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CARBON SEQUESTRATION IN COAL MEASURES ROCKS

Ian Porter¹, Jae Dawes¹, Long Nghiem¹ and Jim Somerville²

ABSTRACT: It is a widely held belief that anthropogenic addition of greenhouse gases to the atmosphere will lead to rapid climate change. It is also a widely held belief that the burning of fossil fuels for power generation is a major contributor to these anthropogenic additions, predominantly the emission of carbon dioxide and the consequential increase in carbon dioxide levels in the atmosphere. Irrespective of the cause and effect of the increased carbon dioxide levels it is in the interest of the coal mining industry to work with various partners to develop methods of reducing the rate of emission of carbon dioxide to the atmosphere. Carbon capture and sequestration has the potential to meet that objective. This paper looks at the various methods of carbon sequestration and presents the published results from various laboratory and theoretical studies to determine the potential for carbon sequestration in coal measures rocks.

INTRODUCTION

A common consensus is that human development has greatly contributed to increased atmospheric levels of greenhouse gases - particularly methane and carbon dioxide (the 2nd and 3rd most prolific greenhouse contributors, behind water vapour). It has been determined that greenhouse gas emissions have increased since the Industrial Revolution with concentrations of carbon dioxide having risen from 280 ppm to approximately 350-380 ppm (Bachu, 2003; Normile, 2009). With these rises in greenhouse gases it is believed that rapid climate change will occur, leading to unacceptable social and economic problems. The emission of carbon dioxide and the consequential increase in carbon dioxide levels in the atmosphere is of particular concern, and irrespective of the cause and effect of the increased carbon dioxide levels it is in the interest of the coal mining industry to work with various partners to develop methods of reducing the rate of emission of carbon dioxide to the atmosphere.

The most theoretically promising option to reduce carbon dioxide is Carbon Capture and Sequestration (CCS) (Metz, *et al.*, 2005). CCS involves capturing CO₂ from large point emitters (such as power stations) and storing it for a geological time frame. CCS involves:

- separation of carbon dioxide from industrial point sources;
- transport to site;
- long term storage in atmospheric isolation.

According to Metz *et al.* (2005), CCS could reduce CO₂ emissions from pulverised coal power stations by 80-90%. Figure 1 shows the amount of carbon dioxide emitted from a regular power plant in comparison to CO₂ emitted from a power plant with a carbon capture regime implemented. Although more carbon dioxide is produced, due to the increased fuel required per unit of electricity released to the grid, the amount of carbon dioxide emitted is significantly less. The widespread application of CCS depends on future advances in technology. At present the technology exists to allow for each individual stage of CCS, e.g. CO₂ has been pumped underground for use in Enhanced Oil Recovery (EOR), the technology to drill wells deep into the earth has been proven by the successful exploitation of natural oil and gas reserves, the use of pipelines to distribute oil and gas is a routine occurrence, however, the various technologies have not been put together into a system that can economically and safely sequester carbon dioxide on a geological timeframe.

Studies have shown that CO₂ can be theoretically stored in the ocean (Marchetti, 1977; Holloway and Savage, 1993), soil (McCarl, *et al.*, 2007), saline aquifers (Allen, *et al.*, 2005; Bachu, *et al.*, 1994), coal (Bachu, 2000; Larsen, 2004; Fitzgerald, *et al.*, 2005; Shackley and Gough, 2006; Orr, 2009) and sandstone (Korbol and Kaddour, 1995; Nowak, 2007).

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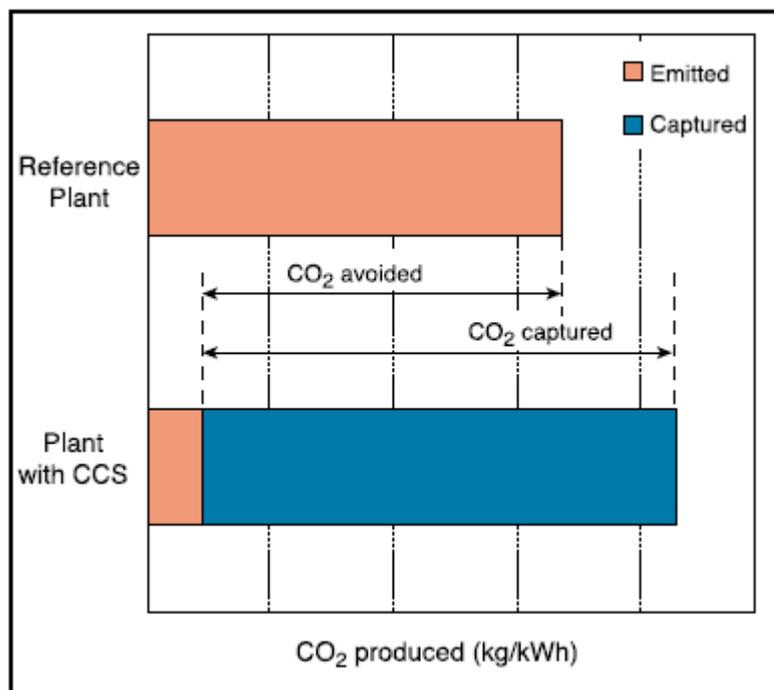


Figure 1 - CO₂ emissions for power stations with and without capture. (Metz, *et al.*, 2005)

OCEAN SEQUESTRATION

According to Metz *et al.* (2005) there are several viable methods for ocean sequestration:

- dissolution - CO₂ is injected by ship or pipe at depths greater than 1000 m where the carbon dioxide dissolves into the water and becomes part of the equilibrium;
- lakes - carbon dioxide is deposited onto the sea floor deeper than 3000 m. At these depths CO₂ is denser than sea water and is capable of forming lakes which settle on the floor and prevent the spread of CO₂ into the environment;
- conversion - converting CO₂ into solid bicarbonates (with the use of limestone) and storing them in the ocean. Bicarbonates are extremely stable and would be capable of withstanding the erosion and pressures that they would be subjected to at great depths that are required for sequestration;
- store CO₂ in solid clathrate hydrates already on the ocean floor, or create new hydrates.

Each of these systems has merit, but an obvious demerit is the potential effect on the ocean equilibrium and subsequent effect on ocean ecosystems.

SOIL SEQUESTRATION

It has been suggested (McCarl, *et al.*, 2007; Parliamentary Library, 2011) that sequestering carbon into soil could improve the health and productivity of the soil. There are two ways to sequester carbon in soils; increase carbon levels in soils or convert organic matter into a more stable form of carbon. The former involves returning plant material to the soil. McCarl *et al.* (2007) concluded that when plant material was returned to the soil or the amount of lost carbon reduced, or both, then the overall health of the soil would improve. For the latter biochar can be considered as a viable option (Krull, 2010; McCarl, *et al.*, 2007; Parliamentary Library, 2011; Victoria Parliament, 2010). Biochar is a charcoal that is produced when organic materials (manure, wood chips, crops, coal) are burned in an oxygen-depleted environment; the process is conducted at high temperatures and is known as pyrolysis (Krull, 2010). It utilises production processes similar to coke making.

The sequestration of carbon dioxide into soils is dependent on a number of factors. The climate (temperature and precipitation), geology of the surrounds (texture, mineralogy), vegetation types and the land management practises employed all affect the ability of soil to sorb and store carbon dioxide (McCarl, *et al.*, 2007; Victoria Parliament, 2010). These factors all affect the soil residence time (Jastrow, *et al.*, 2007), i.e. the time for the stored carbon to be released back into the atmosphere. There is evidence that the residence time associated with biochars can range from hundreds of years to several thousand of years (Lehman, *et al.*, 2008). Figure 2 (Lehman, 2007) shows the natural carbon cycle in comparison to the biochar carbon cycle.

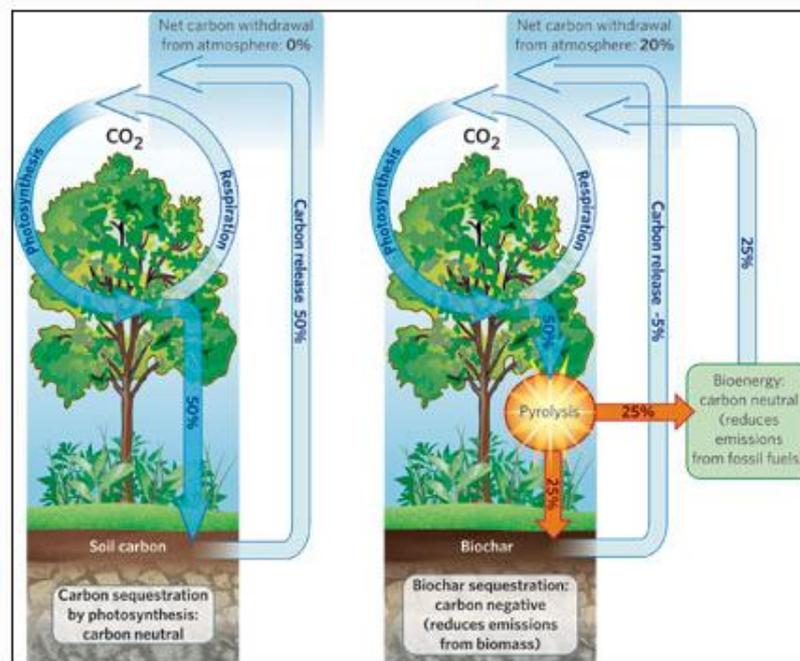


Figure 2 - Natural carbon cycle verse biochar carbon cycle (Lehmann, 2007)

Whatever the merits or demerits of soil sequestration there must be an ability to store CO₂ for geologically significant periods.

GEOLOGICAL SEQUESTRATION

There is general agreement (Metz, *et al.*, 2005; IEA, 2004; Bachu, 2003; Holloway, 1997; Orr, 2009) that the most suitable geological formations, as illustrated in Figure 3, for carbon sequestration are:

- deep saline aquifers;
- depleted oil and gas reservoirs (including coal bed methane);
- rock formations, such as sandstone, limestone and shale, and uneconomical coal seams.

Sequestration in deep saline aquifers and depleted oil and gas reservoirs

There are currently two significant commercial projects; Sleipner, Norway and In Salah, Algeria, that utilise deep saline aquifers for CO₂ storage. These two projects are similar, in that they harvest the CO₂ from extracted CH₄, and store it in deep saline aquifers. The Sleipner project is the world's longest running sequestration project, with the first CO₂ sequestration occurring in 1996 (Korbol and Kaddour, 1995). Sleipner utilises a deep saline aquifer for sequestration that sits around 1700 m above the gas deposit, Figure 4, while In Salah exploits a saline aquifer down dip of gas production. Both of these projects sequester approximately 1Gt CO₂ annually (Metz, *et al.*, 2005).

Storage of CO₂ is a 'by-product' of EOR at the Weyburn oil field in Canada. The CO₂ is pumped into the formation to drive out the last drops of oil that would otherwise be irrecoverable and in the process is captured in the sandstone formation.

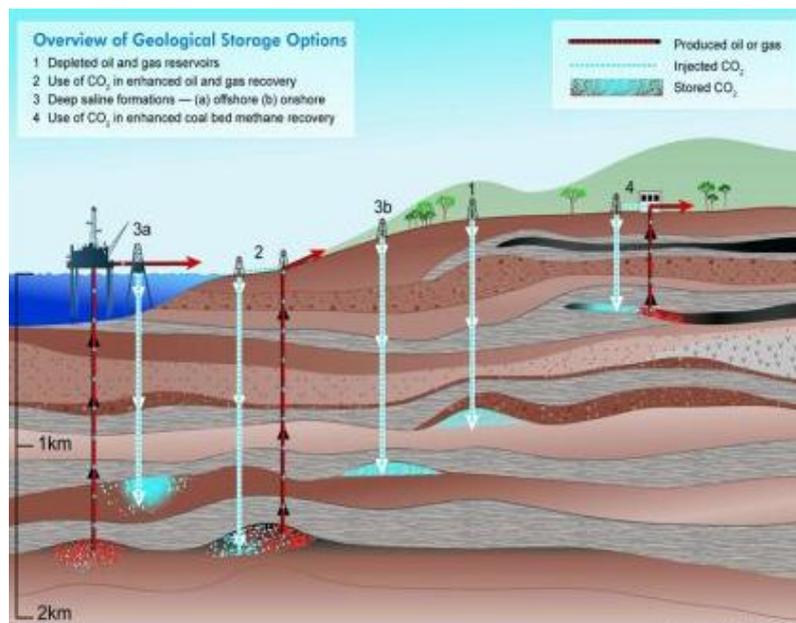


Figure 3 - Methods for storing CO₂ in underground geological formations (Metz, *et al.*, 2005)

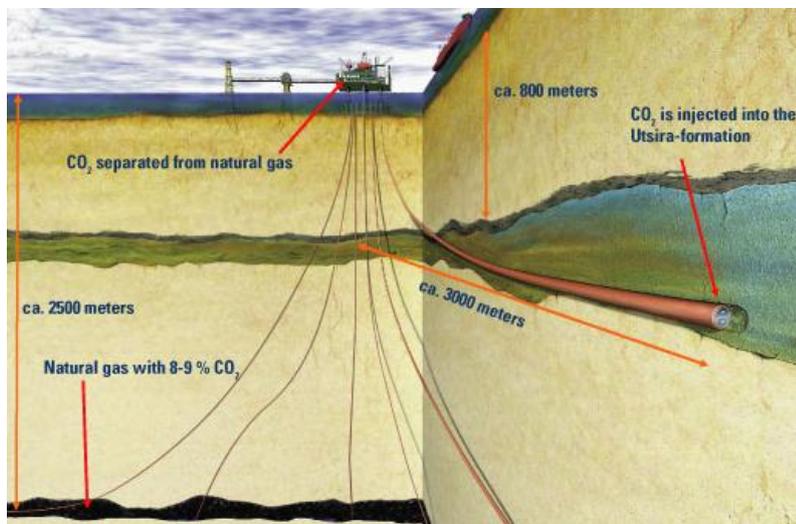


Figure 4 - Carbon dioxide injection at the Sleipner gas field (image courtesy of Statoil)

Sequestration in coal measures rocks

For CO₂ to be held in geological formations some form of trapping mechanism must be present, this may be in the form of an impermeable caprock, dissolution in formation water or geochemical trapping where the CO₂ is mineralised. Geochemical trapping results in the most stable form of carbon storage, but this may take thousands of years and as such some form of lower order trapping must initially take place.

Carbon dioxide is stored in coal measures rock by the process of adsorption (often simply termed sorption), where the gas is held on the rocks' surface. With this process, coal for example, may theoretically hold 40 to 60 m³ of CO₂ per tonne of coal at 5 - 6 MPa (Gaucher, *et al.*, 2011). In addition to storing CO₂ in uneconomical seams, the CO₂ can be used in an Enhanced Coal Bed Methane (ECBM) operation, Figure 5, as CH₄ is preferentially displaced by CO₂ in coal.

Charrière *et al.* (2010) conducted tests on the effects of temperature and pressure on the diffusion of CH₄ and CO₂ into coal. They found that at a pressure of 5.15 MPa a coal from the Lorraine Basin, France, could adsorb 1.55 mmol/g of CO₂, approximately 34 m³/t. Similar experiments have been conducted elsewhere (Busch, *et al.*, 2003) where over 54 m³/t was adsorbed on a coal from Illinois. A common element in these works is the use of a small sample mass, 5 g or less, of finely powdered coal, and

temperature and pressures that are sub-critical for CO₂. The results are then used to infer what would happen *in situ*.

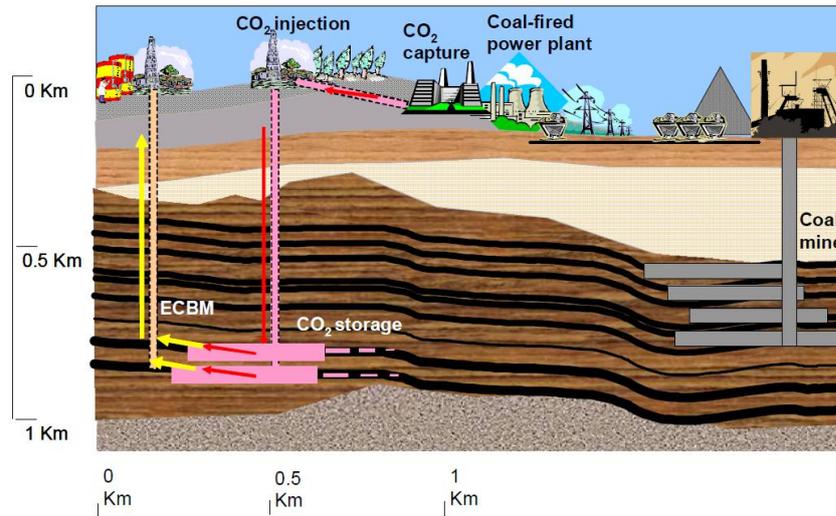


Figure 5 - Enhanced coal bed methane operation (Gaucher, *et al.*, 2011)

Florentin *et al.* (2009) reported on work conducted on coal samples of varying particle size, up to 54 mm diameter core. The findings showed similar total sorption capacities between the varying particle sizes, however the time to reach equilibrium was longer in the solid core samples. It takes at least three days to reach the equilibrium pressure in coal fragments and four days in 54 mm core. In coal fragments, around 70% of the equilibrium pressure was reached during the first 360 min, while in the 54 mm core only 30% of the equilibrium pressure was reached in the same period. As before these results were obtained from samples at sub-critical temperatures and pressure.

Supercritical fluids are substances that are at a temperature and pressure greater than the critical point, Figure 6, such that they cannot be identified as a liquid or a gas. They are capable of effusing through solids (like a gas) and dissolving materials (like a liquid). The most important sequestration property of supercritical fluids is the greatly increased density. When the density increases, the volume of a gas reduces, as such, high density CO₂ takes up much less volume than low density gas, allowing more carbon dioxide to be stored in the same volume. Due to the pressure and temperature requirements to keep CO₂ in the supercritical phase, carbon sequestration occurs at depths greater than 800 m. This is the approximate depth at which the earth's temperature and pressure matches that of supercritical CO₂ - 31.1°C and 7.39 MPa. There are, of course, variations in this due to geothermal gradients and density of surrounding rocks (Bachu, *et al.*, 1994).

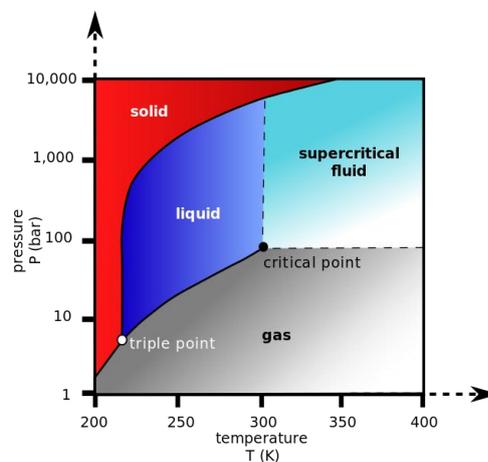


Figure 6 - Phase diagram for carbon dioxide (open source)

Zhang *et al.* (2011), shows that the sorption capacity for CO₂ increases with pressure, but at some pressure beyond the critical pressure it then decreases. This may be a result of the swelling behaviour of the coal under different gas pressures (Day, *et al.*, 2010). These studies and the work reported previously have all been in coal, but the rocks of the coal measures all have the ability to sequester CO₂, obviously to varying degrees.

Sequestration in sandstone and limestone is usually in the context of storage in deep saline formations as discussed previously, however, other coal measures rocks such as carbonaceous shales, particularly those rich in carbonaceous matter (MRSCP, 2005) have the ability to adsorb CO₂ in a manner similar to that of coal. It was reported (MRSCP, 2005) that these shales are often multifunctional; acting as seals for underlying reservoirs, as source rocks for oil-and-gas reservoirs, and are unconventional gas reservoirs themselves. As with ECBM extraction, CO₂ injection into unconventional carbonaceous shale reservoirs could be used to enhance existing gas production. It is believed the carbonaceous shales would adsorb the CO₂ into the shale matrix, similar to coal, permitting long-term CO₂ storage, even at relatively shallow depths (Nuttall, *et al.*, 2005).

Testing core samples under supercritical conditions

As stated previously, most laboratory testing is conducted on small samples of powdered coal, this is particularly true of tests conducted at supercritical conditions. In order to test core samples at supercritical conditions a new high pressure bomb had to be designed. The bomb was based on those used by Florentin *et al.* (2009). Australian Standard, AS1210-2010 Pressure Vessels (Standards Australia Limited, 2010), was used to guide the design process and the final design is shown in Figure 7. For the design and construction of the bomb, marine grade aluminium was chosen as it is resistant to gas erosion. To conform to Australian Standards:

- the bomb must be categorized according to its hazard level;
- the design pressure and operating pressure must allow for a reasonable factor of safety;
- extra precautions must be in place for materials that undergo a phase change whilst contained in the vessel;
- design drawings must be to Australian Standards;
- machining and construction must be done by a competent person;
- the final design needs to be audited by a qualified certifier.

In order to avoid weak spots caused by welds or stress concentrations where the base meets the side wall, the vessel was machined from solid bar and incorporates a hemispherical finish to the base. The bomb is capable of taking 100 mm long 54 mm diameter cores at pressures up to 10 MPa.

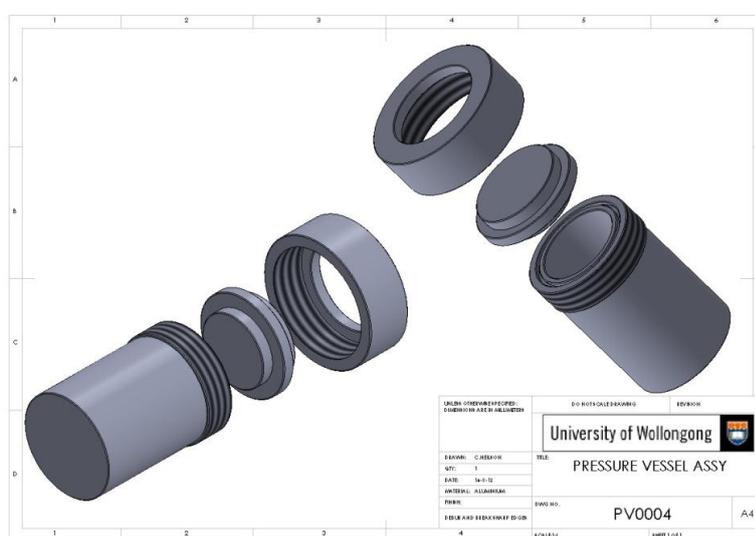


Figure 7 - Pressure vessel for use at supercritical pressures

CONCLUSIONS

At present the only practical application of carbon sequestration is in deep saline aquifers and depleted oil reservoirs. Initial findings have shown that carbon sequestration in these sinks is both feasible and practical. The problem with sequestering carbon dioxide from coal fired power stations in large saline aquifers and depleted oil fields is that these sites are often at some distance from the power station, requiring expensive and expansive transport solutions to get the carbon dioxide to the site.

A potential solution is to utilise coal measures rocks to sequester CO₂. As coal has the greatest sorption capacity by some measure over other coal measures rocks, it would appear that storing CO₂ in uneconomical coal seams, possibly as a by product of enhanced coal bed methane extraction, is the most cost effective and efficient option. The major problem with this approach is that a coal seam that is uneconomical at present may not be in the future. Some countries, the UK for example, have for that reason specifically legislated that CO₂ cannot be sequestered in coal seams.

Although both sandstone and limestone can sequester about 30% the volume of CO₂ that coal can, there is much more of it and, at depth, it is not constrained by being a potential resource. It is necessary to test samples of these rocks under supercritical conditions as they may behave differently to that of coal at elevated pressures. Another potential coal measures rock is carbonaceous shale, where evidence suggests that some shales with high organic content adsorb carbon in a manner similar to coal. Thus, even if legislation or future potential prevents the use of uneconomical coal seams for sequestering CO₂, other coal measures rocks may provide a local solution for coal fired power stations.

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REFERENCES

- Allen, D E, Strazisar, B R, Soong, Y and Heges, S W, 2005. Modelling carbon dioxide sequestration in saline aquifers: significance of elevated pressures and salinities, *Fuel Processing Technology*, 86:1569-1580.
- Bachu, S, 2000. Sequestration of CO₂ in geological media: criteria and approach for site selection in response to climate change, *Energy Conversion and Management*, 41(9):953-970.
- Bachu, S, 2003. Screening and ranking of sedimentary basins for sequestration of CO₂ in geological media in response to climate change, *Environmental Geology*, 44:277-289.
- Bachu, S, Gunter, W D and Perkins, E H, 1994. Aquifer disposal of CO₂: hydrodynamic and mineral trapping, *Energy Conversion and Management*, 35(4):269-279.
- Brennan, S T and Burruss, R C, 2003. Specific sequestration volumes: a useful tool for CO₂ storage capacity assessment, in *Proceedings 2nd annual conference on Carbon Sequestration*, pp 1-12 (National Energy Technology Laboratory, USA).
- Busch, A, Gensterblum, Y and Krooss, B M, 2003. Methane and CO₂ sorption and desorption measurements on dry Argonne premium coals: pure components and mixtures, *International Journal of Coal Geology*, 55:205-224.
- Charrière, D, Pokryszka, Z and Behra, P, 2010. Effect of pressure and temperature on diffusion of CO₂ and CH₄ into coal from the Lorraine basin (France), *International Journal of Coal Geology*, 81:373-380.
- Day, S, Fry, R, Sakurovs, R and Weir, S, 2010. Swelling of coals by supercritical gases and its relationship to sorption, *Energy Fuels*, 24:2777-2783.
- Fitzgerald, J E, Zhejun, P, Sudibandriyo, M, Robinson Jr, R L, Gasem, K A M and Reeves, S, 2005. Adsorption of methane, nitrogen, carbon dioxide and their mixtures on wet Tiffany coal, *Fuel*, 85:2351-2363.
- Florentin, R, Aziz, N, Black, D and Nghiem, L, 2009. Sorption characteristics of coal, particle size, gas type and time, in *Proceedings Underground Coal Operators Conference COAL2009*, pp 208-216 (The Australasian Institute of Mining and Metallurgy: Melbourne). <http://ro.uow.edu.au/coal/102/>.
- Gaucher, E C, Défossez, P D C, Bizi, M, Bonijoly, D, Disnar, J R, Laggoun-Défarge, F, Garnier, C, Finqueneisel, G, Zimny, T, Grgic, D, Pokryszka, Z, Lafortune, S and Vidal Gilbert, S, 2010. Coal laboratory characterisation for CO₂ geological storage. *Energy Procedia*, 4:3147-3154.

- Holloway, S and Savage, D, 1993. The potential for aquifer disposal of carbon dioxide in the UK, *Energy Conversion and Management*, 34(9-11):925-932.
- Holloway, S, 1997. Safety of the underground disposal of carbon dioxide, *Energy Conversion and Management*, 38(Supplement):S241-S245.
- International Energy Agency, 2004. Prospects for CO₂ capture and storage, (online). Available from: <<http://www.oecd-ilibrary.org/docserver/download/6104281e.pdf>> [Accessed: 19 December, 2012].
- Jastrow, J D, Amonette, J E and Bailey, E L, 2007. Mechanisms controlling soil carbon turnover and their potential applications for enhancing carbon sequestration, *Climate Change*, 80(1-2):5-23.
- Korbol, R and Kaddour, A, 1995. Sleipner vest CO₂ disposal - injection of removed CO₂ in the Utsira Formation, *Energy Conversion & Management*, 36(6-9):509-512.
- Krull, E, 2010. Biochar, (online). Available from: <www.csiro.au/files/files/pnzp.pdf> [Accessed: 19 December, 2012].
- Larsen, J W, 2004. The effects of dissolved CO₂ on coal structures and properties, *Coal Geology*, 57:63-70.
- Lehmann, J, 2007. A handful of carbon, *Nature*, 447:143-144.
- Lehmann, J, Skjemstad, J, Saran, S, Carter, J, Barson, B, Falloon, P, Coleman, K, Woodbury, P and Krull, E, 2008. Australian climate - carbon cycle feedback reduced by soil black carbon, *Nature Geoscience*, 1:832-835.
- Marchetti, C, 1977. On geoengineering and the CO₂ problem, *Climatic Change*, 1(1):59-68.
- McCarl, B A, Metting, F B and Rice, C, 2007. Soil carbon sequestration, *Climatic Change*, 80(1-2):1-3.
- Metz, B, Davidson, O, de Coninck, H, Loos, M and Meyer, L, 2005. *IPCC special report on carbon dioxide capture and storage*, 431 p (Cambridge University Press, New York.)
- Midwest Regional Carbon Sequestration Partnership, 2005. Phase 1 Final Report, (online). Available from:< http://216.109.210.162/userdata/Phase%201%20Report/MRCSP_Phase_I_Final.pdf> [Accessed: 19 December, 2012].
- Normile, D, 2009. Round and round: a guide to the carbon cycle, *Science*, 325:1642-1643.
- Nowak, 2007. Biggest carbon burial test will hunt for leaks, (online). Available from: <www.newscientist.com/article/dn11197?DCMP=Matt.Sparkes&nsref=carbon-burial> [Accessed: 19 December, 2012].
- Nuttall, B C, Drahovzal, J A, Eble, C F, and Bustin, R M, 2005. CO₂ sequestration in gas shales of Kentucky, In *Abstracts Volume AAPG 2005 Annual Convention*, pp A101-A102 (American Association of Petroleum Geologists: Tulsa).
- Orr Jr, F M, 2009. Onshore geologic storage of CO₂, *Science*, 325: 1656-1658.
- Parliamentary Library, 2011. Carbon Sequestration, (online). Available from: <<http://www.aph.gov.au/library/pubs/ClimateChange/responses/mitigation/carbon.htm>> [Accessed: 19 December, 2012].
- Standards Australia Limited, 2010. AS1210- 2010 Pressure Vessels, 420 p (SAI Global Limited, Sydney).
- Shackley, S and Gough, C (eds), 2006. Carbon capture and its storage: an integrated approach, Ashgate Publishing Limited, Hampshire.
- Singh, T, 2010. Carbon sequestration- is burying carbon dioxide a realistic option? (online). Available from: <<http://oilprice.com/Environment/Global-Warming/>> [Accessed: 19 December, 2012].
- Victoria Parliament, 2010. Inquiry into soil sequestration in Victoria, Parliamentary Paper 362, (online). Available from: <<http://www.parliament.vic.gov.au/images/stories/committees/enrc/20100902.enrc.scsv.FINREP.pdf>> [Accessed: 19 December, 2012].
- Zhang, D F, Cui, Y J, Liu, B, Li, S G, Song, W L and Lin, W G, 2011. Supercritical pure methane and CO₂ adsorption on various rank coals of China: experiments and modelling, *Energy Fuels*, 25:1891-1899.