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Murad S. Abuaisha, Ahmed Rouabhi, Faouzi Hadj Hassen, David W Eaton, Firas Tayeb, et al.. Thermal effects on the kinetics of gas dissolution in brine: A case study of CO₂ storage in salt caverns (a non-dimensional model). DECOVALEX 2023, Nov 2023, Troyes, France. hal-04297315

HAL Id: hal-04297315

<https://hal.science/hal-04297315v1>

Submitted on 21 Nov 2023

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Thermal effects on the kinetics of gas dissolution in brine: A case study of CO₂ storage in salt caverns (a non-dimensional model)

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Abstract

Understanding the dissolution kinetics of CO₂ in brine is important for efficient management of cycled CO₂ in underground salt caverns.

The kinetics is strongly influenced by natural convection, which is caused by density changes related to the concentration of dissolved gas as well as thermal changes driven by CO₂ cycling and the effects of the geothermal gradient. Based on a novel non-dimensional model that couples cavern thermodynamics with the dissolution mechanisms, this study investigates how the natural convection related to mass and thermal changes impacts dissolution kinetics. The numerical model is validated using laboratory measurements, and thereupon upscaled to typical dimensions of a salt cavern. Although our simulations show that the total dissolved mass is relatively minor compared to the initial stored mass/cycled mass, it is nevertheless important to quantify other loss phenomena, particularly gas permeation into the host rock salt. The developed model can also be adapted easily to study the kinetics of dissolution of other gases, such as hydrogen and methane.

Introduction and Methodology

One promising clean energy resource is methanation, a technique used to convert carbon dioxide to methane using hydrogen. Methanation is supposed to reply to the energy fluctuating demand, which necessitates large storage of its production elements (H₂ and CO₂). This study focuses on CO₂ storage and cycling in salt caverns in the context of clean energy transition [1].

The underground storage system can be divided into three domains: (1) the well which extends from the ground surface to the cavern; (2) the cavern itself; and (3) the rock mass that surrounds the cavern and the well (Fig.1).

The cavern brine has initially low concentration of CO₂. As CO₂ is injected into the cavern, a concentration gradient develops at the brine interface. This concentration gradient forms the driving force for the Fickian diffusion.

Consequently, convection is created in the brine due to changes in the density field. A thermal convection is further developed due to cycling temperature changes at the brine interface, which interacts with the geothermal gradient of the brine. The velocity field that emerges due to both types of convection is expected to enhance the dissolution of CO₂ in the brine.

This research introduces a non-dimensional model that couples cavern thermodynamics [2] with the dissolution mechanisms in the brine. Since the model is non-dimensional, once validated by the laboratory measurements, it can be applied to other scales. This model can also be adapted to study the dissolution of other gases. Experimental work is required, some of it considered in this study, in order to establish the key model parameters.

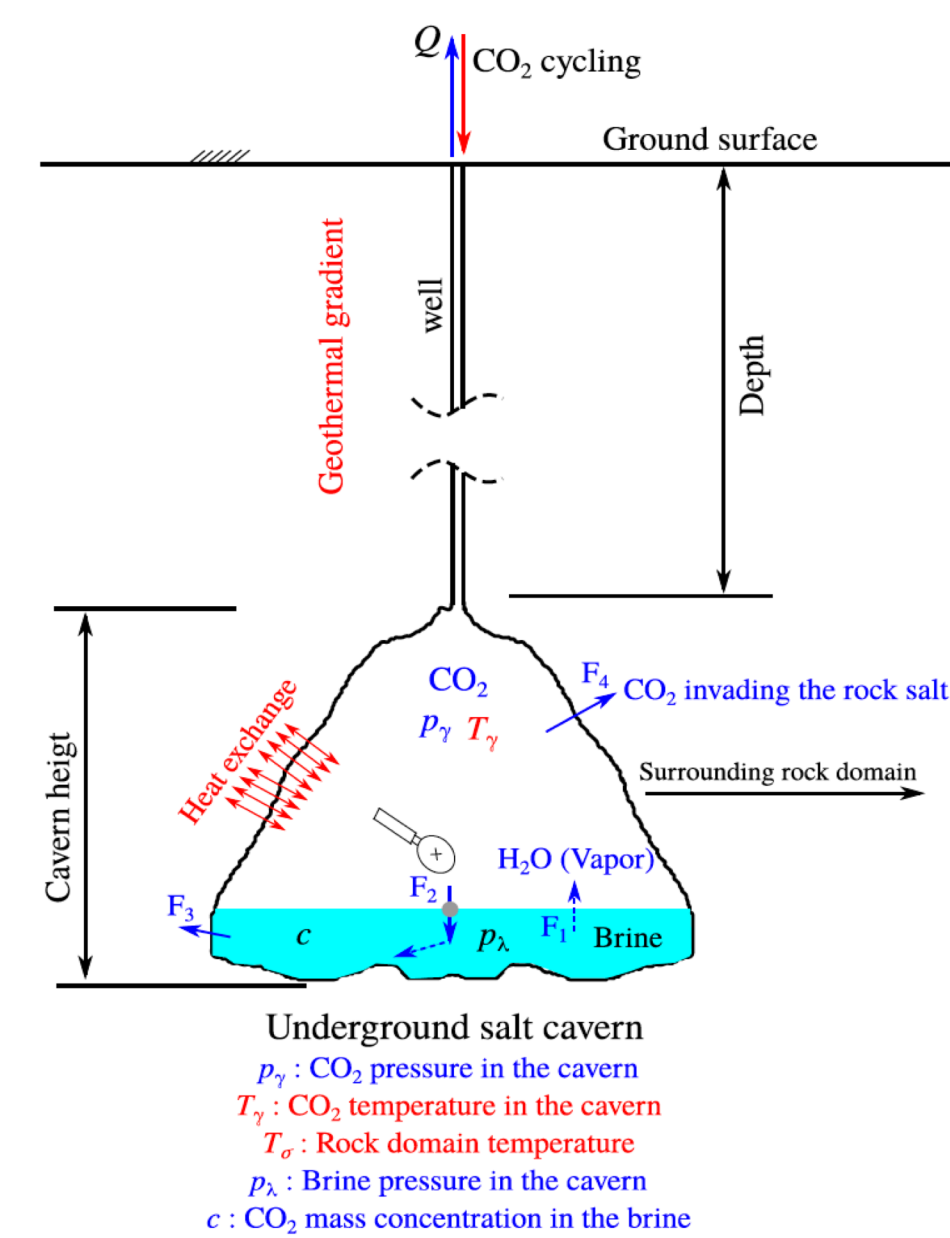


Fig. 1. Schematic representation of CO₂ storage in salt caverns and its transport mechanisms between the cavern phases

Methodology and Laboratory Work

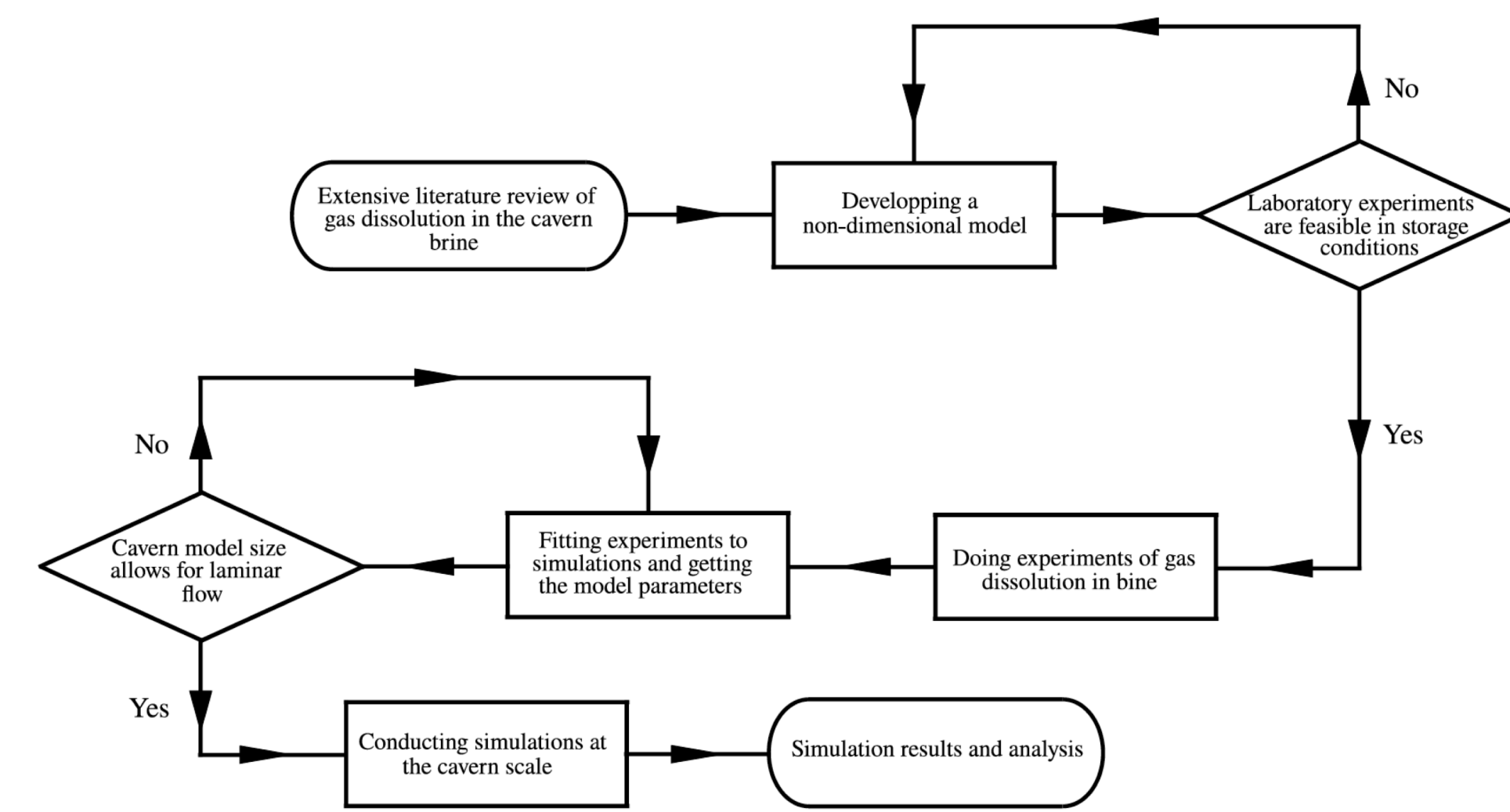


Fig. 2. The workflow of this study

Before applying the non-dimensional model on the cavern scale, experimental validation is needed. Laboratory experiments are performed based on a pressure decay test using a PVT (Pressure-Volume-Temperature) cell. In this setup, the CO₂ gas is injected from a tank into the cell that is partially filled with saturated brine. The cell temperature is controlled by placing it into a temperature-controlled basin (Fig. 3). Once gas is injected, it starts dissolving in the liquid phase.

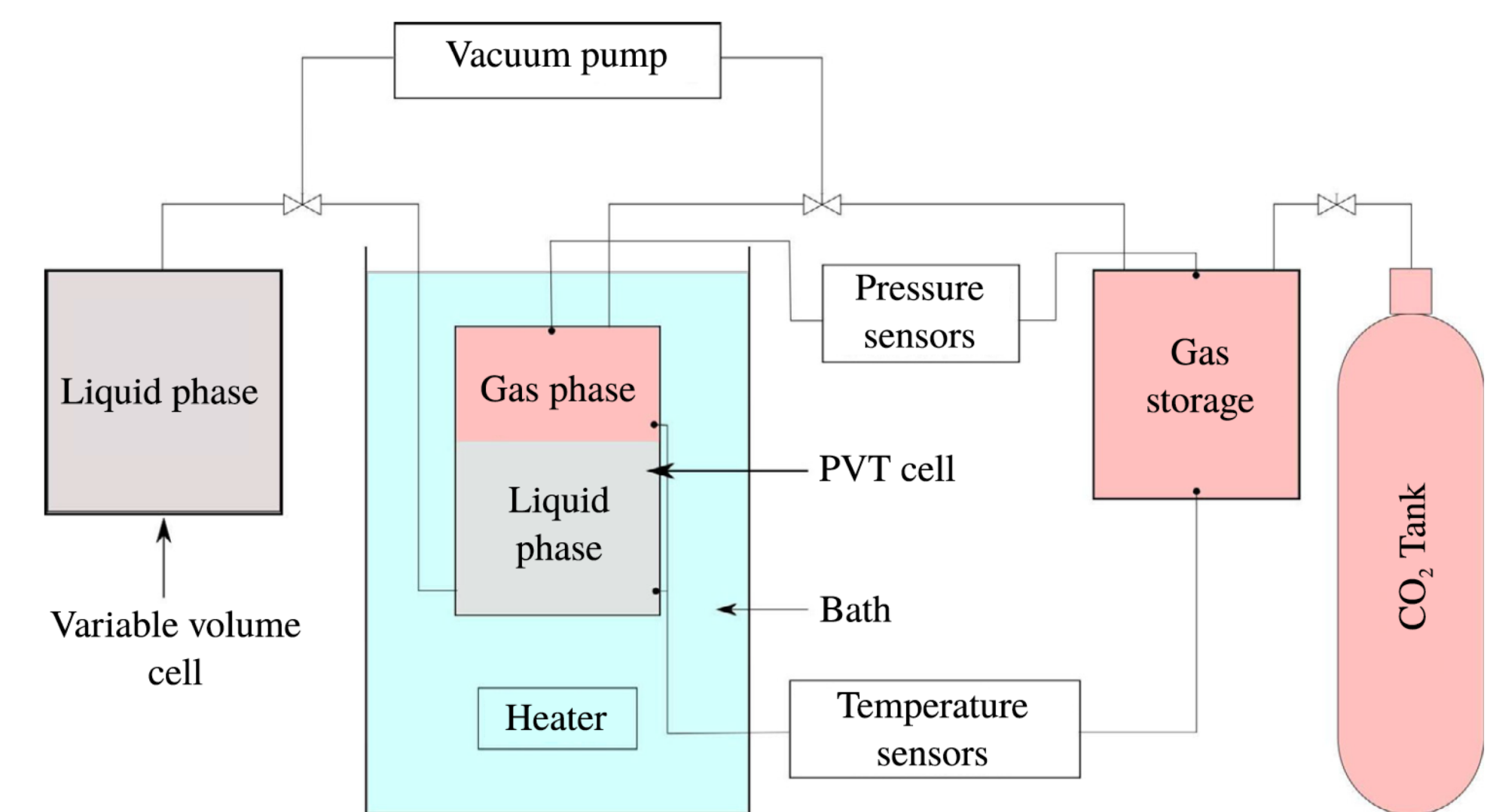


Fig. 3. Schematic representation of the pressure decay PVT cell used to study the CO₂ dissolution in the brine and to validate the mathematical model

Validation – Laboratory Scale

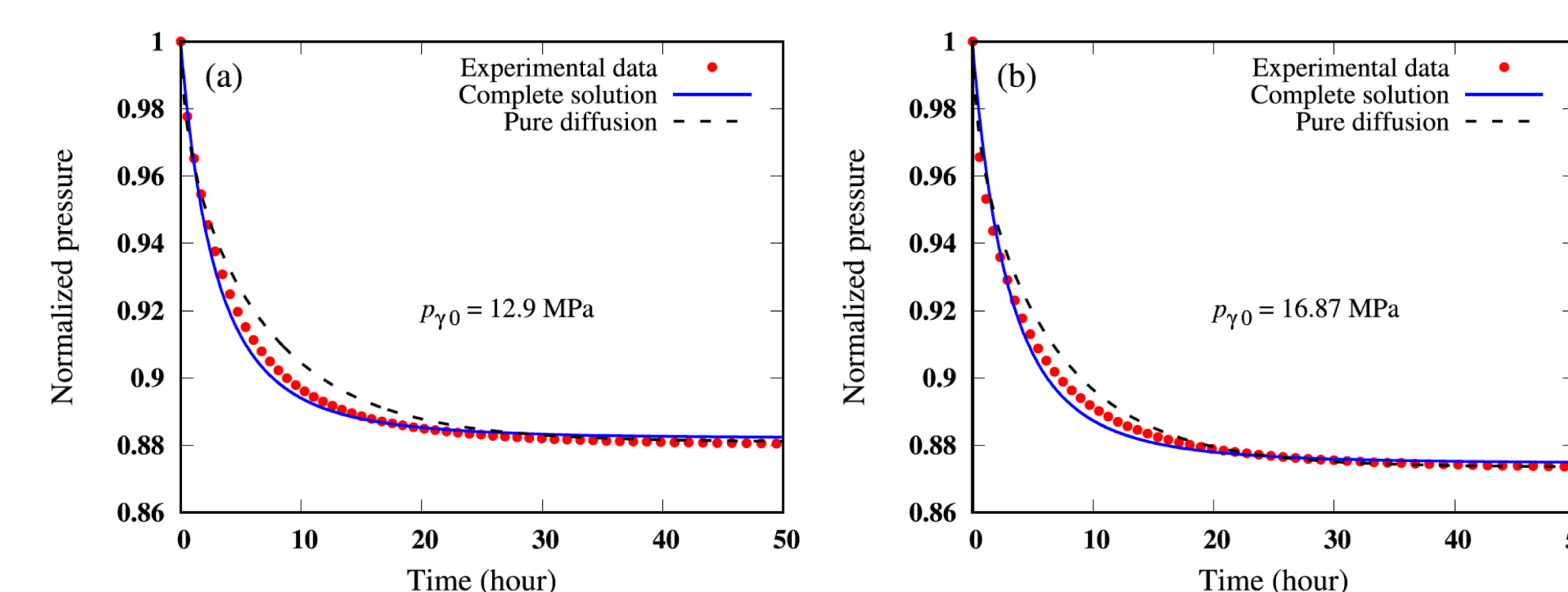


Fig. 4. A comparison between the numerical and the experimental pressure histories for the two laboratory tests: (a) $p_{y0} = 12.9$ MPa and (b) $p_{y0} = 16.87$ MPa. The black curves represent the numerical solutions while considering only pure diffusion ($D_{app} = 8.6 \times 10^{-8}$ m²/s)

The results of the numerical model correlate well to the experimental data. The black curves represent the numerical solutions while considering only the diffusion phenomenon. For these solutions to correlate to the experimental data, the diffusion coefficient needs to be enhanced.

Thermal Effects

Thermal effects are quantified numerically. Fig. 5 shows the numerical model that is used for this purpose. This model represents a cylinder with a radius that is 3 times the radius of the laboratory cell. Nevertheless, the same height-radius ratio is adopted. In the first scenario, a temperature difference of 3 °C is applied over the model height. In the second scenario, the difference is reduced to 0.1 °C.

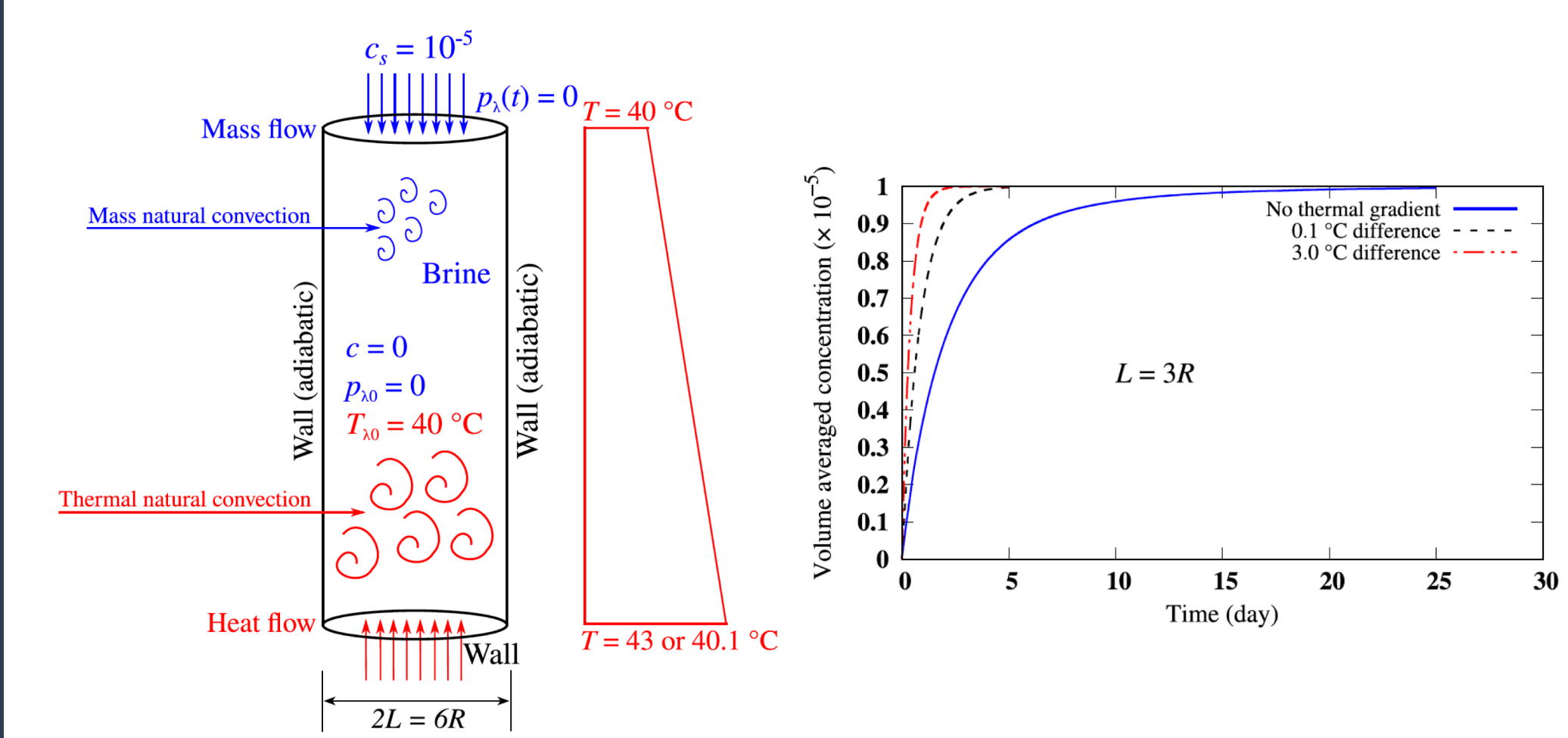


Fig. 5. Schematic representation of the numerical model. Evolution of the volume averaged CO₂ concentration. The figure shows how the thermal effects due to different temperature gradients accelerate the dissolution process.

Fig. 5 shows how even a small temperature difference of 0.1 °C accelerates the dissolution process.

Simulations – Cavern Scale I

To consider the full cavern scale, the numerical model represents a cylindrical volume ~42412 m³ in an infinite rock salt domain (Fig. 6). The cavern brine occupies almost 11% of the volume over a height of 1.5 m.

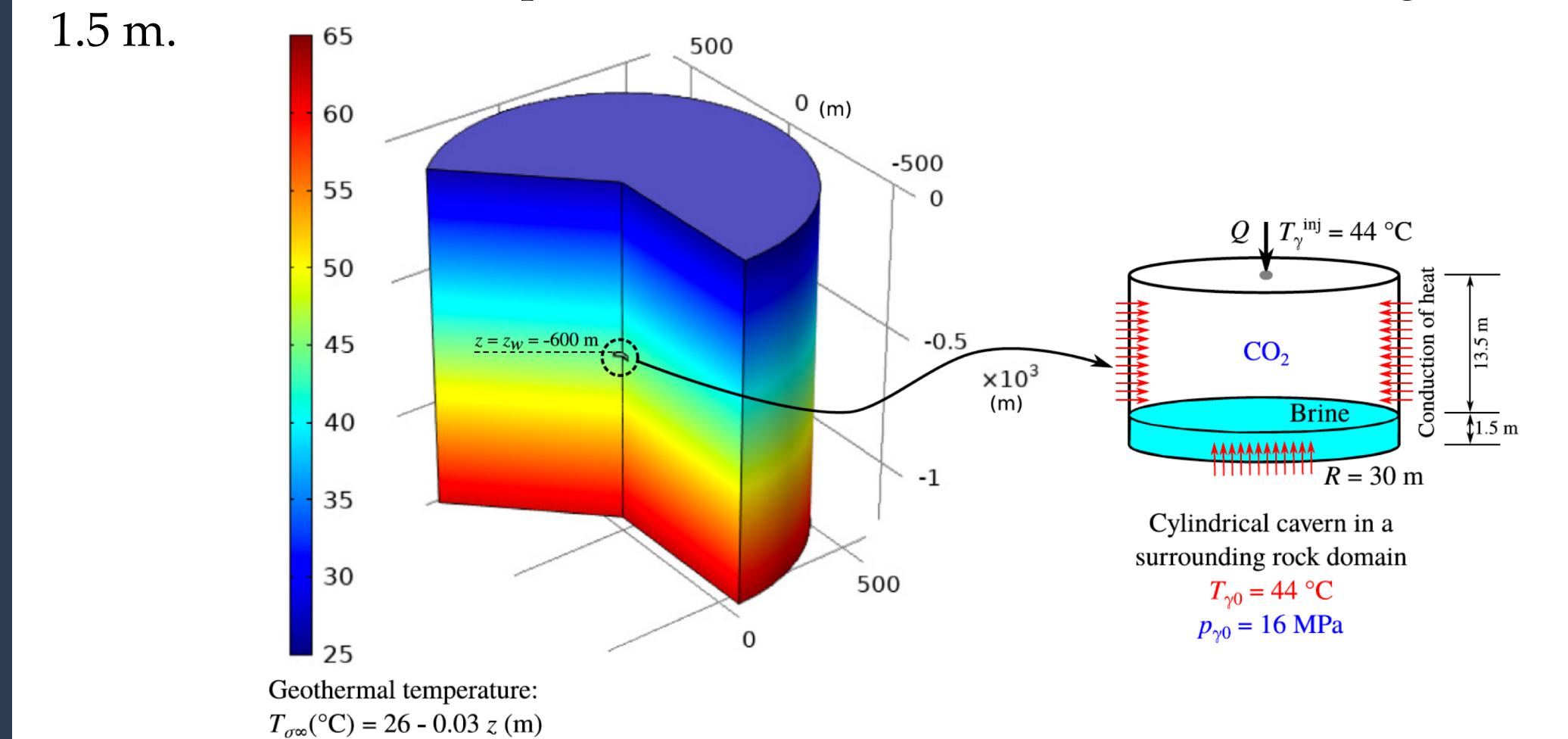


Fig. 6. Schematic diagram of the boundary value problem: it represents a cylindrical cavern created at a depth of 600 m in a surrounding rock salt domain

The cycling program envisaged for this research: a weekly utilization of the cavern with four cycles spreading over ~28 days. Cycling leads to reasonable mass variations where the cycled CO₂ stays always in its supercritical state.

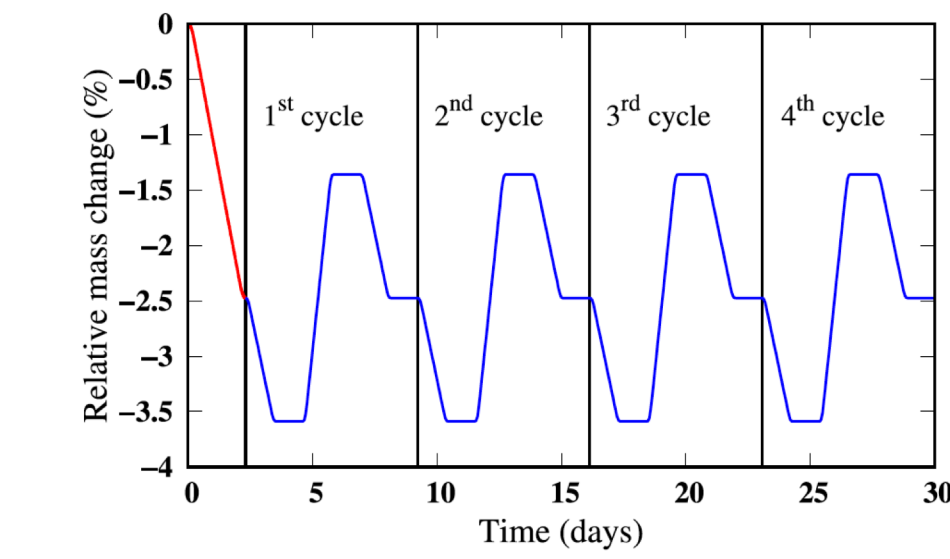


Fig. 7. Cycling program

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Simulations – Cavern Scale II

Fig. 8 shows the cavern thermodynamics as well as the brine CO₂ volume averaged concentration as function of cycling. Fig. 8(c) shows how the brine CO₂ concentration increases linearly due to cycling.

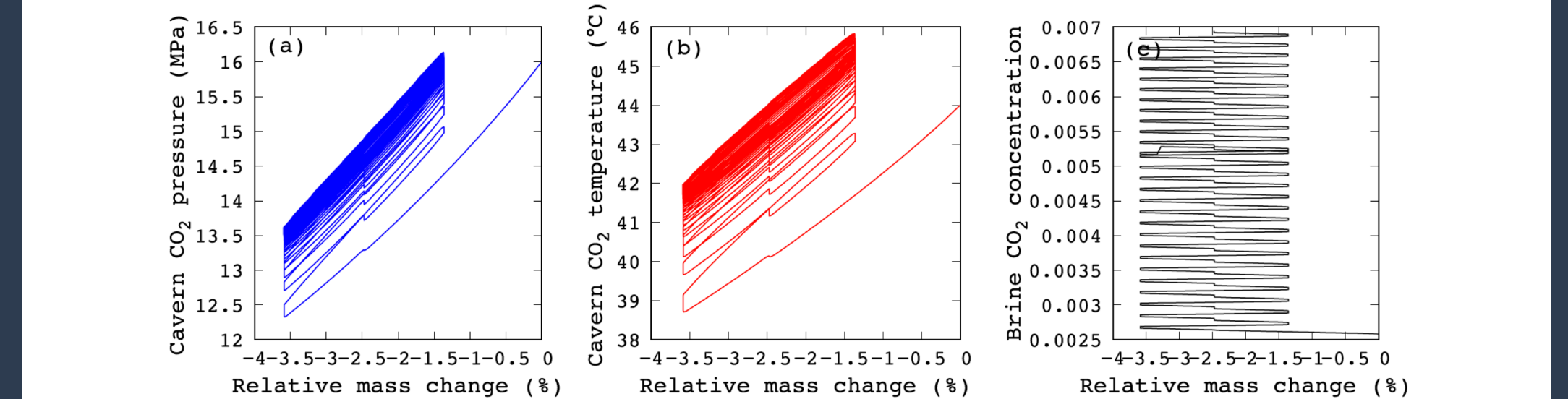


Fig. 8. Cavern CO₂ pressure (a) and temperature (b) as function of the relative mass variations during 6.5 months

Fig. 9(a) shows how the brine volume averaged temperature is impacted by cycling. Fig. 9(b) shows the brine CO₂ concentration averaged over its surface and volume.

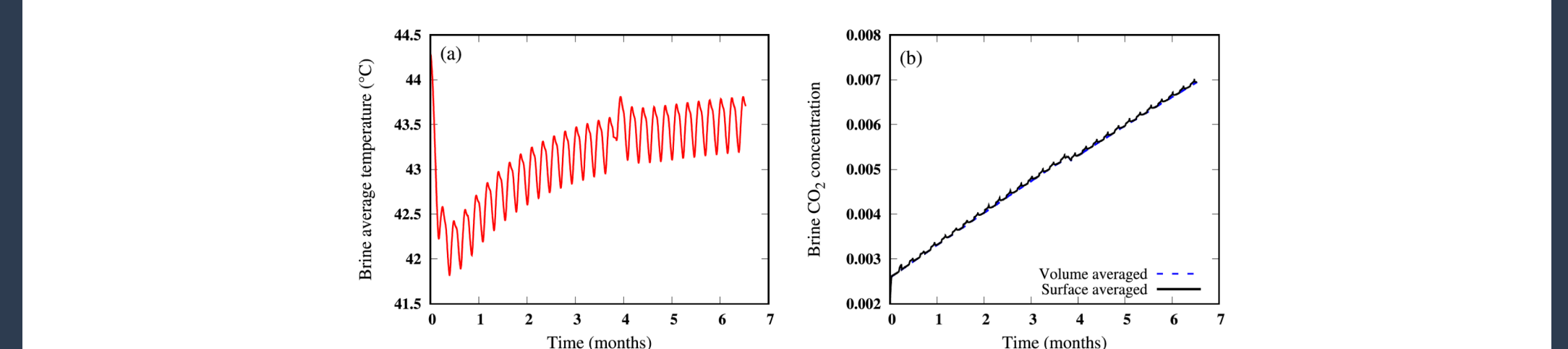


Fig. 9. The evolution of the brine average temperature during the CO₂ cycling. (a) The brine CO₂ concentration averaged over its surface and volume. The surface averaged concentration shows clearly the effects of cycling

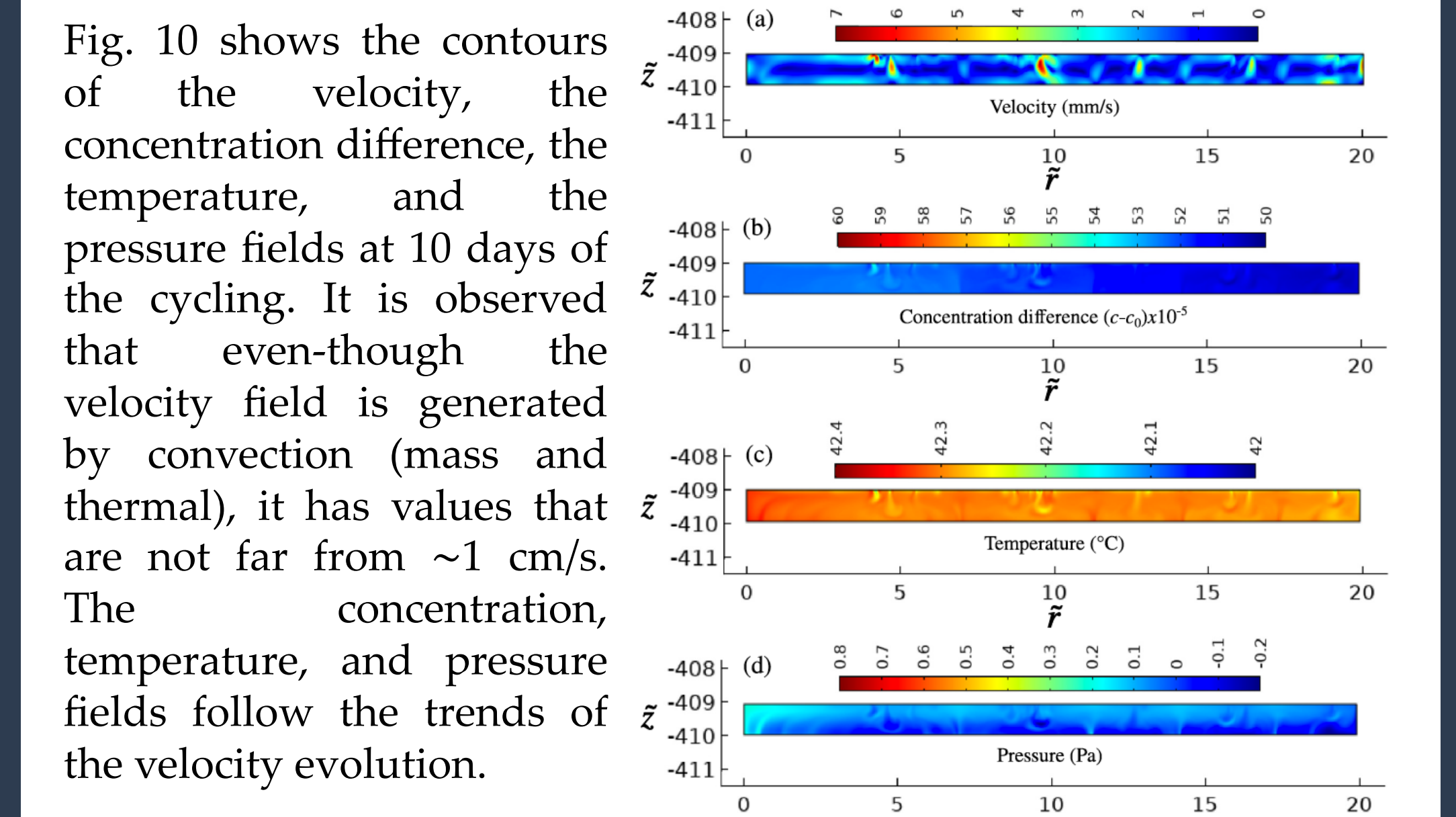


Fig. 10. Contours at 10 days of weekly cycling

Conclusion

The novelty of this research stems from the fact that it presents and quantifies the effects of thermal convection on the gas-liquid dissolution process. Simulations demonstrate that the ultimate total dissolved mass is approximately 0.32% of the initial stored mass. Although this loss may seem insignificant, the quantification of this loss, during cycling becomes important when other loss mechanisms are studied, particularly the permeation of gas into the rock domain.

Acknowledgements

Authors would like to thank the French Association Nationale de Recherche (ANR) for their financial support via the project HyTrend.