

Carbon Dioxide Capture for Storage in Deep Geologic Formations – Results from the CO₂ Capture Project

**Capture and Separation of Carbon Dioxide
from Combustion Sources**

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Volume 1



ELSEVIER

2005

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First edition 2005

Library of Congress Cataloging in Publication Data

A catalog record is available from the Library of Congress.

British Library Cataloguing in Publication Data

A catalogue record is available from the British Library.

ISBN: 0-08-044570-5 (2 volume set)

Volume 1: Chapters 8, 9, 13, 14, 16, 17, 18, 24 and 32 were written with support of the U.S. Department of Energy under Contract No. DE-FC26-01NT41145. The Government reserves for itself and others acting on its behalf a royalty-free, non-exclusive, irrevocable, worldwide license for Governmental purposes to publish, distribute, translate, duplicate, exhibit and perform these copyrighted papers. EU co-funded work appears in chapters 19, 20, 21, 22, 23, 33, 34, 35, 36 and 37. Norwegian Research Council (Klimatek) co-funded work appears in chapters 1, 5, 7, 10, 12, 15 and 32.

Volume 2: The Storage Preface, Storage Integrity Preface, Monitoring and Verification Preface, Risk Assessment Preface and Chapters 1, 4, 6, 8, 13, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33 were written with support of the U.S. Department of Energy under Contract No. DE-FC26-01NT41145. The Government reserves for itself and others acting on its behalf a royalty-free, non-exclusive, irrevocable, worldwide license for Governmental purposes to publish, distribute, translate, duplicate, exhibit and perform these copyrighted papers. Norwegian Research Council (Klimatek) co-funded work appears in chapters 9, 15 and 16.

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Chapter 4

POST-COMBUSTION CO₂ SEPARATION TECHNOLOGY SUMMARY

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ABSTRACT

The post-combustion technology team (the Team) found a number of interesting ways in which the cost of carbon dioxide removal could be reduced from the base case. A number of technologies have been investigated. It is the feeling of the Team that there are many good research opportunities with respect to reducing the total cost of carbon dioxide removal. Extensive studies were completed in these areas, all based on absorption:

- Base case.
- Nexant low-cost design.
- Nexant integrated low-cost design.
- Kvaerner-Mitsubishi (MHI): a new contactor combined with a new absorbent.

The Team has also evaluated:

- Better adsorbents, and the potential of adsorption processes.
- The basis for dismissal of membranes and cryogenics for this application.
- New chemical approaches, producing a few research proposals.
- Alternative equipment technologies.

Finally, the Team has defined what is referred to as best integrated technology (BIT) based on the studied absorption technologies. BIT is not ready to be built, as some features need to be checked. The BIT concept must not be seen as the ultimate post-combustion solution as there are identified research opportunities available, and more are foreseen for the future.

INTRODUCTION

In post-combustion capture, CO₂ is recovered from the exhaust gases of emission sources such as boilers, heaters, and turbines. These sources are present everywhere around the world in refineries, power plants, gas processing plants and chemical plants. Improving the economics of post-combustion capture was a critical goal for the entire CO₂ Capture Project (CCP). The CCP's goal was to achieve a 50% cost reduction

Abbreviations: ANSI, American National Standards Institute; API, American Petroleum Institute; ARI, Adsorption Research Inc; BIT, best integrated technology; ESA, electrical swing adsorption; HRSG, heat recovery steam generation; MEA, monoethanolamine; MHI, Mitsubishi Heavy Industries; NTNU, Norwegian University of Science and Technology; ORNL, Oak Ridge National Laboratory; PSA, pressure swing adsorption; QP, quick place (here: the CCP data base); S&T, Shell and Tube; SINTEF, A Norwegian research organisation (<http://www.sintef.no>); TNO, A Dutch research organisation, (<http://www.tno.nl>); UOP, Universal Oil Products; VSA, vacuum swing adsorption.

in retrofits and a 75% cost reduction in new built plants. This section of the current volume summarises the results of our research over the past 3 years. Progress towards the cost-reduction goals was reviewed in Chapter 4 of this volume.

The CCP post-combustion technology team was formed at the beginning of the CCP, and has since evaluated numerous technologies that were believed to be valuable for CO₂ separation from flue gases.

Current post-combustion capture practice is to install an amine separation unit at the flue gas source. This is a very difficult separation, since the gases are hot, dilute in CO₂ content, near atmospheric pressure, high in volume, and often contaminated with other impurities (SO_x, NO_x, and ash). Oxygen present in the flue gas is problematic for conventional amine plants because of oxidative degradation of the amine. Collectively, these factors result in enormous amine circulation rates, large equipment, and large energy requirements. In the case of CO₂ capture from power plants, the heat duty of the amine stripper places a substantial burden on the low-pressure steam supply. Despite the maturity of amine technology, there appears to be ample opportunities for finding improvements.

Flue gas separation at the scale envisioned by the CCP is a new research area. Earlier such carbon dioxide separation technologies were designed to work at much smaller scale to provide dry ice, food-grade carbon dioxide and for chemical processes. The largest plants in operation were too small by a 100-fold compared to the needs in flue gas capture (100 tonne/day versus greater than 10,000 tonne/day).

The Team was charged with evaluation of technologies that might be useful at the large scale needed and with stimulating development of technologies of appropriate scale. There was little R&D in evidence that would lead to significant cost reductions. The Team pursued those opportunities that were identified, and gradually found further opportunities as work progressed. Some high-risk research projects were found to balance the study work that by nature was less adventurous. We sought to reduce the cost of CO₂ capture through:

- Step-change cost reduction by improving existing amine technology (e.g. better solvents, better solvent contactors, cost-effective plant design).
- High-risk, entirely novel approaches to post-combustion capture (e.g. DOE-funded work in self-assembled nanoporous adsorbents).

The CCP funded several engineering studies and technology development programs in the post-combustion area. Work began by inviting proposals from universities and research institutes in Europe and USA. These invitations were directed to researchers thought to be engaged in relevant research for carbon dioxide separation from flue gas. For a number of reasons few proposals materialised. Open invitations at meetings were made without result. Later attempts to stimulate research into high-risk early-stage novel technologies were made through existing networks. This led to two projects. The choice of projects pursued must be seen in the context of the CCP's goal of achieving a 50% cost reduction in retrofits and a 75% cost reduction in new built plants. Technologies without the potential to meet these targets were not considered for CCP support.

ABSORPTION PROCESSES

Absorption–desorption processes can be designed to handle large volumes of gas and have been used in the past to treat flue gas. Consequently, absorption technology was considered to be the default process that would have to be used at this time.

The Base Case

The Team had the responsibility of overseeing development of the so-called base case. This base case is Fluor's Econamine absorption–desorption process that uses a 30% (weight) aqueous solution of monoethanolamine (MEA) plus additives as the absorbent. The technology is commercially available. It was originally developed to recover carbon dioxide from flue gas to produce carbon dioxide for the industrial gases market. Commercial availability of this process does not however mean that the process is mature in

this context. Carbon dioxide sequestration requires processing of huge gas volumes, and represents a new challenge for cost reduction. Engineering studies were made for the CCP “Alaska Gas Injection Facility”, “Norwegian Power Plant”, and “European Refinery” scenarios.

Simplified Engineering Standards for Cost Reduction—Nexant Study

The Team agreed that the base case process should be analysed carefully to uncover all possibilities to reduce costs. The base case design used the oil industry’s API standards that put very stringent demands on reliability and operability that are well beyond those needed for flue gas treatment. The Team believed that there should be significant cost reduction opportunities by re-optimisation with simpler engineering standards. Nexant was contracted to perform this analysis for the CCP independent of Fluor’s process information. Nexant produced a generic MEA process for comparison. The next step in their work was done in two phases, first to reduce costs as much as possible for a stand-alone separation plant and second to exploit possible synergies in integrating the separation and power plants. Their findings are summarised as follows:

- Improved absorber design based on structured packing with a shorter column height and lower pressure drop.
- Elimination of the flue gas cooler by allowing warmer gas into the absorber.
- Smaller flue gas fan due to reduced pressure drop.
- Replacement of the S&T water wash cooler, lean amine cooler and economiser with plate and frame heat exchangers.
- Changing from API to ANSI classified pumps.
- Using live steam from the power plant directly into the desorber, thus reducing reboiler load.
- Going from two parallel to one carbon dioxide compressor.
- Cheaper PSA-based carbon dioxide gas dryer to replace glycol-based absorption.
- Integration of the carbon dioxide recovery plant with the power plant to allow flue gas recirculation into the gas turbines to reduce flue gas volume to be treated.
- Integration of the carbon dioxide recovery plant with the power plant by relocating 75% of the amine reboiling duty directly into the heat recovery steam generator (HRSG).

These recommended process changes will require tests before acceptance, except for simple equipment swaps of pumps and heat exchangers. Some of Nexant’s proposed cost reductions could have a potential impact on unit availability (e.g. single train compression and lower cost pumps). At this point, it is difficult to accurately assess the impact of these changes and evaluation of the overall impact would be required in the next phase of research. Clearly, plant availability will potentially also be reduced when the plants are integrated and thus made more interdependent.

Alignment of costs was needed to ensure that all cost estimates were made on the same basis. The estimates have an accuracy of $\pm 35\%$. The final conclusions regarding the improvements in carbon dioxide separation costs achieved by Nexant, after alignment of content and cost data with the other post-combustion projects, are given in Choi, et al of this volume [1].

Kvaerner–Gore Membrane Contactor Combined with MHI’s Novel KS-1 Absorbent

This project combined two established technologies for the capture of carbon dioxide from industrial flue gas streams—Mitsubishi Heavy Industries’ (MHI) KS-1 hindered amine solvent and Kvaerner Process Technology’s membrane contactor [2,3]. The Team believed that the combination of the two technologies would result in a smaller and less-expensive plant for CO₂ capture. The technology was evaluated in the CCP’s Norwegian Power Plant scenario and required a separate base case to allow comparison. Conceptually, the Kvaerner–Gore membrane contactor replaced the conventional absorption tower and amine wash section.

Experimental results showed that the KS-1 solvent can migrate through the membrane into the flue gas stream. A water-wash section was needed to minimise solvent loss and carryover. This extra section unfortunately added to the overall cost of the facility.

Analysis of the combined process showed that capital cost savings (versus conventional absorber/desorber equipment) are within the uncertainty of the estimates. The Kvaerner membrane contactors' biggest potential advantage is that of lower weight and space requirements. The main advantage of this combination lies in the lower energy consumption (and hence operating cost) required for the KS-1 solvent regeneration. In comparative studies against conventional MEA processes, a reduction in regeneration energy of over 25% is reported for KS-1.

The combined process has not been used commercially, but both the membrane and the solvent have on their own been installed in one commercial facility.

TNO Membrane Absorber

The Dutch research institute, TNO, has developed a membrane absorber similar to that of Kvaerner–Gore that uses polypropylene membranes that are believed to be less expensive [4]. The starting polymer is less expensive. However, this cost advantage will be diluted by the time all necessary equipment is added to make it a working process. The Team believes that the investment cost of the TNO process would be similar to the Kvaerner–Gore process and found no merit for the CCP in pursuing this line of investigation, since step-changes were targeted.

Best Integrated Technology

The Team found that some technologies could be combined in such a way that the combination became better than the best of the individual technologies. It was clear that the MHI's KS-1 absorbent could be combined with the "Nexant low cost" and "Nexant integrated" designs. The lower energy required to regenerate the KS-1 absorbent when compared to MEA will give further energy savings for the two Nexant processes. The best integrated technology (BIT) is defined by the Team as the Nexant low cost and Nexant integrated designs combined with the MHI absorbent. The estimates presented in Chapter 4 are based on the capital investment of the Nexant designs with the operating cost of the Nexant Integrated process reduced by the savings in operating costs allocated to the Kvaerner–MHI design. Since the absorbent circulation rate is lower leading to reduced pumping costs and smaller heat exchangers, there are further savings in this concept, but these are not included in the rough cost estimate.

Time and resources did not allow a deeper analysis but we believe that further savings would accrue from changes to individual equipment items such as:

- eliminating the reclaimer that is needed in MEA operations;
- auxiliary equipment reductions that handle make-up, bleed and solution quality management;
- smaller circulation pumps;
- smaller coolers for lean amine and wash water;
- smaller economiser (lean/rich heat exchanger);
- smaller reboiler for desorber, and thus smaller overhead condenser;
- smaller steam system since less is used.

The BIT as a concept can be further improved as technologies become available. It should be pointed out that BIT, as defined, refers to those technologies included in the final CCP studies. Better absorbents could well be found as pointed out in the "Radical Chemistry" project, and the "Rotating Absorber and Desorber" (RAD) project preliminary study indicated cheaper equipment. Hence, there are already ideas in existence that could further improve the BIT to achieve even lower capture costs for the post-combustion carbon dioxide removal.

ADSORPTION PROCESSES

Novel and existing adsorbent processes were evaluated by the CCP. All alternatives studied were found to be too expensive to be of interest for flue gas carbon dioxide removal. The Team's opinion is that further development of adsorbents for CO₂ sequestration are futile unless a break-through occurs on the process side. The Team attempted, unsuccessfully, to commission an adsorption process analysis to determine what level of improvements was needed to make an adsorption process economically competitive.

Such a far-ranging study would be valuable and could contribute by setting research targets for future adsorption projects.

Electrical Swing Adsorption

The Team worked with Oak Ridge National Laboratory (ORNL) to evaluate their electrical swing adsorption (ESA) process for CO₂ capture. Limited tests were undertaken to assess the loading capacity of a carbon fibre composite molecular sieve (CFCMS) material, to develop adsorption/desorption curves and to assess the benefit of the electrical swing effect. A process scheme was developed from the laboratory scale test results and some preliminary economics were generated for the system. It is clear from the simple cost analysis that there is no economic incentive to take this idea any further for post-combustion separation. The cost is likely to be higher than the base case and there are several technical obstacles still to be overcome (e.g. low carbon dioxide loading and high pressure drop for CFCMS material), which can only result in an even higher cost for a facility based on this technology. Finally, the “Electrical Swing Effect” was not clearly demonstrated during the CCP test program.

Self-Assembled Nanoporous Materials for CO₂ Capture

This project resulted from an active search for radically different approaches to carbon dioxide separation. A partnership was entered into the Stanford Research Institute to design self-assembled, nanoporous adsorbents for improved adsorption of carbon dioxide from low-pressure flue gas streams. The study comprised thermodynamic assessment of cooperative bonding in adsorption processes, modelling cooperative bonding effects with copper dicarboxylate materials, and laboratory performance testing of such materials. The study also included development of a preliminary process design to adsorb carbon dioxide from low-pressure, dilute, flue gas streams.

The theoretical and laboratory results were evaluated based on a simple adsorption cycle (PSA/VSA) concept. Simple cost estimates showed that the approach, while scientifically interesting, would be very expensive, because of large adsorbent beds and significant energy requirements for blowers and vacuum compressors foreseen for the process. Further work in this area is not recommended unless a break-through process design and an adsorbent with higher capacity is found.

MEMBRANE PROCESSES

CO₂ membrane separation processes are based on permeation and are dependent on a CO₂ partial pressure difference across the membrane to drive the process. For flue gas carbon dioxide capture, the feed has a partial pressure of carbon dioxide in the range 0.04–0.12 bar. Efficient separation at economic rates would require higher pressures, and the entire flue gas stream would most likely have to be compressed. It is deemed by the Team that compression costs would make membrane separation processes unsuitable for flue gas carbon dioxide removal. Hence, no action to pursue such processes was taken. At the end of the CCP program, we still stand by this decision. A new approach would be needed to make permeation membrane technology interesting.

CRYOGENICS OR REFRIGERATION-ORIENTED PROCESSES

Cryogenic separation processes are a major workhorse in gas separation processes. However, cryogenic processes need nearly complete water removal before cryogenic temperatures are reached. Carbon dioxide itself would pose a challenge with respect to freezing, but processes have been described that solve this problem. The drying requirements and extensive heat exchange needed in a cryogenic process lead the Team to conclude that such technologies were not worth pursuing. A CO₂ hydrate process was, however, evaluated briefly, but the cooling needs and partial pressure required for hydrate formation make hydrate formation impractical for CO₂ separation from flue gases.

NOVEL PROCESS CONCEPTS

Exhaust Gas Recycle Prior to Amine Separation of CO₂

The resulting increase in carbon dioxide partial pressure after exhaust gas recycle would not increase the rich-amine loading when using MEA. There was thus general consensus within the Team from the start not

to study recycling. The question was later raised again after studying the MHI absorbent KS-1. The effect of exhaust gas recycle was evaluated by Nexant in their absorption process studies. The conclusion drawn was that the ensuing reduction in gas flow to the separation plant makes the concept very interesting, irrespective of the solvent available, because it reduces the gas volume to be processed. It is strongly recommended to pursue this concept in later research.

Rotating Absorber and Desorber Technology

Norsk Hydro offered this technology for consideration by the CCP. A preliminary study showed a potential investment saving of more than 50% relative to the base case as it was then defined. Further savings might accrue if MHI's KS-1 absorbent was used, but this combination was never evaluated. Further work on this line of research is recommended by the Team.

Creative Chemistry Approaches for CO₂ Separation

This project, managed under the Norcap/Klimatek project, produced three novel process concepts as well as new separation chemistry ideas. This project was operated by Norsk Hydro and involved cooperation with Norwegian universities and SINTEF. Formal cost estimates of proposed solutions are impossible at such an early stage of idea development. Assessment of potential was however, made for all ideas based on informed assumptions. These assumptions will become natural targets for later research. The Team found all three concepts interesting and worthy of further investigation since all three were thus assessed to have cost-saving potentials around 50%.

Fast shaking truck (FST)

Three new ideas are incorporated into this exploratory project, including a new direction in separation chemistry. In the proposed concept, the carbon dioxide carrying material ("truck") is expected to be based on transition metal complexes. If necessary, the mass transfer could be made faster by adding a biomimetic compound as a catalyst ("fast"). Finally, it was foreseen that sonic chemistry could be used to help the desorption ("shaking"). A rough flowsheet was prepared and assessed.

pH swing

This process requires a chemical system that allows pH control so that the solution is alkaline when it absorbs carbon dioxide while the desorption is facilitated by lowering the pH to separate the CO₂. A process with absorption and desorption columns is foreseen, but there is a crystalliser added where the pH lowering chemical is precipitated before the solution is recycled to the absorber. Capital investment will not be reduced, but there is potential for substantially lower operating costs. A similar process is used to recover SO₂ from flue gas [4]. Assessment of the process indicates an interesting potential for reducing energy consumption.

Melting point swing (MPS)

This process is based on the use of salt hydrate melts to absorb carbon dioxide, and desorption being effected by solidifying the melt. Such chemicals have been identified with melting points allowing the use of waste heat to operate the cycle [5]. An elegant all-in-one apparatus was conceived.

NEW CO₂ CAPTURE CHEMICALS

MEA as an absorbent has a few undesirable properties. Significant degradation of the amine occurs in commercial processes. It is not quite clear how much of that is thermally induced and much is chemically caused by components in the gas such as NO_x and oxygen. MEA's vapour pressure is high enough to necessitate a water wash step downstream of the absorber. Furthermore, MEA is very reactive with a high heat of absorption, which contributes to the high-energy demands for regeneration. A new absorbent that alleviates one or more of these drawbacks would represent a substantial improvement in CO₂ separation science. The Team has identified three alternative absorbents that are, or could soon be, commercially available, but where testing is not yet complete. They are:

- Mitsubishi's KS-1, which is offered commercially (Mimura et al., 1999) [3].
- University of Regina's PSR [7].
- Praxair's solvent [8].

All have energy consumption about 30% lower than that of MEA and have degradation rates that are lower by factors of 3–10 when compared to MEA. It is clear that the regeneration energy and degradation rates differ significantly between the absorbents. Clearly, this should be an inspiration to widen the field when choosing an absorbent for CO₂ separation. Early in the CCP, a decision was taken to concentrate on reducing investment cost rather than operational costs, where novel chemistry fits in, so little work was done in the field of absorbent improvements. In retrospect, this was unfortunate, because changing absorbents has proved to lower capital investment as well as reducing energy consumption.

ACKNOWLEDGEMENTS

The dedicated effort of the CCP post-combustion separation technology team through the 3 years project has made the work successful. The team members were: Daniel Chinn, Chevron/Texaco (throughout), Dag Eimer, Norsk Hydro (throughout), Odd Furuseth, Statoil (from autumn, 2001), Mike Haines, Shell (until May 2002, replaced by Mariette Knaap), Paul Hurst, BP (from July 2001), Mariette Knaap, Shell (from mid-2002), Svein Lange, Norsk Hydro (only associated member in 2003), Curtis Munson, Chevron/Texaco (until January 2001), Vernon Schievelbein, Texaco (first half year until C/T merger), Mark Simmonds, BP (from 2001 to 2003/1), Mike Slater, BP (replaced by Paul Hurst in mid-2001), Henriette Undrum, Statoil (replaced by Odd Furuseth), Piergiorgio Zappelli, ENI (from 2001), Eivind Aarebrot, Statoil (until 2002).

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