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Hydrogen migration in rock salt during cycling in underground caverns

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Abstract
For good management and precise tracks of hydrogen quantities stored in salt caverns, this abstract presents a study on hydrogen transport in rock salt during cycling. It provides a novel mathematical-numerical model that couples the cavern thermodynamics with the transport mechanisms of hydrogen in the saturated rock salt in a fully coupled thermo-hydraulic framework. Both the two-phase Darcian percolation and the Fickian diffusion are used to account for hydrogen migration in the interstitial brine of the rock salt. Due to the absence of experimental data, a parametric study is furnished. The effect of cycling within the cavern on the migration mechanisms is discussed in detail. Simulations have confirmed the dependency of the Darcian percolation on the model parameters. However, for similar applications, this dependency might be limited. The value of the Fickian diffusion coefficient affects indirectly the Darcian percolation. The two-phase percolation becomes more of a piston-like for very small values of the diffusion coefficient. On a real-scale typical cavern, and over a period of 40 years, simulations have proven that the quantity of hydrogen lost into the surrounding rock salt is slight. Besides, cycling renders this quantity more unimportant.

1. Introduction

During cavern operation, stored hydrogen undergoes changes in its temperature and pressure. Such changes are expected to affect its migration into the surrounding rock domain. Since rock salt contains interstitial brine and are characterized by extremely low permeabilities and porosities, the application of one-phase generalized Darcian flow to describe hydrogen transport is questioned [1]. Moreover, none of the available literature is addressing the effect of cavern cycling on the migration mechanisms. Indeed, the cavern thermodynamic state is function of cycling. Consequently, in the cavern vicinity, both the interstitial rock salt brine pressure and temperature evolutions depend on cycling as well. These changes in the interstitial brine pressure and temperature must influence the migration of hydrogen into the rock domain. The novelty of this research stems from providing a mathematical-numerical model that couples the cavern thermodynamics with the non-isothermal transport mechanisms of hydrogen into the rock salt. Both the Darcian two-phase percolation and the Fickian diffusion are considered as well as the interaction between them. This model addresses as much as possible of the problem complex physics for good estimation of the exchanged hydrogen mass. Besides, it precisely presents the effect of cycling.

Due to the absence of any literature or experimental data treating hydrogen migration in saturated rock salt, we offered a parametric study that investigated the effect of the two-phase van Genuchten model parameters as well as the Fickian diffusion coefficient on the total mass of the exchanged hydrogen. The van Genuchten model allows to describe the two-phase percolation of hydrogen (characterized by very low viscosity) in rock salt (characterized by very small pore size). Eventually, an overestimating scenario of the model parameters, of the diffusion coefficient, and of boundary conditions, was considered to estimate the amount of hydrogen migrated during a 40-year period of hydrogen cycling in a typical spherical cavern.

2. Hydrogen migration in rock salt

Figure 1 shows the mechanisms related to hydrogen transport in the saturated rock salt while assuming a van Genuchten two-phase percolation. When hydrogen pressure within the cavern exceeds the rock salt pore pressure plus the entry pressure, hydrogen invades the rock salt in a two phase percolation type flow. However, when hydrogen pressure becomes less than the interstitial brine pressure, brine moves towards the cavern and tends to pour down the cavern wall. Fickian diffusion of hydrogen in the rock salt interstitial brine is function of hydrogen cycling. This diffusion is related to the hydrogen mass concentration gradient between the cavern and the rock salt domain. The temperature changes within the cavern lead to temperature changes in the rock salt domain. In this study, the rock salt, the interstitial brine, and the invading hydrogen are assumed to be in local thermal equilibrium. Besides, a temperature continuity is assumed on the cavern wall. The thermo-hydraulic coupling in the rock domain is accounted for through the pressure and temperature effects on the hydrogen and the brine densities.

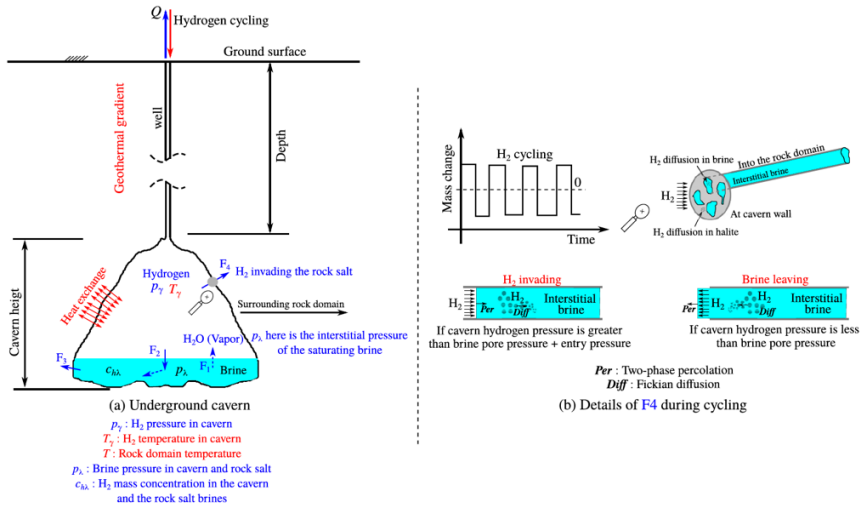


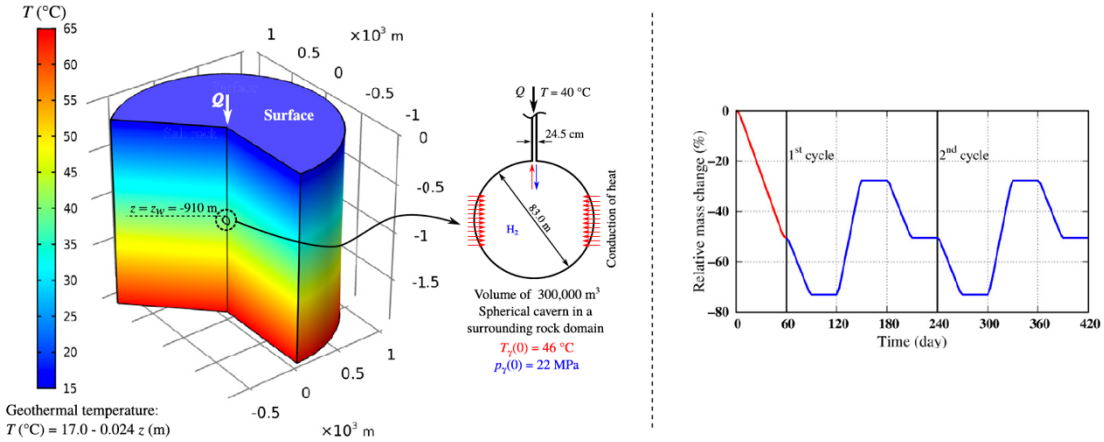
Figure 1: Schematic representation of hydrogen storage in salt caverns and its transport mechanisms between phases. (b) Transport of hydrogen into the rock salt domain (flux F_4).

To simplify the problem, the following assumptions are made. Some assumptions are adopted for an overestimating study:

1. The underground cavern is assumed to be filled with a mono-component single-phase hydrogen;
2. Hydrogen cycling is seasonal/slow, consequently, a uniform thermodynamic state is assumed within the cavern [2];
3. The rock salt domain is assumed saturated with brine;
4. For an overestimating study, brine pressure within the rock salt domain is assumed halmostatic;
5. For an overestimating study, rock salt creep due to cavern operation is neglected;
6. Hydrogen diffusion in the halite structure is neglected;
7. Hydrogen concentration is initially disregarded in the rock salt domain;
8. The rock salt, the interstitial brine, and the invading hydrogen are assumed in thermal equilibrium;
9. Hydrogen invasion into the rock salt domain is assumed to follow the van Genuchten percolation model [3];
10. For an overestimating study, hydrogen entry pressure into the saturated rock salt is neglected.

3. Results of the simulations

The numerical model represents a spherical cavern of volume 300,000 m^3 in a surrounding rock domain. The cavern has been leached (full of brine) and is initially in thermal equilibrium with the surrounding rock domain at 40 °C. The brine pressure within the cavern is also in equilibrium with the rock salt pore pressure at the halmostatic value of 11.2 MPa. The cavern brine is then replaced by hydrogen during a filling phase of 90 days where hydrogen pressure of 22 MPa is attained within the cavern. The cavern is later left unsolicited for a similar period of time. Consequently, after 90 days of standstill, the cavern volume averaged temperature and pressure are 46 °C and 22 MPa respectively. The cavern then undergoes seasonal cycling following the program shown in Figure 2(b) for a 40-year time period.



(a) Typical spherical cavern (b) Imposed mass variations (cycling)

Figure 2: (a) Schematic diagram of the boundary value problem: it represents a spherical cavern created at 910 m depth in a surrounding rock salt domain. (b) Imposed cavern relative mass variations; only the first two cycles are shown.

Figure 3(a, b) shows the cavern temperature and pressure as functions of cycling. Hydrogen pressure is not showing any changes during cycling as the cavern volume is assumed constant. However, temperature is showing some changes due to the energy exchanged with the surrounding rock salt domain. Figure 3(c) shows the hydrogen pseudo-capillary pressure averaged over the cavern surface during cycling. The capillary pressure does not exceed 2 MPa for the working and boundary conditions of this typical spherical cavern. Hydrogen invades the rock domain when the capillary pressure is positive.

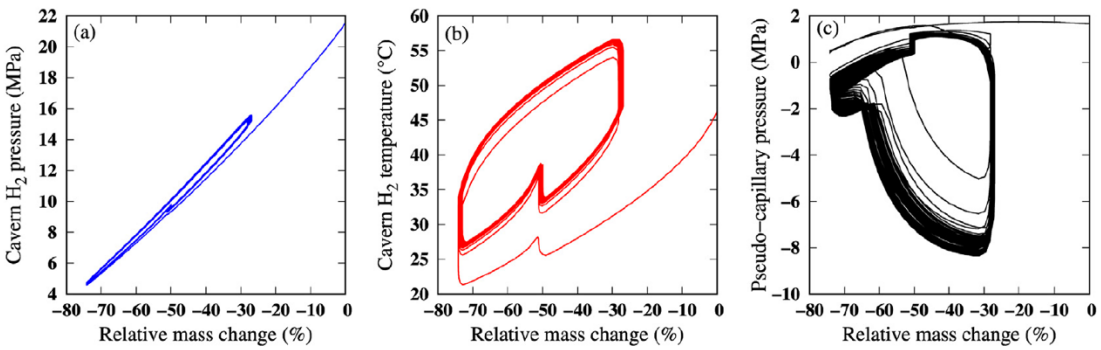


Figure 3: (a) cavern pressure, (b) cavern volume averaged temperature, and (c) hydrogen pseudo-capillary pressure averaged over the cavern surface. The three quantities are presented as functions of cycling/relative mass changes within the cavern.

Figure 4 shows the hydrogen mass exchanged with the surrounding rock domain due to the two-phase percolation and the Fickian diffusion. Figure 4(a) is a zoom-in of Figure 4(b) for the first four cycles/800 days. Both the percolated and diffused hydrogen masses tend to reduce during withdrawal. Even-though the Fickian diffused mass is initially smaller than the percolated mass, after ~25 years of cycling, the diffused mass becomes more significant than the two-phase percolated mass (Figure 4(b)). Eventually, the percolated and the diffused hydrogen masses sum up to ~93 kg after 40 years of the cavern operation.

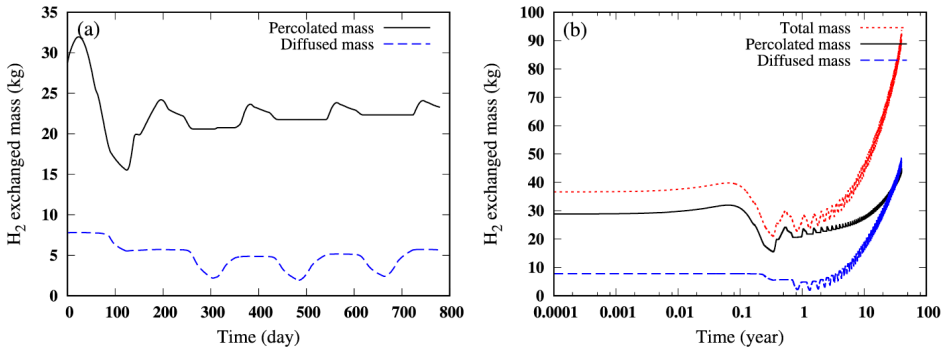


Figure 4: Darcian percolated and Fickian diffused hydrogen masses during 40 years of cycling. Figure (a) is a zoom-in of figure (b) for the first four years. The debrining and the standstill phases of 180 days have led to the initial percolated and diffused masses observed in Figure (a).

4. Discussion and conclusion

In general, for an overestimated diffusion of hydrogen in brine, a null pressure entry, and overestimated van Genuchten parameters, the hydrogen mass lost to the rock domain remains very negligible. Yet, increasing energy demands necessitate fast utilization of underground caverns. The severe utilization of salt caverns exposes them to considerable pressure and temperature changes throughout short periods of time, which may affect the development of the damaged zones. The introduction of a damaged zone, during cycling and at the end of leaching, can affect the total mass of the lost hydrogen. Future studies will include, in a first step, laboratory work to develop mass transport laws that would describe precisely hydrogen migration into the rock salt. In a second step, constitutive laws that quantify the permeability and porosity evolutions of rock salt due to mechanical, hydraulic and thermal charges would be needed. Once these transport and constitutive laws are developed, they can be integrated in our current frame work to estimate their effect on the hydrogen percolation and diffusion during rock salt damaging/severe exploitation of caverns. Our simulations have demonstrated that cycling reduces the total amount of transported hydrogen (Figure 4).

References

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