

(ID Modèle = 2081337)

Ineris-206731-2736278-v1.0

05 avril 2022

State of knowledge on the storage of hydrogen in salt caverns

English version of Deliverable L6.3 of the ROSTOCK-H research project











controlling risks for sustainable development

FOREWORD

This document has been prepared as part of the support Ineris provides to administrative authorities, by virtue of Article R131-36 of the French Environmental Code.

Ineris shall not be liable, whether directly or indirectly, for any inaccuracies, omissions or errors or any similar occurrences relating to the information used.

The accuracy of this document must be assessed on the basis of the objective knowledge available and, where appropriate, of the current regulations at the date of the approval of this document. Accordingly, Ineris shall not be held liable due to the evolution of these knowledge or regulations after this date. Its mission shall not entail any obligation for Ineris to update this document once approved.

Given the missions entrusted under article R131-36 of the French Environmental Code, Ineris is not a decision-maker. Hence, the opinions, advice, recommendations or equivalent that will be provided by Ineris as part of its missions are solely aimed at assisting the decision-making process. As a result, the responsibility of Ineris cannot replace that of the decision-maker that is therefore, notably, solely responsible for any interpretations made on the basis of this document. Any recipient of the document shall use its results in their entirety or at least in an unbiased manner. The use of this document in the form of extracts or summary notes will be the sole and entire responsibility of the recipient. The same applies for any amendments that may be made thereto. Ineris declines all responsibility for any use of the document outside the purpose of the mission.

List of persons involved in the drafting of this document:

AGNOLETTI Marie-France, PErSEUs (University of Lorraine)

- BARBIER Laura, PErSEUs (University of Lorraine)
- COQUELET Christophe, Mines ParisTech
- CORVISIER Jérôme, Mines ParisTech
- DE DONATO Philippe, University of Lorraine

GOLFIER Fabrice, GeoResssources (University of Lorraine, CNRS)

GOMBERT Philippe, Sites & Territories Direction, Ineris

GRGIC Dragan, GeoResssources (University of Lorraine, CNRS)

HADJ-HASSEN Faouzi, Mines ParisTech

JALLAIS Simon, Air Liquide

JORAND Frédéric, LCPME (University of Lorraine, CNRS)

KARIMI-JAFARI Mehdi, Geostock

LACROIX Elodie, Sites & Territories Direction, Ineris / GeoResssources (University of Lorraine)

LAHAIE Franz, Sites & Territories Direction, Ineris

REVEILLERE Arnaud, Geostock Green Storage

THORAVAL Alain, Sites & Territories Direction, Ineris

TRUCHE Laurent, ISTerre (UGA, CNRS, USMB, IRD, University Gustave Eiffel)

WEINBERGER Benno, Fire-Explosion-Dispersion Direction, Ineris

Table of Contents

1	Intro	oduction	7
2	Und	erground hydrogen storage concept (A. Réveillère, M. Karimi-Jafari)	8
	2.1	Development of underground storage in salt caverns.	8
	2.2	Creation of a storage salt cavern	9
	2.3	Advantages of the underground storage of hydrogen	10
	2.4	Integrity of a storage salt cavern	11
3 B.	App Weinb	licable regulatory framework and associated administrative procedures (P. Gombert, F. La erger, S. Jallais)	ahaie, 13
	3.1	Current thinking at European level	13
	3.2	French regulations	13
	3.2.	1 Legislation applicable to underground hydrogen storage in a mining title context	13
	3.2. auth	2 Legislation applying to underground storage sites in the context of environm norisation	iental 14
	3.3	Regulations governing the underground storage of gas in other countries	16
	3.3.	1 Regulations governing the underground storage of gas in Germany	16
	3.3.	2 Regulations governing the underground storage of gas in the United States	17
	3.3.	3 Great Britain's regulatory framework	18
4 J.	Spe Corvis	cific features of underground hydrogen storage in salt caverns (F. Hadj-Hassen, C. Coq ier, F. Golfier, D. Grgic, L. Truche, F. Jorand)	uelet, 19
	4.1	Thermodynamic properties of hydrogen	19
	4.2	Interaction of hydrogen with materials present in the cavern	20
	4.3	Interaction with the well completion (steel, concrete).	22
	4.4	Impact of bacterial activity in a salt cavern	24
	4.5	Hydrogen leaks and the impact of hydromechanical couplings	29
5 Ja	Des fari, A.	ign of an underground hydrogen storage facility in a salt cavern (F. Hadj-Hassen, M. Ka Réveillère)	arimi- 34
	5.1	Design of the surface installations	34
	5.2	Characterisation of the salt	37
	5.3	Coupled modelling	38
	5.4	Laboratory storage pilot	41
6	Tec	hnical-economic analysis (S. Jallais, F. Hadj-Hassen)	43
	6.1	Description of markets and scenarios	43
	6.2	Spindletop storage feedback	45
7	Risł 48	analysis and environmental monitoring (A. Thoraval, P. Gombert, P. de Donato, E. La	croix)
	7.1	Risk analysis	48
	7.1.	1 Description of the storage and its environment	48
	7.1.	2 Identification, characterisation, and reduction of potential hazards	51
	7.1.	3 Feedback from accidents and incidents	51
	7.1.	Approach to the identification of accident sequences	53
	7.1.	5 Central Feared Events related to the well	53
	7.1.	6 Finalisation of the risk analysis for a real storage site	54
	7.2	Expected impact of a hydrogen leakage in a shallow aquifer	55

	7.2.1	Experiment carried out	55
	7.2.2	Results obtained	55
8	Social ad	cceptability of underground hydrogen storage (L. Barbier, M.F. Agnoletti)	57
8	8.1 Am	ultifaceted concept	57
	8.1.1	Different levels of acceptability analysis	57
	8.1.2	Measuring social acceptability	57
8	3.2 Hov	v is the underground geological storage of hydrogen perceived?	58
	8.2.1	Perception of hydrogen and its storage	58
	8.2.2	Effects of individual characteristics on acceptability	59
8	8.3 Whi	ch factors affect the social acceptability of the underground storage of hydrogen?	59
	8.3.1	Unacceptability factors	60
	8.3.2	Acceptability factors	60
8	3.4 Cor	clusion on the social acceptability of the underground geological storage of hydrog	en. 61
9	Conclusi	ons regarding the storage of hydrogen in salt caverns	62
10	Bibliog	raphic references	65
11	Gloss	ary	70

Table of figures

Figure 2-1. Example of the positioning of underground storage caverns (in green and purple) in a layer Figure 2-2. Diagram of the well completion of a salt cavern during the different phases (GEOSTOCK) Figure 2-3. Aerial view of the Geosel (30 salt caverns storing about a third of France's annual consumption of liquid hydrocarbons) and Geomethane (7 salt caverns storing natural gas) sites in the Lubéron Regional Nature Park (Google Earth)11 Figure 3-1. Administrative procedures applicable to a request for the underground storage of H_215 Figure 4-1. Variation of temperature as a function of pressure during isenthalpic relaxation for hydrogen Figure 4-2. Cavern volumes required to store the same mass of gas M_o as a function of depth (M_o: mass of methane for $P_0=22$ MPa and $T_0=40$ °C).....20 Figure 4-3. Equipment used in laboratory to measure the solubility of hydrogen in aqueous solutions and comparison of the results obtained with other gases (ARMINES/CTP work)......21 Figure 4-4. Molar fractions of the gases formed in the cases of H₂, CH₄, and O₂, depending on the level Figure 4-5. Evolution over time of the concentration of sulphides in solution during the leaching experiment of hydrated CEM V Rombas cementitious pastes at 20 and 90°C under Ar or H₂ (10 bar). Figure 4-6. Simplified diagram of global bacterial activity showing the chemical species that can potentially be mobilised to ensure energy conversions (dissimilatory pathways) or the fixation of constitutional elements for biomass (assimilatory pathways) (Jorand, 2018)25 Figure 4-7. Schematic representation of Table 4-2 reactions expected in the context of the geological Figure 4-8. Maximum values in salt (NaCl) where bacterial activity has been observed, according to their

Figure 4-9. Growth curves as a function of the culture time for Shewanella oneidensis MR-1 (a) and Klebsiella mobilis (b) cultured in a rich medium (tripticase soy broth, aerobic) under different concentrations of NaCl (from 5 to 100 g·l ⁻¹ in addition to the concentration of the TSB medium)
Figure 4-10. Left: SEM analyses of a salt sample (a, b: high magnification images; c: BSE map; d: EDS map). Middle: photo of a cylindrical salt sample fitted with strain gauges for hydromechanical testing. Right: distribution of voids in a cylindrical salt sample obtained from 3D X-ray tomography
Figure 4-11. Stress-strain curve for uniaxial (tests 3 and 4) and triaxial (tests 5 and 6) compression tests
Figure 4-12. Evolution of P and S elastic wave velocities, dynamic elastic constants (E11, E33, v13, v31, and v12) and Thomsen's anisotropy factors (ϵ and γ) during a uniaxial compression test
Figure 4-13. Evolution of the apparent gas permeability as a function of the deviatoric stress and the pressure for sample 5 (triaxial compression test at 1 MPa of confinement, on the left) and for different samples (uniaxial and triaxial compression tests at different confining pressures, on the right)
Figure 5-1. Production of hydrogen by electrolysis and fields of application (source: SMRI)34
Figure 5-2. Thermodynamic hydrogen injection and withdrawal chains in the cavern for the scenario chosen in the Power to Mobility application
Figure 5-3. Results of a triaxial compression test with measurement of the dilating deviator37
Figure 5-4. Creep test on salt with three deviator stages and at constant temperature (5, 10, 15 MPa, T = 30°C)
Figure 5-5. Salt cavern storage system and coupling of associated phenomena
Figure 5-6. Behaviour of hydrogen storage in a salt cavern compared with the storage of other gases.
Figure 5-7. 2D asymmetric numerical model, zooming in on the cavern mesh
Figure 5-8. CFD model of the Staßfurt cavern in Germany42
Figure 5-9. Storage pilot developed in the laboratory42
Figure 6-1. Costs involved for each storage configuration considered, with a detailed CAPEX breakdown
Figure 6-2. Profile of the Spindletop cavern recorded by the August 2004 sonar scan (Source: Air Liquide)
Figure 6-3. Brine and hydrogen pressures after the cavern filling phase (on the left) and the selected operation cycle (on the right) (source: Air Liquide)46
Figure 6-4. Maximum value of the major principal stress (on the left) and maximum deviation between the deviatoric stress and the dilatancy upon contraction criterion (on the right) (Source: Air Liquide)47
Figure 7-1. Diagram of the well as considered in this study (Source: Ineris)
Figure 7-2. Location of the cavern in its environment (Source: Ineris)
Figure 7-3. Simplified representation of an accidental sequence and its potential barriers
Figure 7-4. Accident sequences related to ERC1 "Loss of hydrogen containment at the production wellhead"
Figure 7-5. Schematic cross-section of the Catenoy site during the injection of dissolved hydrogen55
Figure 8-1. Answers to the questionnaire on the social acceptability of underground H_2 storage58
Figure 8-2. Simplified diagram of the factors influencing individuals intending to act against underground hydrogen storage
Figure 8-3. Simplified diagram of the factors influencing individuals intending to act in favor of underground hydrogen storage60

Executive summary

This document is the English version of deliverable L6.3 of the research project ROSTOCK-H, translated in the framework of the French Ministry of Ecological Transition support program "SIT04: Safety and Impact of Underground Storages". It deals with the specificities of storing hydrogen in salt caverns with the particular aim of buffering the sometimes-intermittent production of renewable sources of energy currently under development. It thus reviews, in turn: the concept of the underground storage of hydrogen in salt caverns, the design of this storage and its techno-economic analysis, the associated risks, the environmental monitoring to be established at a future site, and finally, the social acceptability of this emerging technology. It is based both on experiments and original models, designed and implemented as part of the project, but also on feedback from the storage of various products in salt caverns which have been operational since the mid-20th century.

The pertinence of the ROSTOCK-H project rests on the fact that, among the 2000 existing salt caverns in operation around the world, only 6 are used to store hydrogen. It therefore proved necessary to produce an overview of the existing knowledge and contribute to new results. These results confirm that salt constitutes a sufficiently leak-tight formation for storing hydrogen, that the risk of accident - leakage in particular - is limited, that the hydrogen sector is already viable for needs related to mobility, and that its social acceptability is starting to increase.

In France, salt cavern storage technology is therefore sufficiently mature to now be used for hydrogen, despite the specific properties of this gas (high mobility and diffusivity, chemical and biochemical reactivity, flammability, etc.). By offering a synthesis of the main work carried out throughout the ROSTOCK-H project, this document also makes it possible to draw up guidelines for the design and management of future underground hydrogen storage in salt caverns.

Use the link provided below for quotations:

French national institute for industrial environment and risks (Ineris), State of knowledge on the storage of hydrogen in salt caverns, Verneuil-en-Halatte : Ineris - 206731 - v1.0, 05 avril 2022.

Keywords:

Underground storage, Salt cavern, Hydrogen, Mechanical behaviour, Risks, Social acceptability

1 Introduction

The ROSTOCK-H research project, entitled "Risks and Opportunities of Geological Storage of Hydrogen in salt caverns in France and Europe", was subsidized by the GEODENERGIES Scientific Interest Group from 2016 to 2021. It intends to improve knowledge of the phenomena involved in the underground storage of hydrogen in salt caverns with the aim of making their optimisation and safe operation possible. The technical-economic, regulatory and social conditions for the deployment of the technology in France and Europe are also discussed. Led by Air Liquide, this project brought together research teams from Geostock, ARMINES (via the MINES ParisTech – ARMINES Joint Research Centres for Geosciences and Thermodynamics of Processes), the University of Lorraine and Ineris.

The French energy-climate law of 8 November 2019 plans to increase the share of renewable energies to at least 33% of gross final energy consumption by 2030. This ambitious goal testifies to France's commitment to decarbonise its energy model and particularly its electricity system. For certain renewable sources of energy (solar, wind...) with an intermittent or fluctuating nature, increasing their share in the electrical system requires the use of bulk energy storage solutions. One of these solutions consists of converting this electricity into hydrogen by water electrolysis, then storing the hydrogen thus produced. For this purpose, the underground environment offers numerous advantages: less ground area used, distancing the product from surface concerns, storage in large volumes, possibility of storage under high pressure. In addition, the underground storage of liquid or gaseous products has been practised since the beginning of the 20th century in the United States, and since the 1950s in France. There are thus more than 600 underground storage sites in the world, mainly used to store natural gas or hydrocarbons. In France, 23 storage sites are currently operational, housing a total of about a hundred underground reservoirs - the majority being salt caverns - as well as aquifers, mined caverns, and a depleted reservoir (Ineris, 2016; Géologues, 2018). Although all of these options are currently under consideration, storage in salt caverns seems today to be the most mature solution, at least for the short term, to address the energy transition needs - in particular owing to the remarkable natural tightness provided by the salt and the possibility of carrying out rapid cycles (daily or weekly for example).

This document constitutes the English version of deliverable L6.3 of the ROSTOCK-H research project. It summarizes most of the work carried out throughout the project and offers guidelines for the design and management of underground hydrogen storage in a salt cavern, based particularly on the results obtained. Its goal is to present the concept of underground hydrogen storage (chapter 2), the applicable regulatory framework and associated administrative processes (chapter 3), the specificities of underground hydrogen storage in a salt cavern (chapter 4), the design of underground hydrogen storage in a salt cavern (chapter 5), the techno-economic analysis of such storage (chapter 6), the analysis of the risks and environmental monitoring (chapter 7), and, finally, the social acceptability of this emerging technology (chapter 8). Its main contributors are Géostock for chapter 2, Ineris for chapters 3 and 7, Mines Paris Tech for chapters 4 and 5, Air Liquide for chapter 6, and University of Lorraine for chapter 8.

2 Underground hydrogen storage concept (A. Réveillère, M. Karimi-Jafari)

2.1 Development of underground storage in salt caverns.

"Until about 100 years ago, when modern geology revealed its prevalence, salt was one of the world's most sought after commodity. A substance so valuable that it served as currency, salt has influenced the establishment of trade route and cities, provoked and financed wars, secured empires and inspired revolutions". This quote by the writer and journalist Mark Kurlanski (2002) reflects the importance of salt in the history of humanity. This has all changed and today salt crystals have a value equivalent to that of sand used for construction, with dissolved salt being significantly less expensive again.

The industrial revolution in fact made it possible to develop an industry to extract salt from the subsoil in the 18th century in the United Kingdom and in the 19th century in the rest of Europe. This was enabled through mining techniques (chamber and pillar mines) as well as through the progressive development of "leaching" (injection of fresh water and recovery of salt water), a technique which leaves an underground volume filled with brine.

This leaching started where salt was the most accessible, on the surface of outcropping layers (essentially halite or "rock salt" with the formula NaCl), accelerating, as it were, the natural erosion, and inducing subsidence and surface collapses. However, this only allowed a limited extraction because only the surface of the layer was then exploited. To manage these risks and improve the mining ratio, operators decided to drill wells down to the bottom of the salt layer, and then to limit the height of exploitation of the salt mass in order to leave a layer of salt intact at the top. Caverns of several tens of metres in diameter and sometimes several hundreds of metres high, entirely contained in a geological salt layer were thus created by this extraction activity.

These underground caverns left filled with brine by mining activity started to be reused eight decades ago: in the early 1940s for the storage of crude hydrocarbons in Canada in the context of the Second World War, and in 1961 in the United States for the storage of natural gas. Caverns specifically designed for storage were then created, including for new products such as hydrogen, for example, starting in 1972 in England.

This underground storage industry became progressively more structured. Specialised companies appeared, a regulatory framework was drafted, research laboratories took an interest in the problems encountered, and know-how gradually spread, particularly under the aegis of the Solution Mining Research Institute (SMRI¹), which has been organising conferences and training for more than 50 years, and which brings together more than 2000 articles on salt caverns. Underground storage in salt caverns is therefore a technology that has now reached maturity.

All types of hydrocarbons (whether in liquid, gaseous, liquefied or supercritical state), as well as other products such as hydrogen, nitrogen, helium, and compressed air can be stored in salt caverns. In France, there are 78 salt storage caverns, some of which have been in operation for more than half a century. Globally, there are about 2000 salt storage caverns in operation, of which only 6 are for hydrogen. Figure 2-1 presents a recent example of the sizing of a natural gas storage project site, where the planned salt caverns are spaced away from each other and placed at a distance from the salt roof. The site is in Cheshire (UK), close to where the first underground industrial operations for the mining of salt resources were developed.

¹ <u>www.solutionmining.org</u>



Figure 2-1. Example of the positioning of underground storage caverns (in green and purple) in a layer of salt (in yellow) on a project site in Cheshire in the United Kingdom (Geostock, 2014)

2.2 Creation of a storage salt cavern

The salt formations sometimes appear in layers of varying thickness, which can reach or exceed a thousand meters, and other times in domes with often very high vertical extension. Storage caverns are generally dug into rock salt because of:

- its thickness, sufficient to create large caverns;
- its high solubility in water which facilitates the excavation of caverns by dissolution, requiring much less manpower and heavy equipment compared to conventional mining excavation techniques;
- its very low porosity and permeability which guarantee the tightness of storage caverns for nonwetting fluids (gas, hydrocarbons), as well as its chemical non-reactivity with these products;
- its good mechanical strength, in particular its resistance to compression.

Creation of a salt cavern starts with the drilling of an oil-type well down to the salt layer at a depth of several hundred or several thousand metres. Starting from the base or shoe of the last cemented casing of this well in the salt formation, a cavern of one or a few hundred meters in height is formed by leaching. To do this, fresh water is injected either at the base of the cavern through a central tubing (direct leaching), or via the annular space formed between this central tubing and an external column (inverse leaching). The water is forced to circulate between the points of injection and extraction so that it leaches the wall of the cavern which is thus enlarged by dissolution, and the water takes on salt to become brine.

In order to control the dissolution of the salt in the upper part of the cavern, a fluid that is inert and lighter than brine (such as nitrogen) is injected into the annular space between the production casing and the external column. Developments in the cavern roof are closely dependant on the way in which the depth of the interface between this inert fluid and the brine is set. Thus, the geometry of the excavated cavern depends on the depth of this interface and that of the water injection and brine extraction points. The duration of leaching of a cavern is roughly proportional to its final volume and inversely proportional to the water injection rate. A few years of leaching are thus needed to obtain a cavern of several hundreds of thousands of m³ in volume.

In the case of the storage of a gas such as hydrogen, the well equipment is presented in Figure 2-2. It includes:

- a completion installed in a cemented production casing; the annular space between the two is filled with a so-called "completion" fluid, which isolates the cement casing from the gas, erosion and potentially damaging pressure variations, and constitutes a second containment barrier for better safety;
- a central column through which the brine is discharged from the cavern during 1st gas fill;
 a production wellhead.



Figure 2-2. Diagram of the well completion of a salt cavern during the different phases (GEOSTOCK)

The well completion and the cavern are subject to tightness tests prior to the start of gassing. This is done by injecting gas through the annular space between the production column and the central column. The cavern is not lined; its tightness is guaranteed by the salt itself. In the case of storage of gas, a minimum gas pressure will always be maintained in the cavern in order to ensure its mechanical stability. The gas used to maintain this minimum pressure is referred to as "cushion gas". The amount of gas injected into the cavern in addition to the cushion gas until the cavern reaches its maximum pressure (typically between 100 and 200 bar) is called the "working gas". The storage is exploited by compressing and decompressing this working gas.

2.3 Advantages of the underground storage of hydrogen

In the first chapter of Réveillère et al. (2017), David Evans shows that – in the case of petroleum products – underground storage is the least accident-prone link in the production-storage-distribution chain. As emphasised by Bérest et al. (2019), there are several reasons for this:

- flammable products cannot explode or burn at depth, in the absence of oxygen in the air;
- these products are protected from external disruptions (tornado, fire, aircraft crash, attack) by several hundred metres of ground;
- Underground storage sites are barely affected by earth tremors, since the maximum effect most often occurs near the surface.

Furthermore, their surface footprint is low as shown, for example, in Figure 2-3.



Figure 2-3. Aerial view of the Geosel (30 salt caverns storing about a third of France's annual consumption of liquid hydrocarbons) and Geomethane (7 salt caverns storing natural gas) sites in the Lubéron Regional Nature Park (Google Earth)

More than 10% of the global annual production of natural gas is now stored in 661 underground storage sites, whether in salt caverns, aquifers, or depleted deposits, representing a total of 422 billion m³ (NPTC)². Underground storage has become so important because it is cheaper, safer, and has less impact on the environment than alternative surface storage techniques. These advantages also remain equally valid for hydrogen; its underground storage could also take a major place in the hydrogen energy ecosystem which is developing today.

2.4 Integrity of a storage salt cavern

A gas storage cavern is generally, with some exceptions, operated by the working gas compressionexpansion cycles. It is possible to manage the main risks by complying with the operating limits regarding pressures and their variations over time. These risks included:

- massive rupture of the salt formation;
- excessive loss of cavern volume, which may be significant for a deep cavern at very low pressure (the minimum allowable pressure is determined based on laboratory test results and geomechanical modelling);
- loss of the salt's tightness, particularly through development of microfracturing, which is a risk at high gas pressures. This risk is managed by complying with a maximum allowable pressure which may be based on hydraulic fracturing test results in the borehole (in particular, it must be much lower than the fracturing pressure obtained from the borehole tests);
- leakage through the cement around the shoe of the casing. This risk is estimated by a "Mechanical Integrity Test" or "MIT" type of tightness test.

The creation and operation of the cavern modifies the initial stresses existing in the salt. In extreme cases, these perturbations may lead to damage of the salt and its rupture on a more or less large scale. It is necessary to apply the sizing and operating rules for gas storage caverns to prevent such undesirable phenomena (see chapter 4). The principle mechanisms underlying the rupture of the salt

² One m³ (at NPTC) corresponds to the volume occupied by 1 m³ of gas under Normal Temperature and Pressure Conditions, i.e. 1013 hPa and 0°C; one sometimes encounters the designation Nm³ or (N)m³ for "Normo-m³" or "Normal-m³".

are tensile failure (in particular, due to thermal effects), and short-term shear failure (very often in extensional mode) and long-term shear failure (tertiary creep):

- at the scale of a cavity, the most likely rupture scenario generally occurs at very low pressure in the cavern with an extension stress state (when the pressure in the cavern is lower than the stresses perpendicular to the cavern wall), either via shear or tensile failure because of irregularities in the shape of the cavern;
- thermal tensile stress due to cavity gas cooling can occur during fast withdrawals at low pressure; the extension of the tensile zone around the cavern may vary depending on the operating parameters, in particular the rate of variation of the cavern pressure;
- for an effective tensile stress to appear in the salt around the cavern, the gas pressure must exceed the least compressive tangential stress added to the salt tensile strength which is often considered to be null for the sake of conservatism;
- before the short-term rupture occurs, the salt undergoes a dilatancy state in which the damage increases the salt permeability; various authors have shown that the onset of dilatancy can be considered to be a criterion for long term stability.

Variation in the internal pressure during operation of a storage salt cavern therefore has a decisive effect on the local or global stability of the structure. The minimum operating pressure depends on the state of lithostatic stresses (in other words, the depth of the cavern and the weight of the overlying rock mass). A very low pressure in the short term can cause local instabilities which change the initial shape of the cavern (for example, spalling of the walls, rise of the roof, excessive convergence of the walls). In the long term, this can lead to the overall collapse of the cavern due to reach a tertiary creep stage. Thus the duration of the low pressure operations in gas storage caverns must be limited.

One problem associated with the maximum operating pressure is the gradual increase in the permeability of the salt which can occur long before fracturing. The maximum allowable pressure depends not only on the lithostatic stress state but also on the history of the internal pressure. In fact, the lithostatic stress state of the rock mass changes over time because of the viscous nature of the salt.

Today, thanks to numerical modelling, the compliance of a salt cavern with the stability and tightness criteria can be checked, starting at the design stage. Once the construction is completed, the tightness of the cavern/well system is checked by means of MIT tests which make it possible to check, in particular, the quality of the cementation at the level of the last cemented casing. During operation, regular checks by sonar survey of the cavern and the condition of the casings (steel and cementation) are performed in order to verify the integrity of the cavern. A micro-seismic monitoring network also makes it possible to continuously detect any rock falls or other rupture phenomena, and thus to implement corrective actions if necessary.

3 Applicable regulatory framework and associated administrative procedures (P. Gombert, F. Lahaie, B. Weinberger, S. Jallais)

To promote the development of an underground hydrogen storage sector in France, it is important to ensure that the regulatory framework is appropriate and, if necessary, to identify the changes necessary to ensure a controlled development of this sector - in particular regarding safety and the environment. We address here the regulatory framework applicable to the underground part of underground storage systems for hydrogen or similar gases, given that the surface installations necessary to operate this type of storage (compressors, exchangers, drying and purification stations, pipelines, temporary storage, possibly electrolysers, etc.) are not specific to underground storage and are already well covered by established regulations.

3.1 Current thinking at European level

The European Commission has established several texts to encourage energy transition in Europe, including directives 2009/28/EC and 2012/27/EC and more recently, the Green Deal for Europe (COM(2019) 640). A European strategy for hydrogen was defined on 8 July 2020 (COM(2020) 301) but few concrete measures were taken, at that stage, in the area of underground storage.

In addition, various other European research projects have been carried out (or are underway) on the topic of "hydrogen", including:

- the HyUnder project (2012-2014) which assessed the potential for underground storage of renewable electricity in the form of hydrogen in the European Union but without addressing the regulations of each Member State;
- the HyLaw project (2017-2018) which studied laws and regulations concerning fuel cells and hydrogen applications, as well as the legal barriers to their deployment and commercialisation but without however addressing underground storage;
- the HyPSTER project (2021-2023) which aims in particular to assess, on a European scale, the regulations and standards applicable to the safety of hydrogen storage in salt caverns. However, the results of this study will only be available in 2023.

3.2 French regulations

Although some projects are underway, no underground hydrogen storage demonstrator nor industrial site has been implemented on French territory to date³. As a result, the legal framework applying to this activity has yet to be tested. However, the study carried out by Ineris within the framework of the STOPIL-H₂ project (Djizanne & Lahaie, 2020) provided, through the analysis of the concrete case of the Etrez pilot (Eastern France), a more precise vision of the applicable regulatory framework in France.

3.2.1 Legislation applicable to underground hydrogen storage in a mining title context

Since the publication of Ordinance No. 2021-167 of 17 February 2021 relating to hydrogen, the underground storage of hydrogen is governed by the Mining Code (Book II) regardless of its origin (fossil, renewable or low carbon) or its use (chemical industry, mobility, injection into the gas network, etc.)⁴. As such, the research, creation, testing, development, and operation of an underground hydrogen storage facility requires the procurement of a mining title (an exclusive permit for exploration or exploitation concession), according to the procedures defined in the Mining Code (Book II – Titles II to IV) and specified in Decree No. 2006-648 of 2 June 2006 relating to mining titles and underground storage titles. Nevertheless, the ordinance of 17 February 2021 stipulates that "the holder of a

³ The first EU supported project aiming for large scale green hydrogen underground storage in salt caverns is HyPSTER, with a demonstration facility in progress in France. The project is launched since 2021 with an engineering study, which precedes the field-testing phase (see https://hypster-project.eu/)

⁴ Before the publication of this ordinance, only the storage of hydrogen for industrial use was covered by the mining code as a "chemical product for industrial use" (art. L211-2).

combustible or natural gas storage concession is exempt from the obligation to obtain a new mining title for the storage of hydrogen, as long as the geological formations in which the storage of hydrogen is envisaged are included in the perimeter(s) covered by the title already in his/her possession".

3.2.2 Legislation applying to underground storage sites in the context of environmental authorisation

Once the mining title is granted for the underground storage of hydrogen (or in the context of an existing concession if it is exempt pursuant to article 6 No. 2 of the ordinance of 17 February 2021), the project promoter must obtain authorisation for the implementation of underground storage works (research, creation, testing, development or operation). Three scenarios can then arise:

- 1. Exploration works are planned for a new site and will involve a quantity of hydrogen that is less than 100 kg, which is the ICPE classification threshold (Installation Classified for the Protection of the Environment) under section 4715; this is the case for drilling, creation or development of the caverns, or tests that involve small amounts of hydrogen; these works are governed the Mining Code and must comply with title VI of Book II of the Mining Code, decree no. 2006-649 of 2 June 2006 relating to mining works, underground storage works and the policing of mines and underground storage, decree no. 2016-1303 of 4 October 2016⁵ relating to research work by drilling and exploitation by well of mining substances (known as the "drilling" decree), and the ministerial order of 14 October 2016⁵ relating to research work by drilling and exploitation by well of mining "order);
- 2. Exploration works are planned for a new site and will involve a quantity of hydrogen that is greater than the ICPE classification threshold under section 4715 (i.e. 100 kg); in this case:
 - if they are undertaken on a new site, they pertain to the ICPE regulations under the declaration regime (if the quantity of hydrogen is between 100 kg and 1 ton) or the authorisation regime (if the quantity of hydrogen is greater than 1 ton, with a low SEVESO threshold of 5 tons and a high SEVESO threshold of 50 tons);
 - if they are undertaken within an existing underground storage, they must be treated as a modification of the existing ICPE; the nature of the modifications made as well as a presentation of the associated vulnerabilities, risks, and disadvantages must be brought to the attention of the Prefect; the Prefect then judges the nature of these modifications, according to the criteria set out in article R.181-46 of the environmental code, and decides on the administrative follow-up to be given: if the modifications are deemed to be substantial, it will be necessary to file a new application for environmental authorisation, otherwise an update of the requirements applicable to the site by a complementary or amending prefectural decree will suffice;
- 3. Underground storage operation works (first filling of the cavern, injection/withdrawal operations, maintenance and monitoring of the facilities) are planned; pursuant to the provisions of Directive 2012/18/EU of 4 July 2012, known as the "Seveso III Directive", the operation of hydrogen storage is covered by ICPE legislation under section 4715 and is therefore governed by the environment code (Book V Title 1).

In view of the quantities of hydrogen expected in future underground hydrogen storage sites, these will most likely fall under the SEVESO high-threshold authorisation regime. In this case, any modification made (creation of a new cavern, drilling of a new well, etc.), as well as the final shutdown of the storage site, will be governed by the ICPE regulations and not by the mining code.

Figure 3-1 presents a summary of the administrative procedures currently applicable in France to an underground hydrogen storage project.

⁵ These texts will only be applicable after amendment of article 2 to incorporate hydrogen into the legislation.



Figure 3-1. Administrative procedures applicable to a request for the underground storage of H₂

3.3 Regulations governing the underground storage of gas in other countries

3.3.1 Regulations governing the underground storage of gas in Germany

Contrary to France, where article L 211-2 of the mining code defines the list of products authorised in an underground storage, there is no such restrictive list in Germany. Each new product intended for storage must first undergo an assessment of its impact on the environment. Subsequently, the legislation that applies to this new product is the same as that which currently applies to the underground storage of natural gas. This is the reason why Germany was able to implement industrial storage of helium in Epe (North Rhine) in 2016, with no legislative modification. It is the first commercial site in the world for the underground storage of pure helium within a salt cavern⁶, located at a depth of 1,300 m.

The main texts that govern the underground storage of natural gas in Germany, which are equally applicable to the future storage of hydrogen, are the Federal Mining Act of 13 August 1980 - Bundesberggesetz⁷ (BBergG) and the Order on the Environmental Impact Assessment of Mining Projects (UVP V Bergbau).

<u>The BBergG Act</u> covers mining rights and, in particular, underground storage in paragraph 126, which specifies the necessary conditions to determine the suitability of an underground storage site:

- Article 51 stipulates that the authorisation for the establishment and operation of underground storage should be carried out on the basis of the operational plans drawn up and updated by the operator, which are then approved by the competent authority; it concerns:
 - <u>master operating plans</u> necessary for the establishment and operation of an underground storage; they are valid for a period generally limited to two years and specify the various operations planned by the operator and, in the case of redevelopment, all the work and new installations planned;
 - <u>special operation plans</u> which may be required by the competent authority for certain parts of the project; these deal with special works and installations that fall outside of the master operating plan, which are excluded because the necessary operations exceed the two-year limitation previously set;
 - Framework Operating Plans that outline all the installations to establish a framework for future development (for approval by the Master and Special Operating Plans); they give the administration an overview of the project as a whole, in order to assess potential sources of conflict in relation to public safety, public order, and the environment; they allow the operator to obtain an approval in principle for the execution of the mining project, their execution requiring the approval of the Master and Special Operating Plans;
- Article 55 stipulates that the approval of an operation plan can be granted by the administration if the following points are verified, and if its operation is not contrary to public interest:
 - 1. the extraction of mineral resources provided for in the plan is permitted;
 - 2. the operator has the necessary expertise and reliability;
 - 3. all precautions against injury to life and health and for the protection of property are taken both with regard to employees and third parties;
 - 4. mineral resources of public interest will not be damaged;
 - 5. the surface is protected in the interest of public safety and transportation;
 - 6. the waste produced by the operation will be properly used or disposed of;
 - 7. some precautionary measures for reuse of the surface after mining are planned;
 - 8. the existing storage or mineral exploitation installations are not endangered by the new installation;
 - 9. no harmful effects are expected from the operation or extraction phases.

^{6 &}lt;u>https://fr.media.airliquide.com/actualites/premiere-mondiale-air-liquide-met-en-service-le-premier-site-de-stockage-dhelium-pur-f2ed-1ba6d.html</u>

⁷ http://www.gesetze-im-internet.de/bbergg/

- Additionally, in the event of an installation in a littoral or coastal area, it is necessary:
 - 1. that it does not affect the operation and functioning of maritime installations;
 - 2. that it does not affect the use of shipping lanes, airspace, fisheries, nor is it inappropriate for the maintenance of plant and animal life;
 - 3. that it does not affect the deployment, maintenance, and operation of submarine cables and pipelines, as well as oceanographic or other scientific research:
 - 4. that it limits harmful effects on the sea as much as possible.
- the authorisation for shutdown of an operation also requires a shutdown plan based on points 2 to 13 above, with the additional requirement that the following be ensured:
 - the protection of third parties against dangers to life and health caused by the operator's activities, even after these have ceased;
 - the restoration of the surface conditions in the area affected by the operation;
 - in littoral or coastal areas, the complete removal of operational installations from the seabed.

<u>The UVP-V Bergbau order</u> specifies the scope of the environmental impact study for the products stored, as well as its domain of applicability. The content of this study is specified in paragraph 16 of the Environmental Impact Assessment Act "Gesetz über die Umweltverträglichkeitsprüfung (UVPG)". The operator must submit a report to the competent authority on the probable impact of the project on the environment, containing at least the following information:

- a complete description of the project;
- a description of the environment and its components affected by the project;
- a description of the characteristics of the project and the site, with the aim of excluding, reducing, or offsetting the occurrence of any major adverse environmental effects of the project;
- a description of the measures intended to eliminate or reduce the major adverse environmental impacts of the project, and the remedial measures planned;
- a description of the main expected environmental impact of the project;
- a description of the reasonable alternative solutions in relation to the project and its specific characteristics, as well as the reasons for the choice made by the project promoter to take into account their respective effects on the environment;
- a general non-technical summary of the report.

The following are subject to UCP-V Bergbau: natural gas storage facilities with a capacity greater than 100 million m³, as well as oil, petrochemical and chemical storage facilities with a capacity of:

- 200,000 t or more;
- 50,000 t to 200,000 t based on a general preliminary review on a case by case basis;
- 10,000 to 50,000 t based on an on-site specific preliminary review on a case by case basis.

3.3.2 Regulations governing the underground storage of gas in the United States

In the United States, the federal regulations refer to regulatory texts (codes, standards). Nevertheless, each state can decide whether to apply these regulations or use its own. The regulatory requirements applicable to the storage of gas⁸, which fall under both federal and state jurisdiction, have evolved considerably over the past twenty years.

At the federal level, this involves:

- the FERC (Federal Energy Regulatory Commission), to which the Energy Policy Act of 2005 specifically assigned market authority over gas storage, but not operational or safety authority;
- the PHMSA (Pipeline and Hazardous Material Safety Administration) which is an entity of the Department of Transportation and which has operational and security authority;
- the EPA (Environmental Protection Agency) which has jurisdiction over all environmental aspects associated with the discharge of gas in air, soil, or water.

In 2016, the PHMSA was tasked with developing regulatory texts on the safety of underground gas storage under the Pipeline and Enhancing Safety Act of 2016 (PIPES Act). It has also published an Interim Final Rule (IRF) and, in January 2020, minimum federal safety standards for gas storage sites.

⁸ Which do not depend on the type of gas stored nor on its use (industrial or energy provision).

In this context, State entities will have to inspect the storage facilities and apply these rules. The bulk of the IFR, now published as a Final Rule, consists of two Recommended Practices (RPs) by the American Petroleum Institute (API):

- API RP 1170 "Design and Operation of Solution-mined Salt Caverns used for Natural Gas Storage" (September 2015);
- API RP 1171 "Functional Integrity of Natural Gas Storage in Depleted Hydrocarbon Reservoirs and Aquifer Reservoirs" (July 2015).

We have seen that operational and safety considerations related to gas storage fall under the jurisdiction of both the States and the Department of Transportation, in particular the PHMSA. However, the States have authority over intra-state underground gas storage sites and they can implement their own rules applicable to these facilities, provided they are stricter than the minimum standards, and compatible with them. Many states, especially those with a long history of oil and gas exploitation, thus have specific regulatory programs for storage sites that focus on well integrity issues. Due to the greater public attention on problems resulting from plant failures, and in conjunction with recent improvements in federal regulations, the States are now required to review their rules with the aim of strengthening them and to enact new autonomous rules. This is notably the case in California following the Aliso Canyon⁹ accident in 2015: the California Geologic Energy Management Division recently finalised new rules which have served as models for certain other States undertaking the process of updating their underground gas storage programs. In addition to following the PHMSA's Final Rules, incorporating API RP 1170 and 1171, a number of practices have also been incorporated into the new rules, in particular requirements for risk management, emergency response planning and the execution of robust testing. Other regulation programmes at the federal and state level deal with the discharge of products of concern into the air, groundwater, surface water, and soil (methane¹⁰, volatile organic compounds, toxic substances, etc.).

The technologies for creating and operating storage facilities are mature, and technical and regulatory recommendations exist to help industries and regulators design and operate this type of facility. As noted earlier, recent American federal regulations specifically reference the API RP 1170 and 1171 practices. Two documents drawn up in the wake of the Aliso Canyon accident also provide an excellent toolkit for the design and operation of the facilities, the assessment of the problem, and the response to be provided following a failure. In 2016, a report entitled "Ensuring Safe and Reliable Underground Natural Gas Storage" was prepared by a federal task force formed by the Departments of Energy and Transportation, with recommendations to reduce the likelihood of leakages in gas storage facilities. Another report, published in 2019, presents the results of a detailed investigation assessing the Aliso Canyon incident and the root causes of the failures.

Finally, API RP 1170 makes recommendations for solution-mined salt caverns. This covers the main stages of their design, construction, and operation. It details specific engineering practices, in particular the mechanical integrity tests, geological and geomechanical assessments, well design, drilling operations, saline solution extraction operations, gas storage operations, monitoring of the integrity of the cavern, and its abandonment procedure.

3.3.3 Great Britain's regulatory framework

The competent labour inspection authority for occupational health and safety, the Health and Safety Executive or HSE, regulates safety at salt cavern gas storage sites using legislation covering four distinct areas.

<u>The first area is land use planning</u>: establishments wishing to hold stocks of certain hazardous substances above a threshold quantity¹¹ must contact the "Hazardous Substances Authority" or HSA which is the local planning authority.

⁹ As a reminder, this was a massive gas leaks from a storage facility in a depleted reservoir, brought under control several months later by injection of mud through an emergency drilling intersecting the leaking well.

¹⁰ In the U.S., methane is considered to be an atmospheric pollutant and is regulated under the *Clean Air Act* (which is not the case for hydrogen).

¹¹ For information, this threshold quantity is 15 tons for natural gas and 3 tons for hydrogen.

<u>The second area is the control of major accident hazards</u>: the main health and safety legislation covering gas storage facilities is the 1999 regulation COMAH (*Control of Major Accident Hazards Regulations*). COMAH creates a framework for the regulation of facilities where there is a risk of major accident for people and the environment. Operators of COMAH facilities are required to demonstrate that they are taking all necessary measures to prevent major accidents involving dangerous substances and limit the consequences to people and the environment of any major accidents which occur. Regarding the storage of gas in salt caverns, the HSE recommends that the European norm BS EN 1918: 1998 be adopted, in particular parts 3 (functional recommendations for storage in leached salt caverns) and 5 (functional recommendations for surface facilities). Finally, the safety reports must take into account all external events that could lead to a major incident, and in particular the following elements:

- the effects of foreseeable hazards, such as earthquakes and ground movements, on salt caverns and surface facilities;
- the geological characteristics of the region, which must be considered in a sufficiently detailed manner to provide a clear understanding of the geological processes that have formed the area in question, the relation between the cavern and other existing or future caverns, and the potential for seismic activity.

<u>The third area is control and containment of pipeline risks</u>: this area is not relevant to the subject of this report.

<u>The fourth area is well and drilling safety</u>: a drilling site is defined as any place where there is an activity or operation related to the extraction of minerals by drilling. Since the drilling to realise a salt cavern was initially intended for salt extraction, the regulation of 1995 on the safety and operation of boreholes applies from the beginning of the on-site operations, and continues to apply throughout the life of the facility until the abandonment of the well.

4 Specific features of underground hydrogen storage in salt caverns (F. Hadj-Hassen, C. Coquelet, J. Corvisier, F. Golfier, D. Grgic, L. Truche, F. Jorand)

4.1 Thermodynamic properties of hydrogen

The thermodynamic behaviour of hydrogen and helium is characterised by the Joule-Thomson effect. Contrary to other gases (methane, compressed air, etc.), the temperature of these two gases increases slightly during isenthalpic expansion, as shown by Figure 4-1.



Figure 4-1. Variation of temperature as a function of pressure during isenthalpic relaxation for hydrogen and other gases (P varying from P_o=22 MPa to 0.2P_o, T_o=40°C)

The difference in density and compressibility between these gases means that to store the same mass of gas (in this case a reference mass of methane in a cavern of 300,000 m³ at a pressure of 22 MPa and at a temperature of 40° C corresponding to a depth of 1000 m), the volumes required for each gas

are very different. In particular, for hydrogen, a cavern with a volume that is 10 times larger would be needed to store the same mass at this depth. Figure 4.2 gives the evolution of the volume ratio required for each gas according to the depth of the storage. By way of comparison, the same evolution is shown for a perfect gas.



Figure 4-2. Cavern volumes required to store the same mass of gas M_o as a function of depth (M_o : mass of methane for $P_o=22$ MPa and $T_o=40^{\circ}C$)

4.2 Interaction of hydrogen with materials present in the cavern

At the end of leaching and gas filling, a quantity of saturated brine remains in the cavern, the volume of which is estimated at approximately 5 to 10% of the total volume of the cavern. This brine and the insolubles exchange heat with the stored gas, but other exchanges can also occur in the form of dissolution of the gas in the brine, humidification of the gas, and chemical reactions with the insolubles.

The solubility of hydrogen in an aqueous solution characterised by a given salt concentration has been measured in the laboratory at equilibrium under different pressure and temperature conditions. A comparison was made of the results obtained with the other stored gases, especially CO_2 , which has the highest solubility. Figure 4.3 shows the experimental setup used and the main results obtained. The measurement principle is based on the static-analytic method, with liquid phase sampling, and analyses by gas chromatography in order to determine the solubility of the gas in the solution. The experimental data were used to parametrize f three equations of state. The first is based on the use of a cubic equation of state much used in chemical engineering (the binary interaction parameter incorporates the salting out-effect). The second model is a molecular based model where all molecular interactions resulting from the presence of water and electrolytes are taken into account (note that this model allow a very accurate water content prediction of the gas phase). The third model is a geochemical modelling of the gas-water-salt balance using Henry's law and a specific activity coefficient model coupled to the Debye-Hückel Activity Coefficient model. These equations of state can be integrated directly in the gas storage model, or in post-processing in order to assess gas losses by dissolution.



Figure 4-3. Equipment used in laboratory to measure the solubility of hydrogen in aqueous solutions and comparison of the results obtained with other gases (ARMINES/CTP work).

Figure 4.3 highlights two important findings:

- in the conventional range of gas storage pressure in salt caverns (i.e., up to 25 MPa), the dissolution of hydrogen is lower than that of the reference gas which is methane; beyond this pressure, the trend is inversed and the dissolution of hydrogen becomes greater;
- the solubility of the gases is about three times greater in pure water than in saturated brine (the *salting out* phenomenon).

Knowledge of the thermodynamic properties in terms of gas dissolution (composition at equilibrium) and density makes it possible to precisely calculate the quantity of gas stored in the cavern. In the same way, knowledge of the water content of the gas and its potential reactivity with the insolubles allows to determine any changes in the composition of the gas as well as the risks of degradation of the cavern and the well.

Numerical simulations have been made with the help of a geochemical code (i.e. without incorporating transport). They represent the interaction between the gas reservoir and the well by bringing together in the same cell a saturated brine, solid phases (halite and insolubles, in particular sulphates and iron), and a large quantity of gaseous hydrogen. For comparison, additional simulations were made involving other gases of interest in the context of energy storage (CH_4 , CO_2 and O_2). The initial mineral assemblage is composed mainly of halite, but also of anhydrite, dolomite, pyrite, quartz, barite and siderite. The possible formation of secondary minerals such as calcite, hematite and mackinawite (a sulphide of iron and nickel) is also considered.

Halite, which is the main component of salt, is only slightly impacted by the presence of gas, whatever its nature, temperature and pressure. The modelling carried out therefore proves reassuring in terms of safety since in almost all the simulations, the halite precipitates very weakly, presumably after the vaporisation of water in the gas phase. The only cases where halite dissolves (weakly) are those with CH_4 and the highest level of insolubles (11 and 21 % in volume).

As for the insolubles, they are the substances that induce reactivity when combined with the stored gases. The elements that will react are carbon, sulphur, and iron:

- sulphates (barite and anhydrite) will be transformed into sulphides (pyrite and mackinawite) under the action of reducing gases (CH₄ and H₂);
- the carbonates containing magnesium and iron (dolomite and siderite) will transform into calcite under the action of CH₄ and CO₂;

iron will remain as Fe(II) (pyrite and mackinawite) in reducing environments (CH₄ and H₂) or oxidize to Fe(III) (hematite) in oxidizing environments (CO₂ and O₂).

Water appears very logically in the gas phases in variable quantities depending on the affinity of the different gases stored. As for the other secondary gases, CH_4 appears to a minor extent in the storage of H_2 , whereas H_2S appears in the storage of CH_4 in larger proportions (Figure 4.4). As this type of storage in salt caverns has been in operation for several decades, this phenomenon is fairly well known and the facilities are therefore adapted accordingly.



Figure 4-4. Molar fractions of the gases formed in the cases of H₂, CH₄, and O₂, depending on the level of insolubles

4.3 Interaction with the well completion (steel, concrete).

The reactivity of hydrogen as a reducing agent in the natural environment is still poorly known because, until recently, it was assumed to be insignificant. However, there has been a resurgence of interest in recent years within the geoscientific community on the question of redox processes induced by hydrogen, particularly in three main areas of research: i) natural hydrogen production in mid-ocean ridges and in certain continental environments such as ophiolitic massifs and cratons (Abrajano et al., 1990; Salvi et al., 1992; Charlou et al., 2002, 2010; Potter et al., 2013), ii) the synthesis of abiotic hydrocarbons and carbonaceous-nitrogenous organic molecules as well as the development of deep ecosystems (Scherwood Lollar et al., 2002; Takay et al., 2004; Kelley et al., 2005; Proskurowski et al., 2008; Prinzhofer et al., 2018), and iii) the reactivity and transport of hydrogen produced by corrosion in geological storage sites for radioactive waste, or artificially injected in salt caverns or aquifers for energy storage purposes (Truche, 2009; Truche et al., 2013; Reitenbach et al., 2015). These topics share an interest in the mobility of hydrogen in the natural or human-modified geological environment, and all require multidisciplinary approaches that consider both the kinetics of production, the two-phase transport of the molecule in a porous environment, its solubility, and its reactivity in the presence of bacterial and surface catalysts. The scientific motivations are different depending on the field of investigation considered: origin of life, transport of metals, evaluation of energy resources and material flows, destabilisation of the geological environment with modification of the prevailing geochemical conditions. loss or alteration of resources via leaks, and biotic or abiotic interactions with the surrounding formation, etc.

A bibliographical review on the abiotic reactivity of hydrogen in deep geological environments makes it possible to establish an initial inventory of useful knowledge for questions on the safety of the geological storage of hydrogen, and to feed into scientific investigations by analogy with natural cases of hydrogen migration through the lithosphere. It thus appears that the abiotic reactivity of hydrogen is very weak at low temperatures (<100°C) and that there is still no known natural catalyst that is capable of accelerating the sulphate and carbonate reduction reactions in this temperature range. The reduction of Fe(III) to Fe(II), which only requires the transfer of a single electron, has never yet been measured in a concrete

manner at the time scales accessible to the laboratory (up to 6 months at 200°C). The Fe(III) of clays or iron oxides remains stable under these temperature conditions (<200°C), even in the presence of high H₂ partial pressures (up to 100 bar). Currently, only three reactions involving H₂ as a reducing agent are known to be effective at less than 100°C: i) the reductive dissolution of pyrite which can lead to the precipitation of pyrrhotite and release of H₂S, ii) the reduction of nitrates catalysed by stainless steel or native iron, and iii) the reduction of uranyl (UO₂²⁺).

To these redox reactions, it is also necessary to add another form of reactivity that can impact the behaviour of H_2 in the natural environment: the electrostatic interactions with the charged surfaces of the minerals inducing a possible adsorption of the hydrogen molecule. This adsorption has been measured experimentally and observed in its natural environment (clayey halo surrounding the Cigra Lake deposit, Athabaska, Canada). This phenomenon is certainly of low intensity (<0.5% by mass of clay or rock at 25°C) but, taken to the scale of the geological formation, it becomes quite significant. This should be taken into account in the case of cements, where lamellar minerals such as hydrated calcium silicates (CSH) can easily act as a trap for H_2 .

At ambient temperature, the oxidation of hydrogen can only be observed in the presence of microbial catalysts via the hydrogenase enzymes, capable of persisting in the environment. Apart from nitrates, only Fe(III), sulphates and carbonates can act as hydrogen oxidants. The free energies of these reactions, calculated based on the pH, make it possible to estimate the probability of the oxidation of hydrogen via microbial catalysis. It is high at a neutral pH in the presence of bioavailable Fe(III) and sulphates, but remains less probable in the presence of carbonates (formation of CH₄). At a higher pH (>10), a reaction of hydrogen with sulphates and carbonates is not possible.

Regarding the special case of the cement medium, it must be recognised that there is to date no data in the extreme pH range (>11) exhibited by these materials. Concretes seem to be very insensitive to variations in oxidation-reduction conditions due to a fairly limited presence of reactive compounds from this point of view. Moreover, most of the behaviour of these materials takes place in an aerated environment. For conventional civil engineering, there has therefore never been a need to describe these materials under conditions that may be imposed by the geological storage of hydrogen. The presence of hydrogen originating from radiolysis and/or corrosion has never been the subject of a study regarding its impact on the physics and chemistry of hydrated cements. The only studies conducted related to bacterial activity and the presence of nitrates. Many cements contain varying proportions of sulphur at different degrees of oxidation, as well as iron and other metals. However, based on existing work, it is reasonable to assume that the sulphates and carbonates will not be destabilised. Bacterial activity will in all cases be inhibited by the PH level induced by cementation materials, at least as long as the concretes have not been degraded or neutralised to the point of no longer sustaining an alkaline environment. Excluding the presence of nitrates, the main uncertainties are related to a more detailed understanding and the quantification of hydrogen's reactivity in the presence of Fe(III) in a cement medium. At any rate, given the masses of steel - and therefore of Iron (0) - present in the storage site, it is unlikely that the Fe(III) in cement and the clays is sufficient to reduce all of the hydrogen generated by radiolysis and corrosion.

The results of H_2O /cement/ H_2 interaction tests (Truche and Bertrand, 2019) have led to some useful conclusions regarding the reactivity of well casings in a geological hydrogen storage site. A simple dissolution in anoxic conditions of a hydrated powder of CEM V cement showed a rapid release of sulphides in solution (Figure 4-5). Whether at 20°C or at 90°C, this production of sulphides was identical under argon and under hydrogen (10 bar). It stabilises at 0.04 ± 0.01 mol_{S2}/kg_{cement} at 20°C after 800 h of reaction and at 0.06 ± 0.01 mol_{S2}/kg_{cement} at 90°C after 400 h. An increase in the temperature therefore leads to an increase in the concentration of sulphides. It seems that a state of equilibrium between mineralised sulphides and sulphides in solution had been reached. When the cement is in the form of a block, and not a powder, no production of sulphides was measured over a period of 3 months. The very weakly reactive surface of a block of cement compared to powder certainly explains this observation. It would be necessary to reach the liberation mesh dimensions of the micro (nano) phases of sulphides for these to react in solution.



Figure 4-5. Evolution over time of the concentration of sulphides in solution during the leaching experiment of hydrated CEM V Rombas cementitious pastes at 20 and 90°C under Ar or H_2 (10 bar).

(Experiments carried out with powders (100 g/l, particle size: 250 µm) or 1 cm³ blocks)

Analyses of volatile acid sulphides (VAS) carried out on the CEM V cement paste before reaction revealed sulphide contents of around 0.11 mol_{S2}/kg_{cement}.: The mass balance performed on these VAS before and after leaching at 20°C and 90°C was consistent with the production of sulphides in solution by dissolution of the cement VAS in a hyperalkaline condition. Quite clearly, the accessibility of the solution to the micro/nano sulphide phases (elemental sulphur and iron sulphides such as mackinawite, greigite and pyrite) present in CEM V cement is a determining factor in the process of releasing dissolved sulphides in an anoxic alkaline medium (pH>12). Studies on the composition of cementitious pore waters have already revealed significant concentrations of sulphides, sulphites and thiosulphates and concluded that reducing conditions predominate in an anoxic cementitious environment (Lothenbach et al., 2012; Stephan, 2015). The nature of the sources of reduced sulphur remains very poorly identified.

Finally, the presence of hydrogen at 20°C as well as at 90°C has no impact on the mineralogical transformations that cement paste undergoes during leaching. These mineralogical transformations are mainly distinguished by the disappearance of portlandite¹² and ettringite¹² in favour of the precipitation of well-crystallised silico-aluminous phases (katoite¹²).

4.4 Impact of bacterial activity in a salt cavern

The impact of microorganisms on the geological storage of hydrogen in salt caverns remains unknown. The fact that microbial populations are harboured deep underground is beyond doubt. This has started to be documented in particular following the studies aimed at disposing radioactive waste deep underground (Pedersen, 1999). Concentrations of bacterial biomass generally vary at around 10^8 to 10^{10} cells·g⁻¹ of surface soil, diminishing more or less gradually to reach 10^2 to 10^6 cells·g⁻¹ at 2 to 3 km of depth (Lipp *et al.*, 2008, cited by Onstott *et al.*, 2010). These microorganisms are mainly located on the surface of solid elements, within rock pores and fractures. In order to be active, they must be in contact with water where they encounter all the elements necessary for their growth and activity. Outside of an aqueous phase or in conditions incompatible with their multiplication, certain microorganisms can survive for years or even thousands of years (Rothschild and Mancinelli, 2001) but whether such

¹² Respectively a calcium hydroxide, a hydrated calcium and aluminium sulphate, and a hydrated calcium and aluminium silicate.

populations found deep underground remain active or dormant is still poorly documented¹³. To ensure their growth, like all living beings, microorganisms need to find nutritive elements under a form they can assimilate in their environment, such as carbon (about 50% of the dry mass), nitrogen (10-15%, a major constituent of proteins and nucleic acids) or phosphorous (less than 1%, in the form of phosphate) (Figure 4-6).



Figure 4-6. Simplified diagram of global bacterial activity showing the chemical species that can potentially be mobilised to ensure energy conversions (dissimilatory pathways) or the fixation of constitutional elements for biomass (assimilatory pathways) (Jorand, 2018)

<u>Note</u>: a wide variety of microorganisms are capable of maintaining a hydrogenotrophic metabolism (using hydrogen as an electron donor) to satisfy all or part of their energy needs (in red); the required acceptors can equally well be found in oxic, sub-oxic or anoxic environments (in blue).

The microbial world is able to assimilate a wide variety of chemical species such as organic or inorganic molecules (CO_2) for the carbon needs, or nitrates, dinitrogen, or ammonium for the nitrogen needs. If these molecules are not limiting in concentration and if the other physicochemical factors are favourable (in terms of temperature, pressure, pH, etc., see Table 4-1), then growth of microorganisms would be possible. The nutrients must also provide them with the energy necessary for the synthesis of new constituents. This is the role of redox reactions, catalysed by bacteria, which involve an electron donor (the reducer) and an electron acceptor (the oxidizer). If the reaction is thermodynamically possible, it is catalysed by bacteria which convert a variable proportion (about 50 to 98%) to cellular energy. Depending on their metabolic specificity, the bacteria are thus able to draw from a very wide range of organic or inorganic compounds, which is undoubtedly one of the singular features of the microbial world (Figure 4-6). *Bacillus infernus* was thus isolated at a depth of about 2700 m in Virginia: this microorganism is strictly anaerobic, thermophilic (61°C) and halotolerant (growth up to 35 g·l⁻¹), and

¹³ Assuming that these cells are indeed active, their generation time would be on the scale of decades or centuries according to Onstott *et al.* (2010).

capable of developing on lactate or formate by respiring Fe^{III} or Mn^{IV} (Boone *et al.*, 1995). For its part, thermosulfidibacter takaii, a hydrogenotrophic bacterium isolated from the seabed, uses elemental sulphur as an electron acceptor (Table 4-1). Bacterial structures organised as biofilms have also been shown to exist in the subsoil at a depth of several hundred metres (Brown et al., 1994). Hydrogen is also involved as an electron donor in a very wide variety of bacterial species which thus correspond, by definition, to the chemolithotrophs (users of inorganic sources of electrons). Nevertheless, since the use of hydrogen as a source of electrons via the hydrogenases is optional, the majority of these bacteria are considered to be chemoorganotrophs (Gottschalk, 1986). Hydrogen can in this way counterbalance a deficiency in organic sources of electrons and provide the energy needed for the assimilation of CO₂ or N₂ in oligotrophic environments¹⁴. So that, according to Pedersen (1999), "the ultimate limitation for active microbial life is the availability of hydrogen as a source of energy over time". It is therefore legitimate to think that an external supply of hydrogen in environments such as the earth's subsoil, where microbial growth is undoubtedly limited, is likely to stimulate the activity of microorganisms. A recent bibliographic review on the subject (Dopffel et al., 2021), reports studies which show, by isotopic analysis (¹³C), a conversion into methane of 45 to 60% of the hydrogen present in town gas, stored for 7 months in a tank, and incubated with an enrichment of methanogenic bacteria from groundwater (Smigan et al., 1990). It is therefore necessary, as far as possible, to predict the disturbances that these microbial activities would be able to generate in geological hydrogen storage sites.

Factors	Extreme values	Example of bacteria
Hydrostatic pressure (marine or terrestrial)	1 - 1,000 atm (130 MPa): - 5,000 m (terrestrial) - 10,000 m (seabeds) - 1,600 m (marine subsoil)	Bacillus infernus (Boone et al., 1995), potential electron acceptors (Fe ^{III} , SO_4^{2-} , NO_3^- , Mn^{IV}), organotrophs (organic acids, sugars, etc.), halotolerant (0.6 M), thermophilic (65 – 76°C)
Temperature	-5 °C +113 °Cª	Colwellia psychrerythraea Pyrolobus fumarii
Salinity	~ 0 (ultrapure water) 5 M (NaCl)	Rouxiella chamberiensis, Haloarcula marismortui, Dunaliella salina
Acidity/alkalinity (pH)	~ 0 12	Picrophilus spp., Cyanidium caldarium Natronomonas pharaonis
Availability in water	0.75 - 1 0.6 – 1	Bacteria Fungi
Dry region	Sahara desert	Ramlibacter tataouinensis
Radioactivity	10,000 grays ^ь	Deinococcus radiodurans

Table 4-1. Main extreme values of physical and chemical parameters for which bacterial activity has been recorded (Jorand, 2018)

^a the water can remain liquid up to 400°C depending on the pressure. ^b i.e. 1000 times more than what humans can bear.

As already noted, the relative abundance of microorganisms in terrestrial subsoil, as well as their activity can be potentially high, depending on the availability of nutrients (Figure 4-7). Thus, a supply of hydrogen is a source of electrons that is particularly appreciated by many microbial groups, which can lead to undesirable effects in the context of gas storage such as the loss of H_2 , the formation of H_2S (which is corrosive and toxic), CH_4 , or acid, or even to clogging as pointed out by Dopffel *et al.* (2021).

¹⁴ The needs in phosphorus (essentially, phosphates), another major nutrient, can be satisfied by minerals such as apatite or vivianite (Pedersen, 1999).



Figure 4-7. Schematic representation of Table 4-2 reactions expected in the context of the geological storage of hydrogen

<u>Note</u>: C and N are naturally present in the subsoil but in very "energy-intensive" forms so they can be easily assimilated (CO₂, N₂). A supply of hydrogen would thus constitute a sufficient source of energy to allow the growth of various hydrogenotrophic bacteria.

Depending on the nature of the oxidized species present, hydrogen can be used at different rates that can be related to their redox potential and thus to the variation in free energy associated with the hydrogen oxidation reaction. Thus, in the presence of oxygen or nitrate, the expected rate of hydrogen consumption is much higher than with sulphate or Fe^{III} as oxidant (Table 4-2). However, in underground storage sites, the presence of significant concentrations of oxygen and nitrate is not expected, and it is therefore more likely that Fe^{III} and Mn^{IV}, sulphate, and CO₂ constitute the potential electron acceptors for the bacteria in the sites considered.

Table 4-2. Major metabolic groups known to be able to use H₂ as an electron source, with an example of the bacterial genus or species of the group (based on data from Amend and Shock, 2001; Konhauser, 2007).

Hydrogenotrophic metabolic groups	Reaction	∆G⁰ kJ/mole electron (25°C)
Hydrogen-oxidising aerobic bacteria (e.g.: <i>Ralstonia eutropha</i>)	$2H_2 + O_2 = 2H_2O$	-132
Denitrifying bacteria (e.g.: <i>Paracoccus denitrificans</i>)	$5H_2 + 2 NO_3^- + 2H^+ = N_2 + 6H_2O$	-127
Fe/Mn-reducing bacteria ^a	$H_2 + MnO_{2(s)} + 2H^+ = Mn^{2+} + 2H_2O$	-83
(e.g.: Shewanella putrefaciens)	H_2 + 2FeOOH _(s) + 4H ⁺ = 2Fe ²⁺ + 4H ₂ O	-55
Sulphate-reducing bacteria (e.g.: <i>Desulfovibrio</i> <i>desulfuricans</i>)	$4H_2 + SO_4^{2-} + 2H^+ = H_2S + 4H_2O$	-38
Methanogenic bacteria (e.g.: <i>Methanobacterium subterraneum</i>)	$4H_2 + HCO_3^- + H^+ = CH_4 + 3H_2O$	-17
Acetogenic bacteria (e.g.: <i>Clostridium acetium</i>)	4H ₂ + 2 HCO ₃ ⁻ + H ⁺ = CH ₃ COO ⁻ + 4H ₂ O	-13

<u>Key</u>: ΔG^0 = Gibbs free energy under standard conditions

^a The bacteria respiring Fe^{III} and Mn^{IV} are generally the same, there is virtually no specificity with respect to these chemical species, unlike sulphate-reducing bacteria with sulphates.

The kinetics of hydrogen consumption under pressures of 3 to 100 bar, by *Shewanella oneidensis* in the presence of Fe^{III} citrate as an electron acceptor, vary from 5.4 to 5.6 10^{-13} mM_{H2}·h⁻¹·cell⁻¹, at 30 °C independently of pressure (Hazotte, 2012)¹⁵. To date, no measurements of this rate of consumption in a saline environment have been published (studies in progress). Although bacterial activities have been documented for extreme salinity values (Figure 4-8), the data are much more limited when it comes to anaerobic metabolisms such as those that interest us here for hydrogenotrophic organisms. Indeed, since osmolarity regulation is very energy-intensive, bacterial growth in a hypersaline environment is only efficient for energetically "profitable" metabolisms such as oxygen respiration (Oren, 2011). It is therefore expected that with regard to hydrogenotrophic anaerobic metabolisms, which, moreover, are autotrophic for the most part, this growth is very weak and/or concerns little or no bacterial activity, at least under hypersaline conditions. As such, methanogenic activities have been described up to 100 g·I⁻¹ in NaCl while acetogenic, sulphate-reducing or iron-reducing bacteria are shown to be active up to about 250 g·I⁻¹ (Figure 4-8) and up to 90°C (Dopffel *et al.*, 2021).



Figure 4-8. Maximum values in salt (NaCl) where bacterial activity has been observed, according to their metabolic group, in laboratory or field investigations (adapted from Oren, 2011)

The bacteria thus react differently, depending on the species, to high NaCl concentrations. As an example, Figure 4-9 shows the effect of salinity on the growth of *Shewanella oneidensis* MR-1 and *Klebsiella mobilis* in a rich medium, clearly demonstrating that *Klebsiella* has a stronger tolerance to high concentrations than *Shewanella*.

As a result, the possibility of anaerobic hydrogenotrophic activities occurring in the saline storage conditions cannot be excluded, but these should be much lower than in low osmolarity media.

¹⁵ These values should be treated with caution since they were carried out in the laboratory under optimal conditions with, in particular, a soluble form of Fe³⁺ (Fe^{III} citrate). They therefore greatly overestimate the suspected field values.



Figure 4-9. Growth curves as a function of the culture time for <u>Shewanella oneidensis</u> MR-1 (a) and <u>Klebsiella mobilis</u> (b) cultured in a rich medium (tripticase soy broth, aerobic) under different concentrations of NaCl (from 5 to 100 g·l⁻¹ in addition to the concentration of the TSB medium)



Another point to consider in salt caverns, apart from the high salinity, is the low specific surface of the solid supports likely to accommodate active bacterial biofilms, in contact with the aqueous phase and the gas. The specific surface area relative to the volume of gas stored, compared to the specific surface area available in the porous media, allows a much lower exchange surface, which should result in a much lower quantity of biomass. Consequently, the bacterial problem of hydrogen storage in a salt cavern will concern more the production of corrosive H_2S than an exhaustion of the stored gas. Controlling the availability of sulphates may be an approach to follow in order to limit the activity of sulphate-reducing bacteria. Certain solutions often proposed in the context of bacteria-related biocorrosion concern the supply of nitrate which will most certainly impact the activity of sulphurogenic bacteria but which risks promoting the activity of other hydrogenotrophic bacteria such as denitrifying bacteria (Table 4-2) or nitrite-producing bacteria, which can lead to certain corrosion mechanisms in steels (Etique et al., 2014). Consequently, limiting all bacterial activity in the storage conditions should be the basic rule. For this, and insofar as possible, we must limit on the one hand any intrusion of exogenous microorganisms to the site and, on the other, any nutrient supply (including carbonate, N_2 , phosphate, and organic matter). Since sulphates are relatively abundant in the underground environment and are highly water-soluble, it is undoubtedly illusory to hope to eliminate them entirely. On the other hand, the addition of ferrous salts can help to precipitate the sulphides into FeS and thus limit their diffusion with the gas.

4.5 Hydrogen leaks and the impact of hydromechanical couplings

The mechanical behaviour of rock salt is dominated by viscoplastic deformation processes with a marked tendency towards positive strain hardening. At low average effective stress, the salt exhibits dilatant deformation due to the damage it has suffered (opening of cracks), which can thus lead to an increase in permeability by several orders of magnitude (Popp et al., 2001; Peach and Spiers, 1996). The dilatancy threshold which corresponds to the threshold for the unstable propagation of cracks is often used for the dimensioning of salt caverns. Conversely, the closing process ("self-healing") of cracks, resulting from dislocation movements such as sliding, diffusion at grain boundaries, and the recrystallisation processes at a microscopic scale, can induce a significant and almost irreversible reduction in permeability (Sutherland and Cave, 1980; Peach, 1991; Chen et al., 2013; Zhu and Arson, 2015). This delayed process must therefore also be taken into account via creep tests when studying the integrity of the rock salt seal.

Moreover, salt is an unconventional reservoir rock with very low permeability and porosity, and micrometric to nanometric pore sizes. When gas flows at low pressure through these small-diameter pores, the collision of the molecules with the walls induces slippage. The lower the pore size and gas pressure, the greater the gas slippage effect. The apparent permeability (k_a) measured by Darcy's law

is then greater than the intrinsic permeability (k_{∞}) related to the connected geometric porosity. Therefore, the study of gas flow in such a material must take into account this effect, also called the Klinkenberg effect (Klinkenberg, 1941; Letham and Bustin, 2016). Because of the very low porosity of salt, the gas pressure must only very slightly modify the effective stress and more especially the value of the Biot coefficient which defines the extent of the poroelastic coupling within the material. This poroelastic parameter should also be characterised in order to correctly interpret the influence of the mechanical loading on the salt's gas permeability.

We thus conducted a series of laboratory experiments on samples of rock salt to study its sealing property under mechanical loading (isotropic, deviatoric) in order to assess the potential risks of leakage. The salt samples used were taken at a depth of 530 m from the salt bed of the Alsace potash mines within the Grand Est region, dating from the Sannoisian-Oligocene geological stage. This salt is mainly made up of halite crystals, ranging in size from several millimetres to several centimetres, with a random orientation, interspersed with marls and anhydrite crystals. The mineralogical analyses show on the Figure 4-10 (left) images of clear and phenoblastic or milky and xenomorphic halite crystals, some flat and elongated anhydrite crystals, and occasional dark marls . The porosity corresponds mainly to crystal joints and isolated microcracks. The middle picture shows a cylindrical salt sample (100 mm diameter, 200 mm length) fitted with strain gauges for hydromechanical testing. The colour varies from pale yellow to brown and dark grey, depending on the impurity content (up to 5%). On the right is the distribution of voids in this salt sample, obtained from 3D X-ray tomography. These voids are oriented at random and occur mainly in isolated clusters.



Figure 4-10. Left: SEM analyses of a salt sample (a, b: high magnification images; c: BSE map; d: EDS map). Middle: photo of a cylindrical salt sample fitted with strain gauges for hydromechanical testing. Right: distribution of voids in a cylindrical salt sample obtained from 3D X-ray tomography.

Figure 4-11 represents the evolution of the deformations during the cyclic compression tests in uniaxial and triaxial conditions on four different samples (denoted 3, 4, 5, and 6). Our experimental curves show the same mechanisms and present the same characteristics as those of other similar laboratory tests described in the literature. The mechanical behaviour of this salt is (visco)plastic with a marked tendency towards positive strain hardening. The axial deformation is linear from the start of loading regardless of the confining pressure (0, 1, 5 MPa), thereby indicating the absence of significant initial cracks. The elastic-plastic limit is very low in all tests and the axial deformation curve is non-linear practically from the start of mechanical loading.



Figure 4-11. Stress-strain curve for uniaxial (tests 3 and 4) and triaxial (tests 5 and 6) compression tests

The crack initiation threshold corresponds to the end of the linearity of the lateral deformation curves. At the point of inversion of the volumetric deformation curve, there is a transition between compression (closure of cracks and reduction in pore volume) and dilatancy (reopening of cracks, microcracking and increase in pore volume). Starting from this dilatancy threshold, crack propagation becomes unstable and the deformation is localised. In this unstable condition, the crack propagation process is then controlled by the rate of growth of the crack, with this growth progressing even if the applied stress remains constant. This threshold then corresponds to the long-term resistance of the rock. The crack initiation threshold and the unstable fissuring threshold, corresponding to the dilatancy limit, both tend to increase with the confining pressure, which is a classic phenomenon for rocks. Before the dilatancy threshold, the irreversible deformations are isochoric (without volumetric deformation) since they are essentially plastic, as already shown by Thorel (1994) on the same rock salt. Starting from the dilatancy threshold, the damage to the material is responsible for the emergence of irreversible dilating volumetric deformations. The induced cracks are generally vertically oriented and cylindrical symmetry is globally preserved. Under uniaxial loading conditions, the dilatancy threshold of this rock salt is about 10 MPa which is consistent with the results obtained by Thorel (1994). In the dilating domain, the progressively accumulated damage during loading is responsible for the reduction in positive strain hardening (Schulze et al., 2001), which increases the permeability. In the non-dilating compaction domain, the microcracks are compacted, closed or even healed, and further microcracking is no longer possible, thus decreasing the permeability (Schulze et al., 2001).

The microcracking mainly develops at low confining pressures (0 and 1 MPa). When rock salt sample 4 (under zero confining pressure P_c) was completely unloaded at the end of the test, considerable permanent (irreversible) deformations were observed, as previously shown by Liu, et al. (2014) using cyclic uniaxial compression tests. These non-recoverable inelastic deformations can result from the propagation and movement of dislocations in the crystals, from the mass diffusion, as well as from microcracking for the low confining pressures. Since non-recoverable volumetric deformation is dilatant, dilatancy damage by microcracking is the dominant mechanism during loading. For a higher confining pressure ($P_c = 5$ MPa), the dilatancy associated with the volumetric deformation decreases to almost zero, thus indicating the absence of microfracturing. When this sample was completely unloaded at the end of the test, highly significant irreversible deformations were observed. Since the non-recoverable volumetric deformation is almost equal to zero, only the isovolumetric plastic mechanism was involved during loading. In the ductile regime (fully plastic), the propagation and movement of dislocations within the crystals lead to strain hardening of the material which is due, at the microscopic level, to the

increasing number of dislocations and interactions developing between them as well as with various microstructural obstacles during the deformation (Yahya et al., 2000). The behaviour of halite (rock salt) can become fully plastic (ductile), even at room temperature, if the average stress is high and the deviatoric stress much lower than the maximum strength, which can virtually eliminate microfracturing (Thorel, 1994; Aubertin et al., 1993) and therefore the dilatancy of the material. From the ductile transition onwards, macroscopic rupture of the samples tested was no longer observed despite very significant axial deformations. The ductile transition in halite is obtained at confining pressures of the order of 5-10 MPa (Senseny et al., 1992; Handin et al., 1986; Liang et al., 2007), which is coherent with the value of approx. 10 MPa found by Thorel (1994) for rock salt from the Alsace potash mines. For higher values, the influence of the confining pressure on the plastic deformation decreases rapidly (the stress-strain curves are scarcely higher).

The unloading-reloading curves are very linear and much more rigid than the loading curve, even for the lower cycle, which means that irreversible mechanisms (plasticity, closure of voids) develop almost from the start of loading. The elasticity constants (Young's Modulus, Poisson's coefficient) estimated from the unloading curves are dispersed from one sample to another. There is no obvious influence of the stress (confining pressure, deviatoric stress) on these elastic constants, as many authors have already shown (Senseny et al., 1992; Schulze et al., 2001; Thorel, 1994), even though the slopes of the unloading stress-strain curves are expected to decrease due to microcracking damage.

It is not easy to determine the dilatancy threshold using strain gauges during a uniaxial/triaxial compression test, especially with a material such as salt. To improve the determination of this threshold, the velocity of the ultrasonic waves was measured during a uniaxial compression test on the salt (sample 9 of Figure 4-12). These wave velocities are very sensitive to microcracking and allow the onset of dilatancy be detected more precisely, and earlier than with conventional measurements of change in volume. It is supposed that the material, which is initially isotropic, becomes transverse isotropic when damaged, thus requiring the measurement of five independent wave velocities in order to determine the five independent elastic coefficients. Figure 4-12 shows the evolution of the P and S elastic wave velocities, the dynamic coefficients of the elastic compliance tensor (Young's moduli E11 and E33, Poisson's coefficients v_{12} , v_{13} and v_{31}) and the Thomsen dimensionless anisotropy wave factors $P(\epsilon)$ and S(y) during the uniaxial compression test on the sample denoted 9. The decrease in the VP₀ $^{\circ}$ velocity (direction perpendicular to the axis of the specimen and therefore to the applied axial stress) based on an axial stress of 11 MPa indicates that the main deformation mechanism is the opening of the microcracks, oriented vertically in the direction of axial stress, at least for the low confining pressures. The dilatancy threshold, which can be detected very precisely with this method, is a tipping point from which a significant increase in the permeability (several orders of magnitude) may occur as a result of the unstable propagation and coalescence of the cracks. After full unloading, VP_{0^o} was less than its initial value, which confirms that the damage is irreversible. The VP_{90°} velocity (in the direction of the applied axial stress) begins to decrease very slightly starting from a much higher stress (18 MPa) because this velocity hardly registers the opening of microcracks that are parallel to the direction of the applied axial stress. The evolution of E_{11} and E_{33} indicates that the material becomes more rigid in the axial direction and more flexible in the lateral direction, which confirms the opening of microcracks parallel to the direction of the applied axial stress. Thomsen's dimensionless anisotropy factors gradually increase during loading (especially ε) due to all the deformation mechanisms (closing and self-healing of pre-existing microcracks, plasticity with hardening in the axial direction, preferential opening of axially oriented microcracks).

The poroelastic coupling is almost negligible in this salt because the Biot coefficient is close to zero. Indeed, an increase in the gas pressure in the pores does not induce any significant increase in volume and does not decrease the average stress *via* the concept of effective stress. Therefore, this concept does not apply for this salt for which effective stress = total stress, meaning that during the gas permeability measurements, the increase in the pressure of the gas in the pores and cracks does not decrease the effective confining pressure, which remains equal to the total confining pressure.

Figure 4-13 on the left shows for sample 5 (triaxial compression test at Pc = 1 MPa) the evolution of the apparent permeability to gas (helium) as a function of the deviatoric stress and pressure. We can clearly observe the Klinkenberg effect for all of the deviatoric stress values, i.e. a decrease in permeability with the increase in gas pressure. Figure 4-13 on the right shows the evolution of the apparent permeability to gas as a function of the deviatoric stress for different samples (uniaxial and triaxial compression tests at different confining pressures). For all of these tests, the permeability was measured at the same gas pressure (5 bar). For some tests, the permeability was measured continuously (samples 7 and 8) while for others (samples 2, 5, 6, 10), it was only measured for certain deviatoric stress levels.



Figure 4-12. Evolution of P and S elastic wave velocities, dynamic elastic constants (E11, E33, v13, v31, and v12) and Thomsen's anisotropy factors (ε and γ) during a uniaxial compression test.

It should be noted first, in Figure 4-13 on the right, that the initial permeability of the salt samples tested exhibits a large dispersion over almost three orders of magnitude. In some cases, the initial permeability is very high (up to more than 10⁻¹⁷ m²) compared to the classic expected value for healthy saline rock. These significant differences are related to a very random initial cracking of the salt samples. For the most initially damaged samples, and therefore the most permeable ones, the Klinkenberg effect was absent in the permeability measurements. In addition, a moderate increase (of one order of magnitude) of the permeability is observed with the increase in the deviatoric stress when the confining pressure is low (0-1 MPa). On the other hand, for a higher confining pressure (5 MPa), the permeability barely changes with the increase in stress because the salt becomes even more viscoplastic and is much less damaged.

In the context of hydrogen storage in deep salt caverns, an increase of the permeability of about one order of magnitude may favour hydrogen leaks at the walls of the salt cavern, that is, where the average stress is lowest. However, this must be put into perspective given the low extension of the damaged zone near the wall.



Figure 4-13. Evolution of the apparent gas permeability as a function of the deviatoric stress and the pressure for sample 5 (triaxial compression test at 1 MPa of confinement, on the left) and for different samples (uniaxial and triaxial compression tests at different confining pressures, on the right).

5 Design of an underground hydrogen storage facility in a salt cavern (F. Hadj-Hassen, M. Karimi-Jafari, A. Réveillère)

5.1 Design of the surface installations

The hydrogen discussed in our study comes solely from the electrolysis of water powered by renewable electricity (wind and solar). The electrolyser is therefore a central element of the installation. Currently, there are three main electrolyser technologies: alkaline, PEM (Proton Exchange Membrane), and SOEC (Solid Oxide Electrolyser Cell). The first is a mature and widely distributed technology in the industry The second is more recent and based on fuel cells. It is currently only available for small installations. The last technology operates at high temperatures and is still at the laboratory stage: it differs mainly by the substitution of a part of the electricity required to dissociate water by heat, which improves the chemical energy/electrical energy efficiency ratio.

As an energy carrier, hydrogen can be used in a wide range of applications, in particular for the production of electricity (Power to Power), transportation (Power to Mobility), industry (Power to Industry), and the natural gas network (Power to Gas). Figure 5-1 shows the hydrogen circuit studied, with the upstream production by water electrolysis; the storage in salt caverns in the centre; and the four downstream industrial applications.



Figure 5-1. Production of hydrogen by electrolysis and fields of application (source: SMRI)

<u>Power to Power (P2P)</u>: this configuration involves hydrogen storage in times of electricity overproduction compared to demand, with the aim of producing electricity again (via turbines or fuel cells) when demand increases. The periods of high consumption are also accompanied by an increase in the market price of electricity, which is consistent from an economic point of view. Two peaks in consumption and price often recur daily, in the morning and the afternoon. The variant chosen corresponds to the consumption of 200 MW with the two following time slots: from 7:00 to 11:00 and from 17:00 to 21:00. The cycling is thus daily and rapid, with 8 hours of withdrawal (2 x 4 h) and 16 hours of injection.

<u>Power to Mobility (P2M)</u>: hydrogen is currently used for so-called captive fleets, such as forklifts, busses, or taxis, it is an excellent fuel and seems to be a cleaner alternative to energy storage by battery for electric vehicles. This involves installing a fuel cell that uses ambient air and hydrogen transported at

very high pressure (350 or 700 bar) in the vehicle, to supply the electricity needed to run the electric engine. The short recharge time (3 min to fill up) and the great autonomy with reduced weight and size make it a competitive solution compared of electric vehicles. The chosen variant also corresponds to a daily cycle, but slower, with 12 hours of withdrawal and 12 hours of injection (for example, withdrawal in the evening and injection during the day). This cycle intends to meet a demand of 310 t of hydrogen per day for a market such as Germany.

<u>Power to Industry (P2I)</u>: the hydrogen is mainly used in refineries for oil desulphurization, but also in the production of electronic components, ultra-flat glass, metallurgy, and the food industry. In total, industries consume 1 Mt/year of hydrogen in France and 61 Mt/year in the rest of the world. The chosen variant corresponds to the situation where a cavern replaces a hydrogen reforming plant that has been shut down, with a flow rate of 100,000 Nm³/h. The period needed to resume the plant activity has been set at one month and this scenario is repeated twice a year (1 month of withdrawal and 5 months of injection).

<u>Power to Gas (P2G)</u>: this process consists of reinjecting small amounts of hydrogen into the gas network, or transforming it into methane by recycling industrial carbon dioxide (methanation). The chosen configuration consists of withdrawing hydrogen during the 6 months of high demand (winter), and injecting it during the 6 months of lowest demand (summer), which corresponds to a seasonal cycle. A variant involving storage at low depth has also been considered for this configuration.

Table 5-1 summarises the characteristics of the chosen storage scenarios for each industrial application after their validation by numerical simulation of the geomechanical behaviour of the storage.

Configuration	Power to Power	Power to Mobility	Power to Industry	Powe	r to Gas
Scenario	4+4 h withdrawal 16 h injection	12 h withdrawal 12 h injection	1 month withdrawal 5 months injection	6 months 6 month	withdrawal as injection
Cavern depth (m)	800	1300	1300	350	1300
Power delivered (MW)	200	-	-	-	-
Electrolysis power (MW)	260	1100	60	3	25
Withdrawal flowrate (kg/s)	2.68	5.7	1.54	0.015	0.13
Hydrogen mass (t)	77	246	3992	241	2 022
Injection flowrate (kg/s)	1.34	5.7	0.31	0.015	0.13
Initial stress (MPa)	17.6	28.6	28.6	7.7	28.6
Cavern maximum pressure (MPa)	10	17.5	17.5	5.5	19
Cavern minimum pressure (MPa)	6	13	9.5	2.5	7
Cavern volume (m³)	60000	116000	798000	100000	300000
Tubing diameter (")	8.5	8.5	7.5	7.5	7.5
Cushion gas mass (t)	222	1186	4708	200	1684

Table 5-1. Parameters of the storage scenarios chosen for the four industrial applications

The hydrogen leaves the electrolyser at 3.5 MPa and 25°C. Since the hydrogen injection pressure in the salt cavern is greater than that of the electrolyser outlet, the gas must be compressed. Compression is sometimes also necessary at the cavern outlet in order to facilitate the transport of the gas and prepare it for its dedicated application. The compression ratio should not exceed 3, that is, the outlet pressure should be less than three times the inlet pressure. The temperature in the compressor must not exceed 150°C to prevent damage. As a result, in some configurations, it will be necessary to cool the hydrogen at the compressor outlet with heat exchangers, and to use several compression stages.

The hydrogen withdrawn from the cavern must first be treated in order to eliminate the sulphur, then dried. A purifier and a dryer are therefore installed at the outlet of the cavern, which operate with the heat recovered from the compression outlet by the exchangers. In the P2P configuration, a turbine is used - more economically attractive than a fuel cell - to convert the hydrogen back into electricity.

The modelling of the surface facilities was done with the ASPEN HYSIS software which is commonly used in the industry, particularly in the field of process thermodynamics.

By way of illustration, Figure 5-2 gives the thermodynamic hydrogen injection and withdrawal chains for the scenario chosen in the Power to Mobility application. This scenario was the subject of a further study by Geostock.



Figure 5-2. Thermodynamic hydrogen injection and withdrawal chains in the cavern for the scenario chosen in the Power to Mobility application

Table 5-2 brings together the parameters of each storage scenario, the surface facilities associated with it, and the electrical power mobilised.

Configuration	Power to Power	Power to Mobility	Power to Industry	Power	to Gas
Scenario	4+4 h withdrawal 16 h injection	12 h withdrawal 12 h injection	1 month withdrawal 5 months injection	6 months withdrawal 6 months injection	
Cavern depth (m)	800	1300	1300	350	1300
Injection flowrate (kg/s)	1.34	5.7	0.31	0.015	0.13
Withdrawal flowrate (kg/s)	2.68	5.7	1.54	0.015	0.13
Power consumed during injection (MW)	2.6	17.4	0.9	0.1	0.4
Power consumed during withdrawal (MW)	175.0	21.0	-	-	-
	5 compressors 12.5 MW 1 compressor 2.6 MW	2 compressors 8 MW 2 compressors 7 MW 1 compressor 9 MW	2 compressors 0.5 MW	1 compressor 11 kW	2 compressors 0.2 MW
	7 exchangers	6 exchangers	2 exchangers	1 exchanger	3 exchangers
	1 combustion room	-	-	-	-
	1 treatment device NOx	-	-	-	-
Surface installation	1 condenser	-	-	-	-
	1 pump 1MW	-	-	-	-
	1 turbine 185 MW	-	-	-	-
	1 turbine 40 MW	-	-	-	-
	Purifier	Purifier	Purifier	Purifier	Purifier
	Dryer	Dryer	Dryer	Dryer	Dryer
	Safety valves	Safety valves	Safety valves	Safety valves	Safety valves

Table 5.2. Energy balance of the chosen storage scenarios

5.2 Characterisation of the salt

The advantages of salt, which are at the origin of the interest in it for the underground storage of energy, must not overshadow its geomechanical behaviour, which is very specific and marked, above all, by the creep phenomenon. Below, we examine this thermomechanical behaviour more closely, knowing that until now the assumption of sealing was generally accepted. The diffusion and permeation aspect of hydrogen in salt was addressed in the project and the preliminary results obtained are presented.

In addition to measuring physical properties (density and speed of sound) and thermal properties (conductivity, volumetric capacity, expansion coefficient), the characterisation of salt in laboratory also includes Brazilian tests¹⁶ to measure the tensile strength, uniaxial and triaxial compression tests with loading/unloading cycles to determine the elastic parameters as well as the damage caused by dilatancy, and finally triaxial creep tests using deviatoric stress and temperature stages. Figure 5-3 illustrates the results of a triaxial compression test.



Figure 5-3. Results of a triaxial compression test with measurement of the dilating deviator

The triaxial creep test consists of several conventional steps performed on the same sample at constant stresses and temperatures, and separated by phases of changes in the deviatoric stress or temperature.

¹⁶ This is a diametral compression test, also known as "indirect tensile test".

The duration of each step can vary from one to several weeks. An example of a creep test is shown by Figure 5-4

There are many laws to describe salt creep as a function of time, the deviatoric stress, and the temperature. Lemaître's power law is commonly used. Thus, during a multi-stage creep test, the total axial deformation corresponds to the sum of an elastic deformation, an irreversible viscoplastic deformation (given by Lemaitre's law), and a thermal expansion deformation:

$$\epsilon = \frac{q}{E} + \left(\frac{q}{K}\right)^{\beta} ex \, p\left(A\left[\frac{1}{T_{ref}} - \frac{1}{T}\right]\right) t^{\alpha} + \alpha_{th}(T - T_0)$$

with:

- ϵ : the total axial deformation;
- q: the deviatoric stress;
- E: Young's modulus;
- α , β and K: the parameters of Lemaitre's law describing salt creep;
- A: the Arrhenius factor accounting for the effect of temperature on creep;
- T: the temperature, its initial value is T_0 , and T_{ref} is a reference temperature;
- t: time

5.3 Coupled modelling

Modelling a gas storage in a salt cavern is a complex problem, especially when account for a field comprising several types of operations: caverns in brine or in liquid hydrocarbons or in gas. In our particular case, the problem consists in simulating the thermodynamic behaviour of an isolated salt cavern in interaction with the surrounding massif. As shown in Figure 5-4, the storage system can be divided into four subsystems: the well (1) connecting the cavern (3) to the surface, and the surrounding massif, itself divided into two subsystems, the block around the well (2) and the block around the cavern (4).



Figure 5-4. Creep test on salt with three deviator stages and at constant temperature (5, 10, 15 MPa, $T = 30^{\circ}C$)



Figure 5-5. Salt cavern storage system and coupling of associated phenomena

A dedicated software, DEMETHER, was developed by Armines/Géosciences to solve thermodynamic problems related to the storage of fluid energy carriers in underground caverns.

In order to demonstrate the principle of modelling, comparative simulations of the storage of difference gases (methane, helium, hydrogen and compressed air) were carried out for the case of a spherical cavern with a volume of $300,000 \text{ m}^3$ at a depth of 1000 m. Common values were used to describe the salt rheology and the initial stresses and temperature conditions. The effects of the well and the insolubles and residual brine are disregarded. Leaching is not taken into account and at time t_o , the temperature and the pressure in the cavern are equal to the initial values, the cavern being full of gas whose mass is determined according to the volume of the cavern and the properties of the gas stored at these pressure and temperature values. The loading imposed on the four storages consist of a discharge of 50% of the initial mass of each gas in 60 days, then a cycle around this mass is run for 40 years with withdrawal, injection, and pause phases having a unit duration of 30 days.

The analysis of the modelling results and the stability assessment are usually done according to the following criteria (Figure 5-6):

- the pressure in the cavern must remain within the authorised range defined by a minimum value estimated at 0.2 P_o (P_o being the geostatic pressure assuming an isotropic initial state) in order to avoid excessive salt creep and the risk of damage by dilatancy, and a maximum value estimated at 0.8 P_o in order to avoid hydrofracturing of the salt, particularly at the roof of the cavern and in the chimney;
- the absence of tensile stresses noting that salt, like all geomaterials, does not withstand tension;
- the absence of dilatancy by examining the average and deviatoric stress diagram;
- the loss of cavern volume must be limited and must be below the threshold of 1% per year;
- the extension of viscoplastic deformations in the cavern walls.

As shown in this figure, the tangential stress at the cavern wall becomes positive after a few operating cycles and therefore reflects the development of tension. The amplitude of this tension increases as cycling progresses. The two extreme storages situations are always with methane which leads to the strongest amplitude of this tension, and hydrogen which leads to the lowest. This result is a direct consequence of the thermal behaviour where the sharp drops in temperature during the withdrawal lead to the development of this tension. One of the possible solutions to overcome the risk of salt fracturing by tension consists in changing the stresses applied to the cavern. This can be done in two different ways: change the amplitude of these stresses or their period, or even both at the same time.



Figure 5-6. Behaviour of hydrogen storage in a salt cavern compared with the storage of other gases.

Modelling the storage site using the DEMETHER software makes it possible to establish pressure and temperature profiles at the cavern walls throughout its lifetime. From a design perspective, it must be supplemented by 2D or 3D thermomechanical modelling to account for the complex geometry of the cavern or network of caverns as well as the contribution of the other rocks around the salt formation and the movements induced on the surface. The obtained temperature and pressure profiles will then serve as input data. Based on the same reasoning, Armines/Géosciences developed the 3D finite-element numerical code PHIMEF which ensures a strong coupling of the different physics involved (thermal, hydraulic, and mechanical). Figure 5-7 illustrates the modelling carried out using this software for the Spindletop hydrogen storage cavern in the U.S. (Air Liquide).



Figure 5-7. 2D asymmetric numerical model, zooming in on the cavern mesh

5.4 Laboratory storage pilot

All the models currently used to study the thermodynamics of gas storage in salt caverns assume that the cavern is in thermodynamic equilibrium with a constant spatial distribution of pressure and temperature. The heat exchanges taking place between the insolubles, the residual brine, and the stored gas are taken into account in the modelling, but not the mass exchanges described in chapter 4 (dissolution of the gas in the brine and its humidification by evaporation of water). The instrumentation of the caverns being difficult to achieve, and in the best case could only be done vertically along the well axis, a reduced-scale storage pilot was developed in laboratory. The replica comprises at its base a brine container and is equipped with thermocouples and humidity sensors to describe the thermodynamic behaviour during the different injection/withdrawal cycles applied.

One of the main difficulties in designing this pilot was to find the appropriate similarity laws that would make it possible to reproduce the behaviour of a real large-scale cavern in a reduced-scale pilot. This task was achieved using the available data for an instrumented cavern in Germany (Staßfurt S107 with a volume of 288,000 m³) which has been subjected to injection and withdrawal cycles separated by periods of pause. A numerical CFD model on COMSOL was developed to reproduce the measurements obtained on site (Figure 5-8), and then used to define the specifications of the laboratory pilot (Figure 5-9).



Figure 5-8. CFD model of the Staßfurt cavern in Germany



Figure 5-9. Storage pilot developed in the laboratory

6 Technical-economic analysis (S. Jallais, F. Hadj-Hassen)

6.1 Description of markets and scenarios

In the previous chapter, four main storage configurations in salt caverns were identified in relation to the possible applications of hydrogen produced by water electrolysis. These are the production of electricity to meet peak demand (Power to Power, P2P), mobility with the use of fuel cells (Power to Mobility, P2M), the needs of the chemical industry (Power to Industry, P2I) and finally the injection of hydrogen into the gas network or the production of syngas by methanation (Power to Gas, P2G). For each of these configurations, variants were investigated in order to achieve storage at different depths, and with variable operating scenarios ranging from a daily to a seasonal cycle. The detailed analysis of each variant and the numerical simulation of the geomechanical behaviour of the storage site made it possible to retain, for each envisaged configuration, at least one solution for which the parameters have been adjusted in the different operating phases of the storage site: flowrates, pressures, temperatures, tubing, etc.

This work was pursued in order to determine, for each industrial application, the required surface installations and their interactions with the salt cavern storage site. With the exception of the first application which specifically uses a turbine or a fuel cell for the production of electricity, most of the equipment used in all the applications consists of compressors, exchangers, and drying and purification stations (Table 5.1).

This section will continue analysing these storage configurations, starting with the economic assessment of each adopted scenario. This assessment was based on a simple model in which the expenses were divided into two parts:

- operational expenses (OPEX): the cost of the electricity consumed was estimated based on the power rating of the machines used and the current price per kWh; annual maintenance costs are also included in the OPEX;
- investment expenditures (CAPEX): the investments correspond to the price of the machines, calculated based on their respective power ratings (using a model where costs per kWh are estimated) for each type of machine (compressor, electrolyser, etc.); the equipment renewal expenses based on the estimated lifetime of the various components are also taken into account.

The revenues are estimated based on the hydrogen sales price (or price of electricity in the case of Power to Power) multiplied by the production rate of the installation. To assess the profitability, the Total Levelized Cost (TLC) is determined. This cost corresponds to the price at which hydrogen must be sold to achieve an Internal Rate of Return (IRR) of 10%.

The costs retained for the infrastructures differ from one configuration to the other, depending on the industrial application envisaged. The CAPEX for these infrastructures includes their purchase and installation, and their OPEX include maintenance and renewal at end of life. The costs of the civil engineering installations and their management have also been taken into account (40 % of initial CAPEX). Table 6-1 summarises the main infrastructures, their costs, and their lifespans.

Cost (€/kW)	Lifespan (h)
16,000 P ^{0.44}	300,000
300	300,000
100	40,000
negligible	300,000
negligible	300,000
600	80,000
1000	80,000
negligible	300,000
100 €/m³	300,000
	Cost (€/kW) 16,000 P ^{0.44} 300 100 negligible negligible 600 1000 negligible 100 €/m ³

Table 6-1	Infrastructure	costs and	lifespans	(source: Air	· Liquide)
	masuucuie	costs and	mespans	(Source, Ail	Liquide)

The selected electrolyser is of alkaline type, requiring 50 kWh to produce 1 kg of hydrogen. This choice corresponds to the most mature technology and the most economic one, since a proton exchange polymer electrolyte membrane (PEM) electrolyser is at least twice as expensive for the same power generated. The price of electricity per MWh was set using intraday data provided by the Energy Regulation Commission, based on the EPEX SPOT stock market¹⁷. The injection and withdrawal phases were designed to take advantage of the lowest possible electricity purchase prices in order to make the hydrogen storage as profitable as possible. These prices vary from one application to the next. The averages used are respectively €43.1/MWh for the first two applications (P2P and P2M), €52/MWh for P2I and finally €55/MWh for P2G. The duration of the project was set at 30 years with 8000 hours per year of operation.

The initial investments at year zero represent a significant part of the sales price of hydrogen and electricity. On the one hand, this significant investment is partly due to the costs of electrolysers and/or fuel cells, particularly in the P2P configuration. Added to this is the limited lifespan of these installations, the replacement of which (up to 4 replacements in 30 years for the electrolyser membranes) significantly increases the CAPEX.

For configurations requiring lower hydrogen flow rates (P2I and P2G), the power requirements of the electrolysers are lower, hence their smaller contribution to the CAPEX. An equipment price sensitivity study was carried out. The TLC for selling electricity in the P2P configuration was established as a function of the cost of the electrolyser per unit of power. It appears that the TLC of the electricity can only fall below ≤ 100 /MWh when the electrolyser is purchased at less than ≤ 160 /kW (compared to a current purchase price of ≤ 600 /kW).

The maintenance of equipment, calculated at 4% of CAPEX per year, has been included in the OPEX, as well as the purchase of electricity according to the prices given above. Figure 6-1 summarises the various costs involved for each storage configuration considered, and shows the CAPEX breakdown.



Configuration	P2P/PAC	P2P/Turbine	P2M	P2I	P2G/350 m	P2G/1300 m
OPEX (M€)	45	40	191	9	0.4	4

Figure 6-1. Costs involved for each storage configuration considered, with a detailed CAPEX breakdown.

¹⁷ EPEX SPOT is the exchange-based electricity market covering France, Germany, Austria and Switzerland (<u>https://www.statkraft.fr/produits- et-services/glossaire/</u>)

After having updated the OPEX and the replacement CAPEX, the TLC for the sale of hydrogen or electricity produced by each configuration was defined. The calculated prices include the compression and expansion at the cavern outlet to achieve the parameters defined in the study of the surface installations (Table 6-2).

Configuration	P2P/PAC	P2P/Turbine	P2M	P2I	P2G/350 m	P2G/1300 m
CTA (€/kg)	-	-	3,5	3.3	9	5,4
CTA (€/MWh)	250	188	-	-	-	-

Table 6-2. TLC for selling hydrogen and electricity for each configuration

The main conclusions to be drawn from this economic assessment can be summarised as follows:

- in the P2P configuration, with minimal sales prices of €188 and €250 /MWh respectively (depending on whether a turbine or a fuel cell is used), profitability is not assured; according to the French transmission system operator RTE, the price of wholesale electricity on the EPEX-SPOT market in France exceeded €150/MWh only 6 times in 2018, and for periods not exceeding 4 hours; this configuration is therefore positioned on energy that is significantly more expensive than the annual price peaks;
- with a hydrogen selling cost of €3.50/kg at 70 MPa, the P2M configuration proves to be interesting; at the end of 2018, the pump price per kg of hydrogen was around €12 after transport and compression (source: Air Liquide); hydrogen at €3.50/kg, excluding transportation and margin, is therefore already a competitive solution for the mobility market;
- for the P2I configuration, hydrogen is resold at a minimum price of €3.30/kg; however, hydrogen for industry is today mostly produced by SMR (Steam Methane Reforming) where the costs are estimated at €2/kg on average (source: Air Liquide); added to this price is a carbon tax of €0.03/kg of CO₂ produced, i.e., at the rate of 10 kg of CO₂ for 1 kg of H₂, €0.30/kg of carbon tax; the salt cavern solution for hydrogen is currently not optimal but the outlook is optimistic;
- for the P2G configuration, the hydrogen can be sold at between €9/kg and €5.40/kg for caverns respectively situated 350m and 1300m deep, which makes this method poorly competitive vis-à-vis current methods; indeed, the price of SMR hydrogen being €2.30/kg on average (carbon tax included), it appears that the distribution of hydrogen in the gas network and for methanation is still not economically efficient.

Today, therefore, the storage of hydrogen produced by electrolysis and stored in a salt cavern only appears to be truly profitable for the P2M configuration. The P2P and P2G configurations are very uncompetitive in the current state of technological progress, and the P2I configuration may well become more interesting in the future - particularly in a scenario where the price of the carbon tax increases.

6.2 Spindletop storage feedback

The Spindletop cavern, located in Texas (United States) is currently the largest hydrogen storage facility in the world with a volume of 906,000 m³ and an energy capacity of 274 GWh. The 8,230 t of stored useful hydrogen mass makes it possible to replace the production of a steam reforming unit of 120,000 Nm³/h for a duration of 30 days (back-up of the pipeline network). The cavern is located to the east of the Spindletop oil field which also includes gaseous and liquid hydrocarbon storage. It is connected to Air Liquide's network of hydrogen pipelines in Texas, which is 545 km long.

The salt dome within which the cavern is located is almost circular with a diameter of about 1600 m. The cap rock of the Spindletop salt formation is a disk-shaped mass of anhydrite, gypsum, and limestone, but it does not completely cover the salt. The salt roof lies at a depth of 549 m, while that of the cap rock lies at 485 m. As for thecavern, which is cylindrical, its roof lies at about 1220 m depth and its footwall at about 1486 m. Its centre therefore lies at an average depth of about 1350 m with a maximum diameter of about 71 m (Figure 6-2).



Figure 6-2. Profile of the Spindletop cavern recorded by the August 2004 sonar scan (Source: Air Liquide)

Creation of the cavern began in March 2010 and ended in August 2014, i.e. a total leaching duration of about 1620 days. Some very limited interruptions occurred following the obstruction of the tubing by insoluble sulphates. In September 2010, a cavern reconnaissance operation was carried out for two weeks. The hydrogen filling phase began in June 2015, i.e. after a rest phase of about 310 days following the end of leaching, and it continued for 460 days until September 2016. The first withdrawal took place in November 2016 following a second rest phase of 65 days.

Figure 6-3 shows the brine and hydrogen pressures measured from the start of filling in June 2015 until the end of 2017. The history of the cavern and its operating mode were established based on this figure. The annual cycle used includes two withdrawal phases of 1 month each separated by a rest phase, also of 1 month. The second withdrawal is followed by an 87-day pause before the injection is carried out for 130 days. The cycle ends with another pause of 58 days.

In addition to the wellheads and GPS stations available on the Spindletop site, forty subsidence measurement stations have been installed in the storage areas. Between January 2010 and February 2018, eight measurements were carried out with an almost annual frequency. The average speed of subsidence of the surface at the hydrogen storage cavern is about 1.6 cm/year.



Figure 6-3. Brine and hydrogen pressures after the cavern filling phase (on the left) and the selected operation cycle (on the right) (source: Air Liquide)

A numerical modelling of the hydrogen storage was carried out in two phases, assuming that the cavern is totally isolated in view of the distances that separate it from the other hydrocarbon storage caverns. The purpose of the first phase is to establish the history of the pressure and temperature at the wall of the cavern with the cycling envisaged, from its creation until the end of its operation, that is to say over a period of 30 years. During the second phase, the real geometry of the cavern is considered, and the profiles of pressure and temperature previously calculated are imported in order to study the behaviour of the cavern while ensuring a strong coupling between the thermal and the mechanical aspects.

The assessment of the stability of the cavern is based on the following criteria:

- the pressure must fall within the two thresholds that guarantee its integrity, that is, the maximum filling pressure P_{max} and the minimal emptying pressure P_{min} as a function of the initial stress P_0 : $P_{max} < 0.8P_0$ and $P_{min} > 0.2P_0$
- the major principal stress must always remain compressive and no tensile stress is tolerated;
- the dilatancy criterion must be validated at any time during operation of the storage;
- the cavern must not lose more than 20% of its volume over the 30 years of operation envisaged;
- any expansion of the viscoplastic deformation induced within the rock mass must be limited.

The results obtained show that the annual operating scenario envisaged (two successive withdrawal phases of 1 month and a long injection phase of approximately 4 months, all separated by pause phases) allows to ensure the stability and the integrity of the cavern over the required period. Figure 6-4 gives an illustration of this result.



Figure 6-4. Maximum value of the major principal stress (on the left) and maximum deviation between the deviatoric stress and the dilatancy upon contraction criterion (on the right) (Source: Air Liquide)

7 Risk analysis and environmental monitoring (A. Thoraval, P. Gombert, P. de Donato, E. Lacroix)

7.1 Risk analysis

In spite of the many advantages, the gas storage in salt cavern is not exempt from risks, due in particular to the significant gas pressures (about 200 bar for a cavern located at 1000 m depth), highlighted by some past accidents (Brouard, 2002; Bérest and Brouard, 2003; Evans and West, 2008; Bérest et al., 2019). In the case of hydrogen, risks will be amplified due to its high mobility, its reactivity with certain materials and chemical species as a reducing agent, as well as its flammability.

The risk analysis carried out for the project has made possible the identification of dangerous phenomena likely to occur in such a storage site, as well as the barriers that make it possible to avoid or limit the consequences of the identified accident sequences (Balouin et al., 2015). It fits here in the following framework:

- we consider a storage site in the current operating phase that is to say only the injection and withdrawal operations; the storage construction and testing phases (drilling, leaching, tests), the phases of shutdown or work on wells during operation (sonars, workovers, etc.), and the postexploitation phase (including the well closure) will be the subject of specific analyses;
- we consider a generic storage site model; the analysis performed is not intended to replace that specific to each site that the operator has to operate; as a result, we will not carry out a detailed risk assessment, which would entail taking into account the real vulnerabilities specific to each site, but instead will carry out an assessment of the accident sequences that may possibly occur on a typical installation;
- the generic vulnerabilities that are of interest in this study are those related to public safety and the environment i.e. the vulnerabilities external to the site. As such, the vulnerabilities internal to the company (health and safety of workers, goods and infrastructures internal to the site, activities internal to the site, etc.) are not taken into account here.

The analysis is based on a detailed description of the storage site, the operations carried out during the operating phase, the vulnerabilities (vulnerable entities in the event of major accidents), potential sources of disruption or danger (as well as the possibility of overcoming them), and on the feedback available. In a second part, we will discuss the impact on a water table of any potential hydrogen leaks from deep cavern storage.

7.1.1 Description of the storage and its environment

The "system" on which the analysis is carried is shown schematically in Figure 7-1. It is composed of the following elements:

- <u>a cavern</u>, created by dissolution within a mass of salt and used for the storage of hydrogen; it may have been originally used for the storage of another gas (as natural gas) and then converted to hydrogen storage, or it may be a newly created cavern for this purpose; being in the operating phase, the cavern is considered to be filled with hydrogen, apart from its lower part, where insolubles and residual brine are found;
- overburden, assumed to be made up of alternating layers of low-permeability formations (of the clay or marl type) and more permeable formations (of the limestone or sandstone type), the latter possibly hosting aquifers; to cover the variety of hazards likely to be encountered, we will assume the presence of two aquifers (one, located immediately at the top of the salt, containing salt water, and the other, in the subsurface, containing fresh water) and a water-sensitive formation, which may be at risk of dissolution (e.g. an salt formation located at an intermediate level) or at risk of hydration (e.g. anhydrite);
- <u>a well</u>, connecting the surface to the cavern, through which the cavern has been leached; it is covered, over most of its height, with steel casings, cemented to the ground and suspended on the surface in a drilling wellhead (DW); here we assume the presence of three casings: a surface casing (SC) protecting the surface aquifer, an intermediate casing (IC) protecting the water-sensitive formation, and a production casing (PC) anchored in the salt; under the shoe of the last cemented casing, we assume the presence of a section left uncovered and leading into the cavern; the well is equipped with a simplified well completion arrangement, similar to those used for the storage of natural gas, namely:

- a work column (WC), anchored to the base of the PC, through which hydrogen circulates during the injection and withdrawal operations;
- an annular obturator, making it possible to isolate a space between the PC and the WC, used in particular for the detection of leaks; it is assumed that this annular space is filled with a completion fluid such as inert water;
- a safety valve located about 30 m below the surface, controlled from the surface via a hydraulic line (SCSSSV - surface-controlled subsurface safety valve), and intended to secure the well during an incident or intervention at the wellhead;
- an operating wellhead (OW) or "Christmas tree", fixed on the DW, which includes the suspension device for the work column, all the outlets, valves and sensors necessary for controlling the flow of products entering and leaving the well; it makes it possible to lower the tools needed for inspection and maintenance operations (corrosion inspections, cavern sonar scans, etc.).



Figure 7-1. Diagram of the well as considered in this study (Source: Ineris)

Figure 7-2 situates the cavern in its environment by distinguishing between the surrounding hydrogeological context, the storage site and the environment outside the storage site. This figure also details the vulnerabilities, external disruptions and potential dangers.



Figure 7-2. Location of the cavern in its environment (Source: Ineris)

The sources of disruptions that could affect the safety of a hydrogen storage facility are comparable to those of a natural gas storage facility. They can be grouped in two categories:

- <u>sources of aggressions linked to the natural environment</u>, such as floods, rising water tables, lightning, forest fires, seismicity, landslides of natural origin (shrinkage-swelling of clays, landslides, instability of natural underground caverns), extreme weather conditions (frost, heat wave, wind, etc.). Examples can include: a dangerous phenomenon (ignited jet, explosion) occurring on an installation or infrastructure likely to reach the wellhead by domino effect; a third-party drilling operation reaching of the cavern or the well; a ground movement likely to destabilise the cavern or damage the well (whether of natural origin or related to the mechanical instability of a neighbouring cavern);
- <u>sources of aggression linked to the human environment</u>, coming from infrastructures (roadway, railway, or airway networks), industrial activity, existing or closed mines, intrusions, malice, etc. Examples include: a mechanical disruption following the fall of an object (vehicle, machine, crane, plane, wind turbine in the case of a wind farm built near the cavern); a thermal disruption; wellhead overpressure linked to an ignition or explosion affecting a hydrogen pipeline or tank, an electrolyser, a compression unit, a processing unit, etc.

Because this is a generic study, the surface installations will not be described in detail, but it is necessary to identify those likely to interact with the system being studied, either as a source of aggression, or as a generic issue to be addressed. The installations most likely to be encountered within the surface area affected by the hydrogen storage cavern are:

- installations not specific to underground hydrogen storage:
 - o compressors;
 - o treatment units (drying, purification, etc.);
 - storage of hydrocarbons;
 - o brine basins;
 - storage of nitrogen (for inerting);
 - o pipelines (overhead or underground) carrying water or brine;

- overhead or underground electrical lines and equipment;
- installations specific to underground hydrogen storage:
- electrolyser (if the hydrogen production is carried out at the storage site);
- hydrogen buffer storage;
- renewable energy generation unit (photovoltaic panels, wind turbines), if the electricity production is carried out at the storage site;
- o overhead or underground hydrogen pipelines;

7.1.2 Identification, characterisation, and reduction of potential hazards

The main potential hazards can be listed as follows:

- <u>hydrogen</u>: its minimum ignition energy in air is 17 µJ (in stoichiometric proportion), which is significantly lower than that of natural gas (290 µJ), and its ignition range is very wide (from 4 % to 75 % in volume), resulting in a very high probability of ignition in air; hydrogen also has other characteristics: it leaks easily due to its small size and low viscosity, permeates through materials, causes embrittlement of certain metallic materials (ferritic steels), and interacts with the underground environment in a way that is likely to produce ancillary gases (including H₂S);
- <u>gas pressure</u>: excessive pressure in the cavern may provoke rupture of the salt at the walls, which would favour leakage of the stored product; on the other hand, if the gas pressure is too low, the cavern can close on itself (by rupture or excessive creep of the wall salt), leading to a risk of deformation (or even rupture) of the overburden and consequent subsidence or collapse of the ground at the surface;
- <u>geological formation that can induce toxic secondary gases by interaction with hydrogen</u>: these may be banks of insolubles on the cavern wall or in the cover, containing sulphur-containing minerals in particular (gypsum, anhydrite, pyrite, etc.);
- <u>other potential hazards not specific to hydrogen storage in salt caverns</u>: particularly watersensitive geological formations likely to come into contact with the aquifers via the cementation of the well (see chapter 7.1.5).

The approach to reducing potential hazards specific to any risk analysis involves examining the possibility of:

- reducing the volume of stored hydrogen by choosing a smaller cavern;
- reducing the maximum operating pressure or the time spent by the hydrogen in storage at this pressure;
- increasing the minimum operating pressure or reducing the time spent by the hydrogen in storage at this pressure;
- limiting the possibilities for the stored hydrogen to interact by the choosing a salt layer containing few insolubles, operating conditions that inhibit or reduce the presence of bacteria, and/or materials that are inert to hydrogen;
- avoiding, when designing the storage site, choosing a drilling route that traverses water aquifers
 of different qualities, or formations sensitive to water.

7.1.3 Feedback from accidents and incidents

There are only six hydrogen storage caverns in the world: three large caverns in the United States (Texas), managed by Conoco Philips, Praxair and Air Liquide, and three small caverns in the United Kingdom (Teesside) managed by Sabic. They store hydrogen intended for industrial use (chemical industry), with much longer cycles (seasonal and even annual) than those expected for the storage of hydrogen of renewable origin (weekly or even daily). To our knowledge, there are no documents in the public domain mentioning incidents or accidents connected with these storage sites. Furthermore, Air Liquide, partner of the ROSTOCK-H project and operator of the Spindletop storage, has confirmed that there have been no incidents on their site.

This feedback, even if minor, shows that hydrogen can be stored in salt caverns in safe conditions and for long periods of time¹⁸, at least for caverns subject to long-period cycles. This has led us to extend the risk analysis to the field of underground gas and hydrocarbon storage, which are good analogues of underground hydrogen storage. Based on the literature published on the subject (Brouard, 2002; Bérest

¹⁸ Since 1972 in the United Kingdom and since 1983 in Texas (for the first cavities).

& Brouard, 2003; Evans & West, 2008; Charmoille & Thoraval, 2010; Prats, 2013; Bérest et al., 2019), a database was established which lists, to date, 85 noteworthy accidents or incidents that have occurred in the field of gas and hydrocarbon storage. The 11 accidents that seemed most relevant in the context of the underground storage of hydrogen (accidents occurring in salt cavern storage facilities, and originating in the well or the cavern) are presented in Table 7-1. They can be divided into 5 categories:

- risks related to the gas column (4 cases),
- risks related to the brine column (3 cases),
- risks related to the wellhead (2 cases),
- risks related to cementation (1 case),
- risks related to the cavern (1 case),

The main causes of these accidents are: corrosion of the casings, poor cementing, over-filling of the cavern, a leak from the central column and excessive pressure in the cavern leading to geomechanical disorders.

The main measures taken after these accidents were: controlling the state of the casings and cementation section before the caverns are put into operation, standardization of tightness tests (MIT) tests, setting of a pressure limit to ensure a safety margin versus the maximum working pressure, and implementation of a dedicated safety procedure regarding brine completion in case of gas intrusion.

Table 7-1. External accidentology on underground storage of natural gas or hydrocarbons in salt caverns

Risk category	Date, Place, Product	Description of the facts	Consequences	Cause	References
Risks related to the brine column	2012, Manosque,France - Diesel	Rupture of the central column	Not applicable (incident)	Not specified	ARIA n°41801
Risks related to the wellhead	2007, Manosque, France - Fuel oil	Leakage on wellhead equipment	Oil spillage on the surface	Not specified	ARIA n°38242
Risks related to the wellhead	2004, Odessa, Texas, USA - Propane	Failure of a wellhead flange	Release of gas to the atmosphere - discharge of 116.5 m ³ of gas over 4 hours	Not specified	Evans et al. (2008), Hazard. Cargo Bull. (June 2004), N Riley (pers com, 2007)
Risks related to the brine column	2004, Moss Bluff, Louisiane, USA - LPG	Leakage in the brine circuit	Explosion, fire, release of 170 m ³ of gas over 6.5 hours, 360 people evacuated	Rupture by water hammer of the corroded tubing following closure of the well safety valve, linked to the intrusion of gas in the central column	ARIA n°23709, Evans et al. (2008), Hopper (2004), Seni et al. (2005)
Risks related to the gas column	2003, Magnolia, Texas, USA - Natural gas	Leakage through casing	Release of 9.9 Mm ³ of gas in a few hours, 30 people evacuated	Not specified	Evans et al. (2008), Hopper (2004)
Risks related to the brine column	1992, Brenham, Texas, USA - LPG	Well blowout	Explosion, fire, 3 fatalities, 23 injuries, 50 people evacuated, 500 to 1600 m ³ of gas released	Intrusion of gas into the central column due to overfilling of the cavern Failure of safety systems	ARIA n°5244, Evans et al. (2008), NTSB (1993), Berest and Brouard (2003)
Risks related to the gas column	1988, Clute, Texas, USA - Ethylene	Leakage through casing	Underground dispersion lasting 25 days	Not specified	Evans et al. (2008),
Risks related to the gas column	1985, Mont Belvieu, Texas, USA - Propane	Leakage through casing	Explosion, fire, 2 fatalities, 2000 people evacuated	Corrosion of the casing	Brouard (2002), Evans et al. (2008), Berest and Brouard (2003)
Risks related to the gas column	1980, Mont Belvieu, Texas, USA - LPG	Leakage through casing	Explosion, fire, evacuation of 50 families (300 people)	Corrosion of the casing	ARIA n°20712, Brouard (2002), Evans et al. (2008), Berest and Brouard (2003)
Risks related to cementation	1980, Mississippi, USA - Natural gas	Upsurge through the cementation	Underground dispersion	Bad cementation	Evans et al. (2008), Pirkle (1986), Pirkle and Jones (2006)
Risks related to the cavern	1980, Stratton Ridge, Texas, USA - Natural gas	Leakage through cavern walls	Underground dispersion	Rupture of cavern walls	Evans et al. (2008), Hopper (2004)

7.1.4 Approach to the identification of accident sequences

This involves exhaustively identifying the Hazardous Phenomena (HP) that can lead to major accidents induced by scenarios identified in the workgroup: each HP is the result of a Central Feared Event (CFE), located at the heart of the accident sequence and linked to different causes or Initiating Events (IE). Secondly, we provide a list of the (technical or organisational) prevention or protection measures that can be implemented by the operator to deal with the accident scenarios identified. Based on lessons learned from accidentology, the scientific literature, and the expertise of Ineris and other project partners, a generic analysis of the hazards associated with the system studied was then carried out, leading to the development generic "bow tie" methodologies (Figure 7-3) that indicate:

- the feared hazardous phenomena;
- the accident sequences likely to lead to these HPs, highlighting in particular the IE, CFE, and the Secondary Feared Events (SFE) that may follow;
- the main barriers or risk control measures that can be implemented as prevention (prior to a CFE) or protection (following the CFE) in order to block the development of these accidental sequences, to reduce the effects of the HPs potentially generated, or to protect the vulnerable entities before they are affected by these HPs.



Figure 7-3. Simplified representation of an accidental sequence and its potential barriers Key: PRE = preventive barrier, PRO = protective barrier

7.1.5 Central Feared Events related to the well

Because of its complexity and dimensions (several km in length), the well is the weak link in the accident sequence for underground storage. The main CFEs are the following:

- CFEs related to hydrogen leakage:
 - o CFE1 "Loss of hydrogen containment at the production wellhead";
 - CFE2 "Loss of hydrogen containment at the gooseneck";
 - CFE3 "Loss of hydrogen containment at the drilling wellhead";
 - CFE4 "Limited hydrogen leakage into the overburden".
- the CFEs related to the formation of toxic products from the hydrogen:
 - CFE5 "Formation of toxic gases from the hydrogen in the cavern or in the gas completion";
 - CFE6 "Formation of toxic gases at the casings or cementation, or in the overburden" (following a hydrogen leakage); CFE4 is an IE of this CFE.

In addition, there are CFEs related to geomechanical risks (rupture, loss of volume):

- CFE7 "Rupture of the salt in the wall of the cavern exceeding the limits of the layer or the salt dome" (this is a complete rupture of the wall of the cavern up to the roof of the salt);
- CFE8 "Rupture of the pillar between caverns".

There are other CFEs that not related to the stored products, but instead to the quality of construction and the ageing of the outer cementations of the well: "Undesired hydraulic communication between two aquifers of different qualities", "Uncontrolled hydration of a swelling formation (anhydrite, clay)" and "Uncontrolled dissolution of a soluble formation present in the overburden". These CFEs therefore concern any type of well and are not specific to the storage of hydrogen.

7.1.6 Finalisation of the risk analysis for a real storage site.

As an example, Figure 7-4 represents the CFE1 case "Loss of hydrogen containment at the production wellhead". This analysis was carried out for a generic storage site and includes:

- <u>a list of possible causes of each CFE</u>: external aggressions (shock of external origin, external overpressure or thermal domino effects, malice, works, etc.), internal aggressions (on site shock, internal overpressure or thermal domino effects, etc.), procedural causes (operator error, material fault, water hammer, etc.);
- <u>a list of preventive measures</u> (closures, prevention plan, speed limit, anti-water hammer device, thermal protection, pressure detection, etc.) <u>and protective measures</u> (shut-off valves, flame detection, internal operation plan, specialised team, etc.);
- <u>a list of expected consequences</u> (torch fire, explosion of an unconfined cloud of H₂ or UVCE [Unconfined Vapour Cloud Explosion], slow combustion of a cloud of flammable vapours or flash-fire, etc.).



Figure 7-4. Accident sequences related to ERC1 "Loss of hydrogen containment at the production wellhead"

In a real situation, it will be necessary to rank the identified HPs in terms of probability of occurrence and severity of the consequences. A ranking matrix should thus be chosen by the operator in the upstream phase of the risk analysis. This shows two classes of HP: those with estimated effect distances within the site and those with distances beyond the site boundaries. This first classification reveals the critical HPs and those that may have effect distances (as stated in the Order of 29 September 2005) outside of the site.

Certain HPs related to the underground environment are very difficult to quantify in terms of kinetics and/or effect distances, or else they have a negligible probability of occurrence in some contexts. This is the risk of ground collapse (localised or generalised) linked to geomechanical disorders at the level of the cavern, as well as the risk of fire or explosion on the surface linked to gas rising from the ground following a leakage at the well or the cavern. The circular of 10 May 2010¹⁹ proposes, for the storage of gas, hydrocarbons, and chemical products for industrial use (including hydrogen) in the operating phase, a specific treatment of these risks that allows them to be eliminated, subject to compliance with a certain number of criteria or the establishment of a certain number of barriers listed on pages 119 to 122 of the aforementioned circular.

¹⁹ <u>https://aida.ineris.fr/sites/default/files/fichiers/Circulaire_COB_V5b_compact.pdf</u>

7.2 Expected impact of a hydrogen leakage in a shallow aquifer

7.2.1 Experiment carried out

In the event of a leakage in a deep storage site, the hydrogen will migrate towards the surface and meet, in most cases, an aquifer where it can dissolve into the groundwater. Aquifers therefore represent levels of vigilance and alert that are all the more interesting in that, by transporting information from hydrogeological upstream to downstream, they allow the implementation of integrated monitoring downstream of a deep storage site. In the Parisian basin, the aquifers that could fulfil this role are those of the Albian-Neocomian sansdtones and chalk formations. We studied the latter, more superficial one. It contains an aquifer used to supply drinking water to many cities, including Paris. When this aquifer is unconfined, the groundwater is oxygenated and enriched in nitrates from agricultural inputs as well as, locally, in sulphates from the overlying tertiary cover. The intrusion of hydrogen into this type of water will tend to reduce its oxidation-reduction potential and its level of other dissolved gases (N₂, O₂ and CO_2). It could also result in the reduction of nitrate ions to nitrite or ammonium ions (more toxic), especially if the groundwater is in contact with a catalyst metal such as steel or stainless steel of the casings of the drinking water wells, or agricultural or industrial boreholes.

To study this risk, we simulated a sudden and brief release of hydrogen into the chalk aquifer at the Ineris experimental site in Catenoy (Oise). Located about 50 km north of Paris, this site provides access to the unconfined chalk aquifer at a depth of 12 m. The water there has calcium bicarbonate facies and contains nitrate and sulphate ions (Table 7-2). It is also neutral, moderately mineralised and oxygenated, that is, in an oxidizing condition.

The experiments consisted in extracting 5 m^3 of water from this aquifer into a tank, then bubbling hydrogen into it until a concentration of 1.76 mg/L was reached, i.e. 95% of the water saturation under these conditions. This hydrogenated water was then injected into the aquifer at a depth of between 12 and 24 m, and its impact was monitored using 6 piezometers at distances of 5 to 60 m, plus an upstream control piezometer (Figure 7-5).





7.2.2 Results obtained

The bubbling of hydrogen in the tank induced quasi-saturation of the water with hydrogen and a partial degassing of the gases initially dissolved in it, in particular O_2 and CO_2 . Once injected into the aquifer, the concentration of dissolved O_2 in this water was reduced and its calco-carbonic equilibrium was shifted, causing the predominant ions to precipitate and concomitantly varying the conductivity and pH. The main effect, however, was the drastic drop in the redox potential and the presence of dissolved

hydrogen detectable up to 10 m downstream. The maximum impact on the aquifer was observed at the PZ2BIS piezometer located 5 m downstream the injection well (Table 7-2). In addition, the PZ2TER piezometer, located 7 m downstream the injection well, was equipped with an experimental device consisting of infrared and fibre Raman sensors, which also made it possible to monitor the dissolved hydrogen plume with a detectability in the order of 1.7 10⁻¹ mg·l⁻¹.

 Table 7-2. Characteristics of the water chalk 5 m downstream the injection well before and after the experiment

<u>Key</u>: B.Line = baseline, Peak = peak of hydrogen plume, Cond. = electrical conductivity, ORP = oxydo-reduction potential

	H ₂ (mg/L)	NO ₃ - (mg/L)	NO ₂ - (mg/L)	NH₄⁺ (mg/L)	SO ₄ ²⁻ (mg/L)	SO ³⁻ (mg/L)	Cond. (µS/cm)	рН	ORP (mV)	O ₂ (mg/L)
B.Line	0.00	33	<0.02	0.10	27	<0.01	558	7.2	+192	6.24
Peak	0.63	33	<0.02	0.10	27	<0.01	490	7.4	-139	4.17

In contrast, no significant variation, other than natural, was observed in the concentrations of nitrate and sulphate ions nor in their expected metabolites (nitrite, ammonium, sulphide). Note, however, that no element likely to act as a catalyst came into contact with groundwater adjacent to the site, and that the brevity of passage of the hydrogenated plume (a few days) did not allow the development of bacteria likely to react with these elements. During this experiment, the dissolved hydrogen thus behaved as a conservative tracer. However, this result is only valid under the experimental conditions of the test: in the case of a larger and/or longer leakage, it is likely that the physicochemical and hydrogeochemical impacts would be greater in space and in time.

Nonetheless, this shows that it is feasible to monitor aquifers adjacent to future underground hydrogen storage sites. It should take into account and favour *in situ*, continuous and synchronous measurement systems related to the physicochemical parameters of water (redox potential, pH, conductivity, temperature), as well as, in particular, the quantification of dissolved gaseous phases (H_2 , O_2 , CO_2 , N_2). The work done as part of Elodie Lacroix's thesis (2021) on the Catenoy site made it possible to specify the main phases of this metrological approach relating to the potential monitoring of a hydrogen migration in an aquifer. It is summarised here below:

- before any activity, it is necessary to establish a geochemical baseline over several seasonal cycles in order to quantify the natural variations, geometry and dynamics of the system; this phase is essential to assess any subsequent changes linked to a diffuse leakage of hydrogen in connection with the exploitation of the underground storage;
- besides classic physicochemical monitoring, infrared and fibre Raman sensors should be included in a dedicated piezometer, fitted in particular with a semi-permeable membrane (impermeable to water but permeable to gas) with rapid response kinetics (< 5 minutes);
- the installation of the various monitoring piezometers must take into account the flow velocity of the aquifer; in the case of slow flow (< 1 m·d⁻¹), the main piezometers must be placed less than 10 m downstream of a potential leakage zone, taking into account the zones of varying permeability (fractured or cracked zones, paleochannels, etc.);
- the coupled measurements from the infrared and Raman sensors should focus on the quantification of N₂, O₂, CO₂, CH₄, H₂ and H₂O-vapour gases if necessary; with regular maintenance, the detectability of dissolved H₂ is around 1.7 10⁻¹ mg·l⁻¹.

8 Social acceptability of underground hydrogen storage (L. Barbier, M.F. Agnoletti)

8.1 A multifaceted concept

8.1.1 Different levels of acceptability analysis

The concept of social acceptability belongs to the vocabulary of the social sciences, but it is difficult to define unequivocally. It is not very well framed and highly dependent on the situation, the actors (public, business, politician) or the discipline in which it is studied (Batel et al., 2013; Batellier, 2015; Huijts et al., 2012). It has often been considered in societal terms by being qualified as "free, prior and informed consent" (Leblanc & Massé, 2013), as "consensus between the parties involved" (Owen & Kemp, 2013), as a "social license to operate" (Portales & Romero Casteñada, 2016), or as a "process of political assessment" (Fortin & Fournis, 2014). The aspects relating to the user rarely feature in these definitions.

The literature also shows that social acceptability is treated at different levels depending on the objective of the study. Some authors (Fortin & Fournis, 2014) make reference to macro levels where the stakes are directly related to the global development of the project, to economic interests, or to the resolution of conflicts between stakeholders (politician, business, public). In this case, social acceptability can be measured in terms of the benefit-cost or benefit-risk ratio, used to refer to the meso or microsocial levels. Indeed, it can be considered as a process rooted in the individual perception of an object which depends on the social relations, values and meanings, shared between individuals and between groups, in which the object can be integrated or not (Thomas et al., 2019; Wolsink, 2018; 2019).

These different levels must be taken into account and complexify the study of social acceptability. To these difficulties is added the question of the object to which the concept refers. It is not possible to treat in the same fashion the acceptability of using an object or tool (e.g., UTAUT; Vankatesh et al., 2003) and the acceptability of more ambitious projects such as the underground storage of hydrogen. For the first case, social acceptability must be addressed from an individual perspective, that is, related to the intention of use (Terrade et al., 2009), to the utility and usability (Nielsen, 1993). In the second case however, social acceptability must be treated at the meso social level and take into account groups and/or stakeholders, as well as inter-individual factors.

The question of the novelty of the object is crucial. Currently, the issue of underground hydrogen storage is an unprecedented subject of study in the social sciences. There is no scientific literature on the subject and no assumptions can be made about its social acceptability. Thus, it is necessary to study the social acceptability of underground hydrogen storage through related hydrogen research while taking into consideration the inter-individual and/or group dimensions that will or will not lead to acceptability.

8.1.2 Measuring social acceptability

To our knowledge, no studies have been conducted on the social acceptability of large-scale underground hydrogen storage facilities. Studies addressing the public perception of energy storage (Ambrosio-Albala et al., 2019) are still rare and mainly deal with CO₂ storage (Upham and Roberts, 2011; van Alphen et al., 2007). They are often carried out with the help of surveys and press or focus group analysis, thus making it possible to observe a general trend concerning the favourability towards a project. Nevertheless, other research has been conducted on social acceptability and its components on the subject of hydrogen usage: cars (Molin et al., 2007), public transport (O'Garra et al., 2007) or refuelling stations (Huijts and Van Wee, 2015). Some of the methods used by these researchers not only make it possible to observe the favourability of individuals towards an object, but also to determine the factors underlying social acceptability through the use of structural equation modelling.

The research done by Huijts et al. (2012, 2014, 2018) appears to be the most complete and closest to the issue of the underground storage of hydrogen. Their work on hydrogen refuelling stations resulted in a robust conceptual model to study social acceptability that integrates different factors such as trust, affects, attitudes and norms. Other work has highlighted the importance of taking certain sociodemographic variables into account. Indeed, Flynn et al. (2010) and others (Terrade et al., 2009) have shown that individual characteristics (e.g., gender, age, etc.) directly influence social acceptability. In view of these various studies, it seemed to us relevant and appropriate to measure the factors identified by Huijts et al. (ibid.) and to use the socio-demographic variables of the participants to account for the elements that influence the social acceptability of underground hydrogen storage.

8.2 How is the underground geological storage of hydrogen perceived?

As part of the ROSTOCK-H project, a survey was carried out to understand the perception of underground hydrogen storage. The results presented below were produced by processing data collected through an online questionnaire. One hundred and seventy-six participants responded: 99 men, 59 women, and 18 unspecified, with a mean age of 42 years and a standard deviation of 12.1 years. The questions focused on the different factors and variables presented in Table 8-1.

Table 8-1. Set of factors and individual characteristics used to measure the social acceptability of
underground hydrogen storage

Factors	Individual characteristics			
Trust in government	Gender			
Trust in the managing company (of the	Age			
underground hydrogen storage)	Level of education			
Trust in industry	Has (had) a work-related experience of hydrogen			
Trust in scientists	Level of knowledge on hydrogen			
Negative affects	Live near salt cavern used to store hydrocarbons			
Positive affects	Live near underground natural gas storage			
Perception of environmental issues				
Perception of fossil fuels issues				
Importance perceived hydrogen's utility				
Attitudes toward behavioral intention				
Behavioral intention (e.g., acting against/in favor)				
Personal norm (e.g., self-expectation)				
Subjective norm (e.g., social groups' influence)				

8.2.1 Perception of hydrogen and its storage

Of the 176 participants, 85.2% attach importance to environmental problems such as global warming, air pollution, depletion of fossil fuels, and the need to develop renewable energies. However, 4.5% give this little or no importance and 10.2% have no opinion. Furthermore, 79.5% believe that hydrogen can have a positive impact on the energy mix, 6.3% believe not, and 14.2% have no opinion. (Figure 8-1a). With regard to effects on the population, (Figure 8-1b), 71.0% of the participants believe that an underground hydrogen storage infrastructure will be useful to people living nearby. The impact on the local economy is believed by 88.6% to be beneficial but 22.7% of the sample believe that hydrogen could have a negative effect on the health of people living nearby. Then, 28.4% believe that there could be dangers inherent in the infrastructure will have a positive effect on the environment.



Figure 8-1. Answers to the questionnaire on the social acceptability of underground H₂ storage

The majority of individuals are concerned about the environment and see hydrogen as a solution for the energy transition. Nevertheless, 43.8% said they were against an underground hydrogen storage facility, against 37.5% who said they were in favour of it (18.7% have no opinion).

8.2.2 Effects of individual characteristics on acceptability

Among the individual characteristics of the participants, some effects on the factors related to the social acceptability were observed (Table 8-2).

	Gender	Age	Level of education	Work- related experience of hydrogen	Level of knowledge	Live near hydrocarbo ns storage
Trust in government	3 > ♀					
Trust in the managing company		Under 35 yo > over 47 yo > 35-47 yo		Yes > No		
Trust in industry	♂ > ♀					
Trust in scientists				Yes > No	- 🗡 +	
Negative affects	♀ > ♂		- 🔪 +	No > Yes	- 🔨 +	No > Yes
Positive affects				Yes > No	- > +	Yes > No
Perception of fossil fuels issues	♀ > ♂		- 🔪 +			No > Yes
Importance perceived of hydrogen's utility	3 > ♀			Yes > No	- / +	
Subjective norm		Over 47 yo > under 35 yo				

Table 8-2. Effects of individual characteristics on social acceptability factors (only significant effects are presented)

We note that gender influences certain factors since the men have more confidence in politicians and industrialists and they perceive more utility in hydrogen than the women. The latter group express more negative affects towards the underground storage of hydrogen than men, and they have a greater perception of the problems linked to fossil fuels. Regarding age, we observe that those under 35 have the most trust in the managing company and that those over 47 are more sensitive to the influence of those around them (i.e., subjective norm). In addition, the perception of problems linked to fossil fuels tends to decrease as age increases. The higher the educational level, the lower the negative affects towards the underground storage of hydrogen. When the participants' work-related experience of hydrogen, they perceive hydrogen to be of greater utility and put more trust in the managing company and the scientists. They also express more positive affects than people that have absolutely no professional connection with hydrogen. Conversely, the latter group express more negative affects towards the underground storage of hydrogen. When the participants have a good knowledge of hydrogen, they put more trust in scientists, perceive a greater utility for hydrogen and express more positive affects than people with little knowledge on the subject. The latter express more negative affects towards the underground storage of hydrogen. Finally, individuals who live near a hydrocarbon storage site express more positive affects towards the installation of an underground hydrogen storage facility, and less negative affects. In addition, they perceive the problem of fossil fuels as being less important than people living at a distance do.

8.3 Which factors affect the social acceptability of the underground storage of hydrogen?

To identify more precisely the factors that negatively or positively affect the acceptability of the underground storage of hydrogen, we performed structural equation modelling. This was subdivided into two models in order to specify the factors that lead individuals to a positive or negative behavioural intention to the underground storage of hydrogen. This method makes it possible to account for the causal relationships between the different factors (featured in Table 8-1) and to understand their influence on the positive (acceptability) or negative (unacceptability) behavioural intentions of individuals.

8.3.1 Unacceptability factors

The following diagram (Figure 8-2) presents the links between the factors that affect the participants with a negative behavioural intention to underground hydrogen storage. The factors in red are those that actively participate in individuals' negative intention, and the factors in blue are those that moderate this negative intention. The uncoloured factors are variant factors, i.e. they can participate in both the negative intention of individuals, and in its moderation.



Figure 8-2. Simplified diagram of the factors influencing individuals intending to act against underground hydrogen storage

The results show that the subjective norm (i.e. what individuals perceive to be acceptable or unacceptable behaviour by those around them) is the main direct factor that leads individuals to have a negative behavioural intention towards the storage facility. It also seems that the affects (especially the negative ones) have direct impact on the subjective norm.

Conversely, knowledge seems to moderate negative behavioural intentions to underground hydrogen storage. This same factor is positively affected by the personal norm (i.e., the moral and individual obligation to adopt a behaviour according to one's personal belief system; Thøgersen, 2006), and by the perception of the degree of utility of hydrogen. Indeed, this last factor seems to have a key impact within the model since it moderates the negative affects while positively impacting knowledge. In addition, it makes it possible to directly diminish the negative behavioural intention to the underground storage of hydrogen. It results from the (positive) effects of trust and environmental problems, and helps moderate negative affects while positive affects.

8.3.2 Acceptability factors

The diagram of factors affecting the positive behavioural intention of individuals to the underground storage of hydrogen is presented in Figure 8-3. The factors in blue indicate this positive intention.



Figure 8-3. Simplified diagram of the factors influencing individuals intending to act in favor of underground hydrogen storage

Trust in the managing company and politicians has (indirect) positive effects on social acceptability. The perception of hydrogen as being of great utility seems to decrease the negative affects and increase the positive affects. The latter points, as in the personal and the subjective norms, have a direct positive impact on the individuals with a positive behavioural intention. However, no link between the preceding factors and the two types of norm was observed.

In spite of the presence of negative affects in the model, it appears that these have no direct or indirect link to the social acceptability of the underground storage of hydrogen. Finally, no factor moderating the positive behavioural intention was observed.

8.4 Conclusion on the social acceptability of the underground geological storage of hydrogen.

The results revealed that 70.5% of the individuals in the sample surveyed are in favour of underground hydrogen storage, but 62.5% declare a negative behavioural intention towards the installation of such storage. This shows that individuals can be inconsistent with their previously stated attitudes (Ricci et al., 2008). This observation demonstrates the importance of understanding social acceptability as reliant on a set of factors and not on a single dimension. In the case of the underground storage of hydrogen, social norms seem to significantly affect social acceptability, in particular through the subjective norm which has direct effects on both the acceptability and unacceptability of this type of storage in salt caverns The research by Poortinga et al. (2004) has already established that behaviour in connection with the environment may be motivated by contextual and social factors.

In agreement with our study, other research on hydrogen shows that knowledge can influence acceptability (Schmidt and Donsbach, 2016). The perception of hydrogen and/or its underground storage is based on a combination of the experience and knowledge of individuals (Sherry-Brennan et al., 2010). Knowledge constitutes a moderating factor for the participants with a negative behavioural intention to the installation of an underground hydrogen storage facility. We can assume that providing knowledge to individuals reduces their uncertainty (Ambrosio-Albalà et al., 2019) and modulates certain elements of their perception that could lead to decrease social unacceptability. This observation is reinforced by the perception of the utility of hydrogen in the two models, where it acts both as a mediator for social acceptability and a moderator for people with negative behavioural intentions. Furthermore, trust has indirect positive effects on social acceptability, notably through the affects. Positive affects enhance the perception of utility and beneficial consequences on the environment, while reducing the perception of risks (Huijts, 2018). Among the participants with a positive behavioural intention to the installation of an underground hydrogen storage site, the positive affects directly influence their intention. Indeed, these allow individuals to form representations when their level of knowledge is too low (Sütterlin and Siegrist, 2017).

The social acceptability of the underground storage of hydrogen is therefore the result of several factors and characteristics (norms, affects, trust, gender, proximity, etc.) that interlink with each other and are embedded in a specific social or technological context (Sherry-Brennan et al., 2010). Studying the acceptability of the underground storage of hydrogen, and in general of the installation of infrastructures to manage or generate energy, demonstrates the great variety of positions taken by the public. Abric's (2001) research makes it possible to explain these differences in perception by means of three aspects: knowledge, involvement, and experience. As highlighted by Agnoletti (2019), when there is a lack of knowledge on the underground storage of hydrogen, individuals call on knowledge of subjects they are more familiar with (e.g. the storage of gas, shale gas, or nuclear materials) even if these are not factually representative of the underground storage of hydrogen. As a result, the lack of experience and knowledge related to hydrogen prevents the elements constituting the perception from modulating in favour of the installation of underground hydrogen storage (Sherry-Brennan et al., 2010), which can reinforce the activation of social norms (Bordarie and Gaymard, 2015) against the idea of such storage. The study results lead us to conclude that the social acceptability of the hydrogen underground storage by individuals depends on their knowledge (e.g. of infrastructure, applications). In the absence of knowledge, it depends on affects, trust in the parties involved, environmental impact, utility for the planet and individuals, as well as the prevailing social norms.

9 Conclusions regarding the storage of hydrogen in salt caverns

This document constitutes deliverable 6.3 of the ROSTOCK-H project. It summarises most of the work carried out throughout the project and makes it possible to draw up guidelines for the design and management of underground hydrogen storage in salt caverns. There is some feedback from salt caverns that have been storing hydrogen for a few decades already, but it is scant in view of the small number of existing caverns. In addition, feedback from hundreds of salt caverns around the world that store natural gas can also be considered, since it appears that this technology is mature and applicable to the case of hydrogen.

Salt caverns

Salt caverns for the extraction of salt started to be industrially developed in the 18th century in Europe. They have been used to store hydrocarbons since the middle of the 20th century in North America. Today, salt caverns are used to store all types of hydrocarbons (liquid, liquified, gaseous, or supercritical) as well as other products that do not dissolve in salt such as hydrogen, nitrogen, helium and compressed air. In France, there are 80 salt caverns used for storage, some for more than half a century. Worldwide, there are approximately 2,000 storage salt caverns in operation, including 6 for hydrogen. Salt caverns are created by dissolution after an oil drilling-type operation. Several months of "leaching" of the geological salt formation are then necessary to obtain a cavern measuring a few hundred thousand m³. The caverns are not lined, the tightness being ensured by the salt itself. Salt caverns allow gas to be stored at economically attractive densities because the pressures can be high if the salt layer is deep enough: typically up to 200 bar for a cavern at 1100 m depth. Hydrogen storage in a salt cavern, which has existed since 1972, is now subject to a strong resurgence of industrial interest, and new pilot caverns are being built in France, Germany and the Netherlands.

A geomechanical retro-analysis was carried out for the Spindletop salt cavern (Texas, USA) which is currently the largest hydrogen salt cavern storage site in the world. This analysis began by specifying the geological and geomechanical characteristics of the terrain as well as the geometric properties of the cavern. A case history of the leaching, gassing, and first cycle applied made it possible to define a regular operating scenario over a period of 30 years. The geomechanical modelling was carried out in two phases: a first in order to establish the temperature and pressure profiles from the creation of the cavern to the end of operation, and a second in which the true geometry is taken into account, and the previously calculated pressure and temperature records are applied to the wall of the cavern in order to study its thermomechanical behaviour. The results obtained show that the annual operating scenario envisaged (two successive withdrawal phases of 1 month and a long injection phase of about 4 months, all separated by rest phases) makes it possible to ensure the stability and the integrity of the cavern.

Behaviour of salt

The characterization of the thermomechanical behaviour of salt is an essential step in the geomechanical modelling of the storage site. This operation is carried out through a laboratory test campaign on samples taken from the site. Besides the thermoelastic properties and the dilatancy damage criteria of salt, its most striking feature relates to creep. This is described by viscoplastic laws that take into account the effect of the stress deviator and of temperature. The modelling of the storage site itself is difficult to carry out because the only known parameters are measured at the wellhead and it is necessary to apply multiphysics couplings to access the specific parameters of the cavern throughout its history (leaching, gassing, operation and abandonment). A program developed by Armines/Mines Paristech specifically for this application was used to compare the storage of hydrogen in salt caverns to that of other gases (CH₄, compressed air, He). Under the assumption of perfectly sealed salt, the results obtained show that the storage of hydrogen is not particularly different from conventional methane storage. For fast cycles, it may even lead to more favourable stability conditions.

All models currently used to study the thermodynamics of gas storage in salt caverns assume a cavern in thermodynamic equilibrium with a constant spatial distribution of pressure and temperature. It is difficult to install instruments in the caverns, and in the best of cases this is only possible vertically along the well axis. As such, a reduced-scale storage pilot with a container holding brine at its base was developed in the laboratory to verify this hypothesis. This pilot is spatially instrumented with thermocouples as well as humidity sensors in order to describe the thermodynamic behaviour during the various injection/withdrawal cycles. Several storage scenarios have been simulated with this pilot and the results obtained show that for fast cycles, the currently accepted uniformity hypothesis is not entirely coherent and that it is necessary to account for the velocity field in the cavern.

Specificities of hydrogen

The thermodynamic behaviour of hydrogen is characterised by the Joule-Thomson effect in which, contrary to other stored gases (methane and compressed air), an isenthalpic relaxation is accompanied by a slight increase in temperature. However, during the operating cycles of the cavern, the temperature increases during the injection phase and decreases during the withdrawal phase. The difference in density and compressibility between hydrogen and methane means that to store the same mass of gas, a hydrogen cavern with a volume ten times greater than that of methane will be necessary.

At the end of leaching and gassing, a quantity of saturated brine remains in the cavern. This brine and the insolubles stagnate at the bottom of the cavern exchange heat with the stored gas, but other exchanges can also take place in the form of dissolution of the gas in the brine, humidification of the gas and chemical reactions with the insolubles. The solubility of hydrogen in an aqueous solution characterised by a given salt concentration was measured at equilibrium in the laboratory and modelled under different pressure and temperature conditions, then compared to other gases, in particular CO₂, which has the highest solubility. The state equations established can be incorporated directly in the storage model, or be included in post-processing in order to calculate the gas losses by dissolution. Within the conventional range of gas storage pressures in salt caverns, the dissolution of hydrogen is lower than that of methane, which is the reference gas. However, beyond this pressure, the trend is reversed and the dissolution of hydrogen becomes higher than that of methane. The other important result is that the solubility of gases is about three times higher in pure water than in saturated brine.

Furthermore, knowledge of the water content of the stored gas and its potential reactivity with insolubles makes it possible to determine any changes in the composition of the gas and the risks of degradation of the cavern and the well. Numerical simulations using geochemical code have shown that halite is only slightly impacted by the presence of gas, whatever its nature, temperature, and pressure. Water appears in the gas phases in variable quantities depending on the affinity of the different gases stored, CH_4 is in the minority in hydrogen storage, and H_2S appears in larger proportions in methane storage. As this type of storage in salt cavern has been in operation for several decades, this phenomenon is fairly well known and the facilities are therefore adapted accordingly. As for the risk of hydrogen leaks, an increase in permeability of the salt by one order of magnitude could favour leakage at the walls of the salt cavern, i.e. where the average stress is the lowest. However, this must be put into perspective given the small size of the damaged zone near the wall. Thus, although the gas injection/withdrawal conditions may induce damage to the salt near the wall, this remains limited by the salt's highly viscoplastic nature and its capacity to self-heal. Despite the specific properties of hydrogen (high mobility, high diffusivity), storage in a salt cavern remains the safest solution.

Impact of microorganisms

Among the many technological challenges posed by the geological storage of hydrogen in salt caverns, the impact of microorganisms remains unknown. In the absence of clearly established data, it is prudent to follow the recommendations of Dopffel et al. (2021): "For safe underground storage of H_2 gas, we recommend to analyse the relevant microbiological and geochemical characteristics of each site to be able to predict the most suitable storage strategy and establish a good monitoring and mitigation approach to follow and counter potential microbial side effects." However, it seems obvious that the main risk associated with the storage of hydrogen in salt cavern will probably not concern the quality of the gas – contamination by sulphide chemical species, however, cannot be ruled out. Instead, the risk will reside in the impact on the biodegradation of the boreholes and operation structures (casings, strainers, pipes, etc.) made of materials containing iron or concrete, and occurring in particular in the suboxic zones and/or the transition between the anoxic and surface aerobic zones.

Applications of hydrogen

As an energy carrier, hydrogen can be used in a wide range of applications, in particular for the production of electricity, transportation, industry, and the natural gas network. This study concerned green hydrogen produced by the electrolysis of water from renewable electricity. Typical storage scenarios were established for each of these applications based on market need and the depth of the saline formation. The parameters for each scenario were then refined by geomechanical modelling of the storage site.

The storage scenarios corresponding to the four industrial applications of hydrogen (Power to Power, Power to Mobility, Power to Industry, Power to Gas) were the subject of an economic evaluation in which the cost of the electricity consumed was estimated from the power rating of the machines used and the current price per kWh. The investments correspond to the prices of the machines, calculated on the

basis of their respective power ratings, and on the material renewal expenses - themselves based on the estimated lifetime of the various components. The revenues were estimated based on the hydrogen sales price (or price of electricity in the case of this application) multiplied by the production rate of the installation. To assess the profitability, the Total Levelized Cost (TLC) was determined, corresponding to the price at which hydrogen must be sold to obtain an Internal Rate of Return (IRR) of 10% over 30 years with 8000 hours per year of operation. The main conclusion of this evaluation is that the storage of hydrogen produced by electrolysis in a salt cavern only appears to be truly profitable, at present, for the mobility configuration (Power to Mobility). The configurations for the production of electricity and the gas network are highly uncompetitive in the current state of technological progress, and the industry configuration will only tend to be attractive in a scenario where the price of the carbon tax increases.

Risk analysis

Although the gas injection-withdrawal conditions may induce damage to the salt near the wall, the risk of hydrogen leakage remains limited due to the salt's highly viscoplastic nature and its capacity to self-heal. Despite the specific properties of hydrogen (high mobility, high diffusivity), the risk of leakage is limited and storage in a salt cavern remains the safest solution.

Nevertheless, the specificities of hydrogen (high mobility, reactivity with materials, ease of ignition) have led to a revision of the risk analyses traditionally carried out for the storage of gas. This approach made it possible to characterise the main accident scenarios specific to the storage of hydrogen in salt caverns. The potential hazards identified are those related to the characteristics of the products used and their pressurisation, and, depending on the context, those related to the possibility of the formation of toxic gases by interaction with hydrogen (H_2S). The main feared event is the loss of hydrogen containment following the failure of a wellhead element, which is likely to lead to an explosion or fire on the surface. A detailed analysis, taking into account the environment of the storage (surface installations, vulnerable entities), will have to be carried out for each case in order to specify the probability of occurrence and the severity of the consequences of the various hazardous phenomena and to identify the measures needed to prevent or limit the consequences of the identified risks.

Environmental monitoring will have to be set up around the underground hydrogen storage site using the overlying aquifers. These act as levels of vigilance and alert to signal any potential leakage making its way towards the surface. By transporting information from upstream to downstream, these aquifers constitute ideal environments for an integrated monitoring immediately downstream of a deep storage site. Since hydrogen is not normally present in groundwater, its concentration and that of other dissolved gases (N₂, O₂ and CO₂) must be continuously monitored, as will the values of the main physico-chemical parameters (redox potential, pH, conductivity), favouring *in situ*, continuous, and synchronous measurement systems. Work done as part of the project on the Catenoy site has made it possible to specify the main phases of this metrological approach relating to the potential monitoring of a hydrogen migration in an aquifer: establish a multi-annual geochemical baseline, implement traditional physicochemical monitoring, as well as infrared and fibre Raman sensors in a dedicated piezometer, install several monitoring piezometers taking into account the flow rate of the aquifer and its heterogeneities (fractured or cracked zones, paleochannels, etc.).

Social acceptability of the underground storage of hydrogen

The scientific literature on social acceptability often presents attitudes as the main weighed factors. These are very good indicators to assess the favourability of individuals to an object (Eagly & Chaiken, 1993) and are often used in studies on large-scale gas storage, such as CO₂ storage, or for applications using hydrogen. Nevertheless, they do not make it possible to predict a behavioural intention and, as such, an a priori social acceptance. When it comes to effectively using or accepting the introduction of a new element in daily life, individuals may act inconsistently with their previously stated attitudes (Ricci et al., 2008). Social acceptability can therefore not be reduced to a favourability index. Some authors (Hienuki et al., 2019; Itaoka et al., 2017) have shown that the acceptability process is much more complex and intricately tied to other elements. In the case of the study on the underground storage of hydrogen, the gender of the individuals and their familiarity with the object were identified as interindividual variables having direct effects on social acceptability. In addition, inter-individual factors such as trust, affects, knowledge, perceived importance of hydrogen, and social norms appeared as levers that can be used to alter the perception of individuals vis-à-vis the underground storage of hydrogen in order to improve its social acceptability. We have thus been able to show that this is not limited to observing the a priori favourable disposition of individuals, but that it is closely linked to individual variables as well as to intra- and inter-individual factors that can be correlated with each other.

10 Bibliographic references

Abrajano TA, Sturchio NC, Kennedy BM, Lyon GL, Muehlenbachs K, Bohlke JK, 1990. Geochemistry of reduced gas related to serpentinization of the Zambales ophiolite, Philippines. Appl Geochem. 5, 625-630

Abric JC, 2001. A structural approach to social representations. In K. Deaux & G. Philogène (Eds.), *Representations of the social: Bridging theoretical traditions* (pp. 42–47). Blackwell Publishing.

Abuaisha M, Billiotte J, 2021. A discussion on hydrogen migration in rock salt for tight underground storage with an insight into a laboratory setup, Journal of Energy Storage 38, 102589

Abuaisha M, Rouabhi A, Billiotte J, Hadj-Hassen F, 2021. Non-isothermal two-phase hydrogen transport in rock salt during cycling in underground caverns, International Journal of Hydrogen Energy 46(9): 6632-6647. <u>https://doi.org/10.1016/j.ijhydene.2020.11.152</u>

Agnoletti MF, 2019. Effet de la pratique sur les représentations sociales : l'exemple du renouveau minier en Lorraine. In P. Raggi (Ed), Un Après-Mine Imprévu, PUN, 115-132.

Ambrosio-Albalá P, Upham P, Bale CS, 2019. Purely ornamental? Public perceptions of distributed energy storage in the United Kingdom. Energy Research & Social Science, 48, 139-150.

Amend JP, Shock EL, 2001. Energetics of overall metabolic reactions of thermophilic and hyperthermophilic Archaea and Bacteria. FEMS Microbiology Reviews, 25: 175–243.

Balouin T, Kribi S, Prats F, 2015. Formalisation du savoir et des outils dans le domaine des risques majeurs - Étude de dangers d'une installation classée – Ω 9. Rapport INERIS-DRA-15-148940-03446A.

Batel S, Devine-Wright P, Tangeland T, 2013. Social acceptance of low carbon energy and associated infrastructures: A critical discussion. Energy Policy, 58, 1-5.

Batellier P, 2015. Acceptabilité sociale, cartographie d'une notion et de ses usages, Montréal: UQAM.

Bérest P, Brouard B, 2003. Safety of Salt Caverns Used for Underground Storage Blow Out; Mechanical Instability; Seepage; Cavern Abandonment. Oil & Gas Science and Technology Rev IFP, 58, 3 361-384.

Bérest P, Louvet F, 2019. Aspects of the thermodynamic behavior of salt caverns. Proc. SMRI Fall Meeting, Berlin, Germany.

Bérest P, Brouard B, Réveillère A, 2018. L'étanchéité des stockages. Le cas des cavernes dans le sel. Revue Géologues n° 196.

Boone DR, Liu Y, Zhao ZJ, Balkwill DL, Drake GR, Stevens TO, Aldrich HC, 1995. Bacillus infernus sp. nov., an Fe(III)- and Mn(IV)-reducing anaerobe from the deep terrestrial subsurface. International Journal of Systematic Bacteriology, 45: 441 – 448.

Bordarie J, Gaymard S, 2015. Social representations and public policy: Influence of the distance from the object on representational valence. Open Journal of Social Sciences, 3(9), 300-305.

Brouard B, 2002. Etude bibliographique sur les accidents dans les bassins salifères résultants d'une dissolution volontaire ou non. Rapport GEODERIS, 18 mars 2002

Brown DA, Kamineni DC, Sawicki JA, Beveridge TJ, 1994. Minerals associated with biofilms occurring on exposed rock in a granitic underground research laboratory. Applied and Environmental Microbiology, 60: 3182 – 3191.

Chabab S, Théveneau P, Coquelet C, Corvisier J, Paricaud P, 2020. Measurements and predictive models of high-pressure H2 solubility in brine (H2O+NaCl) for underground hydrogen storage application, Int J of Hydrogen En, 45(56):32206-32220, <u>https://doi.org/10.1016/j.ijhydene.2020.08.192</u>.

Charlou JL, Donval JP, Fouquet Y, Jean-Baptiste P, Holm NG, 2002. Geochemistry of high H_2 and CH_4 vent fluids issuing from ultramafic rocks at the Rainbow hydrothermal field (36° 14°N, MAR). Chem Geol 191, 345- 359.

Charlou JL, Donval JP, Konn C, Ondréas H, Fouquet Y, 2010. High production and fluxes of H_2 and CH_4 and evidence of abiotic hydrocarbons synthesis by serpentinization in ultramafic-hosted hydrothermal systems on the Mid-Atlantic Ridge. In: Diversity of Hydrothermal Systems on Slow

Spreading Ocean Ridges. Rona PA, Devey C, Dyment J, Murton BJ (eds). Am Geophys Union, Washington, DC, pp. 265-296

Charmoille A, Thoraval A, Lahaie F, 2010. Synthèse de l'état des connaissances et des pratiques en matière d'abandon des stockages souterrains. Rapport INERIS-DRS-08-86168-00481D.

Chen J et al., 2013. Self-Healing Characteristics of Damaged Rock Salt under Different Healing Conditions. Materials, 6(8), 3438–3450.

Djizanne H, Lahaie F, 2020. Analyse du cadre réglementaire des essais à l'hydrogène prévus dans la cavité EZ53 du site d'Etrez. Rapport Ineris-178376-2224229.

Dopffel N, Jansen S, Gerritse J, 2021. Microbial side effects of underground hydrogen storage – Knowledge gaps, risks and opportunities for successful implementation. International Journal of Hydrogen Energy, 46: 8594 – 8606.

Eagly AH, Chaiken S, 1993. The psychology of attitudes. Harcourt Brace Jovanovich College Publish.

Etique M, Jorand FPA, Zegeye A, Grégoire B, Despas C, Ruby C, 2014. Abiotic process for Fe(II) oxidation and green rust mineralization driven by a heterotrophic nitrate reducing bacteria (Klebsiella mobilis), Environmental Science & Technology 48: 3742–3751.

Evans DJ, West JM, 2008. An appraisal of underground gas storage technologies and incidents, for the development of risk assessment methodology. Research rapport RR605, prepared by the British Geological Survey for the Health and Safety Executive.

Flynn R, Bellaby P, Ricci M, 2010. The 'value-action gap'in public attitudes towards sustainable energy: the case of hydrogen energy. The Sociological Review, 57(2), 159-180.

Fortin MJ, Fournis Y, 2014. Vers une définition ascendante de l'acceptabilité sociale: les dynamiques territoriales face aux projets énergétiques au Québec. Natures Sciences Sociétés, 22(3), 231-239.

Geostock, 2014. KGSP assessment of geological suitability, preliminary design and safety. Rapport Geostock IEL / F / J / 0001, <u>http://www.kgsp.co.uk/wp-content/uploads/2015/12/9.2-KGSP-Sub-surface-Safety-Assessment-Report.pdf</u> (consulté le 25/02/2021).

Gombert P, Lafortune S, Pokryszka Z, Lacroix E, de Donato P, Jozja N, 2021. Monitoring Scheme for the Detection of Hydrogen Leakage from a Deep Underground Storage. Part 2: Physico-Chemical Impacts of Hydrogen Injection into a Shallow Chalky Aquifer. Appl. Sci. 11 :2686. https://doi.org/10.3390/app11062686

Gottschalk G, 1986. Bacterial Metabolism, Series: Springer Series in Microbiology, 2nd ed. 1986, XIII.

Ham M, Jeger M, Frajman Ivković A, 2015. The role of subjective norms in forming the intention to purchase green food. Economic research-Ekonomska istraživanja, 28(1), 738-748.

Handin J, Russel JE, Carter NL, 1986. Experimental deformation of rocksalt. In Mineral and Rock Deformation: Laboratory Studies (Edited by B. E. Hobbs and H. C. Heard), pp. 117-160. Americal Geophysical Union Monograph, Washington, DC.

Hazotte A, 2012. Cinétique de l'utilisation microbienne du di-hydrogène sous hautes pressions. Rapport de Master 2, GPRE-SEE, Université de Lorraine, 42 p.

Hienuki S, Hirayama Y, Shibutani T, Sakamoto J, Nakayama J, Miyake A, 2019. How knowledge about or experience with hydrogen fueling stations improves their public acceptance. Sustainability, 11(22), 6339.

Hopper JM, 2004. Gas Storage and single-point failure risk», Energy Market, October.(<u>http://www.falcongasstorage.com/_filelib/FileCabinet/Articles/article_singlepointfailurerisk.pdf</u>).

Huijts N, Molin E, Chorus C, van Wee B, 2012. Public acceptance of hydrogen technologies in transport: a review of and reflection on empirical studies. In Shiftan Y, Geerlings H, Stead D (eds) Transition towards sustainable mobility (pp. 137–164). Routledge.

Huijts NM, 2018. The emotional dimensions of energy projects: Anger, fear, joy and pride about the first hydrogen fuel station in the Netherlands. Energy research & social science, 44, 138-145.

Huijts NM, van Wee B, 2015. The evaluation of hydrogen fuel stations by citizens: The interrelated effects of socio-demographic, spatial and psychological variables. International journal of hydrogen energy, 40(33), 10367-10381.

Huijts NM, Molin EJ, van Wee B, 2014. Hydrogen fuel station acceptance: A structural equation model based on the technology acceptance framework. Journal of Environmental Psychology, 38, 153-166.

Huijts N, Molin E, Chorus C, van Wee B, 2012. Public acceptance of hydrogen technologies in transport: a review of and reflection on empirical studies. In Shiftan, Y., Geerlings, H., & Stead, D. (Eds) Transition towards sustainable mobility (pp. 137–164). Routledge.

Ineris, 2016. Le stockage souterrain dans le contexte de la transition énergétique. Maîtrise des risques et impacts. Ineris références, septembre 2016, 40 p., <u>www.ineris.fr</u>

Itaoka K, Saito A, Sasaki K, 2017. Public perception on hydrogen infrastructure in Japan: influence of rollout of commercial fuel cell vehicles. International Journal of Hydrogen Energy, 42(11), 7290-7296.

Jorand FPA, 2018. The bacterial cell: the functional unit of biofilms, Chap. 4. In: Interactions Materials – Microorganisms. Concrete and Metals more Resistants to Biodeterioration (C. Lors, F. Feugeas and B. Tribollet, Eds), EDP Sciences, Les Ulis, FR, 63 – 93.

Kelley DS, Karson JA, Früh-Green GL, Yoerger DR, Shank TM, Butterfield DA, Hayes JM et al., 2005. A serpentinite-hosted ecosystem: the Lost City Hydrothermal Field. Science 307, 1428–1434.

Klinkenberg LJ, 1941. The permeability of porous media to liquids and gases. In: Drilling and Production Practice.

Konhauser K, 2007. Introduction to geomicrobiology. Blackwell Publishing, Malden, MA, USA, 425 p.

Kurlansky M, 2002. Salt: a world history. London: Penguin Books.

Lacroix E, 2021. Développement d'outils de monitoring pour la détection des fuites d'hydrogène (H₂) à l'aplomb des sites de stockage géologique. Thèse, Université de Lorraine, Nancy, Octobre 2021.

Lacroix E, de Donato P, Lafortune S, Caumon MC, Barres O, Liu X, Derrien M, Piedevache M (2021). In situ continuous monitoring of dissolved gases (N_2 , O_2 , CO_2 , H_2) prior to H_2 injection in an aquifer (Catenoy, France) by on-site Raman and infrared spectroscopies: instrumental assessment and geochemical baseline establishment. Anal. Methods, 2021, 13, 3806-3820, DOI: 10.1039/D1AY01063H

Lafortune S, Gombert P, Pokryszka Z, Lacroix E, Donato P, Jozja N, 2020. Monitoring Scheme for the Detection of Hydrogen Leakage from a Deep Underground Storage. Part 1: On-Site Validation of an Experimental Protocol via the Combined Injection of Helium and Tracers into an Aquifer. Appl. Sci. 10:6058. <u>https://doi.org/10.3390/app10176058</u>

Leblanc J, Massé B, 2013. Acceptabilité sociale : Pour qui ? Pour quoi ? Actes du forum Acceptabilité sociale, Mars 2013, Rimouski.

Liang W, Yang C, Zhao Y, Dusseault MB, Liu J, 2007. Experimental investigation of mechanical properties of bedded salt rock. Int. Journal of Rock Mechanics & Mining Sciences, 44, 400–411.

Lipp JS, Morono Y, Inagaki F, Hinrichs KU, 2008. Significant contribution of Archaea to extant biomass in marine subsurface sediments. Nature 454: 991-994.

Lothenbach et al., 2012. Hydration of low-alkali CEM III/B-SIO₂ cement (LAC). Cement and concrete research 42, 410-423.

Mauger S, 2011. Essais de performance comparatifs sur une gamme de détecteurs de flamme. Rapport d'étude INERIS-DRA-11-117743-08553A, 29 juillet 2011,

Molin E, Aouden F, van Wee B, 2007. Car drivers' stated choices for hydrogen cars: evidence from a small-scale experiment (No. 07-0547).

Nielsen J, 1993. Usability engineering. Boston: Academic Press.

NTSB, 1993. Highly volatile liquids Release from Underground Storage Cavern and Explosion, Mapco Natural Gas Liquids Inc., Brenham Texas, April 7, 1992, NTSB/PAR-93/01, PB93-916502.

Nunoura T, Oida H, Miyazaki M, Suzuki Y, 2008. Thermosulfidibacter takaii gen. nov., sp. nov., a thermophilic, hydrogen-oxidizing, sulfur-reducing chemolithoautotroph isolated from a deep-sea

hydrothermal field in the Southern Okinawa Trough. International Journal of Systematic and Evolutionary Microbiology, 58, 659–665.

O'Garra T, Pearson P, Mourato S, 2007. Public acceptability of hydrogen fuel cell transport and associated refuelling infrastructures. In Risk and the public acceptance of new technologies (pp. 126-153). London: Palgrave Macmillan.

Onstott TC, van Heerden E, Murdoch L, 2010. Microbial life in the depths of the Earth, In: Les frontières géologie – biologie, Geosciences, la revue du BRGM, nº11, 52 - 69.

Oren A, 2011. Thermodynamic limits to microbial life at high salt concentrations. Mini Review. Environmental Microbiology, 13: 1908–1923.

Owen JR, Kemp D, 2013. Social licence and mining: A critical perspective. Resources policy, 38(1), 29-35.

Peach CJ, 1991. Influence of deformation on the fluid transport properties of salt rocks. Geologica Ultraiectina, volume 77, 1-238.

Peach CJ, Spiers CJ, 1996. Influence of crystal plastic deformation on dilatancy and permeability development in synthetic salt rock. Tectonophysics, 1(4), 101-128.

Pedersen K, 1999. Subterranean microorganisms and radioactive waste disposal in Sweden. Engineering Geology, 52: 163–176.

Popp T, Kern H, Schulze O, 2001. Evolution of dilatancy and permeability in rock salt during hydrostatic compaction and triaxial deformation. J. of Geophysical Research: Solid Earth, 106(B3), pp. 4061-4078.

Pirkle RJ, 1986. Near Surface Geochemical Monitoring of Underground Gas Storage Facilities. Presented at: The American Gas Association Meeting, Chicago, IL, April

Pirkle et Jones (2006). Applications of Petroleum Exploration and Environmental Geochemistry to Carbon Sequestration, Fifth Annual Conference on Carbon Capture & Sequestration held in Alexandria, Virginia, january.

Poortinga W, Steg L, Vlek C, 2004. Values, environmental concern, and environmental behavior: A study into household energy use. Environment and behavior, 36(1), 70-93.

Portales L, Romero Castañeda S, 2016. Inconsistencies and limitations of the Social License to Operate: the case of Mexican mining. Humanistic Management Network, Research Paper Series, 1(16).

Potter J, Salfi S, Longstaffe FJ, 2013. Abiogenic hydrocarbon isotopic signatures in granitic rocks: Identifying pathways of formation. Lithos, 183-183, 114-124.

Prats C, 2013. Règles méthodologiques applicables aux études de danger des stockages souterrains. Rapport INERIS-DRA-13-133158-12929A.

Prinzhofer A, Cissé CST, Diallo AB, 2018. Discovery of a large accumulation of natural hydrogen in Bourakebougou (Mali). International Journal of Hydrogen Energy.

Proskurowski G, Lilley MD, Seewald JS, Früh-Green GL, Olson EJ, Lupton JE, Sylva SP, Kelley DS, 2008. Abiogenic hydrocarbon production at Lost City Hydrothermal Field. Science 319, 604-607.

Réveillère A, Bérest P, Evans DJ, Stöwer M, Chabannes C, Koopmans T, Bolt R, 2017. Past Salt Caverns Incidents Database Part 1: Leakage, Overfilling and Blow-out. SMRI Research Report RR2017-2

Réveillère A, 2021. The development of the concern for tightness of Salt Caverns, of Mechanical Integrity Tests techniques and of their accuracies. In preparation.

Ricci M, Bellaby P, Flynn R, 2008. What do we know about public perceptions and acceptance of hydrogen? A critical review and new case study evidence. International Journal of Hydrogen Energy, 33(21), 5868-5880.

Rothschild LJ, Mancinelli RL, 2001. Life in extreme environments, Nature, 409: 1092

Salvi S, Williams-Jones AE, 1992. Reduced orthomagmatic C–O–H–N–NaCl fluids in the Strange Lake rare-metal granitic complex, Quebec/Labrador, Canada. European Journal of Mineralogy 4, 1155–1174.

Schmidt A, Donsbach W, 2016. Acceptance factors of hydrogen and their use by relevant stakeholders and the media. International Journal of Hydrogen Energy, 41(8), 4509-4520.

Schulze O, Popp T, Kern H, 2001. Development of damage and permeability in deforming rock salt. Engineering Geology, Volume 61, pp. 163-180.

Seni SJ, Johnson DO, 2005. Regulatory Response to Recent Events Effecting Three Gas Storage Facilities in Texas. AGA Annual Meeting 2005.

Senseny PE, Hansen FD, Russell JE, Carter NL, Handin JW, 1992. Mechanical behaviour of rock salt: phenomenology and micromechanisms. Int. J. Rock Mech. Min. Sci. & Geomech. Abstr. 29/ 4, 363-378.

Sherry-Brennan F, Devine-Wright H, Devine-Wright P, 2010. Public understanding of hydrogen energy: a theoretical approach. Energy Policy, 38(10), 5311-5319.

Smigan P, Greksak M, Kozankova J, Buzek F, Onderka V, Wolf I, 1990. Methanogenic bacteria as a key factor involved in changes of town gas stored in an underground reservoir. FEMS Microbiology Ecology, 73: 221-224.

Sutherland HJ, Cave SP, 1980. Argon Gas permeability of New Mexico rock salt under hydrostatic compression. Int. J. Rock Mech. Min. Sci. & Geomech. Abstr., 17(5), 281-288.

Sütterlin B, Siegrist M, 2017. Public acceptance of renewable energy technologies from an abstract versus concrete perspective and the positive imagery of solar power. Energy Policy, 106, 356-366.

Terrade F, Pasquier H, Reerinck-Boulanger J, Guingouain G, Somat A, 2009. L'acceptabilité sociale: la prise en compte des déterminants sociaux dans l'analyse de l'acceptabilité des systèmes technologiques. Le travail humain, 72(4), 383-395.

Thøgersen J, 2006. Norms for environmentally responsible behaviour: An extended taxonomy. Journal of environmental Psychology, 26(4), 247-261.

Thomas G, Demski C, Pidgeon N, 2019. Deliberating the social acceptability of energy storage in the UK. Energy Policy, 133, 110908.

Thorel L, 1994. Plasticité et endommagement des roches ductiles. Application au sel gemme, s.l.: s.n.

Truche L, Bertrand M, 2019. Réactivité de l'hydrogène dans le système H₂O/ciment/H₂ Livrable n°3 - Rapport final du contrat de collaboration de recherche, Université Grenoble Alpes-ANDRA, Référence ANDRA N° 20075412.

Upham P, Roberts T, 2011. Public perceptions of CCS in context: Results of NearCO2 focus groups in the UK, Belgium, the Netherlands, Germany, Spain and Poland. Energy Procedia, 4, 6338-6344.

Van Alphen K, tot Voorst QVV, Hekkert MP, Smits RE, 2007. Societal acceptance of carbon capture and storage technologies. Energy Policy, 35(8), 4368-4380.

Vankatesh V, Morris M, Davis G, Davis FD, 2003. User acceptance of information technology: toward a unified view. Mis Quarterly, 27(3), 425-478.

Wolsink M, 2018. Social acceptance revisited: gaps, questionable trends, and an auspicious perspective. Energy research & social science, 46, 287-295.

Wolsink M, 2019. Social acceptance, lost objects, and obsession with the 'public'—The pressing need for enhanced conceptual and methodological rigor. Energy Research & Social Science, 48, 269-276.

Zhu C, Arson C, 2015. A Model of Damage and Healing Coupling Halite Thermo-mechanical Behavior to Microstructure Evolution. Geotech Geol Eng , 33(2), 389–410.

11 Glossary

NTPC: Normal Temperature and Pressure Conditions (i.e. 1013 hPa and 0°C).

Well completion: the assembly of equipment required to complete the well to enable optimal operation.

Focus group: a technique for collecting qualitative data whereby individuals are brought together in order to study their attitudes regarding a product, subject, or concept.

MIT: "Mechanical Integrity Test", terminology used in the salt cavern industry to refer to a leak test. This comes from American regulations which require wells injecting into the subsoil (in particular those of salt caverns for the production of brine or for storage) to test their "mechanical integrity".

*Nm*³: Gas volume (in m³) under Normal Conditions of Pressure and Temperature (see this term)

Personal norm: personal norms make the individual feel moral obligation to behave in accordance with his/her personal beliefs (Thøgersen, 2006)

Subjective norm: the belief that a specific behaviour will be approved and supported by a person or a group of persons (Ham et al., 2015)

Object: that to which action, thought, or feeling is directed (OED2, 1989)

Tool: anything used as a means of performing an operation or achieving an end (Collins, 2022)

Usability: extent to which a system, product or service can be used by specified users to achieve specified goals with effectiveness, efficiency and satisfaction in a specified context of use (ISO 9241-210, 2010)

Utility: the quality of practical use, usefulness, service ability (Collins, 2022)



French National Institute for Industrial Environment and Risks (Ineris) Parc technologique Alata • BP 2 • F-60550 Verneuil-en-Halatte +33 3 44 55 66 77 • ineris@ineris.fr • www.ineris.fr/en