Producing hydrogen from natural gas using hot slag from steel plants

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Abstract

A process for thermal decarbonization of methane to hydrogen and solid carbon is presented and examined. It utilizes the high temperature heat from the slag by-product of ironmaking to drive a thermal decomposition reaction, making it a waste-heat-to-hydrogen technology. This is accomplished *via* dry granulation of molten slag that feeds a fluidized bed reactor to effect methane-slag contact. First, the proposed process and the heat and mass balances are presented. Then, a techno-economic analysis investigates the capital and operating costs of the process, compares the hydrogen production cost to that of other processes, and examines cost sensitivity to the prices of process inputs and outputs. Finally, a computational fluid dynamics (CFD) modelling study of the fluidized bed reactor examines the thermal decomposition of methane and its dependence on reaction kinetics as well as reactor design and operation.

Keywords: thermal decomposition, decarbonization, pyrolysis, methane, iron

1. Introduction

The steel industry has been considering heat recovery from hot slag to improve efficiency (*e.g.*, as described in [1]). Slag is usually disposed of in pits or it undergoes a wet granulation process to convert the material for use in cement manufacture. In either case, the high-quality energy of the slag is severely downgraded or completely lost. The recovery of high temperature heat from the slag requires dry granulation, for which methods have been proposed and studied (*e.g.*, [2], [3]). Typically, the intention has been to capture the slag energy as high-temperature air (at 500-600 °C) for use in the plant, and to achieve the required properties of the slag by-product for use in cement manufacture [1], [3].

Climate change concerns have spurred interest in low-carbon fuels and feedstocks, particularly for high temperature industrial processes that are difficult to decarbonize [4], [5]. Hydrogen is carbon-free, but it needs to be produced, usually from water or a hydrocarbon. A production pathway that has gained attention recently is to decompose natural gas (which consists mostly of methane) into gaseous hydrogen (or a hydrogen-rich gas) and a solid carbon (or carbon-rich) by-product. An advantage of methane decomposition is that the carbon-containing by-product is potentially easier to store or utilize and, in addition, the revenue from utilization would potentially offset the CO₂-avoidance cost. A disadvantage is that, compared to steam-methane reforming, it produces half the hydrogen per unit methane input. Nonetheless, about 60% of the chemical energy of the methane is retained after decarbonization and, unlike combustion, a carbon dioxide by-product is avoided. When carbon dioxide utilization and/or

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storage (CCUS) is an option, the solid carbon can feed a direct carbon fuel cell to generate power and produce a pure CO₂ stream [6], [7].

Reviews of methane thermal decomposition technologies have been carried out. Canada's Oil Sands Innovation Alliance (COSIA) commissioned reviews of technology pathways for natural gas decarbonization with the aim to generate hydrogen for steam production, along with value-added byproducts to offset the CO₂-avoidance cost [8], [9]. These studies examined a wide range of processes including the use of catalysts, membranes, and electricity. Schneider et al. [10] reviewed the state of the art in decarbonization of natural gas by thermal, electrical, and catalytic methods, including fluidized bed configurations.

The United States Department of Energy (DOE) commissioned a study and testing [11] on the production of hydrogen and carbon from thermal decomposition of natural gas and other hydrocarbon fuels. The carbon produced in the process acted as a catalyst for the decomposition reaction. Various concepts and designs for the hydrocarbon pyrolysis reactor were evaluated and tested, including packed bed, tubular, free volume, fluid wall, and fluidized bed configurations. The hydrogen concentration in the effluent gas varied in the range of 30-90% by volume, depending on operating parameters and the hydrocarbon feedstock. The study concluded that the fluidized bed reactor was the most suitable for decomposition of both methane and propane.

The production of hydrogen using hot slag represents a conversion of waste heat to chemical energy, as opposed to its physical capture in heated air. Li [12] showed that combined physical and chemical conversion of the slag energy is more efficient than physical capture alone, both for quantity of energy captured and for making the highest quality use of it. In addition, Barati et al. [13] showed that recovery of slag energy in chemical form offers a higher energy density than in physical form and a much better ability to transport the energy. Purwanto and Akiyama [14] performed experiments to study the kinetics of the decomposition of methane and carbon dioxide (representing biogas) using granulated slag to produce hydrogen and carbon monoxide. Their apparatus was an electrically heated, fixed bed of about 2 grams of slag particles, whose average diameter was 2 millimeters. They observed a catalytic effect from the presence of the slag particles that was higher than for an iron catalyst but not as high as for nickel and cobalt catalysts.

This work proposes using the hot slag from ironmaking to drive the endothermic methane decomposition, using a fluidized bed to effect contact between slag and natural gas. This requires that the molten slag undergo dry granulation. As mentioned, the scientific literature reports both experimental and modelling studies of dry granulation by rotary atomization [1], [2], [3]. This type of granulation method is envisioned for the process proposed here, although the granulation itself is not examined in detail.

2. Proposed Process

Figure 1 shows the proposed process. The rotary atomizer granulates liquid slag to solid particles with expected diameters ranging from 100 μ m to 3 mm [1]. The granulated slag falls into a fluidized bed and contacts methane travelling upward from the bottom. The methane undergoes thermal decomposition to gaseous hydrogen and solid carbon. These products flow through a heat exchanger where they cool to 500 °C, quenching the decarbonization reaction and heating the incoming methane. Most of the slag

particles, along with deposited carbon, are discharged at the bottom of the fluidized bed. Some of the slag particles are entrained by the gas flow, exit the top of the fluidized bed, and are separated from the product stream in cyclones. The product gas is cooled to 150 °C in a gas cooler and then entrained solid carbon is separated from the gas stream using a fabric filter. Hydrogen is separated from any remaining methane using a palladium membrane. The remaining methane is returned to the fluidized bed, where it is combined with the supplied methane prior to entering the heat exchanger.



Figure 1: Proposed process to produce hydrogen from methane using molten slag

Aspen Plus[®] was used to analyze the process shown in Figure 1 for a case corresponding to the blast furnace production (specifically, the slag production rate and temperature) of a Canadian steel plant. The results are shown in Table 1. It is expected that typically about 20% of the reductant could be replaced by the produced hydrogen, although this percentage would depend on the steel-to-slag ratio of a given process. There are other ways the hydrogen could be used to reduce CO₂ emissions. In many slag-producing processes (whether blast furnace, basic oxygen furnace, or electric arc furnace), slag-tapping is not continuous. The slag transfer to the proposed process would need to have buffer capacity in order for it to operate continuously.

Slag input, tonnes/year	367000
Slag temperature, °C	1300
Natural gas input*, tonnes/year	45000
Natural gas temperature, °C	20
Hydrogen produced, tonnes/year	9354
Carbon black produced, tonnes/year	4814
Slag plus deposited carbon black, tonnes/year	389450

Table 1: Hydrogen production using slag from a Canadian blast furnace

* Natural gas is approximated as methane.

3. Techno-economic Analysis

Keipi et al. [15] studied the cost of hydrogen production by thermal decomposition of methane (TDM) using a fluidized bed reactor, which they called a "regenerative heat exchanger reactor" (RHER). The main difference between the present process and that of Keipi et al. [15] is that the thermal energy for this decomposition process comes from the hot slag rather than from burning a portion of the natural gas. Like the present system, their process filtered carbon from the gas stream and separated hydrogen from methane using a palladium membrane filter. Results of their capital cost analysis are shown in Table 2, along with the corresponding equipment of the present system.

Table 2: Estimated cost of Keipi et al. [15] for thermal decomposition of methane (TDM) with hydrogen production of 529 tonnes/year (60 kg/h), along with the corresponding equipment of the present system

Keipi et al. [15] TDM equipment	Capacity	Cost (thousand EUR)	Corresponding equipment of the present system
RHER reactor	60 kg H₂/h	2740	Fluidized bed reactor
Membrane reactor	60 kg H₂/h	98	Membrane reactor
Membrane tube	60 kg H₂/h	273	Membrane tube
Carbon pelletizer	252 kg C/h	121	Carbon pelletizer
Waste heat boiler	60 kg H₂/h	25	Heat exchanger

The costs from Keipi et al. [15] were scaled according to equipment capacity using the relationship recommended by Peters and Timmerhaus [16], which is expected to provide capital cost estimates within \pm 50% [15].

(2)

Table 3 provides the scaled capital cost for the present system based on this scaling relationship, converted to Canadian dollars.

Equipment unit	Capacity	Scale exponent, n^{*}	Cost (thousand CAD)*
Fluidized bed reactor	1067.8 kg H₂/hr	0.6	23035
Membrane reactor	1067.8 kg H ₂ /hr	0.6	824
Membrane tube	1067.8 kg H ₂ /hr	0.6	3059
Carbon pelletizer	549.5 kg C/hr	0.7	290
Heat exchanger	1067.8 kg H ₂ /hr	1	663
			27871

Table 3: Estimated capital cost for major equipment of the present system with hydrogen production of 9354 tonnes/year (1067.8 kg/h)

*EUR to CAD exchange factor of 1.5 *Refers to Equation (1)

To convert the total capital cost to an annual cost, the standard capital recovery factor (CRF) was employed:

$$CRF = i(1+i)^N / ((1+i)^N - 1)$$

where i is the annual interest rate and N is the investment period in years.

Keipi et al. [15] recommended an annual operation and maintenance (O&M) cost of 2% of the annual capital cost, except for the membrane reactor for which they recommended an annual O&M cost of 4% of its total capital cost. Their O&M cost did not include the costs of electricity or methane, which were accounted for separately. This approach is employed in the present analysis.

Additional assumptions were required for the cost estimates. The investment period was assumed to be 15 years, with no remaining value at the end. The annual interest rate was 10%. Given that the scaling relationship of Equation (1) is expected to provide a capital cost estimate within ±50%, and that the proposed system is novel, a 50% contingency was added to the capital cost estimates, which is higher than the 30% used by Keipi et al. [15]. Based on information from Canadian steel producers, the sale price for slag that is uncontaminated by carbon (and therefore useful for cement production) was assumed to be CAD 25/tonne and the sale price for slag that is contaminated by carbon (and therefore only useful as aggregate) was assumed to be CAD 5/tonne. Like Keipi et al. [15], it was assumed that 80% of the carbon black would deposit on the slag, with the rest entrained by the product gas stream. Again, natural gas was represented by 100% methane.

Results of the analysis are shown in Table 4, which presents the net annual hydrogen production cost capital, maintenance, and natural gas costs minus the slag and carbon black revenue. Table 4 (a) examines the effect of natural gas purchase price on hydrogen production cost and Table 4 (b) examines the effect of carbon black sale price on hydrogen production cost. In all cases, the capital cost with contingency was CAD 41.8 million and the capital cost recovery factor was 0.131 (corresponding to the 10% annual interest rate and 15-year investment period), for an annual capital cost of CAD 5.50 million. The annual O&M cost was CAD 83500. The annual natural gas input was 45000 tonnes/year, the annual slag output was 389450 tonnes/year, the annual carbon black output was 4814 tonnes/year, and the annual hydrogen output was 9354 tonnes/year. Note that the annual slag output includes the slag and 80% of the carbon generated in the process.

As mentioned earlier, throughout Table 4, it was assumed that 80% of the solid carbon would deposit on the slag, resulting in 6% carbon-in-slag and, consequently, the slag sale price would drop from CAD 25/tonne to CAD 5/tonne. There are interesting economic considerations with respect to the carbon and the slag. For example, with a carbon black sale price of CAD 0.7/kg, the reduction in the sale price of the slag from CAD 25/tonne to CAD 5/tonne (367000 tonnes × CAD 25/tonne - 389450 tonnes × CAD 5/tonne = CAD 7.2 million lost revenue) would result in a net income reduction of CAD 3.8 million as the sale of carbon black (CAD 700/tonne × 4814 tonnes = CAD 3.4 million) failed to compensate for the reduction in slag revenue. If the hydrogen were to be sold, the mark-up over its production cost would have to be CAD 0.41/kg (CAD 410/tonne × 9354 tonnes = CAD 3.8 million) to compensate for the reduction in slag revenue. In the extreme, if neither the carbon black nor slag had any value (representing a CAD 9.2 million loss of slag revenue), the hydrogen mark-up would have to be CAD 0.98/kg to compensate.

The estimated hydrogen production cost varied from CAD 0.62 to 3.25/kg, driven strongly by the price of natural gas, which was varied from CAD 1.55/GJ to CAD 11.38/GJ. Natural gas price constituted 40% to over 80% of the hydrogen production cost—the higher the natural gas price, the higher its percentage of the production cost. The O&M cost accounted for less than 5% whereas the capital cost accounted for 20% to 60%. The carbon black sale price had a relatively small impact on the hydrogen production cost.

Variable	Unit	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
Natural gas price	CAD/kg	0.086	0.176	0.320	0.417	0.528	0.632
Natural gas annual cost	CAD Million	3.87	7.92	14.41	18.78	23.78	28.42
Carbon black price	CAD/kg	0.35	0.35	0.35	0.35	0.35	0.35
Slag & carbon black annual sale	CAD Million	3.63	3.63	3.63	3.63	3.63	3.63
Net H ₂ annual production cost	CAD Million	5.82	9.86	16.36	20.73	25.72	30.37
Net H ₂ cost per kilogram	CAD/kg	0.622	1.055	1.749	2.216	2.750	3.247

Table 4 (a): Hydrogen production cost estimate at different natural gas purchase prices with fixed carbon black sale price

Table 4 (b): Hydrogen production cost estimate at different carbon black sale prices with fixed natural gas purchase price

Variable	Unit	Case 1	Case 2	Case 3	Case 4
Natural gas price	CAD/kg	0.239	0.239	0.239	0.239
Natural gas annual cost	CAD Million	10.74	10.74	10.74	10.74
Carbon black price	CAD/kg	0.000	0.250	0.500	0.700
Slag & carbon black annual sale	CAD Million	1.95	3.15	4.35	5.32
Net H ₂ annual production cost	CAD Million	14.38	13.17	11.97	11.01
Net H ₂ cost per kilogram	CAD/kg	1.537	1.408	1.280	1.177

It is important to compare the proposed process to alternative hydrogen production processes. Table 5 compares the proposed methane thermal decomposition process using hot slag to the methane decomposition, steam-methane reforming, and water electrolysis scenarios presented by Keipi et al. [15]. This comparison assumed 15 years of operation, a 10% annual interest rate, a natural gas price of CAD 0.463/kg, an electricity price of CAD 45/MWh, a CO₂ emissions cost of CAD 15/tonne of CO₂, and no sale of carbon black or slag. It was found that the proposed slag-to-hydrogen process is competitive with large-scale steam-methane reforming with integrated CCS and much less costly than small-scale steam-methane reforming. This supports a conclusion similar to that of Keipi et al. [15] in that it would be most suitable for on-site hydrogen production and use at a plant. In addition, the present process using slag waste heat reduced the hydrogen production cost by approximately 25% compared to methane thermal decomposition driven by combusting a portion of the natural gas itself.

Table 5: Hydrogen production cost of this process (bold type) compared to that of the processes presented by Keipi et al. [15]. TDM is thermal decomposition of methane, SMR is steam-methane reforming, SS is small-scale, LS is large-scale, and CCS refers to integrated CO₂ capture and storage. The CO₂ emissions cost was CAD 15/tonne.

	Slag-TDM	TDM-SS	SMR-SS	SMR-LS	SMR- LS+CCS	Water electrolysis
H ₂ production, tonnes/year	9354	529	529	52900	52900	529
H ₂ production cost with CO ₂ cost, CAD/kg H ₂	2.80	3.88	6.00	2.49	3.25	3.93
H ₂ production cost without CO ₂ cost, CAD/kg H ₂	2.80	3.86	5.80	2.31	3.17	3.93

4. Technical Analysis of the Reactor Concept

To examine the fluidized bed reactor concept, a simplified CFD modelling study was performed. The technical aspects of interest were the gas and particle flow, heat transfer, thermal decarbonization reaction, and fate of the solid carbon. A possible fluidized bed configuration is shown in Figure 2. The percent conversion of methane to hydrogen is of key interest because, referring to Figure 1, higher conversion percentage means that less methane needs to be recycled back to the reactor, resulting in a smaller reactor size for a given hydrogen production rate.



Figure 2: A possible fluidized bed reactor configuration. The reactor portion, indicated by the dashed lines, was examined by CFD modelling

A simplified fluidized bed reactor configuration was created for the CFD analysis. This configuration, of pilot-scale size, is shown in Figure 3. To simplify the model, solid slag particles were injected into the reactor at the indicated location (although, if designed as in Figure 2, they would be falling into the reactor fom the top). The slag discharge pipe was not used for reasons that will be explained. The reactor walls were assumed to be adiabatic. Methane was injected from the bottom and the reactor outlet was at the top. Process modelling using Aspen Plus suggested the operating parameters shown in Table 6, which were implemented for the CFD simulation. The reactor diameter was chosen such that the fluidized bed would operate as a bubbling bed in order to provide a long residence time for gas-solid contact. The reactor height, as well as the location of hot slag injection, were adjusted based on preliminary CFD simulation results to reduce the reactor size and improve performance, resulting in the configuration shown in Figure 3.



Figure 3: Simplified fluidized bed reactor configuration for CFD analysis. The diagram is not to scale.

$\cdot \cdot $	Table 6: Fluidized bed read	ctor operation suggested	d by Aspen Plus proce	ss modelling
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Solids holdup	7.9 kg
Overall bed pressure-drop	0.2 bar
Bed temperature	1083 °C
CH₄ injection rate	1.5 kg/h
CH ₄ injection temperature	500 °C
Slag injection rate	37.8 kg/h
Slag injection temperature	1300 °C

The methane decomposition reaction was modelled by a particle surface reaction,

 $CH_4 + C_{core} \rightarrow (C_{core} + C) + 2 H_2$

(3)

where C_{core} represents nucleation particles of carbon-black-size but with very small mass, which were injected into the bed with the methane. Carbon was then added to these particles as the methane decomposed and, as the carbon deposited, the particle diameters were assumed to remain constant while the particle densities increased. The resulting density of carbon particles in the simulation was somewhat high but within the right order of magnitude. It will be shown that this over-prediction of carbon particle density did not affect the gas flow's ability to suspend the carbon particles.

No kinetic data were available for methane decomposition driven by hot slag particles so the results presented here used kinetic data from two alternative sources. The first was based on test results from a fluidized bed using different carbon particles as bed materials [11] and the second was derived from experiments by Kashiwaya and Watanabe [17] in which methane was injected into molten slag.

The multiphase flow was modelled using the dense discrete phase model in ANSYS-Fluent[®]. Particleparticle interactions were modelled using the kinetic theory of granular flow. There were three discrete phases: slag particles initially in the bed, injected slag particles, and carbon particles. Preliminary simulations indicated that particles with diameter less than 140 µm would likely be blown out of the bed. Accordingly, the slag bed and injected slag consisted of particles with diameters of 150, 210, 270, 330 and 390 µm, evenly distributed. Carbon particle diameters ranged from 0.2 µm to 3 µm with mean diameter of 1 µm and spread factor of 3.1 according to carbon black samples measured by CanmetENERGY-Ottawa. The P1 radiation model of ANSYS-Fluent was used to model the radiation heat transfer and the weightedsum-of-gray-gases model was used for the absorption coefficient of the gas phase. The particles participated in the heat transfer by convection, radiation, and, in the case of the carbon particles, an endothermic methane-decomposition surface reaction, using the framework provided in ANSYS-Fluent. Table 7 provides key heat transfer parameters for the particles.

Slag particle heat capacity, $J/(kg \cdot K)$	960
Carbon particle heat capacity, $J/(kg \cdot K)$	1680
Endothermic heat of methane decomposition, kJ/mol_{CH_4}	76
Fraction of endothermic reaction heat taken from particle	03
(remainder taken from gas phase)	0.5
Radiative surface emissivity of slag particles	0.9
Radiative surface emissivity of carbon particles	0.9

Table 7: Particle heat transfer parameters

The CFD model simulated 16 seconds of operation time and achieved steady state with respect to fluidization and methane decomposition. These 16 seconds of simulated operation took 26 days to compute using an HP Z840 workstation with 24 cores. The relatively short, simulated operation time, due to the computational intensity, meant that the simulation could not achieve steady state with respect to solids inventory in the bed. It was, therefore, decided to not allow solids to exit the reactor through the discharge pipe. As mentioned, hot slag particles entered through the injection location to compensate for heat consumed by methane decarbonization. As a result, the bed solids inventory increased by 2% during the 16 seconds. In real operation, the slag discharge rate would be controlled to match the injection rate. This could be accomplished in a CFD simulation, albeit at high computational cost.

The CFD simulation results confirmed that the fluidized bed operates in the bubbling regime, which is consistent with the earlier process design. The bubbling bed was found to have a height of approximately 1 meter. A plot of instantaneous particle volume fraction at steady state is shown in Figure 4. The average bed temperature at steady state remained between 1020 and 1060 °C, which is close to that predicted by the Aspen Plus simulation (referring to Table 6), and the average temperature of the gases exiting the reactor was approximately 850 °C. In the configuration studied here, the resulting mole fraction of hydrogen depended noticeably on the decomposition reaction kinetics employed. Using the kinetics from [11], the hydrogen mole fraction was 0.7, whereas using the kinetics from Kashiwaya and Watanabe [17], it was 0.9. The 0.7 hydrogen mole fraction implies a 53.8% conversion of methane to hydrogen (each mole of converted methane results in two moles of hydrogen) whereas the 0.9 hydrogen mole fraction implies an 82% methane conversion. As discussed at the beginning of this section, higher conversion percentage means that less methane needs to be recycled back to the reactor. The more methane that needs to be recycled back, the larger the required reactor for a given hydrogen production rate. There is clearly a significant difference between the conversion percentages involving the two sets of reaction kinetics. The variation of reaction kinetics in the literature is not unexpected, given that the decomposition of methane is influenced by catalytic effects that depend on the composition of the material in contact with it, particularly when the material contains metal compounds.



Figure 4: Instantaneous solid volume fraction of the fluidized bed at steady state. The slag injection location is circled. The height [meters] is indicated.

Figure 5 (a) shows the predicted hydrogen mole fraction (the balance being methane) and gas temperature in the reactor when kinetics from [11] were used. The hydrogen mole fraction reached approximately 0.7 at the reactor midpoint and then the reaction stalled. Figure 5 (b) shows the predicted gas temperature. As expected, the heating by slag particles occurred mostly in the bottom 1 meter of the reactor, where the bubbling bed is located. The decarbonization reaction continued to progress above the bed for another 2 meters until it stalled. The temperature plot indicates that, as the endothermic decarbonization reaction proceeded, the gas temperature dropped to a level at which the reaction rate became very slow. Thus, little was accomplished in the top 3-meters of the reactor. This was confirmed by examining plots of the reaction rate. It is possible that, with further optimization of the design and operating conditions, the methane decomposition percentage could be improved. Increased bed height would create a longer high-temperature region, which would likely improve the methane conversion.





The rate of carbon particles exiting the reactor at the gas outlet was found to be 0.61 kg/h, representing essentially all the converted carbon (the mass flow rate of carbon in the methane times the 53.8% conversion rate). This implies that the flow was able to suspend the carbon particles and carry them to the gas outlet, despite their excessive density mentioned earlier. Whether the carbon particles would exit through the gas outlet in reality, given that carbon might stick to the slag particles instead and exit with the slag discharge, requires further (and likely experimental) investigation.

Conclusion

A new process was presented for thermal decarbonization of methane to gaseous hydrogen and solid carbon in which a hot, dry-granulated slag feeds a fluidized bed reactor to effect methane-slag contact and drive the methane decomposition. It is expected that about 20% of the reductant could be replaced by the produced hydrogen, depending on the steel-to-slag ratio of a given process. (There are other ways the hydrogen could be used to reduce CO_2 emissions.) A techno-economic analysis was performed, suggesting that the proposed hydrogen production process would be competitive with large-scale steammethane reforming with integrated CCS and much less costly than small-scale steam-methane reforming, making it suitable for on-site hydrogen production and use at a plant. In addition, using the hot slag to drive the methane decomposition would reduce hydrogen production cost by 25% compared to combusting a portion of the natural gas itself. A CFD analysis was carried out to examine the fluidized bed reactor. It suggested that the fluidized bed approach is an appropriate method to provide methane-slag contact. It operated in the bubbling bed regime and resulted in significant decarbonization of the methane. Further work would be required to optimize the design and operation, and to control the solids inventory in the fluidized bed. Further (likely experimental) investigations would be required to reduce the uncertainty of the chemical kinetic rate for methane decomposition as well as to determine the fate of the produced carbon.

Acknowledgment

Funding for this work was provided by Natural Resources Canada through the Program of Energy Research and Development.

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