PART 1

Transport Processes
1.1. Introduction

Prior to any CO$_2$ geological storage project, an assessment is needed to confirm the ability of the formations overlying the storage reservoir to act as a seal with respect to the underlying pressurized CO$_2$. In the case of hydrocarbon reservoirs, these formations typically consist of low-permeability caprocks: the ability of these tight rocks (usually imbibed with water) to act as a seal with respect to an underlying buoyant phase is primarily a capillary effect, which is indeed proven in the case of the hydrocarbon phase present in the reservoir, but not in the case of a CO$_2$-rich phase. In the case of deep saline aquifers, the sealing efficiency of the overlying tight formations (referred to as aquitards or aquicludes) with respect to a non-aqueous buoyant fluid is not known.

Excessive pressurization of the CO$_2$ phase, due to the injection operations and to the buoyancy effect, induces leakage through the seal rock(s) by a variety of mechanisms, each being active above a specific pressure threshold. The lowest of these thresholds should indeed not be exceeded during the CO$_2$ injection and storage operations – unless the associated leakage rate is extremely small.

The failure mechanisms are conventionally divided into two types: mechanical and capillary. Seals that fail because of mechanical and capillary mechanisms are referred to as hydraulic seals and membrane seals, respectively [WAT 87].
Mechanical failure comprises the development of high-permeability pathways through a variety of processes, such as tensile fracturing or shear slip reactivation of pre-existing faults, as well as thermal fracturing. This is considered to be the most dangerous failure mode, with high associated leakage rates, and it is constantly monitored in most ongoing CO$_2$ storage projects, e.g. by passive microseismic techniques (see [RUT 12] for a recent review).

Capillary failure (or capillary invasion), in which the rock pore structure is left unaltered during (and after) the gas leakage process, is a more pervasive process than mechanical failure, and lower leakage rates are expected – the effective gas permeability that follows gas capillary breakthrough is only a fraction of the intrinsic permeability of the seal rock, which itself does (or should) not exceed a few micro-Darcy ($10^{-18}$ m$^2$). It is expected to occur at a lower CO$_2$ (pore) pressure than the pressure required for tensile fracturing, except in extremely tight rocks such as evaporites [GLU 04]. This pressure corresponds to the capillary entry pressure, or the minimum overpressure in the CO$_2$ phase (compared to brine pressure) required to intrude into the seal rock and to displace the saturating brine phase. From the Laplace–Young equation, this overpressure, hereafter denoted by $P_c$, is inversely proportional to a characteristic pore throat size $R$ and proportional to the interfacial tension (IFT) between the brine and the stored CO$_2$-rich gas ($\gamma_{w,g}$) and to the cosine of the contact angle $\theta$ characterizing the wettability of the seal rock:

$$P_c \sim \gamma_{w,g} \cos(\theta)/R$$

More precisely, $\theta$ is the water-receding (or gas-advancing) angle on the rock substrate, which is referred to in the following as the drainage angle. This angle is often significantly smaller than the imbibition angle corresponding to the reverse displacement (water displacing the CO$_2$-rich fluid from the substrate): the difference, or contact angle hysteresis, is related in a complex manner to the interactions of CO$_2$ with the substrate, and to substrate chemical and/or geometrical heterogeneities.

In practice, the sealing capacity of a tight rock (sampled for instance from the overburden of a potential storage site) initially saturated with brine is appreciated in the laboratory from breakthrough or displacement experiments. This sort of experiment is carried out in a more general context, e.g. for qualifying the confining properties of the engineered (compact) clay barriers used for underground nuclear waste disposal (here, the gas is the hydrogen generated by the waste) [GAL 98, HAR 99, HOR 99, GAL 00], or of the upper layers of underground reservoirs for seasonal storage of natural gas. The “standard” procedure [THO 68] for running such experiments consists of increasing stepwise the gas pressure on one face of a brine-saturated rock sample until the gas starts penetrating into the sample and thus displaces the saturating brine, which defines the gas entry pressure, and then, for a slightly larger pressure threshold called breakthrough or displacement pressure,
comes out on the other face of the sample; then, the effective gas permeability after breakthrough can also be determined. The gas (CO₂) breakthrough (or displacement) pressure, hereafter denoted as \( P_d \), and the effective gas permeability after breakthrough are very important in practice, as they are related, respectively, to the maximum sustainable overpressure in the storage reservoir and to the leakage rate of the CO₂-rich fluid following breakthrough (\( P_d \) can be exceeded, however, if this rate is extremely small). If it is assumed that the capillary invasion process described in the previous paragraph is the dominant failure mechanism (i.e. the seal rock is a membrane seal), then \( P_d \) should obey the following Laplace–Young equation:

\[
P_d - P_w \sim \gamma_{w,g} \cos(\theta)/r \sim \gamma_{w,g} \cos(\theta)/k^{1/2}
\]  

[1.2]

where \( P_w \) is the pressure in the brine that imbibes the seal rock, \( r \) is the size of the narrowest pore throats along the leakage pathway in the seal rock, and \( k < r^{1/2} \) is the intrinsic (or absolute) permeability of the seal rock. In addition to being very tight (\( r \) and \( k \) small), the interfacial tension between brine and the CO₂-rich gas must be high enough and the seal rock must be water-wet (i.e. small \( \theta \)) if the brine-saturated rock is to function as an effective (membrane) seal with respect to the buoyant CO₂-rich phase. On the one hand, the available evidence is that IFTs between brine and CO₂ in geological storage conditions are higher than \(~23 \text{ mN/m}~\) – this value has to be corrected for the effects of the impurities co-injected with CO₂ and of the mixing with indigenous hydrocarbons (if storage is in a hydrocarbon reservoir). On the other hand, the water-wet character of many rocks and rock-bearing minerals seems to be preserved in the presence of CO₂, at least in the drainage conditions (CO₂ displaces water from the substrate) representative of the leakage process. This subject is currently being intensely investigated.

The outline of this chapter is as follows: the “standard” procedure for determining gas breakthrough (or displacement) pressures in water-saturated tight rocks and the ensuing gas effective permeability is presented in section 1.2. This determination, which is extremely time-consuming, is mandatory in the case of deep aquifers where there is no \textit{a priori} knowledge of the sealing behavior of the top aquitard (or aquiclude), but it can be skipped in the case of hydrocarbon reservoirs where a lower bound on breakthrough pressure can be simply inferred from the reservoir discovery pressure and the relevant interfacial properties. These properties, namely the IFTs between brine and CO₂-rich fluids and the wetting (contact) angle \( \theta \), are reviewed and discussed in section 1.3, and the method for inferring a lower bound on the CO₂ breakthrough pressure of depleted hydrocarbon reservoirs is presented in section 1.4. In section 1.5, the gas breakthrough experiments available in the literature are further examined and discussed. It turns out that the mechanism of capillary invasion in a rigid porous medium (i.e. membrane seal failure) does not account for many of the observed
features, which witness some quasi-permanent deformations of the porous structure.

The important topics of mechanical failure processes such as thermal fracturing, reconnection of fracture networks, and shear reactivation of faults (especially when a reactive fluid, i.e. CO$_2$ and water, changes the slip conditions) are not addressed in this chapter (see [RUT 12] for a review).

1.2. Gas breakthrough experiments in water-saturated rocks

The standard procedure [THO 68] for determining the gas breakthrough (or displacement) pressure in a given rock sample fully saturated with brine and placed in representative temperature and confining pressure conditions consists of increasing the gas pressure in a stepwise fashion (each step lasting a few hours to a few days) on one face of the sample until the breakthrough pressure is reached, i.e. gas displaces the saturating brine phase across the sample [LI 05, HAR 09, TON 10, TON 11, BOU 11, SKU 12]. Gas entry (but no breakthrough) may occur for the pressure step previous to breakthrough, i.e. the gas phase enters the rock, but does not come out on the other face of the sample. Most gas displacement experiments are conducted with nitrogen as the gas phase rather than with CO$_2$: then, the CO$_2$-breakthrough overpressure is obtained by multiplying the nitrogen-breakthrough overpressure by the ratio of the brine-CO$_2$ IFT to the brine-nitrogen IFT (assuming contact angles to be the same for the two gases: this point is discussed at the end of the following section).

In practice, the cylindrical (thin) rock sample to be tested is placed in a Hassler-type cell and fully saturated with brine under a prescribed representative confining pressure (this confining pressure or, more precisely, the effective pressure, i.e. the difference between the confining and pore pressures, has a strong impact on the rock petro-physical properties of interest; see [TON 10, TON 11, SKU 12]). The intrinsic permeability can then be determined, e.g. from steady-state measurements of the brine flow rate versus pressure drop across the sample [BOU 12]. Then, the gas phase is brought into contact with the inlet face of the sample and compressed in a stepwise manner, while a constant backpressure $P_w$ is imposed in the brine phase on the outlet face, e.g. by means of a syringe pump working in the constant-pressure mode (the flow line from the sample outlet to the syringe is filled with the brine phase). Figure 1.1 displays the volume readings of the downstream pump and the upstream pressure in the CO$_2$ phase obtained in the course of a breakthrough (or displacement) experiment. The sample in this example comes from the carbonate-rich overburden of a depleted gas reservoir currently used as a CO$_2$ storage reservoir in the southwest of France. The absence of brine flow (as indicated by the volume readings in the downstream pump)
indicates that CO₂ breakthrough has not yet occurred for the maximum investigated CO₂ (upstream) pressure of \( P_g = 8.6 \) MPa (the experiment was stopped at that pressure), which is 7.6 MPa above the pore (brine) pressure \( P_w = 1 \) MPa (see [TON 10, TON 11] for details): the CO₂ entry and breakthrough overpressures are thus higher than 7.6 MPa.

**Figure 1.1.** In this experiment, breakthrough has not yet occurred for gas (upstream) pressures as high as 8.6 MPa. Rock permeability is in the nano-Darcy range, and the brine pressure downstream is equal to 1 MPa [TON 10, TON 11]

Figure 1.2 displays the results obtained in a similar experiment carried out with a more permeable caprock sample, in which CO₂ entry (but not breakthrough) is apparent for the first pressure step investigated (equal to 1.5 MPa, i.e. 0.5 MPa above the brine pressure), whereas CO₂ flows continuously through the sample for the following pressure step \( P_g = 2.5 \) MPa, as manifested by the uninterrupted displacement of the piston of the downward pump. The displacement pressure \( P_d \) is thus intermediate between 0.5 and 1.5 MPa. The gas effective permeability for the subsequent pressure step can be inferred from the measured steady flow rate \( Q \) and the equation

\[
k_g = 2 \mu_g Q L P_w/A (P_g^2 - P_w^2)
\]  

[1.3]

where \( L \) is the sample thickness and \( \mu_g \) the viscosity of the gas (assumed to be a perfect gas). In this experiment, \( k_g = 1.5 \) nano-Darcy.
Figure 1.2. In this experiment, carried out with the same setup as that used in Figure 1.1 and a more permeable rock sample, CO₂ entry into the rock sample is apparent when $P_g = 1.5$ MPa (first pressure step), and CO₂ breakthrough pressure is intermediate between $P_g = 1.5$ MPa and $P_g = 2.5$ MPa, where gas flows through the rock sample at a steady rate, from which an effective gas permeability of 1.5 nano-Darcy is inferred (see text, and [TON 10, TON 11]).

The accuracy of the standard procedure is directly related to the amplitude and duration of the pressure steps and to the ability of the downstream syringe pump to record small volume variations or piston displacements. In the experiment of Figure 1.2, gas entry is apparent only 200–300 hours after the upstream pressure has been raised to 1.5 MPa, which suggests that in the experiment of Figure 1.1 all pressure steps except the last step were too short. An experiment typically takes weeks or even months; more than a year has been necessary to determine the consolidation behavior and gas (nitrogen) breakthrough parameters of a specimen of Nordland shale overlying the Sleipner CO₂ storage site [HAR 09].

The time required for accurate measurements is longer when the rock is less permeable. Experimental time can be saved, however, if mercury intrusion capillary pressure (MICP) data are available for the rock investigated (the reliability of such data is questionable for very fine-grained sediments [HIL 03, NEW 04]; they can only be used to give an order of magnitude). MICP measurements consist of forcing mercury (Hg) into an evacuated (dried) core sample under increasing pressure while monitoring the volume of Hg (the non-wetting phase) that is injected at each pressure step (the wetting phase is the mercury vapor). An estimate of the mercury capillary entry pressure is obtained as the intersection of the low Hg-saturation ($S_{Hg} \approx 0$) and intermediate Hg-saturation parts of the logarithm of the capillary pressure versus Hg-saturation [CAR 10]: this entry pressure must then be converted to that of the
brine/gas system interest by using the proper interfacial tensions and contact angles (usually assumed to be equal to 140° in the mercury phase and to 0° in the brine).

A few alternate procedures have been proposed to evaluate breakthrough pressures in a shorter amount of time; they have recently been discussed and compared by Boulin et al. [BOU 11], who concluded that the standard procedure is the most representative of the leakage process of interest, even though duration is its main drawback. One of these alternate procedures [HIL 02, HIL 04] consists of forcing the gas into the brine-saturated rock at a pressure much higher than the breakthrough pressure, and then letting the system relax until the pressures in the inlet and outlet faces are stable: the residual pressure difference is considered to be a proxy of the breakthrough overpressure. This procedure provides meaningful trends but, as argued by Zweigel et al. [ZWE 04], the residual pressure difference (also called “snap-off pressure” [AMM 12]) results from a reimbibition process and is therefore expected to be smaller than the true breakthrough (or displacement) overpressure corresponding to a (first) drainage process. A factor of ~2 between the two quantities has in fact been measured by Harrington et al. [HAR 09] with the Nordland shale mentioned above, by Boulin et al. [BOU 11] with two other rock samples (one being the caprock of the Ketzin storage site in Germany), and by Amman-Hildenbrand et al. [AMM 12] as well.

1.3. Interfacial properties involved in seal rock integrity

As emphasized above, a key role is played by the brine/gas IFT and the wetting (or contact) angle $\theta$, which have been thoroughly investigated over the past decade.

1.3.1. Brine-gas IFT

A large amount of brine/gas IFT values are available in the literature, which also provides simple methods for evaluating this property, including when the gas not only contain CO$_2$ but also some other “impurities” (such as H$_2$S, CH$_4$, N$_2$, etc.). One of these methods has recently been proposed by Duchateau and Broseta [DUC 12] and is detailed below.

Early measurements of IFT between pure water and CO$_2$ in pressure and temperature conditions relevant to geological storage have been critically reviewed by Georgiadis et al. [GEO 10] and Bikkina et al. [BIK 11a], who also presented some new measurements. Starting at low pressures from values close to the surface tension of pure water at the temperature of interest, the IFT between pure water and CO$_2$ decreases with increasing pressure, first linearly (for $P$ below 3–5 MPa), then more slowly until it reaches a pseudo-plateau of about 23–30 mN/m for high-enough
$P$ (larger than about 15–20 MPa at $T = 100^\circ$C, and 8–10 MPa at $T = 40^\circ$C), with the lowest values obtained after long (~1 day) equilibration times [BIK 11a]; this pseudo-plateau value turns out to be fairly independent of temperature up to 110°C.

![Figure 1.3](image.png)

**Figure 1.3.** Pressure dependence of the interfacial tension between CO$_2$ and pure water at $T = 35, 50, 70, 90$ and $100^\circ$C, and between CO$_2$ and a 20 g/L NaCl brine at $35^\circ$C [CHI 07a]

When salts are added to the water, the IFT between the aqueous phase and the gas increases by an amount that is independent of pressure: this amount is thus equal to the increase in water surface tension (i.e. the IFT between the aqueous phase and its vapor or air). It can be rigorously shown from an analysis of Gibbs adsorption equation [DUC 12] that the effects of gas pressure (which tends to lower the IFT) and brine salinity (which tends to increase the IFT) on IFT are independent. Hence, the brine/gas IFT at given $T$ and $P$ is equal to the pure water/gas IFT at the same $T$ and $P$, plus the surface tension increase induced by the addition of the brine salts to the water at $T$. This result holds for any gas that is sparingly soluble in water (including CO$_2$, and possibly H$_2$S) and for any non-adsorbing salt species [DUC 12]. The surface tension values of many salted aqueous solutions are available in the literature [ABR 93], as are the IFTs between pure water and CO$_2$ mixed with the following “impurities”: CH$_4$ [REN 00] and N$_2$ [YAN 01], which tend to raise the IFT, and H$_2$S [SHA 08], which tends to lower the IFT. If one (or two) of these two tensions is (are) not available, it is easier to measure it (them) than the brine/gas IFT itself, as it does not require the use of corrosion-resistant equipment.

### 1.3.2. Wetting behavior

No clear-cut answer has yet been given as to whether or not rocks and rock-forming minerals remain water-wet in the presence of CO$_2$ in geological...
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storage conditions, i.e. the contact angle $\theta$ in equations [1.1] and [1.2] remains low. Most of the recent experimental work in this domain consists either of contact angle measurements by the sessile drop method, or of direct observations of CO$_2$ and brine distributions in silica or glass micromodels. Using the sessile drop method, Yang et al. [YAN 08] observed that $\theta$ increases to values above 90° at high pressures in the CO$_2$ phase (>10 MPa) for a brine and a limestone rock from the Weyburn CO$_2$ storage reservoir; however, Espinosa and Santamarina [ESP 10] did not find any significant angle variation for quartz and calcite when increasing the pressure to 10 MPa at 23.5°C; Bikkina [BIK 11b] observed an increase in $\theta$ – by more than 40° – after repeated exposure of calcite and quartz to CO$_2$ at 25 and 50°C and pressures up to 20 MPa; Jung and Wan [JUN 12] observed for silica substrates at 45°C that $\theta$ increases with pressure by slightly less than 20°, much of this increase taking place between 7 and 10 MPa; more moderate variations have been recently observed for other water-wet substrates by Mills et al. [MIL 11] and by Wang et al. [WAN 12]. In direct observations in silica or glass micromodels, Chalbaud et al. [CHA 09] did not notice any significant change in wetting angles when displacing water with CO$_2$ under various subcritical (gaseous or liquid) and supercritical conditions (up to 60°C and 10 MPa), but Kim et al. [KIM 12] observed values as high as $\theta \approx 80°$ for strongly salted (5 M NaCl) brine droplets left in the micromodel after drainage with CO$_2$ at 8.5 MPa and 45°C.

A possible reason for measurement discrepancies is contact angle hysteresis: any intermediate value between the advancing and receding angles can be observed by using the sessile drop method, unless specific procedures such as increasing or decreasing the pressure are adopted to shrink or swell the water drop. This difficulty can be circumvented by using the captive drop method, in which a drop of water (surrounded by the CO$_2$ phase) or a bubble of CO$_2$ (immersed in water) is held against the substrate and alternatively expanded and retracted by fluid injection and withdrawal to get the advancing and receding angles (see Figure 1.4). This method has been used by Chiquet et al. [CHI 07b] and Broseta et al. [BRO 12] to investigate the wetting behavior of a variety of substrates including quartz, muscovite mica, calcite, and a carbonate-rich rock (similar to that used in the breakthrough experiments presented in section 1.3 and Figures 1.1 and 1.2) over a large range of temperatures (up to 140°C), pressures (up to 150 bar), and brine salinities (up to NaCl saturation) representative of various geological storage conditions. In these measurements, a bubble of CO$_2$ was held beneath the substrate (immersed in water) and its volume varied by means of a capillary connected to a syringe (see Figure 1.4). The contact angle of interest, namely the drainage angle $\theta_d$, was observed to be low and not significantly altered under high CO$_2$ pressure [BRO 12]. While this needs to be further checked with other known water-wet minerals, we assume in the following that rock-forming minerals remain water-wet in the presence of CO$_2$ under geological storage conditions and incipient leakage (i.e. drainage) conditions (of CO$_2$ displacing water).
1.4. Maximum bottomhole pressure for storage in a depleted hydrocarbon reservoir

Acceptable bottomhole pressures are those that preserve the integrity of the seal rock: the maximum bottomhole pressure, which should not be exceeded during the CO₂ injection and storage process, corresponds to the breakthrough pressure of the seal rock. When the storage reservoir is a deep saline aquifer, the ability of the top low-permeability aquitard (or aquiclude) to confine a buoyant non-wetting fluid is not known in the first place, and the breakthrough pressure needs to be determined (see section 1.2). When the storage reservoir is a depleted hydrocarbon reservoir, the sealing capacity of the top seal is indeed proven up to the reservoir discovery pressure, but with respect to the hydrocarbons initially present in the reservoir (i.e., at reservoir discovery). It is not correct to assume that this seal will also be able to trap over the scheduled storage timescale (a few hundreds to thousands of years) of the CO₂-rich fluid that results from the injection process: refilling the depleted reservoir with CO₂ up to the reservoir discovery pressure will inevitably give rise to leakage through the top seal if the hydrocarbon reservoir was close to leakage conditions at discovery, because of the lower IFT between brine and the CO₂-rich fluid as compared to that between the brine and the hydrocarbon fluid [LI06] – the drainage contact angles are supposed to be unchanged (see section 1.3).

A lower bound for the CO₂-rich fluid breakthrough (or displacement) pressure \( P_d \) (see equation [1.2]) is obtained from the following conservative hypothesis: the conditions for incipient hydrocarbon leakage by capillary failure were met at reservoir discovery, i.e., the discovery pressure \( P_{\text{dis}} \) is also the hydrocarbon breakthrough (or displacement) pressure, which thus exceeds the pressure \( P_w \) in the brine phase imbibing the seal rock by an amount proportional to \( \gamma_{w,\text{hc}} \cos(\theta)/r \), where \( \gamma_{w,\text{hc}} \) is the interfacial tension between brine and the hydrocarbon phase, \( r \) corresponds to the narrowest pore throats along the leakage pathway, and \( \theta \) is the drainage contact angle. At some (small) distance from the reservoir boundary, the pore pressure \( P_w \) within the seal rock is the same as when the reservoir was discovered (because of the very low pressure diffusivity in low-permeable rocks).
Therefore, by using equation [1.2] and assuming that the drainage contact angles for the hydrocarbon- and CO₂-rich phases are equally low, the displacement pressure $P_d$ is related to the discovery pressure $P_{\text{dis}}$ as follows:

$$P_d - P_w < (P_{\text{dis}} - P_w) \frac{\gamma_{\text{CO}_2,g}}{\gamma_{\text{CO}_2,hc}}$$ [1.4]

Since IFTs between water and hydrocarbons are at their maximum – equal to about 50 mN/m in geological storage conditions – for alkane-rich hydrocarbons, which is about twice the IFT between brine and CO₂ under similar conditions, the following simple rule of thumb can be enunciated: the pressure in a depleted hydrocarbon reservoir refilled with CO₂ should not exceed a value approximately half-way between the pore (i.e. brine) pressure in the seal rock and the reservoir discovery pressure. More precise estimates can be obtained from the IFTs between the formation brine and the hydrocarbons under the conditions of reservoir discovery and between the same brine and the CO₂-rich fluid present in the reservoir under storage conditions.

1.5. Evidences for capillary fracturing in seal rocks

“The view that gas breakthrough in water-saturated tight rocks leaves the porous skeleton intact is a simplification.” The available experimental and theoretical evidences are that, in the course of the gas breakthrough process, some degree of rock deformation occurs that provides preferential pathways for gas migration; in addition, deformation often proceeds in a somewhat irreversible or quasi-permanent manner. The experimental evidences for the “formation, aperture and propagation of gas preferential pathways” [GAL 98] during gas breakthrough are numerous, starting with the early gas migration studies by Gallé, Harrington, and coworkers [GAL 98, HAR 99, HOR 99, GAL 00] in compacted clays (mostly bentonites) fully saturated with water. Intermittent gas outflow is apparent in some experiments, suggesting that “these pathways are relatively unstable” [GAL 98], and the gas breakthrough pressure needed to re-establish gas flow after a first gas breakthrough test (followed by flow arrest) is lower than the first gas breakthrough pressure (i.e. in the virgin clay), suggesting that the flow pathways opened under high gas pressure partially close when pressure falls [HOR 99]. Similar observations were made for gas (nitrogen) breakthrough experiments in the water-saturated Nordland shales overlying the Sleipner CO₂ storage site by Harrington et al. [HAR 09], who found some moderate alteration of the sealing capacity (i.e. a lower breakthrough pressure) upon repeat breakthrough measurements with the same sample.

In two recent studies of gas (CO₂ or N₂) breakthrough experiments [WOL 10, TON 10, TON 11] in various carbonate-rich rocks with permeabilities well below the micro-Darcy, a measurable loss in the rock sealing capacity (as quantified by a lower breakthrough over-pressure and a higher effective gas permeability) was
observed again upon repetitive breakthrough experiments (in which rock samples were re-saturated with brine prior to starting a new breakthrough test).

In another recent study [SKU 12] of CO$_2$ breakthrough conducted in a micro-fractured shale (sampled from the Draupne formation in the North Sea) with permeability of a few nano-Darcy, the breakthrough pressure was observed to be sensitive to the confining and effective pressure conditions and to the compaction history, with a higher confining pressure and loading/unloading cycling of the sample leading to a higher breakthrough pressure; in addition, strain measurements indicated some pronounced dilation of the rock sample during breakthrough, presumably due to the decrease in effective pressure. Evidence for pressure-induced opening of micro-fractures by the intruding gas was provided by fitting the measured effective gas permeability versus volumetric strain to a simple model of fracture aperture [OLI 08].

In all the above-mentioned gas breakthrough experiments in clays or tight rocks, the alteration of the sealing capacity consecutive to gas breakthrough (i.e. the decrease in gas breakthrough pressure and the increase in clay or rock permeability) was moderate. However, these experiments are not numerous, and as mentioned by Harrington et al. [HAR 09], “a satisfactory explanation for how gas pathways develop (still) remains elusive.” Clearly, some research effort should be devoted to the understanding of these phenomena, which have recently been referred to as “capillary fracturing” and they should be examined closely in non- or weakly-consolidated porous media, both numerically through 2D micro-mechanical simulations [JAI 09] and experimentally [HOL 12]. At small (pore) size, micro-fracturing or fracture opening arises from the difference in pressures between neighboring pores or grains when two different fluids (brine and gas) are present, and it is indeed more pronounced when capillary pressure forces are stronger, that is, for smaller grain sizes (i.e. smaller permeabilities) and larger brine/gas IFT. Capillary fracturing is ubiquitous in nature, and has been noted to occur in various contexts: gas venting from marine sediments, oil and gas generation and escape from the source rocks, or the vertical migration of gas through water-saturated tight sediments (e.g. mudstones) that is observed in many sedimentary basins [APL 99].

1.6. Summary and prospects

The typical procedure for assessing seal rock integrity with respect to an underlying over-pressurized gas consists of running gas breakthrough experiments, from which the gas breakthrough over-pressure and the effective gas permeability are determined. These two key parameters are related, respectively, to the maximum sustainable bottomhole pressure and to the gas leakage rate through the seal rock when this pressure is exceeded. These experiments are mandatory for the rocks
(aquiclude or aquitard) overlying an aquifer used as a CO₂ storage reservoir, but not for the seal rock of a depleted hydrocarbon reservoir, where a lower bound for the maximum bottomhole pressure can be inferred from the discovery pressure of the reservoir and two IFTs: the IFT between brine and the hydrocarbons initially in place, and the IFT between brine and the stored CO₂-rich fluid.

The latter result holds provided the seal rock remains water-wet in the presence of CO₂, which seems to be the case if this rock was water-wet in the first place (i.e. in the presence of hydrocarbons). However, some seal rocks, such as mudstones, contain a non-negligible amount of organic matter [APL 11]: in this case, some significant wettability alteration in the presence of CO₂ is to be expected (see [BRO 12] for a review of the literature evidences) and the above simple approach might not hold. More generally, a critical issue is to understand and predict the distribution and connectivity of wettability and permeability heterogeneities within seal rocks [APL 11].

As emphasized in section 1.5, capillary fracturing is apparent in most gas breakthrough experiments, even though its (negative) effects on seal capacity have been shown to be rather limited in the cases investigated up to now. More work needs to be done in order to understand and predict the magnitude of these effects as a function of fluid properties and seal rock type and fracture mechanics. Another important topic that deserves to be investigated further with the above approaches and concepts is how fault seals (e.g. cataclastic fault rocks or permeability “baffles”) function in the presence of CO₂ [TUE 12].

1.7. Bibliography


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