A Zero CO_2-Emitting Process for Transportation Fuels from Coal and Natural Gas Resources

Jin-Ping Lim[1], Daniel Steele[1], Diego del Rio Diaz-Jara[1], Donald J. Eckstrom[1], Robert B. Wilson[1], Stephen Niksa[2] and Ripudaman Malhotra[1],*

[1]*SRI International, Menlo Park, CA*
[2]*Niksa Energy Associates, LLC, Belmont, CA*

Abstract: Simultaneous steam reforming of natural gas and conversion of coal to syngas was achieved in a gas-blown, electrically heated, entrained-flow gasifier. Conversions > 95% for both coal and natural gas were achieved at temperatures above 1450°C with a product molar ratio of H_2:CO equal to 2. Fully validated kinetic models were used to extrapolate these results to commercially relevant gasifier conditions, and an Aspen Plus process model was developed for producing diesel fuel from the syngas via methanol. If CO_2-free electricity is used to drive gasification, producing hydrocarbon fuels entails no CO_2 emission or consumption of water. Process economics analysis predicts diesel fuel can be produced for $2.81/gallon with electricity at $100/MWh. The capital cost for a plant producing 100,000 barrels/day of diesel fuel is estimated at $3.18 billion or $31,800/daily barrel of capacity, less than half of the estimated $70,000/daily barrel for a coal-to-liquids plant based on Fisher-Tropsch synthesis.

Keywords: Coal-to-liquids, gasification, methanol, natural gas, steam-reforming

---

1 Introduction

Production of liquid fuels from domestic resources is important for enhanced energy security, particularly in petroleum-importing countries. Liquid fuels from petroleum furnish over 90% of transportation energy needs, which are vital for all aspects of the economy. As recently as 2011, the United States (US) consumed on average 18.8 million barrels (bbl) of petroleum each day, while domestic production was only 7.8 million barrels per day (bpd); the shortfall being made up by importing 11 million bpd [1]. Even after allowing for the 3 million bpd of finished petroleum products, the net imports amounted to 43% of the consumption.

US petroleum reserves are estimated at 30.5 billion barrels, which corresponds to 1.2 cubic miles of oil (CMO) [2]. For reference, global reserves of petroleum are 53 CMO, global consumption of petroleum is about 1.1 CMO/yr, and US consumption is about 0.29 CMO/yr. At the current rate of domestic production, the 1.2 CMO of petroleum reserves will be exhausted in less than 15 years, and despite recent discoveries, such as Bakken shale oil (an estimated resource volume of 0.7 to 0.9 CMO), domestic petroleum production cannot keep up with increasing demand, and for long-term energy security alternate methods for producing liquid hydrocarbons will be needed.

Like the US, many countries such as Russia, China, India, Australia, and South Africa face similar shortages of domestic petroleum supplies, which they need for liquid fuels. However, many of them have abundant coal and natural gas—particularly shale gas, for which recovery has been greatly improved by horizontal drilling and hydraulic fracturing—resources and a coal-and-gas-to-liquids (CGTL) technology could greatly improve their energy security. US coal reserves are estimated at 107 billion tons of bituminous and anthracite coals and 129 billion tons of low-rank coals [1]. Together, they are equivalent to 35 CMO of energy. Global coal reserves are estimated at 120 CMO, whereas the annual consumption is 0.8 CMO. The US also has excess natural gas supplies. Its reserves are estimated at 273 trillion cubic feet (tcf), and an additional 862 tcf are deemed technically extractable from shale gas resources. Combined, they are equivalent to 7.4 CMO. Global shale gas resources are estimated at 5760 tcf, or 37.6 CMO. As domestic coal and natural gas supplies far exceed those of oil, it would greatly improve the energy security of the country if these resources could be used for producing transportation fuels.

Conversion of coal-to-liquid fuels (CTL) has been known since the 1920s. Conventional Fisher-Tropsch synthesis (FTS) was first commercialized in Germany in 1936. During World War II about 25% of automobile fuel in Germany was produced using FTS process. More recently, South Africa adopted FTS process to produce liquid fuel during the years of the Apartheid. Currently, Sasol operates the largest FTS facilities in Secunda, South Africa with a capacity of 160,000 barrels per day (bpd). Despite this history, CTL is not widely practiced because of a number of factors, including: (i) high capital cost; (ii) high CO_2 emissions; and (iii) high water consumption. The FTS process is capital intensive with estimated capital costs in excess of \$70,000/daily bbl [3]. The large environmental footprint in terms of CO_2 emissions and water consumption also render the process unsustainable: producing 1 kg of hydrocarbon fuels by FTS requires 5 kg of water and produces 4.5 kg of CO_2.

In this paper, we describe a scalable and environmentally "no worse than petroleum" CTL process that is economical and environmentally benign. Specific attributes of the process include: (i) zero CO_2 emissions; (ii) zero consumption of water; (iii) production cost of diesel less than \$3.00/gal; and (iv) capital cost of \$32,000 per daily barrel. We achieved these targets with the process depicted in Figure 1, wherein coal and natural gas are simultaneously converted into syngas; the syngas
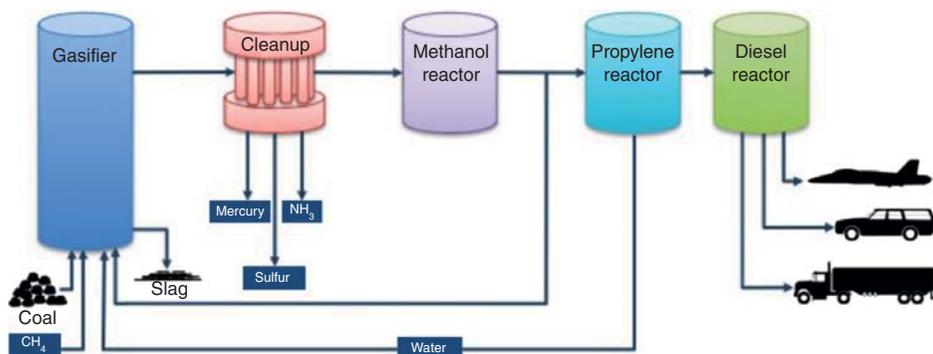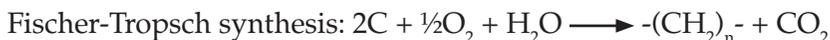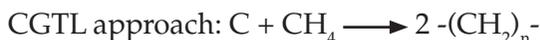
Figure 1 Overall process diagram for simultaneous coal gasification and steam reforming, followed by production of hydrocarbon fuels via methanol.

is converted to methanol, which is then used to produce transportation fuels. We refer to this coal-and-gas-to-liquids process as the CGTL process. The advantage of the CGTL scheme over the conventional FTS process is immediately apparent when we consider the overall equations:

$$\text{CGTL approach: } C + CH_4 \longrightarrow 2\text{ -}(CH_2)_n\text{-}$$

$$\text{Fischer-Tropsch synthesis: } 2C + \tfrac{1}{2}O_2 + H_2O \longrightarrow \text{-}(CH_2)_n\text{-} + CO_2$$

In the FTS process, for every carbon atom of coal that is converted into liquid product, at least one carbon atom is converted into CO$_2$. Often, to drive the endothermic processes additional carbon is burned in the reactor with the result that, for every carbon in the liquid fuel, 1.5 to 2 carbons end up as CO$_2$. The hydrogen required for the conversion into liquid fuels is provided by water, and hence it entails consumption of water. In contrast, the CGTL process uses natural gas as the source of hydrogen. As can be seen in the overall equation, no CO$_2$ is emitted and the yield of liquid product from *solid* phase carbon is at least four times that relative to the FTS process, because for every coal carbon, this process also converts one carbon from gaseous methane into liquid. This process intensification is key to the vastly reduced capital requirement. Use of natural gas instead of water as the primary source of hydrogen also reduces the overall energy requirement for the CGTL process [4], and makes feasible the use of electrically heated gasifiers. By simultaneously steam reforming natural gas and gasifying coal, we achieve a CO:H$_2$ ratio of 1:2, which is required for methanol production. The water that we use in the gasifier is regenerated in the methanol-to-olefin conversion step and is recycled.

Several other researchers have also recognized that controlling the CO:H$_2$ ratio is crucial to reducing the CO$_2$ emissions associated with a CTL approach. One

method for increasing the hydrogen ratio is catalytic dehydrogenation of FTS light ends [5]. Hydrogen gas is recycled, and the resulting solid carbon product is recovered for sale. Two other groups have proposed the use of coal gasification and methane steam reforming in parallel to achieve the desired CO/H$_2$ ratio without relying on a water-gas shift reaction [6, 7].

Although the co-reforming of methane and coal in a gasifier with steam as described in this paper has not been previously demonstrated, researchers in China are investigating a related approach. For example, Cao *et al* [8] described experiments on gasification of coals of three ranks in the presence of steam and CH$_4$ and provided the justification for the basic innovation of our effort. However, their work was conducted at atmospheric pressure in a fluidized bed reactor with oxygen or air feed to supply the energy through partial oxidation. Experiments showed that char particles catalyzed the conversion of methane to syngas and, thus, the degree of conversion depended on the coal suspension loading in the reaction zone.

Zhou *et al* further elaborated on the role of coal char as a catalyst in methane reforming by steam and/or CO$_2$ [9]. This study, also conducted in a fluidized bed reactor at 1 atm pressure, shows that the dominant variable in methane conversion is temperature, with conversion rising rapidly above 800°C. Several other papers from China, US, and Australia have also investigated the catalytic activity of coal char and coal ash for the steam reforming of methane although none achieve the operating temperatures we targeted [10–16].

Use of an electrically heated gasifier reduces the carbon-footprint of the process. Since the energy requirements for the gasifier are reduced by the use of methane, it becomes more feasible to consider using electricity to drive the process. In essence, the proposed CGTL process can be considered as a means of storing green-electricity as liquid hydrocarbon fuels. As discussed below, the well-to-wheels carbon footprint of fuels from this process is lower than of petroleum-derived fuels. This is particularly true if carbon-neutral electricity is available, but also when natural gas is used in a combined cycle process to produce electricity. The process intensification afforded by this scheme reduces the gasifier size and thereby the capital expenses. Furthermore, the process would not create any CO$_2$ emissions in the production of fuel, nor would it consume any water.

In view of these attractive features of this scheme, we undertook a laboratory proof-of-principle demonstration of co-gasifying coal and natural gas with steam in an electrically heated gasifier, which is the unique and therefore the highest-risk aspect of the process. We were concerned that the presence of hydrogen might adversely affect the kinetics of steam gasification of the coal as has been reported [17].

2   Experimental Section

The laboratory work was conducted in SRI's high-pressure entrained-flow gasifier that captures all products and produces reliable kinetic data [17–20]. In this
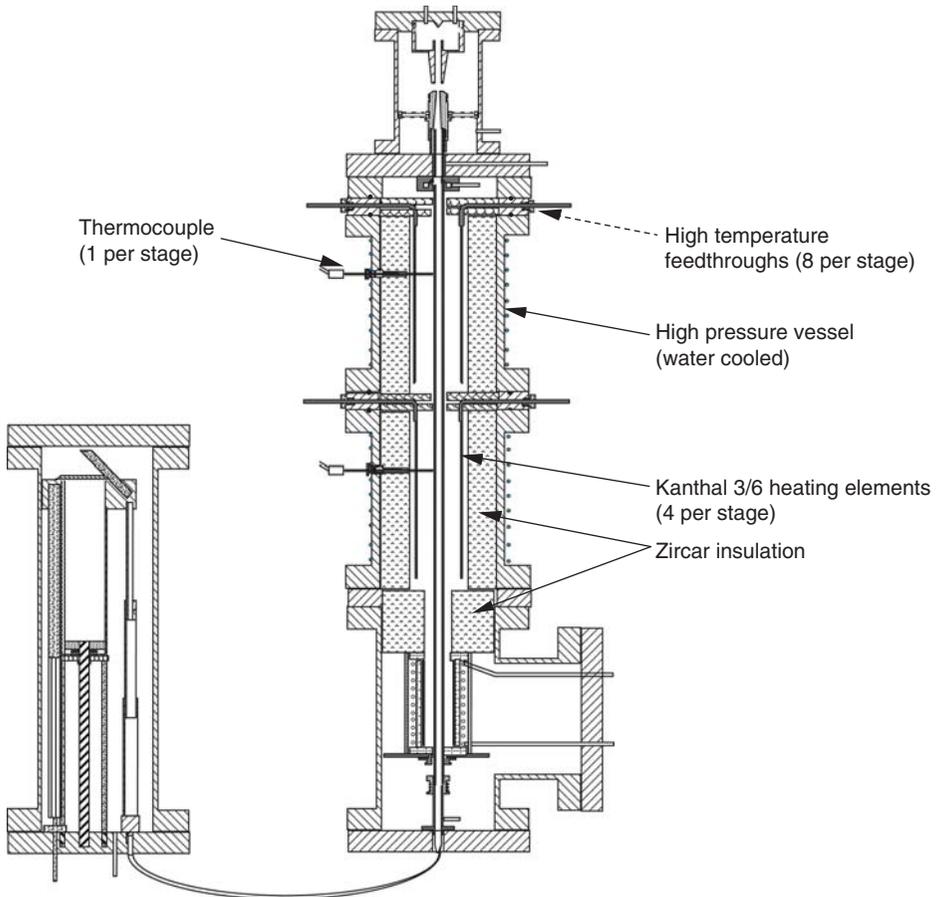
Figure 2 Schematic of SRI's pressurized radiant coal flow reactor (p-RCFR).

system, coal particles pass upward through a 134-cm mullite tube that passes through an injector section consisting of an inductively heated graphite furnace (17 cm) followed by up to three furnaces (30 cm ea.) that are resistively-heated with Kanthal heaters. Figure 2 shows the schematic of the apparatus with only two of the three Kanthal-heated stages. The thermal and fluid dynamic characteristics of this reactor have been well characterized, and we have used it successfully to build simulation models for various coal utilization schemes [21–26].

The test coal used in this study was Powder River Basin (PRB) (DECS-26) sub-bituminous. The coal samples were ground, sieved, and aerodynamically sized to between 75 and 105 μm. Coal properties are listed in Table 1.

In a typical run, the coal loading was between 3 and 6% by weight of the total gas flow, and the test pressure was 3.0 MPa (30 atm) for a nominal residence time

Table 1 Proximate and ultimate analysis of the subbituminous, DECS 26 used in this study.

Proximate Analysis (air dried)	
Moisture	0.1
Volatile matter	39.4
Ash	5.0
Fixed Carbon	55.5
Ultimate Analysis (dry ash-free basis)	
Carbon	73.7
Hydrogen	5.6
Oxygen	19.0
Nitrogen	1.1
Sulfur	0.6

of 1.5 seconds. The Kanthal furnace temperature was varied between 1100°C to 1600°C, and the injector temperature was typically 50°C to 100°C higher to bring the gas and particle temperatures up to the Kanthal temperatures at the interface between injector and downstream furnaces. The gas feed consisted of a mixture of methane (0–10%), steam (10–30%), hydrogen (0–40%), and carbon monoxide (0–15%), with argon being the balance. In most cases, the methane to coal ratio (by carbon) is maintained at 40:60. In order to prevent problems with sooting of methane, we developed a test procedure wherein the Ar, H_2, and H_2O flows were established, and all reactor conditions were stable before introducing methane and coal.

To demonstrate the feasibility of this approach with higher coal loadings, we built a new injector with a heating shell to vaporize water into steam and to heat the gas stream to simulate the temperature and composition of a recycle stream. The injector also included a water-cooled inner tube through which dry coal could be fed without having it soften and clog up the system before entering the gasifier. With this injector, we conducted tests with coal loadings as high as 20 wt%.

3 Experimental Results

3.1 Effect of Temperature

Temperature is the single most critical factor for achieving high conversion of both methane and coal. We performed two series of tests to determine the effect

of temperature on methane and char conversions. The first series of tests was per-
formed at temperatures between 1200°C and 1500°C. As expected, our findings
demonstrated that char conversion increased with increasing temperature. The
second series of tests was performed controlling other experimental variables,
such as the gas environment of the entrained coal. For this series of experiments,
the coal and methane ratio was fixed to maintain the 60:40 carbon ratio required to
produce the correct 2:1 H$_2$:CO ratio used by the downstream process. The amount
of steam was adjusted to give a steam-to-carbon ratio of 1.8. We used a hydrogen
concentration of 20% to limit sooting to within an acceptable level. We used argon
to make up the balance of the gas feed. Test conditions have been the same for all
experiments with the main reactor temperature between 1400–1500°C, an operat-
ing pressure of 30 atm, and a suspension loading of 6.2%.

The coal and methane conversions are shown in Figure 3. Coal conversion
increased from 85% at 1400°C to 92% at 1500°C. We also observed a higher meth-
ane conversion at the higher temperatures. Methane conversion went up from 35%
at 1400°C to 65% at 1500°C.

The distribution of carbon in the feed and products of these tests is shown in
Figure 4. As listed in Figure 4, the observed carbon closures were good, ranging
between 87% and 100%. It is worth mentioning that the CO to CO$_2$ ratio increases
with temperature from 5.3 (at 1400°C) to 7.2 when operating at 1500°C, while keep-
ing the amount of water constant. This result suggests that at higher temperatures
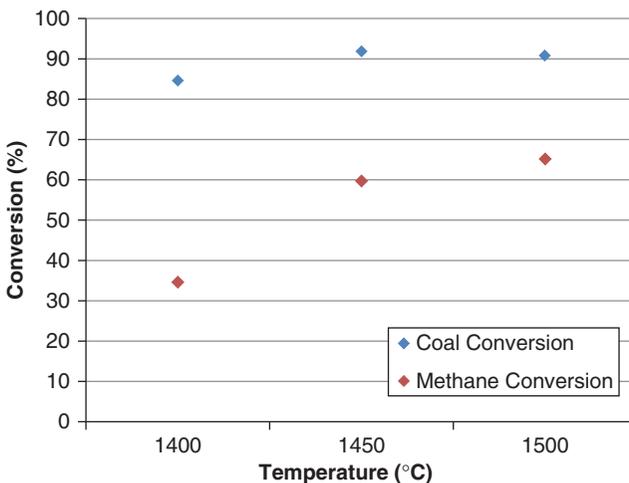we could operate with higher water concentrations without risking production of
too much CO$_2$.

Figure 3 Temperature effects on steam gasification of PRB coal at 20% H$_2$ and 1.8 steam-to-
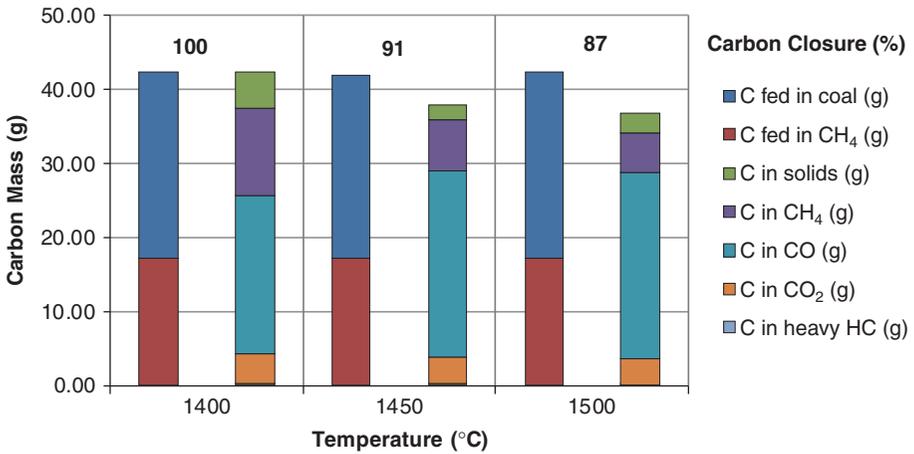carbon ratio; 6.2% suspension loading and P = 30 atm.

Figure 4 Temperature effects of carbon distribution in feed (left bar) and product (right bar) streams of gasification of PRB coal at 20% H$_2$ and 1.8 steam-to-carbon ratio; 6.2% suspension loading and P = 30 atm.
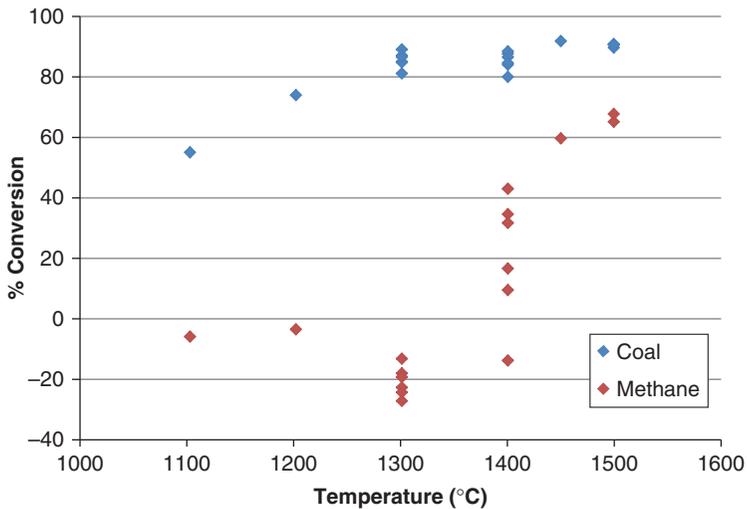

Figure 5 Temperature effect on conversion of coal and methane.

The importance of temperature with regard to achieving high overall conversions can be seen upon examination of several experiments with similar reaction conditions (Figure 5). Coal conversion increases monotonically with temperature, while methane conversion is observed to be highly dependent on reaction environment. There appears to be a temperature threshold for the conversion of

methane – in order to effect conversion rates high enough to consume both the fed methane and that produced by the gasification of coal, the reactor temperature must exceed 1400°C. In this figure, the negative values of methane conversion indicate that the amount of methane produced during coal gasification exceeded the amount of methane consumed during the steam reforming reaction.

3.2 Effect of Hydrogen

In preliminary scouting experiments, we observed that in the absence of hydrogen there was excessive sooting of methane to preclude completing a successful run. The sooting was kept to a minimum with the addition of about 20% hydrogen. Hydrogen has also been reported to inhibit char gasification under certain conditions [27], and thus while we may want to include hydrogen in the gasifier stream to minimize sooting, it could have the adverse effect of slowing down char gasification.

We conducted a series of experiments at different gasifier temperatures to investigate the effect of hydrogen on solids recovery (expressed as a percentage of carbon recovered as char plus soot relative to the carbon fed as coal and methane) in which PRB coal was injected into the pressurized reactor (30 atm) along with steam (25%), methane (5%), and a variable amount of H$_2$ (from 0 to 30%) using argon as the balance. The coal to methane ratio in these experiments was 1:1 on carbon basis and suspension loadings between 2.4 and 3.6 wt% depending on the amount of argon. Runs were conducted with the Kanthal-heated gasifier sections at 1200°C and 1400°C.

The solids recoveries for these series are presented in Table 2. Increasing hydrogen concentration helps to reduce the amount of solid carbon monotonically at

Table 2 Solids recovery and methane conversion as a function of hydrogen concentration at 1200 and 1400°C.

% Hydrogen	Solids Recovery (%)		Methane Conversion (%)	
	1200°C	1400°C	1200°C	1400°C
0	22.6	14.9	29	66
5	22.3		−6	60
10	15.9	7.2	−15	
20	16.6	7.5	−16	44
30	12.5	6.3	−20	43

[a] Solids recovery is the percentage of carbon in char and soot product relative to the total carbon fed as coal and methane.
[b] Total pressure was 3.0 MPa; Gas stream consisted of steam (30%), methane (25%); hydrogen as indicated and argon was the balance; coal:methane feed was 1:1 on carbon basis.

both these temperatures. There was no reversal, and thus any reduction in char conversion due to the presence of hydrogen was more than offset by inhibition of soot formation. While addition of hydrogen did not have an adverse effect on solids conversion, it negatively impacted methane conversion, particularly at 1200°C. Indeed, with increasing hydrogen, the methane conversion at 1200°C dropped from 20% to -29%, meaning methane was produced in those runs. At 1400°C, the methane conversions fell from 66% in the absence of hydrogen to 43% when 30% hydrogen was included in the gas stream.

The results show a trend toward higher coal and methane conversions at higher temperatures. Accordingly, we conducted another series of tests at 1500°C. This time, we increased the coal to methane ratio to 0.6:0.4 as required for a 1:2 CO:H_2 ratio of the product syngas. The suspension loading was also increased, and it varied from 4.8 wt% to 6.2 wt% depending on the argon content. The amount of steam was fixed during these experiments to give a steam-to-carbon ratio of 1.8.

The observed coal and methane conversions are displayed in Figures 6 and the distributions of carbon in the feed and product streams are shown in Figure 7, along with the carbon closures. The results show that char conversion goes up from 90% to around 95% with decreasing hydrogen concentration. The effect of hydrogen on methane conversion is more marked, increasing from 68% at 20% hydrogen concentration to 86% at 5% hydrogen concentration. Under these conditions, a total conversion (coal and methane combined) greater than 90% is observed. Diminishing the amount of hydrogen also has an effect on the water-gas shift reaction and the resulting CO to CO_2 ratio. As expected, the ratio diminishes when lowering the hydrogen concentration, and it varies from 7.2 at 20% hydrogen to 5.6 when no hydrogen is
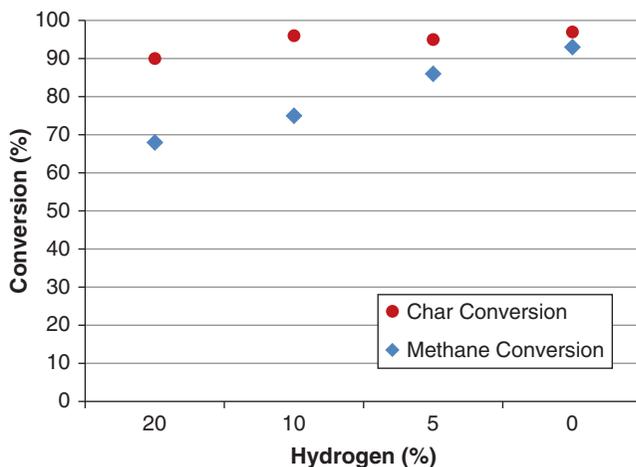

Figure 6 Hydrogen concentration effects on steam gasification of PRB coal at 1500°C and 1.8:1 steam-to-carbon ratio.
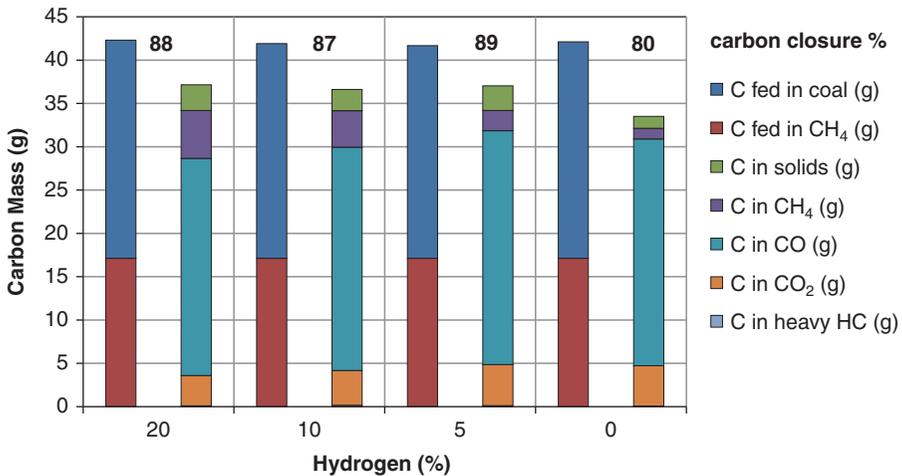
Figure 7 Hydrogen concentration effects on carbon distribution in feed (left bar) and product (right bar) streams on steam gasification of PRB coal at 1500°C and 1.8:1 steam-to-carbon ratio.

present. This amount of CO$_2$ presents no particular problem for the catalytic conversion of syngas to methanol, which is the next step in our scheme.

Finally, diminishing the amount of hydrogen present during the reaction increases the amount of soot formed. At 5% hydrogen concentration, as much as 10% of the methane is converted into soot. Also, at hydrogen concentrations of 10% or less, soot deposits were seen in the mullite tube. Although the deposits are far from being a concern at these intermediate solid loadings, they could be of concern at higher solid loadings.

To test the ability to feed high loadings of coal entrained with methane, steam and recycled hot gas from the methanol synthesis unit, we built a new injector with a heating shell to vaporize water into steam and to heat the gas stream to simulate the temperature and composition of a recycle stream. The injector also included a water-cooled inner tube through which dry coal could be fed without having it soften and clog up the system before entering the gasifier. With this injector, we were able to feed coal loadings as high as 20% and achieved coal conversions in the 90% range when operating at 1500°C.

4   Process Simulation

4.1   *Extrapolation to Industrially Relevant Conditions*

There are three important differences between the operating conditions evaluated in the lab-scale tests and commercial processing: (1) Much larger coal suspension loadings must be imposed at commercial scale; (2) The reactor pressures are much higher at 50 to 70 atm; and (3) The feed stream cannot be diluted with inert gases.

In addition, the residence times through the gasifier will probably be longer than the 1.4 s imposed in the lab-scale tests, because commercial entrained coal gasifiers typically operate with residence times of 2.5 to 4 s. Since such conditions could not be tested directly in the laboratory, we used simulations to estimate conversions and product distributions across a broad domain of conditions. We have developed and validated an approach to predict conversions in industrial-scale operations based on lab-scale experiments for several coal-conversion technologies [24–26]. We begin by interpreting the laboratory data quantitatively using fully validated reaction mechanisms, detailed operating conditions, and computational fluid dynamic (CFD) codes to extract the rate parameters that can be used to estimate conversions under commercially relevant gasifier conditions.

Fully validated reaction mechanisms for each chemical process were used to address important technical questions and, ultimately, to quantitatively interpret lab-scale gasification tests. Primary devolatilization was described with Niksa Energy Associates FLASHCHAIN®, and the distributions of volatile products were adjusted for instantaneous hydrogenation. Consequently, volatiles contained an abundance of oxygenated gases and gaseous hydrocarbons but no products heavier than oils. Char gasification by steam with inhibition by H$_2$ and CO was described with CBK/G—a mechanism previously validated for applications at elevated pressures in complex syngas mixtures. Reforming chemistry in the gas phase was described with 566 reactions among 154 species as heavy as phenol that were assembled from literature submechanisms, which were extensively validated for applications at reducing conditions under elevated pressures.

Due to the emphasis on comprehensive chemistry, these mechanisms were implemented in simulations that suppressed all aspects of turbulent fluid mechanics and convective transport phenomena. Series of continuous stirred tank reactors (CSTRs) were used to coarsely resolve the time evolution of the reforming chemistry in nominal plug flow along the test reactor. The first reactors in the series were fed with increments of the hydrogenated volatiles. Since char conversion is much slower than devolatilization, the last reactors in the series were fed with only char conversion products. Intermediate reactors were fed with both types of products. Finite-rate kinetics for both devolatilization and char gasification determined the incremental additions to each CSTR in the series. This simulation strategy gave converged solutions for nearly every case in the test matrix within 15 min or less.

4.2 Process Model

The predicted composition of the gas exiting the gasifier forms the starting point of a large model that incorporates the entire CGTL process that we assembled in Aspen Plus to estimate the mass and energy flows in a plant capable of producing 100,000 barrels per day of diesel fuel. In order to reflect reality as closely as possible, process-operating parameters for each unit operation in the process were based on our best understanding of commercial operations. To acquire this information,

we utilized publically available sources such as patents, company releases, and Department of Energy (DOE) reports. We supplemented these sources by purchasing the more in-depth information provided by several SRI Consulting (SRIC) Process Economic Program reports. The process conditions, conversion rates, capital and operating costs, and scaling factors gained from these sources were utilized in the design of the process model.

The output from the Aspen model is a summary of the mass and energy balances around each unit. In this process, we have three primary inputs to the plant: coal, natural gas, and electricity. The only product is diesel fuel. From the model, we calculated that for every gallon of diesel produced, the plant would consume 7.7 lbs of coal, and 94 scfs (or 4 lbs) of natural gas, and 17.7 kWh of electrical energy. In terms of energy, the material inputs correspond to 19.1 kWh of coal and 27.6 kWh of natural gas. Since a gallon of diesel is equivalent to 43.3 kWh, the net efficiency of the plant is 67%. We then used the model to generate the capital and production cost. We used these balances to generate the capital and process cost estimates.

We used published performance and cost data for the different components to estimate the cost of our system. Where multiple systems were available, we chose the ones that have been deployed at large scales and for which reliable performance data were available. For gas cleanup, we used data for the Selexol process from National Energy Technology Laboratory (NETL) reports [28, 29]; for syngas to methanol, we used data from SRIC reports based on a Lurgi Mega-Methanol plant [30]. We chose Lurgi's technology for methanol-to-propylene conversion, and UOP (a Honeywell company) technology for propylene to hydrocarbon fuels. The capital costs for the different units of the CGTL process are shown in Table 3.

Table 3 Capital cost breakdown for a CGTL plant ($ per daily barrel of diesel).

	Current SRI Estimate	Current State-of-the-Art Estimate
Gasification	$13,200	$29,890
Air Separation	0	6,790
Syngas Cleanup	3,400	3,220
Methanol Synthesis	2,130	
Olefin Synthesis	3,160	
Oligomerization	1,780	
Syngas to diesel	7,070	7,630
Product Upgrading	820	2,730
Offsite	7,300	19,740
Capital Cost Total:	**$31,790**	**$70,000**

Table 4 Breakdown of cost for producing diesel from coal and natural gas.

		Current SRI Estimate	Current State-of-the-Art Estimate[1]
Capacity	BPD	100,000	20,000
Total Fixed Capital (TFC)	$ Millions	3,179	1,867
Production Costs			
Raw Materials	$/gal	0.49	0.73
Utilities	$/gal	1.77	0.04
Labor Cost	$/gal	0.07	0.14
Maintenance Materials, Operating Supplies	$/gal	0.10	0.12
Plant Overhead	$/gal	0.05	0.11
Taxes Insurance (2%/yr of TFC)	$/gal	0.04	0.14
Depreciation (10%/yr of TFC)	$/gal	0.21	0.68
G&A, Sales, Research	$/gal	0.09	0.19
Net Production Cost	**$/gal**	**$2.81**	**$2.14**

[1] Reference [30].

For comparison, the table also lists the corresponding costs for a conventional FTS process.

The breakdown of the production cost is shown in Table 4. For a baseline case, we priced the PRB coal at $14/ton, and natural gas at $4.81/MMBtu. The total production cost also includes capital depreciation at 10%/yr of total fixed capital. This depreciation adds 0.21 ¢/gal to the cost of the finished fuel. The largest component of the production cost is the price paid for the electricity. In order that the process be truly zero CO_2, the electricity must not be produced from a fossil fuel. For that reason, we used a premium price of 10¢/kWh for electricity. It is noteworthy that if the restriction of zero CO_2 emissions is relaxed, we could reduce the production cost to about $1.60/gallon with the electricity from the grid. This action does not affect the plant layout, and therefore will not increase the capital cost.

5 Carbon Footprint

We also performed a "well-to-wheel" analysis of the fuel produced by our process to assess its "greenness [31]." We used the Argonne National Laboratory's GREET Model to make this assessment. Table 5 compares CO_2 emissions from the use of

Table 5 Well-to-wheel comparison of diesel from the CGTL process and its variants with those of conventional diesel and a coal-based FT product.

Conventional Diesel	389	Conventional diesel from crude oil	Argonne National Labs GREET[a] 1.8d.1
Coal-Based Fischer-Tropsch	830	Diesel from coal using a Fischer-Tropsch process	R. Smith, *et al*, + GREET 1.8d.1
SRI Process (Carbon-Neutral Electricity)	**326**	**SRI's product from coal and natural gas using the SRI approach and carbon-neutral electricity**	**SRI Estimate + GREET 1.8d.1**
SRI Process (NGCC Electricity)	536	SRI's product from coal and natural gas using the SRI approach and electricity from a natural gas fired power plant	SRI Estimate + GREET 1.8d.1
SRI Process + Biogas (Carbon-Neutral Electricity)	190	SRI's product from coal and biogas using the SRI approach and carbon-neutral electricity	SRI Estimate + GREET 1.8d.1

[a] GREET - Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation Model

fuel from the CGTL process with those from normal petroleum-based diesel and a conventional FTS diesel. For this comparison, we assumed hydrocarbon fuels to have the same characteristics as the diesel fuel. From an environmental standpoint, diesel from the SRI process is somewhat better than petroleum diesel and emits about half as much CO_2/mile as conventional FTS-diesel. In this analysis, we have assumed the availability of CO_2-neutral electricity to drive the process. However, if we use a more realistic scenario of electricity being produced by a natural gas combined-cycle turbine, the SRI process produces about 38% more CO_2 per mile than petroleum diesel.

The carbon footprint of this scheme could be substantially reduced by blending in bio-derived carbons. A recent study by RAND Corporation concluded that FTS fuels derived from coal and biomass mixtures offer the most promising near-term option for meeting the Department of Defense's needs for clean and affordable

fuels [32]. The blending of biosourced carbon in the CGTL process can be achieved by either using biomass (or biochar) along with coal, or by using biogas from anaerobic digesters instead of natural gas. If we use biogas, a carbon-neutral source of methane, the resulting CGTL diesel would emit less than half the amount of CO$_2$ as petroleum diesel. Thus, the diesel produced using this process with biogas as the source of methane would qualify under the revised Renewable Fuels Standard (RFS 2) of the Energy Independence and Security Act of 2007 (EISA).

6   Summary and Conclusions

We have described here an environmentally benign process for producing liquid hydrocarbon fuels from coal and natural gas via methanol. The conversion process does not consume water and does not emit CO$_2$. A critical step in this process is simultaneous steam reforming of methane and gasification of coal in an electrically heated, gas-blown, entrained-flow gasifier, and we provided proof-of-principle for this step in laboratory tests. Based on the conversion data from laboratory tests, we estimate the capital cost for such a plant to be less than half that of a conventional FTS-based CTL plant and, even using a relatively expensive electricity price, the projected production cost of diesel fuel is less than $3.00/gallon.

Acknowledgment

References

1. US Energy Information Administration, Annual Energy Outlook, http://www.eia.gov/forecasts/aeo/ (2011).
2. H.D. Crane, E.M. Kinderman, and R. Malhotra, *A Cubic Mile of Oil*, Oxford University Press, New York, NY (2010).
3. R. Smith, M. Asaro, and S. Naqvi, Fuels of the future: technology intelligence for coal to liquids strategies, Process Economics Program, SRI Consulting, (2008).
4. D. Hildebrandt, D. Glasser, B. Hausberger, B. Patel, and B.J. Glasser, Converting coal into liquid fuels with less work. *Science* **27**, 1680–1681 (2009).
5. G. Huffman, Incorporation of catalytic dehydrogenation into fischer-tropsch synthesis of liquid fuels from coal to minimize carbon dioxide emissions. *Fuel* **90**, 2671–2676 (2011).
6. T. Adams and P. Barton, Combining coal gasification and natural gas reforming for efficient polygeneration. *Fuel Process. Technol.* **92**, 639–655 (2011).
7. M. Sudiro and A. Bertucco, 2009. Production of synthetic gasoline and diesel fuel by alternative processes using natural gas and coal: process simulation and optimization. *Energy* **34**, 2206–2214 (2009).

8. Y. Cao, Z. Gao, J. Jin, H. Zhou, M. Cohron, H. Zhao, H. Liu, and W. Pan, Synthesis gas production with an adjustable H$_2$/CO ratio through the coal gasification process: effects of coal ranks and methane addition. *Energy Fuels* **22**, 1720–1730 (2008).

9. H. Zhou, Y. Cao, H. Zhao, H. Liu, and W. Pan, Investigation of H$_2$O and CO$_2$ reforming and partial oxidation of methane: catalytic effects of coal char and coal ash. *Energy Fuels* **22**, 2341–2345 (2008).

10. Y. Li, R. Xiao, B. Jin, and H. Zhang, Experimental study of the reforming of methane with carbon dioxide over coal char. *Int. J. Chem. Reactor Eng.* **6**, A16 (2008).

11. Z. Bai, H. Chen, W. Li, and B. Li, Hydrogen production by methane decomposition over coal char. *Int. J. Hydrogen Energy* **31**, 899–905 (2006).

12. W. Chen, F. Sheu, and R. Savage, Catalytic activity of coal ash on steam methane reforming and water-gas shift reactions. *Fuel Process. Technol.* **16**, 279–288 (2006).

13. M. Haghighi, Z. Sun, J. Wu, J. Bromly, H. Wee, E. Ng, Y. Wang, and D. Zhang, On the reaction mechanism of CO$_2$ reforming of methane over a bed of coal char. *Fuel Process. Technol.* **16**, 279–288 (2006).

14. H. Zhou, Y. Cao, H. Zhao, H. Lui, and W. Pan, Investigation of H$_2$O and CO$_2$ reforming and partial oxidation of methane: catalytic effects of coal char and coal ash. *Energy Fuels* **22**, 2341–2345 (2008).

15. M. Halmann and A. Steinfeld, Hydrogen production and CO$_2$ fixation by flue-gas treatment using methane tri-reforming of coke/coal gasification combined with lime carbonation. *Int. J. Hydrogen Energy.* **34**, 8061–8066 (2009).

16. Z. Sun, J. Wu, and D. Zhang, CO$_2$ and H$_2$O gasification kinetics of a coal char in the presence of methane. *Energy Fuels* **22**, 2160–2165 (2008).

17. J. Cor, N. Manton, G. Mul, D. Eckstrom, W. Olson, R. Malhotra, and S. Niksa, An experimental facility for the study of coal pyrolysis at 10 atmospheres. *Energy Fuels* **14**, 692–700 (2000).

18. Y.-L. Liu, R. Malhotra, and S. Niksa, Impact of pressure variations on coal devolatilization products. 1. detailed product distributions from 0.1 MPa. *Energy Fuels* **18**, 508–519 (2004).

19. N. Manton, J. Cor, G. Mul, D. Eckstrom, R. Malhotra, and S. Niksa, Impact of pressure variations on coal devolatilization products. 2. detailed product distributions from 1.0 MPa. *Energy Fuels* **18**, 520–530 (2004).

20. D.J. Eckstrom, A.S. Hirschon, R. Malhotra, and S. Niksa, Kinetics of pulverized coal combustion at high pressures. *Prepr. Pap.-Am. Chem. Soc. Div. Fuel Chem.* **49**, 811–816 (2004).

21. G.-S. Liu and S. Niksa, Pulverized coal flame structures at elevated pressures. Part 1. Detailed operating conditions. *Fuel* **84**, 1563–1574 (2005).

22. S. Niksa and G.-S. Liu, Pulverized coal flame structures at elevated pressures. Part 2. Interpreting NO$_X$ production with detailed reaction mechanisms. *Fuel* **84**, 1575–1585 (2005).

23. S. Niksa, Chemical reaction kinetics for the initial stages of entrained flow gasification, paper presented at the 238th ACS National Meeting, Washington, DC, 16–20 August 2009, http://oasys2.confex.com/acs/238nm/techprogram/P1279071.HTM (2009).

24. D. Eckstrom (SRI), S. Niksa (NEA), et al., High pressure coal combustion kinetics, Final Technical Report (DOE Award No. DE-FC26-01NT40777, Fluent Subcontract No. FY00012SRI September 2004).

25. G.-S. Liu and S. Niksa, Coal conversion submodels for design applications at elevated pressures. Part II. Char gasification. *Prog. Energy Combust. Sci.* **30**, 697–717 (2004).

26. D.J. Eckstrom, A.S. Hirschon, R. Malhotra, and S. Niksa, Kinetics of pulverized coal combustion at high pressures, paper presented at the *12th International. Conference on Coal Science*, Cairns, Australia, 2–6 November 2003.
27. M. Lussier, Z. Zhang, and D. Miller, Characterizing rate inhibition in steam/hydrogen gasification via analysis of adsorbed hydrogen. *Carbon* **36**, 1361–1369 (1998).
28. Department of Energy, National Energy Technology Laboratory, Baseline Technical and Economic Assessment of a Commercial Scale Fischer-Tropsch Liquids Facility, (2007).
29. Department of Energy, National Energy Technology Laboratory, Cost and Performance Baseline for Fossil Energy Plants, Volume 1: Bituminous Coal and Natural Gas to Electricity, Revision 1, (2007).
30. V. Wan, R. Smith, Methanol from Coal, Process Economics Program SRI Consulting, (2006).
31. Argonne National Laboratory, GREET Model 1.8d.1.
32. J.T. Bartis and L. van Bibber, Alternative fuels for military applications, RAND Corporation, (2011).