG-2008**		**EOS – CG**	
CO (%)	**P Range (MPa)**	**Exp. data number**	**MAPE (%)**	**RMSE (kg/m³)**	**MAPE (%)**	**RMSE (kg/m³)**	**MAPE (%)**	**RMSE (kg/m³)**
5.03	1.96 - 48.65	162	2.46	452.18	1.35	89.18	0.23	2.50
10.11	1.90 - 48.20	169	2.16	267.36	2.51	399.90	0.34	9.87
25.25	2.06 - 48.06	188	1.62	114.02	5.30	1506.19	0.23	4.56
50.18	2.03 - 48.17	187	0.92	16.28	3.55	245.12	0.27	1.60
General results:			1.76	202.43	1.76	582.20	0.27	4.58

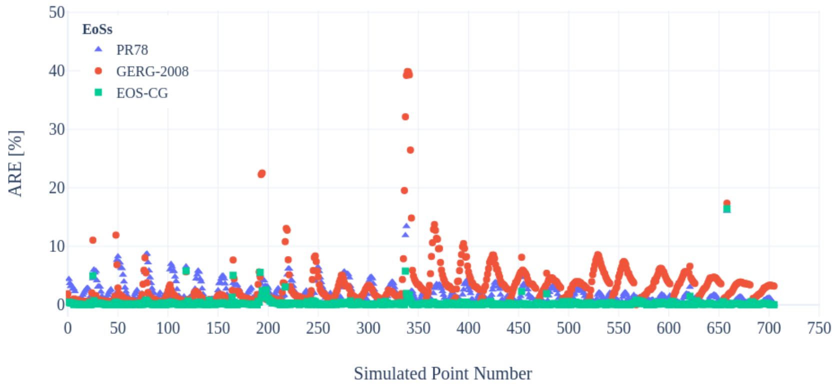


Figure 1. ARE analysis for PR78, GERG-2008, and EOS-CG according to the simulations performed.

As shown in Table 1, PR78 presented better suitability as the CO concentration increased, with MAPE less than 2% and RMSE approximately 200 kg/m³. In Figure 1, ARE has been decreasing from the initial points to the final points (lowest to highest concentrations of CO), and PR78 has only 3 points with ARE greater than 10%.

As for the accuracy of GERG-2008, it has a different behavior, which is not so efficient for higher concentrations of CO. Another consideration is that in the molar concentration of 25.25 % for CO, the error was considerably more prominent than in other situations. This occurrence contributes to the higher global value of errors for GERG-2008, with MAE less than 3.5% and RMSE of approximately 580 kg/m³, more than double the PR78. Some justifications for this happening are some evident outliers from the simulated data for GERG-2008 (Figure 2), which may have affected the error values, mainly for the RMSE. Another meaningful consideration is made by Kunz and Wagner (2012) about GERG-2008's few experimental data for binary mixtures with secondary components (the case of CO). Furthermore, for the CO_2 + CO system, the model uses the Lorentz-Berthelot combination rule without adjustment.

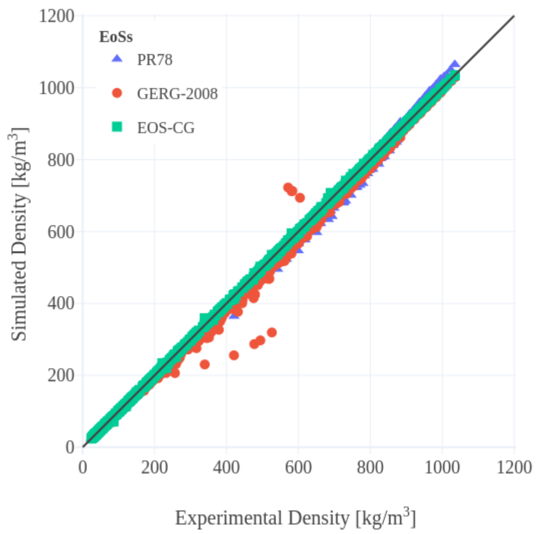


Figure 2. Cross plot between the experimental and simulated density for PR78, GERG-2008, and EOS-CG.

The GERG-2008 equation of state presented a greater ARE than the other models (Figure 1), where, at some points, they reach approximately 40%. Still, on GERG-2008, it was noted that the final points had a higher ARE than the initial ones.

Finally, EOS-CG presented the best fit to the system with MAPE less than 0.3% and RMSE close to 4.5 kg/m³. As shown in Figure 1 and Figure 2, the efficiency of the model was significantly better when compared to the others. This result was already expected, as the model development focuses on predicting the thermodynamic properties of components - CO_2-rich mixtures, humid and combustion gases - common in CCS processes, including CO as an impurity (Gernert and Span, 2016). That said, the EOS-CG was the model chosen to carry out the analyses of the CO influence on CO_2-rich mixtures when compared to pure CO_2.

4.2 Influence of CO on CO_2-rich mixtures

To evaluate the impact of the CO presence on CO_2-rich mixtures, one of the factors analyzed was the influence of different CO concentrations on the equilibria conditions on the phase envelope. The result is shown in Figure 3. The curves that consider the presence of carbon monoxide ($CO \neq 0$) were generated using the EOS-CG model, and the curve referring to pure carbon dioxide (Pure CO_2) was generated with the Spam & Wagner model.

Considering the curves with carbon monoxide, it was possible to verify that its presence causes the fluid to have a biphasic region. The higher the concentration of CO, the greater the biphasic region and, consequently, the greater the bubble pressure. This important finding is relevant to consider when planning CCS/CCUS processes. Usually, the transport of CO_2 is carried out with the fluid in a supercritical phase, when the fluid has the density of a liquid and the viscosity of a gas, according to Blanco et al. (2014). The authors also affirm that the transport of carbon dioxide in a two-phase flow can increase the minimum operating pressure because the last one must be greater than the bubble pressure, which also generates a higher cost.

The phase envelope of each case and Table 2 show that the critical pressure increases, making it necessary to operate at higher pressures to meet a supercritical state. During the construction of Table 2 and Figure 3, it was noted that critical point values are not calculated for the EoS-CG model at a CO concentration of 50.18 %. The same happens for the GERG-2008 model. The PR78 model calculated the critical point pressure and temperature values, P = 15.18 MPa and T = 241.71 K.

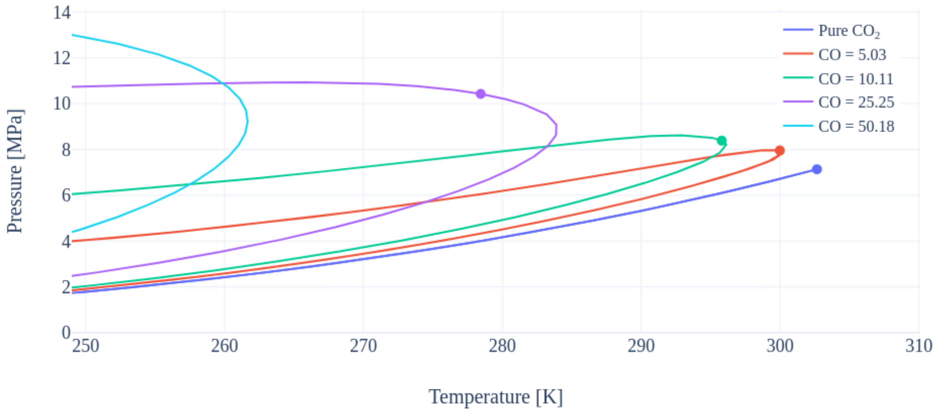


Figure 3. Phase envelope for Pure CO_2 and different CO concentrations in the system CO_2/CO using the model EOS-CG.

Table 2. Critical point according to CO concentration in the system CO_2/CO from EOS-CG.

CO (%)	P_c [MPa]	T_c [K]
0.0	7.13	302.66
5.03	7.95	299.98
10.11	8.38	295.80
25.25	10.42	278.45
50.18	15.18$^{(1)}$	241.71$^{(1)}$

$^{(1)}$Values calculated using PR78 model.

Modifying the critical point can impact changes in the fluid's thermodynamic properties, phase behavior (as already seen), and transport properties (coefficients of heat and mass). In addition, it can generate variations in the speed of sound in the fluid, affecting the compressibility of the fluid and flow velocity. According to Vitali et al. (2023), the bubble point line is one of the most relevant properties for predicting shear fracture in CO_2 pipelines.

Another relevant consideration for the performance of the process is the presence of a biphasic region under operating conditions. This implies a possible hindrance of the fluid flow due to the presence of flow patterns that consecutively interfere with the efficiency of the process. For example, the emergence of slug may cause pressure instability along pipelines, damaging the equipment.

Using the simulations to define the best equation of state for the CO_2 + CO system, Figure 4 was constructed, which compares the density behavior (considering the results generated by the EOS-CG) of pressure and temperature for each molar fraction studied. Thus, the presence of a two-phase region was also evident, and it was found that the density of the mixture was lower at higher temperatures, as expected.

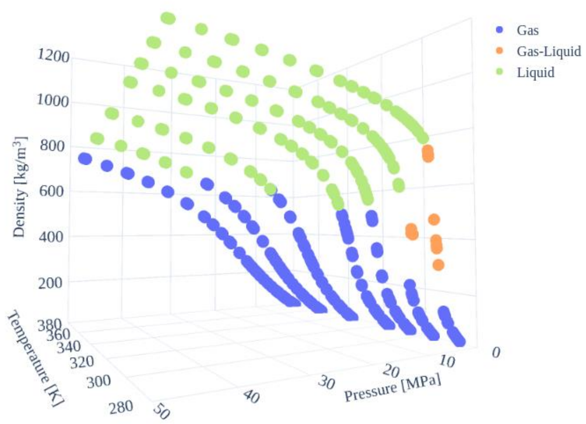
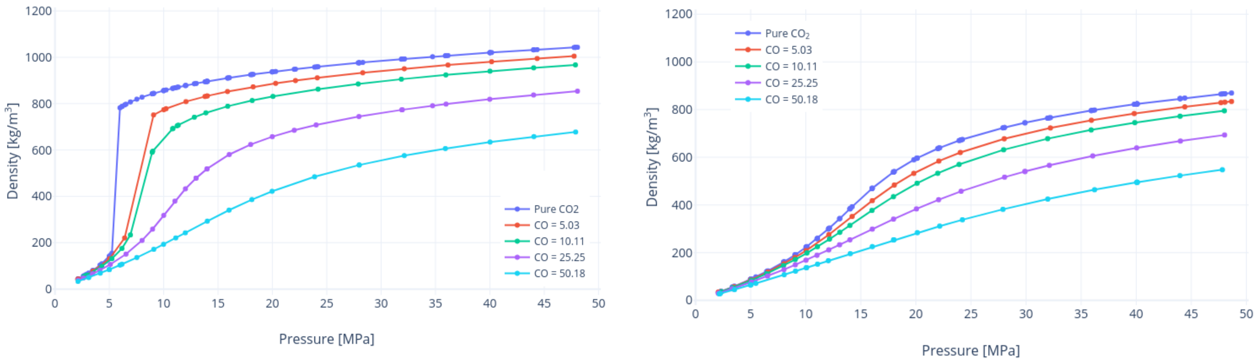


Figure 4. Density behavior at the function of pressure and temperature for CO concentration of 10.11% in the system CO_2/CO using the model EOS-CG.

In Figure 4, it was evident that in operating conditions close to the critical point of the mixture, the values for density change quite expressively, even with minor changes in pressure and temperature due to the phase transition. In Figure 5a, it is better to understand the density sensitivity close to critical conditions when compared to Figure 5b. Still, regarding Figure 5, it was found that the presence of CO in the mixture affects the density, and the higher the concentration of CO, the lower the density, affecting the PVT properties.



(a) T = 293.15 K (b) T = 353.15 K

Figure 5. Density behavior as a function of impurity concentration and temperature (a) 293.15 K and (b) 353.15 K.

One of the impacts of reducing the density in a horizontal flow is that it will increase the fluid flow velocity, resulting in a pressure drop in the CO_2 transport pipes and thus making the process more expensive since it is necessary, for example, to use compressors acting with high pressures along the pipeline (Onyebuchi, 2017).

This factor may directly imply the transport lines dimensioning, such as the size of the pipe, and the lower the density of the fluid, the greater its volume, also impacting the storage capacity of CO_2 in geological formations, causing the formations to become saturated more quickly and storing less CO_2 (Li and Yan, 2009).

5. CONCLUSION

This paper evaluated three equations of state (PR78, GERG-2008, and EOS-CG) for the prediction of CO_2-rich mixture phase behavior and density. The results showed that the EOS-CG is the best EoS among the others. This EoS returned a MAPE of 1.76% and an RMSE of 202.43 for a total of 706 points investigated. Furthermore, the PR78 performed better than the GERG-2008 overall.

The presence of CO makes it have a biphasic region in the phase envelope, and the higher the CO concentration, the greater this region. In addition, the critical point also increases, which implies the need to work with high pressures, which leads to higher costs.

Finally, it was possible to verify that it is necessary to consider the presence of impurities in the CCS processes, as they impact the sizing of pipes and pumps to the planning of the volume that is expected to be stored.

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