

# A site assessment tool for H<sub>2</sub> storage in depleted hydrocarbon reservoirs

Moein Jahanbani Veshareh<sup>a</sup>, Hamidreza M. Nick<sup>b</sup>, Mohammad Reza Alizadeh Kiapi, Ali Mahmoodi<sup>d</sup>

<sup>a</sup> Denmark Technical University, Kongens Lyngby, Denmark, [Moein@dtu.dk](mailto:Moein@dtu.dk), CV

<sup>b</sup> Denmark Technical University, Kongens Lyngby, Denmark, [Hamid@dtu.dk](mailto:Hamid@dtu.dk)

<sup>c</sup> Denmark Technical University, Kongens Lyngby, Denmark, [Mraki@dtu.dk](mailto:Mraki@dtu.dk)

<sup>d</sup> Denmark Technical University, Kongens Lyngby, Denmark, [Alimhm@dtu.dk](mailto:Alimhm@dtu.dk)

## Abstract:

Hydrogen storage is a key component in realization of an emission free future. Depleted hydrocarbon reservoirs offer a low cost medium for large-scale hydrogen storage. While the effect of hydrogen in triggering some chemical and biochemical reactions has established some screening criteria to choose a suitable underground storage site according to reservoir geochemistry, there is no screening criteria based on the effect of variables such as pressure, temperature and composition of the residual hydrocarbon on hydrogen recovery. In this work, we first investigate the cost required for hydrogen compression in terms of the work required for compressors. Then we investigate the effect of reservoir pressure, storage pressure, reservoir temperature and residual composition on hydrogen recovery. Our results show that on one hand the work required for pressurizing hydrogen does not increase linearly with pressure, and on the other hand, hydrogen recovery increases with storage pressure. Additionally, Hydrogen recovery was shown to decrease by increase in reservoir initial pressure before hydrogen storage. Therefore, it seems that hydrogen storage will be more efficient if it is conducted at the highest possible pressure in a reservoir with low initial pressure (either a shallow reservoir, or a depleted reservoir). Our results did not show any strong relationship between hydrogen recovery and temperature. Hydrogen recovery showed to increase slightly with increase in residual hydrocarbon density. However, the effect of residual hydrocarbon was observed to be significant on purity of the produced hydrogen. In this sense, depleted black oil reservoirs seem to be the best and dry gas reservoirs the worst choice.

Keywords:

Energy storage; Hydrogen; Underground storage; Screening; Depleted hydrocarbon reservoirs

## 1. Introduction

Hydrogen is a clean energy carrier that can decarbonize transport, heating and other energy intensive sectors [1-3]. United Nation Industrial Development Organization (UNIDO) has reported hydrogen as “a true paradigm shift in the area of more efficient energy storage, especially for renewable energy on industrial scale” [4] and the IPCC’s 1.5°C report recognizes hydrogen as a major player in restricting global warming by reducing CO<sub>2</sub> emission of energy demanding industries [5].

The downside of many renewable energy resources is their intermittent nature, being dependent on seasonally fluctuating events such as wind and sunshine [6, 7]. This intermittency leads to periods of energy excess and deficit, disabling renewable energy to satisfy energy demands [8, 9]. Energy storage can alleviate the intermittency of renewable energies by saving the produced energy in the energy excess periods for the energy deficit periods. Energy can be stored as hydrogen either through water electrolysis or by fossil fuel reformation coupled with carbon capture and storage [10].

While energy in MWh scale can be stored in surface facilities such as tanks and pipes, underground storage is required for GWh or TWh scales. Salt caverns are the main subsurface setting that have been utilized for hydrogen storage commercially in the past [11, 12]. Salt caverns offer a very low theoretical hydrogen loss of only 0.01% [13] since they are inert to hydrogen and very gas-tight. Additionally, salt caverns allow high injection and production rates, making them suitable for short to medium term storage (where multiple storage-withdrawal cycles are required) [13]. However, hydrogen storage in salt caverns is restricted by pressure (below 20MPa [14]) and the presence of salt caverns with a suitable thickness, limiting their storage capacity to GWh scale [15].

Saline aquifers and depleted hydrocarbon reservoirs are other subsurface settings that can be used for underground hydrogen storage. Compared to salt caverns, saline aquifers and depleted hydrocarbon reservoirs can be found easier geographically and they have orders of magnitude higher storage capacity that

can even facilitate energy storage in TWh scale. Hydrogen storage in saline aquifers compared to depleted hydrocarbon reservoirs requires exploration and drilling costs. Moreover, saline aquifers are saturated with water at hydrostatic pressure. Displacement of the saturating water may require high injection pressures [16]. Besides, the presence of water leads to water coning in hydrogen production stage that limits hydrogen production rate and reduces hydrogen recovery [17]. Therefore, hydrogen storage in depleted hydrocarbon reservoirs is more appealing.

In the literature screening criteria have been well established for CO<sub>2</sub> capture and storage in depleted hydrocarbon reservoirs, e.g. [18]. For hydrogen storage, some screening criteria have been suggested according to research studies that have studied the influence of chemical and biochemical reactions that can occur due to interactions between hydrogen and reservoir rock minerals and indigenous microbial community. These criteria are usually environmental parameters that control microbial growth such as temperature, pH and formation brine salinity [19, 20]. However, to the best of our knowledge no research work has screened depleted oil reservoirs from reservoir engineering point of view.

Screening a reservoir for hydrogen storage from reservoir engineering point of view can include macroscopic characteristics such as pressure and temperature and microscopic factors such as wettability. While understanding the effect of microscopic properties requires dynamic simulations, by considering some simplifying assumptions, the effect of macroscopic properties can be estimated using static experiments. The aim of this work is to screen oil reservoirs according to their macroscopic properties, namely, temperature, pressure and residual oil composition.

## 2. Materials and methods

### 2.1. Calculation of compression energy consumption

As the high-pressure water electrolysis technology is still immature, we assume that compression is necessary to pressurize hydrogen into desirable pressures required for injection and storage. We can divide compressors into three categories of positive displacement, dynamic and injectors. Positive displacement or intermittent flow compressors confine an amount of gas within a specific volume. By decreasing the volume, they rise the pressure of the confined gas. Positive displacement compressors are available in two types: Rotary and Reciprocating. Dynamic or turbo compressors increase the gas pressure by using inertial forces that increase gas velocity and convert energy into pressure. Dynamic compressors can be found either as centrifugal compressors or axial compressors. The compressor type choice is mainly according to discharge pressure and inlet flow. Given the discharge pressure and inlet flow scales that we deal with in hydrogen storage projects, centrifugal compressors are more likely to be suitable. Therefore in this work we only focus on centrifugal compressors.

Compressors are usually connected to another machine that derives the shaft. While various compressor deriving machines such as gas turbine, electric motors, steam turbine and turbo expanders are available, in this work we only focus on electric motors as electricity is the form of energy that is available for us in terms of surplus renewable energy. Therefore we assume no energy loss in compressor derive machine.

Isothermal gas compression can be proved to require the least amount of energy to raise gas pressure from suction pressure ( $P_{\text{suction}}$ ) to discharge pressure ( $P_{\text{discharge}}$ ). However, an isothermal compression is practically impossible. Adiabatic gas compression is a term used when no heat transfer occurs during the compression process between the system and the surrounding. If we consider hydrogen molecular weight and gas constant to be 2.016 g/mol and 8.3145 J·mol<sup>-1</sup>·K<sup>-1</sup>, respectively, the work ( $W$ , kJ/kg) required to compress a kg of hydrogen gas from  $P_{\text{suction}}$  to  $P_{\text{discharge}}$  can be calculated using the following equation:

$$W = 4.124 \cdot T_{\text{suction}} \cdot \left( \frac{Z_{\text{discharge}} + Z_{\text{suction}}}{2} \right) \cdot \frac{1}{\eta} \cdot \left( \frac{K}{K-1} \right) \cdot \left[ \left( \frac{P_{\text{discharge}}}{P_{\text{suction}}} \right)^{\frac{K-1}{K}} - 1 \right] \quad (1)$$

Where  $T_{\text{suction}}$  is the suction temperature in °K,  $K$  is the adiabatic gas component,  $Z$  is the compressibility factor and  $\eta$  is the thermal efficiency of compression and is in the range of 0.75-0.8. According to equation 1, the work required for compression does not increase linearly by increasing the compression ratio ( $P_{\text{discharge}}/P_{\text{suction}}$ ) as the power of compression ratio is below one. However, compression cause a significant increase in temperature. Compressor manufacturers normally advice a maximum discharge temperature of 177 °C for oxygen free gasses. Additionally, for safety concerns, manufactures normally recommend compression ratios of below six. Therefore, depending on hydrogen injection pressure, several stages of compressors maybe required. The discharge temperature can be found using the following equation:

$$\frac{Z_{\text{discharge}} \cdot T_{\text{discharge}}}{Z_{\text{suction}} \cdot T_{\text{suction}}} = \left( \frac{P_{\text{discharge}}}{P_{\text{suction}}} \right)^{\frac{K-1}{K}} \quad (2)$$

Note that  $Z_{\text{discharge}}$  depends on  $T_{\text{discharge}}$ ; therefore,  $T_{\text{discharge}}$  can be calculated by assuming  $Z_{\text{discharge}}$  to be equal to one, then in an iterative process values of  $Z_{\text{discharge}}$  and  $T_{\text{discharge}}$  can be updated until  $T_{\text{discharge}}$  converges. In order to find out the optimum number of compressors we use the theory of equal compression ratios. For example if hydrogen pressure needs to increase 15 times, then 3 compression stages (that each pressurize the gas 5 times) are used. We assume that by using pre and intercooling units  $T_{\text{suction}}$  for every compression stage is 20°C. We ignore the pressure drop that occurs in cooling stages. If achieving a certain compression ratio increases gas temperature to above 177°C, then more compression stages are used.

## 2.2. Hydrogen loss and recovery

A unit volume (1m<sup>3</sup>) of a storage medium with a storage pressure of  $P_{\text{storage}}$  will store  $n_{\text{stored}}$  moles of hydrogen according to the following equation:

$$n_{\text{stored}} = 1.2 \times 10^5 \frac{P_{\text{storage}}}{Z_{\text{storage}} T_{\text{reservoir}}} \quad (3)$$

Note that in equation 3 we have assumed that temperature of the subsurface medium ( $T_{\text{reservoir}}$ ) does not change due to hydrogen storage. If we ignore water coning or any other mechanism that might cause hydrogen trapping, and if we assume that no derive mechanism (e.g. rock expansion and aquifer derive) will change the volume that hydrogen occupies, a fraction of hydrogen is produced due to expansion when storage medium pressure is allowed to drop to  $P_{\text{abandonment}}$ , and a fraction of hydrogen will remain in the reservoir. The remaining hydrogen can be calculated via the following equation:

$$n_{\text{trapped}} = 1.2 \times 10^5 \frac{P_{\text{abandonment}}}{Z_{\text{abandonment}} T_{\text{reservoir}}} \quad (4)$$

The ratio of  $100 \times (1 - [n_{\text{trapped}}/n_{\text{stored}}])$  indicates maximum percent of hydrogen that can be recovered.

## 2.3. Residual oil composition effect on recovery

In order to investigate the effect of residual hydrocarbon type on hydrogen recovery, we used the workflow shown in Fig. 1. Hydrogen production was modeled by stepwise removal of 5% gas phase volume. In each step Peng-Robinson equation of state (EoS) was utilized to calculate the new pressure and vapor liquid volume and composition. Gas phase removal was stopped when pressure dropped to the abandonment pressure. Various initial hydrocarbon compositions were generated by randomly mixing C1, C3, C5 and C7+ with various molar fractions. Hydrogen recovery was calculated by dividing total hydrogen production at abandonment pressure by initially stored hydrogen.

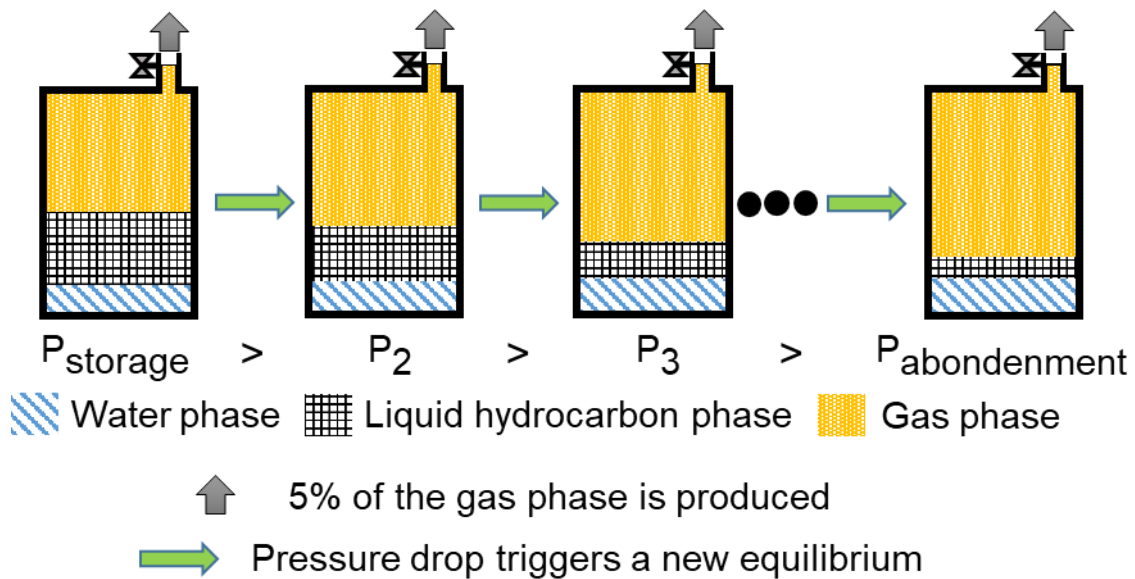


Fig. 1. Schematic representation of the mathematical model and workflow to study the effect of residual hydrocarbon composition on hydrogen recovery.

### 3. Results

#### 3.1. Compression energy consumption

If we assume that the discharge pressure of the electrolyzer is 0.2 MPa, Fig. 2A and B show the required work for compressing a kg of hydrogen and the minimum number of compressors required. Note that Fig. 2A merely demonstrates the adiabatic power consumption; it does not take into account any power consumption due to pressure losses such as the pressure loss in intercooling stages or valves. According to Fig. 2A the required work does not increase linearly with pressure. Considering the combustion energy of  $H_2$  that is 33.6 kWh/kg, Fig. 2A illustrates that depending on the required discharge pressure, between 2 to 10% of energy is lost for compression. Fig. 2B shows that Except for very low (below 17 bar) or very high (above 80 bar) discharge pressures, 4 compression stages are required.

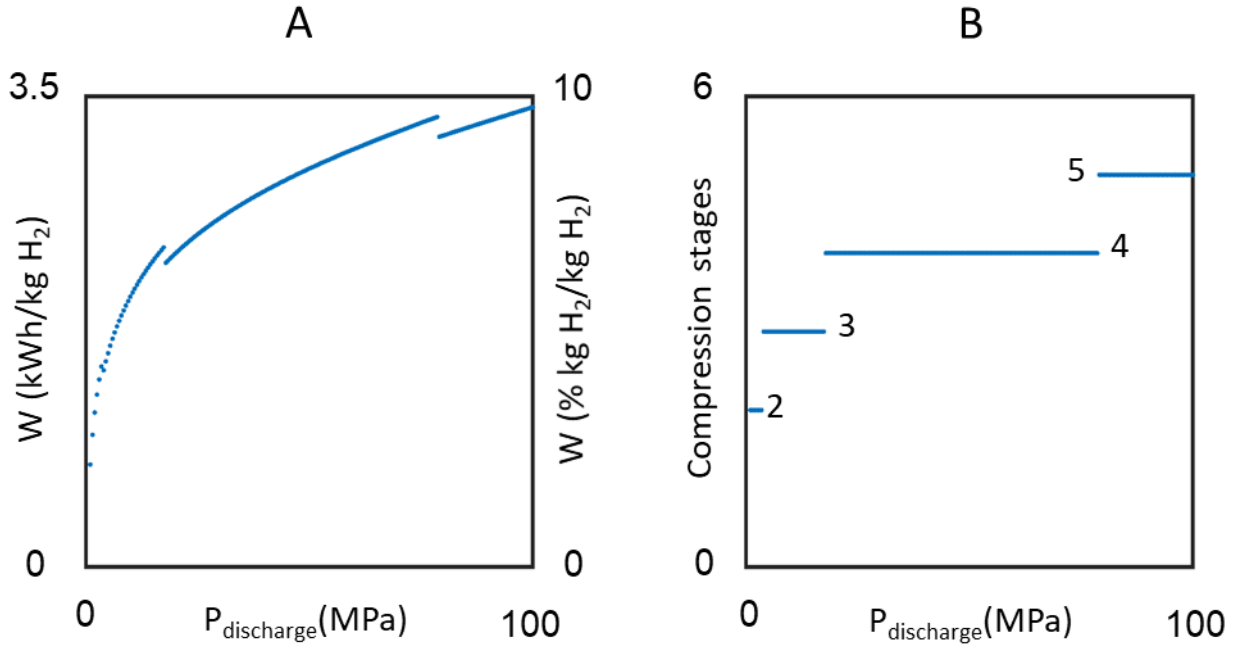


Fig. 2. A) Work required (in kWh  $H_2$ , left y axis and in % kg  $H_2$ , right y axis) for pressurizing a kg of hydrogen from 0.2 MPa adiabatically, and B) Minimum number of compression stages such that discharge pressure to suction pressure in none of the compression stages is higher than six, and discharge temperature does not exceed 177°C.

#### 3.2. Hydrogen loss and recovery

If we consider reservoir temperature to be 50°C and reservoir initial pressure to be 10 MPa, Fig. 3A and B show the recovery and hydrogen loss ( $n_{trapped}$ ), respectively. While temperature effect is negligible, by an increase in injection pressure, the recovery is increased (Fig. 3A). Fig. 3B shows that by increase in temperature, the trapped hydrogen decreases from 7.5 to 5.5 kg. Therefore, reservoir temperature does not seem to influence hydrogen recovery or loss. Fig. 3A shows that hydrogen recovery increases significantly with pressure at low pressures and at higher pressures the dependency of recovery on storage pressure decreases. Fig. 3C shows that after a certain pressure the increase in work required for compression is more than the increase in recovery factor. Note that the intersection point in Fig. 3C is only an optimum point in terms of cost benefit, as hydrogen compression requires always less than 10% of the injected hydrogen.

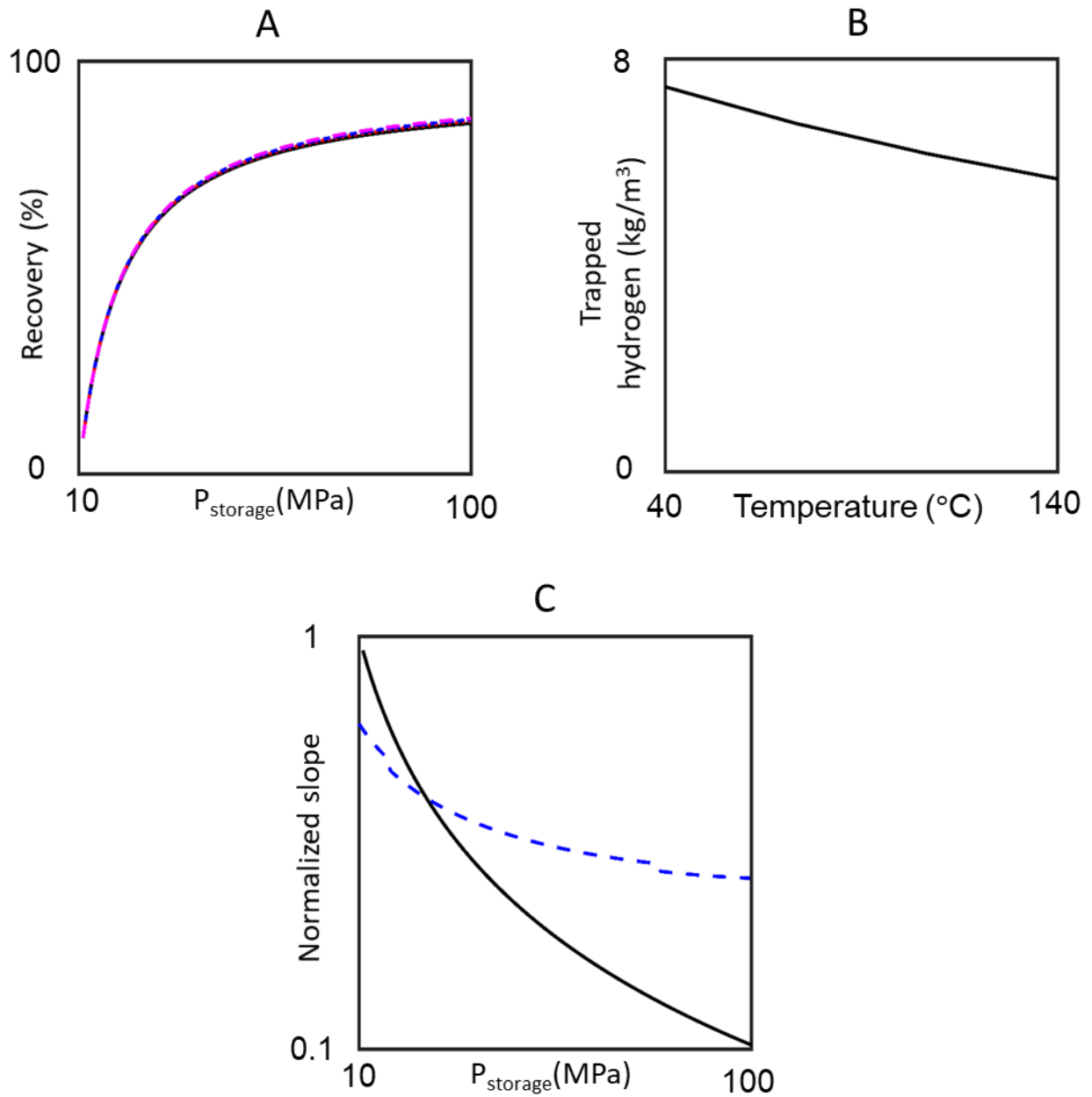


Fig. 3. A) Percent of the injected hydrogen that is recovered for various storage pressures and temperatures (solid, dotted, dotted-dashed and dashed lines for 40, 73.3, 106.7 and 140°C, respectively). B) Trapped hydrogen in a unit volume ( $1\text{m}^3$ ) for various temperatures considering an abandonment pressure of 10 MPa. C) The slope of normalized recovery percent ( $[ \text{Recovery} - \min(\text{Recovery}) ] / [ \max(\text{Recovery}) - \min(\text{Recovery}) ]$ ) and normalized compressor work ( $[ W - \min(W) ] / [ \max(W) - \min(W) ]$ ) (Fig. 2A) for different pressures.

If we assume the injection pressure of 50 MPa, Fig. 4 shows the influence of various initial pressures (or abandonment pressures) on trapped hydrogen and recovery. According to Fig. 4A, by an increase in abandonment pressure from 11 to 40 bar, recovery factor decreases from 75 to 17%. While temperature effect on recovery factor is negligible, Fig. 4B shows that for a given abandonment pressure, trapped hydrogen is less at higher temperatures.

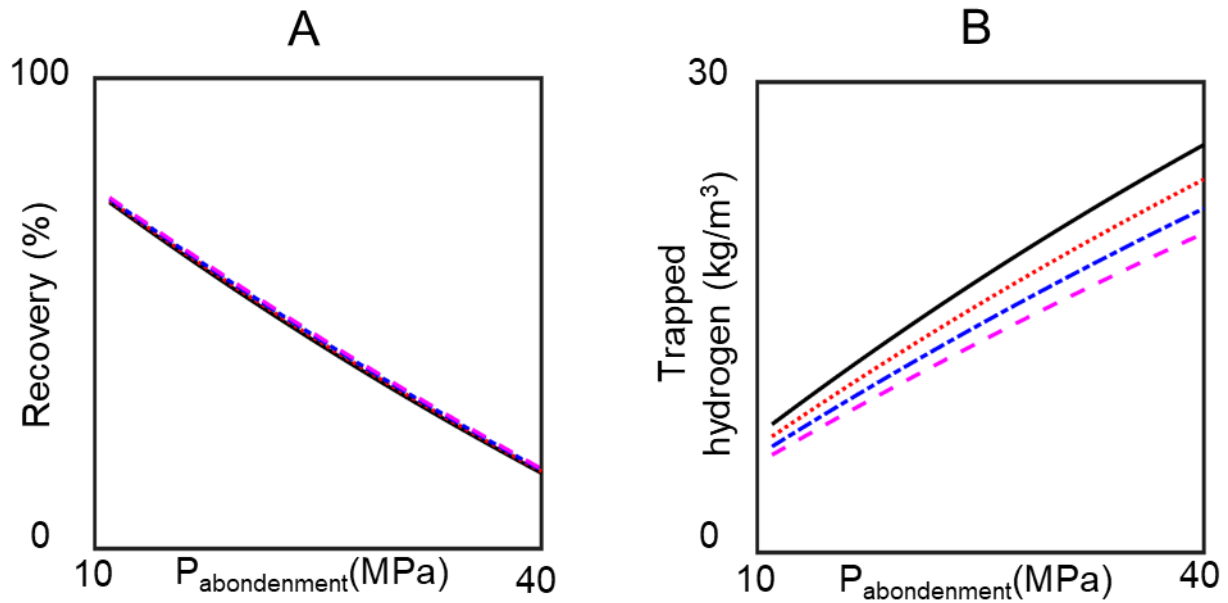


Fig. 4. A) Percent of the injected hydrogen that is recovered from a hydrogen storage with a storage pressure of 50 MPa for various abandonment pressures between 10 to 40 MPa and different reservoir temperature of 40, 73.3, 106.7 and 140°C (represented by solid, dotted, dotted-dashed and dashed lines, respectively). B) Trapped hydrogen in a unit volume (1m<sup>3</sup>)

Fig. 5 shows the effect of temperature on storage capacity at various temperatures. The dependency of storage capacity on temperature is more significant in higher pressures (e.g. 100 MPa) and is negligible in low pressures (e.g. 10 MPa).

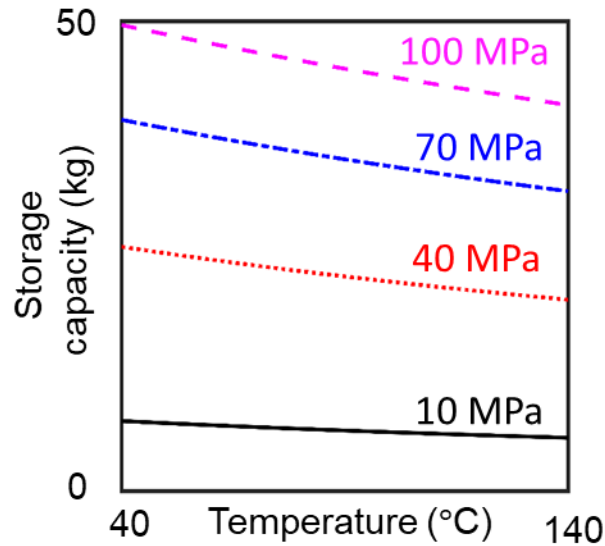


Fig. 5. Storage capacity in terms of kg hydrogen that can be stored in a unit volume (1m<sup>3</sup>) at temperatures between 40 to 140°C and pressures between 10 to 100 MPa.

### 3.3. Residual oil composition effect on recovery

In order to visualize various compositions, we used hydrocarbon density at storage condition. Lower densities represent compositions with higher fraction of C1 and C3, whereas higher densities represent compositions with higher fraction of C5 and C7+. According to Fig. 6, there is no strong relationship between residual oil composition and hydrogen recovery. The recovery seems to increase slightly by increase in residual hydrocarbon density. At a given density, up to 5% difference in recovery can be observed for various compositions. While 5% recovery is a small difference, in reservoir scale it can cause a significant energy loss.

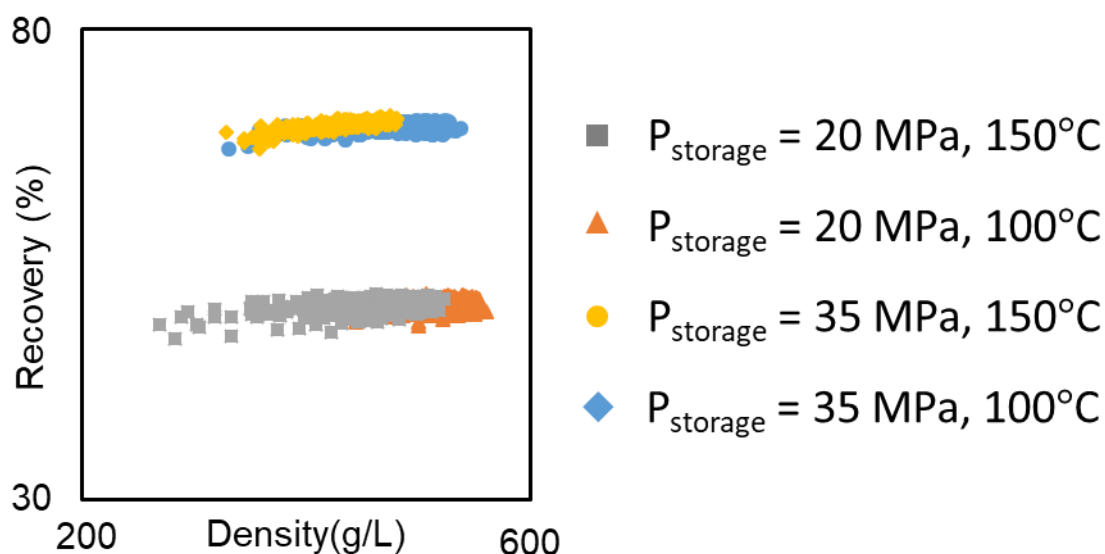


Fig. 6. The percent of stored hydrogen that is produced due to sequential gas phase removal – pressure drop (as shown in Fig. 1) from two storage pressure of 20 and 35 MPa to an abandonment pressure of 10 MPa at two different temperatures of 100 and 150°C.

While residual oil composition effect on ultimate recovery is small, it influences purity of the produced gas significantly. For four different residual hydrocarbons, Fig. 7 shows that purity of the produced hydrogen is higher when residual oil contains heavier components. Therefore, hydrogen storage in depleted reservoirs with lighter hydrocarbons may be slightly more beneficial in terms of ultimate hydrogen recovery, but associated with lower purities of hydrogen in production fluids.

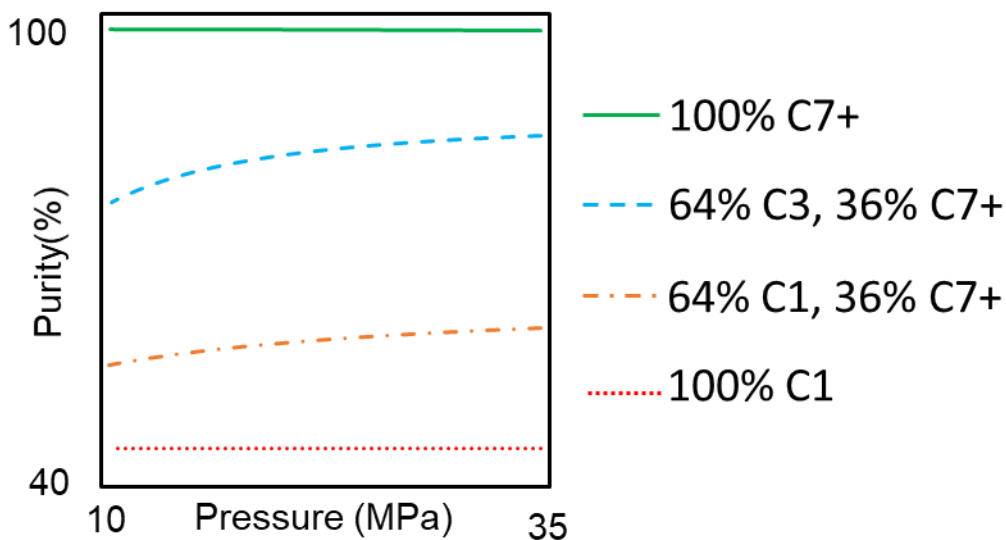


Fig. 7. Purity of the produced hydrogen as a function of pressure drop. Pressure of 35 MPa is the storage pressure and then pressure drops stepwise to an abandonment pressure of 10 MPa. Therefore, higher pressures represent early stages of hydrogen recovery, while lower pressures represent older stages.

## 4. Discussion

Our results show that in on the one hand higher storage pressures are associated with a higher recovery, and on the other hand a lower cost in terms of the required work for compression. However, increase in pressure is restricted by the pressure above which cap rock becomes permeable. Abonnement pressure is another property that influences recovery factor. Abandonment pressure is a pressure below which either no fluid is produced, or the pressure at which hydrogen production is not significant. Note that abandonments pressure cannot be higher than reservoir initial pressure. This is one reason why hydrogen storage is more efficient in shallower reservoirs, or reservoirs that their pressure has fallen to sufficiently low pressures due to depletion. Note that the aforementioned features, allowing high storage pressures, and having a low initial pressure are two advantages of hydrogen storage in depleted hydrocarbon reservoirs compared to salt caverns, which do not allow storage pressures of more than 20 MPa, and saline aquifers, which have a high initial pressure equal to hydrostatic pressure as they are not depleted.

According to our results, temperature influence on hydrogen storage from reservoir engineering point of view is small. Considering the higher risk of microbial hydrogen loss in lower temperatures, hydrogen storage seems to be more efficient in reservoirs with higher temperatures. Note that higher temperatures usually are associated with higher depths and therefore higher hydrostatic pressures. This is another reason why using depleted hydrocarbon reservoirs is advantageous compared to saline aquifers, since depleted hydrocarbon reservoirs have a lower pressure due to depletion.

Based on our study, hydrogen recovery showed to be not significantly related to the composition of the residual oil. However, the purity of the produced hydrogen will be higher if hydrogen storage is done in a hydrocarbon reservoir with heavier hydrocarbons. The reason is that heavier hydrocarbons tend to stay in the liquid phase and do not contaminate the gas phase (hydrogen), while lighter hydrocarbons will vaporize to the gas phase as the pressure drops and reduce the mole fraction of hydrogen (purity). Therefore, hydrogen storage in depleted black oil reservoir seems to result in the highest hydrogen purity, while dry gas reservoirs should be associated with a low hydrogen purity.

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