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The influence of temperature on wettability alteration during CO₂ storage in saline aquifers

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Abstract

The wettability of a formation is defined as the tendency of one fluid to spread on a surface in competition with other fluids which are also in contact with it. However, the impact of temperature on wettability in an aquifer and the modification of relative permeability curves based on the temperature variation in aquifers is not well covered in the literature. This study redresses this dearth of information by investigating the impact of temperature on wettability distribution in a reservoir and updating the relative permeability curves based on its temperature propagation. The impact of the latter is studied in relation to the solubility of CO₂ injected into an aquifer using the numerical methods (i.e. ECLIPSE). If the CO₂ injected has a temperature higher than the formation geothermal temperature, it can change the wettability of the formation further to a more CO₂ wet condition. This increases the risk of leakage and also changes the relative permeability curves as the CO₂ moves through the reservoir, a situation that needs to be considered in reservoir simulations. The results show that updating and modifying the relative permeability curves with temperature variation in an aquifer can increase the amount of CO₂ dissolution there.

Keywords: CO₂ storage, wettability alteration, CO₂ solubility, relative permeability

1. Introduction

The concentration of CO₂ in the atmosphere has increased up to 45% since the industrial revolution (Celia et al., 2015). However, due to Covid-19 pandemic in 2020, as industries, transportation

systems and all other businesses have shut down it has caused a sudden drop in CO₂ emission. Compared to this time last year, levels of air pollution in New York has dropped at almost 50% due to restrictions. In China, emission data shows a 25% decrease at the starting point of the year and coal usage has decreased by 40% at china's largest power plant since the last three months of 2019 (Saadat et al., 2020). Carbon capture and storage (CCS), that comprises the separation of CO₂ from the gaseous exhaust of power plants and other heavy industries and its safe and secure long-term storage in geological formations is considered as the most applicable method for mitigating the rise of CO₂ concentrations in the atmosphere (Metz et al., 2005; Bachu, 2008; Jiang, 2011). The best storage sites are those that trap the CO₂ as an immobile phase under a ultra-low permeability confining caprock where it is subjected to further gradual physical and chemical trapping mechanisms (Metz et al., 2005). In the long-term, several trapping mechanisms are active in the aquifer which are categorised as: structural trapping, residual trapping, solubility trapping and mineral trapping (Garcia et al., 2010). Since the CO₂ leakage from a storage site can create harmful environmental defects, its long-term storage security is of a great importance (Abbaszadeh and Shariatipour, 2018; Metz et al., 2005; Gasda et al., 2004; Nordbotten et al., 2005; Burton and Bryant, 2007; Celia et al., 2011).

Ali et al., (2019) investigated the CO₂ wettability of sandstones exposed to traces of organic acids. Their results indicate that the quartz surface turned significantly less water wet with increasing organic acid concentration which lowers the residual trapping capacity of the formation in comparison with a water wet condition. Their results are in line with the results of our study. In another study, they investigated the effect of nanofluids on CO₂ wettability reversal of sandstone formation (Ali et al., 2020; Ali, 2018). The results of their study again showed that the quartz surface turned hydrophobic by exposing to organic acids and turned to hydrophilic when nanofluids were used which increases the CO₂ storage capacity. These findings are in agreement with the results of our study.

Researchers have studied the impact of different parameters, such as the heterogeneity of the permeability, absolute permeability, porosity and mineralogy of the pore surface, on fluid flow and

investigated experimentally (Chaudhary et al., 2013; Rahman et al., 2016), it has received far less attention in simulation studies and there is a lack of information on the effect on CO₂ solubility in the aquifer. Previous simulation studies have shown that wettability of a sand surface drastically impacts the relative permeability curves (McCaffery and Bennion, 1974; Heiba et al., 1983; Krevor et al., 2012; Levine et al., 2013), capillary pressure curves (Heiba et al. 1983; Anderson, 1987) and the extent and distribution of fluids in a porous media (Morrow, 1990). Thus, in this work we have attempted to investigate the impact of reservoir wettability on CO₂ dissolution in an aquifer.

Jha et al., (2019) investigated the wettability alteration of quartz surface by low salinity surfactant nanofluids at high pressure and high temperature conditions. They showed using contact angle wettability measurements that initial weak water wet quartz surface become more water wet when Zirconia nanoparticles used in low salinity formulation (Jha et al., 2018).

Farokhpoor et al., (2013) have shown that the changes in the wettability condition of a reservoir rock could lead to a decrease in the capillary entry pressure and consequently the sealing capability of the cap rock. They investigated the impact of pressure, temperature and salinity on the wettability of different minerals by measuring the contact angle. Alnili et al., (2018) compared the wettability conditions of the action of CO₂-brine on porous sandstone and pure quartz. Their results show that the contact angle of porous sandstone is higher than for pure quartz due to the presence of pores in the sandstone. Moreover, the pressure, temperature and salinity have the same effect on pure quartz and sandstone as the contact angle increases with increasing pressure and temperature and decreases with increasing salinity. The water contact angle on the quartz surface under CO₂ geological conditions was probed using experimental and molecular dynamic methods by Chen et al., (2015). Their results indicated that the water contact angle increases with ionic strength that the impact of pressure and temperature is very weak and the dependence of the pressure, temperature and salinity is the same for monovalent and divalent ionic solutions. Sarmadivaleh et al., (2015) demonstrated the impact of pressure and temperature on quartz-CO₂-brine contact angles. In their experiments the contact angle was found to be zero under ambient conditions and seen to increase significantly due to the increase in pressure and temperature, which

results showed that the petrophysical characteristics of the storage reservoir, the rock type and the state of the gas determines the distribution of the residual trapping in the reservoir. The results also showed that the injection of supercritical CO₂ into the reservoir can change the wettability of the reservoir rock towards CO₂ wet conditions.

The results of wettability measurements in calcite is different to some extent compared with sandstone. Arif et al., (2017) investigated the wettability of calcite, which is the representative of limestone rocks, using contact angle measurements. Their results indicated that the calcite is strongly water wet at 0.1 MPa and 25 °C and it turns towards CO₂ wet with an increase in pressure. Under high pressure storage conditions the wettability of the calcite surface changes towards slightly CO₂ wet, recalling a low structural and capillary trapping capability. The low structural and capillary trapping ability caused the CO₂ plume to move upwards easily and increased the risk of leakage. However, the contact angle decreased with the increase in temperature implying that the calcite surface becomes more water wet and additionally, the system moves towards a more CO₂ wet condition at high salinities. The wettability of calcite at storage conditions was investigated by Stevar et al., (2019). They measured both static and dynamic contact angles at temperatures between 298 to 373 K and pressures up to 30 MPa. As observed above, they also found that the calcite surface became strongly water wet at ambient conditions and was seen to change to weakly CO₂ wet at high pressures and low temperatures.

A study conducted by Gershenson et al., (2016) compared the CO₂ trapping in highly heterogeneous reservoirs using Brooks and Corey and Van Genuchten type capillary pressure curves (Onoja and Shariatipour, 2018). They showed that when heterogeneity and hysteresis are represented, the two conventional approaches for defining saturation functions, Brooks and Corey and Van Genuchten represent fundamentally different physical systems. In another work, Gershenson et al., (2017) presented the CO₂ trapping with fluvial architecture and investigated the sensitivity to heterogeneity in permeability and constitutive relationship parameters for different rock types. They suggested that the larger the contrast in permeability between rock types, the larger the CO₂ plume and the larger the rate of capillary trapping and dissolution.

pressure and temperature performed to investigate the impact of natural capillary heterogeneity in a sandstone rock on CO₂ saturation buildup and trapping. Their results showed that a CO₂ plume can be immobilized behind capillary barriers as a continuous phase at saturations higher than would be possible as isolated ganglia. In another study, Krevor et al., (2012) probed the impact of relative permeability on trapping of CO₂ and water in sandstone rocks at reservoir conditions. Their results demonstrated that petrophysical properties of multiphase flow of CO₂/water through sandstone rocks is, for the most part, typical of a strongly water wet system and that analog fluids and conditions may be used to characterize these properties.

The CO₂ plume behavior for a large scale pilot test of geological carbon storage in a saline formation was investigated by Doughty (2010). Their model results suggested that the injected CO₂ plume is immobilized at 25 years. At that time, 38% of CO₂ was in a dissolved form, 59% immobile free phase, and 3% was in a mobile free phase. The plume footprint was seen to be roughly elliptical and extended much farther up-dip of the injection well than down-dip.

In a study by Juanes et al., (2006) the impact of relative permeability hysteresis on geological CO₂ storage was investigated. In their study, they evaluated the relevance of the relative permeability hysteresis when modeling geological CO₂ sequestration processes. They concluded that the modeling of relative permeability hysteresis is required to assess accurately the amount of CO₂ that is immobilized by capillary trapping and is therefore not available to leak. In another work, MacMinn et al., (2010) studied the impact of CO₂ migration on capillary and solubility trapping in saline aquifers. They demonstrated that solubility trapping can greatly slow the speed at which the plume advances and they derived an explicit analytical expression for the position of the nose of the plume as a function of time.

In a recent work by Al-Khdheawi et al., (2017b) the influence of CO₂ wettability on CO₂ migration and trapping capacity in deep saline aquifers was studied. They showed that CO₂ wet reservoirs are most permeable for CO₂; CO₂ migrates furthest upwards and the plume has a candle-like shape. In a water wet reservoir the plume was seen to be more compact. They also investigated the effect of wettability heterogeneity and reservoir temperature on CO₂ storage efficiency in deep saline

capillary and dissolution trapping mechanisms. There are other important affecting parameters on trapping mechanisms and storage efficiency such as: CO₂-rock wettability (Al-khdheewi et al., 2017c), injection well configuration (Al-khdheewi et al., 2017d), CO₂-water injection scenario (Al-khdheewi et al., 2018b; Al-khdheewi et al., 2018c; Al-khdheewi et al., 2018d; Al-khdheewi et al., 2019) and brine salinity (Al-khdheewi et al., 2018e; Al-khdheewi et al., 2017e).

Researchers have proposed different methods for the calculation of relative permeability. A method developed by Purcell (1949) to calculate the pore size distribution of a porous media from mercury injection capillary curves was utilized to calculate the multiphase relative permeabilities. Burdine (1953) presented similar equations to Purcell's method and introduced tortuosity as a function of wetting phase saturation. Another equation was presented by Corey (1954) based on the capillary pressure curves as a power law function of wetting phase saturation. Because of the limitations of Corey's method in presenting relative permeability curves for the wide saturation range, Brooks and Corey (1966) modified the equations for the relative permeability and included the pore size distribution index.

The wettability state of the formation directly impacts on the relative permeability, which also controls the distribution of fluids and plume migration in porous media. In order to consider the five wettability conditions, Al-Khdheewi et al., (2017) used five different relative permeability curves. To create these curves they used the data provided by McCaffery and Bennion, (1974). Then they used the Didger software (Golden Software Inc., 2013, Colorado) to digitize the curves and obtain the relative permeability values as a function of water saturation. They considered S_{wr} less than 15% for CO₂ wet systems and more than 25% for water wet systems. Furthermore, the S_w at which the relative permeability of the wetting phase and the non-wetting phase are equal is considered to be more than 50% for water wet systems and less than 50% for CO₂ wet systems. Finally, the curves were fitted using the Van Genuchten-Maulem model (Van Genuchten, 1980; Mualem, 1976) and fed to the simulators. Table 1 shows the parameters used to make the relative permeability curves.

Table 1: Parameters used to create the relative permeability curves (Al-Khdheewi et al., 2017a).

Wettability Scenario	S_{gr} (Drainage)	S_{wr} (Drainage)	λ (Drainage)	S_{gr} (Imbibition)	S_{wr} (Imbibition)	λ (Imbibition)
SWW	0	0.26	0.78	0.35	0.26	0.58
WW	0	0.25	1.05	0.30	0.25	0.95
IW	0	0.22	1.22	0.25	0.22	1.17
CW	0	0.15	1.41	0.15	0.15	1.51
SCW	0	0.1	1.7	0.10	0.10	1.90

As previously mentioned, the structural and capillary trapping capability of the formation will tend to decrease if the wettability changes towards CO₂ wet. In this regard, Al-Anssari et al., (2017a) and Al-Anssari et al., (2017b) proposed changing the wettability of the formation to strongly water wet through the use of nano fluids in the oil wet reservoirs (Al-Anssari et al., 2018; Al-Anssari et al., 2017; Al-Anssari et al., 2019). In this study, the impact of the temperature on altering the injected CO₂ on wettability and the modification of relative permeability curves for reservoir simulations was investigated for the first time in the literature.

2. Description

2.1 Impact of temperature on the wettability in the Containment and Monitoring Institute geological CO₂ storage project, Calgary, Canada

In this part of the study the impact of temperature on the alteration of wettability using the Containment and Monitoring Institute (CaMI) project in Canada is investigated. The aim of the CaMI project is to test different monitoring techniques for CO₂ storage projects. Usually CO₂ is injected into the formations with a depth greater than 800 metres to meet the supercritical state conditions. In the CaMI field research station, however, since the target formation of Phase 1 is located at a

is 6 metres and there is a uniform geothermal temperature equal to 12.6 °C (Dongas and Lawton, 2014). It should be noted that the temperature difference between the injected CO₂ and the reservoir's geothermal temperature was the reason for choosing this project for investigating the impact of temperature on the wettability alteration of the reservoir rock (Abbaszadeh and Shariatipour, 2020). Bringing to mind that the wettability will change with temperature; thus, the temperature distribution in the reservoir will change the wettability conditions in some parts of the formation and create a wettability profile. Solving the equations related to the temperature distribution in the porous media determines the temperature profile in the reservoir (Equation 1). Since the reservoir is thin and the entire formation is perforated, the one-dimensional solution to Equation 1 with the constant temperature heat source in the wellbore was considered for the temperature profile in the reservoir (Lauwerier, 1955; Barends, 2010). Fig. 1 shows the temperature profile for steady-state heat propagation in the reservoir considering both conduction and convection within the 100 m distance from the wellbore. It should be noted that there are two observation wells at 20 m (Obs. Well 1) and 30 m (Obs. Well 2) from the injection well, respectively in the CaMI project which are demonstrated in Fig. 1.

$$\frac{\delta T}{\delta t} = D \frac{\delta^2 T}{\delta x^2} - u \frac{\delta T}{\delta x} \quad (1)$$

where, D is the thermal diffusivity (m²/s) and u is the thermal convection velocity (m/s). Equation 1 is solved based on the following boundary conditions:

$$T(x, 0) = T_i, \quad x \geq 0$$

$$T(0, t) = T_0, \quad t \geq 0$$

$$\frac{\delta T}{\delta x_{x \rightarrow \infty}} = 0, \quad t \geq 0 \quad (2)$$

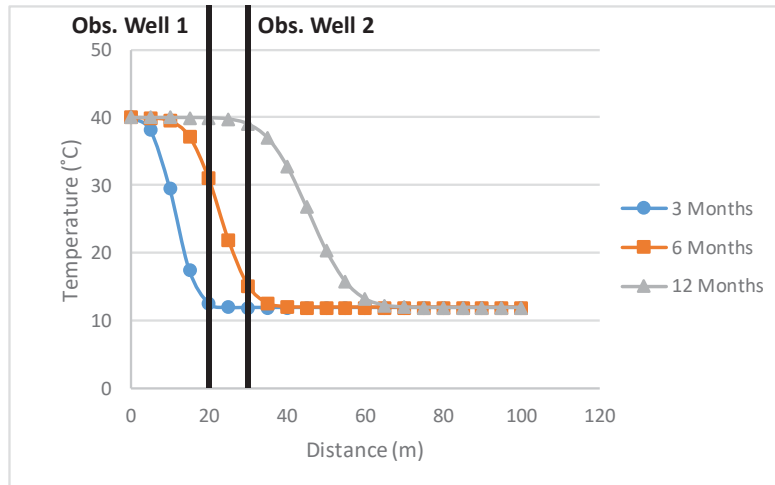


Fig. 1: temperature profile within the reservoir.

As already mentioned, temperature changes in the reservoir change the wettability state of the formation rock. Since changes in the wettability have been investigated by measuring the contact angle (Farokhpoor et al., 2013), using the data provided by Alnili et al., (2018), we can consider the wettability profile in the reservoir with regard to distance from the wellbore in terms of contact angle. Fig. 2 shows the contact angle profile in the reservoir against by the distance from the wellbore.

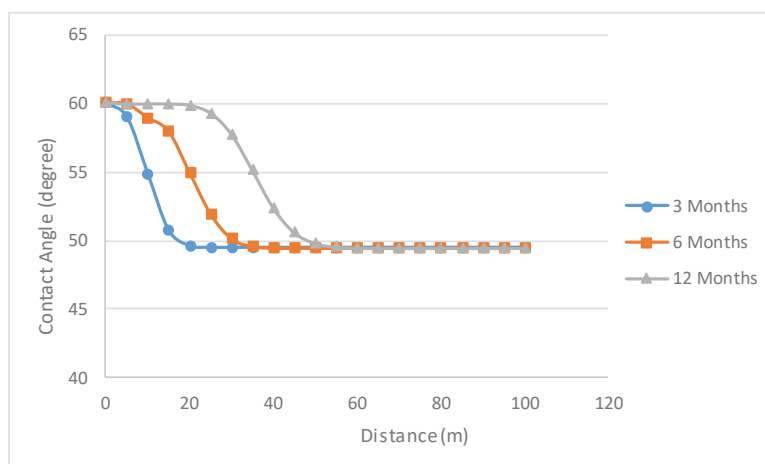


Fig. 2: The contact angle profile in the reservoir.

Fig. 2 shows that as the CO₂ is injected with a higher temperature than the formation geothermal temperature, it changes the wettability conditions of the formation towards a more CO₂ wet

leakage. It should be noted that since the formation in CaMI project is sandstone we used the data by Alnili et al., (2018), which they have studied CO₂/brine wettability of porous sandstone. It has been shown in the literature that the CO₂ contact angle may have different behavior with temperature in different rocks. For example, while contact angle decreases with temperature for quartz, it increases for coal (Al-Yaseri et al., 2016; Arif et al., 2016).

2.2 Impact of temperature on relative permeability curves

The studies above show that the wettability changes with temperature. Additionally, the change in temperature can also change the relative permeability curves. By solving the equations related to the temperature distribution in the reservoir, the temperature profile in a reservoir can be calculated and consequently the wettability profile in the reservoir and different relative permeability areas can be determined.

First the relationship between the temperature and relative permeability needs to be investigated. In this regard, the relative permeability curves which have been created based on the experiments at 102 bars and 19, 31, 38 and 41 °C are considered (Liu et al., 2010; Chen et al., 2014). Then a curve is fitted on the experimental relative permeability of the wetting phase (water) and the resulting curve is considered equal to the Van Genuchten formula at each temperature. Then the Lamda (λ) factor of the Van Genuchten formula is calculated at each temperature (Fig. 3). Now the relative permeability curves can be created at any desired temperature and fed to the simulators.

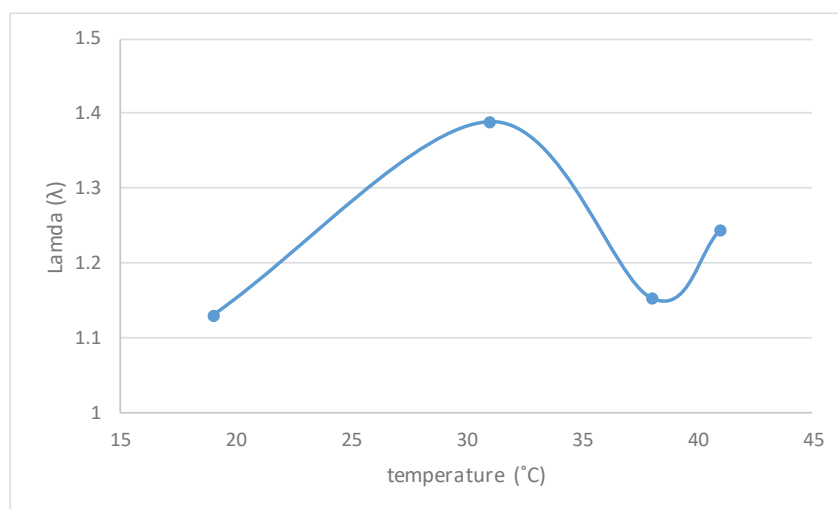


Fig. 3: Lamda (λ) vs Temperature.

is read from Fig. 9 and then the corresponding wetting phase relative permeability curve is created and compared to the relative permeability curve presented by Chen et al., (2014). The results show that this method can accurately predict the wetting phase relative permeability curve (Fig. 4).

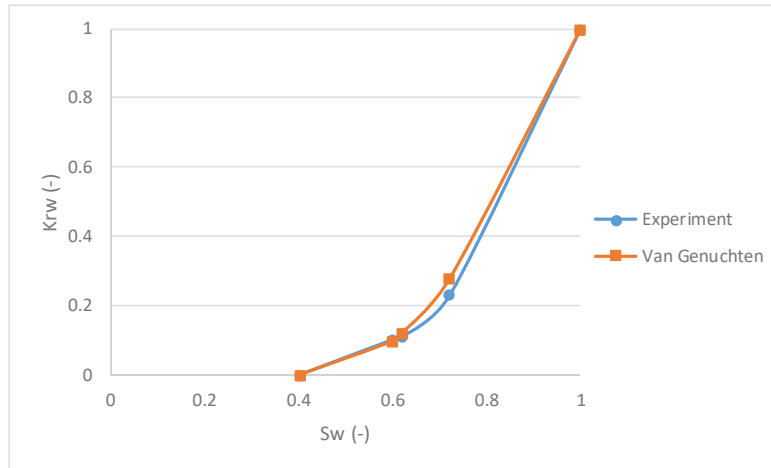


Fig. 4: Prediction of experimental relative permeability values by Van Genuchten formula.

It is already known that if CO₂ is injected with a temperature higher than the temperature of the formation there will be a temperature profile in the reservoir based on the heat conduction and convection of the CO₂ plume in the reservoir. For example, Fig. 1 shows the temperature profile in a typical reservoir when the CO₂ is injected at a temperature of 40 °C and the reservoir temperature is 12.6 °C. This temperature profile will result in a relative permeability profile in the reservoir based on the temperature of the formation at that point.

As discussed, the relative permeability is temperature-dependent, therefore; the relative permeability curve changes continuously in a reservoir where the temperature is changing. In other words, the relative permeability curve will change at the CO₂ plume front in the reservoir and this should be considered in the reservoir simulations. In this regard, the λ factor is read at each temperature from the Lamda-Temperature curve and the corresponding relative permeability curve is created using the Van Genuchten formula. This formula is updated based on the temperature progress in the reservoir. Below, three sample relative permeability curves at three temperatures of 20 °C, 25 °C and 30 °C, respectively, are shown which have been created based on the above method. The λ is equal to 1.15, 1.28 and 1.36 at the temperatures of 20 °C, 25 °C and 30 °C, respectively. The resulting curves are presented in Fig. 5.

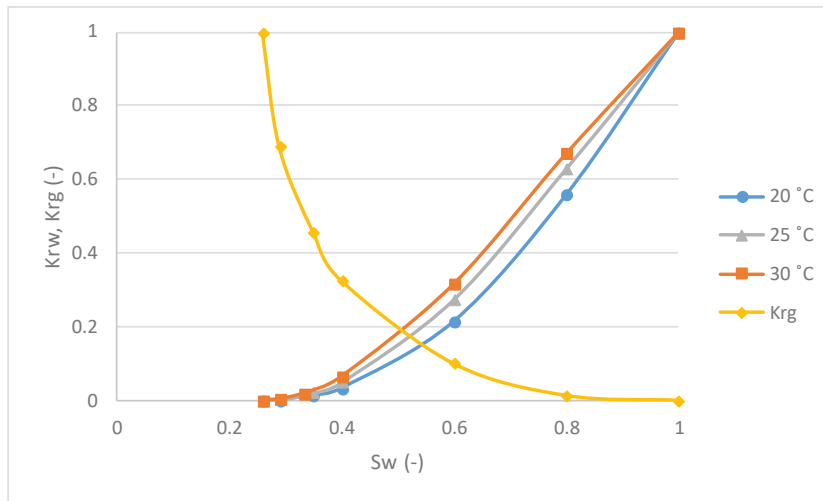


Fig. 5: Relative permeability curves at different temperatures.

3. Results and discussion

In order to see the impact of this phenomena in practice, a synthetic simulation model has been developed based on the reservoir properties of the CaMI project using ECLIPSE 300 through CO2STORE option combined with THERMAL option (Table 4).

Table 2: Input data for simulating CaMI CO₂ injection project

Input Data	Value
Model size (m)	100×40×6
Horizontal Permeability (mD)	0.27
Porosity	0.18
Kv/Kh Ratio	0.1
Depth (m)	300
Rock Compressibility (1/bars)	5.56e-5
Thickness (m)	6

Injection rate (kg/day)	500
Injection Temperature (°C)	40
Injection Pressure (bars)	47

Then the reservoir is divided into four different temperature regions by distance from the wellbore and a relative permeability curve is allocated to each region based on the hypothetical mean temperature of that region (Fig. 6).

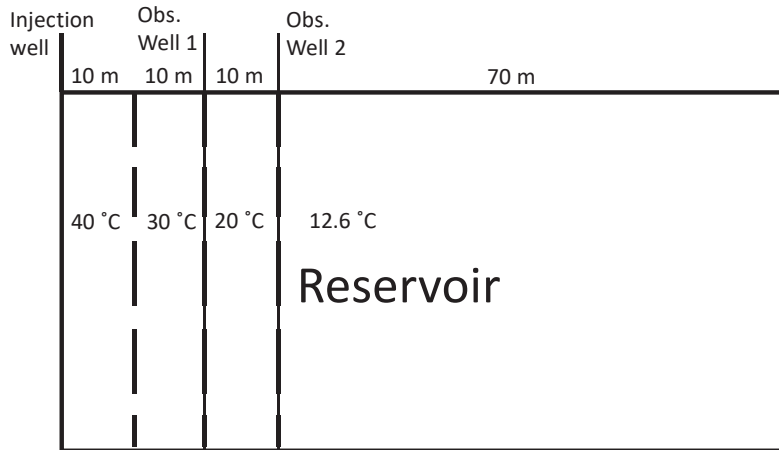


Fig. 6: The division of the reservoir into 4 temperature regions

Because the CO₂ plume moves approximately 30 meters away from the injector in the reservoir during the first six months (Fig. 1), the reservoir would therefore experience different temperature profiles due to this movement. Thus, the reservoir is divided into two main parts. Part one is where there are three sections at 10, 20 and 30 meters away from the injector with temperatures of 40, 30 and 20 °C, respectively. Part two is where the temperature remains the same as the geothermal temperature of the reservoir (12.6 °C). It should be noted that here we do not update the relative permeability curves based on time. The relative permeability curves are only considered based on the location from the wellbore. In this regard, the amount of dissolved CO₂ in the brine for the current method in comparison to considering one relative permeability curve for the reservoir is as follows:

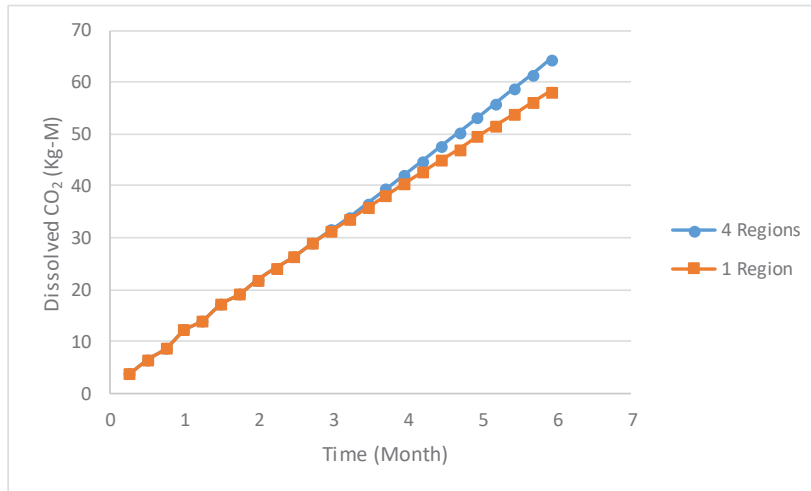


Fig. 7: Comparison between dividing the reservoir into 4 regions and considering the reservoir as one region

Fig. 7 shows that as the reservoir is divided into four regions, the simulation shows a higher CO₂ solubility in the aquifer in comparison to considering the reservoir as one region and allocating one relative permeability to it.

Conclusions

The impact of temperature on wettability in an aquifer and the modification of relative permeability curves based on the temperature variation in aquifers is not well covered in the literature. This study investigated the impact of temperature on wettability distribution in a reservoir and updating the relative permeability curves based on its temperature propagation. The impact of the temperature on relative permeability curves is studied based on the solubility of CO₂ injected into an aquifer using the numerical simulations. The wettability of the formation will change towards a more CO₂ wet condition if the injected CO₂ has a temperature higher than the geothermal temperature of the formation. This increases the risk of leakage and also changes the relative permeability curves as the CO₂ moves through the reservoir, a situation that needs to be considered in reservoir simulations. The results show that the amount of CO₂ dissolution in the aquifer will increase if the relative permeability curves are updated and modified with temperature variation.

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