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Hydrogen underground storage efficiency in a heterogeneous sandstone reservoir

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Abstract:
Underground hydrogen storage has been recognized as a key technology for storing enormous amounts of hydrogen, thus aiding in the industrial-scale application of a hydrogen economy. However, underground hydrogen storage is only poorly understood, which leads to high project risk. This research thus examined the effect of caprock availability and hydrogen injection rate on hydrogen recovery factor and hydrogen leakage rate to address some fundamental questions related to underground hydrogen storage. A three dimensional heterogeneous reservoir model was developed, and the impact of caprock and hydrogen injected rate on hydrogen underground storage efficiency were analysed with the model. The results indicate that both caprock and injection rate have an important impact on hydrogen leakage, and the quantities of trapped and recovered hydrogen. It is concluded that higher injection rate increases H_2 leakage when caprocks are absent. In addition, lower injection rates and caprock availability increases the amount of recovered hydrogen. This work therefore provided fundamental information regarding underground hydrogen storage project assessment, and supports the decarbonisation of the energy supply chain.

1. Introduction

Hydrogen is a clean energy carrier which is produced from renewable sources (Lord et al., 2014; Hanely et al., 2018; Acar and Dincer, 2019). Hydrogen thus has the potential to drastically reduce anthropogenic greenhouse gas emissions which are mainly emitted by burning traditional fossil fuels such as coal, natural gas or oil (IEA, 2020)-and to therefore significantly mitigate climate change (Yartys and Lototsky, 2004; Hanley et al., 2018; Tarkowski, 2019). Hydrogen-essentially a decarbonised energy source-has thus been suggested as the best alternative energy form in the near future (Seo et al., 2020).

However, large-scale hydrogen storage is a key barrier for the implementation of a hydrogen economy (Zhang et al., 2016; Berta et al., 2018). Current storage options include chemical storage (e.g., as a metal hydride; Song, 2013), as a compressed gas (on the surface), or underground geological storage (Zhang et al., 2016; Pan et al., 2021). Based on safety, economic, and technical factors, underground hydrogen storage (UHS)-on which we focus here-has been suggested as the best option for large scale storage (Ozarslan, 2012). In UHS, hydrogen is injected into underground formations, and it can be withdrawn or refilled any time (Lubon and Tarkowski, 2020). As such UHS is conceptually and technically similar to CO_2 geo-sequestration (CGS), albeit here the CO_2 is injected for long term disposal and should not be recovered again (Metz

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et al., 2005). Target formations for UHS include depleted oil and gas reservoirs, saline aquifers, salt deposits reservoirs, permafrost grounds, and deep coal seams (Crotogino et al., 2010; Tarkowski, 2019; Han et al., 2020; Seo et al., 2020; Iglauer et al., 2021), thus storage capacities are potentially very large and such reservoirs are geographically abundant (Fossen, 2016).

So far, countries from worldwide have put great endeavours exploring the practical approaches for large-scale UHS. There are three pilot projects already testing UHS. (1) The 'HyStock project' in Netherlands, which completed in May 2020, has tested pure hydrogen storage in salt caverns and has triggered a sign of forthcoming 'hydrogen economy'. (2) The 'Underground Sun.Storage' project in Austria has assessed the mixed and pure hydrogen storage in gas fields. The completed Sun.Storage project subsequently performed as the fundamental for a further 'Underground Sun Conversion' project to achieve a large-scale storage of solar energy in the form of hydrogen in underground gas reservoirs. (3) The Hychico project in Argentina has investigated the mixed hydrogen storage in gas fields, and has explored the underground methanisation using combined H_2 and CO_2 injection (IEA, 2021). Apart from those pilot projects, large-scale UHS projects such as HyUnder Project in Europe, which was led by 12 countries including Germany, France, U. K. and etc., has thoroughly assessed the large-scale UHS potential throughout Europe which has set an effective model for future demonstrations.

However, due to the low density of H_2, H_2 migrates upwards in a reservoir and could leak back to the surface (similar to CO_2 in CGS) (Matos et al., 2019; Lankof and Tarkowski, 2020; Iglauer et al., 2021). It is thus vital to assess the feasibility of UHS at reservoir (hectometre) scale, to derisk UHS projects, and to avoid H_2 leakage (Lord et al., 2014; Lankof and Tarkowski, 2020; Luboń and Tarkowski, 2020).

Due to the novelty of the UHS concept, there is, however, very little knowledge about how H_2 flows through the reservoir (this knowledge is vital to interpret the feasibility of UHS). This research thus simulated H_2 injection and withdrawal from a three-dimensional (3D) heterogeneous reservoir and analysed various geological settings and H_2 injection scenarios. The results from this analysis were quantified and several conclusions were drawn. This work thus aids the in assessment of UHS projects and the large-scale implementations of a hydrogen economy.

2. Methodology

H_2 injection and withdrawal processes have been simulated for a sandstone reservoir with TOUGH2 software (Pruess et al., 1999), to predict hydrogen recovery factors, H_2 storage capacities and potential H_2 leakage rates. The EWASG (Equation-of-State for Water, Salt and Gas; Battistelli et al., 1997) was used to simulate the thermodynamic behaviour of the three components considered (i.e., water/NaCl/H_2). EWASG considers the effect of salinity on brine enthalpy, brine viscosity, brine vapour pressure, brine density, gas solubility in water, and the heat capacity of the brine. The reservoir had a length of x = 1400 m, a width of y = 1000 m and a depth of z = 500 m (z ranged from 1000 to 1500 m depth);

Table 1. Reservoir model characteristics (Al-Khdheeawi et al., 2017a, 2017b, 2017c, 2018).

Property	Value
Dimensions	1400 m × 1000 m × 500 m
Cell number	19 × 17× 40 = 12920 grid blocks
Vertical to horizontal permeability ratio	10%
Boundary cell volume multiplier	10^{24}
Initial aquifer salinity	60000 ppm
Initial reservoir pressure at 1000 m depth	10 MPa
Pressure gradient	10 kPa/m
Reservoir temperature (isothermal)	333 K
Initial brine saturation	100%
Injection perforation depth	1430 m
Production perforation depth	1280 m
Dip of the strata	0^0 (i.e., horizontal reservoir)

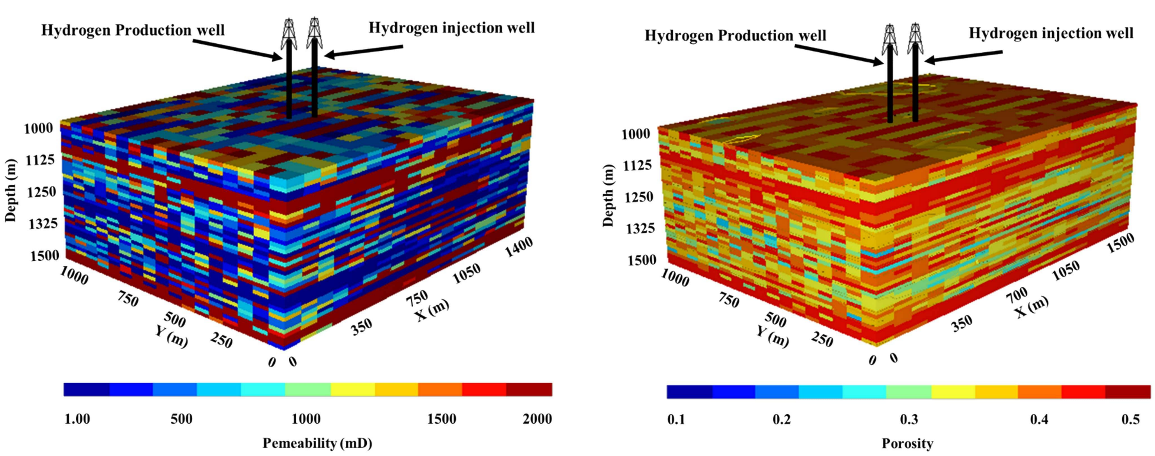

Fig. 1. 3D views of the heterogeneous reservoir model showing the hydrogen injection and production well locations, and the dimensions of the reservoir with different permeability distribution (left) and porosity distribution (right).

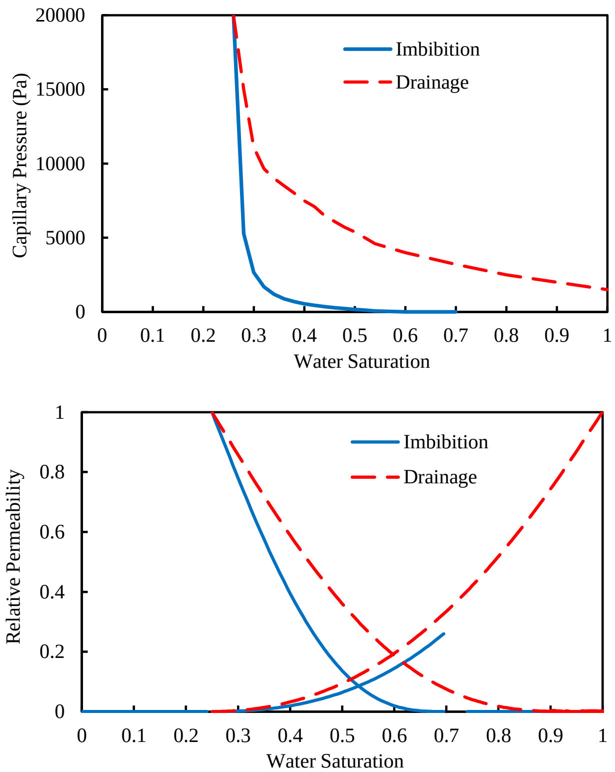

Fig. 2. Capillary pressure (top) and relative permeability (bottom) curves used to simulate the hydrogen injection-withdrawal process in a saline sandstone aquifer.

this resulted in a $19 \times 17 \times 40$ grid (i.e., 12920 cells in total; Fig. 1). Initial water saturation in the reservoir was 100%, and initial pressure was 10 MPa at 1000 m depth (Table 1). The model has been validated by comparing the resulted pressure distribution in the model with the pressure gradient of 10 MPa/km (Dake, 1978). Constant outer pressure boundary conditions (i.e., Dirichlet boundary conditions) were prescribed by multiplying the outer cell volume by a 10^{24} multiplier. Reservoir temperature was isothermal at 333 K, and reservoir heterogeneity was simulated by using the porosity and permeability distribution specified in the 10th SPE comparative solution project (Fig. 1). The ratio of vertical to horizontal permeability (k_v/k_h) was 10%. The effect of caprock availability was also tested. This is important as caprock is not available everywhere. Note that in the context of CO_2 geo-storage, projects have been approved for formations where no caprock is present (and which therefore mainly relies on residual CO_2 trapping) (Stalker et al., 2013). In the model realizations where a caprock existed, a caprock layer was constructed at a depth of 1263 to 1275 m. The porosity and permeability of the caprock were 0.0197 and 0.09 mD, respectively (Tian et al., 2015).

Sandstone (i.e., a weakly water-wet rock; Ali et al., 2021; Iglauer et al., 2021b) capillary pressure and relative permeability curves were used to simulate the hydrogen injection and production processes (Fig. 2; compare also Yekta et al., 2018). These curves were imported into the TOUGH2 code using the Van Genuchten–Mualem model (Mualem, 1976; Van Genuchten, 1980):

$$k_{rw} = \sqrt{S^*}\left\{1-(1-[S^*]^{\frac{1}{\lambda}})^{\lambda}\right\}^2 \qquad \text{if} \quad S_w < S_{ws} \tag{1}$$

$$k_{rw} = 1 \qquad \text{if} \quad S_w \geq S_{ws} \tag{2}$$

$$k_{rg} = 1 - k_{rw} \qquad \text{if} \quad S_{gr} = 0 \tag{3}$$

$$k_{rg} = (1-\hat{S})^2(1-\hat{S}^2) \qquad \text{if} \quad S_{gr} > 0 \tag{4}$$

$$P_c = P_0([S^*]^{\frac{-1}{\lambda}} - 1)^{1-\lambda} \tag{5}$$

$$S^* = \frac{S_w - S_{wr}}{S_{ws} - S_{wr}} \tag{6}$$

$$\hat{S} = \frac{S_w - S_{wr}}{1 - S_{wr} - S_{gr}} \tag{7}$$

where k_{rg} is H_2 relative permeability, k_{rw} is water relative permeability, S_{gr} is H_2 residual saturation, S_w is water saturation, S_{ws} is saturated water saturation, S_{wr} is water residual saturation, P_c is H_2-water capillary pressure, P_0 is capillary pressure scaling factor, and λ is pore size distribution index.

In addition, the influence of permeability and porosity heterogeneity on the capillary pressure for each grid block has been implemented via the Leverett J-function (Leverett, 1941):

$$J = \frac{P_c}{\sigma \cos\theta}\sqrt{\frac{k}{\phi}} \tag{8}$$

where J is dimensionless capillary pressure, k is intrinsic permeability, ϕ is porosity, σ is interfacial tension of H_2-water, θ is contact angle H_2-water-rock, and P_c is capillary pressure.

Note that separate injection and production wells were constructed to reduce the lateral and vertical spread of the hydrogen plume in the reservoir (Panfilov, 2016; Zivar et al., 2020). For both reservoir scenarios (with or without caprock), four different hydrogen injection rates were tested (i.e., 360, 1800, 3600, and 18000 kg/hr) at an injection depth of 1430 m over a 3-year hydrogen injection period. This simulates a scenario where larger amounts of H_2 are stored for longer times, e.g., before shipping/transport infrastructure is put in place for transporting the H_2 further to the end user (Bai et al., 2014; Acar and Dincer, 2019). Thus, different amounts of hydrogen have been injected (i.e., 9467, 47345, 94671, and 473449 tons) over the 3-year injection period. After the 3-year injection period a 1-year hydrogen withdrawal period was simulated, and the hydrogen leakage rate, the percentage produced hydrogen, and the percentage stored hydrogen (i.e., the remaining hydrogen) in the aquifer were computed and quantitatively analysed (see below).

The aquifer modelled here was considered to consist of 100% of quartz and feldspars, which are the main constituents of a sandstone reservoir (Tiab and Donaldson, 2004). As no chemical reaction between H_2 and these minerals was experimentally observed (Yekta et al., 2018a), no H_2-mineral

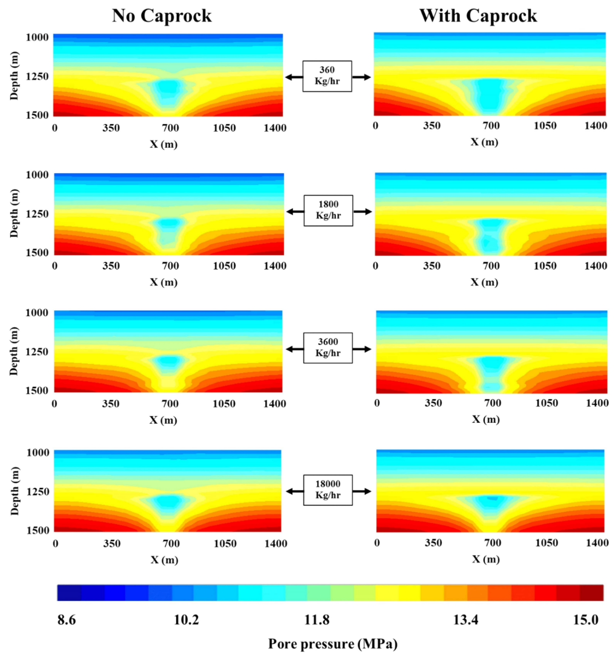

Fig. 3. Pore pressure distribution for the various reservoir and injection settings, at the end of the 1-year hydrogen production period.

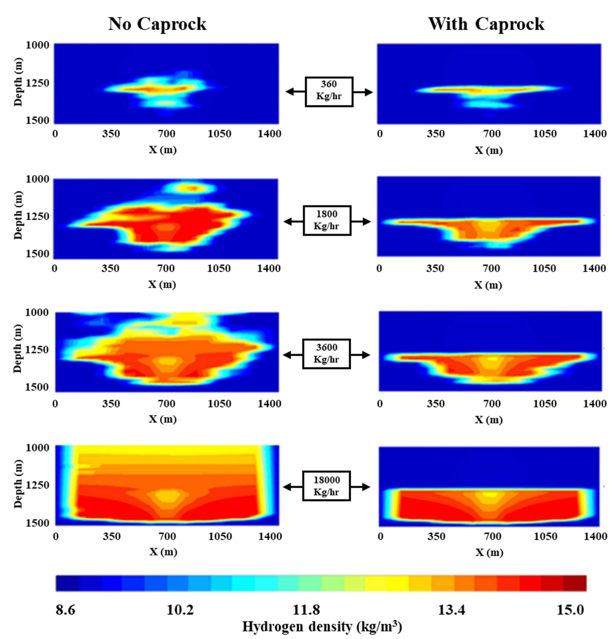

Fig. 4. Hydrogen density distributions for the various reservoir and injection settings, at the end of the 1-year hydrogen production period.

chemical reactions were included in the model.

3. Results and discussion

The pore pressure and hydrogen density distributions for the two reservoir scenarios are shown in Figs. 3 and 4. Fig. 3 shows that the pressure distribution in our model match the pressure gradient of 10 MPa/km (Dake, 1978). Fig. 4 indicates that hydrogen leakage is affected by both injection rate and

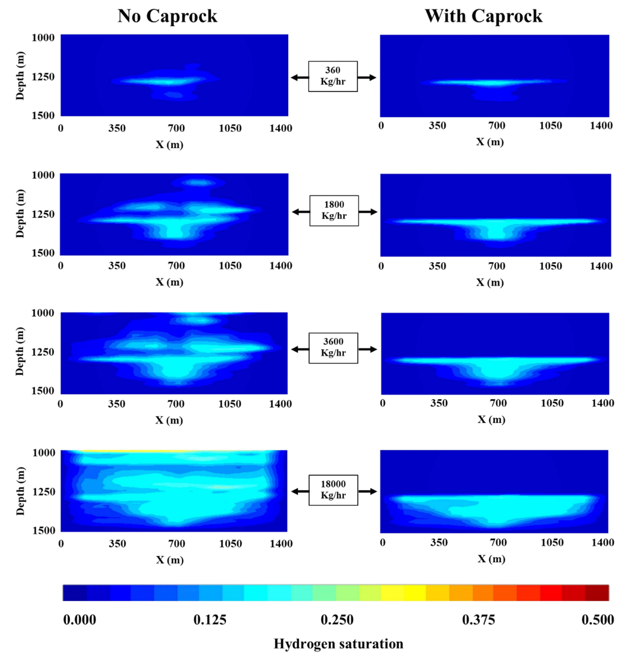

Fig. 5. Hydrogen plume predicted for the two caprock scenarios (with and without caprock) for the four injection rates examined, at the end of the 1-year hydrogen production period.

caprock availability.

For instance, Fig. 4 shows that higher injection rate leads to an increase of the unwanted vertical leakage and lateral spreading of hydrogen. The results indicate that higher injection rate will increase hydrogen losses, and the reason of this is that high injection rate tends to increase the fingering, residual saturation of hydrogen, hydrogen solubility in formation water, diffusion force and viscous force. Thus, this research established the conclusion that injection rate has a significant impact on the hydrogen storage efficiency and that low injection rate is preferable in the underground practice.

Generally, high vertical hydrogen leakage is unwanted in hydrogen underground storage, as it increases H_2 losses and potentially poses a safety problem (Ali et al., 2021b; Pan et al., 2021). Thus, here, the effect of caprock and injection rate on hydrogen leakage was analysed after the 1-year withdrawal period.

Clearly, caprock availability highly affected H_2 leakage, thus caprock prevented H_2 from leaking to the surface (for all injection rates tested). For instance, for an injection rate of 3600 kg/hr or higher, hydrogen leaked to the top of the model (1000 m) in case no caprock was present; otherwise it was trapped below the caprock, Fig. 5. In addition, the injection rate influenced hydrogen leakage in case caprock was absent; and higher injection rates increased H_2 leakage (for example, after the 1-year production period, H_2 depth reached 1200 m for an injection rate of 360 kg/hr, but 1050 m for an injection rate of 1800 kg/hr). The conclusion is that caprock prevents unwanted H_2 leakage, and low H_2 injection rates are preferred in case caprock is absent.

Higher injection rates reduced the hydrogen recovery factor (i.e., the ratio of the recovered hydrogen to the total injected

Table 2. Hydrogen injection and production statistics for different hydrogen injection rates and caprock availability (after the 3-year injection and 1-year withdrawal period).

Injection Rate (kg/hr)	Total injection mass (tons)	Reservoir without caprock		Reservoir with caprock	
		Recovery Mass (tons)	Recovery factor (%)	Recovery Mass (tons)	Recovery factor (%)
360	9467	2835	29.9	3408	36
1800	47345	5254	11.1	10598	22.4
3600	94671	7519	7.9	15110	16
18000	473449	23188	4.9	33331	7

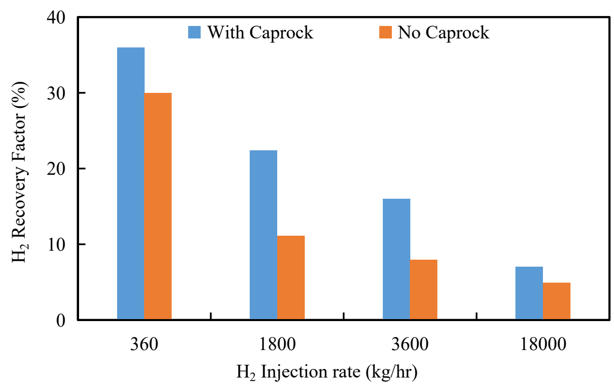

Fig. 6. Hydrogen recovery factor for the two caprock scenarios investigated as a function of hydrogen injection rate at the end of the 1-year hydrogen withdrawal period.

hydrogen). For instance, the recovery factor at a 360 kg/hr injection rate was 36% (with caprock) and 29.9% (no caprock), but only 7% (caprock) and 4.9% (no caprock) at 18,000 kg/hr injection rate (Fig. 6).

Caprock thus also significantly affected the recovery factor, i.e., caprock increased the amount of recovered hydrogen, for all hydrogen injection rates tested. This has been further quantified in Table 2 and is visualized in Fig. 7. Overall, it is concluded that low injection rates and caprock availability are preferred scenarios, due to their higher hydrogen recovery factors.

4. Conclusion and implications

Hydrogen storage is a key barrier to implementing a large-scale hydrogen economy. Currently, hydrogen is stored as a compressed gas, in chemical form (e.g., as metal hydride) or in underground geological formations (Tarkowski, 2019; Lubon and Tarkowski, 2020; Zivar et al., 2020). Underground hydrogen storage (UHS) is considered the best option for large-scale H_2 storage due to safety, economic, and technical factors (Tarkowski, 2019; Seo et al., 2020). However, knowledge about UHS is very limited as it is a new concept. This research thus simulated UHS in a heterogeneous 3D reservoir and examined the effects of caprock availability and hydrogen injection rate on reservoir (hectrometer)-scale H_2 plume dynamics and the amount of recoverable hydrogen. Clearly both, caprock and injection rate, significantly affected vertical hydrogen leakage, and hydrogen recoverability. Based on the simulations results the conclusion is that higher injection rates

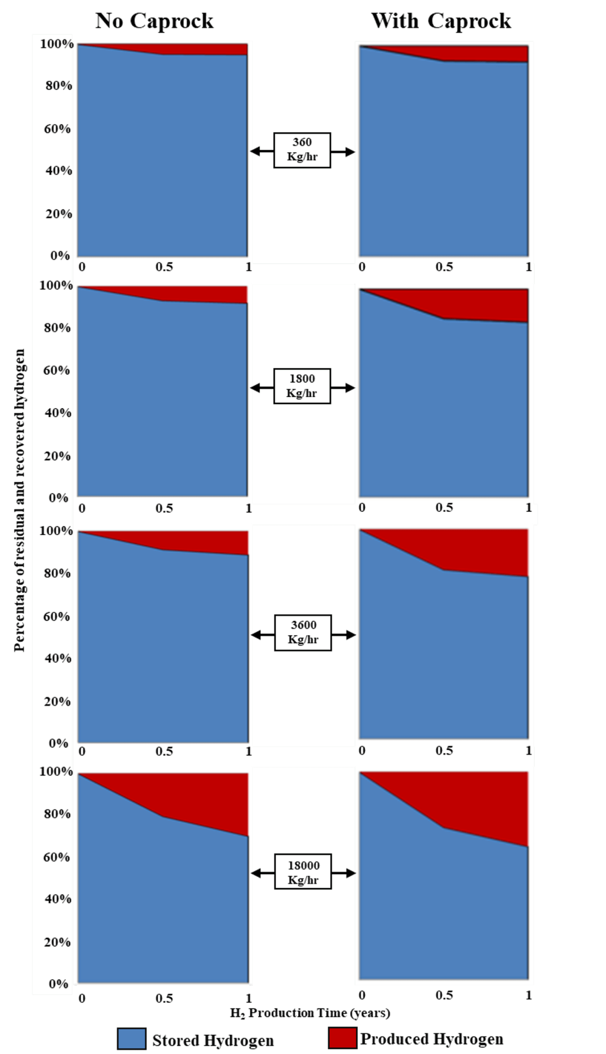

Fig. 7. Percentage of residual and recovered hydrogen as a function of hydrogen injection rate and caprock availability.

lead to a) increased H_2 leakage when caprocks are absent and b) reduce the amount of recovered hydrogen. It is therefore advantageous to operate UHS in reservoirs sealed with a caprock, and to use lower H_2 injection rates. This work thus provides fundamental predictions about H_2 plume dynamics at reservoir scale, and about the recoverability of the H_2- these insights will aid in the large-scale implementation of a hydrogen economy.

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Conflict of interest

The authors declare no competing interest.

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