

DOE'S CARBON CAPTURE AND SEQUESTRATION R&D PROGRAM

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ABSTRACT

Since its inception 10 years ago, the Department of Energy's Carbon Sequestration Program – managed within the Office of Fossil Energy and implemented by the National Energy Technology Laboratory – has been developing both core and supporting technologies through which carbon capture and storage (CCS) will become an effective and economically viable option for reducing carbon dioxide (CO₂) emissions from coal-based power plants. Successful research and development will enable CCS technologies to overcome the various technical, economic, and social challenges, such as cost-effective CO₂ separation and transport, long-term stability of CO₂ sequestration in underground formations, monitoring and verification, integration with power generation systems, and public acceptance. The programmatic timeline is to demonstrate a portfolio of safe, cost-effective greenhouse gas (GHG) capture, storage, and mitigation technologies at the commercial scale by 2012, leading to substantial deployment and market penetration beyond 2012. These GHG mitigation technologies will help slow GHG emissions in the near-term. They also provide the potential to ultimately stabilize and reduce GHG emissions in the United States.

PROGRAM OVERVIEW

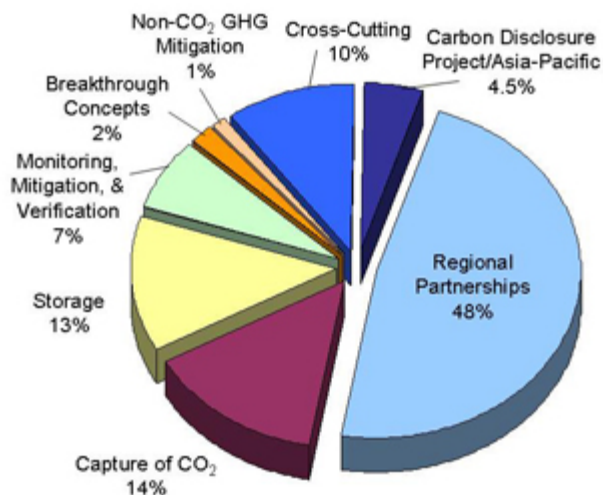
Many climate scientists believe that global climate change is occurring and that a major cause is the anthropogenic emission of GHG into the atmosphere. New analysis by the Intergovernmental Panel on Climate Change¹ indicates that emissions of CO₂ that result from combustion of fossil fuels such as coal or petroleum may alter the global climate. While it is not clear what rules, regulations, or targets may be put in place, it is highly probable that emissions of CO₂ from fossil-fuel-based sources, including coal-fired power plants, will be targeted for reduction. In order to retain fossil fuels as a viable world energy source, cost effective and efficient CCS technologies will need to be developed and tested. By cost-effectively capturing CO₂ before it is emitted to the atmosphere and then permanently storing or sequestering it, fossil fuels can continue to be used in a carbon constrained world without restricting economic growth.

The U.S. Department of Energy's National Energy Technology Laboratory (DOE/NETL) has responded to this challenge through the initiation of a carbon sequestration research and

development (R&D) program in 1997 to develop technologies and approaches to capture and permanently store CO₂ produced during fossil fuel combustion.^{2,3} Today, DOE/NETL's Carbon Sequestration Program plays a key role in several Presidential Initiatives and has widespread support as evidenced in the Energy Policy Act (EPACT) of 2005. The Carbon Sequestration Program also supports FutureGen, a key DOE initiative aimed at building a highly efficient and technologically sophisticated power plant that can produce both hydrogen and electricity while capturing and sequestering CO₂ emissions. FutureGen will serve as a full-scale field laboratory for CCS technologies.

The Carbon Sequestration Program encompasses two main elements: *Core R&D* and *Demonstration and Deployment*. The Core R&D element addresses technology needs in several focus areas and develops technology solutions that can be demonstrated and deployed in the field. Lessons learned from the field tests are fed back to the Core R&D element to guide future efforts. A detailed breakdown of the FY2007 Carbon Sequestration Program budget of \$100 million is presented in Figure 1.

Figure 1 – Breakdown of the FY2007 Carbon Sequestration Program Budget



Core R&D involves laboratory through bench- and pilot-scale research aimed at developing new technologies and new systems for GHG mitigation. The Core R&D portfolio includes cost-shared, industry-led technology development projects, research grants, and research conducted through NETL's Office of Research and Development (ORD). The Core R&D effort encompasses five focus areas: CO₂ capture; CO₂ storage; monitoring, mitigation, and verification; non-CO₂ GHG control; and breakthrough concepts.

The Demonstration and Deployment element of the Carbon Sequestration Program is designed to demonstrate the viability of CCS technologies at a scale large enough to overcome real and perceived infrastructure challenges. The largest component of the Demonstration and Deployment element is the Regional Carbon Sequestration Partnerships (RCSP) Program. Through the RCSP Program, CCS technologies will be tested in the field throughout the United States to identify and eliminate technical and economic barriers to commercialization. Such an effort is necessary to ensure that organizations are prepared to act if future global climate change

policies require large-scale deployment of sequestration technology.

As a technology and a research discipline, carbon sequestration is in its infancy. To guide the Carbon Sequestration Program through this early development period, DOE established the following initial technology goal: “To develop, by 2012, fossil fuel conversion systems that offer 90% CO₂ capture with 99% storage permanence at less than a 10% increase in the cost of energy services.” By simultaneously exploring a number of related R&D pathways, the many challenges confronting carbon capture and storage can be overcome, enabling the Program to achieve this ambitious goal.

TECHNOLOGY DEVELOPMENT EFFORTS

The first three Core R&D focus areas track the life cycle of a CCS system: CO₂ is first captured, then it is stored (sequestered) or converted to a benign or useful carbon-based product, and finally it is monitored to ensure that it remains stored, with appropriate mitigation actions taken as needed. The fourth focus area, non-CO₂ GHG control, primarily involves the capture and reuse of methane emissions from energy production and conversion systems such as coal mine ventilation air. The fifth area, breakthrough concepts, targets novel CO₂ capture, storage, and conversion concepts with the potential to provide step-change improvements in process efficiency, energy use and cost, and those with the potential to expand the applicability of CCS beyond conventional stationary source emissions.

Carbon Capture

The Carbon Capture R&D area accounts for 14% of the FY2007 Carbon Sequestration Program budget of \$100 million. Carbon Capture projects have primarily focused on laboratory through pilot-scale research to develop innovative technologies to capture CO₂. Several projects involve the evaluation of technologies that are currently used in other smaller industrial applications. Innovative concepts are also being developed to increase the performance and lower the overall cost to capture CO₂ from large stationary fossil energy point sources. Carbon capture is an important target area for R&D because current technologies are expensive and very energy intensive, which would seriously degrade the overall efficiency of power plants. Other challenges include: technology scale-up, market considerations/readiness, power plant diversity, reliance on other enabling technologies (e.g., oxygen production), process integration, and an uncertain regulatory framework.

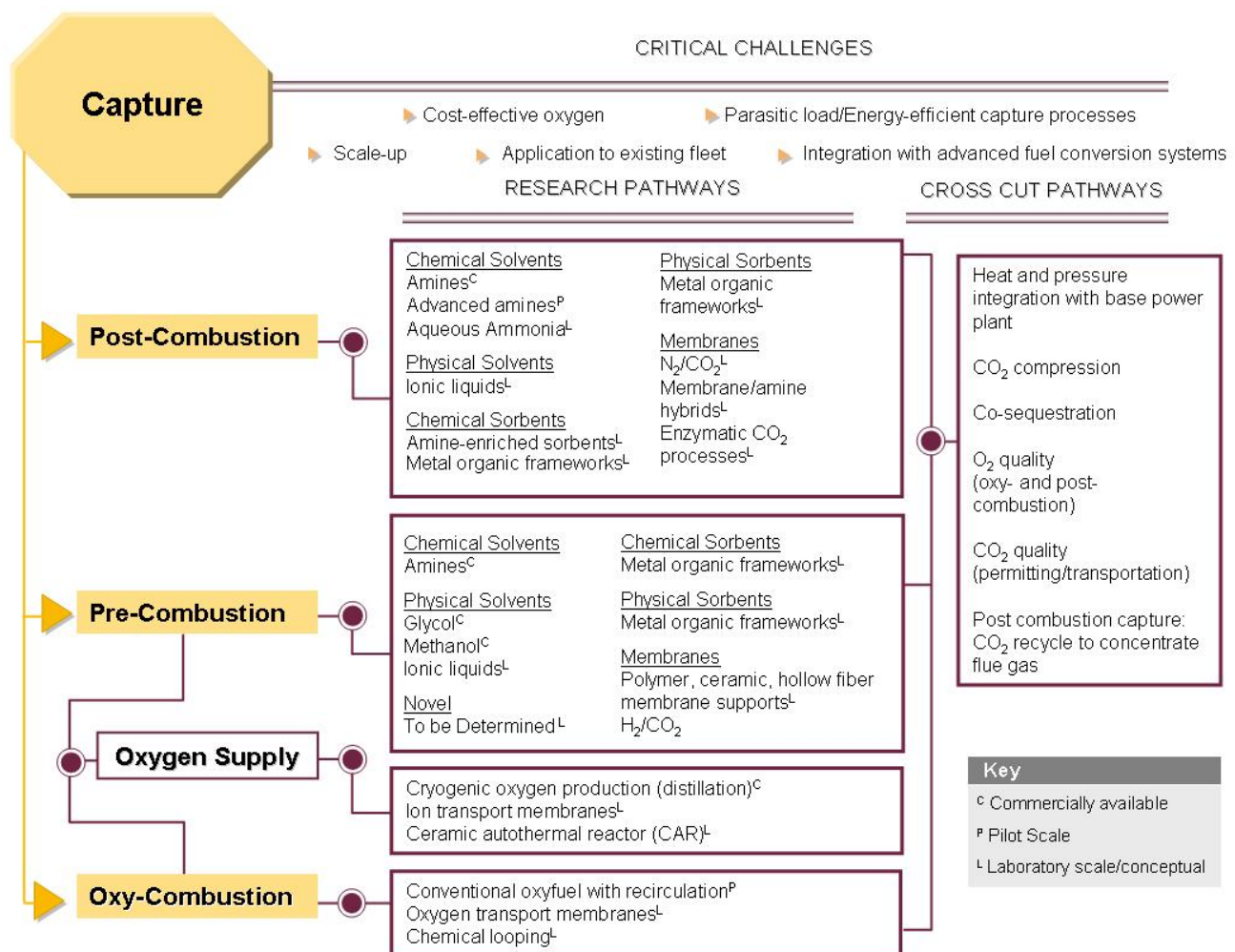
The goals of the Carbon Capture area are directly aligned with the goals of the higher level programs and offices within DOE, as well as key Presidential Initiatives. These goals and the technology research they support are aimed at newly constructed plants. However, funded projects also perform research to meet the needs of the existing power generation fleet as well as other CO₂-intensive industries within the United States.⁴

- By 2007: Identify capture technologies that increase cost of energy services by less than 20% for pre-combustion systems and less than 45% for post-combustion systems and oxy-combustion systems.

- By 2012: Identify capture technologies that increase cost of energy services by less than 10% for pre-combustion systems and less than 20% for post-combustion systems and oxy-combustion systems.
- By 2014: Initiate at least two slipstream tests of novel CO₂ capture technologies that offer significant cost reductions.
- By 2018: Initiate large-scale field testing of promising novel CO₂ capture technologies.

DOE/NETL has categorized carbon capture technologies into three approaches: post-combustion, pre-combustion, and oxy-combustion. Figure 2 shows the portfolio of research pathways that are being investigated in the three technology approaches of the Carbon Capture area. Key enabling technologies such as oxygen supply for oxy-combustion are also included, as are crosscut pathways which can influence technology development efforts. Additionally, the stage of development for the various technology options is also indicated. A key point from Figure 2 is that the majority of the technology options being considered are still in the laboratory stage of development.

Figure 2 - Carbon Capture Program R&D Portfolio



Post-combustion systems separate CO₂ from the flue gas produced by conventional coal combustion in air. In this case, CO₂ is exhausted in the flue gas at atmospheric pressure and a concentration of 10-15 volume percent. This is a challenging application for CO₂ capture because: (1) the low pressure and dilute concentration dictate a high actual volume of gas to be treated; (2) trace impurities in the flue gas tend to reduce the effectiveness of the CO₂ adsorbing processes; and (3) compressing captured CO₂ from atmospheric pressure to pipeline pressure (1,200 - 2,200 pounds per square inch) represents a large parasitic energy load.

Pre-combustion systems are designed to separate CO₂ and hydrogen in the high pressure syngas produced at integrated gasification combined cycle (IGCC) plants. An IGCC plant processes the primary fuel in a reactor with steam and air or oxygen to produce a syngas mixture consisting mainly of carbon monoxide (CO) and hydrogen. Additional hydrogen, together with CO₂, is produced by reacting CO with steam in a 'shift reactor'. Although the initial fuel conversion steps are more elaborate and costly than in post-combustion systems, the high concentrations of CO₂ produced by the shift reactor (typically 15 to 60% by volume on a dry basis) and the high pressures often encountered in these applications are more favorable for CO₂ separation. The advantage of this type of system is the higher CO₂ concentration (partial pressure) and the lower volume of syngas to be handled, resulting in smaller equipment sizes and lower capital costs.

Oxy-combustion involves the use of oxygen, with a purity of 95 to 99% oxygen assumed in most current designs, to combust coal and produce a highly concentrated CO₂ (greater than 60 percent by volume) stream. The CO₂ is separated from water vapor by condensing the water through cooling and compression. Further treatment of the flue gas may be needed to remove pollutants and non-condensed gases (such as nitrogen) from the flue gas before the CO₂ is sent to storage. Oxy-combustion cannot be simply substituted for air combustion in existing fossil-fueled power plants due to differences in combustion characteristics. In order for oxy-combustion to be utilized in existing combustors, a thermal diluent is required to replace the nitrogen in air.⁵ The oxygen produced from air separation would be mixed with recycled flue gas to approximate the combustion characteristics of air.

DOE/NETL is currently funding multiple carbon capture projects within each of these approaches. As shown in Table 1, these carbon capture R&D efforts are being performed both externally by research organizations and academic institutions and internally through NETL's Office of Research and Development, specifically the Separations and Fuels Processing Division and the Office of Computational Dynamics. The primary objective of these projects is to identify carbon capture technologies capable of achieving the goals of the Carbon Capture area.

Table 1 – Carbon Capture Projects Funded by DOE/NETL

Participant	“Project Title”	Research Pathway	Scale
<i>Post-Combustion</i>			
Research Triangle Institute	Dry Regenerable CO ₂ Sorbent	Chemical Sorbent	Bench
Carbozyme, Inc.	Biomimetic Membrane	Membrane	Laboratory
University of Notre Dame	Ionic Liquids (ILs)	Physical Solvent	Laboratory
UOP LLC	Metal Organic Frameworks (MOFs)	Chemical & Physical Sorbent	Laboratory
University of Akron	Amine-grafted Zeolites	Chemical Sorbent	Laboratory
Membrane Technology & Research, Inc.	Novel Polymer Membranes	Membrane	Laboratory
NETL	Solid CO ₂ Sorbents	Chemical Sorbent	Bench
NETL	Solid CO ₂ Sorbent Reactor Design	Chemical Sorbent	Laboratory
NETL	Ammonia-based Process	Chemical Solvent	Laboratory
NETL	Novel Amine Sorbents	Chemical Sorbent	Laboratory
NETL	MOF CO ₂ Membranes	Membrane	Laboratory
NETL	ILs and poly(ionic liquids) (PILs)	Physical Solvent	Laboratory
<i>Pre-Combustion</i>			
SRI International	PBI-based Membranes	Membrane	Laboratory
NETL	Novel Membranes	Membrane	Laboratory
NETL	CO ₂ Capture Solvents	Physical Solvent	Laboratory
<i>Oxy-Combustion</i>			
The BOC Group, Inc.	Pilot Test CAR Oxy-combustion Unit	Oxy-combustion	Pilot
B&W	PC Oxy-combustion Pilot Testing	Oxy-combustion	Pilot
SRI	Oxy-fired CO ₂ Recycle Retrofit	Oxy-combustion	Bench
Praxair, Inc.	Oxygen Enriched Combustion	Oxy-combustion	Laboratory
NETL/Jupiter Oxygen	PC Oxy-combustion with Integrated Pollutant Removal	Oxy-combustion	Bench
NETL	Oxy-combustion Modeling & Optimization	Oxy-combustion	Laboratory
NETL	Chemical Looping	Oxy-combustion	Laboratory

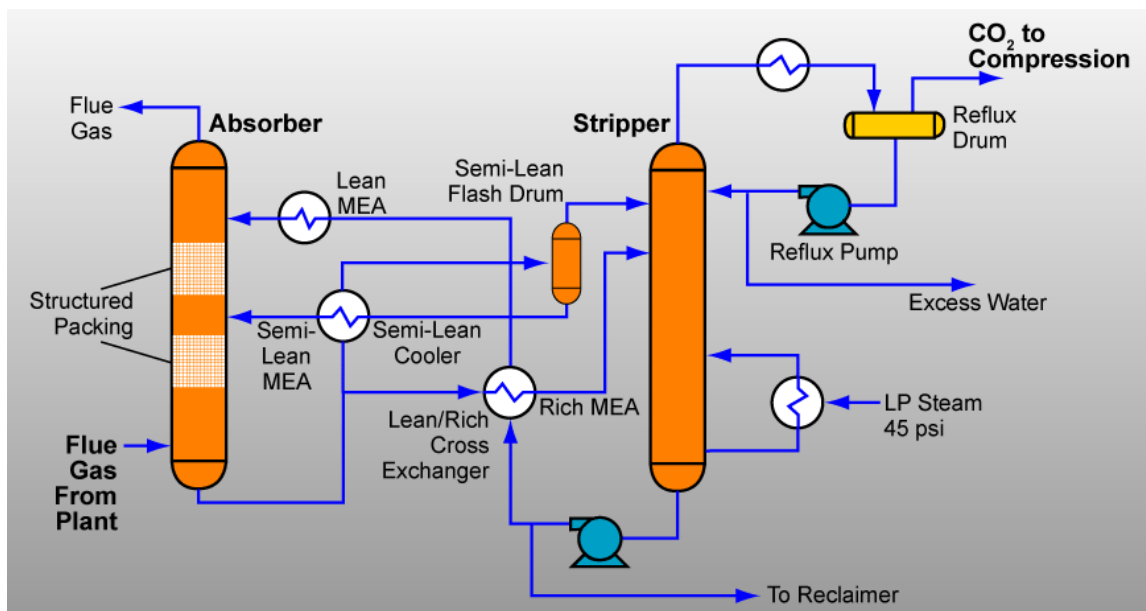
The selection of a capture technology for a given application will depend on many factors.⁵ These can include partial pressure of the CO₂ in the gas stream; extent of CO₂ recovery required; sensitivity of the technology to impurities, such as acid gases and particulates that may be present in the gas stream; purity of the desired CO₂ product; capital and operating costs of the process including cost of additives necessary to overcome fouling and corrosion when applicable; and environmental impacts of the process. The following is a brief description of the research pathways being pursued by DOE/NETL’s Carbon Capture area.

Chemical Solvents

Chemical solvents are currently used in some industrial applications for CO₂ capture. Chemical absorption involves one or more reversible chemical reactions between CO₂ and an aqueous solution of an absorbent, such as an alkanolamine, hindered amine, or aqueous ammonia. Monoethanolamine (MEA) is a widely used solvent in the capture scheme depicted in Figure 3. Although high levels of CO₂ capture are possible, the drawback of this process is that significant

amounts of energy are required in the regeneration step. Chemical solvents are likely to be preferred for cases with low concentrations of CO₂ in the combustion gases. Since they are equilibrium limited, they do not gain significant advantage by operating at elevated pressure.

Figure 3 - Schematic Diagram of the Amine Separation Process



Source: DOE/NETL

Physical Solvents

Another CO₂ capture process currently being used in industrial applications is based on physical solvents, rather than chemical solvents. The advantage of physical solvents is that less energy is required in the solvent regeneration step. Physical absorption is a bulk phenomenon where inorganic or organic liquids may be used to preferentially absorb a gaseous species from the gas mixture. The absorption liquid is regenerated by increasing its temperature or reducing its pressure. This type of process could be a very efficient approach for processing high-pressure CO₂-rich streams, such as those encountered in IGCC systems.⁶

Currently, the state-of-the-art for physical solvents is the glycol-based Selexol™ process, but just like MEA, it has not been built or operated at the scale and conditions needed by the power generation sector. The Rectisol® process is similar to Selexol™, except methanol is used as the solvent. A promising breakthrough concept involves ionic liquids. These organic salts have high thermal stability and low volatility, which is beneficial for CO₂ capture processes, since this can help minimize solvent losses and be a more cost-effective solution than current technologies.

Chemical Sorbents

Currently under development, chemical adsorption involves the bonding of CO₂ with a solid sorbent. A temperature increase facilitates the sorbent regeneration step. A potential complication of thermal regeneration could be the need of a sweep gas during the regeneration step. Some potential chemical sorbents are sodium and potassium oxides, carbonates, and amine-enriched sorbents. Amine-enriched sorbents have an amine that strongly bonds to sites on a solid

substrate. The implanting of the amine on a solid substrate increases the surface contact area of the amine for CO₂, thus requiring less sorbent/amine.

Physical Sorbents

Another process being developed is based on physical adsorption of gases on solid adsorbents in which a few layers of the adsorbed gas are held by weak surface forces. Adsorption capacity increases with CO₂ partial pressure or a reduction in temperature. The key issue for this type of capture system is the surface area required per unit mass or volume of adsorbed gas. The loaded adsorbent can be regenerated via two mechanisms: temperature swing adsorption (TSA) and pressure swing adsorption (PSA). In TSA, the adsorbent is regenerated by raising its temperature.⁷ PSA is currently used to separate hydrogen from CO₂ and other mixed gas streams. The process works by removing the most adsorbable gas species and regenerating the adsorption bed by decreasing the pressure and purging the capture gas. While PSA produces a highly pure hydrogen stream, it does not selectively separate CO₂ from other gases in the stream, and therefore does not produce a pure CO₂ product for storage.

A promising breakthrough concept, metal organic frameworks (MOFs) represent a class of porous materials that offer these advantages for CO₂ capture: ordered structures, high thermal stability, adjustable chemical functionality, extra-high porosity, and the availability of hundreds of crystalline, well-characterized porous structures.

Membranes

The use of membranes to physically separate CO₂ from the other products of combustion/gasification is another capture process under development. Diffusion mechanisms in membranes differ greatly depending on the type of membrane used. Generally, gas separation is accomplished by some physical or chemical interaction between the membrane and the gas being separated, causing one component in the gas to permeate through the membrane faster than another. The quality of the separation is determined by the membrane selectivity, by the ratio of the permeate flow to the feed flow, and by the ratio of permeate pressure to the feed pressure. Usually, the selectivities of the membranes are insufficient to achieve the desired purities and recoveries, therefore multiple stages and recycle streams may be required in an actual operation, leading to increased complexity, energy consumption, and capital costs. Research has been conducted with a number of different types of gas separation membranes, including polymer, palladium, facilitated transport, and molecular sieves.^{8,9} Their potential is generally viewed as good for high-pressure applications, such as IGCC, but not as promising for combustion flue gas.

Gas absorption membrane technologies are also under development, where the separation is caused by the presence of an absorption liquid on one side of the membrane that selectively removes CO₂ from a gas stream on the other side of the membrane. These membranes are essentially used as a contacting device between the gas flow and a liquid flow. Membrane-solvent systems that use an amine as the solvent are one example.

Another novel approach is the use of enzymatic CO₂ processes. In these processes, the enzyme, carbonic anhydrase, acts as a catalyst to enhance removal of CO₂ from mixed gas streams in liquid membrane reactor designs.

Oxy-combustion Systems

For oxy-combustion to be a cost-effective power generation option, a low-cost supply of pure oxygen is required. In the most frequently proposed version of this concept, a cryogenic air separation unit is used to supply high purity oxygen to the boiler. This commercially available technology is both capital and energy-intensive and could raise the cost of electricity from coal-fired plants considerably, in addition to degrading the overall plant efficiency. However, novel technologies currently under development, such as oxygen or ion transport membranes, have the potential to reduce the cost of oxygen production.

Another breakthrough oxy-combustion concept under development is the chemical looping combustion process. Chemical looping splits combustion into separate oxidation and reduction reactions. A metal (e.g., iron, nickel, copper, or manganese) oxide is used as an oxygen carrier which then releases the oxygen in a reducing atmosphere and the oxygen reacts with the fuel. The metal is then recycled back to the oxidation chamber where the metal oxide is regenerated by contact with air. The advantage of using two chambers for the combustion process is that the CO₂ is concentrated, once the water is removed, and not diluted with nitrogen gas. The benefit of the process is that no air separation plant or external CO₂ separation equipment is required.

Carbon Storage

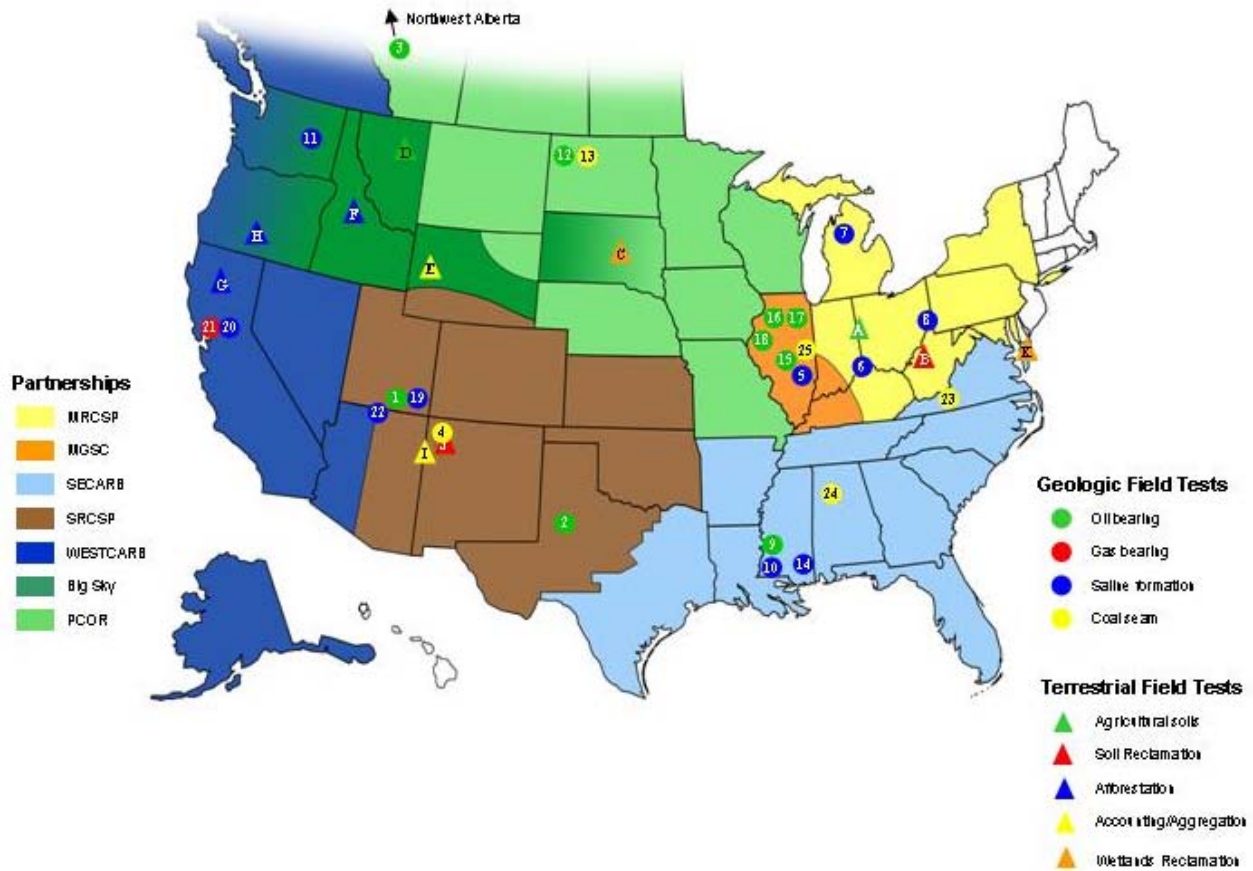
Carbon storage is defined as the placement of CO₂ into a repository in such a way that it will remain stored (sequestered) permanently. It includes geologic and terrestrial sequestration. Geologic sequestration involves the injection of CO₂ into underground reservoirs, such as oil and gas bearing formations, saline formations, oil and gas rich organic shales, basalt outcrops, and unmineable coal seams, that have the ability to securely contain CO₂ over long periods of time. As a value-added benefit, CO₂ injected into oil and gas reservoirs and unmineable coal seams can facilitate enhanced oil recovery (EOR) and enhanced coal bed methane recovery (ECBM). Terrestrial carbon sequestration is defined as the net removal of CO₂ from the atmosphere by the soil and plants and/or the prevention of CO₂ net emissions from terrestrial ecosystems into the atmosphere.

Geographic differences in fossil fuel use and potential sequestration storage sites across the U.S. dictate the use of regional approaches in addressing CO₂ sequestration. DOE/NETL launched the RCSP in 2003 to examine regional differences in geology, land practices, ecosystem management, and industrial activity that can affect the deployment of CCS technologies. The RCSP is being implemented in three interrelated phases: Characterization Phase (FY2003-FY2005), Validation Phase (FY2005-FY2009), and the Deployment Phase (FY2008-FY2017). Although the RCSP includes three phases, it should be viewed as an integrated whole, with many of the goals and objectives transitioning from one phase to the next. Accomplishments and results from the Characterization Phase have helped to refine goals and activities in the Validation Phase, and results from the Validation Phase are expected to flow into and enhance the Deployment Phase.

As shown in Figure 4, the RCSP Program includes a network of seven partnerships: Big Sky Carbon Sequestration Partnership (Big Sky), Midwest Geological Sequestration Consortium

(MGSC), Midwest Regional Carbon Sequestration Partnership (MRCSP), Plains CO₂ Reduction Partnership (PCOR), Southeast Regional Carbon Sequestration Partnership (SECARB), Southwest Regional Partnership on Carbon Sequestration (SWP), and the West Coast Regional Carbon Sequestration Partnership (WESTCARB). Collectively, the seven RCSPs represent regions encompassing 97% of coal-fired CO₂ emissions, 97% of industrial CO₂ emissions, 96% of the total land mass, and essentially all the geologic sequestration sites in the U.S. potentially available for carbon storage.

Figure 4 – Regional Carbon Sequestration Partnerships



The process of sequestering CO₂ involves identifying sources that produce CO₂ and identifying sequestration sites where the CO₂ can be stored. Based on data assembled for the *Carbon Sequestration Atlas of the United States and Canada*,¹⁰ 4,365 identified stationary sources in the seven RCSP regions and the northeastern U.S. generate about 3.8 billion metric tons of CO₂ annually. The aggregate CO₂ sink capacity – including saline formations, unmineable coal seams, and oil and natural gas reservoirs – is estimated to range up to 3,643 billion metric tons, enough to sequester CO₂ emissions at current annual generation rates for hundreds of years.

Monitoring, Mitigation, and Verification

Monitoring, mitigation, and verification (MMV) capabilities will be critical in ensuring the long-

term viability of geological sequestration – satisfying both technical and regulatory requirements. Monitoring and verification encompass the ability to measure the amount of CO₂ stored at a specific sequestration site, to monitor the site for leaks, to track the location of the underground CO₂ plume, and to verify that the CO₂ is stored in a way that is permanent and not harmful to the host ecosystem. Mitigation is the near-term ability to respond to risks such as CO₂ leakage or ecological damage in the unlikely event that they should occur.

In general, MMV research is aimed at providing an accurate accounting of stored CO₂ and a high level of confidence that the CO₂ will remain sequestered permanently. A successful effort will enable sequestration project developers to obtain permits for sequestration projects while ensuring human health and safety and preventing potential damage to the host ecosystem. MMV also seeks to set the stage for emissions reduction credits, if a domestic program is established, that approach 100% of injected CO₂, contributing to the economic viability of sequestration projects. Finally, MMV will provide improved information and feedback to sequestration practitioners, thus accelerating technology progress.

SUMMARY

Currently, in the absence of regulations limiting or taxing carbon emissions, the private sector has little incentive to develop and deploy commercial CCS technologies. However, through cost-shared R&D, the Federal government has a role to play in ensuring the availability of cost-effective technologies for capturing and sequestering CO₂ from fossil fuel use. Commercial availability of CCS technology will provide public benefits in the form of the continued use of low-cost fossil fuels in an environmentally friendly manner. Additional information related to DOE/NETL's Carbon Sequestration Program is available at NETL's web site (http://www.netl.doe.gov/technologies/carbon_seq/index.html).

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DISCLAIMER

References in this paper to any specific commercial product or service are to facilitate understanding and do not imply endorsement by the U.S. Department of Energy.

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