

Experimental evaluation of carbonated brine-limestone interactions under reservoir conditions-emphasis on the effect of core scale heterogeneities



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ARTICLE INFO

Keywords:

Rock-fluids interactions
CO₂ injection
Core flooding
Carbonate formation rocks
CO₂ geosequestration
Wormholes
Dissolution
Compaction
Precipitation

ABSTRACT

CO₂ injection into deep geological structures is very often accompanied by chemical interactions between the host rock and injected fluids and/or the in-situ created solute (i.e. carbonated brine). In fact, the in-situ reactions are considered one way through which the injected CO₂ may be trapped for perpetuity. Depending on the nature and mineralogy of the host rock formation, such reactions may eventually result in a degree of change in the petrophysical properties of the rock. Carbonate formations, due to the presence of highly reactive minerals in their composition, are expected to be more prone to such changes than their sandstone counterparts.

This manuscript presents the results of an experimental study conducted to evaluate possible changes in the petrophysical properties of five heterogeneous limestone samples (calcite concentration > 91 wt%). The study includes five reservoir condition core-flood experiments (i.e. one per each rock sample) complemented by other laboratory measurements/analyses including porosity-permeability measurements, X-ray CT (X-ray Computed Tomography) and SEM (Scanning Electron Microscopy) imaging.

The results show a significant increase in the post-flood permeability of 80% of the samples caused by the dissolution and removal of carbonate minerals. The X-ray CT images reveal signs of significant mineral dissolution and establishment of flow paths through the initial larger pores in the samples leading, eventually, to the formation of wormhole features along the length of the samples. On the contrary, reduction in permeability is observed in one sample which was a relatively long (15.8 cm) composite sample consisting of two core segments placed one after the other in series. The other four samples were shorter with a nominal length of 6.4 cm. This reduction in permeability is observed predominantly in the outlet segment. This change is thought to have been primarily caused by possible migration of carbonate fines released by mineral dissolution in the inlet plug of the long composite core and to a lesser extent by the precipitation of minerals dissolved and transported from the inlet plug. This hypothesis finds further support in the pre- and post-flood dry weight measurements as well as a post-flood SEM image of the plug which reveals signs of fines migration and mineral precipitation. Slight reductions in the porosity and pore sizes are observed in most of the samples. This is likely to have been caused by the combined effect of fines migration, possible mineral precipitation and physical compaction mechanisms. Mechanical compaction is further evident from the reductions in the physical dimensions of the samples.

Overall, the results obtained show that the nature and degree of any change in the petrophysical properties of the rock samples vary to some degree from one sample to the next. This variation is found to depend on the significance and degree of dominance of the three mechanisms of mineral dissolution, mineral precipitation and mechanical compaction if they occur during the flooding process. The migration of carbonate fines also seems to be an important factor in shaping the post-flood sample properties. The presence of any initial core scale heterogeneity in the pre-flood samples is also believed to be a critical factor controlling the eventual outcome.

1. Introduction and background

Approximately half of the world's oil reserves are located in carbonate reservoirs (Mazzullo 2004) and many of these reservoirs suffer from a low recovery factor. Given the current need to reduce the level

of anthropogenic CO₂ emitted into the atmosphere to limit climate change, CO₂ enhanced oil recovery (CO₂-EOR) may be a feasible option to increase the ultimate recovery from carbonate reservoirs and use them as safe CO₂ storage sites at the same time. In fact, there are already numerous cases of application of CO₂ as an EOR agent world-

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<https://doi.org/10.1016/j.ijggc.2017.11.002>

Received 5 April 2017; Received in revised form 30 October 2017; Accepted 1 November 2017

Available online 21 November 2017

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wide most of which are located in the US (Alvarado and Manrique, 2010). One well known example is the Weyburn Field in Canada which has successfully undergone CO₂-EOR (Brown et al., 2017). On the other hand, it is widely acknowledged that, compared to their sandstone counterparts, CO₂ storage/EOR in carbonate reservoirs is more challenging due to their extreme heterogeneous nature (Asghari and Al-Dliwe, 2004), complex fluid-rock interactions and pore network system (Kadhim et al., 2014). Limestone reservoirs, in particular, consist of mainly calcite (CaCO₃) that is very reactive under acidic conditions (i.e., presence of carbonic acid). Such reactions may lead to considerable alteration of the petrophysical properties of the limestone rock formation through a number of mechanisms. These mechanisms may include mineral dissolution, mineral precipitation and mechanical compaction (Egermann et al., 2005; Izgec et al., 2006; Zekri et al., 2009). Very often, the mechanical compaction under overburden stress is a consequence of excessive mineral dissolution and weakening of the rock structure (Khather et al., 2017).

1.1. Fluid-rock interactions in carbonate reservoirs

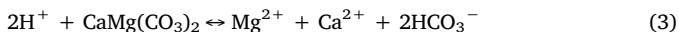
Carbonic acid is the product of the dissolution of the injected CO₂ into the formation water (brine) under reservoir conditions (Luquot and Gouze 2009; Salem and Shedid, 2013) as outlined by the following reaction:



Calcite (CaCO₃) and dolomite (CaMg(CO₃)₂) the two main constituents of carbonate rocks, can readily dissolve in carbonic acid (H₂CO₃) (Salem and Shedid, 2013). However, the reaction rate constant of calcite when in contact with carbonic acid is higher than that of dolomite, and much higher than the reaction rate constant of quartz (Palandri and Kharaka, 2004; Stumm 1992; Wellman et al., 2003; Mohamed et al., 2010). Therefore, if a formation contains a combination of calcite and dolomite, carbonic acid tends to react with calcite at a much faster rate. The reactions between calcite, CO₂ generated acid (H⁺) and formation brine can be defined as follows (Salem and Shedid, 2013):



The reaction of dolomite with CO₂ and formation brine also can be defined as follows (Salem and Shedid, 2013):



The chemical interactions of the underground CO₂-brine-rock system can lead to changes in the petrophysical properties of the rock, altering the storage capacity, injectivity and, in general, the multiphase flow characteristics of the host formation. The extent of such changes in limestone formations, containing a greater proportion of calcite, is expected to be significantly higher than dolostone formations that contain a greater proportion of dolomite (Salem and Shedid, 2013).

Although the dissolution of rock minerals tends to increase the rock permeability, later precipitation of minerals may lead to a reduction in the permeability and effective porosity. Such precipitation and subsequent reduction in permeability has been observed during core-flood experiments by a number of researchers during which, very often, carbonated brine has been used as the injection fluid (Izgec et al., 2005; Oomole and Osoba, 1983). As suggested by these researchers, mineral

precipitation is caused by the pressure drop generated along the length of the rock samples during the flooding procedure. The larger the pressure drop, the more the mineral precipitation and the subsequent reduction in the permeability of the rock (Oomole and Osoba, 1983). Such a pressure drop can lead to change in the pH of the injected brine due to degassing which subsequently impacts on the fluid-rock geochemical interactions and the stability of any previously dissolved minerals in the solution. Mineral dissolution and subsequent precipitation in a relatively long Indiana Limestone core sample has also been reported by Svec and Grigg (2001). As an explanation for this observation, the authors indicate that the injected solution may have become saturated while moving downstream towards the outlet end of the long core, under more neutral conditions and minerals begin precipitating in the void spaces. In other published work it has been argued that the attraction forces between the particles transported and the surface of the pores inside the rock may increase the precipitation rate (Izgec et al., 2006). Furthermore, the reactions and subsequent changes in the petrophysical properties of a rock sample strongly depend on the degree and type of any heterogeneity in the rock and mineral constituents (Oomole and Osoba, 1983). Other important parameters which may impact on the fluid-rock behaviour include the initial permeability of the rock, in-situ temperature, injection flow rate and the duration of the injection period (Moghadasi et al., 2004). When it comes to laboratory core-flood experiments, the flow direction also can be a contributing factor in how the rock properties may change along the length of the rock sample (Khather et al., 2017). With regards to the effect of the flow direction, if a sample is long enough and other experimental conditions (e.g. pressure, temperature, pH, etc.) are favourable, as reported by Svec and Grigg (2001), minerals may be dissolved during the flooding from the inlet section of a sample and precipitate in the outlet section resulting in different variations in the petrophysical properties of the inlet section compared with the outlet section making the flow direction important.

At the injection point where the CO₂-saturated brine comes in contact with the rock, the brine begins to establish preferential flow paths through any pre-existing relatively wider pore channels and relatively high permeability streaks. This behaviour is more pronounced in highly heterogeneous media. Should this be the case, the high flow rate of reactive carbonated brine would rapidly react with the rock minerals within the established flow paths. Subsequently, minerals especially calcite would dissolve in the brine, leading to further increase in the permeability along the established flow paths resulting in the formation of wormholes (Egermann et al., 2005; Izgec et al., 2005; Khather et al., 2017). Also, some clay minerals (if present) react with carbonic acid quite rapidly, which can lead to pore space geometry changes in a relatively short time (Izgec et al., 2008).

In fractured formations, the reactions are likely to occur mainly within the fractures and their close proximity as fractures provide preferential flow paths for the injected fluids. During this process, any change in the fracture surface asperities may subsequently alter the mechanical properties of the fracture, which may lead to the deformation of the fracture aperture (Elkhoury et al., 2013). Depending on the exact nature of such alterations and subsequent events, the fracture aperture may become wider or narrower. Generally, fracture permeability may increase as the fast-flow channels are etched or may decrease as asperities dissolve bringing the fracture walls closer as a result of the overburden stress applied (Polak et al., 2004). Fig. 1

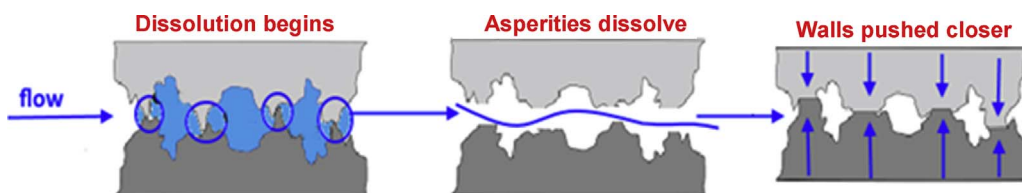


Fig. 1. Dissolution and compaction mechanisms in fractured carbonate rocks.

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