

Hydrogen storage in salt caverns: evolution of salt permeability and hydromechanical modelling at the cavern scale under cycling conditions.

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maîtriser le risque pour un développement durable

GENERAL CONTEXT: energy transition and H₂ storage



Hydrogen: versatile and promising carrier for energy transition

Currently produced from fossil fuels but from renewable energies in the future (green hydrogen)

Clean hydrogen (green and/or blue) should play a key role in the world transition to achieve carbon neutrality before 2050

- ✓ large-scale electricity storage
- $\checkmark\,$ decarbonize uses that are difficult to electrify
- ✓ use in industrial processes



GENERAL CONTEXT: massive H₂ storage

Storing hydrogen in large quantities is the best option

Massive Storage
Underground storage



Storage in salt cavern

Most secure and economical solution for large volumes of H₂

Only 4 salt caverns in operation worldwide (e.g., Spindletop USA, Teeside UK...)

but many industrial pilot sites in preparation worldwide (e.g., Etrez, France)

PROBLEM

Specific constraints compared to conventional gas storage

- High danger and mobility of H₂ (low density and viscosity, high diffusion)
- Need to accurately predict the extent of potential leaks of H₂ to ensure safe storage: sealing of salt?

Strong temperature (0-50 °C) and gas pressure (2-6 MPa for a 350 m deep cavern) variations in the salt cavern depending on the injection-production cycles (depends on usage)

 \rightarrow Thermo-mechanical damage of rock salt in the near field (close to the wall) \rightarrow permeability \checkmark

FOCUS OF THE STUDY

Characterize the ThermoHydroMechanical behaviour of rock salt

- Mechanical properties of initial material
- Impact of damage processes on rock salt permeability
- Viscoplasticity effect (self-healing process)
- Impact of mechanical (static and dynamic) and thermal (dynamic) fatigue

Grgic, D., Al Sahyouni, F., Golfier, F. Moumni, M. & Schoumacker, L., 2022. Evolution of gas permeability of rock salt under different loading conditions and implications on the underground hydrogen storage in salt caverns. *Rock Mechanics and Rock Engineering,* 55(1):1-24.

MATERIALS AND METHODS



Cylindrical sample of rock salt (ϕ = 100 mm; H = 200 mm)



- Salt bed of the Alsace potash mines (530 m depth) in the East region of France (Stocamine site for ultimate waste storage)
- Sannoisian-Oligocene (Cenozoic) geological stage
- Considered as a natural analogue of salt caverns used for in-situ H₂ storage
- Very low initial porosity (~ 1%) composed mainly of infrapores (nanometric size) that connect dispersed cracks and macropores





Experimental device

Experimental device

- Large scale triaxial compression cell
- Continuous measurement of deformations (strain gages)
- Continuous measurement of gas (He) permeability with the steady-state method
- Pressures controlled with precision syringe pumps

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EXPERIMENTAL PROGRAM

Characterisation of the mechanical behaviour

- Hydrostatic compression tests
- Uniaxial and triaxial compression tests
- → Characterization of short-term deformation mechanisms (plasticity, micro-cracking damage)

Gas permeability measurements

- Under hydrostatic loading and triaxial loading with different confining pressures
- ightarrow Analyse the impact of the deformation mechanisms on permeability
 - At different gas pressures
- ightarrow Analyse the slippage (Klinkenberg) effect and obtain the intrinsic gas permeability
 - Under dynamic (cyclic) and static (creep test) triaxial loading, and under dynamic thermal loading
- ightarrow Analyse the impact of mechanical and thermal fatigue

MECHANICAL BEHAVIOUR OF ROCK SALT

Under hydrostatic loading

- Isotropic mechanical behaviour
- The behaviour is elastic (irreversible) and linear up to 30 MPa
- \rightarrow No large initial cracks
 - No poromechanical coupling (Biot's coefficient ~ 0)



Under deviatoric loading

- Very low elastic limit (yield strength)
- Crack initiation and dilatancy thresholds increase with the confining pressure (Pc)
- Dilatant and irreversible microcracking damage develops at low Pc (0 and 1 MPa)
- Behaviour becomes **fully plastic** (ductile) at high Pc (5 MPa)



• Dilatancy threshold ↔ important increase of permeability due to cumulated microcracking damage

INTRINSIC PERMEABILITY OF ROCK SALT

Klinkenberg (slippage) effect

- Klinkenberg effect (i.e., decrease of permeability with the increase in gas pressure) only observed for the less permeable (and then less initially damaged) samples
- When it appears, the gas flow falls in transitional regime in weakly damaged rock salt

Evolution of apparent permeability k_a as a function of mean gas pressure at different deviatoric stress levels during triaxial compression tests



Initial permeability

- The initial intrinsic permeability of the studied rock salt ranges over more than 4 orders of magnitude: 10⁻¹⁶ to 5×10⁻²¹ m²
- Wide permeability range of the as-received samples due to the presence of cracks caused by the stress relaxation (induced by core drilling or cavity excavation) and sample preparation



EVOLUTION OF APPARENT GAS PERMEABILITY WITH STRESS INCREASE

Under deviatoric loading

- At low confining pressure (1 MPa): small increase in gas permeability from the dilatancy threshold due to **microcracking damage**
- At high confining pressure (5 MPa): no increase in permeability because the material becomes **fully plastic** which practically eliminates microcracking and thus dilatancy
 Evolution of apparent permeability k_a



Under hydrostatic loading

as a function of deviatoric stress

- Gas permeability decreases because of the closure of voids (cracks and pores)
- Decrease is irreversible if time of run is high enough
- Due to the **self-healing process** (irreversible closure of cracks)
- → Restoration of the permeability of undisturbed natural rock salt
 - Evolution of apparent permeability k_a as a function of confining pressure *Pc* and time



IMPACT OF MECHANICAL AND THERMAL FATIGUE ON ROCK SALT PERMEABILITY

Static (creep test) and dynamic (cyclic) mechanical fatigue

- Volumetric dilatancy (microcracking damage) develops and increases slightly the permeability during dynamic fatigue
- Self-recovery reduces damage and decreases slightly the permeability during static fatigue (creep)

→ Different mechanisms involved in rock salt deformation during dynamic and static fatigue act in a competitive way to annihilate any significant permeability evolution



Evolution of permeability *k* and deformations as a function of time during a creep test



Evolution of permeability k and volumetric deformation ε_v as a function of time during the cyclic triaxial compression test

IMPACT OF MECHANICAL AND THERMAL FATIGUE ON ROCK SALT PERMEABILITY

Cyclic thermal fatigue

6.E-19

1.E-19

1.E-20

 k_a (m²)

- Small permeability increase due to the microcracking damage that develops at the microscopic scale
- Due to the anisotropy of the thermal deformation of rock forming minerals and to the polycrystalline nature of rock salt
- → Deformation heterogeneities and then differential thermal stresses and microcracking damage



Evolution of volumetric deformation ε_v as a function of time during the thermal cyclic fatigue test

 $1/p_{m}$ (bar⁻¹)

0.15 0.20 0.25 0.30 0.35 0.40 0.45 0.50 0.55 0.60 0.65





EXPERIMENTAL HIGHLIGHTS

- Permeability of the studied rock salt varies over more than 4 orders of magnitude
- Slippage (Klinkenberg) effect only observed for the less permeable and damaged samples
- Deviatoric loading under low confining pressure (1 MPa)
 - ightarrow Small increase in gas permeability from the dilatancy threshold due to microcracking
- Deviatoric loading under high confining pressure (5 MPa)
 - \rightarrow No increase in permeability because the material becomes fully plastic (no more microcracking)
- Hydrostatic loading
 - \rightarrow Gas permeability decreases because of irreversible cracks closure due to self-healing process
- Permeability increases slightly during dynamic/cyclic mechanical and thermal fatigue tests (corresponding to high-frequency cycling in salt caverns) due to microcracking
- Permeability decreases during static fatigue (creep) thanks to the self-recovery process
- Results have to be confirmed on less initially damaged salt samples !!!

The different mechanisms (viscoplasticity with strain hardening, microcracking and cracks healing) involved in rock salt deformation act in a competitive way to annihilate any significant permeability evolution. → Strong confidence in the H₂ storage in salt caverns which remains by far the SAFEST SOLUTION

Numerical modeling of hydrogen storage in salt cavern

- From available experimental results on salt rock, propose/develop a rheological model that reproduces the main features of short and long term behavior
- Validation based on experimental tests and application(s) to salt cavern
- Analysis of the impact of the operational phase:
 - ✓ Mechanical behaviour of the salt cavern
 - ✓ H₂ leakage through the cavern wall (extent of the dissolved H₂ plume)



• HydroMechanical model: viscoplastic and damage model

Coarita-Tintaya E.D., Golfier F., Grgic D., Souley M. and Cheng L. 2023, Hydromechanical modelling of salt caverns subjected to cyclic hydrogen injection and withdrawal, Computers and Geotechnics, 162: 105690, <u>10.1016/j.compgeo.2023.105690</u>

• ThermoHydroMechanical model: Previous model + fatigue + healing + thermal coupling

Elastic tensor

(compressive stresses are negative and $\sigma_1 \leq \sigma_2 \leq \sigma_3$)

$$egin{aligned} C_{ijkl} &= (1 - d) C^0_{ijkl} \ d &= d_i + d_f + d_t - h \end{aligned}; \qquad d \in [0;1] \end{aligned}$$

d_i: instantaneous damage *d_f*: fatigue damage *d_t*: tertiary damage *h*: healing *d*: total damage

Mohr-Coulomb yield surface

$$f_{p} = q + p M_{p} - N_{p}$$

$$M_{p} = \eta_{p}(\gamma^{p}) \frac{\sin \phi}{\left(\frac{\cos \theta}{\sqrt{3}} - \frac{1}{3}\sin \theta \sin \phi\right)} \quad ; \quad N_{p} = \eta_{p}(\gamma^{p}) \frac{(1 - d_{i} - d_{f} + h)c \cos \phi}{\left(\frac{\cos \theta}{\sqrt{3}} - \frac{1}{3}\sin \theta \sin \phi\right)}$$

Hardening rule

$$\eta_{\rho}(\gamma^{\rho}) = 1 - (1 - \eta_{\rho}^{0}) \exp\left\{-\alpha_{\rho}^{0} \gamma^{\rho}\right\}$$

Plastic potential

(Chiarelli *et al* 2003, Souley *et al.* 2017)

$$g = q + \beta(\gamma^{p}) p$$

$$\beta(\gamma^{p}) = \begin{cases} \beta_{m} - (\beta_{m} - \beta_{0}) \exp(-b_{\beta} \gamma^{p}) & ; \quad \gamma^{p} < \gamma^{p}_{ult} \\ \beta_{ult} \exp\left(1 - \frac{\gamma^{p}}{\gamma^{p}_{ult}}\right) & ; \quad \gamma^{p} \ge \gamma^{p}_{ult} \end{cases}$$

$$\beta_{m} = \begin{cases} \beta_{m}^{0} \exp(a_{p} \sigma_{1}) & ; \quad \sigma_{1} < 0 \\ \beta_{m}^{0} & ; \quad \sigma_{1} \ge 0 \end{cases} \qquad \beta \uparrow$$

 $\beta(\gamma^p)$: dilatancy coefficient

. n.

 β_0 ($\beta_0 < 0$) is the initial volumetric contraction, β_m ($\beta_m > 0$) is the volumetric dilatancy at large deformation

We have volumetric contraction when $\beta < 0$, whereas the plastic strains evolve towards volumetric dilatancy if $\beta > 0$.





Fatigue damage (d_f **)** (Zhang et al. 2023)

$$\dot{d}_{f} = \frac{b_{f}}{T_{f}} \left(d_{f}^{max} - d_{f} \right) \qquad ; \qquad d_{f} \in [0; d_{f}^{max}]$$

$$d_{f}^{max} = d_{f0}^{max} \eta_{p} \left(1 + b_{f2} \frac{\langle Y_{d} - Y_{d}^{c} \rangle}{Y_{d}^{c}} \right)$$

Assumption: The value of instantaneous damage is constant during the fatigue test



Tertiary damage (*d*_{*t*}**)**

(Hou et al. 2003)

$$\dot{d}_t = A_T \frac{A_T}{(1-d)^{n_T}} \left\langle \frac{f_{ds}}{\sigma_{ref}} \right\rangle^{n_T}$$

 $q^* = rac{q}{1-d}$ q^* : Mises stress undamaged q : Mises stress damaged

→Damage is activated and increases if the damage limit (dilatance criterion) is exceeded

Healing (*h*)

$$\dot{h} = rac{d}{h_1} \left\langle rac{-f_{ds}}{\sigma_{ref}}
ight
angle$$

Assumption: The healing boundary and the damage threshold are identical



Mathematical formulation

Thermo-hydro-mechanical coupling

$$\nabla \cdot [C : \varepsilon^{e} - b(p - p_{ref})I - \alpha C : (T - T_{ref})I] + \rho_{m}\vec{g} = 0$$

$$\rho_{f} b \frac{\partial \varepsilon_{v}}{\partial t} + \frac{\rho_{f}}{M} \frac{\partial p}{\partial t} + \nabla \cdot (\rho_{f} \vec{q}) - 3\rho_{f} \alpha_{m} \frac{\partial T}{\partial t} = 0$$

$$\rho C_{p} \frac{\partial T}{\partial t} + \rho_{f} C_{p,f} \vec{q} \cdot \nabla T - \nabla \cdot (\lambda \nabla T) = 0$$

$$n \frac{\partial (c_{H_{2}})}{\partial t} + \nabla \cdot (q_{i} c_{H_{2}}) = \nabla \cdot (n D^{*} \cdot \nabla c_{H_{2}})$$

Where:

$$C = (1 - d)C^{0}$$

$$\rho_{m} = (1 - n)\rho_{s} + n\rho_{f}$$

$$\vec{q} = -\frac{k}{\mu_{f}(T)}(\nabla p + \rho_{f}\vec{g})$$

$$\frac{1}{M} = \frac{n}{K_f} + \frac{(1-b)(b-n)}{K_d}$$
$$\alpha_m = (b-n)\alpha + n\alpha_f(T)$$
$$k = k_0 10^{A_k d} \quad \text{(Gawin et al. 2002)}$$

- Fully water-saturated material
- Two-phase flow not considered
- Dissolved H₂ transport by advection and diffusion
- The value of the Biot coefficient is 0.3 if material is damaged

boundary and initial conditions



Radial distance (m)

0

Analysis cases

- Elastic (To verify the HM model: realised) ٠
- Cavern A (seasonal & daily) ٠
- Cavern B (seasonal & daily) ٠

Cavern geometry, boundary and initial conditions and properties





Cavern geometry, boundary and initial conditions and properties

Short-term numerical values

Parameter	Value	Unit	Parameter	Value l	Unit	Parameter	Value	Unit	Parameter	Value	Unit
E	35.0	GPa	α_p^0	70		a _p	0.5	1/MPa	d_{f0}^{max}	0.031	
ν	0.3		$\dot{\beta_0}$	-0.02		b_d	70		b _f	0.036	S
С	12.5	MPa	β_m^0	1.5		d_{i0}^{max}	0.3		b _{f2}	1	
ϕ	28.3	0	b_{eta}	80		a_d	0.5	1/MPa	Y_d^c	0.0278	
η_{ρ}^{o}	0.05		γ^{p}_{ult}	0.09		R_t	3	MPa	Tf	1	у

Long-term numerical values

Parameter	Value	Unit	Parameter	Value	Unit	Parameter	Value	Unit
G_k	12600	MPa	η_k	60000	MPa.d	n _T	3.5	
k_1	-0.18	1/MPa	k 2	-0.15	1/MPa	A_d	4.0E-10	1/d
<i>I</i> 1	0	1/K	A_N	0.005	1/d	β_{d0}	1	
G_{kE}	7560	MPa	B _N	4700	K	at	0.5	1/MPa
k_{1E}	-0.18	1/MPa	n _N	4		h_1	6000	d
I_{1E}	0	1/K	A_T	4.0E-8	1/d	β_h	1	

Cavern geometry, boundary and initial conditions and properties

Ну	/draulic		Т	hermal	Transport			
Parameter	Value	Unit	Jnit Parameter		Unit	Unit Parameter		Unit
k ₀	1.0E-20	m²/s	λ	6.8	W/(m.K)	D*	6.0E-9	m²/s
A_k	90		α	4.0E-5	1/K			
μ_{f}	0.001	Pa.s	C_p	850	J/(kg.K)			
n	0.012		α_{f}	f(T)	1/K			
Ь	0.012							
Kf	2.2E+9	Pa						
ρf	1000	kg/m³						



Stress path at the bottom of the cavern (Deviatoric stress projected to the plane $\theta = \pi/6$)



Relaxation of deviatoric stresses over time

 Seasonal and cavern A: in the first extraction it exceeds the elastic limit. For the following cycles this limit is not exceeded due to creep. In cavern B the dilatancy criterion is reached

Plastic and damage zone



Around **cavern A** there is a small extension of the plastic zone, in the lower and lateral part of the cavern.



Around **cavern B**, the damage zone is initially located at the bottom. The plastic zone is all around the cavern and is₂more extensive for daily cycling

Cavern wall displacements





Around **cavern A**, the displacements are small and in the order of 26 mm

Around **cavern B**, the displacements are important for seasonal cyclage. Maximum displacement of 3.6 m on the lateral side ²⁶

Hydrogen leakage



For both caverns, a similar hydrogen extension is estimated around the cavern. Almost 2.5 m for a daily cycling and almost 15 m for a seasonal cycling. Gas transport mainly by diffusion.

Plastic and damage zones around cavern



Zones at 33 years for (a) Case 1: HM (b) Case 2: THM (c) Case 3: THM (without healing)

- Before the operation phase, only plastic zones are calculated in the analysis cases
- At 3.5 years (first minimum gas pressure), the damage zone appears, and the plastic zone increases
- In the following years, damage zones increased slightly in extension, while the plastic zone increased more

Net damage at the bottom of the cavern



Note: Net damage is defined as $d = d_i + d_f + d_t - h_{r}$

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Cavern wall displacements



- (a) Cavern wall displacements (scaled by a factor of 2.5), (b) horizontal and (c) vertical displacements for Case 1: HM
 - Case 1 has more displacements than Cases 2 and 3 because it has a larger extent of the plastic zone around the cavern
 - Cases 2 (THM) and 3 (THM without healing) exhibit similar displacements
 - Important horizontal displacements are calculated on the bottom wall of the cavern, while significant vertical displacements are calculated on the floor of the cavern

Pore pressure distribution



Pore pressure distribution around salt cavern at 32.5 years for: (a) **Case 1**: HM (b) **Case 2**: THM (c) **Case 3**: THM (without healing)

Pore pressure distributions for Cases 2 and 3 show more modification around the cavern compared to Case 1, due to the temperature effect.

Hydrogen leakage



Hydrogen extension around salt cavern at 33 years for: (a) **Case 1**: HM (b) **Case 2**: THM (c) **Case 3**: THM (without healing)

Hydrogen extensions are similar for the three cases due to transport properties, modified only near the cavern wall

Conclusions

- A THM model describing the main key features of the rock salt behavior has been developed
 - Short term model takes into account elastoplastic and instantaneous damage behaviors
 - ✓ For the long-term behavior, the three creep phases generally observed on creep tests are considered
 - ✓ Fatigue damage and healing have been also implemented
 - Thermal coupling and hydrogen transport are considered
- Numerical simulation results
 - The deep cavern is more susceptible to mechanical stability problems.
 - The daily scenario is also more detrimental to the stability.
 - Even if damage occurs, the extent of damage zone is limited
 - the amount of gas leakage is also limited and both caverns lead to almost the same plume size because of diffusion which is preponderant.
 - Healing contributes to annihilate any significant damage evolution

Thank you for your attention !

Main assumptions of the model

- Rock salt is considered as an isotropic material
- Its elastic limit is very low -> The initial yield strength is assumed to be a fraction of the peak strength criterion
- It has a hardening deformation mechanism and shows a more ductile response than most other rocks
- **Ductile behaviour:** increasing confining stress
- Significant dilatancy at low confining stresses -> To define the dilatancy criterion
- we use the plastic potential: volumetric plastic strain (ε_v^p)
- if $\dot{\varepsilon}_{v}^{p} > 0$: we suppose that the material is in volumetric dilatancy



1 Dilatancy

Axial strain

- Damage initialisation: volumetric dilatation, due to microcracking, lead to a significant increase in the permeability of rock salt
- → Damage initiation is characterised macroscopically by dilatancy
- → The maximum achievable damage is reduced with increasing confining stress, because there is no volumetric dilatancy → ductile behaviour
- Based on continuum of damage mechanics CDM, an isotropic damage variable is considered, in first approximation, which modifies the elasticity and strength parameters





Proposed elastoplastic damage model

Full model equations

(compressive stresses are negative and $\sigma_1 \leq \sigma_2 \leq \sigma_3$)

M-C yield surface:

$$M_p = \eta^p (\gamma_p) \frac{\sin \phi}{\left(\frac{\cos \theta}{\sqrt{3}} - \frac{1}{3}\sin \theta \sin \phi\right)} \quad ; \quad N_p = \eta^p (\gamma_p) \frac{(1-d)c_i \cos \phi}{\left(\frac{\cos \theta}{\sqrt{3}} - \frac{1}{3}\sin \theta \sin \phi\right)}$$

Hardening variable:
$$\eta^p(\gamma_p) = \left\{\eta^p_o + \{1 - \eta^p_o\}\frac{\gamma_p}{\alpha^p + \gamma_p}\right\}$$

 $f_p = q + pM_p - N_p$

(Chiarelli et al. 2003, Zhou et al. 2011)



Rock salt behaviour in long-term



Transient creep (Kelvin-Voigt model)

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Steady-state creep
(Norton law)
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 $\left| rac{f_{ds}}{\sigma_{ref}}
ight|$



Parameters: G_k , k_1 , η_k and k_2

Parameters: A_N and n_N

(Heussermann et al. 2003)

Parameters: A_T and n_T

 $\dot{d}_t = A_T \frac{1}{1}$

→Damage is activated and increases if the damage limit (dilatance criterion) is exceeded



Proposed elastoplastic damage model



Numerical verification of triaxial tests

Short-term parameters values used



