CARBON SEQUESTRATION
RESEARCH AND DEVELOPMENT

Office of Science
Office of Fossil Energy
U.S. Department of Energy

December 1999

Dave Reichle, ORNL
John Houghton, DOE
Bob Kane, DOE
Jim Ekmann, FETC

Sally Benson, LBNL
John Clarke, PNNL
Roger Dahlman, DOE
George Hendrey, BNL
Howard Herzog, MIT
Jennie Hunter-Cevera, LBNL
Gary Jacobs, ORNL
Rod Judkins, ORNL
Joan Ogden, Princeton
Anna Palmisano, DOE
Robert Socolow, Princeton
John Stringer, EPRI
Terry Surles, LLNL
Alan Wolsky, ANL
Nicholas Woodward, DOE
Michael York, DOE

This report is available on the world wide web at
www.ornl.gov/carbon_sequestration/

This report has been reproduced from the best available copy.

Reports are available to the public from the following source.

National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone 703-605-6000 (1-800-553-6847)
TDD 703-487-4639
Fax 703-605-6900
E-mail orders@ntis.fedworld.gov
Web site www.ntis.gov/ordering.htm

Reports are available to U.S. Department of Energy (DOE) employees, DOE contractors, Energy Technology Data Exchange (ETDE) representatives, and International Nuclear Information System (INIS) representatives from the following source.

Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831
Telephone 865-576-8401
Fax 865-576-5728
E-mail reports@adonis.osti.gov
Web site www.osti.gov/products/sources.html

Reports produced after January 1, 1996, are generally available via the DOE Information Bridge.

Web site www.doe.gov/bridge

CONTENTS

FIGURES

TABLES

ABBREVIATIONS, ACRONYMS, AND INITIALISMS

BER	Office of Biological and Environmental Research (DOE)
BES	Office of Basic Energy Sciences (DOE)
CO	carbon monoxide
CO_2	carbon dioxide
DEA	diethanolamine
DOE	U.S. Department of Energy
EOR	enhanced oil recovery
ESA	electrical swing adsorption
FACE	Free Air CO_2 Enrichment
GHG	Greenhouse Gas R&D Programme
GtC	billion tonnes of atmospheric carbon
H_2	hydrogen gas
HNLC	high-nutrient, low-chlorophyll (ocean waters)
IEA	International Energy Agency
IGCC	integrated gasification combined cycle
IPCC	Intergovernmental Panel on Climate Change
m	meter
MBARI	Monterey Bay Aquarium Research Institute
MDEA	methyldiethanolamine
MEA	monoethanolamine
MIT	Massachusetts Institute of Technology
MPa	million Pascal (a measure of pressure)
NASA	National Aeronautics and Space Administration
NEE	net ecosystem exchange
NGO	nongovernmental organization
nm	nanometer
NO_x	oxides of nitrogen
NSF	National Science Foundation
OCMIP	Ocean Carbon-Cycle Model Intercomparison Project
OGCM	ocean general circulation model
PCAST	President's Council of Advisors on Science and Technology
POC	particulate organic carbon
ppm	parts per million
PSA	pressure swing adsorption
R&D	research and development
ROV	remotely operated vehicle
SO_x	oxides of sulfur
TSA	thermal swing adsorption
USDA	United States Department of Agriculture
USGS	United States Geological Survey

EXECUTIVE SUMMARY

Predictions of global energy use in the next century suggest a continued increase in carbon emissions and rising concentrations of carbon dioxide (CO_2) in the atmosphere unless major changes are made in the way we produce and use energy—in particular, how we manage carbon. For example, the Intergovernmental Panel on Climate Change (IPCC) predicts in its 1995 "business as usual" energy scenario that future global emissions of CO_2 to the atmosphere will increase from 7.4 billion tonnes of carbon (GtC) per year in 1997 to approximately 26 GtC/year by 2100. IPCC also projects a doubling of atmospheric CO_2 concentration by the middle of next century and growing rates of increase beyond. Although the effects of increased CO_2 levels on global climate are uncertain, many scientists agree that a doubling of atmospheric CO_2 concentrations could have a variety of serious environmental consequences.

One way to manage carbon is to use energy more efficiently to reduce our need for a major energy and carbon source—fossil fuel combustion. Another way is to increase our use of low-carbon and carbon-free fuels and technologies (nuclear power and renewable sources such as solar energy, wind power, and biomass fuels). Both approaches are supported by the U.S. Department of Energy (DOE) and are not the focus of this report.

The third and newest way to manage carbon, capturing and securely storing carbon emitted from the global energy system (carbon sequestration), is truly radical in a technology context. The development of today's fossil-energy-based system is rooted in the Industrial Revolution. For over 200 years, the development of energy technology has been focused on lowering costs through increased efficiency to support economic growth. Because of their abundance, availability, and high energy content, coal, oil, and natural gas have proved to be attractive energy sources to produce electricity, run industrial processes, propel transportation vehicles, and provide energy for residential and commercial applications. As fossil energy use increased and adverse environmental effects became apparent, energy technology also evolved to minimize them. However, all of this enormous technology development has assumed that the free venting of CO_2 to the atmosphere was environmentally harmless. Only recently has the increasing concentration of CO_2 in the atmosphere been considered to represent a serious environmental problem. The consequence is that we have developed an intricate, tightly coupled energy system that has been optimized over 200 years for economy, efficiency, and environmental performance, but not for the capture and sequestration of its largest material effluent, CO_2.

The goal of this report is to identify key areas for research and development (R&D) that could lead to an understanding of the potential for future use of carbon sequestration as a major tool for managing carbon emissions. Under the leadership of DOE, researchers from universities, industry, other government agencies, and DOE national laboratories were brought together to develop the technical basis for conceiving a science and technology road map. That effort has resulted in this report, which develops much of the information needed for the road map.

This report identifies the R&D topics necessary to understand and develop critical options for the capture, transport, conversion, and sequestration of carbon. It addresses known sources of carbon (industrial sources, power plant flue gases, preprocessed fossil fuels before combustion); carbon forms for sequestration (CO_2, elemental carbon, and minerals that contain carbon); and options for sequestration sinks—oceans, geologic formations, soils and vegetation (see Chaps. 3 through 7).

THE ROAD MAP VISION AND GOALS

The vision for the road map is to

Possess the scientific understanding of carbon sequestration and develop to the point of deployment those options that ensure environmentally acceptable sequestration to reduce anthropogenic CO_2 emissions and/or atmospheric concentrations. The goal is to have the potential to sequester a significant fraction of 1 GtC/year in 2025 and 4 GtC/year in 2050.

The purpose of carbon sequestration is to keep anthropogenic carbon emissions from reaching the atmosphere by capturing them, isolating them, and diverting them to secure storage and/or to remove CO_2 from the atmosphere by various means and store it. Any viable system for sequestering carbon must be safe, environmentally benign, effective, and economical. In addition, it must be acceptable to the public.

Why is carbon sequestration important? Given the magnitude of carbon reductions needed to stabilize the atmosphere, capture and sequestration could be a major tool for reducing carbon emissions to the atmosphere from fossil fuels; in fact, sequestration may be essential for the continued large-scale use of fossil fuels. It will allow greater flexibility in the future primary energy supply. In addition, it could offer other benefits such as the manufacture of commercial products (e.g., construction materials and plastics); improved agricultural practices that could reduce soil erosion, conserve water, and increase the sustainability of food production; the restoration of wetlands, which would help preserve wildlife and protect estuaries; increased biodiversity; enhanced recovery of oil and methane (from coal beds); and the development of exportable technologies to help the U.S. economy.

THE GLOBAL CARBON CYCLE AND FOSSIL FUELS

Most anthropogenic (human-activity-related) emissions of carbon to the atmosphere result from combustion of fossil fuels for the economical production of energy. If the demand for energy continues to increase, it is possible that the only way that fossil fuels can be used for large-scale energy production is through the development and implementation of carbon capture and sequestration options.

Given the magnitude of carbon emission reductions needed to stabilize the atmospheric CO_2 concentration, multiple approaches to carbon management (i.e., improved energy efficiency and clean energy systems) will be needed. All potentially important technical options should be explored.

SCIENTIFIC AND TECHNICAL NEEDS FOR CARBON SEQUESTRATION

STRATEGIC ISSUES

Following are the general recommendations of the report addressing strategic issues regarding a comprehensive carbon sequestration program.

- Sequestration R&D could expand the world's future options for dealing with greenhouse gases.
- Many carbon sequestration options are particularly amenable to improving existing activities—such as CO_2 injection during secondary oil recovery—and often provide important secondary benefits, such as improving ecosystems during reforestation.
- Some carbon sequestration options, such as improved agricultural practices, are available practically immediately. Examining ongoing, field-scale sequestration investigations in terrestrial, geological, and ocean systems can provide critical experience for designing the necessary environmental research programs.
- Some carbon sequestration options that have limited capacity or relatively short carbon residence times could nonetheless make important near-term contributions during a transition to other longer-term carbon management options. Other carbon sequestration options can provide significant long-term contributions.
- For carbon sequestration to be a viable option, it needs to be safe, predictable, reliable, measurable, and verifiable; and it needs to be competitive with other carbon management options, such as energy-efficient systems and decarbonized energy technologies.
- Carbon sequestration is an immature field, so multiple fundamental R&D approaches are warranted and significant breakthroughs can be expected. The federal government is an appropriate sponsor of carbon sequestration R&D.
- Integrated analyses of the carbon sequestration system should be periodically updated to evaluate the potential contributions, costs, and benefits of various carbon sequestration options.
- The information from the R&D program should be provided to policy makers to aid them in developing policy and selecting the most efficient and effective solutions to the issues of climate change.

Separation and Capture of CO_2 from the Energy System

Several currently available technologies can be used to separate and capture CO_2 from fossil-fueled power plant flue gases; from the effluents of industrial processes such as iron, steel, and cement production; and from hydrogen production by reforming of natural gas. CO_2 can be absorbed from gas streams by contact with amine-based solvents or cold methanol. It can be removed by adsorption on activated carbon or other materials or by passing the gas stream through special membranes. Commercial hydrogen production via reforming of natural gas involves separating H_2 from the reformate gases (a mixture of unreacted methane

Geologic or ocean storage sequestration options that use a concentrated source of CO_2 require low-cost carbon separation and capture techniques to be viable options. The scale of the industrial system required to process gigatonnes of carbon warrants investigation into new solvents, adsorbents, and membrane separation devices for either pre- or post-combustion separation.

and other hydrocarbons, CO, CO_2, and water) by adsorption processes such as pressure swing adsorption (PSA). Should fuels decarbonization (e.g., reforming of natural gas to produce H_2) become part of a CO_2 mitigation strategy, the PSA technology could logically be extended to CO_2 separation and capture.

Advanced methods might include adsorbing CO_2 on zeolites or carbon-bonded activated carbon fibers and separating it from flue gases or process gases from industrial operations using inorganic membranes. The use of commercial CO_2-removing processes that scrub gases with amine-based solvents is projected to raise the cost of producing electrical power from coal-fired power plants using existing technology. Capture and sequestration could increase the cost of electrical power generation from coal by as much as 20 to 30 mills/kWh. Thus although CO_2 is separated routinely, dramatic improvements are necessary to make the process economical (Chap. 2). Techniques are needed to transform the captured CO_2 into materials that (1) can be economically and safely transported and sequestered for a long time or (2) can be used to make commercial products (e.g., construction materials) that could offset the costs of separation and capture.

There are numerous options for the separation and capture of CO_2, and many of these are commercially available. However, none has been applied at the scale required as part of a CO_2 emissions mitigation strategy, nor has any method been demonstrated for all the anthropogenic sources considered in this R&D map. Many issues remain regarding the ability to separate and capture CO_2 from anthropogenic sources on the scale required, and to meet the cost, safety, and environmental requirements for separation and capture. In our assessment of the scientific and technological gaps between the requirements for CO_2 separation and capture and the capabilities to meet these requirements, many explicit and specific R&D needs were identified.

- A science-based and applications-oriented R&D program is needed to establish the efficacy of current and novel CO_2 separation processes as important contributors to carbon emissions mitigation. Important elements of such a program include the evaluation, improvement, and development of chemical and physical absorption solvents, chemical and physical adsorbents, membrane separation devices with selectivity and specificity for CO_2-containing streams, molecular and kinetic modeling of the materials and processes, and laboratory-scale testing of the selected processes.

- Field tests are needed of promising new CO_2 separation and capture options in small bypass streams at large point sources of CO_2, such as natural gas wells and hydrogen production plants.

The ocean provides a large potential reservoir. Active experiments are already under way in iron fertilization and other tests of enhanced marine biological sequestration, as well as deep CO_2 injection. Improvements in understanding marine systems will be needed before implementation of major marine sequestration campaigns.

Sequestration in the Oceans

The ocean represents a large potential sink for sequestration of anthropogenic CO_2 (Chap. 3). Two methods are proposed for the sequestration of carbon in the ocean: (1) A relatively pure CO_2 stream that has been generated by a power plant, decarbonized fuel production system, or industrial facility could be injected directly into the ocean. The injected CO_2 may become trapped in ocean sediments or ice-like solids, called hydrates. (2) The net oceanic uptake from the atmosphere could be enhanced through a method such as iron fertilization. These approaches will require better understanding of marine ecosystems to enhance the effectiveness of applications and avoid undesirable consequences.

- Field experiments of CO_2 injection into the ocean are needed to study the physical/chemical behavior of the released CO_2 and its potential for ecological impact.
- Ocean general circulation models need to be improved and used to determine the best locations and depths for CO_2 injection and to determine the long-term fate of CO_2 injected into the ocean.
- The effect of fertilization of surface waters on the increase of carbon sequestered in the deep ocean needs to be determined, and the potential ecological consequences on the structure and function of marine ecosystems and on natural biogeochemical cycling in the ocean need to be studied.
- New innovative concepts for sequestering CO_2 in the ocean need to be identified and developed.

Sequestration in Terrestrial Ecosystems

The terrestrial biosphere is a large and accessible reservoir for sequestering CO_2 that is already present in the atmosphere. Natural carbon fluxes are huge, so even small forced changes resulting from R&D advances would be very significant. It will be important to address the consequences of altering the natural flux.

Terrestrial ecosystems, which are made up of vegetation and soils containing microbial and invertebrate communities, sequester CO_2 directly from the atmosphere (Chap. 4). The terrestrial ecosystem is essentially a huge natural biological scrubber for CO_2 from all fossil fuel emissions sources, such as automobiles, power plants, and industrial facilities. Computer models estimate that terrestrial ecosystems—forests, vegetation, soils, farm crops, pastures, tundras, and wetlands—have a net carbon accumulation of about one-fourth

(1.5 to 2 GtC) of the 7.4 GtC emitted annually into the atmosphere by fossil fuel combustion and land use changes. If there were an increased focus on practices to enhance the natural carbon cycle, the potential for terrestrial ecosystems to remove and sequester more carbon from the atmosphere could be increased by, for example, improving agricultural cultivation practices to reduce oxidation of soil carbon and enhancing soil texture to trap more carbon, and protecting wetlands.

- The terrestrial ecosystem is a major biological scrubber for atmospheric CO_2 (present net carbon sequestration is ~2 GtC/year) that can be significantly increased by careful manipulation over the next 25 years to provide a critical "bridging technology" while other carbon management options are developed. Carbon sequestration could conceivably be increased by several gigatonnes per year beyond the natural rate of 2 GtC per year, but that may imply intensive management and/or manipulation of a significant fraction of the globe's biomass. However, those potentials do not yet include a total accounting of economic and energy costs to achieve these levels. Ecosystem protection is important and may reduce or prevent loss of carbon currently stored in the terrestrial biosphere. The focus for research, however, should be on increasing the rate of long-term storage in soils in managed systems.

- Research on four key interrelated R&D topics is needed to meet goals for carbon sequestration in terrestrial ecosystems:

  — Increase understanding of ecosystem structure and function directed toward carbon allocation and partitioning, nutrient cycling, plant and microbial biotechnology, molecular genetics, and functional genomics.

  — Improve measurement of gross carbon fluxes and dynamic carbon inventories through improvements to existing methods and through development of new instrumentation for in situ, nondestructive below-ground observation and remote sensing to allow aboveground biomass measurement, verification, and monitoring of carbon stocks.

  — Implement scientific principles into tools such as irrigation methods, efficient nutrient delivery systems, increased energy efficiency in agriculture and forestry, and increased byproduct use.

  — Assess ecosystem behavior in response to carbon sequestration strategies in an environment of a changing climate, using a suite of models (including life cycle analysis) to integrate across scales from physiological processes to regional ecosystem management practices.

- Field-scale experiments in large-scale ecosystems will be necessary to understanding both physiological and geochemical processes regulating carbon sequestration based upon integrative ecosystem models. Such carbon sequestration experiments are needed to provide proof-of-principle testing of new sequestration concepts and integration of sequestration science and engineering principles.

Sequestration in Geologic Formations

> Limited geological sequestration is being practiced today, but it is not yet possible to predict with confidence storage volumes and integrity over long time periods. Many important issues must be addressed to reduce costs, ensure safety, and gain public acceptance.

Three principal types of geologic formations are widespread and have the potential for sequestering large amounts of CO_2. They are active and uneconomical oil and gas reservoirs, aqueous formations, and deep and unmineable coal formations. About 70 oil fields worldwide use injected CO_2 for enhanced oil recovery. CO_2 sequestration is already being practiced in a sub-seabed reservoir in the North Sea of Norway. The United States has sufficient capacity, diversity, and broad geographic distribution of potential reservoirs to use geologic sequestration in the near term (Chap. 5). The primary uncertainty is the effectiveness of storing CO_2 in geological formations—how easily CO_2 can be injected and how long it will remain. Only through experience will enough knowledge be gained to assess the ultimate sequestration potential of geologic formations.

- Fundamental and applied research is needed to improve the ability to understand, predict, and monitor the performance of sequestration in oil, gas, aqueous, and coal formations. Elements of such a program include multiphase flow in heterogeneous and deformable media; phase behavior; CO_2 dissolution and reaction kinetics, micromechanics and deformation modeling; coupled hydrologic-chemical-mechanical-thermal modeling; and high-resolution geophysical imaging. Advanced concepts should be included, such as enhancement of mineral trapping with catalysts or other chemical additives, sequestration in composite geologic formations, microbial conversion of CO_2 to methane, rejuvenation of depleted oil reservoirs, and CO_2-enhanced methane hydrate production.

- A nationwide assessment is needed to determine the location and capacity of the geologic formations available for sequestration of CO_2 from each of the major power-generating regions of the United States. Screening criteria for choosing suitable options and assessing capacity must be developed in partnership with industry, the scientific community, and public and regulatory oversight agencies.

- Pilot-scale field tests of CO_2 sequestration should be initiated to develop cost and performance data and to help prioritize future R&D needs. The tests must be designed and conducted with sufficient monitoring, modeling, and performance assessment to enable quantitative evaluation of the processes responsible for geologic sequestration. Pilot testing will lay the groundwork for collaboration with industrial partners on full-scale demonstration projects.

Advanced Biological Processes

> Advanced biological techniques may produce options too radical to predict. Some biologic processes can sequester carbon products at low cost. New carbon sequestration options could become feasible and others could be improved using advanced biological techniques.

Advanced biological processes (Chap. 6) could be developed and implemented to limit emissions and capture and sequester carbon both from relatively concentrated utility and industrial combustion gases, and from dispersed point sources. Bacteria and other organisms could be used to remove carbon from fuels and to recycle carbon from man-made waste streams. In addition, crop wastes and dedicated crops could be used as feedstocks for biological and chemical conversion processes to manufacture fuels and chemicals. Advanced crop species and cultivation practices could be designed to increase the uptake of atmospheric CO_2 by terrestrial and aquatic biomass and decrease CO_2 emissions to the atmosphere from soils and terrestrial and aquatic biomass.

The 21st Century has been referred to as the "Century for Biology." Indeed, many new molecular tools have been developed that will aid in new discoveries and assist in providing solutions to key problems facing humankind and the planet. The difference that advanced biological techniques can make will be evident when they are integrated with land, subsurface, and ocean management practices. The following actions will promote cost-effective and stable biological approaches to carbon sequestration.

- Research should be initiated on the genetic and protein engineering of plants, animals, and microorganisms to address improved metabolic functions that can enhance, improve, or optimize carbon management via carbon capture technology, sequestration in reduced carbon compounds, use in alternative durable materials, and improved productivity.

- The objectives and goals of the advanced biological research should be linked to those specific problems and issues outlined for carbon sequestration in geological formations, oceans, and soils and vegetation so that an integrated research approach can elucidate carbon sequestration at the molecular, organism, and ecosystem levels.

- Short-, mid-, and long-term goals in advanced biological research should be instituted so that scale-up issues, genetic stability in natural settings, and efficacy in the field can be assessed.

Advanced Concepts

Most carbon sequestration options rely on chemical reactions to achieve benign, stable, and inert products. Studies to enhance the relevant chemistry almost certainly will reduce the costs or increase the effectiveness of these options. Results from R&D on advanced chemical topics also may make it possible to generate useful and marketable byproducts.

Many of the sequestration technologies described in this document depend on chemistry. Improved methods of separation, transport, and storage of CO_2 will benefit from research on and development of advanced chemical techniques to address sequestration via chemical transformations (Chap. 7). Any viable sequestration technique must store vast amounts of carbon-rich materials, so environmental chemistry will be valuable to determine whether these materials will be stable when sequestered. Many issues pertaining to aqueous carbonate/ bicarbonate chemistry are relevant to sequestration of carbon in oceans, geological formations, and groundwater. Carbonate chemistry in very basic solutions may lead to a method for extracting CO_2 from air. Clathrates, compounds that can enclose molecules such as CO_2 within their crystal structure, may be used to separate CO_2 from high-pressure systems. Learning clathrate properties may be important to understanding chemical approaches to ocean storage of carbon, and subsurface arctic and marine hydrate formations may also be evaluated as geologic sequestration options.

- The proper focus of R&D into advanced chemical sciences and technologies is on transforming gaseous CO_2 or its constituent carbon into materials that either are benign, inert, long-lived and contained in the earth or water of our planet, or have commercial value.
  - — Benign by-products for sequestration should be developed. This avenue may offer the potential to sequester large (gigatonne) amounts of anthropogenic carbon.
  - — Commercial products need to be developed. This approach probably represents a lesser potential (millions of tonnes) but may result in collateral benefits.
- The chemical sciences can fill crucial gaps identified in the other focus areas. In particular, environmental chemistry is an essential link in determining the impact and consequences of these various approaches. Studies to address the specific gaps identified in Chap. 7 should be conducted to ensure that other focus areas meet their potential.

DEVELOPING A CARBON SEQUESTRATION ROAD MAP

An emerging science and technology road map seeks to identify the scientific and technological developments needed to achieve a specific policy goal. The process of identifying the needed science and technology must be focused by developing a concept of the technological system (Chap. 8). This task is particularly difficult in the case of carbon capture and sequestration because the understanding necessary to design such a system is still immature.

Today, carbon is emitted to the atmosphere from many sources that were not designed to capture, let alone sequester, these emissions. There are many ideas for, and even demonstrations of, technology to capture and sequester carbon from fossil fuel combustion. Many of the requisite new energy production technologies are already under development at DOE. However, the current energy system probably must be modified significantly to make an economical capture and sequestration system possible. Thus, the emerging technology road map for carbon capture and sequestration cannot be constructed apart from consideration of current and emerging energy technologies. It will involve an iterative process to connect this road map with others being developed by DOE for various parts of the energy technology system.

This report is a significant first step toward the development of an emerging technology road map for carbon capture and sequestration. We start from a bold vision of having the scientific and technical knowledge to make carbon sequestration a major carbon management option by 2025. Guided by this vision, each of the technical focus chapters (2–7) identifies key areas for scientific and technical development, including new areas outside traditional energy technology development.

We have begun the process of exploring the mutual relationships and interdependencies of the scientific and technological developments in all these fields by building a series of road map linkages. This process has illuminated how progress in one area affects the total system. However, R&D priorities and performance requirements have not yet been defined. Nor has the phasing of potential R&D schedules been considered. Developing linkages has allowed us to eliminate overlaps to some extent, but gaps in the technology needs have not yet been examined. Before proceeding much further, much more work must be done on specifying the economic constraints and technology needs of the integrated carbon capture and sequestration system. The road map outline presented in this document, especially the research needs delineated in Chaps. 2–7, provides the sound basis for taking these next steps toward a fully realized program in carbon sequestration. This report should be used as a framework in organizing a wider examination by diverse stakeholders of the science and technology required for carbon capture and sequestration.