

Separation and Capture of CO₂ from Large Stationary Sources and Sequestration in Geological Formations

A Summary of the 2003 Critical Review

by Curt M. White, Brian R. Strazisar, Evan J. Granite, James S. Hoffman, and Henry W. Pennline

Increasing amounts of carbon dioxide (CO₂) in the atmosphere, and the resulting global warming effect, is a major air quality concern. CO₂ is the most abundant greenhouse gas emitted by fossil-fuel combustion for power generation, transportation, and heating. Reducing worldwide emissions of CO₂ will require many mitigation measures, including reductions in energy consumption, more efficient use of available energy, renewable energy sources, and carbon sequestration. The feasibility of capturing CO₂ from large point sources and subsequent geological sequestration is the subject of this year's Critical Review.

INTRODUCTION

Carbon dioxide (CO₂) sequestration in geological formations is necessary, along with other measures, to meet the Bush administration's Global Climate Change Initiative target of an 18% reduction in greenhouse gas (GHG) intensity by 2012. GHG intensity is the quantity of GHGs emitted per unit of gross domestic product (GDP). This review concludes that it is practical and feasible to 1) separate CO₂ from other exhaust gases released by fossil-fuel combustion and gasification in stationary sources; 2) capture the CO₂; 3) inject it into the formations; 4) safely maintain it in formations for hundreds of years with negligible leakage back to the atmosphere; and 5) monitor leakage for public safety. This review

describes different technologies for each of these topics and evaluates their applicability to specific situations.

CO₂ CAPTURE AND SEPARATION

Large stationary sources of CO₂ include fossil-fuel-based power generation facilities, natural gas production and upgrading facilities, hydrogen production plants, oil refineries (especially those that use the heavier crude oils), iron and steel plants, and cement and lime production facilities.^{1,2} Other industrial processes, primarily the production of ammonia and ethylene, generate nearly pure CO₂ streams and, therefore, allow for the use of relatively inexpensive methods of CO₂ recovery. Fossil-fuel-based power generation plants contribute approximately one third of the world's CO₂ emissions, but the CO₂ is mixed with other gases from which it must be separated.

CO₂ can be captured during the pre- or postcombustion phase. Precombustion capture occurs in gasification systems, where carbon in the coal is transformed into CO₂ or carbon monoxide (CO) at high pressures. CO₂ concentrations in flue gas from pulverized coal-fired systems, coal-fired integrated gasification combined cycle (IGCC) turbines, and natural gas-fired turbines are 15 vol%, ~9 vol%, and ~4 vol%, respectively, and pressures are close to ambient.³ The low CO₂ fraction requires large volumes of gas to be treated. CO₂ capture in a precombustion setting, as in an IGCC plant, is more energy-efficient and cost-effective than postcombustion capture from a traditional pulverized fossil-fuel-fired power plant.⁴

The location of the capture technology along the gas path can significantly affect CO₂ separation and capture.

The presence of pollutants in precombustion gases (e.g., hydrogen sulfide [H₂S], ammonia [NH₃], and mercury [Hg]) and postcombustion gases (e.g., sulfur dioxide [SO₂], nitrogen oxides [NO_x], and Hg) may influence which method of CO₂ capture is selected.

Currently used CO₂ capture technologies include 1) solvent wet scrubbing with chemical or physical absorbents; 2) solid dry scrubbing with physical adsorbents or chemical absorbents; 3) cryogenic methods; 4) gas membrane separation; and 5) advanced concepts. Factors influencing the practicality of these technologies include 1) partial pressure of the CO₂ in the gas stream; 2) extent of CO₂ recovery required; 3) sensitivity of the technology to impurities, such as acid gases and particles; 4) purity of the desired CO₂ product; 5) capital and operating costs of the process, including cost of additives to overcome fouling and corrosion; and 6) environmental impacts of the process.⁵

Wet scrubbing involves a chemical reaction between the solvent and CO₂ or the physical dissolution of CO₂ into the solvent. Chemical solvents are preferred for low concentrations at low pressures, while physical solvents are favored for high pressures and low concentrations of inert gases, such as nitrogen (N₂). Chemical absorption involves one or more reversible chemical reactions between CO₂ and an aqueous solution of an absorbent, such as an alkanolamine or potassium carbonate. For chemical absorption, significant quantities of heat are needed to dissociate the chemical complex formed by the reaction with CO₂.⁶ Upon heating, the bond between the absorbent and CO₂ is broken, and a CO₂-enriched stream is formed.

Physical absorption uses inorganic or organic liquids to preferentially absorb CO₂ from the gas mixture. The absorption liquid is then regenerated by increasing its temperature and/or reducing its pressure. High boiling-point solvents are preferred to minimize solvent losses and to prevent contamination of the released gas with solvent vapors. Monoethanolamine (MEA) is a widely used wet scrubbing reagent, although significant amounts of energy are required during the regeneration step. To date, all U.S. commercial plants that capture CO₂ from power plant flue gas use processes based on chemical absorption with MEA.⁷⁻⁹ Several other wet chemical absorption processes are currently under development, such as aqueous ammonia solvent for the removal of CO₂ from flue gas.^{10,11}

Because CO₂ is an acid gas, reaction often involves neutralization of the CO₂ with a base compound on or within a solid. Carbonates of lithium, sodium, potassium, and other earth metals are solids that react with CO₂. Another class of reactions involves alkaline metal oxides reacting with CO₂ to form an alkaline earth metal carbonate. A third type of reaction involves the substitution of metals to form a metal carbonate (e.g., lithium and zirconium).

Solid adsorption methods employ a physical attraction between the gas and "active sites" on the solid, while solid

absorption methods employ a chemical reaction to capture the target gas. CO₂ can be physically adsorbed onto high-surface-area solids without undergoing a chemical reaction. Pressure and/or temperature are adjusted during regeneration to repeat the adsorption cycle. Pressure swing adsorption (PSA) entails adsorbing the gas at high pressure, isolating the solid, and then desorbing the sorbed gas by lowering the pressure. Vacuum PSA uses a vacuum during the regeneration step. In temperature swing adsorption (TSA), gases are adsorbed at low temperatures, the solid is isolated, and then the temperature is raised. Cycle time for regeneration is typically much shorter for PSA (order of seconds) compared to TSA (hours).⁶ PSA and TSA are energy-intensive, expensive, and require further research and development to improve their energy efficiency and cost-effectiveness for widespread use.

The capacity of organic and inorganic solvents for CO₂ increases with higher pressures and lower temperatures. Because the partial pressure of CO₂ in flue gas is low, and the flue gas temperature is relatively high, physical absorption is less practical and effective than chemical absorption for these sources. The absorption method is suitable for advanced power generation, such as pressurized precombustion (e.g., IGCC).^{5,12} Physical absorption capacities and kinetics are affected by pore size of the absorbing material, pore volume, surface area, and the affinity of the sorbent for weakly bonding CO₂.

GEOLOGICAL SEQUESTRATION OF CO₂

Bachu¹³ defines geological sequestration as "the capture of CO₂ directly from anthropogenic sources and disposing of it deep into the ground for geologically significant periods of time." Proven geological formations for long-term CO₂ storage include depleted petroleum reservoirs, deep unmineable coal seams, and deep saline aquifers. Other potential storage areas include depleted gas reservoirs, salt domes, salt formations, depleted CO₂ domes, and carbonaceous shales.

There is an incomplete understanding of what happens when CO₂ is injected into a coal seam. This review proposes the following processes that can be inferred from available information:¹⁴

1. The glass-to-rubber transition temperature (T_g) of coal is dramatically reduced by the imbibition of CO₂, and the coal becomes plasticized.^{15,16}
2. The cleat system (open spaces) within the coalbed closes due to swelling, slowing gas flow in that portion of the coal seam.
3. The CO₂ self-diffusivity increases after it plasticizes at temperatures higher than T_g .^{15,16}
4. The diffusivity of CO₂ in coal swollen by high-pressure CO₂ can be described by free volume theory.
5. Plasticization, swelling, increased diffusivity, lowering of T_g , relaxation of the macromolecular network, and depression of the softening temperature are limited by the degree to which the coal is free to swell.

6. CO₂ moving through the coalbed extracts small molecules trapped within the macromolecular network. As the network relaxes, these molecules move with the flowing CO₂, as long as the pressure is above their threshold pressure.
7. Minerals in the coal dissolve in a carbonic acid solution when water and high-pressure CO₂ are present together in the seam.
8. Calcium and magnesium in low-rank coals dissolve in the carbonic acid.
9. Dry CO₂ dries the coal, particularly in those areas where the flow rate of CO₂ is highest.
10. CO₂ pressure, temperature, and pH gradients form between the injection well and the recovery well. When dissolved minerals and organics reach areas of the seam with lower pressure, they precipitate, clogging the coal's pores.

CO₂-enhanced coalbed methane (ECBM) recovery obtains methane gas (CH₄) from unmineable coalbeds by injecting CO₂ to displace the CH₄. This sequesters the CO₂ while producing a useful natural gas product that is a high-efficiency energy source.¹⁷ A large-scale CO₂-ECBM/sequestration project has been performed at Burlington Resources' Allison Unit in New Mexico.¹⁸⁻²⁰ Other examples include British Petroleum's Tiffany Unit in Colorado,^{21,22} the Alberta Research Council's Fenn Big Valley project in Canada,^{23,24} the RECOPOL project in Poland,²⁵ and planned projects in Australia,²⁶ China,²⁷ and Japan.^{28,29} Measurements from these facilities may provide a better understanding of the mechanisms involved.

Deep saline aquifers³⁰⁻³⁴ have large storage capacities and they are widespread across a large portion of the United States and the world. A commercial-scale CO₂ injection project associated with the recovery and purification of natural gas in the North Sea at Sleipner off the coast of Norway has been in operation since September 1996.³⁵⁻³⁷ The natural gas harvested from this field contains up to 9% CO₂. The CO₂ is captured using an amine system to meet a < 2.5% CO₂ product specification. To avoid a steep CO₂ emissions tax imposed by the Norwegian government, one million tons of CO₂ per year is injected into the Utsira sandstone saline aquifer ~ 1000 m below the seabed. This is first time that CO₂ has been stored underground for GHG mitigation. Other brine sequestration projects include the Frio project^{34,38,39} and American Electric Power's project at the Mountaineer Plant.^{40,41}

CO₂ is retained in saline aquifers by 1) hydrodynamic trapping, in which CO₂ remains as an undissolved gas contained by an overlying low-permeability cap rock; 2) solubility trapping, where CO₂ is dissolved in the water; and 3) mineral trapping, which occurs when dissolved CO₂ reacts with either other aqueous species or minerals to precipitate a solid carbonate, most likely a calcium, iron, or magnesium carbonate. Mineral trapping is the least susceptible to leakage.^{30,42}

Simulations of CO₂ injection and storage capacity⁴³⁻⁴⁵ focus on chemical reactions induced by injection of CO₂ and the use of thermodynamic and kinetic information to determine how the chemical makeup of the brine and mineral matrix affect the relative importance of mineral, solubility, and hydrodynamic trapping. The majority of the modeling and simulation work completed so far has been specifically applied to the Alberta sedimentary basin.⁴⁶ Although there is disagreement with respect to the extent of mineral trapping that will occur, the presence of basic minerals and an abundance of calcium, iron, and magnesium favor mineral and solubility trapping.

SEISMIC ACTIVITY CAUSED BY INJECTION OF FLUIDS UNDERGROUND

CO₂ injection into geological formations may cause earthquakes. Induced seismic activity occurs when an external factor is introduced to a local tectonic system that is sufficient to cause a mechanical failure of the rock. Denver, CO, has experienced more than 700 earthquakes, some of which have been attributed to injections at the nearby Rocky Mountain Arsenal.⁴⁷⁻⁴⁹ Small earthquakes (< 5 on Richter scale) occurred one month after injections began into a 12,045-ft wastewater well in an area where there was no previous record of seismic activity. The probability that these were unrelated to the injection well was estimated as 1 in 2.5 million.⁴⁸ An Underground Injection Control (UIC) program was subsequently established by the U.S. Environmental Protection Agency (EPA) to regulate underground fluid disposal. The potential for earthquakes must be examined as part of the planning for any geological sequestration project.

MONITORING OF GEOLOGICALLY SEQUESTERED CO₂

CO₂ is an asphyxiant and is toxic in high concentrations; however, it is nonflammable, nonexplosive, noncarcinogenic, and relatively nontoxic in low concentrations. The current Occupational Safety and Health Administration (OSHA) standard for maximum exposure to CO₂ is 5000 ppmv as an 8-hour time-weighted average concentration. As a result of geologically sequestered CO₂, brine might be displaced into overlying strata, with subsequent contamination of fresh water. Stringent environmental assessments and pre- and postproject monitoring are necessary for sequestration projects.

A monitoring program includes geochemical methods such as tracers, measurement of the CO₂ flux in soil gas, geophysical methods such as four-dimensional (4-D) seismic, and electrical sensing. These measurements can be interpolated and extrapolated with advanced geologic models, flow simulators, and forward seismic models. Each technique yields unique but complementary information that, when combined, provide a more complete picture than is possible from any single technique. Monitoring networks must provide indications of CO₂

leakage into homes, schools, or valleys, where CO₂ may collect at ground level, giving rise to O₂-depleted breathing zones. If leakage is detected at a site, depending on the extent of the leak and its geographic location, sequestration may need to be abandoned and already sequestered CO₂ vented.

THE PATHWAY TO STABILIZATION

Figure 1 depicts GHG emissions scenarios for the United States as a function of time. By 2012, the gap between the two lines shown in Figure 1 is 107 million metric tons of carbon equivalent (MMTCE) and increases to 1100 MMTCE in 2040. For comparison, a new 400-MW pulverized-coal electric power generating station emits approximately 0.6 MMTCE per year. To close this gap, Klara et al.⁵⁰ propose a portfolio of techniques, including increased efficiency and renewable energy, non-CO₂ GHG mitigation, forestry and agriculture, early value-added geologic sequestration, and advanced geologic sequestration technologies. The contribution that each technique will make to close the gap is shown in Figure 2. Figure 2 shows a possible scenario to meet the goal of an 18% reduction in GHG intensity by 2012 that includes a portfolio of technologies, such as

sequestration of CO₂ in geological formations.

Early geologic sequestration includes enhanced oil recovery (EOR) and CO₂-ECBM, which produce value-added oil or gas to offset the overall costs of CO₂ capture and sequestration. Under the scenario shown in Figure 2, EOR will sequester 10 MMTCE/yr and CO₂-ECBM will sequester 2 MMTCE/yr by 2012.⁵⁰ This is achievable from high-purity CO₂ exhaust streams that are currently vented to the atmosphere. In the United States, ~ 44 MMTCE/yr of CO₂ could be easily captured. Figure 2 shows that CO₂ sequestration in geologic media is essential to stabilize the atmospheric concentration of CO₂ and to meet the goals set forth in the Bush administration's Global Climate Change Initiative. According to the modeling platform used by Klara et al.,⁵⁰ an 18% reduction in GHG intensity cannot be plausibly met without including contributions from geological sequestration of CO₂. This review shows how sequestering CO₂ is possible using off-the-shelf technologies.

CONCLUSION

Although CO₂ can be practically separated from effluents, captured, transported, and stored, the overall cost using current technologies must be substantially reduced. Chemical absorbent systems appear to be the most viable option for capturing and separating CO₂ from power station effluent where partial pressures are low and other gases may interfere. Deep saline aquifers provide the most practical method for long-term storage as they underlie much of the continental United States and would minimize transport costs.

Environmental concerns must be addressed during project planning, construction, and execution. Long-term monitoring above and below ground is needed to detect leakage to air and water. The practical applications to date demonstrate that underground CO₂ sequestration provides a safe, verifiable, technologically feasible, and ultimately affordable option to the stabilization of atmospheric CO₂ concentrations.⁵¹

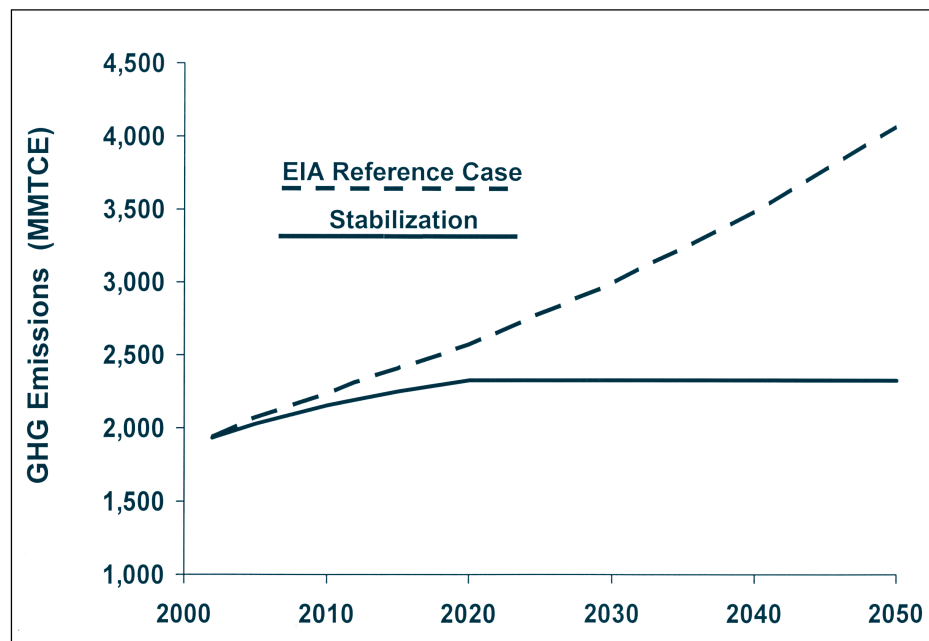


Figure 1. A comparison of two GHG emissions reductions scenarios.

Note: Emissions are in millions of metric tons of carbon equivalent (MMTCE). The top line represents the Energy Information Agency's Annual Energy Outlook 2002 Reference Case (i.e., the projected emissions if the United States conducts "business as anticipated"). This scenario includes reductions accrued by using natural gas instead of coal and assumes that advanced electric power generating technologies begin to be used that include significant advances in energy efficiency, as well as contributions from renewable energy sources and non-CO₂ GHG mitigation. The bottom line represents emissions associated with a "reduced emissions scenario," where the rate of growth in U.S. GHG emissions is slowed and then stopped according to the following schedule:

- 2002–2012: GHG intensity is reduced to 152 MMTCE/\$GDP;
- 2013–2020: emissions growth is reduced to 50% below the Energy Information Agency's 2002 Reference Case; and
- 2021–2040: GHG emissions are stabilized at the 2020 emissions level.⁵⁰

Environmental concerns must be addressed during project planning, construction, and execution. Long-term monitoring above and below ground is needed to detect leakage to air and water. The practical applications to date demonstrate that underground CO₂ sequestration provides a safe, verifiable, technologically feasible, and ultimately affordable option to the stabilization of atmospheric CO₂ concentrations.⁵¹

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DISCLAIMER

Reference to any specific brand name or product is to facilitate understanding and does not imply endorsement by the U.S. Department of Energy.

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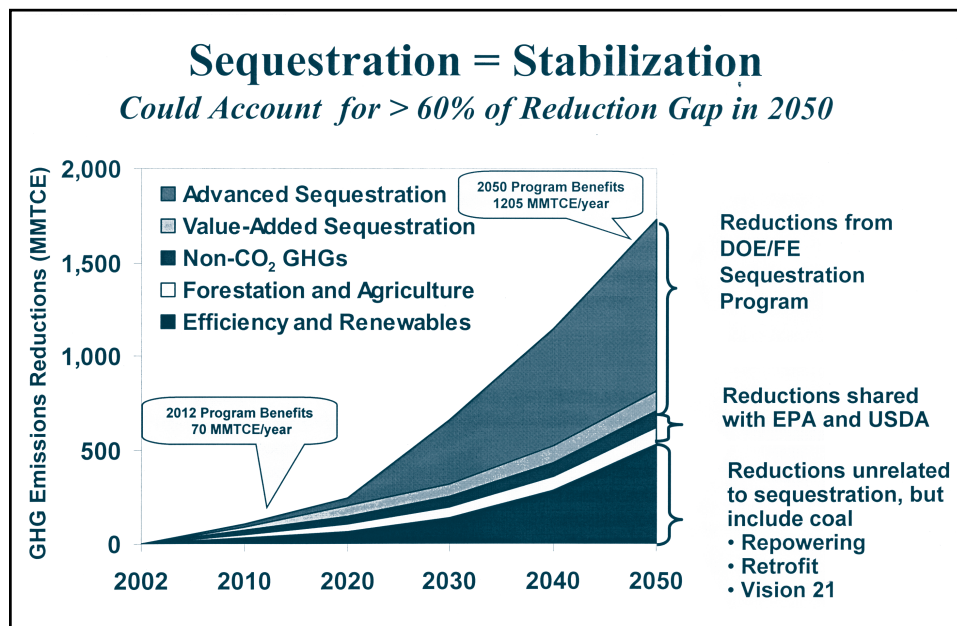


Figure 2. Contribution of technologies to the GHG emissions reductions needed to reach 1110 MMTCE/yr in 2040.

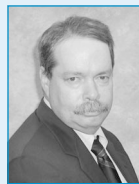
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ABOUT THE CRITICAL REVIEW

The Critical Review program was established by A&WMA (then the Air Pollution Control Association) in 1973 to stimulate discussion of major issues of concern in air pollution control. As the Association grew, the scope of the Critical Review expanded to include waste management and other environmental media. Dr. Curt M. White, Carbon Sequestration Science Focus Area Leader at the U.S. Department of Energy's National Energy Technology Laboratory, will present this year's Critical Review in its entirety at the 96th Annual Conference & Exhibition in San Diego, CA. The complete 2003 Critical Review will also be published in the June issue of the *Journal of the Air & Waste Management Association*. ☺

About the Authors



Dr. Curt M. White is the Carbon Sequestration Science Focus Area Leader at the U.S. Department of Energy's National Energy Technology Laboratory (NETL) in Pittsburgh, PA. The focus area studies the capture and separation of CO₂ from large point sources, such as fossil-fuel-fired electric power generating stations and gasification facilities; oceanic sequestration of CO₂; geological sequestration of CO₂ and geological sequestration modeling; and the development of tools to monitor and verify the integrity of geologically sequestered CO₂. He can be reached via e-mail at curt.white@netl.doe.gov. Dr. White prepared this review in collaboration with the NETL focus area group. **Dr. James S. Hoffman** develops dry, regenerable sorbent processes for CO₂ capture from gaseous streams. **Henry W. Pennline** develops and tests economically and environmentally acceptable flue gas control systems that remove pollutants such as SO₂, NO, PM_{2.5}, Hg, and CO₂ on pilot-scale combustion devices. **Dr. Evan J. Granite** developed a pilot plant design for an integrated gasification process. The design included CO₂ recovery by calcium sorbents, as well as by hot carbonate and MEA scrubber systems. **Dr. Brian R. Strazisar** investigates solvent degradation at CO₂ capture facilities, models geochemical processes resulting from CO₂ sequestration in deep saline aquifers, determines the effects of contaminants on oceanic sequestration, and develops methods to monitor leakage from underground CO₂ sequestration.

Attend the Critical Review presentation

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