Chapter 1

Introduction

In recent years, climate change has become a topic of growing public interest. Many new policies and multilateral agreements have been implemented to reduce the reliance on fossil fuels and to promote the development and use of renewable energy sources. Many countries have established frameworks and guidelines based on the agreements to enable a transition to sustainable sources. The Paris Climate Accord from 2015 set a target to limit the global increase in temperature to 1.5 °C above the levels from the pre-industrial era [76].

The development of renewable energies is primarily focused on the expansion of capacities of solar and wind power, followed by hydropower and nuclear power. The total global solar power is estimated to be approximately 35 PJ, and wind will be nearly 20 PJ by 2050 in the Net-Zero-Emissions scenarios, according to the IEA's World Energy Outlook [49]. A serious drawback of renewable energy sources is their dependency on weather conditions, with a high electricity generation expected during the summer and windy days. This can lead to imbalances between the produced quantities and the actual energy demand. To bridge these gaps and ensure a sufficient amount of energy supply, large-scale energy storage opportunities are necessary. The surplus renewable energy can be converted to other energy carriers, such as hydrogen and ammonia. These products are part of the Power-To-X conversion process, where the X denotes the target energy that is generated.

Hydrogen is gaining significant attention as a chemical energy carrier. The federal government has emphasized its importance in the National Hydrogen Strategy. It is considered a substantial pillar to achieve carbon neutrality by 2045 and also regarded as an energy carrier that can be used to generate electricity or it is immediately used in industrial processes [4, 28, 74]. In 2021, the world demand for hydrogen (H_2) stood at nearly 94 million tonnes (Mt) [48]. The demand forecast for clean hydrogen is dependent on national efforts and technologies to achieve carbon neutrality. When net-zero emissions are to be achieved globally by 2050 and the global increase in temperature is to be kept below 1.8 °C, the annual demand for clean hydrogen can range between 585 and nearly 620 Mt per year by 2050 [34, 86].

1.1 Hydrogen demand and storage capacity

The Federal Government and National Hydrogen Council in Germany estimate the domestic demand to range between 56 to 110 Terrawatthours (TWh) by 2030 [75, 103], with the current hydrogen consumption at around 55 TWh. Other studies have calculated similar values concerning the hydrogen demand in the coming years. A recent study by the BDI conducted in 2021 determined the needed hydrogen to be approximately 240 TWh for all industrial and social sectors (industry, housing, traffic, agriculture) in Germany by 2045 [11]. Another analysis, the EU-funded HyUsPRe project, concluded that the demand in Germany could reach even 1000 TWh by 2050 [35, 37]. Further studies have calculated that the total hydrogen demand in Germany in all examined sectors and industries might range between 250 to 280 TWh by 2050 [57]. Since the European Commission has recognized hydrogen as an essential component of the Green New Deal to reduce emissions in the Member States, much investigation has been directed towards assessing the subsurface storage potential of hydrogen across the continent. The estimated increase in hydrogen demand could reach 4000 TWh in Europe by 2050. Regional demands vary significantly (c.f. Figure 1.1).

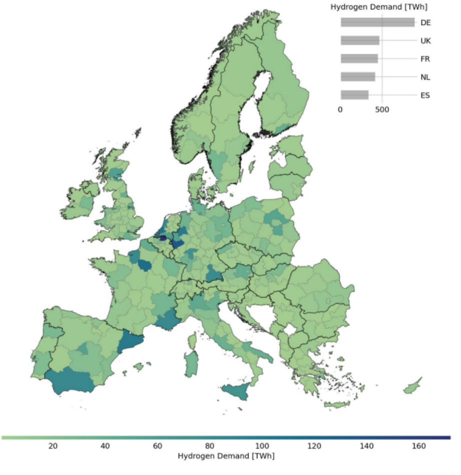

Figure 1.1: Hydrogen demand in the European Union in 2050 [35]

Most of the expected hydrogen demand is projected to be in Central and Western Europe, with the highest local demands forecasted to be in industrial and densely populated areas. Therefore, efficient and large-scale energy storage opportunities are necessary. For many decades, natural gas has been stored in subsurface facilities in times of low demand and was extracted in the colder winter season.

Using those existing facilities is a viable option to store hydrogen in the long run. The most common subsurface storage options are:

- Salt caverns: These are mined or leached large hollow spaces in embedded salt formations, surrounded by a nearly-impermeable salt layer. Due to the vast hollow spaces, salt caverns can operate at a high performance, with short-term high deliverability rates designed to meet temporary peak demands. Salt caverns can also operate at numerous cycles per year and are usually assembled in aggregations of numerous caverns in a field [101].
- Porous formations: The most common subsurface porous storage sites are former hydrocarbon reservoirs. These formations have extensive natural geology and proven structural trapping mechanisms, ensured by the overlying sealing caprock. The stored gas medium is kept inside the porous media and is prevented from migrating into shallower formations. Compared to salt caverns, porous media have larger sizes, which can store a larger volumetric amount of gas. Porous storages have low flow velocities and reduced deliverabilities, making them suitable to meet stable basic demands [101].

Recent studies have estimated the total working gas capacity of hydrogen in existing storage sites to be approximately $260\,\mathrm{TWh}$ across the member states in 2030, assuming all available existing storage facilities are converted. Including non-member EU states, this value is at $349\,\mathrm{TWh}$ (Figure 1.2). The anticipated high storage demand underscores the necessity to use existing storage sites for future hydrogen storage operations.

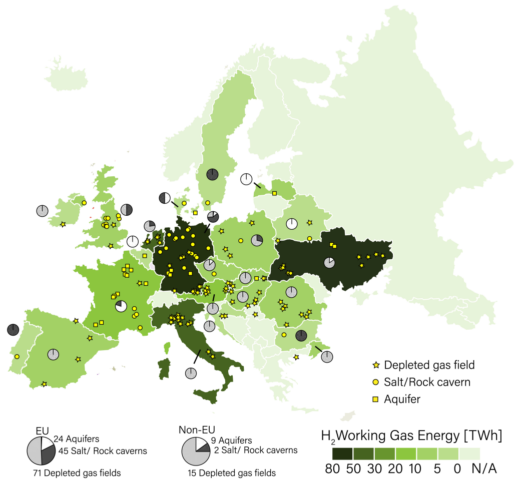

Figure 1.2: Projected hydrogen energy stored capacity in 2030 in the European Union [99] (the pie chart values indicate every subsurface storage type's relative percentage in each country)

Existing porous storage sites (depleted gas or oil reservoirs) possess the largest capacities to store hydrogen, followed by salt or rock caverns. Another type of porous storage site, aquifers, are also considered potential hydrogen storage sites and these could store nearly one-fifth of the entire capacity. The storage sites are distributed all across the European Union, with Germany having the largest hydrogen energy storage capacity (nearly 71 TWh), assuming that all existing storage sites are converted. This value was obtained by using available storage data from public institutions and hydrogen properties (geometrical volume and the density of hydrogen). Another assumption was that the energy of hydrogen is one-quarter of methane and that Germany has nearly one-third of the EU storage capacities [99].

1.1.1. Underground Hydrogen Storage

A result of the increasing necessity to develop long-term storage opportunities for hydrogen in the subsurface is a large number of projects investigating the feasibility of injecting hydrogen into and withdrawing it from the subsurface. Apart from the benefits, which include the use of already existing surface and subsurface facilities and the leveraging of available infrastructure, many challenges facing underground hydrogen storage (UHS) continue to be analyzed. These include integrity issues, geochemical or biochemical reactions or the mixing processes of various gases. This review of past subsurface hydrogen storage tests focuses primarily on the storage of hydrogen in porous formations. The first experience with the injection of hydrogen into the subsurface were gained during the storage of town gas in Ketzin (Germany), Lobodice (Czech Republic) and Beynes (France), beginning in the 1960-ies. Town gas was a gaseous mixture containing H_2, CH_4, CO_2 and nitrogen which was injected into an aquifer for temporary storage. Laboratory analyses have revealed a decrease in the hydrogen and an increase in the methane concentration during the storage period. These changes in the gas composition were attributed to microbial reactions, induced by methanogenic archaea [102]. These microbes consume hydrogen and carbon dioxide to produce methane and water [12, 71].

In the 1970-ies, the first underground hydrogen storage projects were designed and put into operation. In Europe, the first subsurface hydrogen storage test began in Teesside, UK, in 1972 in salt caverns and is still operational with a total storage capacity of nearly 19 t. Larger storage sites (Clemens Terminal) were developed in Texas in 1983 [50]. In the past 10 to 15 years, a large number of research projects focused their investigation on the analysis and feasibility of future hydrogen storage in porous media. A large number of EU consortium projects tackled issues, such as geochemical, microbial risks and deliverability challenges, to improve characterization and developing risk mitigation measures. In the EU projects HyUSPRe (2021-2024) and HyStories (2021-2023), the geochemistry, microbiology and various mixing processes between two gases were analyzed in laboratory experiments. The obtained results were tuned and upscaled to field-scale models to simulate large-scale underground hydrogen storage scenarios. The results improved understanding of the potential challenges and risks associated with hydrogen storage. They also mapped the expected demand, storage capacities, and the forecasted levelized costs of hydrogen storage [14]. Additionally, the geological characteristics of existing gas storage sites were used to draft recommendations that assist in assessing the feasibility of an existing site as a hydrogen storage facility [8].

Another pilot field scale test in a porous formation to store hydrogen was performed in Argentina as part of the company HyChico. Hydrogen was injected with natural gas into a sandstone porous media, shut in for three months and then the gas was withdrawn. The objectives were to analyze the reservoir behavior containing hydrogen and to observe the integrity of the overlying caprock. A subsequent pilot test was designed to analyze methanation processes in the same storage constellation [83].

A field trial in a porous reservoir began in 2013 in Austria as a part in the project Underground Sun Storage until 2017. This project consisted of numerous working packages, which included examining the flow behavior of hydrogen and methane in storage rock samples, the microbial characterization of the formation water, analyzing material integrity in the presence of hydrogen and conducting field trials by injecting 10% of hydrogen in a total volume of $1.22 \times 10^6\,\mathrm{Sm}^3$ into the porous storage site. After a shut-in period of three months, the withdrawn gas composition revealed that 82% of hydrogen was withdrawn and the rest remained in the subsurface. No compromise of the storage site was detected [3]. A follow-up project (Underground Sun Storage 2030) concluded that large-scale subsurface hydrogen storage was feasible on a commercial basis [42].

Extensive investigation of technical and geological integrity of subsurface storage sites has also been carried out. In the project Hy-Integer (2016-2019), the implications of hydrogen and other gas mixtures in the porous subsurface on existing well cement sheaths at reservoir conditions were analyzed in laboratory experiments. The flow of gas through the porous media and the integrity of the structure in the presence of hydrogen in the long-run was evaluated and used for cyclic hydrogen storage simulations [31]. Hydrogen diffusion experiments on pure porous storage rock samples and caprock layers have been performed between 2021-2025 to assess the geological integrity of a storage site to contain hydrogen. The results of the experiments were processed to develop a numerical simulation model to replicate the laboratory works and to predict the reservoir sealability [40]. These diffusion experiments have been extended in a follow-up project (TEN.efzn), which investigates hydrogen diffusion processes through intermediate rock layers consisting of multiple stratigraphic formations [30].

1.1.2. Subsurface methanation

Microbial species, such as methanogenic archaea, by consuming hydrogen and carbon dioxide and producing methane and water (ratio H_2:CH_4 4:1), post one of the most significant challenges to UHS in porous media [38, 102]. Microbes live in the aqueous phase inside the storage site, and accelerate their metabolism when they come into contact with hydrogen and carbon dioxide. According to the Sabatier reaction, hydrogen and carbon dioxide are consumed and methane is produced, which indicates a decrease in hydrogen. In a pure UHS scenario, this would be a disadvantage to the storage efficiency. If the purpose is to develop an underground methanation reactor (UMR), this microbial conversion process forms the basis for this concept. This idea was introduced by Strobel et al. (2020), in which the conversion of hydrogen to methane is intentional, and has a volumetric energy content nearly three times larger than hydrogen. The deliberate methane production results in a higher energy content and provides an important contribution in the energy transition [96].

1.2 Motivation and scope of thesis

The need for large-scale energy storage opportunities to have a sufficient supply available during times of low production of renewable electricity is obvious. So far, the storage of hydrogen in salt caverns has been extensively assessed and investigated. Associated projects have reached a high maturity level with field-scale pilot tests underway in Northern Germany. For instance, a salt cavern in the region Etzel has been filled with $1 \times 10^6\,\mathrm{Sm}^3$ of hydrogen [27]. Another field test is ongoing at the storage site Krummhörn, where a new cavern was leached and filled with $500.000\,\mathrm{Sm}^3$ of hydrogen. Extensive investigations on material compatibility and geological and technical integrity towards H_2 have been successfully performed [43]. In recent years, the utilization of subsurface porous storage sites to achieve this objective has received increased attention. Their large storage capacities, their contribution to the energy base load, their availability and gas storage operational history load make them suitable candidates for large-scale hydrogen storage. Past projects have focused on analyzing the total available capacity and the potential biochemical and geochemical challenges. Different well patterns designed for the cyclic operation have been analyzed in simulations to obtain an optimal well placement strategy to maximize the full-scale deliverability of a storage site. Further analysis also used available existing natural gas withdrawal data to predict the potential hydrogen withdrawal rates from an identical storage site through nodal analysis [52]. However, a big gap remains between the effects of individual gas properties on the withdrawal rates for natural gas and hydrogen. The extent to which gas properties could influence the magnitude of a withdrawal value and its duration also remains unclear. Considering biochemical processes, different microbial development models have been formulated in literature and simulated at large field scales. Replicating microbial cell growth behavior was conducted either in batch experiments