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Gaseous CO₂ Behaviour during Water Displacement in a Sandstone Core Sample

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Abstract: CO₂ injection into subsurface formations involves the flow of CO₂ through a porous

11 medium that also contains water. The injection, displacement, migration, storage capacity and security 12 of CO₂ is controlled mainly by the interfacial interactions and capillary, viscous, and buoyancy forces 13 which are directly influenced by changes in subsurface conditions of pressure and temperature; the 14 impact of bouncy forces is assumed negligible during this study. In this study, gaseous CO₂ is injected into a water-saturated sandstone core sample to explore the impact of fluid pressure (40-70 bar), 15 16 temperature (29-45 °C), and CO₂ injection rate (0.1-2 ml/min) on the dynamic pressure evolution and displacement efficiency. This study highlights the impact of capillary or viscous forces on the two-phase 17 18 flow characteristics and shows the conditions where capillary or viscous forces become more 19 influential. The results reveal a moderate to considerable impact of the parameters investigated on the differential pressure profile, endpoint CO2 relative permeability (KrCO2max), and irreducible water 20 21 saturation (S_{wr}). Overall, the increase in fluid pressure, temperature, and CO₂ injection rate cause an 22 increase in the maximum and final differential pressures, an increase in the KrCO2^{max}, a reduction in the 23 S_{wr} . S_{wr} was in the range of around 0.38-0.45 while $K_{rCO2^{max}}$ was less than 0.25. The data show a significant 24 influence for the capillary forces on the pressure and production behaviour. The capillary forces 25 produce high oscillations in the pressure and production data while the increase in viscous forces 26 impedes the appearance of these oscillations. The appearance and frequency of the oscillations depend 27 on the fluid pressure, temperature, and CO₂ injection rate but to different extents.

29 **1** Introduction

30 Carbon capture and storage (CCS) is regarded as one of the most promising techniques that can 31 deal effectively with the increasing emissions of anthropogenic CO₂ into the atmosphere due to fossil 32 fuel burning and other human activities (Bachu, 2001; Hangx et al., 2013; Kazemifar et al., 2015). The 33 captured CO2 can be sequestered in deep saline aquifers, depleted or abandoned oil and gas reservoirs 34 (Delshad et al., 2010; Gozalpour et al., 2005; Kaveh et al., 2012), or unmineable coal bed seams (Kaveh 35 et al., 2012; Plug and Bruining, 2007) to enhance recovery from hydrocarbon reservoirs, increase 36 methane production from coal beds, or extract geothermal heat from subsurface formations (Kaveh et 37 al., 2012; Tutolo et al., 2015). Figure 1 presents a summary of the pressure and temperature ranges at 38 which saline aquifers are found underground and highlights that CO₂ can exist in a gaseous, liquid or 39 supercritical phase (Bachu, 2000; Espinoza and Santamarina, 2010; Frailey et al.; Nourpour Aghbash 40 and Ahmadi; Saraji et al., 2014; Sohrabi et al.).



- Figure 1: The pressure and temperature ranges at which saline aquifers are found underground
 (Saraji et al., 2014). This study is conducted under pressure ranged from 40 to 70 bar and temperature
 ranged from 29 to 45 °C.
- 45 During CO₂ injection in subsurface formations, the bulk of the injected CO₂ (as a non-wetting fluid)
- 46 will displace the formation water (as a wetting fluid) in an immiscible displacement (<u>Basbug et al., 2005</u>;
- 47 Herring et al., 2014b). The displacement of the injected CO₂ depends on a number of parameters,

namely, the interfacial interactions (e.g. interfacial tension and wettability), solubility of CO2 in 48 49 formation water, densities and viscosities of fluids present, petrophysical properties of the subsurface 50 formation, injection rate and its duration, and more importantly on the capillary and viscous forces 51 (Cinar and Riaz, 2014; Duan and Sun, 2003; Pentland et al., 2011b; Trevisan et al., 2017). The capillary 52 forces at the CO₂-water interface are of considerable importance in determining the nature of the flow 53 through pores (Roof, 1970). Any change in subsurface conditions of pressure and temperature will have 54 a significant impact on the interfacial interactions (Espinoza and Santamarina, 2010; Liu et al.; Plug and 55 Bruining, 2007; Yang et al., 2007), the viscous forces due to the change in viscosity (Bachu and Bennion, 56 2008b) and the capillary forces. The change in interfacial interactions, and viscous and capillary forces 57 due to the change in underground conditions will have a considerable influence on the capillary 58 pressure, relative permeability (Alkan et al., 2010), pore-scale fluid distribution (Al-Menhali and 59 Krevor, 2014), CO₂ injection, fluid migration, capacity and long-term fate of CO₂ storage in saline aquifers (Levine et al., 2011; Saraji et al., 2013; Wang et al., 2015), CO2-enhanced oil and gas recovery 60 61 processes (Gozalpour et al., 2005; Qi et al., 2010). According to Salimi et al., the change in capillary 62 pressure, due to the change in the operational conditions, can have a direct influence the CO₂-storage 63 capacity and the heat recovery due to its impact on the solubility and density of both CO₂ and water 64 (Salimi et al., 2012). Thus, it is of utmost importance to have a deep insight into the dynamic behaviour 65 of CO₂ under different operational conditions.

66

CO₂ has been used in the oil industry for a long time, in particular, to increase productivity through Enhanced Oil Recovery (EOR), and extensive research has been undertaken describing multi-phase flow properties of CO₂-oil systems (<u>Bahralolom et al., 1988</u>). On the other hand, much less laboratory investigations have been done for CO₂-water (brine) systems (<u>Perrin and Benson, 2010</u>). Those published have mainly focused on CO₂ wettability (<u>Al-Menhali and Krevor, 2014</u>; <u>Bikkina, 2011</u>; <u>Farokhpoor et al., 2013a</u>; <u>Kaveh et al., 2012</u>; <u>Sakurovs and Lavrencic, 2011</u>; <u>Saraji et al., 2013</u>), CO₂-water (brine) interfacial tension (<u>Aggelopoulos et al., 2010</u>; <u>Bachu and Bennion, 2008b</u>, 2009; <u>Busch and</u> Müller, 2011; Chiquet et al., 2007; Li et al., 2012; Yu et al., 2012), relative permeability (Bachu, 2013;
Krevor et al., 2015; Liu et al.; Perrin et al., 2009) and capillary pressure (Busch and Müller, 2011; Pini et al., 2012; Plug and Bruining, 2007). Cinar and Riaz showed that much of the research has been directed to investigate the fluid properties rather than studying the multiphase flow properties of the CO₂-water systems (Cinar and Riaz, 2014).

79 The limited investigations of the multiphase flow characteristics of CO₂-water (brine) systems have 80 involved laboratory experiments (Jobard et al., 2013), computational modelling (Jobard et al., 2013; Ma 81 et al., 2013; Xu et al., 2011), and field scale projects (Wang et al., 2015). The CO2-water (brine) 82 investigations included core flooding displacements performed at liquid, supercritical and gaseous CO2 83 conditions. Current literature survey of the CO₂-water (brine) multiphase flow experiments showed that most of these experiments were supercritical (Sc) CO₂-brine (water) displacements studies, which 84 85 were performed on various porous media such as core samples (Berg et al., 2013; Edlmann et al., 2013), 86 micromodels (Cao et al., 2016), and packed beds of glass beads (Song et al., 2012; Suekane et al., 2005). 87 In these studies related to supercritical CO₂ migration, researchers have examined various parameters 88 such as relative permeability curves (Berg et al., 2013; Chang et al., 2013; Krevor et al., 2013; Suekane et al., 2005; Suenaga and Nakagawa, 2011), capillary pressure curves (Herring et al., 2014a; Wang et al., 89 2013), CO₂ residual saturation and distribution (Alemu et al., 2011; Chang et al., 2013; Herring et al., 90 91 2014a; Pentland et al., 2011a; Saeedi et al., 2011; Suekane et al., 2005), heterogeneity impact (Ott et al., 92 2015; Perrin and Benson, 2010; Shi et al., 2011; Wang et al., 2013), water displacement efficiency (Cao et 93 al., 2016), mass transfer (Berg et al., 2013), and formation dry-out (Ott et al., 2011). Some liquid (L) CO₂-94 water (brine) core flooding displacements were conducted to investigate the multiphase flow 95 characteristics of CO₂-water-porous media (Manceau et al., 2015), CO₂ residual saturation and distribution (Alemu et al., 2011), and pore-scale heterogeneity (Zhang et al., 2011). 96

On the other hand, very scarce data was found regarding gaseous (G) CO₂ injection into water
(brine) saturated porous systems (<u>Islam et al., 2013</u>; <u>Jiang et al., 2017</u>; <u>Lassen et al., 2015</u>; <u>Yu et al., 2014</u>).

99 Even though liquid and supercritical CO₂ injection is more efficient, the dynamic behaviour of gaseous 100 CO₂ in reservoir rock is necessary information, particularly considering that many potential saline 101 storage aquifers are within temperature and pressure conditions of the gaseous CO₂ phase (Figure 1) 102 and that any leakage of CO₂ from deeper storage would inevitably result in a phase change to a gaseous 103 CO₂ state (Edlmann et al., 2016; Miocic et al., 2016). The existing GCO₂-water experiments were 104 designed to investigate the crossover zone of flow regimes, impact of capillary number, CO₂ injection 105 rates and permeability on displacement efficiency. Islam et al. conducted GCO₂-water experiments at 1 106 bar and 25 °C using a vertical Hele-Shaw cell filled with micro-beads to investigate the crossover zone 107 from capillary to viscous to fracture fingering. They observed that all the three fingering patterns can 108 occur in the cell but at different heights (Islam et al., 2013). Jiang et al. performed both immiscible and 109 miscible drainage GCO₂-water displacements inside a packed bed filled with quartz glass beads to have 110 a better understanding of the two-phase flow characteristics inside porous media. The experiments 111 were conducted at CO₂ injection rates varying from 0.01 to 3 ml/min and at 60 bar and 24.85 °C. They 112 observed that: (I) at low CO₂ injection rates, the CO₂ dissolution increases; (II) the increase in glass beads 113 diameter (i.e. higher permeability) leads to a decrease in the capillary forces (Jiang et al., 2017). Yu et al. 114 conducted immiscible drainage GCO₂-water displacements at 60 bar and 24.85 °C inside a packed bed 115 of glass beads (0.2 mm diameter) to study the impact of the capillary number on displacement efficiency. They noticed that the increase in the capillary number, when it is between 10⁻¹¹ and 10⁻¹⁰, 116 117 results in a sharp reduction in the residual water saturation as a result of increasing the impact of the 118 viscous forces (Yu et al., 2014).

Despite the considerable research on the CO₂–water (brine) systems and its practical importance, the analysis of the pressure data in core flooding has been widely overlooked (<u>Rezaei and Firoozabadi</u>, 2014). To the authors' best knowledge, there is no detailed investigation into the dynamic pressure evolution and displacement efficiency of gaseous CO₂ during its injection into a water saturated core sample. In this paper, laboratory dynamic drainage experiments were performed by injecting pure CO₂ into the deionised water-saturated sandstone core sample to investigate the impact of fluid pressure, temperature, and CO₂ injection rate on the differential pressure profile, water production, and endpoint effective and relative permeabilities of CO₂. This study also highlights the impact of capillary and viscous forces on the pressure and production data as well as shows the conditions at which capillary or viscous forces become more influential. During these dynamic displacements, the transient pressure at the inlet and outlet sides of the core and the transient outflow rates of water and CO₂ were measured and analyzed. The endpoint water saturations of CO₂ and water were also calculated.

131 2 Materials

A sandstone core sample from the Guillemot A Field in the North Sea was used to perform the unsteady state GCO₂-water drainage experiments. The core sample has a diameter of 2.54 cm and a length of 7.62 cm. The average porosity and absolute water permeability of the core sample were about 14% and 15.8 millidarcys, respectively. This study is one in a series, thus the core sample description, the experimental setup and the CO₂-water displacement procedures can be seen in our recent publication (Al-Zaidi et al., 2018).

138

3 **Results and discussion**

To gain a deep insight into the dynamic behaviour of GCO₂-water drainage displacements under various fluid pressure, temperature, and injection rate conditions; the inlet and outlet pressure, CO₂ and water out flowrate, the irreducible water saturation and endpoint effective and relative permeabilities of CO₂ were measured and analyzed.

In this study, the difference between the pressure transducer readings at the inlet and outlet sides of the core sample has been used to calculate the differential pressure. The differential pressure during horizontal CO₂ injection is largely influenced by the capillary and viscous forces. The capillary forces are controlled mainly by the CO₂-water interfacial tension, contact angle (i.e. wetting status), pore diameter and geometry (Alkan et al., 2010; Bikkina et al., 2016; Chatzis and Morrow, 1984; Fulcher Jr et al., 1985). The wetting status plays an important role in determining the imbibition and the distribution of the wetting and non-wetting phases inside the porous media (Chalbaud et al., 2007; Espinoza and

Santamarina, 2010). The capillary forces, which are responsible for the entrapment of one phase by 150 151 another during immiscible displacements in porous media (Akbarabadi and Piri, 2013; Chatzis and Morrow, 1984), arise from the presence of the interface between the immiscible fluids (Bikkina et al., 152 153 2016) and significantly dominate the multiphase flow, especially in low permeability rocks and 154 fractured reservoirs (Schembre and Kovscek, 2003). On the other hand, the viscous forces are controlled 155 mainly by the viscosity of both displacing and displaced fluids, the fluid velocity in the pores, the 156 amount of each fluid (i.e. saturation) in the pore, and the core sample properties (e.g. frontal area, permeability, and length). Espinoza and Santamarina (Espinoza and Santamarina, 2010) proposed the 157 158 following equation to account for the impact of the capillary and viscous forces on the differential 159 pressure as follow:

160
$$\Delta P = P_{CO2} - P_{water} = 4 \frac{\sigma_{CO2-water} COS\theta}{d} + v \frac{32 L}{d^2} \left(\frac{l_{CO2} \mu_{CO2} + l_{water} \mu_{water}}{L} \right)$$
(1)

161 Where ΔP is the differential pressure across the core sample (Pa). P_{CO2} and P_{water} are the pressures 162 of CO₂ phase and water phase, respectively. $\sigma_{CO2-water}$ is the CO₂-water interfacial tension (mN/m), θ the 163 contact angle, d (m) the diameter of the largest effective pore (Chiquet et al.; Chiquet et al., 2007; 164 Farokhpoor et al., 2013b; Han et al., 2010), *L* (m) the length of the core sample, *l* (m) the length of CO₂ or water phase inside the core sample, v (m/s) the fluid velocity in the pores, and μ (Pa·s) the viscosity 165 of the fluids. The first term of Eq.1 refers to the Young-Laplace equation, which accounts for the 166 167 capillary forces, while the second term refers to the Poiseuille's equation (Espinoza and Santamarina, 168 2010; Li, 2015), which account for the viscous forces. For small injection rate and high viscosity contrast 169 conditions the impact of viscous forces can be neglected, thus Eq.1 can be reduced to the Young-Laplace 170 equation (Li, 2015) as follows:

171
$$\Delta P = P_{CO2} - P_{water} = 4 \frac{\sigma_{CO2-water} \cos \theta}{d}$$
(2)

172 The Young-Laplace equation is used to determine the critical pressure point, which is the 173 differential pressure required for the displacing fluid to enter the core sample for the first time. The non-wetting fluid cannot enter the core sample unless its pressure becomes higher than the critical
pressure point (<u>Han et al., 2010</u>).

In this study, the experimental results have been categorized into two main sections. The first section presents and discusses the impact of the experimental fluid pressure, temperature and CO_2 injection rate on the differential pressure profiles while the second section deals with the impact of the parameters investigated on the endpoint CO_2 effective (relative) permeability and irreducible water saturation.

It should be noted that during this study, the term low and high-fluid pressure refers to the experiments conducted at pressures less and higher than 50 bar, respectively. The low and high temperature refers to the experiments performed at less or higher than 33 °C, respectively. The low, medium and high injection rates refer to the experiments performed at injection rate ranging from 0.1 to 0.2 ml/min, from 0.3 to 0.6 ml/min, and from 1 to 2 ml/min, in sequence. The corresponding time refers to the time required to reach the maximum-differential pressure at the start of the experiment. The quasi-differential pressure refers to the differential pressure at the end of the experiment.

188 3.1 Differential Pressure Profile of GCO₂-Water Drainage Displacements

To investigate the effect of fluid pressure, experimental temperature, and CO₂ injection rate on the differential pressures, series of GCO₂-water displacements were performed at various fluid pressures (from 40 to 70 bar), experimental temperatures (29-45 °C) and CO₂ injection rates (0.1-2 ml/min).

192 3.1.1 Effect of Fluid Pressure on the Differential Pressure Profile of GCO₂-Water Drainage 193 Displacements

Figure 2 presents the impact of increasing fluid pressure on the differential pressure profile of GCO₂-water drainage displacements. A number of trends are identifiable: Firstly, the differential pressure profile at all fluid pressures is characterized by a high initial increase, immediately followed by a steep rapid reduction and then followed by a quasi-differential pressure. Secondly, there are 198 multiple oscillations of these cycles. The frequency of these oscillating cycles increases as fluid pressure 199 increases along with a rise in the values of the maximum and quasi-differential pressures.

200 The high initial increase in the differential pressure can be related to the capillary pressure. The 201 following reduction in the differential pressure profile reflects the impact of the reduction in both 202 capillary forces and viscous forces according to Eq.1. The injection of gaseous CO₂ into the core sample 203 generates the initial increase in differential pressure to overcome the capillary entry pressure for the 204 invasion of the gaseous CO₂ (<u>Chang et al., 2013</u>). The reduction in the capillary forces can be associated 205 with the reduction in the pore resistance to CO₂-water interfaces as the number of pores opened by CO₂ 206 is increased (Kwelle, 2017). This agrees very well with Kwell's finding, who noticed a high reduction 207 in the differential pressure profile as the CO₂-water interface is displaced out of microcapillary tubes (Kwelle, 2017). The reduction in the viscous forces can be related to the combined effect of the dynamic 208 209 change in relative permeability of gaseous CO₂ and water and the high rate replacement of a more 210 viscous fluid (water) with a less viscous fluid (CO₂) (<u>Chang et al., 2013</u>). Replacing water by CO₂ at a 211 high rate can be linked to (a) the high mobility ratio due to the high viscosity contrast and (b) gas 212 expansion effects which generate an increase in volumetric CO_2 injection rate inside the core sample.

213 • The gas expansion can, in turn, be related to the density change of the injected CO₂ due to the 214 temperature difference between inside the water bath (i.e. 29 to 45 °C depending on the experimental 215 conditions) and outside it (room temperature 18-20 °C). The density of the injected CO₂ varies as the 216 CO₂ enters the water bath dependant on the injection rate, fluid pressure and the temperature difference 217 from the pump to the sample. The density ratio (d_r) suggested by Perrin and Benson (Perrin and Benson, 2010) has been used to calculate the injection rate inside the core sample. For instance, at an 218 219 experimental pressure of 40 bar, an injection rate of 1 cm3/min at 20 °C becomes 1.7522 cm3/min at 33 220 °C. However, at an experimental pressure of 70 bar and the same injection rate and temperature 221 conditions, it becomes 5.281cm³/min.

222
$$d_r = \frac{P_1 T_2 Z_2}{P_2 T_1 Z_1}$$
(3)

224 Figure 2 reveals that the differential pressure profiles are characterized by multiple differential 225 pressure (PD) oscillations. The appearance of these PD oscillations can be related to the impact of the capillary forces at the trailing end of each CO₂-water slug during CO₂ flooding (Nutt, 1982) or the 226 227 capillary end effects. According to Nutt, the impact of the capillary forces at the trailing end of the CO2-228 water slug is governed by the wetting status of the injected fluid. If a non-wetting fluid (e.g. CO₂) is 229 injected, then the capillary forces will work in an opposite direction to the applied viscous forces. Thus, 230 as water depletion is progressed, the applied viscous forces will drop until they become less than the capillary forces. Upon reaching this point, the flow of the non-depleted capillaries is potentially blocked 231 232 by the capillary forces (Nutt, 1982). This blockage occurs due to a re-imbibition process of the wetting 233 phase inside the core sample, which was noticed by Hildenbrand et al. (Hildenbrand et al., 2002). 234 Hildenbrand et al. observed that the re-imbibition process occurs when the excess pressure in the non-235 wetting phase declines after the gas breakthrough (Hildenbrand et al., 2002), as shown in Figure 3. This 236 re-imbibition process occurs in a progressive manner starting with the smallest pores and continuing 237 to the larger pores, leading to the successive loss of the interconnected flow-paths, which, in turn, leads 238 to a progressive decline in the non-wetting phase relative permeability. Finally, when the last 239 interconnected flow-path for the non-wetting phase is blocked, the permeability of the non-wetting phase will drop to zero (Hildenbrand et al., 2002). According to Hildenbrand et al., this re-imbition 240 process can result in a residual water saturation when certain-gas filled pores become isolated a result 241 242 of interrupting the flow pathways. The maximum differential pressure required to open the flow paths 243 again can be used to determine the largest effective pore radius and, hence, the sealing efficiency of the 244 rock (Hildenbrand et al., 2002).

Therefore, since our core sample is water-wet, the pressure of the injected CO_2 had to build up to a certain level to overcome the capillary forces that blocked the CO_2 outflow rate (<u>Nutt, 1982</u>). Due to the high compressibility nature of the gaseous CO_2 , the injected CO_2 will accumulate inside the core

sample and the connections pipes until the differential pressure becomes high enough to overcome the 248 capillary forces. Once the blocked capillaries are opened to flow, the cumulative CO₂ will expel the 249 liquid drops that block the pores out of the core sample quickly; the rate of expulsion is expected to 250 251 increase with the fluid pressure. The development of this phenomenon is highly influenced by the core 252 sample properties and the injection rate due to their direct impact on viscous and capillary forces. As a 253 result, this phenomenon is expected to be reduced when the injection rate, i.e. viscous pressure drop, 254 becomes high enough to overcome the capillary forces (Nutt, 1982). However, due to the cyclic 255 reduction of the viscous pressure drop (i.e. viscous forces) to the level that becomes insufficient to 256 overcome the capillary forces, this phenomenon of oscillations can occur frequently.

257 On the other hand, since the GCO₂-water displacements are strongly influenced by the capillary 258 end effects and viscous instabilities (<u>Müller, 2011</u>), it might be suggested that the appearance of the 259 oscillations is due to the impact of capillary end effects. The capillary end effects occur at both inlet and 260 outlet faces of the core sample, but its impact becomes more severe at the outlet face. According to 261 Müller, the capillary end effects can never be entirely prevented but can be corrected for (<u>Müller, 2011</u>). 262 The impact of capillary end effects and viscous instabilities can be reduced when the following scaling 263 coefficient proposed by Rapoport and Leas for stabilized floods becomes greater than one.

$$264 \qquad Lu\mu \ge 1$$

(4)

where *L* is the length of the medium (cm), *u* the Darcy velocity (cm/min), and μ the displacing phase viscosity (cp) (Fathollahi and Rostami, 2015). The scaling coefficients for the 40, 50, and 70 bar displacements are 0.0773, 0.0844, and 0.285, respectively. The scaling coefficients increased significantly as the fluid pressure increased from 40 and 50 bar to 70 bar, which indicates a reduction in the impact of capillary end effects with increasing fluid pressure. However, since the data from Figure 2 reveal an increase in the frequency of the oscillations with increasing fluid pressure, this indicates that the capillary end effects are not responsible for the PD oscillation phenomenon. In addition, the disappearance of the oscillations at lower injection rates as shown in Figure 7 further supports the idea
that the oscillations are not because of the capillary end effects.

274 Figure 2 also shows that increasing fluid pressure leads to an increase in the rate of the differential 275 pressure (PD) oscillations along with increases in the values of the maximum and quasi-differential 276 pressures and a reduction in the corresponding time (the time required to reach the maximumdifferential pressure at the start of the experiment). For illustration, it can be seen that as the fluid 277 278 pressure increased from 40 to 50 bar, the rate of the PD oscillations increased by around 33% and the 279 maximum-differential pressure increased by about 2.50%. The quasi-differential pressure was constant 280 at around 1 bar. The corresponding time declined by approximately 17%. However, as the fluid 281 pressure increased from 50 to 70 bar, the PD oscillations substantially increased by 225%, the maximumdifferential pressure raised by around 9% and the quasi-differential pressure increased by 165%. The 282 corresponding time dropped considerably by around 78%. The high reduction in the corresponding 283 284 time with increasing fluid pressure can be related mainly to the increase in gaseous CO₂ density and 285 the injection rate inside the core sample due to the expansion effects. As gaseous CO₂ becomes denser, 286 it requires lesser time to be compressed to the required pressure.

287 The increase in the maximum and quasi-differential pressures with increasing fluid pressure can 288 be related mainly to the magnitudes of both viscous and capillary forces. According to Eq.1, as the fluid 289 pressure increases the viscous forces increase [due to the increase in CO₂ viscosity and the injection rate 290 inside the core sample due to expansion impact], while the capillary forces decrease [because of the 291 reduction in the CO₂-water interfacial tension (IFT) (Georgiadis et al., 2010) and the increase in the 292 contact angle (Banerjee et al., 2013) due to increasing CO2 solubility (Bennion and Bachu; Yang et al., 2007)]. Thus, the increase observed in the differential pressures is the net result of the increase in the 293 viscous forces and the reduction in the capillary forces. Reducing capillary forces with increasing 294 295 pressure is expected to cause a reduction in the extent of differential pressure increase.

The increase in the PD oscillations means the frequency of liquid drops expelled out of the core sample is increased. This can be associated mainly with the reduction in the capillary forces and the increase in gas density with increasing pressure. Increasing the gas density and reducing capillary forces mean less time was required to reach a differential pressure value which was sufficient to overcome the capillary forces; thus, increasing the frequency of the PD oscillations.





Figure 2: Effect of fluid pressure on the differential pressure profile of GCO₂-water displacements
 conducted at 0.4 ml/min and 33 °C.



304 305

Figure 3: Re-imbibition process in fine-grained rocks (schematic re-imbibition); (A) drainage, (B) initially water-saturated sample, (C) gas breakthrough, (D) re-imbibition (<u>Hildenbrand et al., 2002</u>).

307

3083.1.2Effect of Temperature on the Differential Pressure Profile of GCO2-Water309Displacements

310 Figure 4 presents the impact of increasing experimental temperature on the differential pressure 311 profile. The results demonstrate that the increase in the experimental temperature has a significant 312 impact on the differential pressure profile. Firstly, increasing the temperature increases the frequency 313 of the PD oscillations. At an experimental temperature of 29 °C, the differential pressure profile 314 experienced no oscillations. However, as the temperature increased to 31 °C, the oscillations appeared 315 for the first time. A further increase in the temperature to 33 °C caused the number of oscillations to 316 increase by double. Secondly, the increase in the temperature prompts an increase in the magnitude of 317 the maximum-differential pressure. The quasi-differential pressure was almost constant due to the slight impact of both capillary forces and viscous forces at the end of core flooding. 318

319 The appearance and frequency of the PD cycles with increasing temperature have three potential 320 explanations. The first potential reason behind the onset of the oscillations and their frequency is the 321 increase in the capillary forces despite the slight increase in viscous forces under these conditions. The 322 increase in temperature leads to an increase in the CO₂-water IFT (<u>Iglauer et al., 2012</u>) with a reduction 323 in the contact angle (Yang et al., 2007) due to the decline in the CO₂ solubility (Bennion and Bachu; Yang 324 et al., 2007) as well as a slight increase in CO₂ viscosity, and a slight increase in CO₂ injection rate inside 325 the core sample due to expansion effect. For illustration, as the experimental temperature increased 326 from 29 to 31 °C, CO₂-water IFT increases from to 42.9 to 44.42 mN/m, CO₂ viscosity increases very 327 slightly from 16.72 to 16.755 × $[10^{-6}(Pa \cdot s)]$ and CO₂ injection inside the core sample increased from around 0.45 to 0.46 ml/min. However, a further increase in the experimental temperature to 33 °C 328 329 caused the CO₂-water IFT to decrease to 34.1 mN/m (Bachu and Bennion, 2008a), CO₂ viscosity to 330 increase to $16.805 \times [10^{-6}(Pa \cdot s)]$ and CO₂ injection to increase to 0.466 ml/min.

The second possible reason might be related to the fluctuating behaviour in the CO₂-water IFT when the experimental temperature is around the critical point (<u>Bennion and Bachu</u>), as shown in Figure 5. The third potential reason is that the PD oscillations might occur because of increasing temperature which results in a quicker increase in the movement of CO₂ molecules. This is because each
individual molecule has more energy as it becomes hotter, according to the Kinetic molecular theory
(Physics, 2017). A high energetic CO₂ molecule might open the closed flow path, due to the increase in
capillary forces, quicker.

The results indicate that for the sandstone core sample (from the Guillemot A field, North Sea) used in the experiment and under the aforementioned experimental conditions, the onset temperature point of the oscillations is around 31 °C. The characteristics of the sandstone sample, e.g. pore size distribution, play a key role in the onset of the PD oscillations phenomena as they have a direct influence on the magnitude of the capillary forces as illustrated by Young-Laplace law (Eq.2).

343 The data also reveals that as the experimental temperature increased from 29 to 31 °C, the 344 maximum-differential pressure increased by around 12.5% (from 0.72 to 0.81 bar) and the 345 corresponding time dropped by around 9.1% (from 12.1 to 11 min). However, increasing the temperature from 31 to 33 °C caused the differential pressure to decline slightly by 1.23% (from 0.82 to 346 347 0.81 bar) and the corresponding time dropped by 30% (from 11 to 7.7 min). The increase and decrease 348 in the maximum-differential pressure can be related mainly to the increase or decrease in the capillary 349 forces due to CO₂-water IFT, as stated above. The highest reduction in the corresponding time occurred 350 as the temperature increased to 33 °C. This can be related to the highest reduction in the CO2-water IFT 351 (Bennion and Bachu), as shown in Figure 5.







Figure 4: Effect of temperature on the differential pressure profile of GCO₂-water displacements conducted at 50 bar and 0.4 ml/min.





356 Figure 5: Interfacial tension for CO₂-Pure Water Systems adopted from (<u>Bachu and Bennion, 2008b</u>).

To further investigate the effect of the temperature on the differential pressure profile, and especially on the PD oscillations, additional GCO₂-water displacement experiments were conducted under a high-pressure of 70 bar and higher temperature conditions.

The data from Figure 6 shows that increasing the experimental temperature by 12 degrees (from 361 33 to 45 °C) at a high-pressure caused no further increase in the rate of the PD oscillations. Yet, it 362 instigated a very slight increase in the maximum and quasi-differential pressures with a small reduction 363 in the corresponding time. The maximum differential pressure increased by only 4.2% (from 0.854 to 0.89 bar) and the quasi-differential pressure by 4.81% (from 0.208 to 0.218 bar). The corresponding time
declined by around 17% (from 1.8 to 1.5 min).

- The data showed no further increase in the PD oscillations occurred when there are no fluctuations in the IFT as the temperature increased from 33 to 45 °C, as shown in Figure 5. This suggests that the IFT fluctuations might have highly influenced the frequency of PD oscillations.
- 369 The increase in the maximum and quasi-differential pressures can be related to the increase in the 370 capillary forces (because of the increasing CO2-water interfacial tension and the reducing contact angle 371 (Yang et al., 2007)), and the slight increase in the viscous forces (because of the increasing injection rate). 372 The magnitude of the viscous forces might have slightly declined because of the slight reduction in CO2 373 viscosity with increasing temperature. For illustration, as the experimental temperature increased from 374 33 to 45 °C, the CO2-water IFT increases from around 29.15 to around 33.4 mN/m (Bennion and Bachu), 375 and the CO₂ injection rate inside the core sample increased from 1.315 to 1.748 ml/min but the viscosity 376 decreases from 20.743 to $19.05 \times [10^{-6}(Pa \cdot s)]$.



Figure 6: Effect of temperature on the differential pressure profile of GCO₂-water displacements
 conducted at 70 bar and 0.4 ml/min.

380 3.1.3 Effect of CO₂ Injection Rate on the Differential Pressure Profile of GCO₂-Water Core 381 Floodings

Figure 7, Figure 8 and Figure 9 show the impact of increasing CO₂ injection rate on the differential pressure profile. For Figure 8, the experiments conducted at higher injection rate (2 ml/min) lasted shorter than those conducted at lower injection rate (1 ml/min) to explore the impact of injection volumes on the displacement efficiency. The results reveal that increasing the injection rate has a significant impact on the differential pressure profile, mainly at early stages of core flooding. The data reveal a number of important observations (A-E).

388 A) The data show that the higher the injection rate, the higher the maximum differential pressure 389 is. However, increasing the injection rate caused a slight increase in the quasi-differential pressure; the 390 corresponding time decreased at low injection rates and increased at high injection rates. For 391 illustration, as the CO2 injection rate increased from 0.1 to 0.2 ml/min, the maximum-differential 392 pressure increased by 33.54% (from 0.161 to 0.215 bar), and the quasi-differential pressure by 5.88% 393 (from 0.068 to 0.072 bar) while the corresponding time reduced by almost half (from 13.5 to 6.5 min). 394 However, as the CO₂ injection rate increased from 1 to 2 ml/min, the maximum-differential pressure 395 increased by around 44% (from 0.833 to 1.201 bar), the quasi-differential pressure increased by around 396 15% (from 0.254 to 0.291 bar), and the corresponding time increased by 12% (from 3.3 to 3.7 min). The 397 increase in the corresponding time at high injection rates despite the increase in the CO₂ injection rate 398 can be related to the high increase in the magnitude of the maximum-differential pressure as well as 399 the low-density nature of the gaseous CO₂. Since the injected gaseous CO₂ was at low pressure (40 bar), 400 it needed a longer time to reach the higher maximum-differential pressure of 1.201 bar during the 2 401 ml/min-displacement.

B) The data from Figure 7 and Figure 8 reveals that as the injection rate increased by tenfold (from 0.1 to 1 ml/min, and from 0.2 to 2 ml/min), the quasi-differential pressure increased by only around fourfold, (from 0.068 to 0.254 bar, and from 0.072 to 0.291 bar). This might be related to a potential

increase in the relative permeability with increasing injection rate (<u>Akbarabadi and Piri</u>; <u>Chang et al.</u>,
2013) that leads to a reduction in the viscous pressure drop.

407 C) The data previously shown in Figure 2 reveals that the differential pressure profile of the 40 408 bar-experiments is characterized by PD oscillations at 0.4 ml/min CO₂ injection rate. Surprisingly, the 409 data from Figure 7 and Figure 8 reveal no PD oscillations at lower and higher CO₂ injection rates. The 410 disappearance of the PD oscillations at higher injection rates (e.g. 1-2 ml/min) can be related to the high 411 increase in the pressure drop due to viscous forces. Thus, the viscous forces impeded the capillary 412 forces, which are responsible for the observed PD oscillations phenomenon (Nutt, 1982). On the other 413 hand, at lower CO₂ injection rates (e.g. 0.1 to 0.2 ml/min), CO₂ might flow through preferential inlet and 414 outlet pores (Gunde et al., 2010) that are characterized by low resistance to flow and by less capillary 415 forces. Consequently, CO₂ does not need to pass through the smallest channels that are characterized by higher resistance to CO₂ flow and higher capillary forces, hence avoiding the impact of the capillary 416 417 forces that cause the oscillations.

D) To look in detail at the unexpected results regarding the appearance and disappearance of the PD oscillations and the impact of CO₂ injection rate on the differential pressure profile, further experiments were conducted at 40 bar and over a more detailed range of injection rates, as shown in Figure 9. It should be noted that the 0.4 ml/min GCO₂-water displacement is repeated to make sure that the observations were not an experimental error.

The results from Figure 9 show clearly that the PD oscillations occurred only at 0.4 ml/min for the experiments conducted at a low pressure of 40 bar. Overall, the data confirm that the increase in the injection rate produces an increase in the maximum-differential pressure and a reduction in its corresponding time for this range of injection rates. The quasi-differential pressure reduced slightly due to the potential increase in the relative permeabilities (<u>Akbarabadi and Piri</u>; <u>Chang et al.</u>, 2013).

428 The data from Figure 9 can be divided into two groups. The first group includes the experiments 429 conducted at a CO₂ injection rate of 0.3 and 0.4 ml/min while the second group involves the experiments

430 performed at 0.5 and 0.6 ml/min. As the CO₂ injection rate increased for the first lower injection rate group, the maximum-differential pressure was almost constant at around 0.76 bar, but the 431 corresponding time reduced by 25% (from around 20 to 15 min). The second higher injection rate group 432 433 is characterized by a constant maximum-differential pressure of 0.938 bar and a constant corresponding 434 time of 6.5 min. Thus, the data reveals that shifting the CO₂ injection rate to the second group caused 435 the maximum-differential pressure to increase by 23.42% and the corresponding time to reduce by around 57%. The increase in the maximum-differential pressure associated with shifting the CO₂ 436 injection rate might be related to the properties of the core sample. It might have occurred because as 437 438 the injection rate increased from the first to the second group, the maximum-differential pressure had 439 to further increase to open new preferential flow paths for the injected CO2 (Gunde et al., 2010). The 440 nearly constant maximum-differential pressure for each group might indicate a minimal impact for the 441 viscous forces on the differential pressure at low pressures. It indicates also that the expected increase 442 in the maximum-differential pressure due to increasing injection rate is reduced by the potential increase in the relative permeability due to the increasing injection rate (Akbarabadi and Piri; Chang et 443 444 al., 2013).



446 Figure 7: Effect of CO₂ injection rate on the differential pressure profile of GCO₂-water 447 displacements conducted at 40 bar and 33 °C.





Figure 8: Effect of CO₂ injection rate on the differential pressure profile of GCO₂-water
displacements conducted at 40 bar and 33 °C.



452 Figure 9: Effect of CO₂ injection rate on the differential pressure profile of GCO₂-water 453 displacements conducted at 40 bar and 33 °C.



E.1) The data shows clearly that conducting GCO₂-water displacements at higher pressure (70 bar) 458 caused the PD oscillations to appear over a wider range of CO₂ injection rates (from 0.2 to 1 ml/min). It 459 reveals also that the change in the maximum and quasi-differential pressures, corresponding time and 460 461 PD oscillations depend on the range of the injection rate; the highest change occurred as the injection 462 rate increased from 0.4 to 1 ml/min. For illustration, as the CO₂ injection rate increased from 0.4 to 1 463 ml/min, the maximum-differential pressure increased considerably by around 258% (from 0.845 to 464 3.024 bar) and the quasi-differential pressure increased by around 224.5% (from 0.265 to 0.86 bar). The 465 corresponding time prolonged by 140% (from 1 to 2.4 min), despite the increase in the injection rate, 466 due to the increase in the maximum-differential pressure. The frequency of the PD oscillations was 467 almost constant for the last 20 min of both experiments. The increase in the maximum and quasidifferential pressures can be attributed to the increase in the viscous forces; the increase in the 468 469 corresponding time can be related to the high increase in the magnitude of the maximum differential 470 pressure.

471 E.2) On the other hand, as the CO₂ injection rate increased from 0.2 to 0.4 ml/min, the maximum-472 differential pressure was almost constant at around 0.85 bar, the quasi-differential pressure slightly increased, the corresponding time slightly reduced, and the frequency of the PD oscillations 473 474 considerably decreased but the magnitude of the PD oscillations significantly increased from around 475 0.25 to 0.825 bar. The nearly constant maximum-differential pressure (0.85 bar) at the low injection rates 476 (0.2 to 0.4 ml/min)-core floodings reveals a negligible impact of the viscous forces on the differential 477 pressure at the conditions investigated. However, the reduction in the frequency of the PD oscillations 478 might be attributed to CO₂ flow through preferential flow paths (Gunde et al., 2010).

The frequency of the PD oscillations might depend to a considerable extent on the core sample properties, the change in CO₂ distribution due to the change in the CO₂ injection rate, and the operational conditions. For illustration, as the CO₂ injection rate increased from 0.2 to 0.4 ml/min, the CO₂ might have distributed over a wider range of capillaries. Consequently, as the viscous pressure drop declined because of water depletion, the CO₂ flow inside the smaller capillaries was blocked due to their higher resistance to CO₂ flow. Later, as the pressure drop continued, the CO₂ flow in larger capillaries was blocked, too. Ultimately, it came to the point when all capillaries were blocked by the capillary forces (<u>Hildenbrand et al., 2002</u>; <u>Nutt, 1982</u>). Thus, the increase in CO₂ distribution with increasing injection rate might have led to prolonging the time required for the capillary forces to block the CO₂ production from all opened interconnected flow paths. As a result, since the volume of the opened capillaries were larger with increasing injection rate from 0.2 to 0.4 ml/min; therefore, the frequency of the PD oscillations was reduced.



491

492 Figure 10: Effect of CO₂ injection rate on the differential pressure profile of GCO₂-water displacements
 493 conducted at 70 bar and 33 °C.



494

495

Figure 11: Effect of CO₂ injection rate on the differential pressure profile of GCO₂-water displacements conducted at 70 bar and 33 °C.

In summary, fluid pressure, temperature and CO₂ injection rate exert significant influences on the differential pressure profile of the GCO₂-water drainage displacements. The differential pressure profile at all fluid pressures, temperatures and injection rates is characterized by a high initial increase immediately followed by a steep rapid pressure reduction and then by a quasi-pressure drop.

501 The differential pressure is controlled by the interplay of both capillary and viscous forces. The 502 increase in capillary forces leads to the appearance of the PD oscillations (the onset points) while the 503 increase in viscous forces causes their impedance.

There are multiple cycles of these oscillations and the occurrence and frequency of these oscillations vary with fluid pressure, temperature and injection rate. The frequency of these oscillating cycles increases as fluid pressure and fluid temperature increase but vary with injection rate and seem to be fluid pressure dependent. These oscillations occurred only at 0.4 ml/min at low pressures (i.e. 40 bar), but they appeared over a wider range of injection rates at higher pressures (i.e. 70 bar). The maximum-differential pressure reached during each cycle increases with increasing fluid pressure, temperature and injection rate.

511 3.2 Effect of Fluid Pressure, Temperature, and Injection Rate on Irreducible Water 512 Saturation and Endpoint Effective and Relative Permeabilities of CO₂

The effective and relative permeabilities of CO₂ are significantly important to the determination of 513 514the efficiency and integrity of CO₂ sequestration in subsurface formations (Busch and Müller, 2011; 515 Rathnaweera et al., 2015). At the end of the flooding experiment, the volume of the water produced was measured, and the irreducible water saturation was calculated. Then, the core sample was weighed 516 to confirm the irreducible water saturation calculations. To calculate the endpoint effective (relative) 517 CO₂ permeability using Darcy's law, the average quasi-differential pressure and the average CO₂ 518 outflow rate of the last period were used (Akbarabadi and Piri; Chang et al., 2013). The CO2 viscosity 519 520 at the experimental pressure and temperature was calculated using the Peace software website (Peace 521 software, 2017).

The results from Table 1 shows that both endpoint CO₂ relative permeability (K_{rcO2}^{max}) (Armstrong et al., 2017) and irreducible water saturation (S_{wr}) are dependent on the experimental conditions at which they are measured. The S_{wr} was in the range of around 0.38-0.45 while the K_{rCO2}^{max} was less than 0.25. Busch and Müller obtained a low relative permeability for CO₂, too (Busch and Müller, 2011). Such 526 low relative permeability would tend to decrease injectivity while increasing displacements efficiency

The results from Table 1 reveal that in general the increase in fluid pressure, temperature, and injection rate lead to an increase in the $K_{rCO2}max$ and a decline in the S_{wr} . In case of increasing fluid pressure and temperature, the high increase in the K_{rCO2} can be attributed mainly to the high increase in the injection rate inside the core sample due to the high impact of gas expansion (Rostami et al., 2010; Skauge et al.). This increase in volumetric CO₂ injection rate might result in forcing the CO₂ to flow through a wider range of the core sample pores.

534 The displacements efficiency is controlled by many factors that include relative permeability, 535 wetting conditions, viscous fingering, gravity segregation, channelling, the amount of crossflow/mass 536 transfer (Chukwudeme and Hamouda, 2009), mobility ratio, and capillary number (Kazemifar et al., 537 2015). The capillary number (*Ca*) refers to the ratio of the viscous forces to capillary forces (Lenormand 538 et al., 1988). The mobility ratio (M) refers to the ratio of the displaced to the displacing phase viscosities. 539 Increasing the contrast between the viscosity of the displacing and displaced fluid leads to a higher M 540 which will result in a more unstable configuration front. The following formulas are used to define 541 them:

542
$$Ca = \frac{\mu_2 V_2}{\sigma \cos \theta}$$
(5)

543
$$M = \frac{\mu_2}{\mu_1}$$
 (6)

where μ is the dynamic viscosity, σ the interfacial tension between the displaced and the displacing phases, 1 the subscript of the displaced phase, 2 the subscript of the displacing phase, θ the contact angle between the two fluids and the surface, and V_2 the bulk velocity of the displacing fluid. The flowing equation is used to define the bulk velocity.

548
$$V_{bulk} = \frac{Q}{A \phi}$$
(7)

where *Q* is the volumetric injection rate, *A* the area of the frontal face of the core sample, and ϕ the core sample porosity (Kazemifar et al., 2015). Based on the magnitudes of the *Ca* and the *M*, three different regimes can be defined (Kazemifar et al., 2015). For the GCO₂-water displacement investigated both *Ca* and *M* are small, which suggest a capillary fingering regime.

553 The reduction observed in the S_{wr} can be attributed mainly to the increase in the Ca and the reduction in the M. This is because the Ca and M are the most influential dimensionless parameters that 554 govern GCO₂-water core flooding displacement (<u>Kazemifar et al., 2015</u>). As the *Ca* increases, the impact 555 of the capillary forces compared to viscous forces decreases. The balance between the viscous forces 556 and capillary forces governs the pore scale drainage displacements (Heaviside and Black, 1983). The 557 capillary forces are responsible for the trapping of the injected CO₂ (Akbarabadi and Piri, 2013; Bachu 558 and Shaw, 2003). Thus, decreasing the capillary forces (e.g. due to the reduction in the interfacial 559 tension) will lower the S_{wr} (i.e. enhance the fluid displacements) (Ahmadi et al., 2015). On the other 560 hand, reducing M will result in a more uniform displacement of water by CO₂ (Bennion and Bachu, 561 562 2006), which can result in reducing the S_{wr} . The data from Table 1 show that the increase in the Ca and 563 the reduction in the M can lead to a reduction in the S_{wr} even when the change in both Ca and M is small. Ding and Kantzas observed that the critical Ca for the gas-water system is 2E-8 (Ding and 564 565 Kantzas, 2007).

566 The results from Table 1 reveal that increasing the fluid pressure from 40 to 70 bar at 33 °C and 0.4 ml/min caused the K_{rCO2} ^{max} to increase by around 0.099 and the S_{wr} to decrease by around 0.047. The 567 largest increase in the K_{rCO2} ^{max} and the highest reduction in the S_{wr} occurred as the fluid pressure 568 increased from low-fluid pressure displacements (40 and 50 bar) to high-fluid pressure displacements 569 570 (70 bar). The observed trend of the K_{rCO2} ^{max} and S_{wr} are in agreement with the findings of Liu et al. and 571 Bennion and Bachu (Bennion and Bachu, 2006; Liu et al.). Liu et al also observed an increase in the Krco2 572 with increasing pressure (Liu et al.). Bennion and Bachu observed an increase in the Krco2 and increase in the maximum endpoint CO_2 saturation (i.e. decrease in S_{wr}) with increasing pressure; they attributed 573

that to the reduction in IFT with increasing pressure (Bennion and Bachu, 2006). The observed trend of the $K_{rCO2^{max}}$ and S_{wr} can also be associated with the relatively high increase in the *Ca* and the high reduction in the *M*.

577 The results from Table 1 reveal that increasing temperature led to an increase in the K_{rCO2} ^{max}. On the other hand, increasing temperature caused a reduction in the S_{wr} for the displacements conducted 578 at high-fluid pressure (70 bar) and over a high temperature increase (33-45 °C). Nonetheless, for the 579 580 experiments conducted at low-fluid pressure (50 bar) and over a small temperature increase (29-33 °C), 581 the trend of the S_{wr} depends on the magnitude of the experimental temperature. For the high-fluid 582 pressure displacements, when the temperature increased from 33 to 45 °C at 70 bar, the Krco2max increased by around 0.035 and the S_{wr} decreased by around 0.02. The reduction in the S_{wr} for the 70 bar 583 displacements can be attributed also to the high increase in the *Ca* and the high reduction in the *M*. For 584 the low-fluid pressure displacements, as the temperature increased slightly from 29 to 33 °C at 50 bar, 585 the K_{rCO2} max increased by around 0.016. Nevertheless, the S_{wr} value was between around 0.40 and 0.41. 586 587 The S_{wr} saturation slightly increased by around 0.01 as the temperature increased from 29 to 31 °C, and 588 then slightly decreased by about 0.005 as the temperature increased from 31 to 33 °C. The slight increase 589 in the S_{wr} might be related to the slight reduction in the *Ca* as well as the impact of the capillary forces, 590 which can be seen through the appearance of the PD oscillations when the temperature increased to 31 591 °C, see Section 3.2 for more information; the PD oscillations might result in hindering water production 592 to a slight extent. On the other hand, the slight reduction in the S_{wr} , when the temperature further 593 increased to 33 °C, can be associated with the relatively high increase in the Ca as well as the slight reduction in the *M*. 594

595 Overall, the results from Table 1 shows that the increase in the CO₂ injection rate caused an increase 596 in the $K_{rCO2}max$ and a reduction in the S_{wr} . Increasing the injection rate from 0.1 to 2 ml/min at 40 bar and 597 33 °C resulted in an increase in the $K_{rCO2}max$ by around 0.157 and a reduction in the S_{wr} by around 0.05. 598 These findings agree with those in Chang et al. and Akbarabadi and Piri (Akbarabadi and Piri; Chang 599 et al., 2013). However, for the core flooding at 0.4 ml/min or less, the Swr trend is not clear. Moreover, 600 the Krco2max of the experiments conducted at 40 bar-0.2 ml-33 °C does not fit linearly in the trend. 601 Increasing the injection rate from 0.6 to 1 ml/min resulted in the highest reduction in the S_{wr} . This can 602 be corresponded to the high increase in the Ca from around 7.9 E-8 to 1.3 E-7. For the core flooding 603 performed at 70 bar and 33 °C, increasing the injection rate from 0.2 to 1 ml/min caused a very slight 604 reduction in the S_{wr} by 0.0077. However, the $K_{rCO2^{max}}$ increased substantially as the injection rate 605 increased from 0.2 to 0.4 ml/min. Nevertheless, as the injection rate increased to 1 ml/min, a significant reduction in the K_{rCO2}^{max} happened again, the reason is not clear. The very slight reduction in the S_w 606 607 might be because only a slight increase occurred in the *Ca* and that *M* was constant.

Table 1: Effect of fluid pressure, temperature, and injection rate on endpoint effective and relative permeabilities of gaseous CO₂ and irreducible water saturation

Parameter	Experiment	K)co2	Krco2	Sur	М	Ca
ect	40 bar-0.4 ml/min-33 °C	1.768	0.113	0.4244	46.26	5.265E-08
uid e Eff	50 bar-0.4 ml/min-33 °C	1.987	0.127	0.4089	44.56	6.250E-08
Fl Pressui	70 bar-0.4 ml/min-33 °C	2.613	0.212	0.3779	36.10	2.504E-07
ffect	50 bar-0.4 ml/min-29 °C	1.507	0.096	0.4012	48.69	4.748E-08
ıre E	50 bar-0.4 ml/min-31 °C	1.738	0.111	0.4147	46.57	4.698E-08
peratı	50 bar-0.4 ml/min-33 °C	1.987	0.127	0.4089	44.56	6.250E-08
[Tem]	70 bar-0.4 ml/min-33 °C	2.613	0.212	0.3779	36.10	2.547E-07
L	70 bar-0.4 ml/min-45 °C	3.675	0.247	0.3566	31.34	2.714E-07
	40 bar-0.1 ml/min-33 °C	0.67	0.043	0.38	46.26	1.316E-08
ffect	40 bar-0.2 ml/min-33 °C	1.265	0.081	0.446	46.26	2.632E-08
ate E	40 bar-0.3 ml/min-33 °C	0.955	0.061	0.436	46.26	3.948E-08
ion R	40 bar-0.4 ml/min-33 °C	1.493	0.095	0.4244	46.26	5.265E-08
njecti	40 bar-0.5 ml/min-33 °C	1.528	0.097	0.436	46.26	6.581E-08
ij	40 bar-0.6 ml/min-33 °C	1.535	0.098	0.4167	46.26	7.897E-08
	40 bar-1 ml/min-33 °C	1.793	0.114	0.3837	46.26	1.316E-07

40 bar-2 ml/min-33 °C	3.13	0.20	0.391	46.26	2.632E-07
 70 bar-0.2 ml/min-33 °C	2.421	0.154	0.3798	36.10	1.273E-07
70 bar-0.4 ml/min-33 °C	3.625	0.167	0.3779	36.10	2.547E-07
70 bar-1 ml/min-33 °C	1.976	0.128	0.3721	36.10	6.368E-07

610 **4. Conclusion**

In this paper, the effect of fluid pressure, temperature, and CO₂ injection rate on gaseous CO₂ dynamic behaviour during its flooding of a water-saturated sandstone core sample have been investigated in detail. The results indicate that the parameters investigated have a moderate to significant influence on the differential pressure profile, endpoint CO₂ relative and effective permeabilities and irreducible water saturation.

616 For all fluid pressures, temperatures, and injection rates, the differential pressure profiles are 617 characterized by a sharp increase, immediately followed by a steep pressure reduction, and finally, by 618 a gradual pressure reduction. The differential pressure profiles are controlled by the interplay of both 619 capillary and viscous forces. The capillary forces produce cyclic oscillations within the differential 620 pressure and fluid production data; the increase in the viscous forces impede the appearance of these 621 oscillations. The appearance and frequency of the oscillations depend on the fluid pressure, 622 temperature, and CO₂ injection rates. In general, the frequency of the oscillations increased with 623 increasing pressure and temperature. The differential pressure oscillation cycles exhibit a very interesting response to varying injection rate, they are dependent on the fluid pressure. At 40 bar, the 624 625 oscillations were only observed at an injection rate of 0.4 ml/min, whereas at 70 bar the oscillations 626 occurred at all injection rates tested (0.2, 0.4, and 1ml/min).

In general, the increase in fluid pressure, temperature, and injection rate led to an increase in the maximum and quasi-differential pressures; the extent of the increase in the differential pressure is dependent on the fluid pressure, temperature, and injection rate. Increasing the fluid pressure and temperature caused a reduction in the time required to achieve the maximum-differential pressure at

- 631 the start of the experiment, i.e. corresponding time. Whereas, increasing the injection rate caused the
- 632 corresponding time to decrease at low injection rates and increase at high injection rates.
- 633 In general, the increase in fluid pressure, temperature, and injection rate led to an increase in the
- endpoint CO₂ relative permeability (K_{rCO2}^{max}) and a decline in the irreducible water saturation (S_{wr}). The
- 635 S_{wr} was in the range of around 0.38-0.45 while the K_{rCO2}^{max} was less than 0.25.

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