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**Response to Reviewers**

Dear Editor,

Thank you for allowing us to submit a revised draft of the manuscript “Fluid Property Characterization for Underground Gas Storage” for publication in the Journal of Clean Energy and Energy Storage. We appreciate the time and effort you and the reviewers dedicated to providing feedback on our manuscript and we are grateful for the insightful comments toward valuable improvements to our paper. We have incorporated all of the suggestions made by the reviewers. Those changes are highlighted within the manuscript.

Please see below, in blue, for a point-by-point response to the reviewers’ comments and concerns.

Best regards,

Dr. Marcos Vitor Barbosa Machado

Muhammad M. Alhotan, PhD Candidate

Dr. Gary Russell Jerauld

Prof. Mojdeh Delshad

Prof. Kamy Sepehrnoori

COMMENTS FROM REVIEWERS:

Reviewers' comments:

Reviewer #1: Structurally, I think it would be best to split the section "Collection of experimental fluid data" into separate parts for H2 and for CO2, and then to move this material into the individual results sections for H2 and CO2. There is some confusion in the report as it stands now because of the need to qualify whether CCUS or UHS is being talked about, and sometimes it is not clear from context. The title explicitly refers to aquifer storage. So I think the need to look at mixtures primarily concerns the UHS with cushion gas use case. Separating this out will improve reader comprehension.

Thanks for bringing up this recommendation. We have implemented that in the new version of the manuscript.

A strong argument is made for fluid models that are currently implemented in commercial (reservoir) simulators, i.e., PR/SRK and LBC/Pedersen. However, another possibility that fits within that same frame is to use a Black Oil formalism, at least for the pure component CO2 / H2 cases. You could use reference EOS (Span-Wagner for CO2, Lemmon for H2) to generate tables of gas properties. "Oil" table can then be replaced by the water phase, and you can model mutual solubility (gas-in-water via Rsw and also water-in-gas if desired). It would enable using the most accurate pure component properties, while still staying with functionality that exists in commercial simulators.

We acknowledge the comment, but as mentioned in the Introduction, our assumption was to use conventional equations of state and viscosity models commonly available in commercial and compositional simulators. Therefore, the black-oil approach is not considered in this manuscript. We have made this clear in our revised manuscript by explicitly stating it in our assumption list.

In this paper, our objective is to use the actual phase labeling and corresponding properties. We also are interested in mixtures where CO2 or H2 are stored in depleted gas reservoirs or different gases are used as cushion gas for Hydrogen. It is important to note that using the black-oil approach to represent the aqueous phase as an oil phase can lead to certain issues, including:

* Potential loss of accuracy in simulating the phase behavior transition of CO2.
* Possible impact on the computation of aqueous phase properties as a function of dissolved CO2.
* Inability to model CO2 diffusivity in the aqueous phase if it is represented as an oil phase.
* The density and diffusion factors significantly influence CO2 solubility, thus the alternative approach may affect CO2 entrapment.

Considering our objective to propose a generic approach applicable to a wide range of simulation cases without imposing any restrictions on the simulation model, we have chosen to focus on a compositional approach instead.

In Table 1, please clarify for the binary/ternary mixtures what compositions we are talking about - are they fixed mixtures (which?) or ranges?

The composition of the mixture varies, and this information is provided in detail as depicted in the data. To further clarify this point, we have included a sentence after Table 1 to explicitly state the composition variations.

In 3.2 focusing on JST is OK but I think it would be valuable to discuss Pedersen/CSP also. After all, it has a bit more physics, is very popular, and does offer some tuning parameters also. Readers will be interested in how they compare for these applications. Possibly Pedersen will not work for H2 due to choice of ref fluid? At a minimum add a short exploration of this.

Thank you for bringing up this excellent point. Initially, we utilized the Pedersen CSP model for this study. However, due to the extensive and diverse dataset that we needed to accommodate, it became challenging to reconcile it with a model that had limited degrees of freedom, such as Pedersen's model. We found that the JST model offered greater flexibility in representing the various combinations of pressure, temperature, and composition that we employed. We have discussed this decision and its implications after Figure 9. We appreciate your observation and input on this matter.

In results sections, I would like to see a little more analysis of these results as you are using a large number of data sources. Please include some scatter plots (measured vs simulated) over P,T, composition...and report on average + variance of error in any format you chose.

Instead of utilizing scatter plots, we have now presented the errors in two new tables (Tables 6 and 9 in the revised manuscript). These tables summarize the errors for each property (density, viscosity, and solubility) in both UHS and CCS applications, considering pure components and mixtures. The small spreading observed in the results allows us to rely on the average error across different temperatures over the pressure range as a representative measure for evaluating the quality of the fitting methodology. This approach helps to provide a concise and informative assessment of the fitting methodology's performance.

In Fig 8, is temperature actually 451.55 deg C as stated? This would not be relevant, of course, for practical subsurface application.

Indeed, we acknowledge that the temperature range in this dataset is relatively high and may not be representative of UHS applications. We included this dataset solely for the purpose of comparing it with the predictions of our calibrated model, which, as expected, exhibited significant deviations as discussed after Figure 9. This comparison helped to highlight the limitations and challenges in accurately predicting properties at elevated temperatures. We appreciate your understanding of this context and the discussion surrounding it.

Thanks for your constructive suggestions. We are positive that your recommendations allow us to highlight the scientific aspects of our work, and we incorporated 100% of them.

Reviewer #2: Thank you for reports this important work on the PVT characterisation for H2 and CO2 gases using the PR-EoS while using the experimental data. Enjoyed reviewing your work. I have following comments which will further improve the quality of the paper.

Thanks for your constructive suggestions. We are positive that your recommendations allow us to highlight the scientific aspects of our work, and we incorporated 100% of them.

Comments:

\* Assumptions states that the pure H2 or CO2 streams are injected in the tested numerical models. However, section 2 (collection of experimental data) states, "The data set includes density, viscosity, and solubility in water and brine for pure component gas and gas mixtures". This needs clarity.

Thank you for pointing that out. We apologize for the oversight. In the UHS cases, mixtures of H2 and other gases were indeed considered, while in the CCS cases, only pure CO2 was taken into account. We have made the necessary corrections in our assumption list to clarify this distinction.

\* Add equation of the Li and Ngheim model in the manuscript itself perhaps, after equation 8 to complete the PVT modeling aspects.

We have included the equation (9).

\* Comment on the application of the PR-EoS against the SRK-EoS for hydrogen gas and its mixtures (ref: section 3.1). this justification must be stated in this section

Thank you for bringing up this point. In our revised manuscript, we have discussed the application of the EoS-SRK for H2 in section 3.1.4. We acknowledge that some authors argue that SRK may be more accurate than PR in calculating H2 solubility in water (and brine). However, our findings indicate that both EoS models can achieve a similar level of accuracy when coupled with Li and Nghiem's model, provided that certain tuning steps are performed. We have addressed this observation and the associated discussion in the revised manuscript to provide a comprehensive analysis of the H2 solubility calculations. We appreciate your input and the opportunity to clarify this aspect.

\* How these properties will be reliable at the upscaled P& T reservoir conditions?

When formulating a fluid model, it is crucial to ensure that it accurately represents a reference dataset. In particular, PVT experimental data is often considered the most reliable and accurate dataset for this purpose.

In numerical simulations of flow in porous media, primary variables such as pressure, composition, and temperature are typically calculated and can vary depending on the size of the gridblocks used in the model. However, it is important to develop a suitable upscaled model that can effectively represent these properties at coarser scales. The calibrated fluid model is then used to compute the fluid properties based on the results obtained for pressure, temperature, and composition in a suitable scale.

Therefore, if a coherent upscaled model is available, the same fluid model can be used across different scales, ensuring consistency and accuracy in representing the fluid behavior. This approach helps to maintain the integrity and reliability of the fluid model throughout various scales of the simulation.

\* Figure 10: 8 to 12% error in the pressures ranging from 25 to 35 kPa is observed in Figure 10. Is this acceptable and why?

As mentioned in the manuscript, the high deviations observed in our results are primarily associated with the high-temperature datasets, which are not representative of UHS targets. We specifically addressed this issue and provided a comment after Figure 9 to emphasize the discrepancy between the high-temperature dataset and the intended UHS applications.

\* Have you compared your result against the results reported by Ali Hassanpouryouzband? Please refer to article in "Thermodynamic and transport properties of hydrogen containing streams, Ali Hassanpouryouzband, E Joonaki, K Edlmann, N Heinemann, J Yang, Scientific Data 7 (1), 1-14"

There are three distinct disparities between our study and the mentioned paper:

* Coverage of thermal properties: The referenced work includes thermal properties, such as thermal conductivity, which are beyond the scope of our study.
* Focus on transportation versus storage: The focus of the referenced work is on the surface transportation of H2-rich streams, where the temperatures are expected to be much lower compared to the higher temperatures considered in our study for storage purposes.
* Reservoir type and gas composition: The data and results presented in the referenced work are more relevant to hydrogen storage in depleted gas reservoirs, whereas our study focuses on UHS in aquifers. Consequently, the mixtures evaluated in the referenced work are likely to be richer in gaseous hydrocarbons compared to our study, where the presence of CH4 as a cushion gas results in lower concentrations of gaseous hydrocarbons. We have inserted this comment in our Conclusion with the reference to this paper.

\* It will be beneficial for the readers to carry forward and implement you finding if you can release the output results in the form of tables. Hence this is recommended.

It is indeed valuable to provide methodological steps for calibrating models and the corresponding tuned parameters. We understand that all the outputs, including the tuned parameters, are displayed in table formats within the paper. This allows readers to access and utilize the calibrated parameters (Tables 2, 3, 4, and 8 in the revised manuscript) for the cited models.

Additionally, we decided not to reproduce the data used from other authors in a table format, as they are already cited in your manuscript. This approach maintains the integrity of the sources and acknowledges the contributions of the original authors.

\* Comment on the applicability of your PVT work for oil and gas reservoirs at varying pressure and temperature conditions.

The methodology we have adopted draws inspiration from the classic workflow used in PVT simulation for characterizing oil and gas mixtures. However, when applying this methodology to CCS or UHS in depleted hydrocarbon reservoirs, it becomes crucial to have a more extensive dataset that accurately represents the mixtures between the injection gas and the in-situ hydrocarbon components.

In order to achieve a better representation of these mixtures, it is necessary to provide a more complete dataset. By incorporating this additional data, the same steps can still be followed, ensuring the applicability of the methodology. We have included this important comment in the Conclusions section, highlighting the significance of obtaining a more comprehensive dataset for successful implementation in CCS or UHS applications.

Corrections:

1. Remove footnotes on page 1

We fixed it.

2. Page 3: it seems like verb is missing in the sentence, "The innovative aspect of this work is the accurate correlation of experimental…..! Perhaps add wording "….is development of…..accurate…"

We fixed it.

3. Maintain consistency in stating units in equations. Equation 7 states units for the terms (gas viscosity) whereas the other equations don't.

We fixed it.

4. Nomenclature: unit of volume is stated as L. Is it litre, consistent throughout the manuscript?

It is common practice to use liters (L) as the volume unit when referring to the partial molar volume at infinite dilution in the solubility model. This convention allows for consistency and comparability with other studies in the literature.

However, for volumetric properties such as density, the more commonly used unit is cubic meters (m³). This unit is widely recognized and facilitates easier comparison and integration with other literature sources that employ the same unit.

5. Letters in Figure 1 workflow are not visible

We improved the quality of the figure.