Quantifying Sustainable Development with Sustainable Costs in the Optimization of Chemical Production Complexes with New Plants for Carbon Nanotubes, Carbon Dioxide and Biochemicals

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Introduction

- Industrial processes using biomass and carbon dioxide mitigate global warming

- Objectives are:
  - Identify and design new industrial processes using biomass and carbon dioxide as raw materials
  - Show how these processes are integrated into an existing chemical production complex
  - Obtain the optimal configuration of plants in a chemical production complex using mixed integer nonlinear programming

- Demonstrate that these results are applicable to other chemical production complexes in the world
Base Case of Existing Plants

Plants in the lower Mississippi River Corridor, Base Case. Flow Rates in Million Tons Per Year
Optimal Configuration of Plants in a Chemical Production Complex

► Chemical Complex Analysis System.
  • Determines the optimum structure for a complex
  • Uses the triple bottom line as the objective function
  • Incorporates multi-criteria optimization for Pareto optimal solutions
  • Incorporates Monte Carlo simulation for sensitivity analysis of the optimal solutions

► Triple Bottom Line - economic, environmental, and sustainable costs
  • Economic and environmental cost to the company
  • Sustainable cost to society to repair damage to the environment from emissions within permitted regulations

► Life Cycle Analysis (LCA)
  • Comparative assessment of sustainability using eight impact categories.
  • Evaluates categories based on material and energy balances at stages of the life cycle from collecting raw material from earth and ending when all this material is returned to earth.
Carbon Dioxide as a Raw Material

- Intermediate of fine chemicals for the chemical industry
  - C(O)O-: Acids, esters, lactones
  - O-C(O)O-: Carbonates
  - NC(O)OR-: Carbamio esters
  - NCO: isocyanates
  - N-C(O)-N: Ureas
- Use as a solvent
- Energy rich products
  - CO, CH₃OH

From Creutz and Fujita, 2000
Methodology of Developing New Carbon Dioxide Processes

- Identify potentially new processes
- Simulate with HYSYS
- Estimate utilities required
- Evaluate value added economic analysis
- Select best processes based on value added economics
- Integrate new processes with existing ones to form a superstructure for optimization
New Processes Included in the Complex

<table>
<thead>
<tr>
<th>Product</th>
<th>Synthesis Route</th>
<th>Value Added Profit (cents/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methanol</td>
<td>CO₂ hydrogenation</td>
<td>2.8</td>
</tr>
<tr>
<td>Methanol</td>
<td>CO₂ hydrogenation</td>
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<td>Dimethyl Ether</td>
<td>CO₂ hydrogenation</td>
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<td>Formic Acid</td>
<td>CO₂ hydrogenation</td>
<td>64.9</td>
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<tr>
<td>Acetic Acid</td>
<td>From CH₄ and CO₂</td>
<td>97.9</td>
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<td>Styrene</td>
<td>Ethylbenzene dehydrogenation</td>
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<tr>
<td>Methylamines</td>
<td>From CO₂, H₂, and NH₃</td>
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<td>Graphite</td>
<td>Reduction of CO₂</td>
<td>65.6</td>
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<td>Synthesis Gas</td>
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<tr>
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<td>Propane dehydrogenation with CO₂</td>
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</table>
Application of the Chemical Complex Analysis System to Chemical Complex in the Lower Mississippi River Corridor

- Base case
- Superstructure
- Optimal structure
Processes in the Superstructure

Plants in the Base Case
• Ammonia
• Nitric acid
• Ammonium nitrate
• Urea
• UAN
• Methanol
• Granular triple superphosphate
• MAP and DAP
• Sulfuric acid
• Phosphoric acid
• Acetic acid
• Ethylbenzene
• Styrene

Plants Added to form the Superstructure
• Acetic acid from CO$_2$ and CH$_4$
• Graphite and H$_2$
• Syngas from CO$_2$ and CH$_4$
• Propane dehydrogenation
• Propylene from propane and CO$_2$
• Styrene from ethylbenzene and CO$_2$
• Methanol from CO$_2$ and H$_2$ (4)
• Formic acid
• Methylamines
• Ethanol
• Dimethyl ether
• Electric furnace phosphoric acid
• HCl process for phosphoric acid
• SO$_2$ recovery from gypsum
• S and SO$_2$ recovery from gypsum
Triple Bottom Line

\[ \text{Triplet Bottom Line} = \sum \text{Product Sales} - \sum \text{Raw Material Costs} - \sum \text{Energy Costs} + \sum \text{Sustainable (Credits – Costs)} \]

\[ \text{Triplet Bottom Line} = \text{Profit} - \sum \text{Environmental Costs} + \sum \text{Sustainable (Credits – Costs)} \]
### Some of the Raw Material Costs, Product Prices and Sustainability Cost and Credits

<table>
<thead>
<tr>
<th>Raw Materials</th>
<th>Cost ($/mt)</th>
<th>Sustainable Cost and Credits</th>
<th>Cost/Credit ($/mt)</th>
<th>Products</th>
<th>Price ($/mt)</th>
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<tr>
<td>Natural gas</td>
<td>235</td>
<td>Credit for CO2 consumption</td>
<td>6.50</td>
<td>Ammonia</td>
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<td>Phosphate rock</td>
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<td>Debit for CO2 production</td>
<td>3.25</td>
<td>Methanol</td>
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<td>Credit for HP Steam</td>
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<td>Electro-furnace</td>
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<td>Credit for IP Steam</td>
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<td>Credit for gypsum consumption</td>
<td>5.0</td>
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<td>Debit for gypsum production</td>
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<td>HCl</td>
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<td>Granular triple super phosphate (GTSP)</td>
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<td>MAP and DAP</td>
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<td>Contact process for Sulfuric acid</td>
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<td>Wet process for phosphoric acid</td>
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<td>Ethylbenzene</td>
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<td>Styrene</td>
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<td>Acetic acid</td>
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<td><strong>New Plants in the Optimal Structure</strong></td>
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<td>Graphite</td>
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<td>Hydrogen/synthesis gas</td>
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<td>Propylene from CO₂</td>
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<td>Propylene from propane dehydrogenation</td>
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<td>Methanol (Jun)</td>
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<td>Ethanol</td>
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<td>Dimethyl ether</td>
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<td>Styrene - new method</td>
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<td>Electric furnace process for phosphoric acid</td>
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<td>Haifa process for phosphoric acid</td>
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<td>SO₂ recovery from gypsum waste</td>
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<td>S and SO₂ recovery from gypsum waste</td>
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<tr>
<td>Category</td>
<td>Base Case (million dollars/year)</td>
<td>Optimal Structure (million dollars/year)</td>
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<tr>
<td>-----------------------------------------</td>
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<td>------------------------------------------</td>
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<tr>
<td>Income from Sales</td>
<td>1,277</td>
<td>1,508</td>
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<td>Economic Costs (Raw Materials and Utilities)</td>
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<td>602</td>
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<td>Raw Material Costs</td>
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<td>Utility Costs</td>
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<td>25</td>
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<td>Environmental Cost (67% of Raw Material Cost)</td>
<td>362</td>
<td>385</td>
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<tr>
<td>Sustainable Credits (+)/Costs (-)</td>
<td>-18</td>
<td>-15</td>
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<tr>
<td>Triple Bottom Line</td>
<td>343</td>
<td>506</td>
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</table>
Life Cycle Assessment using TRACI

- Comparative analysis was conducted on the base case and optimal configuration of plants in the chemical production complex using TRACI.
- Scope is “Entry-to-Exit”
  - Classifies resources and releases into various impact categories.
  - Characterization value quantifies the extent of harm stressor can cause.
- Changes from the base case to the optimal configuration:
  - Fossil fuel use increased by 75% from the increase is the energy use of the new plants added to consume excess carbon dioxide being released in the atmosphere.
  - Water use increased by a comparable amount.
  - Global warming category decreased by 66% from base case because new processes consumed carbon dioxide.
  - Small changes in acidification, water use, eutrophication, photochemical smog and human health.
Carbon Nanotubes

- Seamless cylindrical tubes, consisting of carbon atoms arranged in a regular hexagonal structure.
- Consist of carbon filaments with nanoscale (10^{-9} \text{ m}) diameter and micron scale (10^{-6} \text{ m}) length.
- Considered as the ultimate engineering material because of their unique and distinct electronic, mechanical and material characteristics.
- Challenge - production of purified carbon nanotubes in commercial quantities at affordable prices.
  Market price is $100-$400/gm for purified nanotubes.
### Summary of Conceptual Designs of CNT Processes

<table>
<thead>
<tr>
<th></th>
<th>CNT PFR Process</th>
<th>CNT-FBR Process</th>
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<tbody>
<tr>
<td>Catalyst</td>
<td>Fe</td>
<td>Co – Mo</td>
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<tr>
<td></td>
<td>Fe(CO)$_5$ → Fe + 5CO</td>
<td>Silica</td>
</tr>
<tr>
<td>Reactants</td>
<td>CO and Fe(CO)$_5$</td>
<td>CO</td>
</tr>
<tr>
<td>Reactor Type</td>
<td>Plug Flow Reactor</td>
<td>Fluidized Bed</td>
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<tr>
<td>Reactor Conditions</td>
<td>1050 °C @ 450 psi</td>
<td>950 °C @ 150 psi</td>
</tr>
<tr>
<td>Selectivity to CNT</td>
<td>90%</td>
<td>80%</td>
</tr>
<tr>
<td>Purification</td>
<td>- Oxidation</td>
<td>- Leaching</td>
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<tr>
<td></td>
<td>- Acid Treatment</td>
<td>- Froth Flotation</td>
</tr>
<tr>
<td></td>
<td>- Filtration</td>
<td>- Acid Treatment</td>
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<tr>
<td>Production rate (kg/hr)</td>
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</tbody>
</table>
Flow Diagram of CNT-FBR Process
<table>
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<tr>
<th>Economic Analysis</th>
<th>CNT-PFR Process</th>
<th>CNT-FBR Process</th>
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</thead>
<tbody>
<tr>
<td>Total Plant Costs</td>
<td>$4.6 million</td>
<td>$4.4 million</td>
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<tr>
<td>Total Product Costs</td>
<td>$186 million</td>
<td>$124 million</td>
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<tr>
<td>Annual Sales Revenue</td>
<td>$450 million</td>
<td>$450 million</td>
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<tr>
<td>Economic Price</td>
<td>$38/kg</td>
<td>$25/kg</td>
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<tr>
<td>Net Present Value (NPV)</td>
<td>$609 million</td>
<td>$753 million</td>
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<tr>
<td>Rate of Return (ROR)</td>
<td>37.4%</td>
<td>48.2%</td>
</tr>
</tbody>
</table>
Sustainable Chemical Plants using Biomass Feedstocks

Vision

● Convert existing plants to ones that are based on renewable resources requiring nonrenewable resource supplements

● Develop new plants using renewable resources which supply the needed goods and services of the current ones

Essential component of sustainable development

● Embodies the concept that sustainability is a path of continuous improvement, where products and services required by society are delivered with progressively less negative impact upon the Earth

● Consistent with sustainable development is defined as development which meets the needs of the present without sacrificing the ability of the future to meet its needs
Biomass Feedstock

- Virgin Biomass –
  - Specifically grown feedstock for food, animal feed, fuel or chemicals
  - Unit price set by usage as food or animal feed

- Waste Biomass –
  - Typically agricultural residue, municipal solid waste, used cooking oils, fats and grease from animals etc.
  - Used as fuel
  - Unit price set by price of the fuel it replaces

United States can provide about 1.0 billion dry tons of sustainable, collectable biomass and continue to meet food, feed and export demands. Corn led in annual production of grain crops in the U.S. with 330,000 tons per year in 2006.
Biomass Conversion Routes

Common conversion routes for biomass include:

- Fermentation
- Anaerobic digestion
- Transesterification
- Chemical conversion
- Gasification/Pyrolysis
- Liquefaction
Chemicals from Fermentation

- Fermentation is the enzyme-catalyzed transformation of an organic compound to release energy.

- Fermentation feedstock can be starch (corn), sugars (sugarcane) or cellulosic and lignocellulosic biomass (switchgrass, corn stover).

- Fermentation using different enzymes yield different products like succinic acid, butanol etc.

U.S. produced 5.0 billion gallons of ethanol in 2006
Approximately 90% of U.S. ethanol is produced from corn
60% of the world’s biobased ethanol is obtained from sugar cane in Brazil
Ethanol can be the starting material for ethylene which is a major commodity chemical used to produce polyethylene, acetaldehyde etc.
Anaerobic Digestion of Mixed Biomass

Inhibition of the fourth stage of anaerobic digestion produces organic acids

**Hydrolysis** - Complex organic molecules are broken down into simple sugars, amino acids, and fatty acids with the addition of hydroxyl groups.

**Acidogenesis** - Volatile fatty acids (e.g., acetic, propionic, butyric, valeric) are formed along with ammonia, carbon dioxide and hydrogen sulfide.

**Acetogenesis** - Simple molecules from acidogenesis are further digested to produce carbon dioxide, hydrogen and organic acids (mainly acetic acid).

**Methanogenesis** - The organic acids are converted to methane, carbon dioxide and water.

The MixAlco process has a favorable return for producing acetone, diethyl ketone, and dipropyl ketone.
Anaerobic Digestion of Animal Waste

In this process flow diagram, anaerobic digestion of farm waste is used to produce electricity.

The fuel gas (commonly known as biogas) is a mixture of 65% methane and 35% carbon dioxide.
Chemicals from Vegetable oils

Vegetable oils can be directly used as base oil in lubricants or can be converted by transesterification to a wide variety of chemicals.

- Transesterification
- Epoxidation

- Hydroformylation
- Metathesis

- Glycerol
- Fatty Acids
- Surfactants
- Methyl soyate from soybean oil
- Polymers, Resins and Plasticizers
- Base oil in lubricants
- Solvents
- Adhesives

Established Processes for Vegetable oil feedstock

Processes existing in Petroleum feedstocks, need research for Vegetable oil feedstock
Chemicals from Transesterification

Transesterification process is the treatment of vegetable oil with an alcohol, acid or enzyme catalyst to produce esters and glycerol.

If methanol is used, then fatty acid methyl esters (FAME) are produced.

These esters have properties similar to diesel and can be used as fuel (biodiesel) or further transformed to chemicals.
Glycerol or commonly known as glycerin is an byproduct of the transesterification process.

~10% by weight of glycerol is produced as a byproduct in transesterification process.

Glycerin can be used to manufacture numerous commodity chemicals such as propylene glycol.

Bio-PDO (1,3-propanediol) is a proprietary chemical produced from glycerol by DuPont.

New low pressure and temperature (200 psi and 200°C) catalytic process for the hydrogenolysis of glycerol to propylene glycol has been reported.
Chemical Conversion of Biomass to Chemicals – Levulinic Acid

Levulinic acid can be produced from acid treatment of xylose. It is a key intermediate for conversion to commodity chemicals by reduction, oxidation or condensation.
Chemicals from Gasification

● Biomass gasification is the conversion of biomass to synthesis gas (mixture of CO and H₂).

● “Syngas” is a starting point starting material for the manufacture of ammonia, methanol, urea and other important chemicals.

● Gasification can be in absence of oxygen (pyrolysis), in presence of oxygen (partial oxidation) and in presence of steam (steam reforming):

  Pyrolysis: \[ C_{6}H_{10}O_{5} \rightarrow 5CO + 5H_2 + C \]
  Partial oxidation: \[ C_{6}H_{10}O_{5} + O_2 \rightarrow 5CO + CO_2 + 5H_2 \]
  Steam reforming: \[ C_{6}H_{10}O_{5} + H_2O \rightarrow 6CO + 6H_2 \]

● Synthesis gas is used in the chemical production complexes to produce ammonia and methanol

● Nearly 12.2 billion pounds of methanol are produced annually in the USA. Methanol is converted to higher value chemicals such as formaldehyde (37%), methyl tertiary butyl ether (28%) and acetic acid (8%).
Chemicals from Pyrolysis

- Pyrolysis is the direct thermal decomposition of organic components of biomass in absence of oxygen to yield useful solid and liquid products and fuel gases.
- Slow, irreversible pyrolysis (conventional) include processes like carbonization, destructive distillation, dry distillation and retorting.

Chemicals from Thermal Liquefaction

- Thermal liquefaction is the direct chemical conversion of biomass to liquid fuels using a liquid medium (aqueous or non-aqueous).
- Hydriodic acid is used to convert biomass under low temperature and pressure to yield hydrocarbons.
New Processes in the Chemical Complex

- Two carbon nanotube processes – CNT-PFR and CNT FBR

- Biomass processes:
  - Fermentation process for corn to ethanol
  - Fermentation process for waste biomass (sugarcane bagasse) to ethanol
  - Conversion of ethanol to chemicals
  - Fermentation of sugar to succinic acid and derivatives
  - Anaerobic digestion to produce mixed ketones
  - Transesterification to produce FAME and FAEE
  - Conversion of glycerol to propylene glycol and other chemicals
  - Chemical conversion to levulinic acid and derivatives
  - Gasification to produce synthesis gas
  - Conversion of synthesis gas to ammonia, methanol and urea
  - Thermal liquefaction to produce hydrocarbons

- The optimum configuration of plants will be determined based on economic, environmental and sustainable costs using multicriteria optimization and Monte Carlo simulation