

Reservoir Selection for Optimised Geological Injection and Storage of Carbon Dioxide: A Combined Geochemical and Stratigraphic Perspective.

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Abstract

Selection of a suitable reservoir and seal is a critical aspect for maximising the benefits of CO₂ storage and decreasing leakage risk. Key parameters are: CO₂ injection rate; CO₂ vertical and lateral migration; and chemical interaction with formation waters and minerals.

Reservoir quality needs to be high near the injection point (e.g. permeability >100mD) to allow for effective injection. Away from the point of injection, however, lower reservoir quality is more favourable (e.g. permeability <50mD), as this slows the vertical and lateral migration of the CO₂ plume, providing additional time for CO₂ mineralisation. During the injection phase, CO₂-water-rock interaction could decrease injectivity (e.g. fines migration blocking pore throats). Therefore, a chemically-mature reservoir is preferable to minimise these effects. However, in the longer-term, CO₂-water-rock interaction is likely to be beneficial, permanently trapping the CO₂ through mineralisation. To promote geochemical reactions a chemically-immature reservoir would maximise geochemical trapping.

The ideal reservoir system is therefore a chemically-mature, highly permeable formation for efficient injection, overlain by a chemically-immature, low permeability formation (for maximised effective long-term containment), capped by a thick regional seal. Several examples of this scenario are found within Australian stratigraphy. For example, the Otway Basin has lowstand, fluvial braidplain sediments (high permeability, quartz arenites) overlain by transgressive, nearshore marine sediments (low permeability, feldspathic litharenites). Natural CO₂ accumulations in the area demonstrate the enhanced mineralogical trapping of CO₂ in chemically-immature reservoirs. Reservoir systems with an ideal stratigraphic arrangement of this type provide opportunity for high injectivity and high mineralogical trapping of CO₂.

1 Introduction

Typically, geological storage of carbon dioxide (CO₂) in the initial stages relies on an effective top seal containing the immiscible phase CO₂ in the reservoir rock below. However, the risk of CO₂ leakage increases proportionally with the residence time of the structurally or stratigraphically trapped immiscible CO₂ whilst it remains in that phase. Leakage through wells, via faults/fractures and by capillary pressure failure or diffusion through the top seal, are all ongoing risks for CO₂ to escape the containment area of a geosequestration project.

This work looks at ways of optimising the containment of the CO₂ through geochemical and stratigraphic techniques. By understanding the relationship of depositional processes to the mineralogy and chemistry of the reservoir system, an ideal storage setting that is common in many geological basins can be established for geosequestration.

1.1 Traditional Concepts

Traditionally, an effective reservoir system for CO₂ storage is considered to be a highly porous and permeable reservoir overlain by a thick seal. This situation promotes ease of injection, minimised pressure effects and high pore-space storage capacity. A thick seal is essential for all storage scenarios as it prevents leakage to overlying formations. This situation is also the ideal reservoir system for many successful hydrocarbon provinces, which are often areas considered for CO₂ storage.

Whilst effective injectivity and sufficient storage capacity are important for CO₂ storage, containment is the critical aspect for any storage site to be successful. To achieve a successful CO₂ storage project, the CO₂ needs to be contained within the defined storage area with an acceptably low risk of leakage. The CO₂ 'bubble' sitting below the seal, in this scenario, has a moderate rate of dissolution into the formation water. However, the conversion of CO₂ into other geochemical phases is limited.

2 Geochemical Trapping Mechanisms

The geological storage of CO₂ in the subsurface can occur through several different geochemical mechanisms. These include the containment of an immiscible CO₂-rich phase, CO₂ dissolved and speciated in the aqueous phase (CO₂, H₂CO₃, HCO₃⁻, CO₃⁼) and carbonate mineral precipitation (van der Meer 1992; Gunter *et al.* 1993; Bachu *et al.* 1994; Hitchon 1996). Residual gas trapping promotes the conversion to other geochemical trapping phases. The behaviour of CO₂ in each geochemical phase and the expected or predicted retention time varies according to the storage system.

2.1 Immiscible CO₂ and Dissolution Trapping

CO₂ becomes an immiscible phase when the amount of CO₂ exceeds the amount soluble in water. During injection of CO₂ for storage, the CO₂ starts as a separate phase and over time dissolves into the water. This occurs at the CO₂/water interface and is subject to a rate of dissolution that is dependent on the area of interface and rate at which CO₂-saturated water is transmitted away from the interface or by the diffusion rate driven by the introduced chemical gradient (Denbigh 1971; Bachu *et al.* 1994).

The solubility of CO₂ is controlled by the temperature, pressure and composition of the water. Solubility decreases with increasing temperature and salinity, and increases with pressure (Spycher *et al.* 2003). Once the CO₂ is dissolved, the chemistry of the system controls what will happen in the short- and long-term future.

2.2 Residual Gas Trapping

Low permeability formations (e.g. permeability <50mD) in a reservoir system are responsible for reduction in fluid flow as evidenced by poor hydrocarbon recovery. The presence of clays and cements in these low permeability formations ('thief' zone) result in high residual gas trapping and a higher formation water saturation than high permeability formations (e.g. permeability >100mD). While this effect is detrimental to hydrocarbon production, the impact of higher residual gas trapping and water saturation is beneficial to geosequestration (Ennis-King and Paterson 2001; Flett *et al.* 2004). Residual gas trapping slows the migration rate of immiscible gas in hydrodynamic trapping, and in the case of structural and stratigraphic trapping, reduces the amount of immiscible gas accumulating below the seal. Geochemically, higher water saturation allows more CO₂ to dissolve and increase the potential of mineralogical trapping.

2.3 Mineralogical Trapping

With the excess bicarbonate and suitable cations (Ca²⁺, Mg²⁺, Fe^{2+/3+} or Mn²⁺) precipitation of new stable carbonate minerals is likely to occur. The creation of new minerals containing carbon is known as mineralogical trapping for CO₂ and is the most permanent form of geological storage of CO₂ (Hitchon, 1996; Watson *et al.* 2004). While there is abundant bicarbonate from the dissolution of CO₂ to form carbonate minerals, the limiting factor is the cation source, either from the formation water (limited) or the host minerals (time-dependant). In a high permeability reservoir, the mineralogy of the formation is typically quartz-rich with a minor component of feldspar, clays or occasionally rock fragments. This suite of minerals all have relatively low rates of reaction with CO₂, and would not result in any major mineralogical trapping of CO₂. However, reservoirs containing substantial amounts of reactive clays (i.e. chlorite, berthierine), non-typical cement phases (i.e. glauconite, laumontite), fine-grained feldspars and fine-grained rock fragments (i.e. ferromagnesium minerals) all react readily due to the lower pH resulting from the CO₂ dissolution. Examples of sandstone which contain these types of mineral suites include greensands, ironstones and volcanogenic sediments.

3 Ideal Reservoir System

An ideal reservoir system for CO₂ storage, as proposed in this paper, enhances geochemical trapping mechanisms that are unlikely to have a major role in the traditional concepts mentioned above. The unit for injection should have high reservoir quality (e.g. permeability >100mD) to allow for effective injection. It should also have high chemical maturity to prevent CO₂-water-rock interactions that could decrease injectivity (e.g. fines migration blocking pore throats). As the CO₂ will generally migrate upwards due to buoyancy (under typical reservoir conditions), overlying this should be a lower permeability (e.g. permeability <50mD), more heterogeneous formation. This will slow the vertical and lateral migration of the CO₂ plume and allow more time for dissolution and mineralisation to occur, increasing the containment security. Ideally, this formation should also be chemically-immature, as the higher abundance of reactive minerals will promote geochemical reactions and maximise permanent mineral trapping of the CO₂. A thick regional seal should overlie to prevent further vertical migration (Fig. 1).

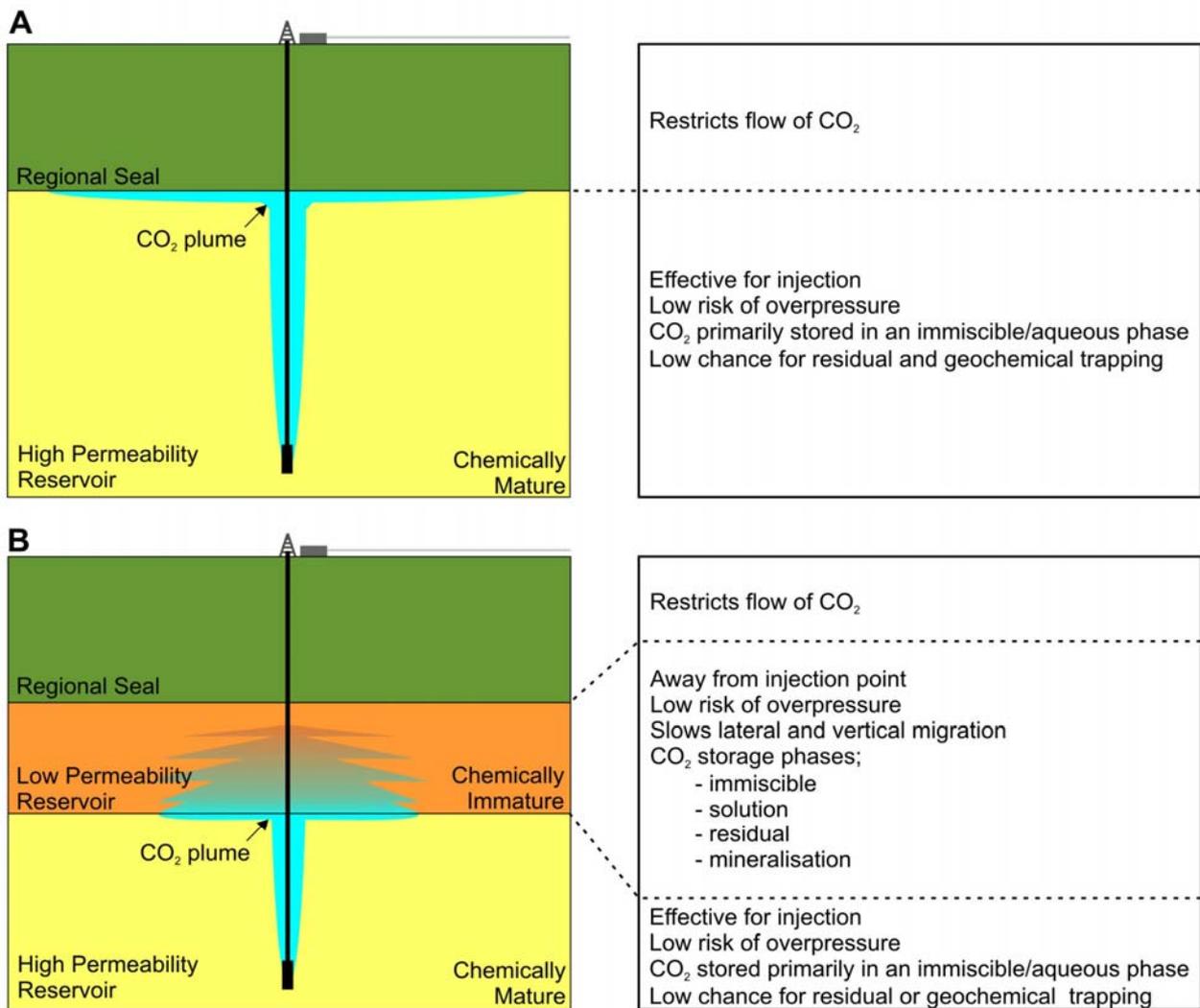


Fig. 1 Conceptual diagram of (A) a traditional storage system for CO₂ showing the migration extent of a CO₂ after time x, vertically due to buoyancy and then laterally along the base of the seal, compared to (B) the optimised storage system for CO₂, where following buoyancy-driven vertical migration the CO₂ encounters a low permeability, heterogeneous zone, slowing both the lateral and vertical migration of the CO₂ plume.

Through knowledge of depositional systems and sequence stratigraphic concepts, the likely stratigraphic arrangements where this type of ideal setting occur can be determined. In the simplest generic terms, a classic example might be a lowstand shoreface succession, overlain by a transgressive marine shelf deposit, topped by highstand marine shales. The fall in relative sea level causes the erosion of exposed sediments, which are then transported to the coastal system. Reworking by currents, tides and waves mean that typically the lowstand shoreface sediments are good quality, chemically-mature reservoir units. A rise in relative sea level would then cause marine shelf sediments to transgress over the top of the shoreface sediments. If the subsequent rise in relative sea level is also accompanied by a reduction in the terrigenous sediment supply, then there is potential for glauconite to develop on the shelf. This would result in finer-grained (lower permeability) greensand sediments, which would have high reactive potential for CO₂ interaction. The continuation of the relative sea level rise would lead to the deposition of highstand basinal marine shales, providing a thick regional seal.

This is only one simple example of how a stratigraphic arrangement of formations can provide an ideal reservoir system for maximised CO₂ storage and containment. For example, high quality reservoir units can be found in most systems tracts depending on where they are located within the depositional system (e.g. transgressive incised valley fills, highstand deltaic sands) and greensands do not always occur only in the transgressive systems tract (e.g. highstand protected bay). However, the simple example discussed above is a common occurrence in passive margin basins around Australia, and in many cases could be the right stratigraphic arrangement to provide the ideal CO₂ storage system.

4 Case Study: Otway Basin, Australia

The Late Cretaceous Waarre Sandstone, Flaxman Formation and Belfast Mudstone of the Otway Basin, Australia, provide an excellent natural analogue for examining the benefits of the ideal reservoir system for CO₂ storage (Fig. 2). Natural CO₂ accumulations in the eastern Otway Basin are commonly reservoirised within the Waarre Sandstone, with the Flaxman Formation sitting stratigraphically above and regionally sealed by the Belfast Mudstone. Understanding how CO₂ storage security can be potentially enhanced is a major benefit when considering successful long-term containment of anthropogenically injected CO₂. To research this issue, core samples and well data over the Waarre Sandstone, Flaxman Formation and Belfast Mudstone interval from the CO₂-rich Boggy Creek-1 well have been studied. Core from the CO₂-free Flaxman-1 well has also been examined as a precursor for the CO₂ reactions.

4.1 Geological Background

The Waarre Sandstone is a mixed lithology which sits unconformably on the Eumeralla Formation. Buffin (1989) has defined the Waarre Sandstone as three sub-units: Unit A (earliest sub-unit), a medium to coarse-grained sandstone inter-fingered with interbedded siltstones; Unit B, a silty, fine-grained sandstone with interbedded siltstone and shale; and Unit C (latest sub-unit), a well developed medium to coarse-grained quartz arenite with excellent reservoir properties. It is within the Waarre C that the majority of CO₂ and hydrocarbon accumulations are found in the eastern Otway Basin (Fig. 2).

The Flaxman Formation is a lower deltaic unit (Boyd and Gallagher, 2001) which sits conformably on the Waarre Sandstone. This formation is comprised of interbedded medium to coarse-grained sandstones, fine-grained sandstone, mudstones, siltstones and thin coal seams, all with common siderite, glauconite, ooids and pellets throughout.

The Belfast Mudstone is a pro-deltaic succession with pale grey to black silty mudstones, interbedded siltstones and fine-grained sandstones (Yu, 1988; Lang *et al.*, 1989). Thin interbedded sandstones within the formation commonly contain authigenic carbonate cements.

Timing of the CO₂ influx into the Waarre Sandstone and Flaxman Formation is yet to be accurately constrained. Helium and carbon isotopic studies of the gas-in-place in the natural CO₂ accumulations have been linked to a magmatic origin (Watson *et al.* 2004). Early volcanism of the Newer Volcanics, to the north

of Boggy Creek-1 is dated at approximately 500,000 years ago. However the most recent volcanic activity in the region is dated at approximately 8,000 years ago (Johnson *et al.* 1989).

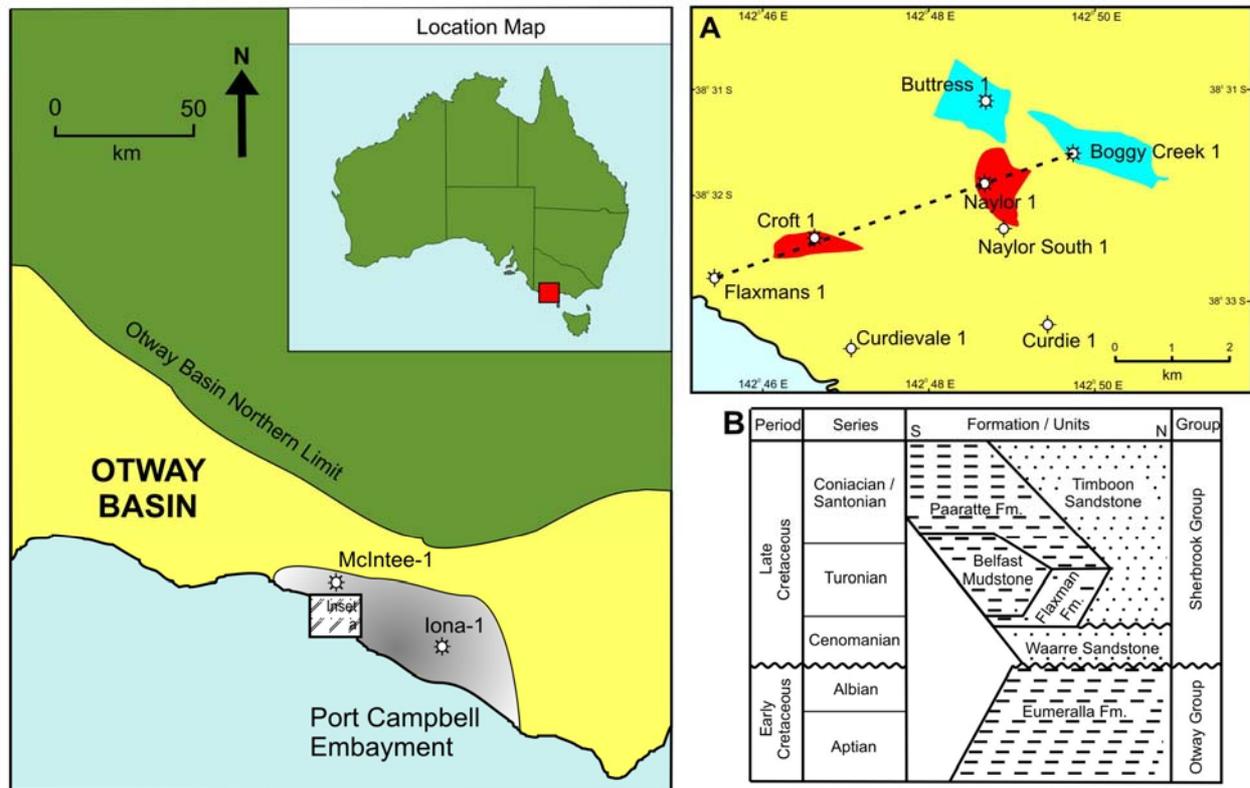


Fig. 2 Location of the Otway Basin, in south eastern Australia, with (A) known wells and gas accumulations (red – hydrocarbon, blue – CO₂) in the study area and (B) stratigraphic table of the Upper Otway Group and Crayfish Group of the Otway Basin.

4.2 Sequence Stratigraphy

Two sequences have been identified over the Waarre Sandstone, Flaxman Formation and Belfast Mudstone interval in the Port Campbell Embayment of the Otway Basin (Fig. 3). Sequence 1 occurs in the lower Waarre Sandstone (units A and B) and comprises a lowstand systems tract (LST) which thickens to the east, overlain by a thick transgressive systems tract (TST) and a thin highstand systems tract (HST). Sequence 1 is interpreted to have been deposited in a restricted tidally-influenced coastal setting (Faulkner, 2000). A fall in relative sea level led to the formation of sequence boundary 2 and a basinward shift in facies. Aggradational coastal-braid plain sediments were deposited during the LST (equivalent Waarre C). This was followed by a marine transgression, during which time TST sediments (Flaxman Formation) were deposited in tidal flat and lagoonal depositional environments (Faulkner, 2000). Continued relative sea level rise led to the HST deposition of prodelta shales of the Belfast Mudstone.

In terms of the ideal reservoir system for CO₂ storage, this stratigraphic arrangement in the second sequence provides high quality, chemically-mature LST sediments suitable for injection, with lower quality, chemically-immature TST sediments overlying for the CO₂ to migrate into. The HST shales provide a thick regional top seal.

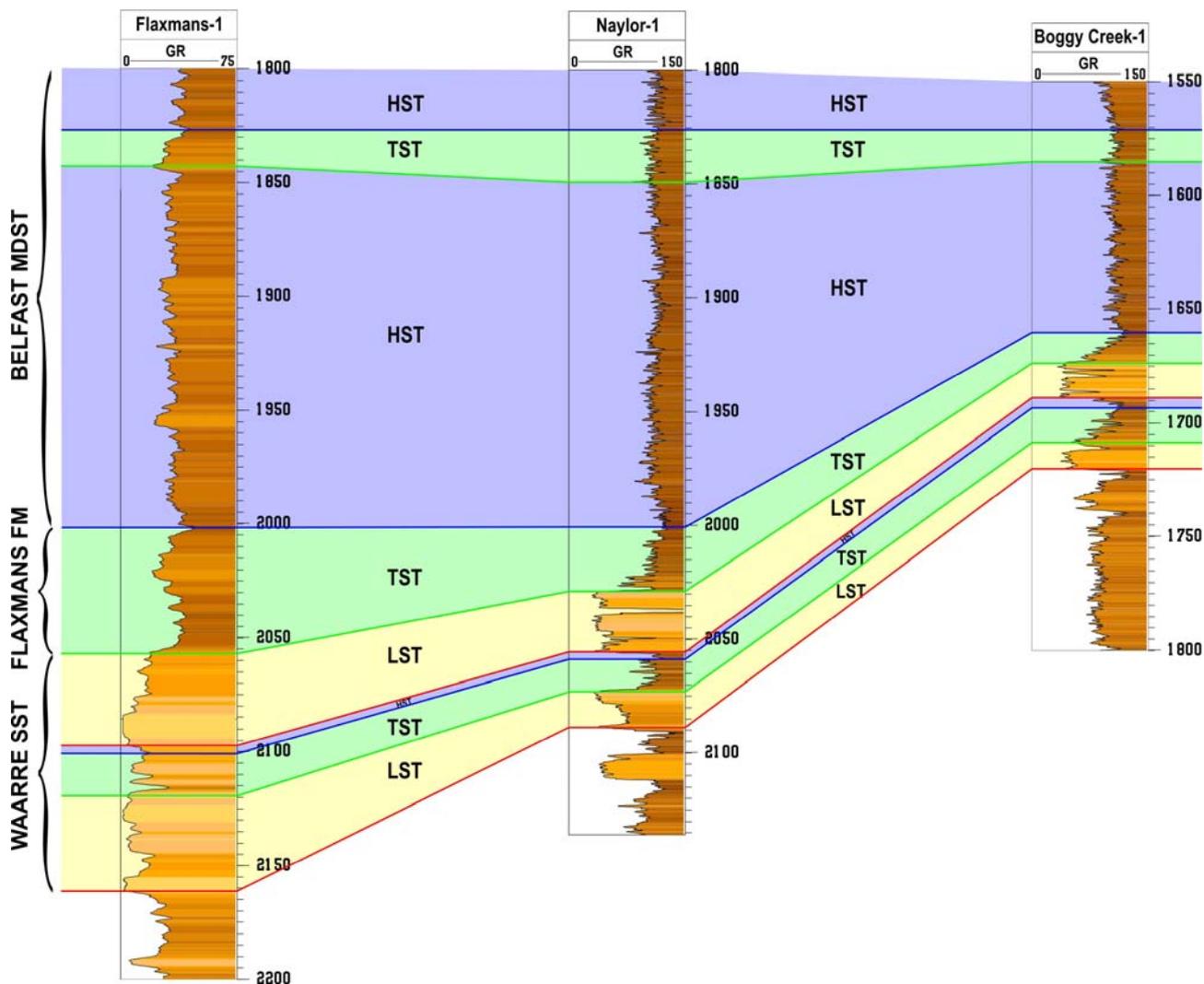


Fig. 3 East-west cross-section indicating sequence stratigraphic interpretation (see Fig. 2 (A) for plan view of well location).

4.3 Geochemical Trapping Potential

Core chips and cuttings were collected from the Waarre Sandstone, Flaxman Formation and Belfast Mudstone from the CO₂-rich (92 mol% CO₂) Boggy Creek-1 well and the non-CO₂ (<1.7 mol% CO₂) Flaxmans-1 well. Petrological evaluation of the samples, including bulk and clay quantitative x-ray diffraction (XRD) analysis, scanning electron microscopy (SEM) examination and thin section examination, were used to quantitatively determine the mineral assemblages of the Waarre Sandstone and Flaxman Formation (Table 1). Through the examination of core from the non-CO₂ precursor well, Flaxmans-1, and comparing it to core from the CO₂-rich well, Boggy Creek-1, CO₂-induced diagenesis was determined and a diagenetic sequence established both pre- and post-CO₂ influx.

Table 1 Mineralogical composition of the Waarre Sandstone and Flaxman Formation pre- and post-CO₂ influx.

Mineral	Waarre Sandstone		Flaxman Formation	
	Pre-CO ₂	Post-CO ₂	Pre-CO ₂	Post-CO ₂
	Flaxmans-1	Boggy Creek-1	Flaxmans-1	Boggy Creek-1
Quartz	89.50%	86.50%	35.50%	31.60%
Potassium Feldspar	2.00%	2.00%	3.00%	-
Albite	1.50%	0.50%	1.00%	-
Anorthite	-	-	1.50%	-
Mica	0.50%	-	3.00%	2.00%
Glauconite	-	-	2.00%	3.00%
Calcite	0.50%	1.00%	1.00%	7.40%
Ankerite	-	0.50%	-	3.00%
Siderite	-	0.50%	23.00%	25.00%
Smectite	0.50%	1.00%	-	5.00%
Illite	0.50%	1.50%	5.00%	9.00%
Kaolinite	3.50%	6.00%	6.00%	12.00%
Chlorite	0.50%	0.00%	-	-
Berthierine	-	-	18.00%	Tr
Pyrite	1.00%	0.50%	1.00%	2.00%

4.3.1 Mineral Reactants and Products: Waarre Sandstone

The Waarre Sandstone is classified as a fine to coarse-grained, poorly sorted quartz arenite sandstone, with large fining-upward sandstone packages and rare interbedded siltstones and mudstones. In the high permeability Waarre Sandstone the Flaxman-1 precursor for CO₂ interaction indicates that there is low reactive potential, due to the dominance of quartz in the mineralogy and limited labile minerals such as feldspars, carbonates and clays. However, CO₂-induced diagenesis was noted in thin section examination of the Boggy Creek-1 Waarre Sandstone, with partial to total dissolution reaction from feldspar to kaolinite, and minor siderite and calcite precipitation (Fig. 4). The dissolution of the feldspar grains has increased the secondary porosity of the formation. However, the occlusion of pore-space through precipitation of kaolinite and carbonate roughly equal the secondary porosity from feldspar dissolution, so porosity variation is minimal (22.5% porosity for Flaxmans-1; 21.7% porosity for Boggy Creek-1).

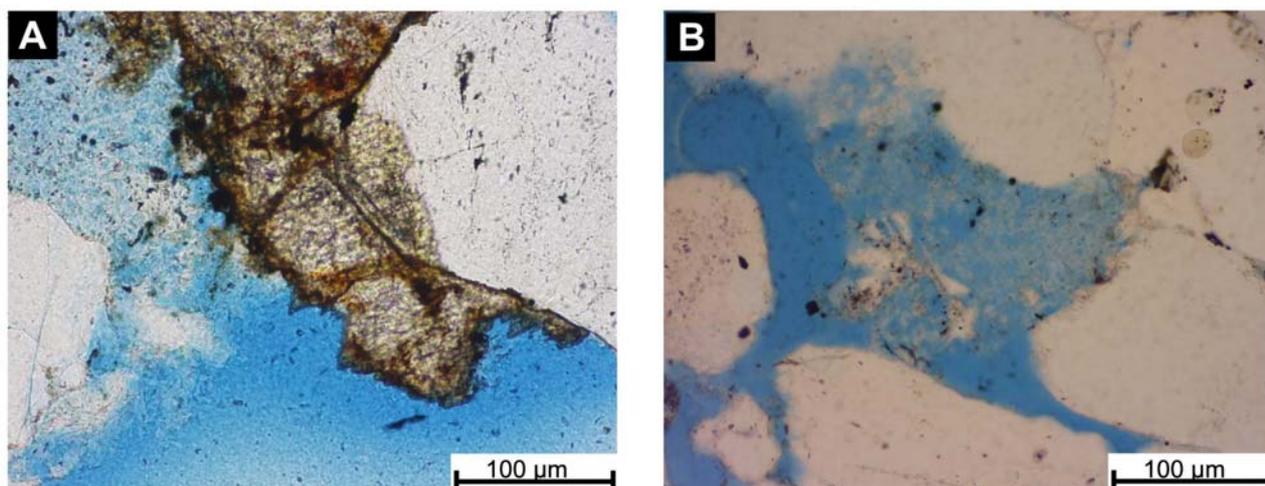


Fig. 4 Thin section photomicrograph of (A: Boggy Creek-1, 1680m) ferroan carbonate (ankerite) and kaolinite precipitation in the pore-space of the Waarre Sandstone as a result of the influx of CO₂ into the formation, and (B: Boggy Creek-1, 1676.4m) a dissolution-precipitation reaction of feldspar to kaolinite, along with the creation of secondary porosity, as a result of CO₂ influx in the Waarre Sandstone.

4.3.2 Mineral Reactants and Products: Flaxman Formation

The Flaxman Formation is a series of mixed interbedded lithologies, including; matrix-rich, very fine to medium-grained, poorly sorted, feldspathic-greywacke greensands; ironstones; glauconitic siltstones; and glauconitic mudstones. The low permeability Flaxman Formation clearly has a chemically-immature mineralogy when compared to the Waarre Sandstone. The formation is co-dominantly quartz and siderite prior to CO₂-interaction, based on Flaxman-1 core samples (Fig. 5a). Berthierine cement is also very high (18%), filling much of the pore-space. Siderite appears to have replaced the syn-depositional glauconite phase from most of the formation and occurs as a framework texture rather than pore-filling cement. The Flaxman Formation in Boggy Creek-1 clearly shows evidence of CO₂ alteration. Berthierine, with its very high surface area, has almost totally been dissolved from the formation at Boggy Creek-1, with only trace amount noted. Siderite, ankerite and calcite have precipitated as a result of the berthierine alteration by CO₂ (Fig. 5b). All feldspar has also been altered to illite (in the case of potassium feldspar) and kaolinite (in the case of plagioclase).

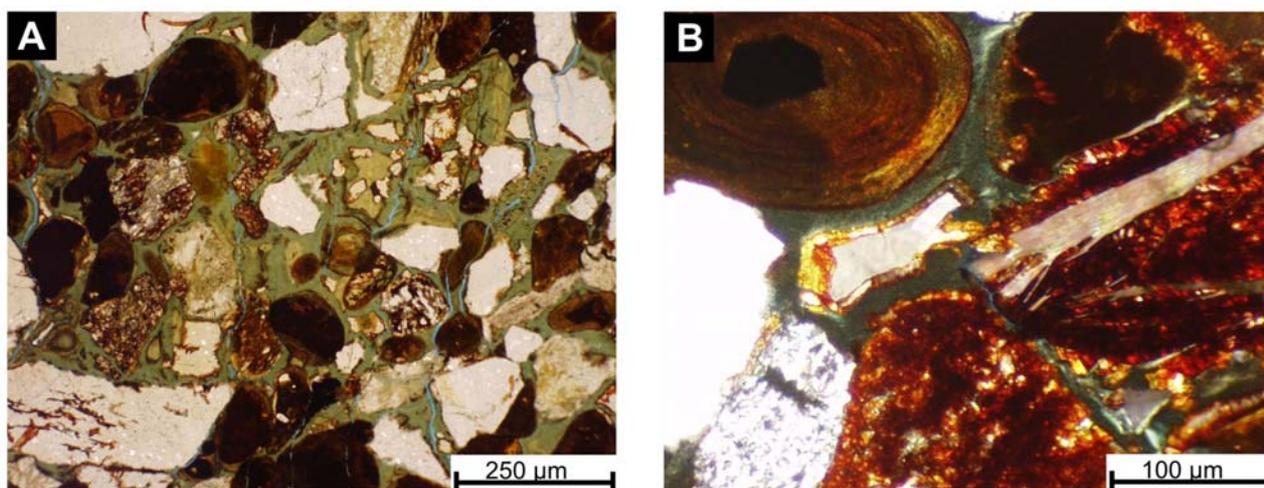


Fig. 5 Thin section micrograph of (A: Flaxman-1, 2020.8m) quartz grains, metamorphic rock fragments and siderite ooids (alteration product of glauconite diagenesis) surrounded by pore-filling berthierine in the Flaxman Formation prior to CO₂ influx and, (B: Boggy Creek-1, 1673m) further precipitation of siderite along grain edges and into pore-space as a result of CO₂ influx into the Flaxman Formation.

4.3.3 Diagenetic Sequence

The detailed paragenetic sequence of the Waarre Sandstone and Flaxman Formation at Boggy Creek-1 is detailed in figure 6. A clear halt to normal diagenesis is observed with the introduction of CO₂, with carbonate cements and kaolinite being the major CO₂-related diagenetic product. Unfortunately, no water chemistry data exists for the formations in this region and the effect of the CO₂ interactions with the water is unknown.

4.3.4 Mineralogical Trapping of CO₂

For mineralogical trapping of CO₂ to occur, reservoir rock requires a sufficient supply of cations to precipitate with the bicarbonate in solution, The lack of labile elements, except for minor amounts of K⁺ (from potassium feldspar) and Na⁺ (from albite and potentially in the formation water) which typically don't form stable minerals in low pH conditions, makes the Waarre Sandstone unsuitable for large-scale mineralogical storage of CO₂ (table 2). However, the Waarre Sandstone did mineralise a minor amount of

carbonate from the CO₂-water-rock interaction, with a carbonate mean increase of 51.3 kg/m³ (CO₂ net storage of 34.3 kg/m³).

Unlike the Waarre Sandstone, the Flaxman Formation had a significantly higher abundance of carbonate and reactive clay (berthierine) minerals that can be reacted to supply Fe^{2+/3+}, Ca²⁺ and Mg²⁺, which can facilitate the development of ferroan carbonate mineralisation. The Flaxman Formation in Boggy Creek-1 demonstrates this mineralisation of CO₂ through the precipitation of additional siderite, ankerite and calcite through the dissolution of berthierine. Earlier siderite does not appear to have been dissolved by the CO₂ influx, preventing the release of additional bicarbonate ions from the mineralogical form. Overall, the Flaxman Formation is a very successful formation for the mineral trapping of CO₂, with a mean carbonate increase of 359.6 kg/m³ (CO₂ net storage of 231.7 kg/m³).

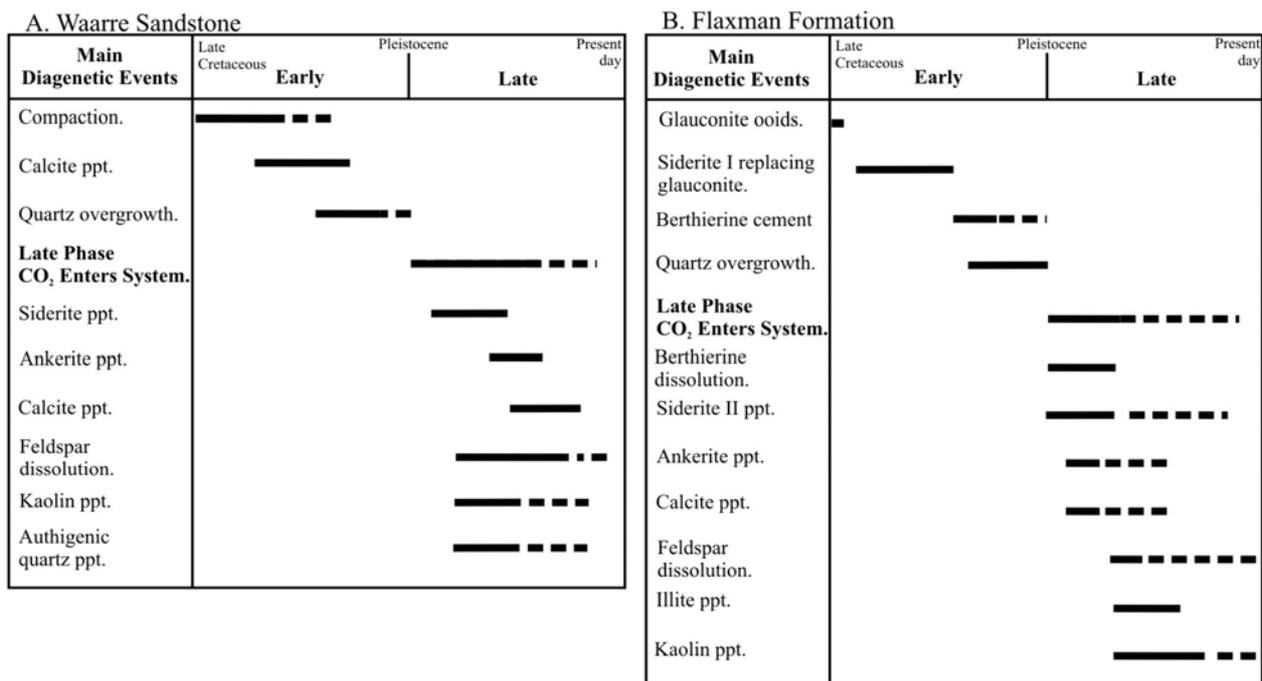


Fig. 6 Paragenetic sequence, with the relative timing of main diagenetic phases and mineralisation in the (A) Waarre Sandstone and (B) Flaxman Formation, pre-and post-CO₂ influx.

Table 2 Calculated carbonate and CO₂ stored, per cubic metre, in the Waarre Sandstone and Flaxman Formation. These calculations are based on mean carbonate compositions, with Flaxmans-1 selected as representative samples for comparison with the carbonate mineralisation in the Boggy Creek CO₂ field.

Formation calculated	Field/Well	Total carbonate in reservoir	Total carbonate content	CO ₂ mineralogically stored	Net CO ₂ storage (early vs. late carbonate)
		%	kg/m ³	kg/m ³	kg/m ³
Waarre Sandstone	Flaxmans-1	0.5	13.5	9.9	Waarre Standard
	Boggy Creek	2	64.8	44.2	34.3
Flaxman Formation	Flaxmans-1	24	946.9	618.1	Flaxmans Standard
	Boggy Creek	34.5	1306.5	849.8	231.7

5 Other Australian Examples

The case study from the Otway Basin highlights how CO₂ storage and containment benefits can be maximised by utilising an ideal stratigraphic and geochemical reservoir system. This example from the Otway Basin is not a unique setting, and many other examples can be found within Australian stratigraphy. Typically, many of these occur in the passive margin basins that are found along the northern, western and southern margins of Australia. In the Petrel Sub-basin, LST-HST shoreface sediments of the Sandpiper Sandstone are overlain by condensed TST marine shelf glauconitic sediments (Euchuca Shoals Formation), and regionally sealed by HST basinal marine shales of the Bathurst Island Group. In the Barrow Sub-basin, LST deep sea fans of the Flag Sandstone are located down-dip from overlying TST Mardie Greensand sediments and regionally sealed by the predominantly HST Muderong Shale. In the Gippsland Basin, nearshore LST-TST coarse clastic sediments of the Latrobe Group are overlain by TST lower shoreface-shelf greensands of the Gurnard Formation, and regionally sealed by HST mudstones and marls of the Lakes Entrance Formation.

6 Future Recommendations/Conclusions

Further investigation is required for the concept of geochemical trapping of CO₂ in typical transgressive formations such as a greensand. With the large degree of mineralisation of CO₂ observed in the Flaxman Formation of the high-CO₂ Boggy Creek-1 well, the rate at which the CO₂ alters the original reservoir mineralogy needs to be considered. If these reactions are very rapid then an impermeable zone could form at the base of the low permeability reservoir, reducing the overall storage volume. If reactions are too slow the CO₂ could migrate through the formation to the base of the seal before any reactions take place. This research could also positively impact enhanced oil and gas recovery in low permeability reservoirs, as the CO₂ could sweep gas trapped in thief zones.

The petrological evidence from the natural CO₂ accumulation at Boggy Creek clearly demonstrates the geochemical benefits of having a lower permeability, chemically-immature formation in the migration pathway for enhancing CO₂ storage. Optimising a geosequestration project through stratigraphic selection of an ideal reservoir system maximises the benefits of geochemical trapping mechanisms. This results in a storage system with increased security compared to traditional geosequestration concepts.

7 Acknowledgements

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