Towards
“Sustainable Carbon Economy”

Feasibility & Technology Options for CO2 Capture from Transportation & Distributed Sources

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Financial support by NASA, DOD, NSF, and GT “Creating Energy Option” Program is acknowledged and appreciated.

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The Need for Sustainable Energy Options

Long term challenges:

- Meet the growing global demand for energy (as fossil fuel reserves become depleted)
- Stabilize atmospheric CO\textsubscript{2} concentration at a “safe” level

Sustainable Future in a carbon-constrained world?

1) Reduce energy consumption (12 TW globally: conservation; efficiency \textbf{but economic growth worldwide}?)

2) Rely on renewable energy (solar; wind; nuclear; carbon-neutral/biofuels \textbf{but accessibility/usability/transportability}?)

3) Sequestration CO\textsubscript{2} (capture & “permanent” storage: oceans, deep earth \textbf{but availability/accessibility/sustainability}?)

Every available options will have to be utilized (no silver bullet)!

What is feasible for mobile, distributed (transportation) applications?
Active Sequestration of CO₂ Emissions

Capture of CO₂ emissions (assume permanent storage capability exists)

Large point sources (e.g. power plants) feature: steady-state, large physical size, economy of scale

→ well covered in literature; area of active research [DOE, industry support]

Small distributed sources (transportation) feature: transient operation, constrained size, convenience, harsh environment

→ neglected in literature; little or no active research [few notable exceptions]
The present carbon-based economy is **unsustainable**!
Electron economy? Hydrogen economy?

Electron Economy
- Solar, Wind, Nuclear
- Fossil Fuel, Coal, Gas
- CO₂ Sequestered

Hydrogen Economy (Does Not Exist)
- Solar, Wind, Nuclear
- Fossil Fuel, Coal, Gas
- Hydrogen (gas)
- CO₂ Sequestered

Review of Proposed Pathways for Mitigation of CO₂ Emissions in the Transportation Sector

+ zero local emissions; CO₂ sequestered centrally; high efficiency
- range, battery lifetime, cost

Hydrogen fueled vehicles
+ zero emissions locally; efficient energy conversion (Fuel Cell)
- onboard storage; distribution infrastructure; commercial viability of FC

Carbon-neutral biofuels (synthetics) [Hoffert (2002)]
+ existing infrastructure (distribution, refueling, IC engines)
- limited capacity/availability; pricing competition from fossil fuels

CO₂ capture from air (downstream) [Lackner, Science (2003)]
+ allows status quo, existing infrastructure; CO₂ sequestered centrally
- lack of sequestration infrastructure; maximize energetic penalty
Enabled by Onboard CO$_2$ Capture and Storage

+ allows status quo, existing infrastructure for refueling/discharging

**LET’S TAKE A DEEPER LOOK...**

- **Is it feasible** (logistics, thermodynamics, size/weight, CO$_2$ liquefaction)?

- **Conceptual Design and Preliminary Analysis** (e.g. Steam reforming with H$_2$ membrane & ATR with O$_2$ membrane)

- **New Perspective** (important!): CO$_2$ capture as **integral to fuel reforming strategy** rather than **added burden**

Sustainable Carbon Economy

CO₂ provides the carbon molecule for liquid energy carriers (analogous to biofuels)!

CO₂ Collected

Energy Conversion (FC w/ CO₂ Capture)

CO₂ Recycled

Energy Conversion (FC w/ CO₂ Capture)

CO₂ Collected

Refueling/Collection

Refueling/Collection

Ground Transportation:
Trucking, Shipping, Trains, etc.

Ground Transportation:
Trucking, Shipping, Trains, etc.

Carbon Economy
(near future)

Carbon Economy
(Sustainable)

Fossil Fuel, Coal, Gas

Distillates (liquid)

Pipeline

Synthetics (liquid)

H₂O

Renewable Energy (Solar, Wind)

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Possible CO₂ Capture Methods

Based on research efforts for sequestration at large-scale point emission sources (power plants) [Bredesen (2004)]

- **Post Combustion Capture in Conventional (Air-Fed) IC Engines**
  - Use existing infrastructure with capture equipment added on
  - Sorbent-based “scrubbers” to remove CO₂ from dilute mixture, requires large (volume), inefficient equipment

  → Not feasible/practical for smaller scale transportation and distributed power generation.

- **Oxy-Fuels** (Combustion/processing in pure O₂)
- **Pre Combustion Capture/Fuel Decarbonization**

  → Call for fuel processing/energy conversion devices which produce “undiluted” CO₂ byproduct stream (most basic requirement!)
Autothermal Reforming with CO₂ Capture

1. Fuel Reforming
2. H₂ Separation
3. O₂ Separation
4. CO₂ Liquifaction
5. CO₂ Storage
6. CO/H₂ “waste” recycling
Steam-Fuel Reforming with CO$_2$ Capture

- Storage Volume (CH$_3$OH:CO$_2$) = 1:1.3 & (CH$_3$OH+H$_2$O:CO$_2$) = 1:1.1
  - Example: 10 Gal. Methanol (57 liters (incl. H$_2$O)) requires 63 liters for storage of CO$_2$

Onboard Storage of Captured CO₂

Energy Penalty for CO$_2$ Liquefaction

State 1

- $T = 25 \, ^\circ C$
- $P = 1 \, bar$
- $C_{CO_2} = 0.04 \, mol / l$

$$dU_{1-2} = \delta Q - \delta W$$

$$W_{1-2} = T \left( S_2 - S_1 \right) - \left( U_2 - U_1 \right)$$

$$W_{1-2} = -30 \, kJ / mol$$

State 2

- $T = 25 \, ^\circ C$
- $P = 100 \, bar$
- $C_{CO_2} = 18.75 \, mol / l$

Compare to energy available in fuel for electrical work (e.g. steam reforming methanol to hydrogen for use in fuel cell)

$$\text{CH}_3\text{OH} + \text{H}_2\text{O} \rightleftharpoons 3\text{H}_2 + \text{CO}_2 \quad \Delta H = 49.4 \, kJ / mol \quad (1)$$

$$\text{H}_2 + \frac{1}{2}\text{O}_2 \rightarrow \text{H}_2\text{O} \quad \Delta G = -228.6 \, kJ / mol \quad (2)$$

- Energy Penalty (%) = $W_{1-2} / (\Delta H + 3\Delta G) = 4.7\%$

CO$_2$ Capture enables recycling of “wasted” fuel products $\rightarrow$ “regenerative” fuel processing
CHAMP Reactor: Regenerative Fuel Processing

Table 1. Composition of Fuel and Exhaust Mixtures

<table>
<thead>
<tr>
<th>Species</th>
<th>Fuel [mol] (w/o recycle)</th>
<th>Exhaust [mol] (w/o recycle)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₃OH</td>
<td>1.0</td>
<td>0.00</td>
</tr>
<tr>
<td>H₂O</td>
<td>1.0</td>
<td>0.035</td>
</tr>
<tr>
<td>H₂</td>
<td>0.0</td>
<td>0.117</td>
</tr>
<tr>
<td>CO</td>
<td>0.0</td>
<td>0.035</td>
</tr>
<tr>
<td>CO₂</td>
<td>0.0</td>
<td>0.965</td>
</tr>
<tr>
<td>H₂ yield</td>
<td>2.848 moles</td>
<td></td>
</tr>
</tbody>
</table>

With recycling

(+) 100% fuel utilization (to H₂)
(-) 15% decrease in specific (per unit volume) throughput

non-ideal ratio

ideal 3/1 ratio
CHAMP Fuel Processing with On-Board CO₂ Capture

**CHAMP** (CO₂/H₂ Active Membrane Piston) fuel processor (compact, scalable, transient, efficient, robust)


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**Diagram**

- **Fuel Cell**
- **Wₑlectric**
- **Air**
- **Cathode**
- **Anode**
- **H₂ Accumulator**
- **H₂ (P = 1.1 atm)**
- **Pure Hydrogen (P = 1.1 atm)**
- **P = 10 bar**
- **CH₃OH + H₂O → 3H₂ + CO₂ + xCO**
- **Pneumatic Piston**
- **Catalyst**
- **Thin Film Heater**
- **Electric heater Input**
- **Fuel mix and recycled product**
- **P = 10+ bar**
- **T = 250°C**
- **Evaporator**
- **Condenser**
- **MeOH/Water**
- **CO₂**
- **Dual Use Tank**
- **CO₂ (liquid) Storage**
- **CO₂ (liquid)**
- **P = 100 bar**
- **T = 25°C**
- **CO₂ + x(H₂ + CO)**
- **P = 10+ bar**
- **P = 10 bar**
- **CO₂ + x(H₂ + CO)**
- **P = 100 bar**
- **Compressio n (x - stage)**
- **Heat Recovery**

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CHAMP (CO2/H2 Active Membrane Piston) Fuel Processor is the key enabling technology!

Summary

- **Sustainable Carbon Economy**: recognize CO$_2$ recycling as integral part of power generation strategy.

- **Feasibility/Practicality established**: CO$_2$ capture with minimal energy penalty & manageable storage volume.

- **Fuel Processing/Energy Conversion identified**: systems that do not dilute CO$_2$ byproduct, take advantage of CO$_2$ capture & enable *regenerative fuel processing*.

- **New reactor designs and system integration strategies** are the key aspects to practical implementation of “sustainable carbon economy”.
Enablers of Distributed Power Generation with CO2 Capture

- **Needs for Research/New Technologies**
  - **Fuel processing systems**: better catalysts (e.g., Yu et al, *J Power Sources* 2005), high surface area catalyst supports (e.g., Fukuhara et al, *Appl. Catalysis A* 2005), new process ideas/reactor designs!!!
  - **Product separation**: H₂ membranes (e.g., McLeod et al, *J. Membrane Science*, 2009), CO₂ membranes (e.g., Huang et al, *J. Membrane Science* 2005), and O₂ membranes (Bouwmeester, *Catal Today* 2003)
  - **CO₂ on-board storage**: lightweight composite tanks for liquefied carbon dioxide storage at elevated pressure
  - **Synthetic renewable liquid fuels**: e.g., photocatalytic F-T synthesis (Ogden at al., *ENIC-2007*), algae, other strategies (Uhrig et al, *Tau Beta Pi Pubs* 2007; Rozovskii et al, *Topics in Catalysis* 2003)