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Research on the Integrity of Underground Salt Cavern Hydrogen Storage

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Abstract. The transition from fossil fuels to renewable energy (especially hydrogen) has become a core strategy for decarbonization and achieving net-zero carbon emissions. Hydrogen storage is crucial in the hydrogen supply chain. Due to the demand for large-scale hydrogen storage, underground hydrogen storage has been explored as an economic method to meet global energy needs. Underground aquifers and depleted oil and gas reservoirs have high costs and immature technologies for hydrogen storage. In contrast, salt cavern hydrogen storage has obvious advantages such as low cost and the most mature technology. However, the safe storage and cyclic utilization of hydrogen in salt caverns require the caprock and reservoir to be highly stable and intact. Currently, research on the integrity of salt cavern hydrogen storage is unsatisfactory and lacks systematic methods. Therefore, this paper aims to review the main challenges related to storage integrity (such as geochemical reactions, microbial activities, geomechanics, etc.), analyze the impacts of various factors on hydrogen storage integrity, and propose feasible methods to mitigate these risks, providing a reference for large-scale underground salt cavern hydrogen storage.

Keywords: salt cavern hydrogen storage, integrity, geochemical reactions, microbial activities, geomechanics

1. Introduction

In response to the escalating climate goals and the recovery of the global economy, renewable energy has developed rapidly. The International Energy Agency (IEA) predicts that by 2026, renewable energy will dominate the global power sector, accounting for 95% of new power capacity. However, the supply of renewable energy represented by solar energy, wind energy, and hydropower is volatile, leading to supply-demand imbalance and posing challenges to the continuous and stable supply of energy [1]. Underground hydrogen storage technology can effectively solve this problem.

Hydrogen energy is a widely recognized renewable, clean, and efficient energy source, with multiple advantages such as abundant sources, environmental friendliness, high energy density, and zero carbon emissions [2]. At present, underground hydrogen storage sites are mainly divided into three types: depleted oil and gas reservoirs, salt caverns, and aquifers [3]. Among them, salt caverns are considered the optimal choice for underground hydrogen storage due to their strong tightness and stability, low buffer gas ratio, and high injection-production frequency [4].

2. Salt cavern hydrogen storage

Salt caverns are man-made underground cavities formed by solution mining in thick underground salt layers or salt domes. The size and shape of salt caverns vary according to different geological conditions, with a volume usually ranging from 5,000 to 1,000,000 cubic meters. Currently, there are only 4 successful cases of large-scale underground storage of high-purity hydrogen worldwide, all of which are salt cavern geological structures, including 1 in the United Kingdom and 3 in the United States.

In 1972, Sabic Petroleum Corporation successfully built the world's first underground salt cavern hydrogen storage facility for pure hydrogen in Teesside, the United Kingdom. The storage facility consists of 3 shallow salt caverns with an effective volume of approximately 70,000 m^3 and a burial depth of 350 m, operating at a constant pressure of 4.5 MPa. In 1983, ConocoPhillips Company of the United States built the Clemens salt cavern hydrogen storage facility on the Clemens Dome salt dome. The salt cavern has a cylindrical structure with a diameter of 49 m, a height of 300 m, a capacity of 580,000 m^3, and a depth of 850-1150 m. In 2007, the salt cavern hydrogen storage facility built by Praxair Corporation of the United States in Moss Bluff was successfully put into operation. The average diameter of the storage facility is 60 meters, the height is 580 meters, the maximum capacity is 560,000 m^3, and the depth is 810-1380 m. The injection pressure during the normal operation of the salt cavern is 7.58-13.45 MPa. In 2014, Air Liquide Corporation of the United States built the world's largest underground salt cavern hydrogen storage facility in Spindletop, which can supply hydrogen for large-scale steam methane reformers for 30 days. The average diameter of the storage facility is about 70 meters, the capacity is 906,000 m^3, and the burial depth is 1500 m. The pressure is maintained at 6.8-20.2 MPa during the operation of the storage facility.

3. Geochemical reactions

When hydrogen is injected into underground salt caverns, it will react with minerals, leading to mineral dissolution and precipitation, which directly affects the integrity and stability of the storage system. Rock salt and its interlayer minerals include rock salt, gypsum, carbonates, sulfates, etc. [5].

Rock salt is generally chemically inert and does not react with H_2. However, factors such as impurities in salt caverns or different hydrogen storage conditions can affect this chemical inertness [6]. In a strongly reducing environment, hydrogen can reduce high-valent ions [7]. For example, S^{6+} in gypsum will be reduced to S^{2-} [8], and Fe^{3+} in clay will also be reduced to Fe^{2+}. On the one hand, the H^+ produced by hydrogen hydrolysis will convert weakly alkaline ions in rocks to neutral, which makes carbonates in calcite (such as CO_3^{2-}, HCO_3^-) react with H_2 to generate CH_4 and CO_2, thereby reducing hydrogen purity. On the other hand, these reactions will consume H^+ and increase the pH value of the cavern, making the environment alkaline, which further promotes hydrogen hydrolysis and aggravates hydrogen loss.

The dissolution of calcite in brine is caused by the decrease in pH value due to water dissociation. Research by Bo et al. [9] shows that calcite, as an impurity in the rock salt structure, its dissolution will lead to significant hydrogen loss. Ahmed et al. [10] conducted experimental studies on the reactions of anhydrite, gypsum, and rock salt in a hydrogen environment and evaluated the impact of hydrogen on the physical and pore structure characteristics of these minerals. The experimental results show that the mineralogical properties of rock salt and anhydrite do not change significantly, but the high concentration of Ca^{2+} and SO_4^{2-} in anhydrite will form H_2S in the cavern.

Geochemical reactions between hydrogen and sensitive minerals can change the porosity and permeability of the caprock, thereby affecting the sealing capacity of the caprock. This is because the containment effect of the caprock on hydrogen seepage and diffusion depends on its physical and chemical properties. For the reservoir, mineral dissolution and precipitation will change the reservoir connectivity and fluid migration characteristics, thereby affecting hydrogen recovery efficiency and final recovery rate.

4. Microbial activities

The number of microorganisms in brine can reach 2-7×10^6 cells/mL. These microbial communities mainly include Actinobacteria, Halobacteria, Desulfobacterota, and Firmicutes [5]. Kirchman's research points out that microbial life requires water, energy, and trace elements such as carbon, nitrogen, and phosphorus [11]. The reservoir temperature of different geological structures is greatly affected by the burial depth. The temperature of depleted gas reservoirs or aquifers is high, reaching 7~174℃, while the temperature of salt caverns is generally 20~35℃ [12]. Microorganisms can survive between -15℃ and 121℃. Since microorganisms are widely present in the formation, their metabolic reactions will cause hydrogen loss and purity reduction. Therefore, it is important to evaluate the impact of microbial activities on the integrity of salt cavern hydrogen storage.

Thaysen's research shows that bacteria obtain metabolic energy through chemical reduction or oxidation, in which electron donors and electron acceptors are essential [13]. The high reducibility and activity of hydrogen make it act as an electron donor in chemical reactions. Bacteria strip electrons from hydrogen atoms and transfer them to the electron acceptor disulfide, and then various hydrogenases activate oxidation reactions to release energy required for bacterial activities. As mentioned earlier, many microorganisms use H_2 as an electron source [14]. They use H_2 to drive their anaerobic respiration. H_2 oxidation in microorganisms is common and can consume a large amount of H_2. All known anaerobic metabolisms can use H_2, such as nitrate reduction, manganese reduction, iron reduction, sulfate reduction, methanogenesis, acetogenesis, etc. However, the content of nitrate, iron, and manganese in salt caverns is extremely low, so the three metabolisms of sulfate reduction, methanogenesis, and acetogenesis are the most important [15].

The conversion of H_2 and CO_2 to CH_4 is one of the common microbial reactions. In an underground coal gas storage facility in the Czech Republic, microbial metabolic activities caused a sharp increase in CH_4 concentration and a significant decrease in H_2 and CO_2 concentrations in the storage facility in a short period of time (H_2 loss was nearly 17%). The amount of CH_4 produced by this reaction is less than the total consumption of H_2 and CO_2, leading to a decrease in the total amount of stored gas and thus a decrease in the gas pressure of the storage facility. Moreover, a major challenge for underground hydrogen storage is microbial sulfate reduction and the generation of hydrogen sulfide. Since even very low concentrations of H_2S can affect gas quality, it is important to analyze underground biological reactions to prevent the formation of hydrogen sulfide. The sulfate reduction process is very efficient, and a small amount of sulfate may generate high concentrations of hydrogen sulfide.

For salt caverns, the risk of microbial activities is usually low because the surface area of salt caverns is much smaller than that of porous media geological structures, which reduces the formation and clogging of microbial biofilms. In addition, the high-salinity environment of salt caverns will increase the osmotic pressure inside microbial cells, greatly reducing microbial activity. However, Hemme et al. [16] found that Halobacteria require more than 100-150 g/L of salt for growth and structural stability, which means that halophilic microorganisms can survive in high-salinity environments. Therefore, although the microbial diversity in salt caverns is affected by high salinity, this does not mean that there is no risk of hydrogen consumption by microbial activities in salt caverns.

5. Geomechanical factors

The cyclic injection and production of hydrogen will cause fluctuations in the storage pressure, leading to periodic changes in the effective stress in the storage facility. The cyclic fluctuation of stress in the reservoir may cause reservoir compaction, resulting in decreased porosity, reduced flow rate, and may trigger land subsidence, fault reactivation, and microseismic activities. Reservoir compaction will promote the settlement of caprock rocks [17], causing bending fractures to appear, and the tightness of the caprock will then fail, leading to hydrogen leakage [18]. In addition, mineral dissolution and precipitation may form fractures and cracks, causing crack closure, rock displacement, and rock mechanical fatigue, resulting in permanent rock deformation and affecting hydrogen storage integrity [19].

Clay minerals in the reservoir, caprock, and faults may undergo stress changes accompanied by clay swelling due to hydrogen adsorption. Although the adsorption capacity of typical swelling clays such as montmorillonite for hydrogen is two to four times lower than that of fluids such as CO_2, and the related stress-strain-adsorption behavior will affect hydrogen injection and production, thereby damaging the stability of the storage system [18], Busch et al. [20] pointed out that fluids and clay water activity significantly affect the swelling potential of clay minerals, and clay swelling is directly related to the water content of clay minerals. In addition, processes such as dissolution-precipitation and crack propagation will accelerate in the presence of water. Therefore, the cyclic injection and production of dry hydrogen during the operation of the storage facility may cause the reservoir to be generally dry, and clay may shrink due to drying, thereby reopening the cracks previously blocked by swelling [21].

Compared with other rocks, rock salt has unique viscoplastic behavior under different pressure and temperature conditions. This characteristic helps prevent crack initiation and propagation and can improve the airtightness of the cavern [22]. However, in some cases, certain stress changes may damage the rock salt and cause it to fracture in a small range. Rock salt fracture is mainly achieved through two mechanisms: tensile failure caused by thermal effects and short-term and long-term shear failure. Among them, short-term shear failure generally occurs in the tensile mode, while long-term shear failure is caused by tertiary creep [23].

6. Conclusions

Salt cavern hydrogen storage is a very promising technical solution that can economically and efficiently store hydrogen and stably and continuously supply electricity to meet future energy needs. Compared with hydrogen storage in other geological structures, salt caverns have low porosity, low permeability, good airtightness, and require a small amount of buffer gas.

Research shows that minerals in salt caverns have a significant impact on the integrity of the hydrogen storage system. This is because these minerals may trigger adverse geochemical reactions, leading to mineral dissolution or precipitation, thereby affecting the injection and production of hydrogen. In addition, minerals such as anhydrite may stimulate the metabolic activities of specific microorganisms to generate hydrogen sulfide, resulting in reduced hydrogen volume and purity. Salt caverns are high-salinity environments, and the activity of most microorganisms is significantly inhibited, but some halophilic microorganisms can still function normally under extremely high salinity. Although there are many studies on underground microbial activities, due to the difficulty in reproducing the complexity of the real environment and most of them being numerical simulation studies, the reliability of experimental conclusions needs to be further verified. From a geomechanical perspective, rock salt has good elastoplastic properties. However, cyclic loading and thermal fatigue may adversely affect the sealing capacity of rock salt, causing cracks and thus hydrogen leakage.

At present, there are few studies on the impacts of geochemical reactions, microbial activities, and geomechanical factors on the integrity of salt cavern hydrogen storage. There is still a lack of sufficient field observations and laboratory data when evaluating these impacts. Before large-scale promotion of salt cavern hydrogen storage technology, more experimental studies and field tests should be carried out to comprehensively evaluate the integrity of the storage facility, thereby providing reference and guidance for the development of actual salt cavern hydrogen storage projects.

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