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## GHGT-9

# Membrane-based, enzyme-facilitated, efficient carbon dioxide capture

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#### Abstract

Carbozyme, Inc. has developed a biomimetic technology that promises significant cost and performance advantages over aminescrubbing systems for the capture of CO<sub>2</sub> from combustion flue gases. CO<sub>2</sub> capture requires that the apparatus be able to accept a wide variety of gas streams, generate a stream acceptable to a pipeline operator, and do so at competitive cost and energy bases. A baseline engineering and economic comparison showed the Carbozyme technology to be an improvement over MEA. A predictive model of the Carbozyme permeator system was validated during a preliminary test of a 0.5-m<sup>2</sup> permeator in which 85% removal of CO<sub>2</sub> from a 15.4% CO<sub>2</sub> feed stream was achieved. Upon fabrication, the next-scale permeator will be shipped to the Energy & Environmental Research Center for testing on coal combustion flue gas.

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#### 1. Introduction

The contribution of greenhouse gases (GHG), in particular carbon dioxide (CO<sub>2</sub>), to global warming is well recognized. Hydrocarbon fuels, coal in particular, are an essential part of the fuel market basket and will be well into the future. Today, electricity generation contributes >40% of the United States' CO<sub>2</sub> emissions and coal accounts for 83% of the electric power sector's CO<sub>2</sub> emissions. [1] The pulverized coal (PC) power plant fleet, both existing and greenfields must include CO<sub>2</sub> capture for GHG management. The key to achieving this goal is the development of cost-efficient CO<sub>2</sub> captures technologies with minimal impact on cost of electricity (COE) by imposing the lowest possible parasitic load for the capture process. CO<sub>2</sub> capture – enrichment and compression to pipeline pressure – is

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the single most expensive and energy-intensive step in the carbon capture and storage (CCS) paradigm. The U.S. Department of Energy (DOE) National Energy Technology Laboratory (NETL) details three key targets of the CCS program: 1) the ability to achieve a 90% capture fraction, 2) the ability to deliver a stream of at least 95% purity, and 3) achieving these goals at less than a 10% COE increase for new facilities or less than a 35% COE increase at existing facilities.

Many adsorptive, absorptive, reactive and membrane-based technologies are under investigation each geared to address these goals [2]. At present, the absorptive approaches, especially those featuring amines, are best established. Amine scrubbing is applicable to low-CO<sub>2</sub> partial pressure gas streams and can produce recovery rates of up to 98% with product purity of greater than 99 vol% [3]. However, amine scrubbing also presents some significant challenges, including a substantial energy penalty associated with regeneration of the amine, solvent degradation, equipment corrosion, large footprint requirements and reagent disposal issues.

In contrast to this approach, Carbozyme, Inc. has developed a biomimetic technology. The Carbozyme technology has three key features: 1) a rapid catalyst, CA, 2) a high efficiency mass transfer hollow fiber design, and 3) low energy requirement that does not use high value steam. CA is the fastest, lowest-energy facilitator known for carbon dioxide (CO<sub>2</sub>) [4, 5]. The Carbozyme permeator consists of two hollow fiber, microporous membranes separated by a thin liquid membrane (CLM). The catalyst, CA, is attached to the hollow fiber wall to guarantee that

the incoming CO<sub>2</sub> contacts the CA at the gasliquid interface to maximize conversion efficiency (Fig. 1). The transport resistance of the gases through the microporous membrane is low. Thus, CO<sub>2</sub> faces little diffusion resistance before being converted to bicarbonate at the feed side. CA facilitates CO<sub>2</sub> uptake by rapid conversion to bicarbonate and the liquid film restricts nitrogen and oxygen entry. CA exhibits a high kinetic rate, low thermodynamic energy demand, and high temperature stability, for selected isozymes [6]. It is a "green" chemical, is found in virtually every organism and consumed daily. CA is suitable for both the absorption and desorption stages, enhancing the

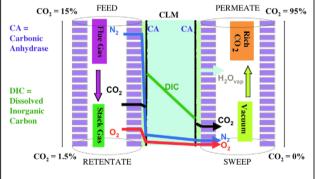


Figure 1. Conceptual diagram for operation of the Carbozyme Permeator.

rate of both processes. The bicarbonate produced by CA is carried in the form of a metal bicarbonate. This design promises significant cost and performance advantages over amine or ammonia-scrubbing systems for the capture of  $CO_2$  from combustion flue gases. Overall, the Carbozyme technology is applicable for moderate temperature (10-75°C) gas flows at low to high pressure with  $CO_2$  concentrations from those found in air to those found in cement plant flue gas (<1% to ~20%) or higher.

### 2. CO<sub>2</sub> Sources and Carbon Capture Targets

#### 2.1. CO2 sources

The Carbozyme  $CO_2$  capture process has been applied to gas streams containing  $CO_2$  at concentrations ranging from that found in breathable air (0.1%) to that found in cement plant stack gas (20%) [7, 8]. The initial development of the process was done at the very low concentrations when the focus was on NASA requirements. The focus was then transferred to capture of  $CO_2$  from flue gas generated at coal-fired power plants (12-15%). Figure 2 shows the projected growth of Greenhouse Gas Emissions as  $CO_2$  equivalents by sector from 2004 to 2030 as given by the IEA [9] as well results of an analysis done at Carbozyme of the impact of electrification of the transport sector. The IEA [9] projects a growth in emissions from all sectors with the power generation sector increasing equivalent  $CO_2$  emissions by 67%. Our analysis was performed by assuming that 90% of road

transportation energy requirements mav be converted from vehicles powered by liquid fossil fuels vehicles powered by electricity from the grid Air, rail, and ship transport would remain unchanged in fuel mix from the IEA reference case. This electrification of the road transport sector results in an increase in electrical power generation requirements and therefore an increase of 130% in GHG emissions from the power generation sector. Our data indicate that not only will size of the power generation fleet grow due to increases in population but that a shift from internal

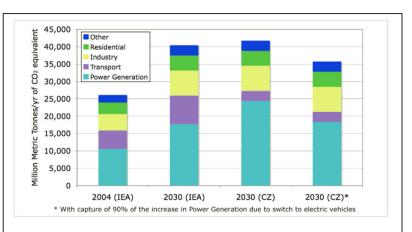


Figure 2. Projected Global CO<sub>2</sub> Emissions by Sector. Electrification of the transportation sector will lead to a reduction in CO<sub>2</sub> emissions from that sector but this decrease will be more than offset by increases in the power generation sector unless CCS is applied to the additional point source emissions. If this is done the result is a net reduction in GHG emissions.

combustion engine to electric powered vehicles will result in a transfer of CO<sub>2</sub> equivalent green house gas (GHG) emissions from transportation (decline by 5360 Mt compared to IEA projections) to power generation (rise by 6708 Mt compared to IEA projections). This is a small additional increase in total potential emissions (1348 Mt). It is 3.3% greater than the IEA total estimate of 40,421 Mt for 2030 but it would provide a potential reduction in emissions of 4689 Mt (11.6% of IEA total estimate for 2030) if 90% of the increased power generation sector potential emissions were captured.

# 2.2. Carbon Capture Targets

The DOE NETL targets for Carbon Capture were noted above. The reasons for these requirements are numerous and varied and represent a mix of technical, economic, and political concerns. For example, the requirement for 90%  $CO_2$  capture is primarily a political one, and might be altered based on economic and technical constraints. In contrast, the requirement for 95%  $CO_2$  purity has a technical basis, which relates to the purity of  $CO_2$  needed to generate a supercritical fluid as these properties are needed for use in enhanced oil recovery and for geologic storage.

The end goals aside, each step in the capture process has its own requirements. For example, under current US EPA regulations there is considerable variance in the flue gas profile. The composition must be managed with an eye towards the acceptance standard for the inlet stream to the CC apparatus. Inasmuch as liquid-based capture methods rely on acid-base chemistry, it is critical to avoid changes in pH of the reaction fluid. In addition, particle load and heavy metal concentrations should be minimized. Other feed stream features include acceptable temperature, pressure, CO<sub>2</sub> concentration, concentration of other major gaseous components, and concentration of contaminants

Feed side acceptance criteria for common flue gas contaminants for the Carbozyme permeator and some details on the reasons for these acceptance limits are given in Jensen et al, 2008 [10]. The conclusion from that work is that  $SO_X$  is controlling contaminant with respect to the need for additional clean up of typical stack gas. Figure 3 illustrates the range of  $SO_X$  concentrations that are typically emitted by existing US power plants. These stack gas  $SO_X$  concentrations range from low values - 300 to 500 ppmv - (for very low sulfur coals conjoined with FGD) to high - above 5000 ppmv - (higher sulfur coals without  $SO_X$  scrubbing). The most conservative acceptance limit for  $SO_X$  ( $SO_2+SO_3$ ) for the Carbozyme permeator is 7 ppmv (18.6 ppmw), a value far below even the low concentration

values for existing power plants. Therefore there is an obvious need for additional flue gas treatment prior to CO<sub>2</sub> capture. The cold ammonia process and the KS-2 process also require SOx values <10ppmv. [11]

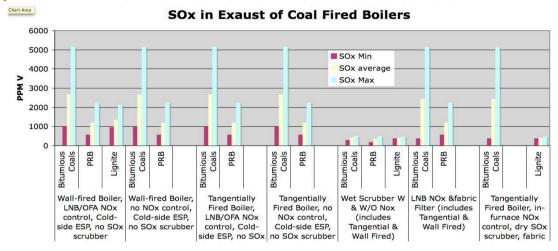


Figure 3. Calculated minimum, maximum and average  $SO_X$  concentrations in exhaust gas from coal fired boilers as a function of the coal, boiler type, and pollution control equipment. All calculations were performed using default conditions of the IECM [12]. Details concerning combinations of coal composition, type of boiler, and pollution control equipment which represent 46.1% of the existing US fleet as of Fall 2007 are available [10].

The first step in determining the most appropriate pretreatment system is to identify the range of combustion flue gas constituents that will most often be encountered. Direct measurements are not feasible because very few power plants will provide detailed compositional data for their flue gases. Instead, we determined that flue gas compositions would have to be estimated for as broad a range of coals and pollution control devices as possible. For this reason, a multistep approach was taken to define the flue gas compositions:

- · The most commonly used coals and their proximate and ultimate analyses were identified.
- The most common plant configurations (i.e., boiler type and attendant pollution control devices) were determined for the entire U.S. power production fleet.
- The composition of the flue gases produced by the most-mined coals when fired in the most common plant configurations was estimated.
- The estimated flue gases were compared to identify a range for each flue gas component that could be expected.
- The expected component ranges were compared with the Carbozyme permeator acceptance criteria to determine
  the cleanup requirements of the pretreatment technology.

The feed-side polishing device that is appropriate for use with the Carbozyme permeator is similar to what others have referred to as a "deep SOx scrubber". To determine the specific type of device that would be needed for the Carbozyme permeator we evaluated the emission composition from about half of the US PC fleet. These values were compared with the Carbozyme stream component acceptance values to allow the CA-based CLM permeator to operate for a minimum of 2500 hours (a conservative value) without need to change enzyme or CLM. More frequent exchange of the CLM or active control of CLM chemistry (e.g., treatment to remove accumulated sulfate) would allow the permeator to accept flue gases containing higher  $SO_x$  concentrations. Addition of EDTA, if needed, to the permeator should permit acceptance of the predicted mercury levels. The flue gas composition data indicated that  $SO_x$  and oxidized mercury levels would likely exceed the acceptance requirements of the CLM permeator, especially for plants with either a dry  $SO_x$  scrubber or without a  $SO_x$  scrubber. The data showed that even the best

fitted plant was still emitting SOx at about 200ppm (vs. 25-fold higher for the worst). A polishing wet scrubber should be able to produce a flue gas stream meeting the liquid membrane permeator's  $SO_x$  requirements.

Currently EERC is preparing a small scale packed column lime scrubber for use in testing of a scaled up model of the Carbozyme permeator. Details on the design of this scrubber are available in [10]. This polishing scrubber was fabricated and installed on the Energy & Environmental Research Center's (EERC) combustion and environmental process simulator (CEPS). Shakedown and testing (late 2008) are expected to validate the polisher's ability to meet the Carbozyme permeator inlet acceptance criteria.

Following CO<sub>2</sub> enrichment the output stream must meet the inlet acceptance criteria of the pipeline carrier. CO<sub>2</sub> product requirements include specific stream composition in addition to compression, drying, and polishing. The most demanding requirement for transport where the amount of moisture, oxygen and certain contaminants with the potential to cause corrosion in steel pipe or would require the use of more expensive pipe materials. Table 1 is a list of the expected Generation 1 Carbozyme permeator performance and a sample pipeline requirement. The final column in Table 1 shows the variance between the anticipated product composition from the Carbozyme permeator before and after compression and drying. Additional polishing will be required to remove the excess oxygen from the purified CO<sub>2</sub> product to meet the pipeline acceptance criteria. A variety of technologies can be employed to remove this oxygen. We anticipate the lowest cost option will be flash vaporization of the O<sub>2</sub> from the product after compression to 8.3MPa (1200 psi) and before final compression to pipeline pressures of 15.2-16.5MPa (2200-2400 psi).

Component	Feed	CZ CO <sub>2</sub> Product Out of Permeator			Pipeline	Variance for CO <sub>2</sub> Product
		Wet	Dry		Requirement	(compare dry CZ product to
	%	%	% volume	% weight	for CO <sub>2</sub>	pipeline requirement)
					Product [13]	
$CO_2$	13.9	43.7	92.4	94.92	95%	0.08% too low
N <sub>2</sub>	72	3.1	6.6	4.3	4%	0.32% too high
$O_2$	5.5	0.39	0.83	0.62	10 ppmw	622 times too high
Ar	0.93	0.07	0.14	0.13		OK
H <sub>2</sub> O	7.7	53.8	0.01	$5.1e^{-5}kg/m^3$	$4.8e^{-4}kg/m^3$	15% of max spec = OK

Table 1. Carbozyme Permeator Product Stream and Pipeline Acceptance Requirements

Over the last several years we have scaled up the design 1,000-fold with no change in performance. This is highly indicative that improvements and development will be linearly scalable. Under current DOE funding we are in a scale-up mode where scale-up involves an increase in both the size of each permeator and in the number of permeators. The tests, which will be run using hot coal flue gas, are central to establishing that the design is fully scaleable within single modules and that multiple modules work harmoniously. Each of these activities will allow progressive scale-up for industrial and power plant operation.

#### 2.3. Performance Testing on Current-scale Permeator

A scalable-design permeator was fabricated and tested. Current results of the shakedown testing indicate that the permeator behavior matched the modeled/expected results. The preliminary test results are summarized in Table 2. Full performance testing is in progress. It will include demonstration of performance under design conditions, optimization of operation to maximize performance, and feedback with respect to design, assembly, and scale-up.

Table 2. Performance Testing on Current-Scale Permeator				
TARGET	OBSERVED			
(DOE CCS goals)	(Shakedown run)			
90% removal of CO <sub>2</sub> from a 15% CO <sub>2</sub> feed	85.3% removal of CO <sub>2</sub> from a 15.4% CO <sub>2</sub> feed			
95% CO <sub>2</sub> dry product gas	81% CO <sub>2</sub> dry product gas (93.6% without argon)			
Flue gas at design load	45.7% of design gas load			

Operating temperature near adiabatic for flue gas	Low operating temperature (prior to temperature control)
Permeate pressure as high as possible	Low permeate pressure (necessary at lower test temperature)
No sweep other than $H_2O$ vapor	Small argon sweep flow (13.3% of dry permeate) used to supplement water vapor sweep (necessary for lower test temperature)

#### 3. COMPETITION

MEA systems have been reported by the Castor Project as requiring 4GJ/t of CO<sub>2</sub> for capture. They anticipate reducing the energy cost to 3.5GJ/t with improved amines and further to 3.2GJ/t with improved mass transfer. Although Le Thiez [14] notes a goal of 2GJ/t, this is assumed to be unrealistic for the moment. These estimations expect the concentrated CO<sub>2</sub> to be discharge from the regenerator at 0.2MPa pressure. Our goal is to compare these energy estimates with ours, in terms of impact on the busbar cost. This is a challenge as the energy consumption of the permeator system is primarily derived from compression operations. Some heat needed by the permeator system to control evaporation of water into the permeate. However, that heat is at such a low temperature that all of it is available from the cooling of the compressor exhaust gas. For this reason it is not included in the evaluation other than to consider the energy necessary to compress the gas to pressure. To put the amine systems on the same basis all of the system costs have been converted to electrical energy. Efficiency assumptions enter into the estimation when converting high value heat to electricity for processes that consume steam power plant energy. To accommodate these differences and to try to be fair to the amine based systems we have assumed very efficient power plants: 40% for now, 45% for the near-future time and 50% in the ultimate system.

Table 3. Energy Use Comparison (GJ/t CO2)

	EU Castor Project [14]			Carbozyme		
	Amine/ Hollow Fiber	Improved Amine	Improved Mass Transfer	Current HFCLM	Near- Future HFCLM	Future HFCLM
Electric energy loss without compression	1.60	1.58	1.60	0.12	0.12	0.11
Electric energy loss for compression to 8.45 MPa including drying	0.30	0.30	0.30	0.57	0.55	0.5
TOTAL Energy Cost	1.90	1.88	1.90	0.69	0.67	0.61
CZ Advantage				2.74	2.80	3.11

The Carbozyme permeator system calculations make no correction for the efficiency of the plant; the compressors are electrically driven and need no conversion. The initial calculations are from a heat and material balance preformed using EPRI case 7C for design conditions. The energy input into the process before compression is a) from a blower to increase the flue gas velocity sufficiently to move the gas though the permeator, and b) from the pump necessary to move the heating fluid and CLM through the permeator. Compressors are used to pressurize the permeate gas from 0.02MPa. Improvements in the design are anticipated, thus capture compression power will be reduced as is shown in Table 5.0.

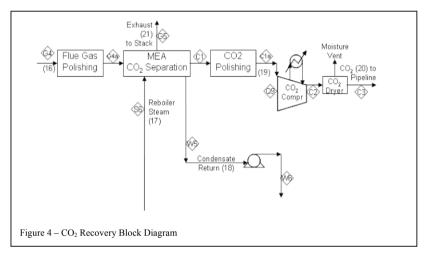
# 3.1. Baseline Cost of CO<sub>2</sub> Capture – MEA-Based Absorption

Evaluation of a novel CO<sub>2</sub>-separation method typically begins with a comparison to the traditional monoethanolamine (MEA) scrubbing process. The purpose of this type of analysis is to establish a baseline set of power plant parameters as a reference method for comparison of CO<sub>2</sub> removal methods. We performed an analysis of this type in order to compare the Carbozyme process with MEA scrubbing. The parameters included in the analysis were thermal performance, capital costs, and operating costs for all major areas of a supercritical pulverized coal (SCPC) steam power plant generating about 500 megawatts of power (the DOE case models).

As part of an ongoing effort to develop fair methods of system evaluation, NETL commissioned a comprehensive comparison of 12 plants representing combinations of fuel type and fuel conversion approach, with and without carbon capture [15]. In this analysis two of those plants are supercritical pulverized-coal (SCPC) plants – one without carbon capture and the other with MEA carbon capture. Those two plants provide the bridge between the 2007 study and the current study of biomimetic carbon capture performed in this project.

The "baseline" effect of MEA CO<sub>2</sub> removal was quantified by comparing its parameters to a corresponding set of parameters for a similar SCPC plant without CO<sub>2</sub> removal. The selected reference plant was a 545-MW supercritical pulverized coal power plant with MEA CO<sub>2</sub> removal. This plant is Case 12 in a published a set of cost and performance baselines for fossil energy plant. [15]

The CO2 recovery block detailed Figure in includes flue gas polishing unit, a  $CO_2$ separation system (MEA, CLM, or other), a CO2 compressor, a CO2 dryer, a CO<sub>2</sub> polishing unit, and a condensate return pump. Inputs to this block are clean flue gas and steam. Outputs from this block are pipeline-quality  $CO_2$ returned condensate, and low-CO2 exhaust gas.



# 3.1.1. Cost of CO<sub>2</sub> Removed and Avoided

Using the method presented in the NETL report [15], the costs of CO<sub>2</sub> removed and CO<sub>2</sub> avoided can be calculated. CO<sub>2</sub> removal and avoidance costs are normally reported as dollars (or euros) per metric tonne, but they are sometimes reported in terms of US tons instead of metric tonnes, or with first-year costs instead of levelized costs. Table 4 shows these variations of CO<sub>2</sub> costs for the reference (MEA) case. Similar calculations for the Carbozyme permeator case will be performed once sufficient data have been collected from scaled up permeators to allow improved estimates from those presented in Table 3.

Table 4 - CO<sub>2</sub> Removal and Avoidance Costs

Cost	Units	Levelized	First-year
CO <sub>2</sub> Removal	\$/tonne (\$/ton)	\$45.00 (\$40.82)	\$41.37 (\$37.53)
CO <sub>2</sub> Avoided	\$/tonne (\$/ton)	\$67.46 (\$61.20)	\$62.02 (\$56.26)

# 4. Conclusions

Carbozyme, under DOE NETL funding, has begun to carry out a systematic analysis of feed gas composition, treatment and requirements for application to its unique enzyme-based, contained liquid membrane permeator. In addition, we have examined the acceptance requirements of the pipeline carrier that would guide the output product. The permeator design uses an efficient catalyst, an efficient mass transfer design and imposes a minimal energy burden on the PC power plant, one that is far less than that imposed by amines. A MEA base case analysis has been developed for direct comparison. Results of shakedown testing of a scalable permeator design indicate that the permeator behavior matches the modeled/expected results.

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