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In-situ and Ex-situ Conversion of Coal to Methane using Hydrogen



Abstract: This paper explains a process to produce methane using coal, steam, and hydrogen by autothermal reactions. In coal gasification, oxygen is used to burn extra coal for providing the necessary energy to the process. The proposed process uses hydrogen in place of oxygen to provide the necessary energy to produce methane. The process uses fossil coal as a feedstock only and not as a fuel. Both ex-situ and in-situ methods are feasible to produce methane/natural gas from coal and green hydrogen. For one unit mass of hydrogen, nearly 24 times methane production is feasible. The required carbon-neutral hydrogen can be generated adequately and economically from water electrolysis by using green renewable electricity from solar PV and wind power potential. The carbon footprint of methane production using coal and green hydrogen is nearly 83% more than that of fossil natural gas but 23% less compared to fossil coal on an equivalent energy basis. The abundant inferior quality coal deposits in India can be used as carbon feedstock to produce methane gas for the fuel requirements of domestic, commercial, and road transport sectors. The existing natural gas infrastructure like cross country pipelines, PNG infrastructure, CNG filling stations, LNG storage facilities, etc remain in use and are further expanded to use the methane produced from coal deposits. The green carbon dioxide gas available from biomass gasification can be sequestered into underground caverns and depleted natural gas or crude oil deposits to brand the grey methane, produced from fossil coal, as carbon neutral fuel. Thus, India can transform into a carbon-neutral robust with economv energy security/independence from a predominantly fossil fuels importing country to an exporter of green fuels, chemicals, and products by harnessing its vast coal, biomass, and renewable energy resources.

Keywords: Coal Gasification, Fischer-Tropsch, Hydrogenation, Methanation, Pyrolysis

Abbreviations:

CNG: Compressed Natural Gas LNG: Liquified Natural Gas SNG: Synthetic Natural Gas FCEV: Fuel Cell Electric Vehicles BEV: Battery Electric Vehicles PNG: Piped Natural Gas PV: Photovoltaic ICE: Internal Combustion Engines

I. INTRODUCTION

Coal had been the predominant energy source from the advent of industrial revolution to a decade ago. It was used as

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a transport fuel by deploying steam engines for nearly 150 years. Even now, it is a major fuel in electricity generation, cement production, and steel production. In the year 2023, energy contributed by coal to total global energy produced was at 26.5% which ranked second position of all fuels [1]. Compared to other fossil fuels such as natural gas and crude oil derivatives on an equivalent energy basis, coal is a major culprit in contributing to global warming, higher water footprint, higher land footprint in the form of open cast mining and ash disposal areas, and air pollution by emitting more particulate matter (PM2.5, PM10, etc.), No_x, So_x, CO, mercury, etc [23]. To reduce the carbon footprint, coal consumption is being discouraged compared to natural gas and crude oil. The prime position of coal in electricity generation is also challenged by the advent of cheaper, renewable, and carbon-neutral wind power and solar photovoltaic (PV) power. However, natural gas and crude oil deposits are not available adequately in all countries. The major populous regions/countries like India, China, East Asia, South Africa, Germany, Columbia, Poland, etc. ought to depend on costly imported liquified natural gas (LNG) or piped compressed natural gas (CNG) and crude oil in the process of reducing coal usage till adequate renewable energy sources are harnessed to achieve carbon neutral status [24]. Dependence on imported natural gas and crude oil is leading to energy security or sourcing problems in energy-importing countries due to transport bottlenecks arising from regional conflicts, international trade restrictions, monopolistic practices of energy-exporting countries, etc. Carbon-neutral hydrogen, produced from renewable energy or biomass, is also emerging as an alternate transport fuel, and feedstock to ammonia/urea and petrochemicals production in place of hydrogen derived from fossil fuels. This paper investigates both ex-situ and in-situ methods to produce methane/natural gas from coal using carbon-neutral hydrogen. In the in-situ method, coal is converted into methane in the original coal seams without mining. In the ex-situ method, coal extracted from mines is used to produce methane in a production plant.

II. METHODS

The proposal is to synthesize methane by combining its elements carbon and hydrogen. It is an exothermic reaction which is possible between 650 to 870°C up to 40 bar pressure [2]. There is feasibility of many possible reactions, but the ultimate product is methane as given below.

 $C + 1/2O_2 \rightarrow CO - 110.5$ Kilo joule/gram mole at 25°C (exothermic reaction above 600°C)

 $CO + 2H_2 \rightleftharpoons CH_4 + 1/2O_2 + 38$ Kj/mole (endothermic equilibrium reaction, reverse dry

reforming of methane reaction up to 850°C. The generated oxygen is recycled) [3].



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Published By: Lattice Science Publication (LSP) © Copyright: All rights reserved. $C + 2H_2 \rightleftharpoons CH_4 - 74.9$ Kj/mole (exothermic equilibrium reaction, methanation reaction between 650 to 870° C up to 40 bar pressure)

The following intermediate reactions can also take place making use of the heat energy liberated in the synthesis process. The ultimate product is methane from CO and H_2 gases [4].

 $C + 2H_2O \rightleftharpoons CO_2 + 2H_2 + 89.8$ Kj/mole (endothermic equilibrium water gas reaction above 600°C)

 $C + H_2O \rightleftharpoons CO + H_2 + 131$ Kj/mole (endothermic equilibrium water gas reaction)

 $CO + H_2O \rightleftharpoons CO_2 + H_2 - 41.2$ Kj/mole (exothermic equilibrium reaction. Water gas shift reaction between 225 to $450^{\circ}C$

 $C + CO_2 \rightleftharpoons 2CO + 172.5 \text{ Kj/mole}$ (endothermic equilibrium Boudouard reaction)

 $CO + 3H_2 \rightleftharpoons CH_4 + H_2O - 206.2$ Kj/mole (exothermic equilibrium reaction, Fischer-Tropsch process methanation reaction)

 $CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O - 130.8$ Kj/mole (exothermic reaction, Sabatier reaction [5])

Raw methane, from the gasifier at temperature 650 to 870° C, is cooled and the available sensible heat is conserved by heating the hydrogen and steam that are to be injected into the coal. Nearly 24 times methane by weight is produced from the hydrogen consumed. In terms of carbon/coal consumed, nearly 1.38 kg of carbon is required for every kg of methane produced. The liberated heat energy by the exothermic methane synthesis reaction is utilized to convert more carbon using H₂O into H₂ and CO₂ as given below.

 $C + 2H_2O \rightleftharpoons CO_2 + 2H_2 + 89.8$ Kj/mole (endothermic reaction above 600°C)

 $[C + 2H_2 \rightleftharpoons CH_4 - 74.9 \text{ Kj/mole}] \times 1.2 \text{ to provide 89.8 Kj}$ heat in an exothermic reaction.

26.4 kgs of carbon + 0.8 kgs of hydrogen (net) + 36 kgs ofH₂O produces 19.2 kgs of CH₄ + 44 kgs of CO₂ undergoing auto thermal reactions without any external heat supplementation. The process is a destructive distillation (pyrolysis) of coal with hydrogen and steam at controlled temperatures and pressure to produce methane predominantly. Potassium carbonate and Fe₂O₃ are found to be better catalysts to enhance methane formation [6]. Highly water-soluble potassium carbonate is separated from the residual coke by water washing. The reactants hydrogen and steam are mostly converted into product gas methane. It has been also observed that irradiation of carbon has increased its oxidation rate at low temperatures [7]. The unreacted hydrogen and steam along with CO₂ are separated from the raw methane and the separated H₂ is reused. CO gas associated with methane is converted to H₂ using a water gas shift reaction before separating CO₂, H₂, etc. from the raw methane gas [8]. Any other organic chemicals such as ethane, propane, etc are also separated from the raw methane while removing impurities from methane. The total carbon footprint is 5.04 kg per kg of CH_4 produced from coal if H_2 is produced by splitting water using green electricity. The carbon footprint in the production of CH₄ is 2.29 kg and upon its utilization is 2.75 kg per kg of CH₄. The total carbon footprint of methane produced from coal is higher by nearly 83% than that of fossil natural gas. However, the carbon footprint of coal/carbon direct burning is 23% more than that of methane produced from coal on an equivalent energy basis. The produced CO_2 gas can be sequestered into underground caverns to make the methane production process carbon neutral.

All the coal fed to the gasifier is not consumed and residual coke (some part of fixed carbon) mixed with ash is extracted from the gasifier. The byproduct coke can be used for generating heat in industrial and commercial applications as it burns smokeless. The residual coke can further be used to convert the available CO_2 gas into CO using O_2 gas available from the water splitting process that is used for producing H_2 from renewable electricity. The following reactions take place in the process.

 $2H_2O \rightarrow 2H_2 + O_2 + 283.4$ Kj/mole (Renewable electricity is used for splitting water. Eight times oxygen by weight is available from the hydrogen generated)

 $C + CO_2 \rightleftharpoons 2CO + 172.5 \text{ Kj/mole}$ (endothermic reaction between 600 to 1400°C)

 $[C + 1/2O_2 \rightleftharpoons CO - 110.5 \text{ Kj/mole}] \ge 1.56 \text{ liberates } 172.5 \text{ Kj in an exothermic reaction.}$

30.72 kgs of carbon + 44 kgs of CO₂ + 24.96 kgs of O₂ produces 99.68 kgs of CO gas undergoing auto-thermal reactions. The high temperature product CO gas available at around 1400°C is used to heat the reactants (CO₂ and O₂ gases) using thermocline/recuperative heat exchangers [9]. The CO gas further reacts with H₂O to produce H₂ and CO₂ gases using water gas shift reaction at around 250°C in the presence of a suitable catalyst. Thus, all the extracted coal is used for producing CH₄ and H₂ gases.

 $CO + H_2O \rightleftharpoons CO_2 + H_2 - 41.2 \text{ Kj/mole.}$ (99.68 kg of CO gas produces nearly 7.12 kgs of H₂ gas and 156.64 kgs of CO₂) The carbon footprint of hydrogen production from coke is 15.82 kg of CO₂ per kg of H₂. The carbon footprint of CH₄ in its production process is 2.95 kgs CO₂ per kg of CH₄ if the needed H₂ is generated from the residual coke.





In the ex-situ method, coal mostly free from physical moisture is fed at the top of the downdraft-type gasifier [10]. Hot hydrogen is fed above the coal column. Superheated steam and oxygen are injected into the coal column near the middle of the coal column. Oxygen is required in small quantities to initiate the partial oxidation of coal into CO gas

[2]. The process diagram is given in Fig. 1. In the methane conversion process, the molar volume of reactants is more than the molar

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volume of product gas methane. Maintaining higher process pressure is conducive to reaction kinetics in the formation of methane but optimum pressure is selected on techno-economic considerations [11]. Residual coke is extracted from the bottom of the gasifier. The product gas/ methane exit from the gasifier is located above the ash extraction point. Process temperature is maintained by controlling the steam input. A regenerative heat exchanger is provided to cool the hot methane gas exiting from the gasifier by the hydrogen and steam that are to be injected into the gasifier for conserving the heat energy to reduce the net hydrogen consumption further. High-temperature nickel alloy material is selected for tubes in the shell and tube heat exchanger to withstand process temperatures up to 870°C. The raw methane is further processed to separate the impurities and pressurized to the end user's CNG requirements. Methane is also liquified into LNG in a methane liquefaction plant.

The in-situ process is like underground coal gasification, but the method resembles shale/tight gas or coal bed methane production. Horizontal drilling in multiple radial directions from a wellhead is carried out to reach a wide area of the coal seam. Hot hydrogen and superheated steam are injected to undergo exothermic synthesis of hydrogen and carbon. Oxygen gas is also made available in small quantities to initiate the partial oxidation of coal into CO gas. The water table in the coal seam area is depressed below the bottom level of the coal seam by injecting air or oxygen into the coal seam which is located below the impervious strata. Hydrogen and oxygen/steam are separately injected through concentric pipes to avoid the occurrence of any fire hazard. The unreacted coal (i.e. residual coke with ash) is a highly porous material without forming huge voids. The left-out carbon/coke is considered sequestered carbon or unused coal. Any unreacted H₂ and CO gases would be associated with the final products methane and CO₂. The in-situ method is shown in Fig. 2.



[Fig.2: In-situ Coal Gasification Using Hydrogen]

There is a possibility of another in-situ method by adopting the underground coal mining method. An underground access gallery is constructed at the edge of the coal seam. Groundwater level is maintained below the coal seam level by pumping out water. Horizontal drilling is done into the coal seam at comfortable spacing. Hot hydrogen, steam, and oxygen at the required pressure are injected into the boreholes through retractable concentrical pipes. Methane along with other gases is extracted from the central pipe. Outgoing hot raw methane gas heats the incoming hydrogen, steam, and oxygen gases to conserve the heat energy and enhance the process efficiency. The raw methane gas is processed to remove the impurities to suit the requirements of end users. In this process, there is no extraction of coal in solid form but in gas/methane form. It is a far less labor-intensive production method.

III. DISCUSSION

In the ex-situ method, biochar or pet coke can also be used in place of coal. When biochar derived from biomass is used, the produced methane is carbon neutral. Hydrogen can also be produced from biomass gasification. The problems encountered in in-situ coal gasification to produce syngas such as unreliable reaction sustainability were not expected as the process in methane production is highly exothermic compared to endothermic reaction in syngas production. The inferior quality coal (low to medium-rank coals with high ash but less sulfur content) available in India is more suitable for methane production economically by in-situ and ex-situ methods.

Presently, the union government of India is offering (excluding taxes) nearly INR 54 (0.62 US\$) per kg of bio-CNG at 250 bar pressure for delivery at the CNG filling stations subject to a maximum 75 km distance from the production point [12]. CNG is sold cheaper than petrol/gasoline to discourage pollution emitting petrol which is a product of imported crude oil. Imported LNG is also used to fuel heavy-duty vehicles and off-the-road construction vehicles to replace pollution emitting diesel fuel which is also a product of imported crude oil. Smaller capacity liquefaction plants of methane are commercially available to produce LNG near the bulk consumption areas and LNG is transported further to nearby fueling stations by road trucks run on LNG boil-off gas [13].

The production cost of methane would be less than the imported cost of LNG (not exceeding US\$ 12 per MMBtu or INR 54 per kg) when hydrogen is produced (below US\$ 10 per kg of H₂) using carbon neutral electricity from solar PV (economical up to INR 9/kWh with pumped hydroelectric energy storage plants or battery energy storage) [14]. Green hydrogen can also be produced economically bv thermochemical conversion of biomass [9]. Methane is a clean fuel as it emits negligible particulate matter when used in internal combustion engines (ICE) since its molecule has no carbon-to-carbon bond [15]. The other pollutants such as No_x, So_x, VOC, and CO emissions are also far less compared to diesel and petrol fuels. CNG or LNG-fueled vehicles excel in the BS-6 (Euro VI) standard related to pollution/emissions [16]. However, methane leakage into the atmosphere leads to global warming 27 to 30 times of CO₂ gas in the first 100 years [17]. Adequate care ought to be taken to prevent methane leakages to air from the production to the

consumption chain. High levels of particulate matter emitted by vehicles (fueled by diesel and petrol) and biomass burning in the

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agriculture fields have been deteriorating the air quality index to hazardous levels particularly during winter months in entire North India due to the accumulation of pollutants at ground level when atmospheric inversions are taking place [18].

Methane is a very good hydrogen carrier at 3.5 times hydrogen gas at STP (0°C at one bar) condition because it is nearly eight times heavier and contains a maximum of 25% hydrogen by weight. The volumetric energy density of LNG (22,200 Kj/liter) is 1.4 times more than methanol (15,800 Kj/liter) [19]. The negative effects of in-situ production of methane from coal deposits, such as gradual land subsidence/degradation, are also applicable to natural gas extraction, underground coal mining, and open-cast coal mining. Groundwater contamination is not expected as there is no injection of chemicals other than hydrogen and oxygen. The groundwater extracted from the coal seam area can be used for agriculture and industrial purposes.

Synthetic natural gas (SNG) which is nothing but methane can also be produced using coal entirely. The heat energy from burning coal is used to split water/steam to provide hydrogen for SNG formation. Nearly two kgs of carbon is required to produce a kg of SNG. The total carbon footprint in the production of one kg of SNG is nearly 7.5 kg of CO₂ [20]. However, SNG production commercially from the high ash and low to medium-rank coal/lignite, which is predominantly available in India, is not established without blending with low ash coal/pet coke [21]. Compared to SNG production at high temperatures exceeding ash fusion temperature (>1050°C), the proposed process operates below 870°C with lesser global warming CO2 gas generation [6].

IV. CONCLUSION

This proposal is an improvement to the established coal gasification technology for methane production to reduce its production cost and carbon footprint. The major difference is that hydrogen is replacing oxygen to provide the necessary energy to the process. The carbon footprint has been reduced comparatively since oxygen is not used for burning the coal. Coal is used as a source of elemental carbon and not fuel. The proposed method/process does not cause any air pollution except CO₂ gas emission if not sequestered. Before the advent of cheaper renewable and carbon-neutral electricity generation from Solar PV and wind power plants, electricity, in high demand, used to be produced from all available fuels like liquid fuels (crude oil), gaseous fuels (natural gas), and solid fuels (coal) including biomass. In addition to catering fully to the ever-increasing electricity needs, renewable solar, and wind electricity potential is so vast to produce the required hydrogen which is essential to produce organic chemicals, urea fertilizer, and transport fuels in case the needed carbon feed/stock is met from fossil coal or non-fossil biomass [22]. Surplus biomass available can also be used for syngas generation economically and syngas can be used further in the production of carbon-neutral fuels (methanol, aviation turbine fuel, etc.) or organic chemicals (ethane, propane, etc). The carbon-neutral CO₂ gas generated from biomass can be sequestered into depleted natural gas and crude oil deposits or underground caverns to brand the produced methane gas, from the coal deposits, as carbon-neutral fuel [9]. Imported liquified petroleum gas (LPG) is also used in large quantities in indoor applications for cooking purposes contributing to substantial particulate matter emissions. Methane produced from coal deposits, by supplying through the available piped natural gas (PNG) network, could replace all the imported LPG in India. CNG and LNG-powered ICE vehicles, hydrogen-powered fuel cell electric vehicles (FCEV), and battery electric vehicles (BEV) could replace all the petrol and diesel vehicles being used in the transport sector including off-road construction, mining, and farming mobile equipment. Methane derived from coal could also be used for meeting peak load power generation by the existing gas turbine power stations to replace imported LNG. The surplus biomass available in India could meet all the hard-to-abate fuels (aviation and marine liquid fuels) and industrial carbon feed/stock requirements by producing adequate organic feedstocks such as methanol, ethylene, etc. as well as providing carbon neutral CO₂ gas for sequestration requirements. In the future, India can avoid the imports of fossil fuels and smoothly transform into an exporter of carbon-neutral organic chemicals, ammonia, urea fertilizer, liquid fuels, etc. by exploiting its vast coal deposits, biomass resources, and solar power potential [23]. Similar energy policies can be followed by other countries that are rich in coal deposits.

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I must verify the accuracy of the following information as the article's author.

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