***THE CARNOL PROCESS SYSTEM
FOR CO₂ MITIGATION AND METHANOL PRODUCTION***

Meyer Steinberg

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Engineering Research and Applications Division

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Abstract

The feasibility of an alternative CO₂ mitigation system and a methanol production process is investigated. The Carnol system has three components: (1) a coal fired power plant supplying flue gas CO₂ to, (2) the Carnol process which converts the CO₂ with hydrogen from natural gas to methanol which is used, (3) as a fuel component in the automotive sector. For the methanol production process alone, up to 100% CO₂ emission reduction can be achieved while for the entire system up to 65% CO₂ emission reduction can be obtained. The Carnol system is technically feasible and economically competitive with alternative CO₂ disposal systems for coal fired power plants. The Carnol process is estimated to be economically attractive compared to the current market price of methanol, especially if credit can be taken for the carbon as a marketable coproduct.

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The Carnol Process System for CO₂ Mitigation and Methanol Production

Introduction

The evidence for greenhouse gas CO₂ warming causing global climate change is continuing to mount, and international agreements are being sought to limit CO₂ emissions (Refs. 1 and 2). The CO₂ emissions are primarily due to fossil fuel combustion in the commercial, industrial and transportation sectors. In the U.S. about one-third of the CO₂ comes from the industrial sector mainly from central power stations which are largely fueled by coal. About an equal amount of CO₂ is emitted in each of the other two sectors. In this paper we describe and develop a system, the Carnol System, which converts CO₂ from coal-fired power plant stack gases with natural gas to produce methanol as a liquid fuel for use in automotive engines. Carbon is produced as a coproduct which is a storable commodity product. The carbon from the coal is used twice and, therefore, the CO₂ is greatly reduced compared to conventional system.

The Carnol System Configuration Development

The basis for the Carnol system depends on integration of the following four developments.

1. A significant amount of effort and improvement has gone into the removal of CO₂ from the stack gas of fossil fuel burning plants particularly for recovery and disposal of CO₂ in the ocean (Refs. 3 and 4). In the Carnol configuration it is proposed to utilize the CO₂ rather than dispose and sequester the CO₂.
2. For purposes of providing the hydrogen to reduce the CO₂ to methanol, it is proposed to thermally decompose methane (from abundant natural gas) to hydrogen and carbon. In order to produce hydrogen without generation of CO₂ it is proposed not to burn the carbon produced, but to use it as a materials commodity (Ref. 5). An alternative hydrogen production process without net CO₂ generation is the gasification of biomass (wood) (Ref. 6).
3. The synthesis of methanol has been significantly improved by using liquid phase slurry catalysts (Refs. 7 and 8). It is proposed to react the CO₂ saturated monoethanolamine (MEA) solvent used in the recovery of CO₂ from coal-fired power plant stack gases to produce the methanol.
4. It is proposed to use the methanol produced in the Carnol system as an alternative fuel in internal combustion automotive engines in the transportation sector. It is recognized that in order to make an impact in CO₂ reduction, a large market for the product methanol must be available. The automotive fuel market provides that possibility. Furthermore, it is shown that methanol is 30% more efficient in IC engines than

conventional gasoline, thus providing further incentives for reducing CO₂ emissions (Ref. 9).

Figure 1 shows a schematic of the overall Carnot configuration for achieving the CO₂ emission mitigation in the industrial and transportation sectors. Each of the process operations are discussed and further developed as follows.

Hydrogen Production with Zero CO₂ Generation

An investigation (Ref. 5) of the basic energy requirements and CO₂ emission for various methods of hydrogen production has indicated that methane decomposition can produce hydrogen with the least CO₂ emission and with the least basic energy requirements (Ref. 5). This evaluation has been extended to include steam reforming of coal and biomass in Table 1. The latter shows that only thermal decomposition of methane and the steam reforming of biomass can achieve zero CO₂ emission. Of these two, the process energy requirement is the least for methane decomposition, however, the total energy requirement per unit of hydrogen is less for biomass gasification. This is so because in the thermal decomposition of methane, the carbon is not used to achieve zero CO₂ emission. In biomass gasification, zero net CO₂ emission is achieved because solar photosynthesis removes CO₂ from the atmosphere. Although the present conventional method for hydrogen production, by steam reforming of methane, has a higher energy efficiency than the thermal decomposition of methane, it generates considerable greater amounts of CO₂. Thus, thermal decomposition of methane competes with biomass gasification. From an economic point of view, natural gas, at present, is less expensive than biomass. However, a balance of not using the carbon and a lower thermal efficiency compensates somewhat for the higher cost of biomass from energy farms. Since the gasification of biomass is much better developed than the thermal decomposition of natural gas, the latter is chosen for further development.

Alternate methane decomposition reactor (MDR) designs have been considered. However, it appears that a molten metal bath which has been proposed for waste processing (Ref. 10) has a number of advantages for a MDR design. For example, bubbling methane through a bath of a low melting metal, such as tin, heated to temperatures in the order of 800-1000° C could decompose over 90% of the methane. The carbon could be trapped and separated from the top of the molten metal bath due to the great density difference between carbon (1.8 gm/cc) and tin (7.3 gm/cc). The partial pressure of tin at 1000° C is less than 10⁻⁶ atm so that there would be no loss of tin or contamination of the hydrogen gas stream with tin. The low viscosity of tin would allow good gas/liquid contact resulting in good heat transfer between the tin and the methane gas. A reactor and process design is shown on the right hand side of Figure 2 which is based on the tin MDR reactor operating at 900° C and 7.5 atm and heat and mass balances for producing a 90% H₂ - 10% CH₄ stream which after pressure swing absorption (PSA) produces a 100% H₂ stream. If the hydrogen is used to produce methanol, the 90% H₂ - 10% CH₄ stream can be used directly and the PSA operation can be eliminated.

The Synthesis of Methanol with Zero CO₂ Emission

The hydrogen produced by methane decomposition is used to react with CO₂ recovered from coal fired power plant.

If one mole of CO₂ is removed from the stack gases to produce 1 mole methanol, then when, the methanol is combusted as fuel, one mole of CO₂ is produced. Therefore, the net generation of CO₂ for production of methanol is zero.

Using MEA solvent gases, it is reported that it requires at least 25% of the capacity of the power plant to remove and recover CO₂ from the stack gases of the power plant (Ref. 11). With improvement in absorber packing, the pressure drop for feeding flue gas through the absorber using a hindered amine solvent the fraction loss of power to recover 90% of the CO₂ from a coal burning plant is reduced to 12% (Ref. 4). About 92% of the energy is needed in the stripper portion of the recovery system which is obtained from the low pressure side of the turbine and reduces the net power output of the plant. However, if the CO₂ is reacted with hydrogen, methanol is produced in an exothermic reaction and that energy can be used to strip out the CO₂ from the MEA and therefore it becomes unnecessary to take steam from the power plant for this purpose. Furthermore, if a liquid phase slurry catalyst system is used with an MEA solvent, the heat of reaction resulting from the synthesis of methanol is sufficient to distill out the methanol from the MEA solvent. The synthesis of methanol has an exothermic heat of reaction of 33 Kcal/gm mol CO₂ (or MeOH), which is more than enough to provide the heat of vaporization of methanol which is only 9 Kcal/mol.

Another important point is that the equilibrium concentration of methanol produced by the reaction of CO₂ with H₂ is favored at lower temperatures. Equilibrium calculation indicates that the concentration of methanol at 120° C is 5.8 and 3.4 times that at 260° C for pressure of 30 atm and 50 atm, respectively. The higher equilibrium methanol concentration results in a lower recycle ratio around the methanol synthesis steps and a higher thermal efficiency. The equilibrium partial pressure of CO₂ can be obtained from the phase equilibrium diagram for the amine H₂O - CO₂ given by Suda, et al. (Ref. 4). The liquid phase methanol slurry catalysts are under investigation by several investigators (Ref. 7). The liquid phase synthesis is favored because its heat transfer characteristics are better than the gas phase heat transfer synthesis.

The Integrated Carnol Process System

The methane decomposition molten metal reactor process can be integrated with the liquid phase synthesis of methanol, using CO₂ removed from a fossil fuel fired power plant. Figure 2 shows the integrated Carnol process. A computer simulation model of the entire process has been developed to obtain material and energy balances over a range of MDR and methanol synthesis reactor (MSR) conditions. A summary of one of the computer runs of the process including the performance parameters are shown in the summary of Table 2. The

thermal efficiency reaches up to 50% while the total efficiency for both carbon and methanol is up to 90%. The CO₂ emission is reduced by 83% compared to that of the conventional methanol process based on steam reforming of natural gas.

Preliminary Economics

A preliminary estimate of the methanol production cost and CO₂ credit is summarized in Table 3 for various production cost factors including natural gas and CO₂ cost. The estimate is based on 90% recovery and utilization of CO₂ (feed) from a 600 MW(e) coal fired power plant. The capital investment is based on an equivalent current steam reforming plant estimated at \$100,000 per daily ton of methanol. Labor, maintenance and utilities are lumped into the fixed charge of 25% of the capital investment. At a natural gas cost of \$2/MSCF, \$10/ton storage of carbon with no income from carbon and if methanol is sold at the historic cost of \$0.45/gal (in the U.S.), the cost of reducing CO₂ by this Carnol process amounts to \$25/ton CO₂. At \$3/MSCF natural gas, the cost of reducing CO₂ increases to \$55/ton. This range of CO₂ emission reduction credit is less than the maximum cost estimated for sequestering CO₂ in the ocean, ~\$60/ton (Ref. 11) not counting the transport of liquid CO₂ from beyond 10 Km inland from the ocean. If the carbon can be sold for \$58 to \$120/ton, the methanol can be produced for \$0.45/gal. However, recently due to the mandated use of methanol for MTBE oxygenation of gasoline, the price of methanol has increased to as high as \$1.30/gal and has then dropped back to about \$0.70/gal. At these prices, the Carnol process can easily meet these market costs, even when charging for CO₂ from the power plant at a rate of as high as \$108/ton. The general conclusion is that the Carnol process is potentially competitive with conventional methanol prices while significantly reducing CO₂ emissions.

Comparison of CO₂ Emission for the Carnol Process and for the Entire Carnol System

Since there are other competitive processes for methanol production using alternative feedstocks, a comparison of CO₂ emissions must be made between processes. Table 4 makes such a comparison. Taking the current conventional process for methanol production by steam reforming of natural gas as the base case, the Carnol process and the BCL biomass steam gasification process are compared (Ref. 6). Biomass is a feedstock of choice because it removes CO₂ from the atmosphere by photosynthesis. The Carnol process obtains an 83% reduction in CO₂ emission with a penalty of only 24% in production capacity of methanol when methane is used to heat the methane decomposition reactor. The CO₂ emission can be eliminated in the Carnol process when hydrogen is used to heat the reactor, in which case the production capacity is reduced by 29%. The steam gasification of biomass process also eliminates CO₂ emission, however, a further reduction in methanol production to 35% is obtained.

While the Carnol process emission reduction can be compared on a methanol production basis as referred to above, it is necessary to estimate the CO₂ emission for the entire system which includes (1) the coal-fired power plant, (2) the Carnol methanol production process and (3) the use of methanol as an automotive IC engine fuel, as shown in Figure 1. The total CO₂ emission for the system, must be based consistently on the HHV of the various feedstock energy

inputs to the system including (a) electricity from the power plant, (b) liquid methanol fuel from the Carnol plant and (c) mechanical energy from the automotive IC engines. This was done in Table 7 for 4 systems: 1) current conventional coal fired power plant, and gasoline driven automobiles; 2) coal fired power plant with conventional natural gas steam reforming for methanol production and methanol fueled vehicles; 3) coal-fired power plant with Carnol process for methanol production and use of methanol fueled vehicles, and 4) coal-fired power plant with biomass gasification methanol production and with methanol fueled vehicles. Several assumptions must be made for a valid comparison.

- The oil refinery produces gasoline at 90% efficiency; the equivalent of 10% C is lost as CO₂.
- The thermal efficiency for steam reforming of methane to methanol is 64%.
- The Carnol thermal efficiency is 90%.
- The net CO₂ emission based on biomass gasification is zero.
- The methanol fueled vehicle is 30% more efficient than gasoline driven vehicle (Ref. 9).

The results shown in Table 5 indicate that the CO₂ emission reduction is reduced by 18% using conventional steam reforming of natural gas for methanol production and its use in vehicles. The CO₂ is reduced by 63% for the Carnol methanol process and 65% reduction when biomass gasification is used. The latter two thus achieves significant reductions in CO₂ emission for the entire system. From an economic point of view, the biomass feedstock is currently more expensive than natural gas in the U.S. and, therefore, for about equal CO₂ emission reduction, the Carnol natural gas process presently should be more economical.

Conclusions

The Carnol process system configuration appears to be technically feasible and economically viable both as a CO₂ mitigation method for coal-fired power plants and as a methanol production process compared to the conventional method especially when considering credit for marketing the carbon coproduct. For process improvement, the development of a molten metal methane decomposition reactor and a CO₂ liquid phase catalytic slurry methanol synthesis reactor are recommended.

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Figure 1

CARNOL System Configuration For CO₂ Emission Mitigation

Figure 2

Carnol VI Process for CO₂ Mitigation Technology

Combining CO₂ Recovery From Power Plants With Liquid Metal Methane Decomposition and Liquid Phase Methanol Synthesis

TABLE 1
HYDROGEN PRODUCTION
 Comparison of Basic Energy Requirements and CO₂ Emission

PROCESS	ENERGY EFFICIENCY		PROCESS FUEL	FEEDSTOCK	RATIO mol H ₂ - mol CH ₄	ENERGY-Kcal/mol H ₂		CO ₂ EMISSION mol CO ₂ mol H ₂
	Process Energy	Overall Fuel				Process	Total	
1. METHANE THERMAL DECOMPOSITION $\text{CH}_4 = \text{C} + 2\text{H}_2$ COMBINED CYCLE POWER FOR ELECTRICAL HEATING	(A) 80% (t)	53.3% (t)	H ₂	CH ₄	1.67	11.3	126.9	0.00
	(B) 80% (t)	58.1% (t)	CH ₄	CH ₄	1.81	11.3	117.1	0.05
	(C) 80% (t)	64.2% (t)	C	CH ₄	2.00	11.3	106.0	0.12
	(D) 55% (t)							
	80% (t)							
	44% (t)	53.9% (t)	CH ₄	CH ₄	1.68	11.3	126.2	0.10
2. METHANE STEAM REFORMING $\text{CH}_4 + 2\text{H}_2\text{O} = \text{CO}_2 + 4\text{H}_2$	80% (t)	94.6% (t)	CH ₄	CH ₄ + H ₂ O	2.95	18.8	71.9	0.34
3. COAL STEAM REFORMING $\text{CO}_{0.8}\text{O}_{0.08} + 1.92\text{H}_2\text{O} = \text{CO}_2 + 2.32\text{H}_2$	80% (t)	92.4% (t)	COAL	COAL + H ₂ O	1.45 H ₂ /Coal	21.9	73.5	0.69
4. BIOMASS STEAM REFORMING $\text{CH}_{1.6}\text{O}_{0.6} + 1.4\text{H}_2\text{O} = \text{CO}_2 + 2.15\text{H}_2$	80% (t)	92.0% (t)	BIOMASS	BIOMASS + H ₂ O	1.54 H ₂ /Biomass	20.3	73.9	0.00

Table 2
Carnol Process Design Parameters
Mass and Energy Balances

UNIT	PROCESS DATA
<u>MDR</u>	
Pressure, atm	7.5
Temperature, °C	900
<u>MSR</u>	
Pressure, atm	30
Temperature, °C	120
<u>PERFORMANCE</u>	
Ratio, Methanol/CH ₄ , kg/kg	1.19
Ratio Carbon/Methanol, kg/kg	0.57
Thermal Efficiency of MeOH,	49.7%
Total Thermal Efficiency, C + MeOH	89.7%
CO ₂ Emissions, lb CO ₂ /MMBTU	30.2
kg CO ₂ /GJ	13.0
CO ₂ Emission Reduction Compared to Conventional Steam Reforming Process	83.4%

Table 3

PRELIMINARY CARNOL PROCESS ECONOMICS
Costs shown in \$10⁶/yr and (Unit Costs)

Investable Capital Cost (IC) = \$8.46 x 10⁸*

PRODUCTION COST FACTORS = INCOME FACTORS

0.25 IC \$10 yr	Natural Gas		C Storage		CO ₂ Cost		C Income		MeOH Income		CO ₂ Credit	
	\$10 ⁶ yr	$\left(\frac{\$}{MSCF} \right)$	\$10 ⁶ yr	$\left(\frac{\$}{ton} \right)$	\$10 ⁶ yr	$\left(\frac{\$}{ton} \right)$	\$10 ⁶ yr	$\left(\frac{\$}{ton} \right)$	\$10 ⁶ yr	$\left(\frac{\$}{gal} \right)$	\$10 ⁶ yr	$\left(\frac{\$}{ton} \right)$
2.12	2.60	(\$2)	0.19	(10)	0	(0)	0	(0)	3.78	(0.45)	-1.10	(-25)
2.12	3.90	(\$3)	0.19	(10)	0	(0)	0	(0)	3.78	(0.45)	-2.40	(-55)
2.12	2.60	(\$2)	0.19	(10)	0	(0)	1.10	(58)	3.78	(0.45)	0	(0)
2.12	3.90	(\$3)	0.19	(10)	0	(0)	2.18	(126)	3.78	(0.45)	0	(0)
2.12	2.60	(\$2)	0.19	(10)	0.42	(10)	0	(0)	3.78	(0.45)	-1.53	(-35)
2.12	3.90	(\$3)	0.19	(10)	0.84	(20)	3.25	(170)	3.78	(0.45)	0	(0)
2.12	3.90	(\$3)	0.19	(10)	0.23	(5)	0	(0)	10.95	(1.30)	+4.47	+103
2.12	3.90	(\$3)	0.19	(10)	0.23	(5)	0	(0)	6.48	(0.77)	0	(0)
2.12	3.90	(\$3)	0.19	(10)	4.69	(108)	0	(0)	10.95	(1.30)	0	(0)

*Based on the following plant capacities

- 1) CO₂ rate, 90% recovered from a 600 MW(e) net (650 MW(e)) gross coal fired power plant = 4.34 x 10⁶ T/Yr
- 2) CH₄ rate = 2.77 x 10⁶ T/Yr = 400,000 MSCF/D
- 3) MeOH produced = 2.78 x 10⁶ T/Yr = 61,100 Bbl/D. = 8460 T/D
- 4) Carbon produced = 1.91 x 10⁶ T/Yr = 5800 T/D

Table 4

Methanol Production and CO₂ Emission Process Comparison

PROCESS	PRODUCTION CAPACITY		CO ₂ EMISSION	
	<u>Moles MeOH</u> Moles Feedstock	% Reduction from Conventional	<u>lbs CO₂</u> MMBTU	% Reduction from Conventional
Conventional Process Steam Reforming of CH ₄	0.76	0%	180	0%
Carnol Process from CH ₄ (and CO ₂)	0.58 ⁽¹⁾	24%	30	83%
	0.54 ⁽²⁾	29%	0	100%
Steam Gasification of Biomass	0.50	35%	0	100%

⁽¹⁾ Heating methane decomposition reactor with CH₄.

⁽²⁾ Heating methane decomposition reactor with H₂.

TABLE 5
CO₂ EMISSION FROM SYSTEM OF COAL FIRED POWER AND LIQUID HC FUELED AUTOMOTIVE POWER

Emission Units in Lbs/MMBTU (HHV)
(Multiply by 0.43 to Convert to Kg/GJ)

SYSTEM	UNIT CO ₂ EMISSION	CO ₂ EMISSION FROM COMBINED SYSTEM	% CO ₂ EMISSION REDUCTION FROM CURRENT SYSTEM
COAL FIRED POWER PLANT	215	205	0%
Gasoline ¹ Fueled Automotive IC Engine (Current System)	196		
COAL FIRED POWER PLANT	215	169	18%
Natural Gas to Methanol ² by Conventional Reforming and with Methanol Fueled Automotive IC Engine ⁵	139		
COAL FIRED POWER PLANT	215	76	63%
Carnol MeOH Plant (Methane Decomp) ³ Plant with Methanol Fueled Automotive IC Engine ⁵	117		
COAL FIRED POWER PLANT	215	71	65%
Biomass MeOH ⁴ (Gasification) Plant with Methanol Fueled Automotive IC Engine ⁵	106		

Assumptions

¹ Oil refinery is 90% efficient for gasoline production - 10% lost to CO₂.

² Thermal efficiency is 64% for conversion of natural gas to MeOH - Conventional steam reforming plant.

³ Carnol plant for methanol is 90% efficient with respect to CO₂ obtained from coal-fired power plant.

⁴ Net CO₂ emission from biomass gasification to methanol is zero due to photosynthesis.

⁵ MeOH fueled IC automotive engine is 30% more efficient than gasoline driven IC engine.