Geological Storage of Carbon Dioxide: The Role of Sedimentary Basins

William D. Gunter, Alberta Research Council, Edmonton, Alberta, T6N 1E4, Canada &

Stefan Bachu, Alberta Geological Survey, Edmonton, Alberta, T6B 2X3, Canada

Abstract

Sedimentary basins, occuring throughout the world, are thick piles of geologically deposited sediments that are the hosts for fossil fuel deposits. They may become even more important in the future if their large storage capacity is utilized for disposing of carbon dioxide. Sedimentary basins are dynamic, in the sense that they have an intricate plumbing system defined by the location of high and low permeability strata that control the flow of fluids throughout the basins and define "hydrogeological" traps. The most secure type of hydrogeological trapping is found in oil and gas reservoirs in the form of "structural" or "stratigraphic" traps, termed "closed" hydrogeological traps which have held oil and gas for millions of years. Obviously, these would be very attractive for CO₂ storage due to their long history of containment. A second type of hydrogeological trapping has been recognized in aquifers of sedimentary basins that have slow flow rates. The pore space in such "open" hydrogeological traps is usually filled with saline ground or formation water. A volume of CO₂ injected into a deep open hydrogeological trap can take over a million years to travel updip to reach the surface and be released to the atmosphere. Although the capacity of structural/stratigraphic traps for CO₂ storage is small relative to open hydrogeological traps in deep sedimentary basins, they are likely to be used first as they are known to be secure, having held oil and gas for geological time. As the capacity of closed traps is exhausted and more is learned about geochemical trapping, the large storage capacity available in open hydrogeological traps will be utilized where security of the geological storage of CO_2 can be enhanced by geochemical reactions of the CO₂ with basic silicate minerals to form carbonates. Potential short circuits to the surface through faults or abandoned wells must be located and their stability evaluated before injection of CO₂. In any event, a monitoring scheme must be put in place. This CO₂ storage technology could be the basis for transition from a fossil fuel based energy economy to one based on renewables over the next 100 years.

Transition of the Global Energy Mix

Reducing GHG emissions to the atmosphere by storage of carbon dioxide in geological formations is a potential bridging technology to allow the continued use of fossil fuels while minimizing their effect on the environment. We think that the world is moving from the current GHG emission-intensive fossil fuel energy-based economy toward a GHG emission-free renewable-based energy economy. Here we lump biomass in with fossil fuels as it is the precursor of fossil fuels and burning of it is emission-intensive. Given that energy use will increase in the future, that the current percentage of clean renewable energy in the energy mix is small and that the total amount of energy consumed by the world is huge, the ramp up to a dominantly renewable energy-based world will take place slowly over the next 100 years. During that time we cannot

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continue to use fossil fuels, particularly coal, in a "Business As Usual" (BAU) approach without irreversible damage to the environment.

The concept of sequestration of CO₂ in biomass or storage in geological formations (reviewed by Gunter et al., 1998) leads to bridging technologies which allow fossil fuels to continue to dominate the energy mix but with lowered GHG emissions, termed here "Zero Emission Fossil Fuels" (ZEFF). We think that this approach will move forward in two sequential "Sequestration" steps as illustrated in Figure 1. The first transition to ZEFF from BAU (i.e. conventional burning of fossil fuels to produce heat and electricity with emissions released to the atmosphere) is modified by the addition of downsteam technologies for the diffuse sequestration of the CO₂ into biomass or by downstream separation of the emissions into a pure CO₂ stream which is injected into and stored in deep geological formations. Both options will be utilized. In the former case, the additional cost is low; land-use constraints are the determining factor. In the latter case, the additional cost is much higher, driven by the expense of capturing a pure CO_2 stream from the emission stream (Figure 2). In the second transition to ZEFF, the fossil fuel is gasified (rather than burned)(which produces additional environmental benefits) to produce heat, electricity and hydrogen, and a pure CO₂ waste stream is created as part of the conversion step, eliminating the expense of downstream separation but still requiring biomass or geological storage. The hydrogen is used to power fuel cells to make electricity in stationary plants or to drive mobile transportation vehicles.

"Making the Energy Transition from Combustion to Zero Emissions"

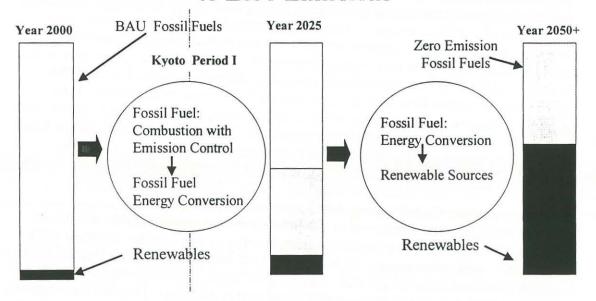


Figure 1: The role of sequestration in the energy path leading to renewables: $BAU \rightarrow$ sequestration through downstream emission control and storage of CO_2 in geological formations \rightarrow sequestration through energy conversion and storage of CO_2 in geological formations \rightarrow renewables.

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In the final transition, the energy mix is dominated by the renewable energy sources: solar, hydro and wind with solar making the largest contribution (Figure 1). Fuel cells will still dominate the transportation market, requiring a hydrogen source but generated from renewables instead of fossil fuels.

The whole process is driven financially by emission trading in the sequestration transition stages where CO_2 emission permits are sold by the government and traded on the open market. A portion of the profits from the sale of these permits could be used to fund renewable energy research, development, demonstration and deployment. This would allow an orderly capacity-building of renewable energy sources without severely affecting our economy and standard of living. If this scenario is correct, geological storage in sedimentary basins of emissions from power plants and other industries will have an important role to play in the future as one of several bridging technologies.

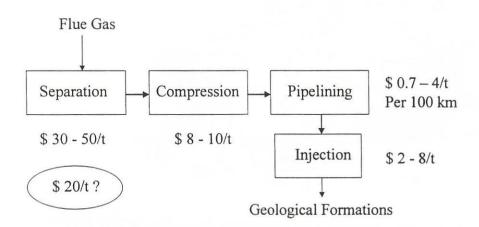


Figure 2: Cost of storage of carbon dioxide extracted from flue gas into deep geological formations. The most expensive part of the process is the separation of CO_2 from the flue gas. It is hoped that new technologies will reduce this part of the cost by more than 50%.

Sedimentary Basins

There is a natural association of sedimentary basins and fossil fuels. Therefore, we should expect a relation between the sedimentary basin, the exploitation of its fossil fuels, and the resulting greenhouse gas emissions. Carbon dioxide is the dominant greenhouse gas resulting from the burning of fossil fuels, and it comprises more than half of all man-made greenhouse gas emissions. Among the methods proposed for the mitigation of greenhouse gas emissions, specifically carbon dioxide, is disposal into porous formations deep in sedimentary basins. This includes injection into hydrocarbon reservoirs to enhance oil and gas recovery (a mature technology), injection into depleted hydrocarbon reservoirs and the long-term sequestration in aquifers (not well understood). The methodology for proving the latter concept has been developed in the Alberta Basin, Canada (Figure 3) and is being practiced in the North Sea. A further development is the concept of injecting carbon dioxide, from the burning of fossil fuels, into coal-beds to remove methane (Gunter et al, 1997a; an immature technology). This would have the dual result of increasing the production of methane, a more environmentally friendly fossil fuel than coal or oil, and using waste carbon dioxide to a useful purpose. While burning the recovered methane will result in more carbon dioxide, clearly this additional carbon dioxide can either be used to recover more methane or be disposed of underground in suitable aquifers. Hitchon et al. (1999) were one of the first to point out this serendipitous association of sedimentary basins, their contained fossil fuels, and the means of exploiting or disposing of the greenhouse gases produced from the fossil fuels.

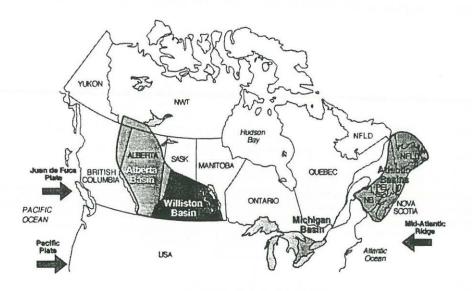


Figure 3: Major Sedimentary Basins in Canada suitable for carbon dioxide storage.

Geological storage requires access to large volume subsurface reservoirs which can act as sealed pressurized containers. The pressure is created by the column of rock overlying the reservoir. For CO₂ storage, the pressure to keep it at liquid-similar densities is found at depths below 800 meters which is also below our precious drinking water sources. The storage volume of the reservoir exists in the pore space between the mineral framework of the rock, and is initially occupied by geofluids such as brines, hydrocarbon and other gases (e.g. H_2S and CO_2). The greatest pore-based void volumes are found in the sedimentary basins of the world. These structures are formed by thick accumulation of sediments (Figure 4, up to 10 kilometers thick) in subsiding basins which are active for many millions of years before they stabilize. Depending on the sediment source and type, the raw materials deposited are detrital sand, silt and mud, chemical precipitated carbonate minerals and organic matter. As the sediments are buried, they start to compact and dewater in the earlier stages of the lithification process. Eventually the grains become cemented together and become rock (e.g. sandstone, siltstone, shale, limestone). The coarser grained sedimentary rocks, such as sandstone, have the highest interconnected porespace (i.e. highest permeability) and therefore the ability for fluid

flow (and are called aquifers). The finer grained sedimentary rocks, such as shales, have poor interconnected porespace and retard flow (and are called aquitards). Sedimentary basins are dynamic, in the sense that they have an intricate plumbing system defined by the superposition of these high and low permeability strata that control the flow of fluids throughout the basins (Figure 4). Ninety-nine percent of the flow takes place in the aquifers. The only flow of importance in the aquitards would be across the aquitards between two aquifers (referred to as cross formational flow) and would be thousands of times slower than flow in aquifers.

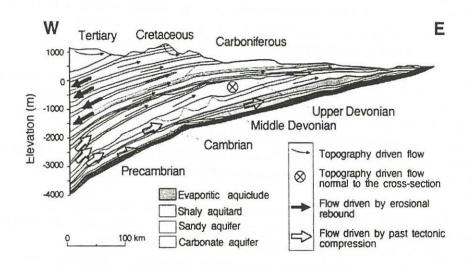


Figure 4: Present flow patterns of aquifers in the south-central part of the Alberta Sedimentary Basin (after Bachu, 1995).

It can be seen by the above description, that sedimentary basins are "leaky" systems (Bachu, 2001a). Where aquifers come to the surface, springs are formed. Only sediments with zero pore space are truly impermeable (e.g. some chemical sediments such as salt, or sediments where the porosity has been filled by cements during diagenesis). Not all sedimentary basins are equal. Younger basins and basins in orogenic regions are deforming throughout geologic time and are seismically active as contrasted to those sedimentary basins on the stable craton. Rates of leakage would be expected to be higher in the former due to higher permeabilities, cross formational flow and faulting associated with earthquakes (Hitchon et.al, 1999). Leakage from the cratonic sedimentary basins are mainly where the aquifer meets the surface (Figure 4).

Even in the case of younger basins, the rate of leakage is insignificant on a human time scale. However, short circuiting the conventional flow paths can lead to rapid release of CO₂. This could occur in orogenic regions by faulting or in non-orogenic regions through improperly abandoned wellbores. Obviously active orogenic regions and parts of sedimentary basins containing transmissive faults should not be targeted for geological storage. Rapid leakage of gas can be detected using monitoring programs which are mature technology in the oil and gas industry (Gunter et al., 1999) Regional flow rates in the deeper part of the sedimentary basins are extremely slow. For example, in the Alberta Basin, flow rate is measured in centimetres per year. In contrast, the distance along the aquifer from the deep basin to where it flows onto the surface is measured in 100's of kilometres (Figure 4). Therefore, a volume of water flowing from the deep basin to the surface would take over 1 million years. The point to be made is that all sedimentary basins are leaky over geologic but not the human time scale. During such a million year time period, other processes such as hydrogeological trapping and geochemical trapping can take place (Bachu et al., 1994).

Hydrogeological Trapping

Sedimentary basins have many physically-bound traps called reservoirs in which the fluid is static; some of which are occupied by oil and gas and the remainder by aqueous fluids. These oil and gas occurrences were formerly also filled by aqueous fluids. Moreover, such closed traps are very small compared to aquifers which are unconfined and eventually release their waters to the surface. But these closed traps have held fluids securely over geological time and obviously would be the first targets for geological storage. In addition, the production of oil and natural gas from sedimentary basins create void or low pressure storage space which can be refilled/repressurized with CO₂. Thus, these depleted reservoirs would be even more attractive for storage. The pore space of sedimentary basins can both be the source and the solution to the problem of GHG emissions.

Hence, the most secure type of hydrogeological trapping is found in oil and gas reservoirs in the form of "structural" or "stratigraphic" traps, termed "closed" traps which have held oil and gas for millions of years. Examples are traps bounded by unconformities, facies change, anticlines and non-transmissive faults. Obviously, these would be very attractive for CO_2 storage due to their long history of containment. A second type of hydrogeological trapping is provided in deep aquifers of sedimentary basins that characteristically have slow flow rates (Figure 4). The pore space in such "open" or "hydrodynamic" traps is usually filled with saline formation water. Carbon dioxide can be injected into these deep aquifers by displacing the saline formation water (Hitchon, 1996). As previously discussed, a volume of CO_2 injected into a deep open hydrogeological trap can take over a million years to travel updip to reach the surface and be released to the atmosphere (Bachu et al, 1994). Over such a long time scale, the CO_2 could be rendered benign by other naturally-acting processess. Consequently, a thorough understanding of the hydrology of the sedimentary basin must be known in order to safely store CO_2 in these hydrodynamic traps.

Geochemical Trapping.

The lifetime and security of hydrogeological trapping can be extended by "geochemical" trapping. There are three types of geochemical trapping: "solubility" trapping, "ionic" trapping, and "mineral" trapping (Gunter and Benson, 2000). Solubility trapping is the dissolution of the CO_2 in the formation water by the chemical reaction:

 $CO_2(gaseous) + H_2O \rightarrow H_2CO_3(aqueous)$

Once dissolved, CO_2 will no longer exist as a free phase that can migrate independently of the surrounding groundwater. Once the CO_2 is dissolved in water, the second type of geochemical trapping can occur, ionic trapping. Dissolved CO_2 acts as a weak acid and will react with the sodium and potassium basic silicate or calcium, magnesium and iron carbonate and silicate minerals in the reservoir as it flows through the pores and becomes neutralized by forming bicarbonate ions or carbonate ions (Gunter et al., 1993) by chemical reactions of the form:

 H_2CO_3 (aqueous) + OH (aqueous) \rightarrow HCO₃ (aqueous)

 $HCO_3^{-}(aqueous) + OH^{-}(aqueous) \rightarrow CO_3^{-}(aqueous)$

Whereas, in solubility trapping the reaction reverses and CO_2 is released when the pressure drops, in ionic trapping when the bicarbonate ion is formed, a drop in pressure does not release CO_2 .

Reaction times vary. Solubility trapping is rapid near the CO_2 -water interface and is limited by the diffusion of the dissolved CO_2 away from the interface. Reaction of the dissolved CO_2 with minerals is rapid (days) in the case of carbonate minerals but slow in the case of silicate minerals (hundreds to thousands of years). Carbonate minerals dissolve in the presence of high levels of dissolved CO_2 and would create additional porosity. However, extent of reaction of dissolved CO_2 with carbonate minerals before reaching equilibrium is small so that the strength of the rock layer is not threatened.

The most secure form of geochemical trapping is by formation of carbonate minerals from continued reaction of the bicarbonate ions with the calcium, magnesium and iron basic silicate minerals such as clays, micas, chlorites and feldspars (Gunter et al, 1993; 1997b; 2000) by reactions of the form:

 $\text{CO}_3^{=}(\text{aqueous}) + \text{Ca}^{++} \rightarrow \text{CaCO}_3 \text{ (solid)}$

Creation of these carbonate minerals, reduces pore space and may lead to plugging of the flow paths in the case of sedimentary rocks containing large amounts of basic silicate mineral assemblages. However, generally the basic silicate minerals represent less than 5% of the total mineral assemblage in an aquifer. The volume increase would be less than 1% if they completely reacted to form carbonate minerals which would reduce the porosity by less than 10%. As the flow rate is much faster than the reaction rate for silicate minerals, injectivity would not be impeded by silicate reactions.

Trapping of CO₂ by carbonate minerals is also possible at the surface as part of an industrial process (Goff et al., 1997). Here, there is a time constraint as long reaction times are not economically feasible. The rates of the carbonate mineral-forming reactions have been improved by artificially increasing the temperature above that commonly found in sedimentary basins. Thus, there are two geochemical pathways (Figure 5) for trapping CO₂ inertly in the form of carbonate minerals. When sedimentary basins are near the source of emissions, geological storage will be favored but as distances increase between the source and the sedimentary basin, inorganic surface storage may be preferred.

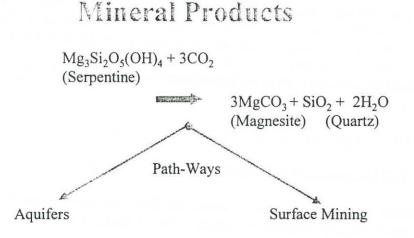


Figure 5: Examples of carbon dioxide trapping reactions which may occur naturally by injection of carbon dioxide in deep aquifers in Sedimentary Basins or may be utilized at high temperatures in an industrial process on the earth's surface.

Discussion

Storage technologies, developed by the oil and gas industry, are fairly mature. The volume of storage depends on the current pressure of the reservoir and the ultimate pressure rating of the reservoir. There are both natural CO_2 reservoirs, and industry experience gained from natural gas storage reservoirs and waste disposal; and injection into these reservoirs is based on being safely below fracture pressure so that the integrity of the seal is maintained. Experience in injection of CO_2 has been gained from repressurizing oil reservoirs using CO_2 in enhanced oil recovery, and similar technology is being developed for production of methane from coal beds (i.e. coalbed methane or CBM).

The global energy mix is becoming increasingly environmentally constrained. It is our opinion, that conversion of fossil fuels to other forms of energy will evolve in a way to minimize release of pollutants. End of the pipe solutions for fossil fuel conversion will gradually be replaced by cleaner conversion technologies. These, in turn, will be replaced by renewable forms of energy. Geological storage of carbon dioxide will play an important role in the transition. In preparation for this, geological mapping of sedimentary basins has to be revisited with more focus on the plumbing system and the safe storage of CO_2 than has been the traditional approach of the petroleum (Bachu, 2000; 2001b). We think that integration of oil and gas recovery operations with energy conversion in scenarios similar to that depicted in Figure 6 (Gunter et al., 1997a, Wong et al., 2000; 2001) will be an early part of the future highway leading to a world energy supply based largely on solar energy.

Green Power Plants

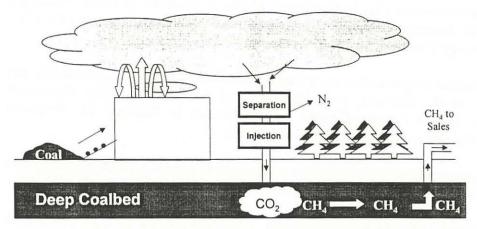


Figure 6: Example of integrated power plants which are fuelled by surface mined coal. The waste flue gas is injected into deep coal seams producing methane which can be used for other purposes. In the storage process, the carbon dioxide sorbs to the coal displacing the sorbed methane in a ratio of two or more molecules of carbon dioxide stored for every molecule of methane released. The benefits are increased gas reserves while decreasing the potential for global warming while developing of technology leading towards long term climate stability.

The ultimate capacity of geological storage of carbon dioxide will be huge (e.g. initial estimates found in Gunter et al.,1998 suggest an Alberta Sedimentary Basin capacity of 20 Gt of CO₂, enough to store all of Canada's CO₂ emissions for half a century), contingent upon identifying secure traps in sedimentary basins. In summary, the most secure hydrogeological traps are closed stratigraphic or structural traps which have been well characterized during their exploitation for oil and gas. Although the capacity of these traps for CO₂ storage is small relative to open hydrogeological traps in deep sedimentary basins, they are likely to be used first as they are known to be secure, having held oil and gas for geological time. Storage of CO₂ as carbonate minerals is the most secure form of storage, but the reactions which trap the CO₂ in carbonate minerals are slow on the human time scale but relatively fast on a geological traps will be utilized where security of the geological storage of CO₂ can be enhanced by geochemical reactions of the CO₂ with basic silicate minerals.

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