Modelling underground hydrogen storage: A state-of-the-art review of fundamental approaches and findings

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ABSTRACT

This review presents a state-of-the-art of geochemical, geomechanical, and hydrodynamic modelling studies in the Underground Hydrogen Storage (UHS) domain. Geochemical modelling assessed the reactivity of hydrogen and respective fluctuations in hydrogen losses using kinetic reaction rates, rock mineralogy, brine salinity, and the integration of hydrogen redox reactions. Existing geomechanics studies offer an array of coupled hydro-mechanical models, suggesting a decline in rock failure during the withdrawal phase in aquifers compared to injection phase. Hydrodynamic modelling evaluations indicate the critical importance of relative permeability hysteresis in determining the UHS performance. Solubility and diffusion of hydrogen gas appear to have minimal impact on UHS. Injection and production rates, cushion gas deployment, and reservoir heterogeneity however significantly affect the UHS performance, stressing the need for thorough modelling and experimental studies.

Most of the current UHS modelling efforts focus on assessing the hydrodynamic aspects which are crucial for understanding the viability and safety of UHS. In contrast, the lesser-explored geochemical and geomechanical considerations point to potential research gaps. A variety of modelling software tools such as CMG, Eclipse, COMSOL, and PHREEQC evaluated those UHS underlying effects, along with a few recent applications of data-driven-based Machine Learning (ML) techniques for enhanced accuracy.

This review identified several unresolved challenges in UHS modelling: pronounced lack of expansive datasets, leading to a gap between model predictions and their practical reliability; need robust methodologies capable of capturing natural subsurface heterogeneity while upscaling from precise laboratory data to field-scale conditions; demanding intensive computational resources and novel strategies to enhance simulation efficiency; and a gap in addressing geological uncertainties in subsurface environments, suggesting that methodologies from oil reservoir simulations could be adapted for UHS.

This comprehensive review offers a critical synthesis of the prevailing approaches, challenges, and research gaps in the domain of UHS, thus providing a valuable reference document for further modelling efforts, facilitating the informed advancements in this critical domain towards the realization of sustainable energy solutions.

1. Background

The transition from fossil fuels to renewable forms of energy is of paramount importance in mitigating possible climate change effects. Burning fossil fuels releases large amounts of carbon dioxide and other greenhouse gases into the atmosphere. Therefore, the development of clean and sustainable energy sources has become a top priority for many countries around the world. According to the International Energy Agency (IEA), hydrogen can play a significant role in achieving global climate goals by reducing carbon dioxide emissions from industry, transportation, and power generation (International Energy Agency, 2022). Hydrogen can also help to increase energy security and reduce the dependence on fossil fuels, particularly in countries that rely heavily on imported oil and gas.

Hydrogen gas as an energy carrier has recently been integrated and used in various applications, including transportation, power generation, and industrial processes (Abe et al., 2019; McCay and Shafiee, 2020). In the transportation sector, hydrogen fuel cells are being developed as an alternative to internal combustion engines. Hydrogen fuel cells are an attractive alternative to traditional fossil fuel-powered

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Hydrogen is also gaining popularity in industrial applications, particularly in the steel and chemical industries. Hydrogen can be used as a reducing agent in steel production, replacing coal and other fossil fuels that are currently used (Liu et al., 2021). The global steel sector is expected to potentially reduce its carbon emissions by up to 50% by replacing coal with hydrogen by 2050 (International Energy Agency, 2022). Hydrogen is also an important feedstock in the production of chemicals such as ammonia, methanol, and other chemicals. The use of hydrogen in these industries has the potential to significantly reduce greenhouse gas emissions.

One key aspect of hydrogen is its potential to act as an energy store for excess energy generated from other renewable sources (Gallo et al., 2016). For instance, during periods of low energy demand, excess energy produced from wind and solar power can be converted into hydrogen gas and stored for later use during peak energy demand periods. This can help to stabilize the energy supply and ensure that energy produced from wind and solar power can be converted into electricity. Electrolysis can use renewable energy sources such as wind and solar power to produce hydrogen without greenhouse gas emissions (Yu et al., 2021). This process produces what is known as green hydrogen and has the potential to become an essential component of the clean energy transition. It is estimated that low-emission hydrogen production could reach 24 million tonnes by 2030, up from the current production of less than 1 million tonnes (International Energy Agency, 2022).

One important aspect of the hydrogen economy and supply chain is hydrogen storage (Gallo et al., 2016). UHS in geological formations is a promising method for storing hydrogen gas during periods of low energy demand and releasing it when energy demand increases (see Fig. 2). Compared to above-ground storage options, UHS offers several advantages, including the ability to store hydrogen in the large quantities necessary for widespread use as an energy source, increased safety due to reduced likelihood of gas leaks, and the potential for high-pressure storage to increase energy density and quality enhancing its suitability for use in fuel-cell cars (Zivar et al., 2021a; Flesch et al., 2018).

Aquifers offer several attractive features for hydrogen storage, such as abundance, stability, and the ability to store hydrogen at high pressures. When hydrogen is injected into an aquifer, it dissolves into the water to some extent, is subsequently stored in the rock’s pores and cracks, then ready for later retrieval to satisfy the energy needs. On the other hand, salt caverns as an underground storage option offer several benefits due to their tightness and low reactivity with the hydrogen, thus reducing the possibility of hydrogen loss due to geochemical reactions. Additionally, their visco-plastic properties make them well-suited for sealing gas movement and avoiding mechanical failures. However, the construction of salt caverns requires a source of fresh water for solution mining (Muhammed et al., 2022; Laban, 2020).

Depleted oil and gas reservoirs also represent viable promising options for UHS due to the industrial operators’ experience in injecting water and CO₂, hence the information on reservoir rock and fluid properties, pressure regime, storage capacity, connectivity and geo-mechanical effects on the storage system (Hematpur et al., 2023; Lewandowska-Śmierzchalska et al., 2018). Possible reactions between hydrogen and residual fluids may affect the purity of the stored hydrogen (Hemme and van Berk, 2018). Therefore, it is necessary to carefully evaluate each storage option to ensure the safety and reliability of hydrogen storage. Nonetheless, UHS remains an attractive option for the transition to a hydrogen-based economy to achieve the ongoing research objectives to further improve and expand the technology.

The present knowledge and understanding of UHS have been derived from underground gas storage of other gases such as carbon dioxide and natural gas (Zivar et al., 2021a; Muhammed et al., 2022). However, hydrogen gas has different properties that require an alteration in the

![Fig. 1. Chart showing the types of hydrogen, their production sources, processes and respective CO₂ mitigation options.](chart_url)
underground gas storage process to prevent metal embrittlement from H₂. Current operating UHS projects are primarily located in salt caverns, however, there is a shift towards aquifers and depleted hydrocarbon reservoirs due to their high capacity and available geological information and models, especially in the case of depleted hydrocarbon reservoirs. More details on the operating and pilot studies of UHS can be found in other review papers that have overall focused on underground hydrogen storage (Zivar et al., 2021a; Hematpur et al., 2023; Muhammad et al., 2023; Sambo et al., 2022).

Due to the growing importance of cleaner energy sources, UHS is becoming increasingly significant. However, as most of the current projects are still in the research or pilot stage, it will be a while before this technology can be implemented on a large scale. To expedite the development of UHS, modelling and simulation studies play a crucial role. These studies offer a cost-effective way to evaluate the feasibility of UHS and optimise the design of storage facilities, reducing the risk of expensive errors during the construction and operation of UHS projects. In addition, these studies can identify areas that require further research to improve the safety and performance of UHS.

This publication focuses specifically on critically reviewing and analysing the existing modelling studies of UHS with the objective of providing a clear and concise understanding of the current state of UHS modelling. The review systematically analyses and categorizes the research into three main categories: geochemical, geomechanical, and hydrodynamic aspects. Within each category, the paper investigates the fundamental concepts, process involving equations, modelling approaches, and findings providing a thorough and accurate overview of the existing literature. Additionally, the review dedicates a section to exploring emerging trends in UHS modelling. By methodically comparing various modelling approaches and studies, this paper effectively highlights the current status of modelling efforts and underscores the gaps and challenges faced in the UHS field. The paper concludes by drawing a well-informed conclusion and suggesting the areas for improvement in future modelling efforts.

This information will prove to be valuable for researchers, engineers, and policymakers who are working on the development and implementation of UHS. By building on the knowledge gained from previous modelling studies, we can accelerate the development of this important UHS technology and help to pave the way for a more sustainable energy future.

2. UHS modelling aspects and approaches

UHS is a complex process that is affected by different mechanisms operational during the geochemical, geomechanical and hydrodynamical interactions in the formation. These aspects have a significant impact on the hydrogen gas injection, storage, and production/withdrawal stages of UHS. Moreover, these mechanisms and aspects interact synergistically during UHS making it more complicated to predict their overall impact on the UHS. Therefore, it is essential to understand the individual and collective effects of these mechanisms to properly predict the performance of UHS. Furthermore, the properties and parameters associated with the studied aspect, geochemical, geomechanical and hydrodynamical, require careful calculation, measurement, or estimation. These properties and parameters can be rock properties, fluid properties, operational parameters, geomechanical properties, reservoir conditions, microbial conditions, among others. A summary of the most important parameters, properties and mechanisms operational during UHS is shown in Fig. 3 and further details are discussed in the next sections. The Various modelling approaches have been proposed and undertaken to investigate these effects. A detailed description of the effects and the undertaken modelling approaches in the previous studies will be presented in the following subsections.

2.1. Geochemical

In this section the geochemical aspects of UHS with respect to fundamentals, modelling approaches, and previous studies findings will be discussed. Various parameters should be considered in geochemical modelling these include pressure and temperature of the reservoir, the resident fluid’s composition, salinity and pH, and the type of microorganisms and their rate of growth. These parameters affect various geochemical phenomena that define the overall geochemical impact during UHS, and these are abiotic reactions, biotic reactions, and mineral dissolution and precipitation reactions. A summary of the most important parameters and phenomena for geochemical modelling is depicted in Fig. 4.

2.1.1. Fundamentals and modelling approaches

Geochemical reactions during UHS may occur between the stored hydrogen gas, cushion gas, reservoir fluids and reservoir minerals (Yekta et al., 2018). A cushion gas is a gas that is injected into the storage zone
before the injection of the hydrogen gas for the purposes of providing pressure support and limiting the spread of hydrogen gas inside the reservoir. Investigated cushion gases involve CH\textsubscript{4}, N\textsubscript{2}, and CO\textsubscript{2} (Saeed and Jadhawar, 2023). The geochemical reactions may lead to losses in the stored hydrogen due to converting hydrogen to other gases such as methane and hydride sulfide. Moreover, geochemical reactions have the potential of reducing the purity of hydrogen gas at the time of production due to mixing with other gases present in the reservoir. The
hydrogen gas purity refers to percentage of hydrogen gas produced during the withdrawal stage. This purity can be affected by the produc-
tion of associated gases such as the utilised casing gas, residual gas in the case of storage in depleted gas reservoirs or gases produced as a result of the geochemical reactions involving hydrogen gas which may happen over a prolonged period of time depending on the storage con-
tions (Muhammed et al., 2023). Another important possible effect of the geochemical activity of hydrogen gas during UHS is the possibility of the geological structure’s mineral dissolution or precipitation leading to a change in its porosity and permeability. Consequently, the geological structure’s geomechanical properties may be altered resulting in possible structure failure or faulting and reducing cap rock integrity (Hemme and van Berk, 2018). Geochemical reactions (see Fig. 5 and Table 1) during UHS can either be abiotic or biotic reactions. Abiotic reactions are those that occur between hydrogen and non-living organisms such as brine, oil, gas, and rock minerals. Abiotic reactions can be equilibrium reactions, mineral dissolution/precipitation reactions, or ion exchange reactions (Zivar et al., 2021a). Equilibrium reactions are fast reactions that are both forward and reverse reactions such that the concentrations of the reactants and products remain constant over time. They can be written in the general form of Eq. (1).

$$ aR + bS \rightarrow cP $$

where $a$, $b$ and $c$ are the stoichiometric coefficient of geochemical species $R$, $S$ and $P$, respectively. Their equilibrium constants ($K$) can be calculated using Eq. 2.

$$ K = \frac{[P]}{[R]^{a}[S]^{b}} $$

where $a$ is the activity of their respective chemical species. The activities are related to the molar concentrations of their species through their individual activity coefficients as shown in Eq. (3).

$$ a_i = \gamma_i C_i $$

WATEEQ Debye-Hückel model (Eq. (4)) can be used to calculate the activity coefficients:

$$ \log \gamma_i = -\frac{A_i^2 \sqrt{I}}{1 + \sqrt{I}} - b_i I $$

where $I$ is the ionic strength and for WATEEQ Debye-Hückel equation, $C_i$ and $b_i$ are ion specific parameters fitted from mean-salt activity coefficient (Parkhurst and Appelo, 2013a). $A$ and $B$ are constant values depend on temperature. The ionic strength is a function of the concentration of all the ions present in the solution and is expressed as in Eq. (5).

![Image](image-url)

**Table 1**

A selection of possible geochemical reactions during UHS (Hemme and van Berk, 2018; Korrani et al., 2016).

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Log $K_{eq}$</th>
<th>Reaction</th>
<th>Log $K_{eq}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_2O \rightarrow H^+ + OH^-$</td>
<td>-14.0</td>
<td>$Na^+ + X^- \rightarrow NaX$</td>
<td>0</td>
</tr>
<tr>
<td>$Al^3+ + 4H_2O \rightarrow Al(OH)_4^3-$</td>
<td>-22.7</td>
<td>$Ca^{2+} + 2X^- \rightarrow CaX_2$</td>
<td>0.8</td>
</tr>
<tr>
<td>$Al^3+ + 3H_2O \rightarrow Al(OH)_3 + 3H^+$</td>
<td>-16.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$Al(OH)_3 + 3H_2O \rightarrow Al(OH)_4^3-$</td>
<td>-10.1</td>
<td>$NaHSiO_3 + 8H_2O \rightarrow Na^+ + Al(OH)_3 + 3H_2SiO_4$</td>
<td>-20.573</td>
</tr>
<tr>
<td>$CO_3^{2-} + H^+ \rightarrow HCO_3^-$</td>
<td>-5.0</td>
<td>$NaHSiO_3 + 8H_2O \rightarrow Na^+ + Al(OH)_3 + 3H_2SiO_4$</td>
<td>-18</td>
</tr>
<tr>
<td>$CO_3^{2-} + H^+ \rightarrow CO_2 + H_2O$</td>
<td>10.329</td>
<td>$Al_2Si_2O_5(OH)_4 + 6H^+ \rightarrow H_2O + 2H_2SiO_4 + 2Al^{3+}$</td>
<td>7.435</td>
</tr>
<tr>
<td>$CO_3^{2-} + Ca^{2+} + H^+ \rightarrow CaHCO_3$</td>
<td>16.681</td>
<td>$SiO_2 + 2H_2O \rightarrow H_2SiO_4$</td>
<td>3.95</td>
</tr>
<tr>
<td>$HCO_3^- + Na^+ \rightarrow NaHCO_3$</td>
<td>11.396</td>
<td>$CaCO_3 + CO_2^{2-} + Ca^{2+}$</td>
<td>8.48</td>
</tr>
<tr>
<td>$CO_3^{2-} + Mg^{2+} + H^+ \rightarrow MgHCO_3$</td>
<td>-0.25</td>
<td>$Fe_2S + 2H^+ + 2e^- \rightarrow Fe^{2+} + 2HS$</td>
<td>-18.479</td>
</tr>
<tr>
<td>$CO_3^{2-} + Na^+ \rightarrow NaCO_3$</td>
<td>11.399</td>
<td>$K_0-Mg_{25,25}Al_{25}Si_{25}O_{52}(OH)<em>{25} + 11.2H_2O \rightarrow 0.68 \times 0.25Mg</em>{2}Si_{2}O_{5}$</td>
<td>-40.267</td>
</tr>
<tr>
<td>$CO_3^{2-} + Ca^{2+} \rightarrow CaCO_3$</td>
<td>1.27</td>
<td>$2.3Al(OH)^{3+} + 3.5H_2SiO_4 + 1.2 H^+ \rightarrow CaMg(CO_3)_{2} + Mg^{2+} + 17.09$</td>
<td></td>
</tr>
<tr>
<td>$CO_3^{2-} + Mg^{2+} + 3H_2O \rightarrow Ca^{2+} + MgCO_3 + H_2O$</td>
<td>3.224</td>
<td>$2CO_2 + 4H_2 + CH_2COOH + 2H_2O$</td>
<td>-12</td>
</tr>
<tr>
<td>$Ca^{2+} + H_2O \rightarrow CaOH^+ + H^+$</td>
<td>2.98</td>
<td>$MgH^+ + H_2O \rightarrow Mg^{2+} + H_2O$</td>
<td>-12.78</td>
</tr>
<tr>
<td>$Mg^{2+} + H_2O \rightarrow Mg^{2+} + H^+$</td>
<td>-12.78</td>
<td>$2CO_2 + 4H_2 + CH_2COOH + 2H_2O$</td>
<td>-12</td>
</tr>
<tr>
<td>$Na^+ + OH^- \rightarrow NaOH$</td>
<td>-11.44</td>
<td>$SO_2^{2-} + 5H_2O \rightarrow H_2S + 4H_2O$</td>
<td>35</td>
</tr>
</tbody>
</table>

Fig. 5. An example of geochemical activity during UHS (after Hemme et al. (Hemme and van Berk, 2018)).
$I = 0.5 \sum_{i=1}^{\infty} c_i ^2 \eta _i$  

where $c_i$ is the molar concentration of ion $i$ and $\eta$ is the valence number of that ion $i$. The hydrogen solubility in water can be written as a geochemical reaction in the form of Eq. (6).

$H_2(g) \leftrightarrow H_2(aq)$

where the subscripts (g) and (aq) denote that the hydrogen is present in gaseous phase or aqueous phase, respectively. The equilibrium constant for this reaction can be considered Henry’s constant which relates the partial pressure of the gas with the concentration of hydrogen dissolved in the solution and depends on both the pressure and temperature conditions. The other equilibrium reactions that occur in a H2 geologic storage system (examples shown Table 1), even if they do not involve hydrogen directly, affect the pH and chemistry of the solution which directly affects the solubility of H2 in water. Increasing the ionic strength of a solution, such as by increasing its salinity, often leads to a 'salting-out' effect for many gases. This effect results in decreased gas solubility in the solution.

Ion exchange reactions (examples in Table 1) are also reversible reactions where a free ion in a solution replaces another ion that is bound to a solid surface or solution. The activity coefficient ion exchange reactions can be calculated using WATEEQ Debye-Hückel model. The use of selectivity coefficients follows the Gaines-Thomas convention (Gaines and Thomas, 1953). The cation exchange capacity (CEC) that quantifies the ion exchange is defined as the measure of how many cations can be retained on soil particle surfaces (Brady and Weil, 2016) and is related to the equivalent fractions using Eq. (7):

$\xi_i = \frac{z_i c_i}{\sum_{j=1}^{N_{ex}} z_j c_j}$  

where $c_i$ is the concentration of the exchange species, $z_i$ is the charge of the exchange species, and $N_{ex}$ is the total number of exchange species.

Mineral dissolution and precipitation reactions are the kinetically controlled heterogeneous solubility product reactions. The rate of mineral dissolution and precipitation reaction per unit bulk volume of porous medium (mol/(m$^3$s)), $r_p$, is expressed in Eq. (8) (Bethke, 1996)

$\dot{r}_p = \dot{A}_p \beta \phi (1 - Q_{i\rightarrow j} / K_{p,i\rightarrow j})$, $\beta = 1, \ldots, n_{j(r,m)}$  

where $\dot{A}_p$ is the reactive surface area of reactant mineral $\beta$ per unit bulk volume of porous medium (m$^2$/m$^3$s), $K_{p,i\rightarrow j}$ is the rate constant of mineral reaction $\beta$ (mol/m$^3$s). $K_{p,i}$ is the chemical equilibrium constant of mineral reaction $\beta$, $Q_{i\rightarrow j}$ is the activity product of reaction and $n_{j(r,m)}$ is the number of mineral reactions.

Hydrogen biotic reactions as a result of microbial activities are one of the key aspects that needs to be considered during UHS. These reactions are dictated by the presence and type of living microorganisms (e.g. bacteria) inside the storing site (Boyden et al., 2021). Some of the know biotic processes involves the consumption of hydrogen are methanogenesis, acetogenesis, sulfate reduction and iron reduction. Methanogenesis is the conversion of hydrogen to methane and water when reacting with CO2 in the presence of methanogens (Panfilov, 2016). It is also known as the Sabatier reaction and is shown in Eq. 9.

$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$  

Acetogenesis is another type of biotic reactions that occurs between H2 and CO2 in the presence of acetogen bacteria. This reaction produces acetic acid and water as shown in Eq. (10) (Rabii et al., 2019; Gregory et al., 2019)

$2CO_2 + 4H_2 \rightarrow CH_3COOH + 2H_2O$  

Sulfate reduction reactions convert H2 and sulfate to hydrogen sulfide (H2S) in the presence of sulfate-reduction bacteria (SRB) (Thayyen et al., 2021; Cavallaro et al., 2005). SRB are mostly active in temperatures around 38 °C (Bernardez et al., 2013) but have been reported to be active at temperatures up to 110 °C (Machel, 2001). However, it is expected that its activity is slower in acidic aqueous and depleted hydrocarbon reservoirs, especially in the presence of high salinity brine.

$SO_4^{2-} + 5H_2 \rightarrow H_2S + 4H_2O$  

Most of the existing studies addressing the modelling of the geochemical and biochemical effects during UHS used US Geological Survey’s PHREEQC. PHREEQC is a state-of-the-art geochemical modelling software that allows for the modelling of aqueous, mineral, ion-exchange and surface complexation reactions (Parkhurst and Appelo, 2013a). To calculate solute activities, PHREEQC uses two ion-association aqueous models (the Lawrence Livermore National Laboratory model and WATEQ4F), Pitzer or, SIT (Specific Ion Interaction Theory) equations. Temperature effects on reactions equilibrium constants are accounted for using Van’t Hoff equation (Eq. (12) (Sandler, 2006)).

$ln K_2 / K_1 = \frac{\Delta H/RT}{R} \left( \frac{1}{T_1} - \frac{1}{T_2} \right)$

where $K_1$ and $K_2$ are the equilibrium constants at temperatures $T_1$ and $T_2$, respectively. $\Delta H$ is the standard enthalpy change of the reaction, R is the gas constant.

It is also possible to use analytical parameters to simulate temperature’s effect on equilibrium constants. PHREEQC enables batch-reaction, 1D transport calculations and utilises the Peng-Robinson equation of state (Eq. (13) (Peng and Robinson, 1976) and Henry’s Law to calculate gas solubilities at high pressures.

$P = \frac{RT V_m}{V_a - b} = \frac{a a}{V_a^2 - 2bV_a - b^2}$

where P is the pressure, T is the temperature, $R$ is the gas constant, V is the molar volume, a and b are empirical constants, and a is a temperature-dependent parameter.

Geochemist’s workbench (GWB), a commercial software developed by Aqueous Solutions LLC, that is also utilised to simulate geochemical and biochemical reactions during UHS. GWB is capable of simulating aqueous, kinetic mineral dissolution and precipitation and surface complexation reactions, 1D and 2D reactive and colloid transport (Bethke and FBISM, 2021). PHREEQC and GWB both are usually used for geochemical modelling, however, GWB’s thermodynamics database is constantly reviewed and updated as opposed to PHREEQC’s database which uses publicly accessible thermodynamics databases. PHREEQC is open source and hence its usage is free but may require some programming knowledge to be able to write the geochemical codes.

2.1.2. Analysis of previous studies’ findings

Amid et al. (2016) modelled the storage of hydrogen in a depleted natural gas reservoir considering the geochemical and biochemical effects. Their results showed that sulfate reduction pose a challenge to the UHS process and is more impactful than methanogenesis. However, hydrogen loss due to dissolution was limited to less than 0.1%. The modelled UHS case could supply 42% of the energy capacity supply compared to the Rough Gas Storage Facility (UK) which is a natural gas facility. This indicates that UHS can indeed provide a viable option for energy storage. Hemme et al. (Hemme and van Berk, 2018) modelled the geochemical and biochemical effects on the underground hydrogen studies. They developed a model using PHREEQC to understand the potential loss of hydrogen and changes in rock properties during the storage process due to solubility, geochemistry, methanation, sulfate reduction and diffusion. The authors included the effect of kinetics for the hydrogen biotic reactions to understand their long-term effects on
the UHS. Results of their work showed that hydrogen loss due to diffusion is minimal. While hydrogen loss as a result of biochemical reactions is dependent on the methanation and sulfate reduction bacteria activity at storage conditions. Another geochemical model was developed by Hassannayebi et al. (2019) to investigate hydrogen geochemical reactions during UHS. They utilised the geochemical software Geochemists Workbench and considered both equilibrium and mineral reactions in a sandstone reservoir at 40 °C and 7.5 bar. Their results show that equilibrium reactions between hydrogen and aqueous-phase components are more dominant during the storage-relevant time span.

Laban (2020) modelled the geochemical aspects of UHS in salt cavities using PHREEQC. Laban’s work showed that bacteria growth rate, sulfate concentration in brine, brine pH and ionic strength, cavern pressure and temperature, and iron ions concentrations are the most influencing factors on hydrogen sulfide production through sulfate reduction bacteria. Bo et al. (2021) developed a geochemical model using PHREEQC to evaluate the losses of hydrogen inside a sandstone reservoir. They found that pressure and temperature play a negligible role in hydrogen loss due to solubility. They also suggested that sandstone reservoirs would be better for hydrogen storage than carbonate reservoirs because water uptake of hydrogen does not react with quartz and clay minerals. Unlike carbonate reservoirs in which calcite dissolves to trigger up to 9.5% hydrogen loss. Moreover, their modelling results show that 87% of calcite would be dissolved over a 30-year period. Therefore, they suggested that deep calcite-free reservoirs with calcite-free caprocks would be better for UHS. Zeng et al. (2022a) investigated the geochemical effects on UHS in Majiagou carbonate reservoir using PHREEQC. They studied hydrogen loss, mineral dissolution and water chemistry over a long period of time. Their results showed that hydrogen loss due to fluid-rock reactions up to 6.6% is expected for the first year compared to 81.1% after 500 years of storage. Calcite dissolution was negligible and limited to only 0.0646% over a 500-year period indicating that the Majiagou formation is a good candidate for UHS. In another work, Zeng et al. (2022b) coupled geochemical modelling using PHREEQC with disjointing pressure calculations to evaluate the effects of various reservoir parameters on the wettability of a hydrogen-brine-calcite system. Their modelling results showed that increasing the temperature and decreasing the salinity increase the disjointing pressure within the hydrogen-brine-calcite system resulting in higher hydrogen gas repulsion from the calcite surface and increasing water wettability. Tremosa et al. (2023) utilised PHREEQC to evaluate the reactivity of hydrogen within UHS in the Lobodice town gas storage site, focusing on its behaviour under various conditions, including microbial influence. They reported that under abiotic conditions, UHS shows minimal hydrogen reactivity. However, microbial-driven processes like methanogenesis, sulfate-reduction, and acetogenesis can lead to hydrogen consumption, yielding methane and hydrogen sulfide. Moreover, methanogenesis, influenced by carbon dioxide and carbonates, significantly contributes to hydrogen losses. Sulfate-reduction has a comparatively smaller effect on hydrogen content, generating hydrogen sulfide and associated corrosion risks. The incorporation of microbial kinetics into models is essential for precise simulations of hydrogen reactivity (Tremosa et al., 2023). The effect of H2-brine-rock interactions on caprock integrity during UHS was investigated by Zeng et al. (2023) using kinetic batch models and analytical estimates. Their findings revealed that the dissolution degrees of tested minerals in different shale types are <1% over 30 years, indicating rock integrity. Their results affirm that H2-brine-shale-rock interactions are unlikely to compromise caprock integrity during UHS. The effects of geochemical reactions on hydrogen and reservoir rock porosity and permeability during UHS in an aquifer have been investigated by Saeed et al. (2023a). Their simulations through PHREEQC revealed that geochemical reactions have a minimal impact on the hydrogen gas in the simulated 30 years period. However, they observed changes in the reservoir rock porosity and permeability as a result of the feldspar and calcite dissolution especially when CO2 was used as a cushion gas.

The results of Amid et al. (2016), Hemme et al. (Hemme and van Berk, 2018) and Saeed et al. (2023a) agree on their prediction of the negligible hydrogen loss due to geochemical reactions and diffusion during UHS which in line with the experimental findings of Hassanpouryouzband et al. (2022) which indicated that there is no risk of hydrogen abiotic reactivity in temperatures up to 80 °C. Unlike the work of Bo et al. (2021) and Zeng et al. (2022a) where they found that hydrogen loss can be up to 9.5% and 81.1% respectively, depending on the storage duration, and reservoir mineral composition and conditions. The variance in projected hydrogen depletion could potentially arise from the inclusion of hydrogen redox reactions, which promote the escalation of hydrogen solubility and subsequent loss. This phenomenon aligns with the investigations conducted by Bo et al. (Saeed et al., 2023a) and Zeng et al. (Hassanpouryouzband et al., 2022), in contradiction to the findings of Amid et al. (Tremosa et al., 2023) and Hemme et al. (Hemme and van Berk, 2018). The adoption of hydrogen redox reactions and their rates is a matter of discussion and research at the moment and requires further experimental work (Hassanpouryouzband et al., 2019), Zhan et al. (2023) investigated the redox reactions’ impact on hydrogen-brine-mineral interactions in UHS. Minerals such as calcite, siderite, quartz, and pyrite at varying dissolved oxygen levels (5.5–5500 ppm), temperature, and pressure were assessed by employing geochemical modelling through PHREEQC. They found that dissolved oxygen concentration amplification yields minimal influence on hydrogen solubility and pH, while siderite and calcite exhibit reactivity, incurring modest hydrogen loss under certain pressures. Similarly, quartz and pyrite displayed limited reactivity. However, they found that carbonate minerals provoke hydrogen dissociation and loss through abiotic geochemical reactions, emphasizing sandstone reservoirs’ role in mitigating hydrogen conversion and contamination concerns in UHS. Calcite dissolution was predicted by Hemme et al. (Hemme and van Berk, 2018), Bo et al. (2021), Zeng et al. (2022a), Zhan et al. (2023), and Saeed et al. (2023a) as a result of the geochemical reactions between hydrogen gas, cushion gas, calcite and brine. This implies that the existence of calcite within the geological formation could induce alterations in the porosity and permeability of the rock, thereby influencing the storage potential and hydraulic conductivity of the reservoir. Moreover, the diminishment of hydrogen resulting from geochemical reactions and diffusion exhibits a direct correlation with variations in pressure and temperature. Elevated pressure and temperature levels are conducive to heightened hydrogen losses during UHS operations (Hemme and van Berk, 2018; Bo et al., 2021). A summary of the studies on geochemical reactions effects during UHS is presented in Table 2.

Geochemical modelling of UHS is an essential aspect of modelling the different aspects of UHS. In addition to the equilibrium reactions, mineral dissolution/precipitation reactions should be properly accounted for in the model. The rates of kinetic reactions are dependent on temperature, pressure and salinity (Hemme and van Berk, 2018). Hence, the adoption of constant reaction rates in the modelling approaches might lead to inaccurate results. Further, geochemical experiments involving hydrogen reactions with various reservoir minerals, brine composition and conditions are needed to calibrate and validate the reaction rates and consequently geochemical modelling of UHS.

2.2. Geomechanical

In this section the geomechanical aspects of UHS will be discussed with respect to fundamentals, modelling approaches, and the finding of previous modelling studies. There are various properties and parameters that impact the geomechanical behaviour of reservoir rock during UHS such as the rock’s tensile, compression and shear stresses, presence of faults and fractures, wellbore geometry, and injection and production rates. As a result, various geomechanical phenomena and effects are modelled including the evaluation of cap rock and wellbore integrity, fracturing of reservoir formation, and faults and fractures reactivation. A
To model geomechanical changes during UHS it is important the determine the stresses exerted on the reservoir rock as a result of the hydrogen gas injection and production. Moreover, it is necessary to determine the rock’s behaviour as a result of these exerted stresses in terms of strength, strain and elasticity. One of the fundamental equations used to model these geomechanical changes is the fluid conservation of momentum equation which is given by Eq. 14.

\[ \rho (\partial \mathbf{v} / \partial t + \mathbf{v} \cdot \nabla \mathbf{v}) = - \nabla P + \mu \nabla^2 \mathbf{v} \]

where \( \mathbf{v} \) is the fluid velocity, and the terms on the right-hand side represent the pressure gradient and viscous stress, respectively (Blyton et al., 2015). This conservation of momentum equation describes the fluid behaviour during H2 injection and production accounting for the pressure gradient, \( \nabla P \). To calculate this pressure gradient, Biot’s equation is needed which describes the interactions between the fluid and rock and can be written as depicted in Eq. (15):

\[ \sigma = \sigma - \beta P \]

where \( \sigma' \) is the effective stress, \( \sigma \) is the total stress, \( P \) is the pore pressure, and \( \beta \) is the Biot coefficient (Gram et al., 2021). Hook’s Law is another fundamental equation utilised to model the geomechanical effects of UHS. It describes the elastic deformation of the rock matrix by relating the stress and strain and it can be written in the following form (Eq. (16))

\[ \sigma = E \varepsilon \]

where \( \sigma \) is the stress, \( \varepsilon \) is the strain, and \( E \) is the elastic modulus of the material. In order to predict the onset of rock failure, Mohr-Coulomb failure criterion can be used. This criterion relates shear strength of the rock to the normal stress and friction angle, and is given by:

\[ \tau = c + \sigma_n \tan(\phi) \]
where $\tau$ is the shear stress, $c$ is the cohesive strength of the rock, $\sigma_n$ is the normal stress, and $\phi$ is the angle of internal friction (Al-Awad, 2002). In UHS modelling, the conservation of momentum equation is used to calculate the pore pressure changes due to hydrogen injection and withdrawal. These changes are then used as input to Biot’s equation to calculate the resulting volumetric strain in the reservoir rock. The strain is then used to calculate the stress changes using Hook’s law. The stress changes are then compared to the strength of the rock, as described by the Mohr-Coulomb criterion, to evaluate the potential for reservoir failure.

Modelling the geomechanical effects of hydrogen gas injection, production, and flow in porous media can be performed in different manners. Two of the techniques to model these effects are one-way coupling or two-way coupling. In one-way coupling, the fluid flow and geomechanical simulations are done sequentially. Typically, the fluid flow simulation is conducted first, generating changes in pore pressure. Then, these changes are fed into the geomechanical model to evaluate stress and deformation. One the other hand, two-way coupling refers to the approach wherein changes in fluid pressures and saturations within the porous medium directly influence the stress-strain behaviour of the solid matrix, and reciprocally, the deformation or mechanical response of the solid matrix concurrently affects the fluid flow paths, saturations, and pressure distributions. Two-coupling ensures that the intricate interactions between fluid dynamics and geomechanical responses are captured in real-time throughout the simulation providing more accurate results (Hawez et al., 2021). Modelling further processes or effects that are co-dependent such as thermal changes and geochemical reactions can also be performed through fully coupled simulations. Including such interconnected processes in fully coupled simulations would provide more accurate results in modelling UHS. However, these types of simulation techniques are much more computationally compared to the one-way coupled technique due to the required iterative technique.

2.2.2. Analysis of previous studies’ findings

Bai et al. (Bai and Tahmasebi, 2022) developed a 3D coupled hydro-mechanical model to evaluate the feasibility of UHS in a saline aquifer at the Powder River Basin of Wyoming State. In their work, they used the one-way coupling approach to simulate the hydromechanical response of UHS in the aquifer, which assumed rock strain and stress are affected by pore pressure without affecting the pore pressure back. Hydrogen properties were calculated based on the Peng-Robinson equation of state. They reported that a maximum of 75% hydrogen could be recovered by the end of the third cycle. Moreover, the integrity of the formation and caprock was evaluated using the Mohr-Coulomb criterion which showed that UHS in the studied structure is geomechanically safe. It was also shown that the possibility of failure decreases during the withdrawal period due to a drop in pore pressure and an increase in the effective stress. Fang et al. (2022) performed numerical simulations using the Thermo-Hydro-Mechanical (THM) coupled simulator FLAC3D-TOUGH2MP to evaluate the applicability of UHS in thin-bedded Anning salt mine (China). TOUGH2MP performs the thermal and hydraulic simulation and FLAC3D performs the mechanical calculations. They demonstrated that the coupled simulator does provide means to evaluate the complex pressure and temperature changes for short term storage in salt cavern. Their work showed that the studied potential salt cavern in Anning mine satisfies the criteria for safe hydrogen storage which are no-spalling, no-creep failure, convergence volume of salt cavern does not affect its storage capacity, and resultant surface subsidence rate should not disturb the surface structures.

Bottcher et al. (Böttcher et al., 2017) carried out numerical modelling through openly available simulator OpenGeoSys to investigate the thermo-mechanical behaviour during UHS in salt caverns. Their results showed that large temperature amplitudes in the working gas may increase the tensile stresses at the cavern boundary. They concluded that reducing the number and frequency of storage cycles can decrease temperature variations to avoid tensile failures. Kumar et al. (2021)
investigated the heterogeneity on the deformation of salt caverns and quantify the state of stress around the caverns by developing an open source 2D finite element simulator. They benchmarked their simulation scheme with experimental data from published literature. Their results showed that salt cavern creep (permanent deformation of solid rocks under the influence of persistent stress) was a slow process and was not affected by the short-term storage of hydrogen. Moreover, the heterogeneity of the salt cavern affects its elasticity differently based on the type (potash, halite) and distribution of impurity. Where the presence of heterogeneous layers was observed to result in much higher local deformations around the cavern compared to a homogeneous formation. Coarita-Tintaya et al. (2023) evaluated the viability and risks associated with utilising salt caverns for UHS. Through a numerical approach, their research integrated hydromechanical characteristics of rock salt encapsulating short- and long-term mechanical behaviours. Their model encompassed elastoplastic, instantaneous damage mechanisms, as well as primary, secondary, and tertiary creep phases. By applying this model, the hydromechanical performance of vertical salt caverns was evaluated for both shallow and deep hydrogen storage scenarios subjected to cyclic injection and withdrawal. They reported that deeper caverns exhibit heightened stability concerns, particularly under daily operational cycles. Nevertheless, gas leakage remains limited due to predominant diffusion transport, even with varying permeabilities. Consequently, they concluded that the stability of salt caverns for green hydrogen storage appears resilient under aggressive operational conditions, boding well for the potential of hydrogen storage initiatives (Coarita-Tintaya et al., 2023). A summary of the modelling studies on geomechanics during UHS is presented in Table 3.

2.3. Hydrodynamic

This section covers hydrodynamic aspects of UHS, including fundamentals, modelling approaches, and findings from previous studies. The main hydrodynamic phenomena observed during UHS can be summarised as H2 lateral spreading, H2 viscous fingering, gas mixing, and permeability of wettability, capillary pressure, and relative permeability also considered properties into hydrodynamic models is essential for accurately predicting UHS performance, making hydrodynamics arguably the most important aspect of UHS modelling.

### 2.3.1. Fundamentals and modelling approaches

Hydrodynamics plays a critical role in understanding fluid flow phenomena within storage formations and is a fundamental aspect of UHS (see Fig. 8). The hydrodynamic behaviour of a fluid in a storage formation is governed by a complex interplay of fluid properties, rock properties, and rock-fluid properties under the prevailing reservoir conditions. In particular, fluid viscosity, density, interfacial tension (IFT), solubility, and diffusivity are important fluid properties that impact fluid flow in storage formations (Muhammed et al., 2022). Rock permeability and porosity are crucial rock properties that determine the ease of fluid flow within the rock and the rock’s storage capacity, respectively. Furthermore, the interactions between rock and fluids under the prevailing reservoir conditions are also critical in determining the efficiency of storage and withdrawal processes (Zivar et al., 2021a; Muhammed et al., 2022; Hashemi et al., 2021a). The rock-fluid properties of wettability, capillary pressure, and relative permeability also significantly influence the amount of hydrogen that can be stored and recovered and the ease of injection and production (Hashemi et al., 2021b; Ali et al., 2021). As a result, the integration of the aforementioned properties into hydrodynamical models is essential for accurately predicting UHS performance.

#### 2.3.1.1. Fluid properties

The most important fluid properties considered in UHS hydrodynamics modelling are viscosity and density. Viscosity is the measure of a fluid’s resistance to flow. Hydrogen’s viscosity is very low compared to other fluids and is 0.0189 cP at standard conditions (Muhammed et al., 2022). This results in challenges during the injection of hydrogen gas into the geological formation. Due to its high mobility, hydrogen gas tends to escape and spread laterally in the reservoir leading to undesirable stored gas losses (Heinemann et al., 2021a). Viscosity can be calculated using the Peng-Robinson EOS which takes into account the intermolecular interactions between the gas molecules (Nasrifar, 2010). Hydrogen gas has a very low density, and it affects the fluids distribution of fluids inside the geological structure leading the hydrogen gas to rise to the top of the structure. Hydrogen gas density and density contrast between the underground fluids become important in the absence of the viscous forces (resulting from injection and production operations) (Nasrifar, 2010). H2 density can also be calculated by first obtaining the molar volume ($V_m$) from the Peng-Robinson EOS and then utilising the ideal gas law:

$$V_m = V/n = RT/P$$
where \( P \) is the pressure, \( V \) is the volume, \( n \) is the number of moles of gas, \( R \) is the gas constant, and \( T \) is the temperature.

2.3.1.2. **Rock properties.** Permeability and porosity are considered the most important rock properties dictating the hydrodynamics of fluids flow in porous media (see Fig. 9). Permeability is the measure of the rock’s ability to permit fluid flow through it. Permeability is calculated
using Darcy’s law and it depends on the properties of the porous media, such as pore size, shape, and connectivity. Darcy’s Law is the basic equation controlling fluid flow through porous media. It connects the pressure gradient and permeability of the porous media to the fluid flow rate (Dake, 1978). Darcy’s law, which describes hydrogen flow in porous media, may be expressed as

\[ Q = -kA \left( \frac{P}{\mu} \right) \]

where \( P \) is the pressure gradient resulting in the flow, \( Q \) is the fluid’s flow rate, \( k \) is the permeability of the porous medium, \( A \) is the cross-sectional area of the media, \( \mu \) is the viscosity of fluid, and \( L \) is the length of the porous media. Permeabilities depend on the rock type and the depositional environment through which the rock was formed. High permeability and homogeneous reservoirs are desirable as they are expected to improve the hydrogen gas’ injectivity and producibility. Another important rock property dictating the hydrodynamics during UHS is the rock’s porosity which is the measure of the rock’s ability to store fluids within its pores. Absolute porosity is a percentage of the pore space available in a rock to the bulk volume of the rock (Dake, 1978). Effective porosity is the percentage of connected pores’ space to the rock’s bulk volume, and it is the porosity that is considered in modelling hydrogen gas, and generally fluids, flow in porous media.

### 2.3.1.3. Rock-fluid properties

Relative permeability and capillary pressure are two essential properties in UHS that influence the flow and distribution of fluids in porous media. Relative permeability is a measure of how easily a fluid can flow through a porous medium compared to another fluid. It is defined as the ratio of the effective permeability of a fluid to its absolute permeability (Pan et al., 2023). It can be calculated using several models such as Stone I, Stone II, and Modified Brooks-Corey Models. The Modified Corey Model can be used to estimate relative permeability in hydrogen-water systems, where the porous media is saturated with the two immiscible fluids (Ahmed, 2010). The model assumes that the relative permeability of each fluid phase is a function of its own saturation and the saturations of the other two phases (wetting and non-wetting phase), and can be written in the general form of equations Eq. 20 to Eq. (21):

\[ k_{r, w} = k_{r, w}^0 \left( \frac{S_w - S_{w,r}}{1 - S_{w,r} - S_{nw,r}} \right)^{n_1} \]

\[ k_{r, nw} = k_{r, nw}^0 \left( \frac{1 - S_w - S_{nw,r}}{1 - S_{w,r} - S_{nw,r}} \right)^{n_2} \]

where \( S_w \) and \( S_{w,r} \) the water saturation and irreducible water saturation, \( S_{nw,r} \) is the residual saturation of the non-wetting phase (hydrogen gas in this case), \( n_1 \) and \( n_2 \) are empirical exponents that depend on the properties of the reservoir and the fluids, and \( k_{r, w}^0 \) and \( k_{r, nw}^0 \) are the end point relative permeabilities for the wetting and non-wetting phase, respectively. Capillary pressure is the pressure difference between two immiscible fluids, such as water and hydrogen, in a porous media. It is an important parameter for predicting fluids distribution in a porous media, and it can be calculated using the Young-Laplace equation (Eq. 22).

\[ P_c = 2\sigma \cos \theta / r \]

where \( P_c \) is the capillary pressure, \( \sigma \) is the interfacial tension between the two fluids, \( \theta \) is the contact angle between the fluid and the solid surface of the porous media, and \( r \) is the radius of curvature of the fluid-fluid interface (Dake, 1978; Ahmed, 2010). Relative permeability and capillary pressure determine the producible hydrogen gas saturation during withdrawal stage.

A few measurements of H2-water relative permeability data have been recorded in the literature. Fig. 10 shows the drainage and imbibition relative permeability curves for H2-water system reported by Boon and Hajibeygi (2022) and Lysny et al. (2022a). It is noted from that a pronounced difference between the drainage and imbibition curves end points exists for both datasets which is a result of the relative permeability hysteresis phenomena. The differences in the residual gas saturation range approximately between 0.28 and 0.42 which then translates to residual gas trapped between the injection and withdrawal phases. The residual trapping of hydrogen gas during the withdrawal phase can be seen on a micro level in Fig. 11. Other forms of hydrogen trapping during UHS are structural trapping due to the entrapment of hydrogen gas within the structures of rocks and dissolution trapping as a result of hydron gas dissolution in water. Hydrogen entrainment as a result of geochemical reactions, biotic and abiotic, is another potential mechanism for losing hydrogen gas during UHS as discussed previously.

The hysteresis between the drainage and imbibition curves of relative permeability result in the entrapment of residual hydrogen gas as a result of the discontinuity in the hydrogen gas phase while the connected hydrogen phase is being produced. This is particularly important in underground hydrogen storage because of the cyclic nature of the operation such that successive injection and withdrawal of hydrogen to and from the porous media result in change in the flow behaviour and consequently relative permeability curves. The hysteresis effects can be assessed by evaluating the amount of the non-wetting phase, here being hydrogen, that stays trapped during a shift in flow from drainage to imbibition. The Land model (Land, 1968) is frequently utilised to investigate this trapping behaviour. This model creates an empirical link between the residual hydrogen saturation \( S_{H_2,r} \) (hydrogen quantity trapped when capillary pressure is null) and the hydrogen saturation at the instant of flow inversion, \( S_{H_2,i} \). The Land model for H2-water system
can be written in the following set of equations Eq. 23 to Eq. 25

\[ S_{H_2,r} = \frac{S_{H_2,i}}{1 + CS_{H_2,i}} \]  

Eq. 23

\[ QC = \frac{1}{S_{H_2,r} + CS_{H_2,i}} \]  

Eq. 24

\[ S_w = \frac{S_w}{1 - S_w} \]  

Eq. 25

where, \( S_{H_2} = 1 - S_w \) and \( C \) is a positive constant, known as the Land coefficient, which is determined experimentally. When \( C = 0 \), it signifies total trapping, whereas \( C \) nearing 1 indicates negligible trapping. For any intermediate value of \( C \), the residually trapped hydrogen gas proportion escalates with an increase in the initial gas saturation. To supplement the Land model, Killough (1976) introduced an interpolation method for identifying scanning curves, which serve as intermediate curves for forecasting the relative permeability curve and its endpoints (Eq. (26) to Eq. (27))

\[ S_{H_2,r} = S_{max,H_2,r} + \frac{S_{H_2,i} - S_{H_2,r,i}}{1 + C(S_{H_2,i} - S_{H_2,r,i})} \]  

Eq. 26

\[ C = \frac{1}{S_{max,H_2,r} - S_{H_2,i}} \times \frac{1}{S_{max,H_2,i} - S_{H_2,r,i}} \]  

Eq. 27

Fig. 10. \( H_2 \)-water relative permeability for drainage and imbibition (from Boon and Hajibeygi (Boon and Hajibeygi, 2022) and Lysyy et al. (Lysyy et al., 2022a)).

Fig. 11. Residual entrapment of hydrogen gas during UHS in an aquifer (modified after Lysyy et al. (Lysyy et al., 2022b)).
By applying a parametric interpolation method, the complete imbibition relative permeability curve for the hydrogen-water system can be derived. This method entails interpolating between the point of flow inversion $S_{hi,i}$ and the corresponding residual saturation $S_{hi,r}$.

### 2.3.1.4. Fluid-fluid properties

IFT is a measure of the attractive forces between the molecules at the interface of two immiscible fluids, and it can affect the capillary pressure and the relative permeability of the fluids within the porous media. H2-water IFT can be directly measured in the laboratory (Hosseini et al., 2022), calculated as a function of capillary pressure using the Young-Laplace Equation (Eq. 22) or modelled through molecular dynamic simulations (van Rooijen et al., 2022). Another fluid-fluid phenomenon that affects the hydrodynamics of UHS is the hydrogen gas’s molecular diffusivity. Diffusivity describes the ability of hydrogen gas molecules to dissipate through a porous media (Charlet et al., 2017). Molecular diffusion is usually calculated using Fick’s law which can be written as follows:

$$ J = \rho_c D_c \nabla c $$

where $D_c$ is the effective diffusion coefficient of component $c$, $c$ is the mole fraction of component $k$ (Hagemann et al., 2015).

Hydrogen dissolves in water in small amounts in it depends on exerted pressure, temperature, and water salinity. Many experimental studies measured the solubility of hydrogen in water in different conditions (Wiebe and Gaddy, 1934; Chabab et al., 2020). Hydrogen gas solubility in water can also be calculated using Henry’s Law as a function of the pressure’s partial pressure, $P_p$, and can be expressed as follows:

$$ C = k_H P_p $$

where $C$ is the concentration of the gas in the liquid, and $k_H$ is the Henry’s law constant, which is specific to the gas and the liquid. The value of $k_H$ for hydrogen gas in water can be obtained from experimental measurements or from thermodynamic databases such as the NIST Chemistry WebBook and the Dortmund Data Bank (Sass, 2013).

### 2.3.2. Fundamental equations

The fundamental equations required to model hydrogen flow in porous media are mass and energy conservations equations, flow equation (Darcy’s Law), and an equation of state. Two of the most widely used Equation of States for PVT behaviour in standard reservoir simulators are Peng Robinson EoS (Eq (12)) and SoaveRedlichKwong EoS. Another important Equation of State that can be implemented for UHS is GERG-2008 equation of state (EOS) named after the group that developed it European Gas Research Group. However, most of the standard reservoir simulators do not include the option to utilise GERG-2008 EOS. The decision to adopt an EOS for modelling UHS depends on the EOS that best predicts the hydrogen gas properties at the desired application conditions. The conservation of mass equation states that the rate of change of mass within a control volume is equal to the net mass flow rate into or out of the control volume and can be written as:

$$ \partial_t (\rho_S \rho_a) + \nabla \cdot (\rho_a \nabla \alpha) = Q $$

where $\rho$ is the porosity, $S_a$ is the saturation of the $a$ phase ($a$ = water, oil, gas), $\rho_a$ is the density, $q_a$ is the fluid flowrate, $Q$ is the source/sink term, and $\alpha$ is the phase (water, oil, or gas). Similar to the conservation of mass, the conservation of energy equation states that the rate of change of energy within a control volume is equal to the net energy flow rate into or out of the control volume and can be written as:

$$ \rho_a c_p \partial_t (T) + \nabla \cdot (\rho_a c_p \nabla T) = -\nabla \cdot \left( k_c \nabla T \right) + H_a $$

where $\rho$ is the density, $c_p$ is the specific heat capacity, $T$ is the temperature, $k$ is the thermal conductivity of the porous medium, $H$ is the rate of heat generation or consumption within the control volume. Darcy’s law (Eq. (32)) describes the fluid flow in porous and can be expressed in a more generic way as (Hagemann et al., 2015):

$$ v_a = \lambda_a (\nabla P - \rho_a g) $$

where $v$ is the velocity, and $\lambda$ is the mobility of phase $a$. In addition to the above fundamental equations, the sum of saturations and sum of concentrations in a phase both are equal to one. Utilising the above-mentioned equations, the compositional transport equation for a component $\gamma$ in a multiphase 3D system can be written as:

$$ \partial_t (\rho_c \gamma_a) + \nabla \cdot (\rho_c \gamma_a \nabla c) = \nabla \cdot (D_{eff} \nabla c) + q_c $$

where $c$ is the concentration of component $\gamma$. Eq. (33) is a partial differential equation (PDE) that can be solved numerically. In order to solve the transport equations, the first step is to discretise them in space and time. After discretising the transport equations in space and time, the initial and boundary conditions are set, and the equations are solved using numerical techniques such as the Gauss-Seidel method and the Jacobi method (Chen, 2007).

Dimensionless numbers are often used in reservoir simulations to characterise the behaviour of reservoirs and to facilitate the analysis of fluid flow through porous media. A group of key dimensionless numbers for reservoir modelling is defined and presented in Table 4.

Multiple software packages have been employed for simulating the hydrodynamic flow of H2 during UHS. Among these packages, DuMuX, SLB’s Eclipse, CMG-GEM, COMSOL, and TOUGH are the most frequently used (Computer Modelling Group, 2022; Schlumberger Ltd, 2014; Pruess et al., 1999; Dickinson et al., 2014; Flemisch et al., 2011). Most of these software packages employ similar principles of fluid flow through porous media. DuMuX is an open-source simulator that models multiphase multicomponent transport in porous media. It is integrated into Distributed and Unified Numerics Environment (DUNE) and solves the pressure, saturation, and flow equations using either the coupled fully implicit approach or the decoupled semi-implicit approach. Being open-source, DuMuX provides an opportunity to adjust the flow equations and couple them with various equations of state to appropriately simulate different flow applications (Flemisch et al., 2011).

SLB’s Eclipse 100 and 300 are reservoir simulation tools that are typically employed for dynamic modelling of oil and gas reservoirs. Eclipse 100 models non-compositional fluid flow in porous media using a black oil model, which simulates the flow of phases as a bulk without considering their respective components. These phases are oil phase, water phase, and gas phase (Schlumberger Ltd, 2014). SLB’s compositional simulator (Eclipse 300) and similarly CMG-GEM account for phase behaviour changes during processes where changes in fluid compositions are expected to take place. In addition to Darcy’s equations, compositional simulators use mass balance for each component and the appropriate equation of state to model multiphase fluid flow in porous media. The most widely used equation of state in petroleum simulation software is the Peng-Robinson EOS (Computer Modelling Group, 2022; Schlumberger Ltd, 2014).

TOUGH (Transport of Unsaturated Groundwater and Heat) is another simulation suite that numerically models fluid flow in porous media and can be used for gas injection and reactive transport in porous media, which is also suitable for modelling the injection of H2 and cushion gases in porous media (Pruess et al., 1999). COMSOL Multi-physics has also been utilised for modelling UHS. It is a simulation environment where multiple scientific models are present to simulate a variety of processes, such as acoustics, fluid flow, and chemical reactions (Dickinson et al., 2014). A comparison of the popular simulators used for modelling fluid flow in porous media and UHS is presented in Table 5.

### 2.3.3. Analysis of previous studies’ findings

Hagemann et al., 2015, 2016 developed a mathematical model and implemented it numerically in DuMuX to investigate the hydrodynamic effects of UHS. Their findings indicated that gravitational forces were...
Table 4
A group of key dimensionless numbers in fluid flow in porous media and reservoir engineering.

<table>
<thead>
<tr>
<th>Dimensionless Number</th>
<th>Equation</th>
<th>Components</th>
<th>Significance and Application</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capillary Number (Ca)</td>
<td>$\frac{\mu_h V}{\sigma}$</td>
<td>$\mu_h$, $V$, $\sigma$</td>
<td>The ratio of viscous forces to capillary forces in a fluid flow through porous media. It gauges the relative importance of viscous forces to capillary forces. A high Capillary Number indicates dominance of viscous forces, which is essential in enhanced recovery processes. In reservoir simulation, this number can help in understanding the displacement efficiency of hydrogen during injection and withdrawal.</td>
</tr>
<tr>
<td>Darcy Number (Da)</td>
<td>$\frac{k}{\lambda L}$</td>
<td>$k$, $\lambda L$</td>
<td>It is used in reservoir models to characterize the porous media and predict how easily fluids can traverse through it, thereby affecting storage and extraction rates of hydrogen.</td>
</tr>
<tr>
<td>Mobility Ratio (M)</td>
<td>$\frac{M_{\text{H}<em>2}}{M</em>{\text{H}<em>2} + M</em>{\text{H}_2}}$</td>
<td>$M_{\text{H}<em>2}$, $M</em>{\text{H}_2}$</td>
<td>The ratio of the mobility of the displacing fluid to the mobility of the displaced fluid and for the injection phase during UHS. It compares the mobility of hydrogen to that of water within the reservoir, which is crucial for the displacement processes.</td>
</tr>
<tr>
<td>Fractional Flow (f)</td>
<td>$f_{\text{H}<em>2} = \frac{\lambda</em>{\text{H}<em>2}}{\lambda</em>{\text{H}<em>2} + \lambda</em>{\text{H}_2}}$</td>
<td>$\lambda_{\text{H}<em>2}$, $\lambda</em>{\text{H}_2}$</td>
<td>The fraction of total flow due to one phase. It can also be written for water by replacing the hydrogen mobility in the numerator with water mobility.</td>
</tr>
<tr>
<td>Leverett J-function (J)</td>
<td>$J = \frac{k P_{\text{H}<em>2} \phi}{\mu</em>{\text{H}_2} \sqrt{\sigma \psi}}$</td>
<td>$k$, $P_{\text{H}<em>2}$, $\phi$, $\mu</em>{\text{H}_2}$, $\sigma$, $\psi$</td>
<td>This function is a dimensionless form of the capillary pressure curve. It is used in reservoir simulation to characterize capillary pressure curves which affect the distribution and movement of hydrogen and water in the reservoir.</td>
</tr>
<tr>
<td>Peclet Number (Pe)</td>
<td>$\frac{UL}{D}$</td>
<td>$U$, $L$, $D$</td>
<td>Represents the ratio of advective to diffusive transport. It is utilised to analyse heat and mass transfer effects, which can impact the behaviour of hydrogen movement and mixing within the reservoir.</td>
</tr>
<tr>
<td>Courant Number (Co)</td>
<td>$\frac{U \Delta t}{\Delta x}$</td>
<td>$U$, $\Delta t$, $\Delta x$</td>
<td>This number is used in setting time step sizes in numerical simulations to ensure stable and accurate solutions, especially in transient simulation scenarios.</td>
</tr>
</tbody>
</table>

dominant at low injection rates, resulting in uniform water displacement, while at higher injection rates, viscous forces became more dominant, leading to unstable water displacement and lateral gas fingering. Stratified aquifers were found to limit the risk of gas loss due to lateral spreading. However, this operation was affected by the velocity at which gas rises toward the cap rock seal and requires gas injection into lower structures. Furthermore, the authors coupled DuMuX with biochemical modelling to investigate the biochemical effects on UHS in a depleted gas reservoir. They observed that microbial growth and decay rates significantly influenced the process of hydrogen conversion into methane and ignoring methanogenesis led to significant differences in the results. Feldmann et al. (2016) also used numerical modelling through DuMuX to investigate hydrodynamics and gas mixing during UHS in a depleted gas reservoir. They concluded that gravity override and viscous fingering phenomena hindered the UHS process in aquifers but had a limited effect on UHS in gas-saturated reservoirs. Additionally, mechanical dispersion amplified the gas mixing process more than molecular diffusion.

Pfeiffer et al. (Sainz-García et al., 2017a; Pfeiffer and Bauer, 2015; Pfeiffer et al., 2016, 2017) utilised SLB’s compositional simulator Eclipse 300 to study the possibility of hydrogen storage in the North German Basin. They found that storage performance increased with the number of cycles, and shut-in periods during the initial filling period improved injectivity by lowering reservoir pressures during these periods. In another scenario, they used five wells for the injection and withdrawal processes, with nitrogen gas injected first to act as the cushion gas. The results confirmed that the storage performance increased with the increased number of storage cycles, and hydrogen loss due to dissolution in connate water was 1.75% of the initial hydrogen storage over a four-year period. In 2017, Pfeiffer et al. extended their efforts by utilising numerical simulation and facies modelling using ECLIPSE 300 simulator to investigate the possibility of UHS in the North German Basin. They showed that on average, UHS can sustain a continuous power output of 245 MW for one week using five storage wells. Enigbokan et al. (2021) used numerical simulation through Eclipse 300 to assess the feasibility of UHS in a depleted gas field, the Viking field in the UK. Their results showed that large-scale seasonal storage can be achieved in the studied field. However, their model did not include hydrogen loss due to solubility, geochemical reactions, or diffusion effects, and can be considered as a best-case scenario in the absence of these effects. Lysy et al. (2021) used the black oil simulator Eclipse 100 to model hydrogen gas storage in the Norne hydrocarbon field (Norway). Their modelling results showed that UHS in the gas zone was the favoured option, resulting in a hydrogen recovery of 87% compared to 77% and 49% hydrogen recovery from oil and water zones, respectively. Injection of formation gas as a cushion gas resulted in improved hydrogen recovery but also affected the purity of the produced hydrogen gas due to gas mixing.

Lubon and Tarkowski (2020) utilised PetramSim-TOUGH2 coupled with the EWSAG module to simulate seasonal hydrogen storage in a deep aquifer structure in Poland. They assumed one well positioned at the summit of the structure was used for both injection and production and that the fracturing pressure and capillary entry pressure would not be exceeded. Their modelling results indicated that water coming and excessive production are the main obstacles for UHS in the aquifer. They also found that hydrogen recovery depends on the initial injection period, where longer injection periods result in lower hydrogen recovery. For example, they found that a 36-month period will result in a 17.75% hydrogen recovery compared to 25.41% recovery in a 24-month period. Lubon and Tarkowski (2021) compared the storage capacity of carbon dioxide and hydrogen in deep aquifer anticline. Their results showed that the structure can store up to 1 million tons of carbon dioxide compared to 4000 tons of hydrogen over a 31-year period. This was due to the higher density of carbon dioxide, which allows a higher mass of carbon dioxide to be stored under the same pressure. They also found that altering the threshold capillary pressure of the cap rock has a
significant impact on the modelling results and suggested that more studies are required to fully understand this effect. Mahdi et al. (2021a) used TOUGH2 coupled with the EWSAG equation of state to evaluate UHS in a heterogeneous sandstone reservoir. Their results showed that higher injection rates lead to increased hydrogen leakage in the absence of a sealing caprock and increases hydrogen losses due to undesirable gas fingering. TOUGH was utilised by Huang et al. (2023) to understand the UHS viability in a depleted gas reservoir. Their simulation encompassed a hydrogen injection-idle-withdrawal operation in an anticline reservoir. They reported that hydrogen leakage through cap-rock and aqueous phase losses (1%) was minimal. Yet, 73% hydrogen recovery without cushion gas was achieved. They investigated the utilization of nitrogen and carbon dioxide cushion gases and reported an improved hydrogen recovery to 91% and 81%, respectively, with nitrogen exhibiting superior performance, enhancing both the hydrogen recovery factor and gas purity.

Heinemann et al. (2021b) investigated the role of cushion gas during UHS in saline aquifer anticline using the commercial compositional simulator CMG-GEM at varying geological parameters (reservoir depth, trap shape, and reservoir permeability). They concluded that cushion gas does not expand the storage capacity, but it can be used to exploit the existing capacity more efficiently. The cushion gas requirement depends on the desired amount of working gas, so studies should be conducted early in projects to determine cushion gas requirements and avoid unnecessary losses. Moreover, their simulation results showed that hydrogen storage in geological structures at greater depths with higher reservoir permeabilities requires lower amounts of cushion gas. They also concluded that tighter anticlines make the injection process more difficult without affecting the production, and more open anticlines result in increasing water production. Kanaani et al. (2022) conducted a study to investigate the impact of various types of cushion gases on the UHS in depleted oil reservoirs using the CMG-GEM simulator. The authors observed that UHS without the use of cushion gas resulted in up to 15.5% losses in the stored hydrogen. Methane was found to be the most effective cushion gas, as it resulted in higher hydrogen recovery rates compared to nitrogen and carbon dioxide. The authors attributed this to the lower molecular weight of methane, which resulted in a smaller density contrast relative to hydrogen. The simulation runs also showed that increased rates of hydrogen injection in short intervals could limit the effects of gravity segregation due to improved sweeping of reservoir fluids. The impact of operational parameters and rates on geologic
Jadhawar and Saeed, 2023b, 2023c investigated the effects of reservoir mechanisms and parameters on the performance of UHS in a deep North Sea aquifer employing CMG-GEM for numerical simulations. Their results showed that with an increased number of storage cycles, there is an upward trend in the efficiency of hydrogen recovery. Moreover, the heterogeneity in aquifer permeability was found to be a pivotal determinant, in defining the magnitude of hydrogen production. In homogeneous aquifers, the cumulative hydrogen production is quantified at 33.53 Bscf, contrasting with 30.38 Bscf in heterogeneous scenarios, thus showcasing a 7% amplified hydrogen recovery in the former case. They also reported that the incorporation of relative permeability hysteresis introduces a tangible impact, leading to diminished volumes of both injected and recovered hydrogen—specifically, a decrease of 0.5 Bscf in injected hydrogen volume and 1.24 Bscf in recoverable hydrogen volume. They also noted that the cumulative influence of hydrogen solubility and diffusion, though individually subtle, assumes greater significance when interwoven with hysteresis within the analytical framework. Saeed et al. (Saeed and Jadhawar, 2023; Saeed et al., 2023b) utilised CMG-GEM to understand the impacts of cushion gas type and physical properties on the efficiency of UHS. Their work showed that the density of utilised cushion gas plays an important role in determining the hydrogen recovery efficiency and that a lighter cushion gas was more beneficial in terms of hydrogen recovery. Thus, CH4 gas was more efficient as a cushion gas compared to N2 and CO2. They also concluded that the utilization of a cushion gas was more favourable for hydrogen recovery compared to operating without a cushion gas. Their results also determined that the purity of recovered hydrogen gas was relatively high at the start of each withdrawal phase in a storage cycle and reduced as the withdrawal continued. However, the overall purity improved with the cycles as the saturation of hydrogen gas near wellbore increased.

In another study, Cai et al. (2022) developed the simulation capabilities of PSFLOW (General Purpose Subsurface Flow Simulator) for modelling hydrogen and gas mixture storage in salt caverns, aquifers, and depleted oil and gas reservoirs. The developed simulator was benchmarked against other simulators and experimental results from published literature. The authors observed that the developed simulator was capable of modelling non-isothermal, multiphase, and multicomponent flow in H2-water, H2-CH4-water, and H2-CH4-CO2-water systems at high pressure and temperature. However, their work did not include geochemical and geomechanical effects.

Lubon and Tarkowski (Lubon and Tarkowski, 2023) conducted a study examining how the duration of the initial filling phase affects the storage of hydrogen in deep aquifers. Their results showed that increasing the length of the initial filling period contributed to greater operational capacity and better overall performance of the storage system. A similar observation was made by Abdellatif et al. (2023a), who confirmed that extending the initial filling period increased the amount of hydrogen gas that could be recovered. They also discovered that raising the rate at which hydrogen was injected actually reduced the efficiency of hydrogen recovery. Specifically, when injection rates were increased from 20 million standard cubic feet per day (MMscf/d) to 30 MMscf/d, the recovery rate dropped from 70% to 54%. This decrease in hydrogen recovery due to higher injection rates was also supported by Mahdi et al. (2021a), who identified factors like gas fingering and the displacement of existing reservoir fluids as causes for this outcome.

Several studies have evaluated the effects of injection and production rates on hydrogen recovery efficiency during underground hydrogen recovery, with a consensus that increasing hydrogen rates negatively affects hydrogen recovery efficiency (Hagemann et al., 2015, 2016; Mahdi et al., 2021a; Wang et al., 2022a, 2022b). This negative impact is attributed to the dominance of viscous forces at higher operating rates compared to lower operating rates, where gravity forces are dominant. Viscous fingering and lateral spreading, both of which result from the low viscosity of hydrogen compared to other fluids in storage, are promoted by the dominance of viscous forces. However, Kanaani et al. (2022) have shown that higher injection rates combined with shorter injection intervals lead to improved hydrogen recovery efficiency due to the minimized effects of gravity segregation resulting from improved sweeping of reservoir fluids.

The expected recovery efficiency of hydrogen during UHS reported in the various modelling studies varies significantly from approximately 30%–90%. The hydrodynamic behaviour of hydrogen and other fluids during storage is the primary factor determining hydrogen recovery efficiency, and this behaviour depends on various operational parameters, reservoir-related properties, and mechanisms, as explained and illustrated in Fig. 8.

Cushion gas plays a beneficial role in UHS and improving the hydrogen recovery efficiency compared to the absence of cushion gas (Sainz-Garcia et al., 2017a; Heinemann et al., 2021b; Kanaani et al., 2022; Abdellatif et al., 2023a). The performance of hydrogen storage depends on the type of cushion gas used, and CH4 and N2 have been found to be the most efficient due to their lower density contrast with hydrogen gas (Kanaani et al., 2022; Abdellatif et al., 2023a). This leads to decreased viscous fingering and lateral spreading, which would occur between hydrogen gas and resident reservoir fluids such as water in the absence of cushion gas. The role of cushion gas is crucial in UHS and evaluating cushion gas volume requirements is essential before undertaking any UHS project.

Among the various reservoir-related mechanisms and phenomena investigated in the literature, relative permeability hysteresis is one of the most important factors affecting the performance of an UHS process (Jadhawar and Saeed, 2023b; Wang et al., 2023a). The hysteresis of relative permeability between hydrogen gas and water in porous media indicates different behaviours of the two fluids during imbibition or drainage processes. This phenomenon results in the retention of some hydrogen gas after injection due to its inability to flow back with the rest of the gas phase during withdrawal. The altered flow behaviour disconnects some of the hydrogen gas phase within the rock, trapping it inside the reservoir. Both Delshad et al. (2022a) and Bo et al. (2023) found that including H2-water relative permeability hysteresis in the simulation improves hydrogen gas injectivity but reduces hydrogen gas recovery efficiency. The increase in injection is attributed to the improved water permeability compared to hydrogen, resulting in easier displacement of water by hydrogen. However, hysteresis leads to higher quantities of hydrogen gas becoming trapped inside the reservoir, leading to lower hydrogen production. Hydrogen gas solubility and diffusion were found to be negligible within the context of UHS (Pfeiffer et al., 2017; Delshad et al., 2022b). In conclusion, several factors influence the performance of UHS, including injection and production rates, the use of cushion gas, and relative permeability hysteresis, among others. Understanding and accounting for these factors is crucial when designing and implementing UHS projects. Moreover, further modelling and experimental studies are essential to properly understand and evaluate the effects of the various hydrodynamic parameters on UHS. A summary of the modelling studies on the hydrodynamics of UHS is presented in Table 6.

2.4. Emerging approaches

Approaches other than classical numerical simulations and geochemical modelling have been used to model various aspects of UHS. These approaches include Machine Learning (ML), Molecular Dynamics Simulations (MD), Computational Fluid Dynamics (CFD), and Pore Scale Modelling. In the following subsections, each approach will be briefly introduced and discussed with the associated published modelling studies.

2.4.1. Machine learning (ML)

ML has emerged as a vital tool in propelling the optimization and modelling capabilities within UHS systems to new horizons. Its competency in managing vast datasets and deciphering complex relationships between variables positions it as a highly effective tool for...
### Table 6
A summary of studies on hydrodynamic modelling of UHS.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Scope</th>
<th>Storage formation type</th>
<th>Approaches and Software packages</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cai et al. (2022)</td>
<td>Numerical simulation of UHS and gas mixture storage</td>
<td>Different scenarios including H2 storage in salt cavern, CH4-saturated aquifer, depleted gas field with CO2 as cushion gas</td>
<td>PSFLOW (General Purpose Subsurface Flow Simulator). Including: 1. Mass and energy conservation equations for gas flow. 2. Thermodynamics modelling.</td>
<td>Simulator was benchmarked against other simulators (TOUGH2/EWASG). Results showed that capable of modelling non-isothermal, multiphase and multicomponent flow in H2-water, H2-CH4-water, and H2-CH4-CO2-water systems at high pressure and temperature.</td>
</tr>
<tr>
<td>Enigbokan et al. (2021)</td>
<td>Numerical modelling to assess the feasibility of H2 storage in depleted gas reservoir</td>
<td>Depleted gas reservoir</td>
<td>Numerical simulation using Eclipse 300</td>
<td>They concluded that large-scale seasonal storage can be achieved in the studied field.</td>
</tr>
<tr>
<td>Heinemann et al. (2021b)</td>
<td>Numerical simulation to assess the role of cushion gas in saline aquifer anticline</td>
<td>Saline aquifer anticline</td>
<td>CMG-GEM</td>
<td>Cusion gas does not expand the storage capacity; however, it can be used to exploit the existing capacity efficiently hydrogen storage in geological structures at greater depths with higher reservoir permeabilities requires lower amounts of cushion gas UHS without the use of cushion gas results of up to 15.5% losses in the stored hydrogen Methane resulted in the higher hydrogen recovery compared to nitrogen and carbon dioxide. Increased rates of hydrogen injection in short intervals limits the effects of gravity segregation due to improved sweeping of reservoir fluids.</td>
</tr>
<tr>
<td>Kanaani et al. (2022)</td>
<td>Used numerical simulation to evaluate the effects of different types of cushion gases on the UHS in depleted oil reservoirs</td>
<td>Depleted oil reservoirs</td>
<td>CMG-GEM</td>
<td>The studied structure can store up to 1 million tons of carbon dioxide compared to 4000 tons hydrogen over a 21-year period. Altering the threshold capillary pressure of the cap rock greatly influences the modelling results.</td>
</tr>
<tr>
<td>Luboi and Tarkowski (2021)</td>
<td>Numerical simulation to compare between the storage capacity of carbon dioxide and hydrogen in deep aquifer anticline</td>
<td>Deep aquifer anticline</td>
<td>PetraSim TOUGH2</td>
<td>Higher injection rates lead to increased hydrogen leakage in the absence of a sealing caprock and increased hydrogen losses due to undesirable gas fingering.</td>
</tr>
<tr>
<td>Mahdi et al. (2021a)</td>
<td>Numerical simulation of UHS in heterogeneous sandstone reservoir</td>
<td>Heterogenous sandstone reservoir</td>
<td>TOUGH2 coupled with EWASG</td>
<td>Storage performance increases with the number of cycles and that shut-in periods during the initial filling period on average UHS can sustain a continuous power output of 245 MW for 1 week using five storage wells. Dimensionless groups including aspect ratio, gravity/turbulence ratio and gravity/ capillary ratio can be used to correlate the hydrogen and carbon dioxide flow between different scales. Heterogenous reservoirs may not be optimal for UHS due to the drop in the purity of the produced hydrogen, especially, at higher injection and production rates.</td>
</tr>
<tr>
<td>Pfeiffer and Bauer (2015)</td>
<td>Numerical simulation of UHS to investigate injection and production aspects</td>
<td>Underground natural gas structure</td>
<td>Eclipse 300</td>
<td>Due to CO2 solubility in brine, the volume of injected carbon dioxide needs to be increased by approximately 30% to result in the same displacement effect. Including carbon dioxide solubility in brine in the model led to an increase in the estimated hydrogen purity.</td>
</tr>
<tr>
<td>Pfeiffer et al. (2017)</td>
<td>Numerical simulation to evaluate feasibility of UHS application</td>
<td>Existing anticlinal structure in the North German Basin</td>
<td>Eclipse 300</td>
<td>Water coning and excessive production is the main hurdle for UHS in the aquifer hydrogen recovery is dependent on the initial injection period, where a longer initial period results in less hydrogen recovery.</td>
</tr>
<tr>
<td>Wang et al. (2022a)</td>
<td>Numerical simulation to investigate the applicability of the scaling theory to hydrogen and carbon dioxide flow inside a hypothetical reservoir.</td>
<td>Hypothetical reservoir</td>
<td>CMG-GEM</td>
<td>UHS in the gas zone was the favoured option which resulted in a hydrogen recovery of 87% compared to 77% and 49% hydrogen recovery from oil and water zones, respectively. A maximum of 75% hydrogen could be recovered by the end of the third cycle the studied structure is geomechanically safe.</td>
</tr>
<tr>
<td>Wang et al. (2022b)</td>
<td>Numerical simulation to investigate the hydrogen and carbon dioxide flow inside a hypothetical reservoir with the effect of carbon dioxide solubility in water on the process</td>
<td>Hypothetical reservoir</td>
<td>CMG-GEM</td>
<td>Water coning and excessive production is the main hurdle for UHS in the aquifer hydrogen recovery is dependent on the initial injection period, where a longer initial period results in less hydrogen recovery.</td>
</tr>
<tr>
<td>Luboi and Tarkowski (2020)</td>
<td>Numerical simulation of UHS to investigate injection and production aspects</td>
<td>Deep aquifer structure in Poland</td>
<td>PetraSim coupled with EWASG module</td>
<td>Water coning and excessive production is the main hurdle for UHS in the aquifer hydrogen recovery is dependent on the initial injection period, where a longer initial period results in less hydrogen recovery.</td>
</tr>
<tr>
<td>Lysyy et al. (2021)</td>
<td>Understand the effects of storing hydrogen in three different zones, gas, oil and water</td>
<td>Norne hydrocarbon field (Norway)</td>
<td>Eclipse 100</td>
<td>UHS in the gas zone was the favoured option which resulted in a hydrogen recovery of 87% compared to 77% and 49% hydrogen recovery from oil and water zones, respectively. A maximum of 75% hydrogen could be recovered by the end of the third cycle the studied structure is geomechanically safe.</td>
</tr>
<tr>
<td>Bai and Tahmasebi (2022)</td>
<td>Investigate the hydro-mechanical aspects of UHS</td>
<td>Saline aquifer at the Powder River Basin of Wyoming State</td>
<td>3D coupled hydro-mechanical model Peng-Robinson equation of state</td>
<td>Water coning and excessive production is the main hurdle for UHS in the aquifer hydrogen recovery is dependent on the initial injection period, where a longer initial period results in less hydrogen recovery.</td>
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</table>

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<th>Approaches and Software packages</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ebigbo et al. (2013)</td>
<td>Investigated the effects of bioactivity on UHS</td>
<td>N/a</td>
<td>Mathematical and numerical modelling</td>
<td>the possibility of failure decreases during the withdrawal period</td>
</tr>
<tr>
<td>Pfeiffer et al. (2016)</td>
<td>Numerical simulation of UHS in an anticline structure</td>
<td>Aquifer</td>
<td>Eclipse 300</td>
<td>Possible competition between three different microbial species: methanogens, acetogens, and acetotrophs. Storage performance increased with the increased number of storage cycles. Hydrogen loss due to dissolution in connate water was 1.75% of the initial hydrogen storage over a 4-year period. Low injection rates result in gravitational forces becoming dominant leading to a uniform water displacement. UHS in stratified aquifers could limit the risk of losing gas due to lateral spreading. Gravity override is more pronounced in aquifers than in gas reservoirs. Mechanical dispersion amplifies the gas mixing process and is more pronounced than molecular diffusion. The number of microorganisms present in the reservoir greatly influence the process of hydrogen conversion into methane. Cushion gas requirements in aquifer is higher than the oil reservoir. The presence of high permeability channel in the oil reservoir requires additional wells to contain hydrogen spreading. Results were sensitive to water/H₂ relative permeability curves used in the aquifer case. Hysteresis consideration resulted in higher injectivity. Molecular diffusion and solubility had minimal impact on hydrogen saturation. Lighter cushion gases results in higher hydrogen production. Hydrogen diffusion is negligible. Hydrogen stored in unconventional gas reservoirs maintains higher purity compared to storage in conventional gas reservoirs due to differential adsorption effect of nanopores. Most of the hydrogen is concentrated in the hydraulically fractured region of reservoir. H₂ loss due to diffusion is minimal (&lt;1%). Hydrogen recovery is significantly improved with the use of N₂ or CO₂ as cushion gases.</td>
</tr>
<tr>
<td>Hagemann et al. (2016)</td>
<td>Investigate biochemical and hydrodynamic behaviour during UHS.</td>
<td>Depleted gas reservoir</td>
<td>Numerical modelling through DuMuX coupled with biochemical modelling</td>
<td></td>
</tr>
<tr>
<td>Delshad et al. (2022b)</td>
<td>Investigated operational parameters effect on UHS</td>
<td>Depleted oil reservoir and saline aquifer</td>
<td>Numerical modelling using CMG-GEM</td>
<td>The presence of high permeability channel in the oil reservoir requires additional wells to contain hydrogen spreading. Results were sensitive to water/H₂ relative permeability curves used in the aquifer case. Hysteresis consideration resulted in higher injectivity. Molecular diffusion and solubility had minimal impact on hydrogen saturation. Lighter cushion gases results in higher hydrogen production. Hydrogen diffusion is negligible. Hydrogen stored in unconventional gas reservoirs maintains higher purity compared to storage in conventional gas reservoirs due to differential adsorption effect of nanopores. Most of the hydrogen is concentrated in the hydraulically fractured region of reservoir. H₂ loss due to diffusion is minimal (&lt;1%). Hydrogen recovery is significantly improved with the use of N₂ or CO₂ as cushion gases.</td>
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<td>Delshad et al. (2022a)</td>
<td>Investigated operational parameters effect on UHS</td>
<td>Depleted oil and gas reservoirs and saline aquifer</td>
<td>Numerical modelling using CMG-GEM</td>
<td></td>
</tr>
<tr>
<td>Abdellatif et al. (2023a)</td>
<td>Numerical simulation of hydrodynamics during UHS</td>
<td>Depleted gas field</td>
<td>CMG Simulation suite</td>
<td></td>
</tr>
<tr>
<td>Wang et al. (2023b)</td>
<td>Multi-scale simulation of UHS</td>
<td>Depleted unconventional gas reservoirs</td>
<td>Monte Carlo Simulation, Numerical modelling using In-house reservoir simulator MSCO2</td>
<td></td>
</tr>
<tr>
<td>Huang et al. (2023)</td>
<td>Assess the viability of UHS</td>
<td>Depleted gas reservoir</td>
<td>TOUGH</td>
<td></td>
</tr>
<tr>
<td>Jadhawar and Saeed (2023b)</td>
<td>Evaluated the effects of reservoir parameters and mechanisms on UHS</td>
<td>Deep aquifer</td>
<td>Numerical modelling using CMG-GEM</td>
<td>Heterogeneity is an important factor in determining the performance of UHS. Relative permeability hysteresis omission in simulation may result in overestimating the recoverable hydrogen gas.</td>
</tr>
<tr>
<td>Luboni and Tarkowski (2023)</td>
<td>Evaluated the impact of the initial filling period on the performance of UHS.</td>
<td>Deep aquifer</td>
<td>PetraSim and TOUGH2</td>
<td>Extended initial filling period resulted in improved operational storage capacity and overall performance.</td>
</tr>
<tr>
<td>Jadhawar and Saeed (2023c)</td>
<td>Evaluated the impact of fluid-fluid and fluid-rock interactions during UHS.</td>
<td>Deep aquifer</td>
<td>CMG-GEM</td>
<td>Hydrogen aqueous diffusion and solubility effects are negligible. The hydrogen recovery efficiency decreases with the increase in operational rates, longer and less frequent storage cycles, presence of shut-in periods between injection and production, and lower number of operational wells.</td>
</tr>
<tr>
<td>Jadhawar and Saeed (2023a)</td>
<td>The effect of operational parameters on UHS</td>
<td>Deep aquifer</td>
<td>CMG-GEM</td>
<td>Hydrogen recovery efficiency was directly related to cushion gas density such that lighter gases result in higher hydrogen recovery. CH₄ resulted in higher hydrogen retrieval compared to N₂ and CO₂. Results showed that 28,282 onshore salt caverns can be developed in Australia with overall capacity of 14,697 PJ of hydrogen energy.</td>
</tr>
<tr>
<td>Saeed and Jadhawar (2023); Saeed et al. (2023b)</td>
<td>Studied the impact of cushion gas type on UHS.</td>
<td>Aquifer</td>
<td>CMG-GEM</td>
<td></td>
</tr>
<tr>
<td>Aftab et al. (2023)</td>
<td>Assessment of hydrogen storage capacity and viability</td>
<td>Salt caverns</td>
<td>AusH₂</td>
<td></td>
</tr>
</tbody>
</table>
modelling the intricate and interconnected processes intrinsic to UHS. A notable application of ML algorithms is showcased in the work of Zhu et al. (2022), who employed Wavelet Neural Networks (WNN) to model hydrogen solubility in water under a variety of conditions, exemplifying the adeptness of ML in accurately predicting complex behaviours in UHS systems. Authors showed that modelling results predicted experimental values with an R-squared value of 0.9999. Their results showed that pressure is the most impactful parameter on the hydrogen’s solubility in brine followed by temperature and NaCl salinity. Ansari et al. (2022) also utilised ML models to predict hydrogen solubility in water. They found that a Radial Basis Function model (RBF) optimised using Cultural Algorithm yielded the most accurate prediction of hydrogen solubility in water under varying conditions. They also concluded that pressure was the most impactful factor on hydrogen solubility which is similar to the observation of Zhu et al. (2022).

ML has been used to model other fluid properties such as H$_2$-water IFT through different approaches. Wei Ng et al. (2022) reported that a Multilayer Preceptor model optimised with Levenberg-Marquardt succeeded in predicting H$_2$-water IFT with an R2 value of 0.9997 in a wide range of salinity (0–4.95 mol/kg), pressure (0.5–4.52 MPa) and temperature (298.03–448.35 K). Ghadamisi et al. (2023) showed that a Gaussian Process Regression model outperformed other evaluated models in predicting the IFT property in a H$_2$-water system with a mean average error of 0.022.

Moreover, ML stands as a robust mechanism for parameter optimization, which is a critical facet for bolstering the safety and efficiency of UHS systems. By scrutinizing historical and real-time data, ML can assist in pinpointing the optimal pressure and temperature conditions that augment hydrogen storage capacity while mitigating the risk of leakage, translating to more efficient operations and potentially diminished operational costs.

One of the significant merits of ML lies in its capability to model and integrate various intricate and interconnected processes like hydrodynamic, fluid behaviour, geochemical, and geomechanical processes, which are cardinal in comprehending the behaviour of hydrogen within underground formations. For instance, in hydrodynamic processes, ML can be employed to model the distribution and movement of hydrogen within underground formations, aiding in predicting how hydrogen will flow under various pressure, temperature, and wettability conditions, which is crucial for the design and operation of UHS systems that aim for efficient storage and retrieval of hydrogen. In this context, a study by Vo Thanh et al. (2023), the use of machine learning algorithms, specifically XGBoost, RF, LGBR, and Adaboost_DT, was investigated to predict hydrogen wettability in underground storage sites using 513 data points. The study found that the XGBoost algorithm was exceptionally proficient, achieving an R2 value of 0.941 and RMSE and MAE values of 4.455 and 2.861 respectively. Substrate types were identified as the most impactful variables influencing the predictions. Furthermore, the study successfully predicted the hydrogen column height in a specific basalt formation in Australia, showcasing the practical applicability and efficiency of using machine learning in predicting hydrogen behaviour and wettability in underground storage.

In tandem, the fluid behaviour of hydrogen within these formations is a complex facet significantly influenced by the geomechanical and geochemical conditions of the storage site. ML can help model and predict this behaviour, thereby aiding in the design of storage caverns and the optimization of storage and retrieval processes. Moreover, ML’s utility extends to modelling geochemical interactions between hydrogen and the surrounding rock formations. By analysing data on rock and hydrogen interactions over time, ML algorithms can aid in forecasting potential alterations in rock chemistry, which is imperative for ensuring the long-term integrity and safety of UHS systems.

Simultaneously, understanding the geomechanical behaviour of rock formations under the pressure and temperature conditions associated with hydrogen storage is quintessential for ensuring the structural integrity of the storage caverns. ML can be employed to model and analyse geomechanical data, aiding in the prediction of rock deformation and stress distributions within the underground formations, which are critical variables for the safe and efficient operation of UHS systems. These comprehensive insights, facilitated by ML, are instrumental in advancing the design, operation, and maintenance of UHS systems, ultimately contributing to the evolution of safer and more efficient UHS solutions.

2.4.2. Molecular dynamic simulations (MD)

MD simulations provide a detailed molecular-level analysis, which is indispensable for understanding the interaction between hydrogen and various materials in UHS systems. For instance, Ghasemi et al. (2022) used molecular dynamic simulations to evaluate hydrogen diffusion in three types of clay minerals (pyrophyllite, montmorillonite, and beidellite) under varying clay charging behaviours. Their simulations were performed using GROMACS. The modelling results showed that hydrogen diffusion in clay minerals is significantly lower than that in bulk water due to the confined environment of clays. Moreover, in negatively charged clays, the increase in pore size up to 2 nm leads to an increase in the hydrogen diffusion coefficient. However, above 2 nm, the hydrogen diffusion coefficient remains constant. They also concluded that the charging behaviour of clay minerals and interlayer cations affects the hydrogen diffusion coefficient. Where the higher polarizability of the O-sheet leads to the attraction of the water molecules and consequently an increase in the diffusion coefficient. Their findings suggest that the reservoir rock and caprock mineralogy need to be properly assessed and evaluated to understand the potential hydrogen diffusion during UHS. Moreover, MD simulations can model the solubility of hydrogen within underground storage, providing insights into how temperature, pressure, and material properties affect hydrogen behaviour on a molecular level. Zhang et al. (2023) utilised a coupled MD-ML approach to evaluate the solubility of hydrogen in brine under varying salinities, pressures and temperatures. Their work indicated that their approach was successful in reproducing the experimental results. Moreover, they found that temperature has a non-linear effect on hydrogen solubility in water.

MD simulations can be utilised to study the interaction between hydrogen molecules and other substances like water or impurities within the storage reservoir, which is crucial for understanding how these interactions may affect hydrogen storage and retrieval. Furthermore, MD simulations have the capability to model the behaviour of sealing materials under different temperatures and pressures, aiding in the selection of materials that can maintain integrity under UHS operational conditions. This molecular-level understanding is essential for designing UHS systems that can safely and efficiently store hydrogen underground.

2.4.3. Computational fluid dynamics (CFD)

CFD models utilise numerical methods to analyse and solve problems associated with fluid flows, allowing for the simulation of fluid dynamics, heat transfer, and chemical reactions within various systems. Sáinz-García et al. (Sáinz-García et al., 2017a; Sáinz-García, 2017) conducted simulations using COMSOL Multiphysics to study the immiscible multiphase flow of water and a methane-hydrogen gas mixture during methane-hydrogen underground storage in the Lower Triassic of the Paris Basin. Their research indicated that both gas and aquifer properties significantly influence the storage process. Moreover, they developed a 3D multiphase numerical model to explore different extraction well configurations, demonstrating that a maximum hydrogen recovery of 78% could be achieved in the studied UHS case. However, they also noted that hydrogen up-coning could pose a substantial challenge for UHS in saline aquifers without the use of cushion gas. By employing tools like COMSOL for CFD simulations, gain crucial insights can be gained into complex multiscale phenomena and optimizing the design and operation of UHS facilities.
2.4.4. Pore network modelling

Pore Network Modelling is a simulation technique that abstracts the intricate structure of porous materials into a simplified network of interconnected void spaces, known as pores, and the constrictions between them, known as throats. This method facilitates the study of fluid flow and transport phenomena within the porous media by capturing essential features such as pore size, shape, and connectivity.

In 2021, Hashemi et al. (2021a) conducted a comprehensive study to understand hydrogen transport characteristics in brine-saturated deep porous rocks for UHS. They utilised pore-scale modelling and performed a sensitivity analysis to quantify the effects of uncertain fluid and rock properties on reservoir-scale functions, such as relative permeability and capillary pressure. The study also explored cyclic hysteretic multiphase flow, relevant to hydrogen-brine energy storage projects. Their results showed that capillary pressure and relative permeability are sensitive to contact angles. Moreover, clay content had a pronounced effect on the endpoint values for relative permeability curves for drainage and imbibition cycles. Wang et al. (2023c) investigated hydrogen transport in sandstone porous media under different wetting conditions using direct numerical simulation. The study found that increased hydrogen wetting reduced the snap-off effect during the primary drainage process, thereby facilitating greater hydrogen storage. However, during the primary imbibition process, increased hydrogen wetting hindered the extraction process, resulting in a recovery factor below 20%.

Bagheri et al. (2023) conducted a pore-scale investigation of hydrogen-water flow dynamics in aquifers using computational fluid dynamics at elevated pressure. The research identified that the optimal rates for hydrogen injection and production differed, and a medium flow rate could minimize both capillary and viscous fingering mechanisms, thereby enhancing storability and recovery factor. The study also noted that neglecting the local compression and expansion of hydrogen could lead to misinterpretations of flow regimes and hydrogen storability. Zhao et al. (2024) used a three-dimensional pore network model to simulate hydrogen-brine two-phase flow in various porous media, such as sandstone, carbonate, and sand packs. The study suggested that a larger contact angle with low water affinity was preferable for UHS due to its low hydrogen trapping rate. Machine learning methods, including least square fitting and Support Vector Machine (SVM), were employed to classify rocks based on their hydrogen trapping rates, providing insights for site selection in UHS projects.

These studies highlight the importance of understanding hydrogen transport and trapping mechanisms in porous media for effective UHS. Both Wang et al. (2023c) and Bagheri et al. (2023) discuss the influence of wetting conditions and flow rates on hydrogen storage and extraction, while Hashemi et al. (2021a) and Zhao et al. (2024) emphasize the need for a systematic understanding of fluid and rock properties on UHS. The integration of pore scale modelling and machine learning methods by Zhao et al. (2024) offers a new approach to predicting hydrogen trapping rates, supplementing traditional pore network modelling techniques. Such integrated approaches contribute to a comprehensive understanding of factors influencing hydrogen storage in porous media and provide valuable insights for UHS site selection and design.

2.4.5. Other approaches

Hassanpouryouzband et al. (2020) modelled thermodynamic and transport properties of hydrogen containing mixtures. They used GERG-2008 Equation of State (EoS) and SuperTRAPP model to predict the thermodynamic and transport properties of hydrogen mixed with methane, carbon dioxide, nitrogen and natural gas (with a typical North Sea natural gas composition). Their modelling results were validated against experimental data from the literature and the model was found to provide accurate predictions at pressures within the range 0.01–100 MPa, temperatures in the range 200–500 K and hydrogen mole fraction in the range 10–90 Mole%. They developed a publicly accessible software (H2Themobank) to calculate hydrogen gas mixture properties such as gas density, thermal capacity, viscosity, entropy among other properties at varying mole fractions, temperatures and pressures. We believe that their valuable effort will be of great benefit in advancing the modelling of UHS and its associated processes. Researchers in HyStorPor project developed an open-access tool to assess the hydrogen storage capacities of both surface and subsurface facilities and formations in varying conditions (Hassanpouryouzband et al., 2021).

Maraggi et al. (Ruiz Maraggi and Moscardelli, 2023) addressed the lack of open-source tools for assessing H2 storage feasibility in caverns by developing the GeoH2 Salt Storage and Cycling App. This web-based thermodynamic simulator comprises modules for H2 physical properties, volumetric capacity, production, injection, and cycling. Validation of physical and volumetric modules with empirical data and comparison of production and injection modules with an open-source simulenhance the tool’s credibility. This App offers a means to assess technical aspects of H2 storage, injection, withdrawal, and cycling in salt caverns. A summary of various studies that utilised modelling approaches other than the geochemical, geomechanical, and hydrodynamic approaches are shown in Table 7.

3. Current status of modelling studies

In reviewing the current landscape of UHS modelling studies as shown in Fig. 12a, it is evident that research efforts have been unevenly distributed across different aspects of the discipline. The analysis of recent modelling studies indicates a pronounced emphasis on hydrodynamic aspects, as they receive the most attention. This focus points to the critical importance placed on understanding the subsurface flow and distribution of hydrogen, a fundamental component in assessing the viability and safety of UHS.

Conversely, geochemical considerations, have been less explored in modelling studies compared to hydrodynamics. These studies are essential for comprehending the chemical interactions between stored hydrogen and the geological media, which have implications for both the integrity of the storage formation and the quality of the stored hydrogen. Geomechanical factors, which assess the mechanical impacts of hydrogen storage on the surrounding geology, have been the subject of relatively few studies, suggesting a potential gap in current research efforts.

The selection of tools for modelling these storage systems (see Fig. 12b) reveals a preference for the Computer Modelling Group’s (CMG) suite of simulation software, which has been the most widely adopted among the tools assessed. Its extensive use implies that CMG’s capabilities are well-aligned with the current focus of the field. Furthermore, the application of Machine Learning (ML) approaches in a substantial number of studies reflects an innovative trend in the field, harnessing the power of data-driven techniques to enhance model accuracy and efficiency. Other tools, including PHREEQC for geochemical calculations and the various other software packages represented in the present modelling studies, show a diversity in the methodologies being employed in current research. This indicates a field that values and requires the application of multiple approaches to understand the multi-faceted aspects encountered during UHS.

4. Challenges and future research

The present investigation into UHS modelling has revealed several issues and challenges that remain unresolved and are summarised below:

- One of the significant challenges in UHS modelling is the pronounced lack of expansive and diverse datasets. Currently, the reliance on limited field trials and laboratory studies for model validation fails to provide the scope and specificity needed for confident application in diverse geological settings. This insufficiency leads to a significant gap between model predictions and their practical reliability. Therefore, a concerted effort is necessary to compile comprehensive
Table 7

A summary of studies that utilised alternative modelling of UHS.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Scope</th>
<th>Storage formation type/ Conditions</th>
<th>Approaches and Software packages</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gajda and Latyniak (2022)</td>
<td>Modelling permeability and hydrogen loss through Polymer Sealing Liners during UHS</td>
<td>Lined Rock Cavern (LRC)</td>
<td>–</td>
<td>Model was successful in modelling permeability and hydrogen loss within studied conditions</td>
</tr>
<tr>
<td>Zhu et al. (2022)</td>
<td>Predict hydrogen solubility in water as a function of pressure, temperature and NaCl salinity</td>
<td>Underground sodium chloride solutions</td>
<td>ML</td>
<td>Used WNN to model solubility. Pressure is the most impactful parameter on the hydrogen’s solubility in brine followed by temperature and NaCl salinity. Hydrogen diffusion in clay minerals is significantly lower than that in bulk water due to the confined environment of clays reservoir rock and caprock mineralogy needs to be properly assessed and evaluated to understand the potential hydrogen diffusion during UHS.</td>
</tr>
<tr>
<td>Ghasemi et al. (2022)</td>
<td>Evaluate hydrogen diffusion</td>
<td>Three types of clay minerals (pyrophyllite, montmorillonite, and beidellite)</td>
<td>MD (GROMACS)</td>
<td>–</td>
</tr>
<tr>
<td>Hassanpouryouzband et al. (2020)</td>
<td>Modelling thermodynamic and transport properties of hydrogen containing mixtures</td>
<td>–</td>
<td>GERG-2008 Equation of State (EoS) and supertrapp model (H2Themonbank)</td>
<td>Modelling results were validated against experimental data from the literature.</td>
</tr>
<tr>
<td>Ruiz Maraggi and Moscardelli (2023)</td>
<td>Modelling of UHS in salt caverns</td>
<td>Salt caverns</td>
<td>Thermodynamical simulator (GeoH2 Salt Storage and Cycling App)</td>
<td>The developed tool used empirical data for validation. Serves as a means for evaluating the physical attributes of hydrogen during UHS, injection, and withdrawal. Optimization of UHS, carbon dioxide sequestration and net present value as objective functions show conflict between the three functions.</td>
</tr>
<tr>
<td>Kanaani et al. (2023)</td>
<td>Underground hydrogen gas storage and carbon dioxide storage optimization</td>
<td>Synthetic reservoir model</td>
<td>ML</td>
<td>–</td>
</tr>
<tr>
<td>Amari et al. (2022)</td>
<td>Modelling hydrogen solubility in water</td>
<td>Varying salinity, pressure and temperature conditions</td>
<td>ML</td>
<td>Radial Basis Function model (RBF) optimised using Cultural Algorithm yielded the most accurate prediction of hydrogen solubility in water. Pressure was the most impactful factor on hydrogen solubility.</td>
</tr>
<tr>
<td>Ng et al. (2022)</td>
<td>Modelled H2-water IFT</td>
<td>Salinity (0.4-9.5 mol/kg), pressure (0.5-45.2 MPa) and temperature (298.03-448.35 K)</td>
<td>ML</td>
<td>Multilayer Preceptor model optimised with Levenberg-Marquardt succeeded in predicting H2-water IFT with an R^2 value of 0.9997.</td>
</tr>
<tr>
<td>Gbadamosi et al. (2023)</td>
<td>Modelled H2-water IFT</td>
<td>Pressure (2.76–34.47 MPa) and temperature (298.15–423.15 K)</td>
<td>ML</td>
<td>Utilised a Gaussian Process Regression model to predict IFT with a mean average error of 0.022.</td>
</tr>
<tr>
<td>Vo Thanh et al. (2023)</td>
<td>Modelled hydrogen wettability in UHS sites</td>
<td>Various sites</td>
<td>ML</td>
<td>Xgboost model successfully predicted the hydrogen column height in a specific basalt formation in Australia. Substrate types were identified as the most impactful variables influencing the predictions.</td>
</tr>
<tr>
<td>Zhang et al. (2023)</td>
<td>Evaluated solubility of hydrogen in brine</td>
<td>Varying salinities, pressures and temperatures</td>
<td>MD-ML</td>
<td>Their approach was successful in reproducing the experimental results. Temperature has a non-linear effect on hydrogen solubility in water. Maximum hydrogen recovery of 74% could be achieved in the studied UHS case. Hydrogen up-coning could pose a substantial challenge for UHS in saline aquifers without the use of cushion gas.</td>
</tr>
<tr>
<td>Sainz-García et al., 2017a; Sainz-García, 2017</td>
<td>Studied immiscible multiphase flow of water and a methane-hydrogen gas mixture</td>
<td>Aquifer in Lower Triassic of the Paris Basin</td>
<td>CFD (COMSOL)</td>
<td>–</td>
</tr>
<tr>
<td>Hashemi et al. (2021a)</td>
<td>Evaluated impacts of uncertain rock-fluid properties</td>
<td>Deep aquifer</td>
<td>Pore Network Model</td>
<td>Contact angle (wettability) effect on relative permeability was observed. Clay content affected the relative permeability curves.</td>
</tr>
<tr>
<td>Wang et al. (2023c)</td>
<td>Hydrogen transport in different wetting conditions</td>
<td>Aquifer</td>
<td>Pore network model</td>
<td>Higher hydrogen wetting reduced the snap-off effect during the primary drainage process and increased storage capacity but decreased the recovery efficiency. Medium flow rate could minimize both capillary and viscous fingering mechanisms. Important to consider local hydrogen compressibility to properly identify flow regime.</td>
</tr>
<tr>
<td>Bagheri et al. (2023)</td>
<td>Hydrogen flow behaviour</td>
<td>Aquifer</td>
<td>Pore network model, CFD</td>
<td>–</td>
</tr>
<tr>
<td>Zhao et al. (2024)</td>
<td>Hydrogen-brine two-phase flow in various porous media</td>
<td>Sandstone, carbonate, and sand pack</td>
<td>Pore network model, ML</td>
<td>Larger contact angle with low water affinity was preferable for UHS due to its low hydrogen trapping rate.</td>
</tr>
<tr>
<td>Hassanpouryouzband et al. (2021)</td>
<td>Hydrogen storage capacity</td>
<td>Surface facilities and subsurface formations</td>
<td>Nobel-Abel Equation of State, C# code in visual studio (H2capES)</td>
<td>This open-source tool allows for quick calculation of hydrogen storage capacity in both surface and subsurface facilities and pore pack.</td>
</tr>
</tbody>
</table>
The accurate representation of UHS systems through modelling necessitates intensive computational resources, particularly when integrating diverse aspects such as hydrodynamics, geochemistry, and geomechanics. High-performance computing (HPC) platforms become a prerequisite as the models strive to capture an array of complex interactions over disparate scales, from the molecular level to the full extent of the storage reservoir. Moreover, there is a pressing need for the development of novel computational strategies, potentially through the application of parallel computing, machine learning and artificial intelligence, to enhance the efficiency of simulations and make the most effective use of available computational resources.

- A notable gap in the current body of research within UHS is the lack of modelling studies that address the geological uncertainties inherent in subsurface environments. Addressing the underexplored area of geological uncertainty is paramount for enhancing UHS modelling. The complexity of subsurface geology, with its unpredictable facies and fault structures, introduces significant variability in predicting porosity, permeability, and containment integrity. Although advanced geostatistical and geophysical methods exist within oil reservoir simulation, their application to UHS remains sparse. Adapting these established methodologies could provide a shortcut to more accurate geological models for UHS. Emphasizing this research gap is essential, as it underpins the accurate assessment of storage capacities and operational safety. By drawing on oil and gas industry experiences, we can potentially fast-track the development of UHS models that effectively navigate geological uncertainties.

This review focused on the technical modelling of UHS; however, another important aspect of modelling UHS is the economic modelling of the process. This type of modelling should involve the full value chain of UHS to allow for a proper comparison between UHS and alternative large scale energy storage systems. There exist some economic studies on this topic, however, they are beyond the scope of this study.

5. Conclusion

The present study conducted an extensive investigation into the current approaches and studies on UHS modelling. The following conclusions summarise the most important findings of this review:

- Modelling UHS is a complex process that requires an understanding and integration of various multi-scale phenomena and aspects, including hydrodynamics, geochemistry, and geomechanics.
- Geochemical studies on UHS predominantly utilised PHREEQC or Geochemists Workbench to describe geochemical reactions. The amount of hydrogen gas loss as a result of abiotic and biotic reactions can range from negligible losses up to more than 80%, depending on the adopted geochemical reactions and kinetic reaction rates, simulated conditions, rock mineralogy, brine salinity, and most importantly, the adoption of hydrogen redox reactions.
- The 3D coupled hydro-mechanical models revealed that rock failure during the withdrawal period in UHS in aquifers decreases compared to the injection period due to a decrease in pore pressure and an increase in effective stress. Additionally, the geomechanical changes during UHS in salt caverns depend on the number of storage cycles, temperature, presence of impurities in the salt cavern, as well as injection and production rates.
- The most widely used software programs for modelling hydrogen hydrodynamics are CMG’s simulation suite, TOUGH, DuMaS, and Eclipse. These simulators use mathematical and numerical models that adopt fundamental fluid flow in porous media equations such as conservation of mass, Darcy’s law, Fick’s law for diffusion modelling, and Peng-Robinson EOS to model hydrogen gas properties. Hydrodynamics simulation studies revealed that the shape and rock properties of the underground storage location play a critical role in determining the hydrogen recovery efficiency. Higher injection and production rates result in viscous forces becoming more dominant
than gravity forces, leading to higher hydrogen gas losses. The utilization of cushion gas was also shown to be preferred, as it improves hydrogen recovery efficiency. It is worth noting that the current simulation modelling approaches are adapted from oil and gas reservoir simulation approaches, as well as underground gas storage, such as natural gas and carbon dioxide. The emerging approaches in modelling UHS systems extend beyond classical numerical simulations and geochemical modelling, encompassing Machine Learning (ML), Molecular Dynamics Simulations (MD), Computational Fluid Dynamics (CFD), Pore Scale Modelling, and other novel techniques. ML has proven invaluable for deciphering complex relationships and optimizing parameters in UHS systems, while MD simulations provide a molecular-level understanding of hydrogen interactions crucial for system design. CFD models offer insights into fluid dynamics and heat transfer, enhancing the optimization of UHS facilities. Pore Network Modelling, on the other hand, abstracts the porous structure of materials to study fluid flow, which is crucial for understanding hydrogen transport in porous rocks.

- Alternative approaches such as the development of open-source tools and software for assessing thermodynamic and transport properties of hydrogen mixtures and storage capacities significantly contribute to the progression and optimization of UHS systems. Collectively, these approaches and tools not only advance the understanding of hydrogen behaviour and interactions in subsurface environments but also play a pivotal role in realizing efficient and safe UHS solutions.

The present study provides crucial insights into the complexities and challenges involved in modelling UHS. The findings of this study can guide future research efforts to improve the accuracy and reliability of the modelling approaches and studies, which is critical for the efficient and safe implementation of UHS as a clean energy solution.

CRediT authorship contribution statement

Motaz Saeed: Conceptualization, Data curation, Formal analysis, Methodology, Visualization, Writing - original draft, Writing - review & editing. Prashant Jadhawar: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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