

Hydrogen geologic storage in China: feasibility and challenges[#]

Zhengyang Du^{1,2}, Zhenxue Dai^{1,2,3,*}, Zhijie Yang^{1,2,3}, Wei Chen^{1,2}, Mingxu Cao^{1,2}, Xiaoying Zhang^{1,2}, Hung Vo Thanh^{4,5}, Mohamad Reza Soltanian⁶, Reza Ershadnia^{6,7}

1 College of Construction Engineering, Jilin University, Changchun, China

2 Institute of Intelligent Simulation and Early Warning for Subsurface Environment, Jilin University, Changchun, China

3 Key Laboratory of Groundwater Resources and Environment, Ministry of Education, Jilin University, Changchun, China

4 Laboratory for Computational Mechanics, Institute for Computational Science and Artificial Intelligence, Van Lang University, Ho Chi Minh City, Vietn

5 Faculty of Mechanical - Electrical and Computer Engineering, School of Technology, Van Lang University, Ho Chi Minh City, Vietnam

6 Departments of Geology and Environmental Engineering, University of Cincinnati, Cincinnati, OH, USA

7 Bureau of Economic Geology, University of Texas at Austin, TX, USA

* Corresponding author: Zhenxue Dai (dzx@jlu.edu.cn)

ABSTRACT

As a clean, efficient energy source, hydrogen is regarded as a promising alternative energy for accomplishing the zero-CO₂ targets. In the longer term, large-scale hydrogen geologic storage (HGS) could reduce the instability of intermittent energy sources, through peak cutting and valley filling. However, the low density and viscosity of hydrogen and its interaction with the surrounding rocks and microbes constrain the effective advancement of large-scale HGS. This paper summarizes the current research status, feasibility analysis, advantages and disadvantages of HGS in the main potential reservoirs (depleted oil/gas fields, salt caverns, and brine aquifers). In addition, the uncertainties and challenges are also addressed for HGS application in the future: 1) Operating parameters, which are difficult to determine and evaluate, have a significant impact on HGS efficiency. For example, the cyclical injection-reproduction and injection rates have large impact on H₂ fingering phenomenon and the geological integrity of the caprocks; 2) Currently, the hydrogen-water-rock geochemical reactions at various temperatures and pressures are not well understood well. There is a lack of a geochemical reaction database to meet the HGS numerical simulation requirements. The associated reactions could cause uncertain changes in porosity and

permeability, which may cause large-scale hydrogen leakage in severe cases; 3) Metabolic mechanisms of subsurface environmental microorganisms have not been thoroughly explored at high temperature and pressure, which poses a related risk of H₂ leakage and contamination for shallow groundwater. Some microorganisms have the ability to consume hydrogen to produce gas mixing (e.g., CH₄), harmful gas pollution (e.g., H₂S), and steel corrosion. This review will provide substantial information for further analyzing the scientific challenges of HGS and promoting the development of HGS simulations and practical engineering applications.

Keywords: hydrogen geologic storage; hydrogen-water-rock geochemical reactions; feasibility analysis; metabolic mechanisms

NONMENCLATURE

<i>Abbreviations</i>	
HGS	Hydrogen Geological Storage
MB	Methanogenic Bacteria
SRB	Sulfate-Reducing Bacteria
<i>Symbols</i>	
a	Year

[#] This is a paper for the 8th Applied Energy Symposium - CUE2022, Sept. 24-27, 2022, Matsue, Japan.

1. Introduction

With the rapid development of the global human population and economy, the world is overly dependent on fossil energy. Nevertheless, the key challenges are the rapid decline of fossil energy and environmental pollution [1-4]. The Paris Agreement set a goal of keeping global average temperature rise below 2 °C [5]. In order to accomplish the zero-CO₂ targets and mitigate climate change, the internationally recognized approach is to reduce the the proportion of fossil energy and accelerate the large-scale development of renewable energy [6, 7]. In particular, renewable energy is the most economical and effective way with little carbon emissions to move away from fossil fuels and improve environmental pollution [8, 9]. As is known, renewable energy sources (such as wind and solar power), which are characterized by instability and intermittency [10, 11], exhibit intermittency plagued by uncertainty on a seasonal and daily scale, leading to energy deficits or surpluses. In deficit periods, this problem can be solved by using other resources to secure supply; nevertheless, during periods of generating excess energy, it will cause unnecessary energy waste without suitable energy storage methods [10, 12-14]. Therefore, it is crucial to find appropriate methods to improve the renewable energy system through peak cutting and valley filling.

Hydrogen has been very popular in recent years. As a clean and efficient energy source, it could be applied as an intermediate transition carrier for efficient energy conversion, and the hydrogen production process and raw materials are uncomplicated and acceptable [15-17]. Hydrogen energy is one of the major energy sources of the future, which will bring opportunities as well as many difficulties, such as hydrogen storage and transportation [18-20]. In order to use hydrogen energy in a sensible way, it is important to store hydrogen in a safe and effective way. There are three methods for storing hydrogen: compressed gas, cryogenic liquid hydrogen, and solid storage, respectively [18]. Solid-state storage is the most efficient compared to other storage methods and could store the maximum amount of hydrogen in a limited volume [21]. It stores hydrogen in the form of nanotechnology or hydride [22]. However, solid-state storage is difficult to apply on a large scale due to its high

economic consumption and technical implementation difficulties [23, 24]. At present, the storage methods of compressed hydrogen and liquid hydrogen are widely used. Due to the low density and high chemical activity of hydrogen, it is difficult to realize the application of large-scale hydrogen storage system on the ground [14, 17].

In recent decades, large-scale hydrogen geologic storage (HGS) in the underground has been considered a feasible method to reduce the instability of intermittent energy sources in the longer term [20, 25-27]. It allows for large-scale hydrogen storage and multiple cyclical injection-reproduction cycles to meet seasonal energy fluctuations[25]. Many scholars propose to store hydrogen in depleted oil/gas fields, salt caverns and brine aquifers [28-31]. [Heinemann, et al. \(2021\)](#) summarized the scientific challenges and possible risks of hydrogen storage in porous media. [Ozarslan \(2012\)](#) analyzed the design for hydrogen storage in a salt cavern and conducted a pre-evaluation for the salt cavern. And HGS could also learn from previous geological storage of carbon dioxide and natural gas.

To reduce carbon dioxide emissions, China invested heavily in wind power generation systems and had a large wind power generation capacity [33-35]. However, because the huge amount of wind energy could not be collected and applied completely, a mean energy value of 17% is lost and wasted due to energy fluctuation and improper storage[36, 37]. At the same time, countries around the world have established or plan to establish a large number of hydrogen stations, including 250 in China (Table 1). Therefore, the building of large-scale HGS sites in China is an unavoidable part of the country's growth.

Table 1 Active hydrogen stations.

Country	Active stations
China	250
Japan	161
South Korea	141
Germany	93
France	21
Switzerland	13
Netherlands	9

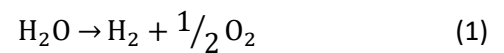
This study provides a concise summary of the

accomplishments made in research pertaining to large-scale HGS and identifies important scientific problems for the foreseeable future. The detailed description will give crucial implications for helping scientists to better conduct a numerical simulation for characterizing the HGS process and promoting real engineering applications. These will be useful in the long run. In the beginning, we went over an analysis of the feasibility, as well as the benefits and drawbacks of using HGS in depleted oil and gas fields, salt caverns, and brine aquifers. After that, we went into great detail to demonstrate the development of hydrogen geochemical processes and the mechanism of microbial metabolism as well as the consequences for HGS. This included the reaction process, experimental investigation, and numerical simulation. At the very conclusion of the presentation, the most significant scientific difficulties and potential outcomes of the large-scale HGS development were discussed.

2. Feasibility analysis of HGS

2.1 Overview of hydrogen properties and storage technologies

Scientists are interested in hydrogen's high energy density, clean combustion products, and production process (120 MJ kg^{-1}), which is approximately three times that of traditional energy (Eq 1)[22]. The density of hydrogen is 0.0899 g/L ($0 \text{ }^\circ\text{C}$ and 0.1 MPa), which is much smaller than other gases. This results in the need for a larger volumetric capacity and higher pressure of hydrogen to store the same mass of gas [38]. The critical pressure and temperature of hydrogen are 1.297 MPa and $-239.97 \text{ }^\circ\text{C}$. Because of the critical temperatures in subsurface conditions is relatively harsh, hydrogen is almost stored in a gaseous state. The low density of hydrogen facilitates the formation of hydrogen caps under the caprock. And the density of hydrogen increases with depth, while the storage efficiency also increases[39, 40]. The low hydrogen viscosity is $0.89 \cdot 10^{-5} \text{ Pa s}$ ($25 \text{ }^\circ\text{C}$ and 0.1 MPa), leading to less residual hydrogen and high recovery. Another advantage of hydrogen is its extremely low solubility, which reduces hydrogen losses in the system [41].



More and more scientists focus on HGS, and have conducted a lot of research, discussion and analysis over the past decades [15]. Many countries and regions have carried out the HGS potential analysis and future development, such as UK [14, 42], Poland [15], Romania [31], China [43] and Europe [44]. And some projects were conducted for laboratory experiments and numerical simulations [45]. At present, only salt caverns have been successfully used for hydrogen storage commercially in the US and UK, respectively [22, 46]. Because of this, there needs to be a lot of research done to improve the theoretical approach for HGS.

2.2 Salt caverns

The salt cave is a cavity volume formed by injecting water into the formation to dissolve NaCl in salt rock [47]. The spatial structure of the salt cavities is controlled by different water injection processes, which are usually cylindrical (Figure1). They can be built at a depth of more than 2000 meters, with about $1,000,000 \text{ m}^3$ volume, enabling certain space for large-scale HGS [26, 48]. The best location for a salt cavern is a salt dome or bedded salt deposit, which facilitates the stability and integrity of the salt cavern [49]. The rock around the salt cavern is very solid and sealed, so hydrogen can be stored safely and there is less chance of it leaking [50].

The internal pressure of the salt cavern will be unbalanced due to the extraction of salt water. It is necessary to inject an appropriate amount of gas to maintain the minimum pressure required internally to ensure the stability of the salt cavern, and the gas is defined as cushion gas [47]. This means the cushion gas is not recoverable, generating economic losses and taking up approximately 22-33% of the storage capacity [14, 51]. Salt caverns could more easily realize cyclical injection-reproduction on a long-time scale without consideration of multiphase phenomena. And the rich brine produced can also bring certain economic benefits after proper treatment. Fortunately, microbial growth and metabolism are inhibited to prevent harmful gases under high salt conditions [52]. However, the storage capacity of salt caverns is small compared to other

geological storage methods, and high-pressure hydrogen is required to balance the cave pressure with the increase in depth. When salt water is around, it speeds up the rusting of steel engineering tools [53, 54].

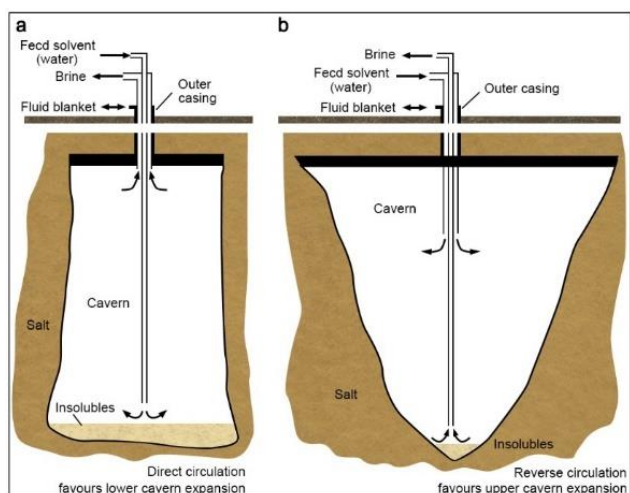


Fig. 1 The manufacturing process of salt caverns [14]

2.3 Brine aquifers

Saturated aquifers are usually composed of porous permeable media and brine and are common in sedimentary rocks throughout the world. As a choice for hydrogen storage, the reservoir formation must meet the requirements of high permeability and porosity, and the isolation boundary must be complete and impermeable. Hydrogen is injected into the formation to replace brine

in the porous media and diffuse beneath the low-permeability cap (Figure 2). The enclosed structure provides a huge space for hydrogen to reproduce and ensures that hydrogen will not escape and leak [14, 25, 42, 51, 55]. Aquifers require approximately 80% of the total reservoir capacity for cushion gas [50, 54].

There have been many successful gas storage sites providing experience for HGS, and the reservoir volume of aquifers is several times larger than salt caverns [47]. So far, except for mixed hydrogen storage, there is no successful case of pure hydrogen storage in aquifers [56, 57]. There is absolutely no risk of explosion due to lack of oxygen. Chemical reactions and microbial metabolism will change the porosity and permeability of the aquifer, thus affecting the efficiency and safety of HGS, which are discussed in detail in Chapter 3 [25]. The fingering phenomenon generated by the lateral diffusion of hydrogen during the injection process also makes hydrogen recovery difficult and leads to hydrogen loss. During the hydrogen recovery process, the movement of the water-gas interface will allow the liquid of the same period to flow back and form a confined space, resulting in a portion of the hydrogen being permanently unrecoverable [58]. Moreover, large-scale drilling boreholes are required for geological investigations to obtain comprehensive geological data [50].

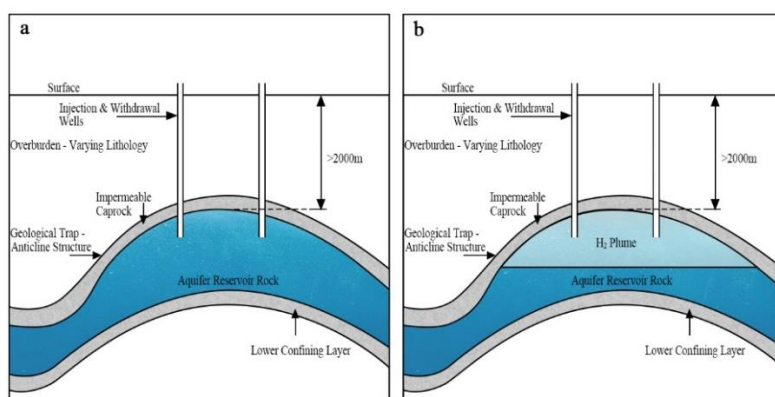


Fig. 2 Hydrogen injection process in aquifers [14]

2.4 Depleted oil/gas fields

Depleted oil/gas fields use geological trap structures to seal gas storage with low permeability cap and edges, which are similar to aquifers. The sealing and

stability of the depleted oil/gas fields are excellent and have been tested over a long period of time. This structure is very common in oil-rich countries. The pore space is filled with a lot of water and residual oil/gas in the depleted oil/gas fields (Figure 3). The presence of

residual gas could replace part of the cushion gas and reduce approximately 50-60% of economic expenses compared with the brine aquifer. Thus, previous extractions should be allowed to finish at the optimal time in order to retain the right amount of cushion gas [59]. And the infrastructure of the depleted oil/gas fields' extraction oil and gas systems could be reused. Detailed geological information can also be easily obtained [60]. Over time, the maximum reservoir pressure will be greater than the initial formation pressure, which means more gas can be stored [26].

The mixing of hydrogen with oil or gas, as well as the presence of an oil-gas-water interface, can complicate the interior of the reservoir, resulting in uncertainty reactions that result in hydrogen loss. There may be contamination in the early work, and site contamination remediation and equipment upgrades are required. Because of the flexible cyclical loop, depleted oil/gas fields and aquifers are good choices for seasonal hydrogen storage [47, 51].

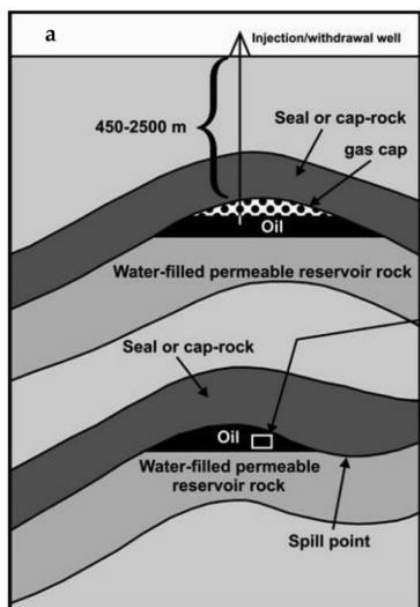


Fig. 3 Hydrogen storage process in depleted oil/gas fields [14]

3. Geochemical reactions and microbial metabolism mechanism

Hydrogen undergoes a series of biotic and abiotic reactions in the subsurface environment (Figure4). Some reactions facilitate hydrogen cycle extraction and storage, while others pose a leakage risk. The quality, safety, and

efficiency of the HGS system cannot be guaranteed without fully understanding the reactions involved. In this section we provided a comprehensive review of hydrogen-related geo-chemical reactions and microbial metabolism from reaction processes, experiments and simulations.

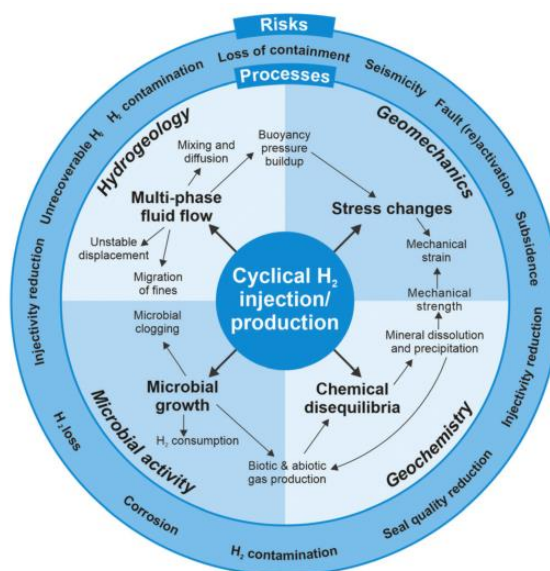


Fig. 4 Reaction processes of HGS

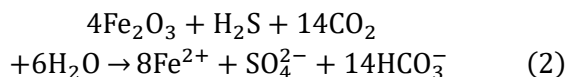
3.1 Hydrogen-water-rock geochemical reactions

3.1.1 Geo-chemical reaction process

As hydrogen is injected into the underground reservoir, the chemical balance between solid, liquid, and gas phases in the system is broken. Causing the following negative effects: (1) A lot of hydrogen loss; (2) Generating other gases to contaminate the hydrogen system; (3) Changes in porosity and permeability due to mineral dissolution/precipitation, affecting recovery efficiency; (4) Influencing the mechanical properties of the reservoir [61].

Hydrogen may react with the initial air-water component to indirectly affect pH and promote mineral dissolution/precipitation. The associated undesirable gas (H_2S) will have a negative effect on hydrogen quality [62]. These harmful gases promote the redox reaction and dissolution of hematite (Eq2) [63]. The reaction product of hydrogen gas and pyrite may also be iron sulfide. And sulfate cements and carbonate cements dissolve under certain temperatures and pressures (10-20 MPa, <40 °C). However, feldspar and quartz hardly react with hydrogen. Mineral dissolution increases the reservoir porosity and

decreases the caprock integrity, which may lead to the risk of leakage channels. In addition, hydrogen can also corrode metal equipment. Although some scholars believed that the hydrogen geo-chemical reaction did not affect the reservoir integrity, it is necessary to understand the mechanism to determine the relevant reaction parameters for safe storage of HGS [25, 64].



3.1.2 Experimental research

The correct experiment is helpful to explore the chemical reaction mechanism of hydrogen, and the identified parameters also provide necessary information for subsequent numerical simulation. At present, the hydrogen experiment focuses more on the safe disposal of nuclear waste than HGS.

Variations in stored gas were observed in both gas storage reservoirs in Ketzin and Beynes as a rough indication of the existence of some geochemical reactions. Especially in Ketzin, microbial metabolism alone could not reasonably explain the large gas loss in the last decades [53]. There is a suggestion that the pyrite reduction is due to the production of hydrogen sulfide in Beynes [65].

[Bourgeois, et al. \(1979\)](#) proposed that redox of pyrite could explain the increased hydrogen sulfide concentration. At specific pressures and temperatures, some researchers proposed the same argument [67]. Under different storage conditions, the comparison of sandstone experiments exposed to hydrogen before and after has also proved the possibility of HGS [29]. [Hassanpouryouzband, et al. \(2022\)](#) performed approximately 250 different types of contrast reaction experiments in sandstone in the presence and absence of hydrogen. The results demonstrated the geo-chemical reaction does not diminish reservoir integrity, and hydrogen storage in sandstone reservoirs is safe from hydrogen loss. However, it is important to note that due to the scale effect, the reaction parameters measured in the laboratory may be significantly different from those in the actual large-scale field.

3.1.3 Numerical simulation

Before HGS applications are actually put into place,

numerical simulations are a good way to predict what will happen at sites so that implementation options can be evaluated and improved..

[Hemme and Van Berk \(2018\)](#) used PHREEQC to develop a one-dimensional reactive model with consideration of the equilibrium of gas-water-rock interactions and evaluated key parameters affecting hydrogen loss. Multi-step geo-chemical modeling was applied to analyze the interaction of hydrogen with minerals over a long time scale. Uncertainty in the kinetic rate parameters of minerals reduced the accuracy of simulation results [69]. [Bo, et al. \(2021\)](#) relied on software for geochemical modeling, the kinetic simulations showed that the presence of calcite causes hydrogen loss in the reservoir. [Ershadnia, et al. \(2022\)](#) investigated the influence of geological parameters and operational process parameters on hydrogen storage in a three-dimensional heterogeneous aquifer.

Some software (e.g., ECLIPSE, TOUGH, COMSOL, OpenGeosys, and PUNQ-S3) was applied to perform HGS simulations. They mainly simulated the effects of temperature, pressure, cushion gas type, injection rate, and geological property parameters on hydrogen simulation. However, they barely consider geo-chemical reactions process. It is essential to establish a chemical reaction database to provide data support for simulation software.

3.2 Microbial metabolism mechanism

3.2.1 Hydrogen-biological reaction process

Microbial metabolism is known to be critical in underground gas storage, and it is also considered to be significant for the HGS. As mentioned in Section 2, salt caverns, deep aquifers, and depleted oil/gas reservoirs are available for HGS, which is also the basis of microbial community phylogeny and metabolic diversity [30, 72]. Microorganisms can obtain energy by oxidizing electron donors and simultaneously reducing electron receptors. Hydrogen has a low reduction potential and can offer high energy, thus it is considered one of the most important electron donors for subsurface microbial cycling [73]. Some classes of microorganisms, which frequently survive in subsurface formations, including methanogens, sulfate reducers, acetogenic bacteria, and

iron (III) reducers, are thought to be major consumers of hydrogen [26]. Subsurface microbes can consume H₂ during their metabolism, leading to the production of a variety of metabolites, such as gases, acids, solvents,

biopolymers, and surfactant biosurfactants. This will cause various adverse side effects such as H₂ loss, H₂S formation, methane formation, acid formation, clogging, and corrosion (Listed in **Table 2**).

Table 2 Overview of the major Hydrogen-consuming processes

Hydrogen-consuming process	Reaction	Specific environmental conditions			
			Temperature (°C)	pH	Salinity (g/L)
Methanogenesis	$\frac{1}{4}\text{HCO}_3^- + \text{H}_2 + \frac{1}{4}\text{H}^+ \rightarrow \frac{1}{4}\text{CH}_4 + \frac{3}{4}\text{H}_2\text{O}$	Optimal	30-40	6.0-7.5	<60
		Critical	122	4.5-9	200
Acetogenesis	$\frac{1}{2}\text{HCO}_3^- + \text{H}_2 + \frac{1}{4}\text{H}^+ \rightarrow \frac{1}{4}\text{CH}_3\text{COO}^- + 2\text{H}_2\text{O}$	Optimal	20-30	6.0-7.5	<40
		Critical	72	3.6-10.7	240
Sulfate reduction	$\frac{1}{4}\text{SO}_4^{2-} + \text{H}_2 + \frac{1}{4}\text{H}^+ \rightarrow \frac{1}{4}\text{HS}^- + \text{H}_2\text{O}$	Optimal	20-30	6.0-7.5	<100
		Critical	113	0.8-11.5	240
Iron (III) reduction	$2\text{FeOOH} + \text{H}_2 + 4\text{H}^+ \rightarrow 2\text{Fe}^{2+} + 4\text{H}_2\text{O}$	Optimal	0-30	6.0-7.5	<40
		Critical	90	1.6-9	200

3.2.2 Experimental research

The topic of HGS increasingly attracts worldwide interest, many research projects are launched for large-scale hydrogen storage, which can provide experience for the follow-up research on HGS. This section mainly reviews some experience from HGS field tests, as well as related experimental research.

(1) Microbial effects in deep aquifers

As mentioned before, a typical investigation is the stored town gas (approximately 45-60% hydrogen content) in Lobodice, Czech Republic [74]. Here, hydrogen is converted to methane or hydrogen sulfide by microorganisms at relatively low temperatures (35 °C). It is observed that the reservoir pressure decreased, the number of Methanogenic bacteria (MB) increased, the emission of CO₂ decreased and the emission of methane increased. There are conflicting reports of town gas storage in the saline aquifer near Beynes, France. On one hand, there are no operational problems and hydrogen loss [75]; on the other hand, other researchers have observed intense microbial activity as a change in gas composition [76]. Panfilov (2010) has presented the population dynamics model of bacteria, which feed on hydrogen and carbon dioxide and produce methane. It demonstrates a possible mechanism for the separation of hydrogen-rich and methane-rich zones observed in

aquifer town gas reservoirs in Lobodice and Beynes.

The Underground Sun. storage project in Lehen, Austria, where hydrogen (12%) is stored with natural gas, saw a drop in hydrogen (18% not recovered) and an increase in methane over a four-month test period [78]. The decrease in carbon dioxide (0.2% to 0.05%) is analyzed as being due to methanogenesis, indicating that very low levels of carbon dioxide were sufficient to activate MB. The reduced sulfate and appearance of acetate indicate methanogenesis and acetogenesis, but there is no H₂S was reported (probably precipitated after reacting with dissolved ferric ions). DNA community analysis for bacteria shows changes from the original bacteria to archaea, the proportion of methanogenic archaea reached nearly 80%, which verified the occurrence of microbial effects. Similar results were obtained from IFA-Tulln laboratory studies using the same field water. Significant sulfate reduction, hydrogen sulfide formation, and subsequent Iron(II) sulfide precipitation are observed when additional barite forms of sulfate are provided.

The HyChico project in Argentina, where hydrogen from a nearby wind farm is stored in a depleted gas reservoir with 10% hydrogen, observed hydrogen loss due to microbial activity. The total gas volume loss, corrosion, and gas composition changes were observed

at the former town gas storage in Ketzin, Germany, varying with the loss of carbon monoxide and the increase of hydrogen, methane, and CO₂. But it is not clear whether or what microbial processes have been active [77].

(2) Microbial effects in salt caverns

Artificial salt caverns are expected to be a practical option for HGS. Because of specific geological structures, the salt cavern has a low risk of microbial activity during hydrogen storage. The surface area of salt caverns is much smaller than that of porous media, which reduces the formation of biofilms and possible clogging. The high-level salinity of brine in salt cavern lead to a large osmotic pressure difference in cells, which reduces the diversity and abundance of microorganisms, especially when the salinity is above 100 g/L. Considering there are still salt-tolerant or halophilic microorganisms active (Halobacteria, for example, can survive in salt at concentrations of 100-150 g/L), microbial effects in salt caverns are still a scientific concern.

The activity of the Sulfate-reducing bacteria can also be observed in salt caverns filled with hydrogen gas. There, the Sulfate-Reducing Bacteria (SRB) live in sumps and salt water, producing biofilms on the walls of the cave [79]. The theoretical evaluation shows that when sulfates are present, there is a high risk of microbial H₂S formation in salt caverns at the brine-gas interface [80].

3.2.3 Numerical simulation

Depleted gas reservoirs or deep aquifers are usually porous and high-permeable, and are considered to have great HGS potential. The technical feasibility of this storage method is still being explored in China [81]. Numerical simulation is a reliable and cost-effective way to understand microbial interactions in reservoirs and how it affects underground hydrogen storage at different scales. The simulation results can be used to measure the effective parameters of H₂ transformation and the kinetic parameters for certain microorganisms[53, 61].

It has been shown that biofilm and mineral deposition can significantly increase the fluid flow resistance in porous media [82]. On the other hand, with the increase of microbial density, microbial biofilm or mineral precipitation may lead to pore blockage, thus reducing the hydrogen injection capacity [83-85].

However, the bio-clogging may also be beneficial. Many models have simulated unidirectional or two-phase flow through porous media containing microbial membranes or suspended bacteria, and applied the multiphase transport model to reduce leakage from geologic carbon dioxide reservoirs [86]. Eddaoui, et al. (2021) established a numerical model of pore plugging, studied the biological plugging process of underground hydrogen storage and its influence on gas migration in the reservoir, and believed that microorganisms accumulated in places with high hydrogen saturation, forcing hydrogen migration, resulting in the uniform distribution of hydrogen stored in aquifers in all directions.

In order to explore the mechanism controlling the conversion of hydrogen to methane, Ebigbo, et al. (2013) proposed a coupled numerical model combining the microbial biofilm scale with the gas flow process at the pore scale, which solved the stokes flow equation respectively for the migration of three gas components H₂-CH₄-CO₂ and concluded that the conversion rate of H₂ and CO₂ to CH₄ was determined by the amount and activity of the existing biomass. Panfilov (2010) established a dynamic coupling numerical model of reaction migration of underground hydrogen storage and methanogenic bacteria communities at the macro scale, which is practical and describes the analytical values of migration parameters at the reservoir scale. The above simulations are mainly based on one-dimensional or two-dimensional problems. Ershadnia, et al. (2022) conducted three-dimensional numerical simulation of aquifer and quantified the sensitivity of a series of different forms of hydrogen injection to various parameters of hydrogen underground.

3.3 Key scientific challenges

In spite of the fact that HGS is realizable, a number of significant scientific obstacles still need to be overcome. There was only a small experiment conducted for HGS under the conditions of the reservoir's temperature and pressure, and the mechanisms of chemical reactions and microbial metabolism have not been thoroughly investigated. It was extremely difficult to differentiate between biotic and abiotic processes in an exact manner. Due to the lack of detailed kinetic and

equilibrium parameters, we discovered that the reaction process was scarcely taken into consideration while the simulations were being run. It is absolutely necessary to create an exhaustive and specific reaction database in order to run simulations. One example of such a database is the CO₂ geological storage database. According to the current state of study, the production of dangerous gases like H₂S is unavoidable, and the question of how to ensure the purity of hydrogen is a serious one. Upscaling research may provide solutions for problems like this one, in which the parameters measured on a smaller scale, such as in a laboratory, yield very different results when applied to a larger, more realistic setting.

4. Summary

This article provides a concise summary of the current status of HGS as well as a study of the practicality, as well as the benefits and drawbacks of three distinct geological situations (depleted oil/gas fields, salt caverns, and brine aquifers) as potential locations for hydrogen reservoirs. In order to expedite the development of HGS, a number of pressing scientific problems have been proposed. These tasks include the investigation of hydrogen's geo-chemical reaction and the process underlying microbial metabolism. It is intended that the further theoretical research and practical implementation of HGS will be able to use this paper as a reference.

REFERENCE

[1] L. Chiari, A. Zecca, Constraints of fossil fuels depletion on global warming projections, *Energy Policy*, 39 (2011) 5026-5034.

[2] J. Ren, N.M. Musyoka, H.W. Langmi, M. Mathe, S. Liao, Current research trends and perspectives on materials-based hydrogen storage solutions: a critical review, *International journal of hydrogen energy*, 42 (2017) 289-311.

[3] N. Endo, K. Goshome, M. Tetsuhiko, Y. Segawa, E. Shimoda, T. Nozu, Thermal management and power saving operations for improved energy efficiency within a renewable hydrogen energy system utilizing metal hydride hydrogen storage, *International Journal of Hydrogen Energy*, 46 (2021) 262-271.

[4] R. Singh, M. Singh, S. Gautam, Hydrogen economy, energy, and liquid organic carriers for its mobility, *Materials Today: Proceedings*, 46 (2021) 5420-5427.

[5] C.-F. Schleussner, J. Rogelj, M. Schaeffer, T. Lissner, R. Licker, E.M. Fischer, R. Knutti, A. Levermann, K. Frieler, W. Hare, Science and policy characteristics of the Paris Agreement temperature goal, *Nature Climate Change*, 6 (2016) 827-835.

[6] Y. Sun, C. Shen, Q. Lai, W. Liu, D.-W. Wang, K.-F. Aguey-Zinsou, Tailoring magnesium based materials for hydrogen storage through synthesis: Current state of the art, *Energy Storage Materials*, 10 (2018) 168-198.

[7] A. Qazi, F. Hussain, N.A. Rahim, G. Hardaker, D. Alghazzawi, K. Shaban, K. Haruna, Towards sustainable energy: a systematic review of renewable energy sources, technologies, and public opinions, *IEEE access*, 7 (2019) 63837-63851.

[8] A. Olabi, M.A. Abdelkareem, Renewable energy and climate change, *Renewable and Sustainable Energy Reviews*, 158 (2022) 112111.

[9] D. Gielen, F. Boshell, D. Saygin, M.D. Bazilian, N. Wagner, R. Gorini, The role of renewable energy in the global energy transformation, *Energy Strategy Reviews*, 24 (2019) 38-50.

[10] Z.-X. He, S.-C. Xu, W.-X. Shen, H. Zhang, R.-Y. Long, H. Yang, H. Chen, Review of factors affecting China's offshore wind power industry, *Renewable and Sustainable Energy Reviews*, 56 (2016) 1372-1386.

[11] M. Zhang, X. Ai, J. Fang, W. Yao, W. Zuo, Z. Chen, J. Wen, A systematic approach for the joint dispatch of energy and reserve incorporating demand response, *Applied Energy*, 230 (2018) 1279-1291.

[12] D.J. Burke, M.J. O'Malley, Factors influencing wind energy curtailment, *IEEE Transactions on Sustainable Energy*, 2 (2011) 185-193.

[13] L. Bird, D. Lew, M. Milligan, E.M. Carlini, A. Estante, D. Flynn, E. Gomez-Lazaro, H. Holttinen, N. Menemenlis, A. Orths, Wind and solar energy curtailment: A review of international experience, *Renewable and Sustainable Energy Reviews*, 65 (2016) 577-586.

[14] R.L. Wallace, Z. Cai, H. Zhang, K. Zhang, C. Guo, Utility-scale subsurface hydrogen storage: UK

- perspectives and technology, *International Journal of Hydrogen Energy*, 46 (2021) 25137-25159.
- [15] R. Tarkowski, Perspectives of using the geological subsurface for hydrogen storage in Poland, *International Journal of Hydrogen Energy*, 42 (2017) 347-355.
- [16] R. Singh, A. Altaee, S. Gautam, Nanomaterials in the advancement of hydrogen energy storage, *Heliyon*, 6 (2020) e04487.
- [17] U. Bünger, J. Michalski, F. Crotogino, O. Kruck, Large-scale underground storage of hydrogen for the grid integration of renewable energy and other applications, in: *Compendium of hydrogen energy*, Elsevier, 2016, pp. 133-163.
- [18] C. Tarhan, M.A. Çil, A study on hydrogen, the clean energy of the future: Hydrogen storage methods, *Journal of Energy Storage*, 40 (2021) 102676.
- [19] L.J. Murray, M. Dincă, J.R. Long, Hydrogen storage in metal-organic frameworks, *Chemical Society Reviews*, 38 (2009) 1294-1314.
- [20] J.M. Miocic, J. Alcalde, N. Heinemann, I. Marzan, S. Hangx, Toward Energy-Independence and Net-Zero: The Inevitability of Subsurface Storage in Europe, *ACS Energy Letters*, 7 (2022) 2486-2489.
- [21] S.H. Ho, M.M. Rahman, Three-dimensional analysis for liquid hydrogen in a cryogenic storage tank with heat pipe-pump system, *Cryogenics*, 48 (2008) 31-41.
- [22] P. Preuster, A. Alekseev, P. Wasserscheid, Hydrogen storage technologies for future energy systems, *Annual review of chemical and biomolecular engineering*, 8 (2017) 445-471.
- [23] N. Rusman, M. Dahari, A review on the current progress of metal hydrides material for solid-state hydrogen storage applications, *International Journal of Hydrogen Energy*, 41 (2016) 12108-12126.
- [24] M.B. Ley, L.H. Jepsen, Y.-S. Lee, Y.W. Cho, J.M.B. Von Colbe, M. Dornheim, M. Rokni, J.O. Jensen, M. Sloth, Y. Filinchuk, Complex hydrides for hydrogen storage—new perspectives, *Materials Today*, 17 (2014) 122-128.
- [25] N. Heinemann, J. Alcalde, J.M. Miocic, S.J. Hangx, J. Kallmeyer, C. Ostertag-Henning, A. Hassanpouryouzband, E.M. Thaysen, G.J. Strobel, C. Schmidt-Hattenberger, Enabling large-scale hydrogen storage in porous media—the scientific challenges, *Energy & Environmental Science*, 14 (2021) 853-864.
- [26] D. Zivar, S. Kumar, J. Foroozesh, Underground hydrogen storage: A comprehensive review, *International journal of hydrogen energy*, 46 (2021) 23436-23462.
- [27] Y. Qiu, S. Zhou, J. Wang, J. Chou, Y. Fang, G. Pan, W. Gu, Feasibility analysis of utilising underground hydrogen storage facilities in integrated energy system: case studies in China, *Applied energy*, 269 (2020) 115140.
- [28] K.S. Basniev, R.J. Omelchenko, F.A. Adzynova, Underground hydrogen storage problems in Russia, in: *18th world hydrogen energy conference*, 2010.
- [29] S. Flesch, D. Pudlo, D. Albrecht, A. Jacob, F. Enzmann, Hydrogen underground storage—Petrographic and petrophysical variations in reservoir sandstones from laboratory experiments under simulated reservoir conditions, *International Journal of Hydrogen Energy*, 43 (2018) 20822-20835.
- [30] N. Dopffel, S. Jansen, J. Gerritse, Microbial side effects of underground hydrogen storage—Knowledge gaps, risks and opportunities for successful implementation, *International Journal of Hydrogen Energy*, 46 (2021) 8594-8606.
- [31] I. Iordache, D. Schitea, A.V. Gheorghe, M. Iordache, Hydrogen underground storage in Romania, potential directions of development, stakeholders and general aspects, *international journal of hydrogen energy*, 39 (2014) 11071-11081.
- [32] A. Ozarslan, Large-scale hydrogen energy storage in salt caverns, *International journal of hydrogen energy*, 37 (2012) 14265-14277.
- [33] X. Zhao, Q. Cai, S. Zhang, K. Luo, The substitution of wind power for coal-fired power to realize China's CO₂ emissions reduction targets in 2020 and 2030, *Energy*, 120 (2017) 164-178.
- [34] L. Hong, H. Lund, B.V. Mathiesen, B. Möller, 2050 pathway to an active renewable energy scenario for Jiangsu province, *Energy Policy*, 53 (2013) 267-278.
- [35] W. Liu, Z. Zhang, J. Chen, D. Jiang, F. Wu, J. Fan, Y. Li, Feasibility evaluation of large-scale underground hydrogen storage in bedded salt rocks of China: A case

study in Jiangsu province, *Energy*, 198 (2020) 117348.

[36] J. Chen, W. Liu, D. Jiang, J. Zhang, S. Ren, L. Li, X. Li, X. Shi, Preliminary investigation on the feasibility of a clean CAES system coupled with wind and solar energy in China, *Energy*, 127 (2017) 462-478.

[37] J. Fan, H. Xie, J. Chen, D. Jiang, C. Li, W.N. Tiedeu, J. Ambre, Preliminary feasibility analysis of a hybrid pumped-hydro energy storage system using abandoned coal mine goafs, *Applied Energy*, 258 (2020) 114007.

[38] A. Lanz, J. Heffel, C. Messer, Hydrogen fuel cell engines and related technologies, in, United States. Department of Transportation. Federal Transit Administration, 2001.

[39] M. Ruith, E. Meiburg, Miscible rectilinear displacements with gravity override. Part 1. Homogeneous porous medium, *Journal of Fluid Mechanics*, 420 (2000) 225-257.

[40] W.L. Vos, L.W. Finger, R.J. Hemley, H.-k. Mao, Novel H₂-H₂O clathrates at high pressures, *Physical Review Letters*, 71 (1993) 3150.

[41] T. Krader, E. Franck, The Ternary Systems H₂O-CH₄-NaCl and H₂O-CH₄-CaCl₂ to 800 K and 250 MPa, *Berichte der Bunsengesellschaft für physikalische Chemie*, 91 (1987) 627-634.

[42] N. Heinemann, M. Booth, R.S. Haszeldine, M. Wilkinson, J. Scafidi, K. Edlmann, Hydrogen storage in porous geological formations—onshore play opportunities in the midland valley (Scotland, UK), *International Journal of Hydrogen Energy*, 43 (2018) 20861-20874.

[43] M. Bai, K. Song, Y. Sun, M. He, Y. Li, J. Sun, An overview of hydrogen underground storage technology and prospects in China, *Journal of Petroleum Science and Engineering*, 124 (2014) 132-136.

[44] D.G. Caglayan, N. Weber, H.U. Heinrichs, J. Linßen, M. Robinius, P.A. Kukla, D. Stolten, Technical potential of salt caverns for hydrogen storage in Europe, *International Journal of Hydrogen Energy*, 45 (2020) 6793-6805.

[45] D. Pudlo, L. Ganzer, S. Henkel, M. Kühn, A. Liebscher, M.D. Lucia, M. Panfilov, P. Pilz, V. Reitenbach, D. Albrecht, The H₂STORE project: hydrogen

underground storage—a feasible way in storing electrical power in geological media?, in: *Clean energy systems in the subsurface: production, storage and conversion*, Springer, 2013, pp. 395-412.

[46] A. Liebscher, J. Wackerl, M. Streibel, Geologic storage of hydrogen—fundamentals, processing, and projects, *Hydrogen science and engineering: Materials, processes, systems and technology*, (2016) 629-658.

[47] R. Tarkowski, Underground hydrogen storage: Characteristics and prospects, *Renewable and Sustainable Energy Reviews*, 105 (2019) 86-94.

[48] J. Michalski, U. Büniger, F. Crocogino, S. Donadei, G.-S. Schneider, T. Pregger, K.-K. Cao, D. Heide, Hydrogen generation by electrolysis and storage in salt caverns: Potentials, economics and systems aspects with regard to the German energy transition, *International Journal of Hydrogen Energy*, 42 (2017) 13427-13443.

[49] G. Han, M. Bruno, K. Lao, J. Young, L. Dorfmann, Gas storage and operations in single-bedded salt caverns: stability analyses, *Spe Production & Operations*, 22 (2007) 368-376.

[50] A.S. Lord, P.H. Kobos, D.J. Borna, Geologic storage of hydrogen: Scaling up to meet city transportation demands, *International journal of hydrogen energy*, 39 (2014) 15570-15582.

[51] C. Sambo, A. Dudun, S.A. Samuel, P. Esenenjor, N.S. Muhammed, B. Haq, A review on worldwide underground hydrogen storage operating and potential fields, *International Journal of Hydrogen Energy*, (2022).

[52] A. Sáinz-García, E. Abarca, V. Rubí, F. Grandia, Assessment of feasible strategies for seasonal underground hydrogen storage in a saline aquifer, *International journal of hydrogen energy*, 42 (2017) 16657-16666.

[53] V. Reitenbach, L. Ganzer, D. Albrecht, B. Hagemann, Influence of added hydrogen on underground gas storage: a review of key issues, *Environmental Earth Sciences*, 73 (2015) 6927-6937.

[54] S.R. Thiyagarajan, H. Emadi, A. Hussain, P. Patange, M. Watson, A comprehensive review of the mechanisms and efficiency of underground hydrogen storage, *Journal of Energy Storage*, 51 (2022) 104490.

[55] K. Luboń, R. Tarkowski, Numerical simulation of

- hydrogen injection and withdrawal to and from a deep aquifer in NW Poland, *international journal of hydrogen energy*, 45 (2020) 2068-2083.
- [56] M. Panfilov, G. Gravier, S. Fillacier, Underground storage of H₂ and H₂-CO₂-CH₄ mixtures, in: *ECMOR X-10th European conference on the mathematics of oil recovery*, European Association of Geoscientists & Engineers, 2006, pp. cp-23-00003.
- [57] O. Kruck, F. Crotagino, R. Prelicz, T. Rudolph, Assessment of the potential, the actors and relevant business cases for large scale and seasonal storage of renewable electricity by hydrogen underground storage in Europe, *KBB Undergr. Technol. GmbH*, (2013).
- [58] W.T. Pfeiffer, S. Bauer, Subsurface porous media hydrogen storage-scenario development and simulation, *Energy Procedia*, 76 (2015) 565-572.
- [59] C.R. Matos, J.F. Carneiro, P.P. Silva, Overview of large-scale underground energy storage technologies for integration of renewable energies and criteria for reservoir identification, *Journal of Energy Storage*, 21 (2019) 241-258.
- [60] R. Tarkowski, B. Uliasz-Misiak, Towards underground hydrogen storage: A review of barriers, *Renewable and Sustainable Energy Reviews*, 162 (2022) 112451.
- [61] N. Heinemann, J. Alcalde, J.M. Miocic, S.J.T. Hangx, J. Kallmeyer, C. Ostertag-Henning, A. Hassanpouryouzband, E.M. Thaysen, G.J. Strobel, C. Schmidt-Hattenberger, K. Edlmann, M. Wilkinson, M. Bentham, R. Stuart Haszeldine, R. Carbonell, A. Rudloff, Enabling large-scale hydrogen storage in porous media – the scientific challenges, *Energy & Environmental Science*, 14 (2021) 853-864.
- [62] N. Heinemann, J. Scafidi, G. Pickup, E. Thaysen, A. Hassanpouryouzband, M. Wilkinson, A. Satterley, M. Booth, K. Edlmann, R. Haszeldine, Hydrogen storage in saline aquifers: The role of cushion gas for injection and production, *International Journal of Hydrogen Energy*, 46 (2021) 39284-39296.
- [63] N. Kampman, A. Busch, P. Bertier, J. Snippe, S. Hangx, V. Pipich, Z. Di, G. Rother, J. Harrington, J.P. Evans, Observational evidence confirms modelling of the long-term integrity of CO₂-reservoir caprocks, *Nature Communications*, 7 (2016) 1-10.
- [64] N. Thüns, B. Krooss, Q. Zhang, H. Stanjek, The effect of H₂ pressure on the reduction kinetics of hematite at low temperatures, *International Journal of Hydrogen Energy*, 44 (2019) 27615-27625.
- [65] C. Gaucher, A.N. Sial, G.P. Halverson, H.E. Frimmel, Neoproterozoic-Cambrian tectonics, global change and evolution: a focus on South Western Gondwana, Elsevier, 2009.
- [66] J. Bourgeois, N. Aupaix, R. Bloise, J. Millet, Proposition d'explication de la formation d'hydrogène sulfuré dans les stockages souterrains de gaz naturel par réduction des sulfures minéraux de la roche magasin, *Revue de l'Institut Français du Pétrole*, 34 (1979) 371-386.
- [67] A. Hassanpouryouzband, K. Adie, T. Cowen, E.M. Thaysen, N. Heinemann, I.B. Butler, M. Wilkinson, K. Edlmann, Geological Hydrogen Storage: Geochemical Reactivity of Hydrogen with Sandstone Reservoirs, *ACS Energy Letters*, 7 (2022) 2203-2210.
- [68] C. Hemme, W. Van Berk, Hydrogeochemical modeling to identify potential risks of underground hydrogen storage in depleted gas fields, *Applied Sciences*, 8 (2018) 2282.
- [69] N. Hassannayebi, S. Azizmohammadi, M. De Lucia, H. Ott, Underground hydrogen storage: application of geochemical modelling in a case study in the Molasse Basin, Upper Austria, *Environmental Earth Sciences*, 78 (2019) 1-14.
- [70] Z. Bo, L. Zeng, Y. Chen, Q. Xie, Geochemical reactions-induced hydrogen loss during underground hydrogen storage in sandstone reservoirs, *International Journal of Hydrogen Energy*, 46 (2021) 19998-20009.
- [71] R. Ershadnia, M. Singh, S. Mahmoodpour, A. Meyal, F. Moeini, S.A. Hosseini, D.M. Sturmer, M. Rasoulzadeh, Z. Dai, M.R. Soltanian, Impact of geological and operational conditions on underground hydrogen storage, *International Journal of Hydrogen Energy*, (2022).
- [72] S.J. Varjani, E. Gnansounou, Microbial dynamics in petroleum oilfields and their relationship with physiological properties of petroleum oil reservoirs, *Bioresource technology*, 245 (2017) 1258-1265.

- [73] F. Freund, J.T. Dickinson, M. Cash, Hydrogen in rocks: an energy source for deep microbial communities, *Astrobiology*, 2 (2002) 83-92.
- [74] P. Amigáñ, M. Greksak, J. Kozánková, F. Buzek, V. Onderka, I. Wolf, Methanogenic bacteria as a key factor involved in changes of town gas stored in an underground reservoir, *FEMS Microbiology Ecology*, 6 (1990) 221-224.
- [75] G. Strobel, B. Hagemann, T.M. Huppertz, L. Ganzer, Underground bio-methanation: Concept and potential, *Renewable and Sustainable Energy Reviews*, 123 (2020) 109747.
- [76] A. Ebrahimiyehta, Characterization of geochemical interactions and migration of hydrogen in sandstone sedimentary formations: application to geological storage, in, Université d'Orléans, 2017.
- [77] M. Panfilov, Underground storage of hydrogen: in situ self-organisation and methane generation, *Transport in porous media*, 85 (2010) 841-865.
- [78] M. Pichler, Underground Sun Storage Results and Outlook, in: EAGE/DGMK Joint Workshop on Underground Storage of Hydrogen, European Association of Geoscientists & Engineers, 2019, pp. 1-4.
- [79] M. Panfilov, Underground and pipeline hydrogen storage, in: *Compendium of hydrogen energy*, Elsevier, 2016, pp. 91-115.
- [80] C. Hemme, W. van Berk, Potential risk of H₂S generation and release in salt cavern gas storage, *Journal of Natural Gas Science and Engineering*, 47 (2017) 114-123.
- [81] A. Amid, D. Mignard, M. Wilkinson, Seasonal storage of hydrogen in a depleted natural gas reservoir, *International Journal of Hydrogen Energy*, 41 (2016) 5549-5558.
- [82] A.B. Cunningham, R. Gerlach, L. Spangler, A.C. Mitchell, Microbially enhanced geologic containment of sequestered supercritical CO₂, *Energy Procedia*, 1 (2009) 3245-3252.
- [83] N. Dopffel, S. Jansen, J. Gerritse, Microbial side effects of underground hydrogen storage – Knowledge gaps, risks and opportunities for successful implementation, *International Journal of Hydrogen Energy*, 46 (2021) 8594-8606.
- [84] Y. Kryachko, Novel approaches to microbial enhancement of oil recovery, *J Biotechnol*, 266 (2018) 118-123.
- [85] Y. Yang, Y. Wu, Y. Lu, M. Shi, W. Chen, Microorganisms and their metabolic activities affect seepage through porous media in groundwater artificial recharge systems: A review, *Journal of Hydrology*, 598 (2021) 126256.
- [86] A. Ebigbo, R. Helmig, A.B. Cunningham, H. Class, R. Gerlach, Modelling biofilm growth in the presence of carbon dioxide and water flow in the subsurface, *Advances in Water Resources*, 33 (2010) 762-781.
- [87] N. Eddaoui, M. Panfilov, L. Ganzer, B. Hagemann, Impact of Pore Clogging by Bacteria on Underground Hydrogen Storage, *Transport in Porous Media*, 139 (2021) 89-108.
- [88] A. Ebigbo, F. Golfier, M. Quintard, A coupled, pore-scale model for methanogenic microbial activity in underground hydrogen storage, *Advances in Water Resources*, 61 (2013) 74-85.