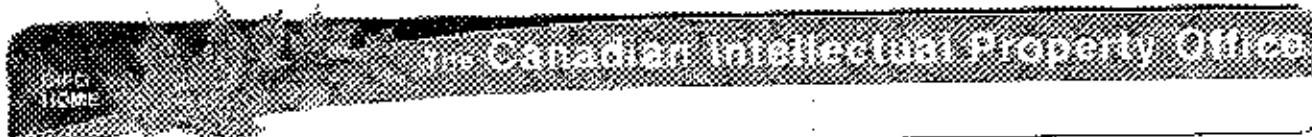


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Strategis Index:

A B C D E F G H I J K L M N O P Q R S T U V W X Y Z

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(12) Patent:

(54) LIQUID HYDROCARBON SYNTHESIZING METHOD

(54) METHODE A SYNTHETISER DES HYDROCARBURES LIQUIDES

More Information:

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ABSTRACT:

CLAIMS: Show all claims

*** Note: Data on abstracts and claims is shown in the official language in which it was submitted.

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Our invention relates to a method of synthesizing liquid hydrocarbons and more particularly to a method in which light hydrocarbon gases such as methane, ethane and the like are converted to a mixture of carbon monoxide and hydrogen which may be synthesized to form liquid hydrocarbons by a catalytic synthesis.

The mixture of carbon monoxide and hydrogen will be referred to as "synthesis gas" for purposes of convenience, and our process relates chiefly to the production of synthesis gas as a subcombination of steps in the broad combination of steps for producing liquid hydrocarbons by catalytic synthesis. It is to be understood that the synthesis gas may be employed for the synthesis of any compounds known to the art which may be synthesized from mixtures of carbon monoxide and hydrogen.

The synthesis gas is preferably a mixture of carbon monoxide and hydrogen having a ratio of two volumes of hydrogen to one of carbon monoxide. This mixture is made from methane, steam and carbon dioxide according to the following reaction:



Methane may be obtained from natural gas wells. Steam may be obtained as a byproduct from the liquid conversion step. It will be obvious, therefore, that the major problem is one of producing the carbon dioxide required for the process. The reaction of converting a mixture of methane, carbon dioxide and steam into synthesis gas takes place at temperatures between 1800° F. and 2200° F. in the presence of a catalyst. Fuel must be burned to achieve these temperatures. The combustion of fuel results in carbon dioxide.

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It is an object of our invention to provide a process in which the carbon dioxide for the reaction is recovered from the flue gases used in heating the catalytic chambers in which the reaction occurs.

5 Another object of our invention is to provide a method of synthesizing hydrocarbons and more particularly synthesis gas, which may be later synthesized to liquid hydrocarbons in a continuous and economical manner.

10 Other and further objects of our invention will appear from the following description:

The accompanying drawing which forms part of the instant specification and is to read in conjunction therewith represents a diagrammatic view of one form of apparatus capable of carrying out the method of our invention.

15 More particularly, referring to the drawing, a light hydrocarbon gas such as methane, for example, obtained from any suitable source, is introduced through line 1. The methane may be under pressure of 400 pounds per square inch and at a temperature in the vicinity of 100° F. It is to be understood that, throughout the specification, particular temperatures and pressures and compositions of gases will be given by way of illustration. These illustrations are not to be construed by way of limitation.

20 The methane passes into an absorber tower 2 which may be a conventional bubble tray tower or a packed tower. The methane is contacted countercurrently in tower 2 with a suitable solvent for removing hydrogen sulphide and other sulphur compounds, which solvent enters the tower 2 through line 3. It is important that sulphur compounds be removed so that in the subsequent conversion of the synthesis gas to liquid hydrocarbons

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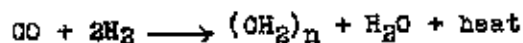
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in the liquid conversion step, the catalyst there will not be poisoned.

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The sulphur compounds in solution in the solvent are withdrawn from the absorber tower 2 through line 4 and pass through heat exchanger 5, through line 6, into the stripping tower 7 in which the sulphur compounds are stripped from the solvent by any suitable means, as for example, a reboiler 8, to which steam is supplied through line 9. The stripper tower may be maintained under a pressure of 10 pounds per square inch and the bottom tower temperature may be maintained approximately 10
225° F. The stripped solution is withdrawn from the stripper tower through line 10 and passed through heat exchanger 5 and then pumped by pump 11 through a cooler 12 in which the absorption menstruum is cooled by heat exchange with water introduced through line 13 to a temperature in the vicinity of 95° F. and 15
is reintroduced into the absorber tower through line 3. The vapors from the stripper tower 7 are removed through pipe 14 and pass through a condenser 15 to which a cooling medium is supplied through a pipe 16. The condensate is removed from the condenser 15 through line 17 and passed into the stripper tower 20
as reflux condensate, it being understood that the stripper tower may be provided with fractionating trays. The sulphur compounds are vented through line 18 and removed from the process. The purified methane, which may contain other light hydrocarbon 25
gases such as ethane and the like, or inert gases such as nitrogen, is withdrawn from the absorber tower 2 through line 19. A portion of the methane passes through line 20 into pipe 21 into which carbon dioxide is introduced through pipe 22. Steam is also introduced into pipe 21 through pipe 23 which is controlled 30
by valve 24 so that the manifold 25 will contain a mixture of

methane, carbon dioxide and steam. The steam may be produced from the liquid conversion step from which it may be withdrawn through pipe 26, controlled by valve 27 and may be under a pressure of 150 pounds per square inch. A pair of catalytic and regenerator chambers 28 and 29 are provided. When one of the chambers is on the make cycle, the other of the chambers is being heated, that is, it may be considered as being on the blast cycle, as will be hereinafter more fully pointed out. In the drawing, chamber 29 is on the make cycle so that valves 30 and 48 will be closed and valve 31 will be opened, permitting the mixture of methane, carbon dioxide and steam to pass through line 32 into the catalyst chamber 29. The catalyst mass in the chamber has been previously heated by the burning of carbonaceous fuel therein to a temperature of the order of 2200° F. The catalyst may be metallic nickel, deposited upon a suitable carrier. The reaction hereinbefore described takes place in the catalyst chamber and the mixture of synthesis gas is withdrawn from the catalyst chamber through pipe 33, it being understood that valve 34 is closed and valve 35 is open. The synthesis gas passes through pipe 36 into the liquid conversion step which is indicated diagrammatically by the reference numeral 37.

This liquid conversion step in which a synthesis gas is converted into liquid hydrocarbons, is described in the art by Fischer and Tropsch and others who have synthesized hydrocarbons from mixtures of carbon monoxide and hydrogen. The reaction takes place in the presence of a catalyst, in the vicinity of 200° C. as follows:



The catalysts used in the above process may be cobalt-thorium-kieselguhr, or nickel manganese alumina kieselguhr or

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cobalt-thorium-copper-kieselguhr mixtures prepared by the reduction of metallic nitrates with hydrogen.

5 The liquid hydrocarbons are withdrawn from the liquid conversion step through pipe 38 which is controlled by valve 39. A gas holder 40 is provided to control the flow to the liquid conversion step. When too much synthesis gas is being made, the excess is diverted through pipe 41 which is controlled by valve 42 into the gas holder 40. When not enough synthesis gas is being made, the deficiency is supplied by removing gas from the holder by blower 43 which passes gas through pipe 44 into the pipe 38 for passage to the liquid conversion step, the flow of synthesis gas being controlled by valve 45. A heat exchanger 36a is positioned in the line 38 to maintain the desired temperature of the gas entering the liquid conversion step 37, which exchange may be either a heater or a cooler.

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brick or catalyst might be damaged. By distributing the heating points, the desired temperature of 2200° F. is achieved.

5 Besides methane, other carbonaceous fuels such as pulverized coal or coke or liquid fuels may be used in the blast cycle. In burning such materials provision would be made for properly introducing the fuels to the chambers.

10 The valves 63 and 64 are so controlled that the products of combustion are cooled to around 100° F. At this temperature most of the steam of the gases of combustion will be condensed and will be withdrawn from the drum 60, together with the cooling water through line 65. The cooled gases leave the drum through line 66 and pass into an absorber 67 which may be a packed or bubble tray absorber similar to the absorber 3. The
15 gases of combustion are contacted countercurrently with a suitable solvent for carbon dioxide, which is introduced into the absorber through pipe 68. The absorber may be operated under a pressure of 25 pounds per square inch. The solution of carbon dioxide in the solvent or absorbing menstruum is withdrawn from the absorber through pipe 69 and is pumped by pump 70 through pipe 71 through
20 heat exchanger 72, through pipe 73 into the stripping tower 74. The stripping tower is operated to maintain a bottom temperature in the vicinity of 235° F. by means of a reboiler 75 to which steam is supplied through pipe 76. The stripped carbon dioxide gas is withdrawn from the stripper through pipe 77 and passed
25 through a condenser 78 to which a cooling medium is supplied through pipe 79. The condensibles are returned to the stripper through pipe 80 as reflux. It is understood that the stripper may be provided with fractionating trays. The carbon dioxide gas is withdrawn from the condenser through pipe 81 and is passed
30 into pipe 82 for passage into the manifold 21 as hereinabove described. A gas holder 82 is provided for the control of the

carbon dioxide stream. When an excess of carbon dioxide is being made, it will pass through pipe 83 controlled by valve 84 into the gas holder 82. When an insufficient quantity of carbon dioxide is being recovered, carbon dioxide is pumped from the gas holder by blower 85 and passed through line 86 into pipe 22 for passage to the catalyst chambers.

The gases of combustion will contain nitrogen, some carbon monoxide and other gases. These are not absorbed by the selective solvent for carbon dioxide and pass from the absorber tower 67 through pipe 87.

The stripped carbon dioxide solvent leaves the stripper through pipe 88, passes through heat exchanger 72 and is pumped by pump 89 through the cooler 90, through line 89, back into the absorber for reuse. A cooling medium is supplied to the cooler through line 91 and serves to reduce the temperature of the absorbing menstruum to about 90° F.

In the drawings, chamber 28 is on the blast cycle and chamber 29 is on the make cycle. When the average temperature of the catalyst mass in chamber 29 drops to about 1800° F. as can be readily determined by pyrometers, thermometers and other temperature recording instruments known to the art, valves 32, 35, 49, 31, and 58 are closed and valves 93, 34, 47, 48, and 30 are opened. This transfers catalyst chamber 29 to the blast cycle and catalyst chamber 28 to the make cycle.

When the changing of valves has been completed, methane gas will flow through the valve 47 and pipe 94 into a manifold 95 and branch pipes 96, 97 and 98 into the chamber 29 for combustion with air furnished by blower 55 through pipe 29, controlled by valve 34, air passing into the catalyst chamber 29 through pipe 33. The products of combustion will leave the

chamber 28 through the pipe 32 and pipe 100, valve 30 being opened for passage into the pipe 59 to the carbon dioxide absorption system. The mixture of methane, carbon dioxide and steam in pipe 25 will pass through pipe 101, controlled by valve 48 into pipe 57 into the catalyst chamber 28, while the synthesis gas formed in the catalyst chamber will be withdrawn through pipe 102, controlled by valve 93 into the pipe 36 for passage to the liquid conversion step or gas holder, or both.

The flow of gases through the catalyst chamber on the make cycle and the flow of methane to the chamber on the blast cycle are so controlled that the heating to the temperature of about 2200° F. of the catalyst mass in the chamber on the blast cycle and the drop in temperature to about 1800° F. of the catalyst mass in the chamber on the make cycle are coextensive so that the process may be operated continuously by alternately shifting one catalyst chamber from make cycle to blast cycle and vice versa.

It will be observed that we have accomplished the objects of our invention. We have provided a novel and expeditious manner of synthesizing liquid hydrocarbons from normally gaseous hydrocarbons including a novel step combination involving the production of synthesis gas from methane, steam, and carbon dioxide in which the carbon dioxide is economically recovered from the gases used in supplying heat for the production of the synthesis gases.

It will be understood that certain features and sub-combinations are of utility and may be employed without reference to other features and sub-combinations. This is contemplated by and is within the scope of our claims. It is further obvious that various changes may be made in details within the

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scope of our claims without departing from the spirit of our invention. It is, therefore, to be understood that our invention is not to be limited to the specific details shown and described.

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Having thus described our invention, what we claim is:

1. A method of converting hydrocarbon gases into liquid hydrocarbons by synthesis of synthesis gas comprising carbon monoxide and hydrogen, said synthesis gas produced in catalytic contacting zones operated alternately on blast and make cycles, the blast cycle comprising burning carbonaceous material with air to raise the temperature between 2200° F. and 1800° F., the make cycle comprising subjecting a mixture of hydrocarbon gas, steam and carbon dioxide to catalytic conversion between temperatures of 2200° F. and 1800° F. in the presence of a catalyst, said carbon dioxide being recovered from the blast cycle, and converting the synthesis gas into liquid hydrocarbons.

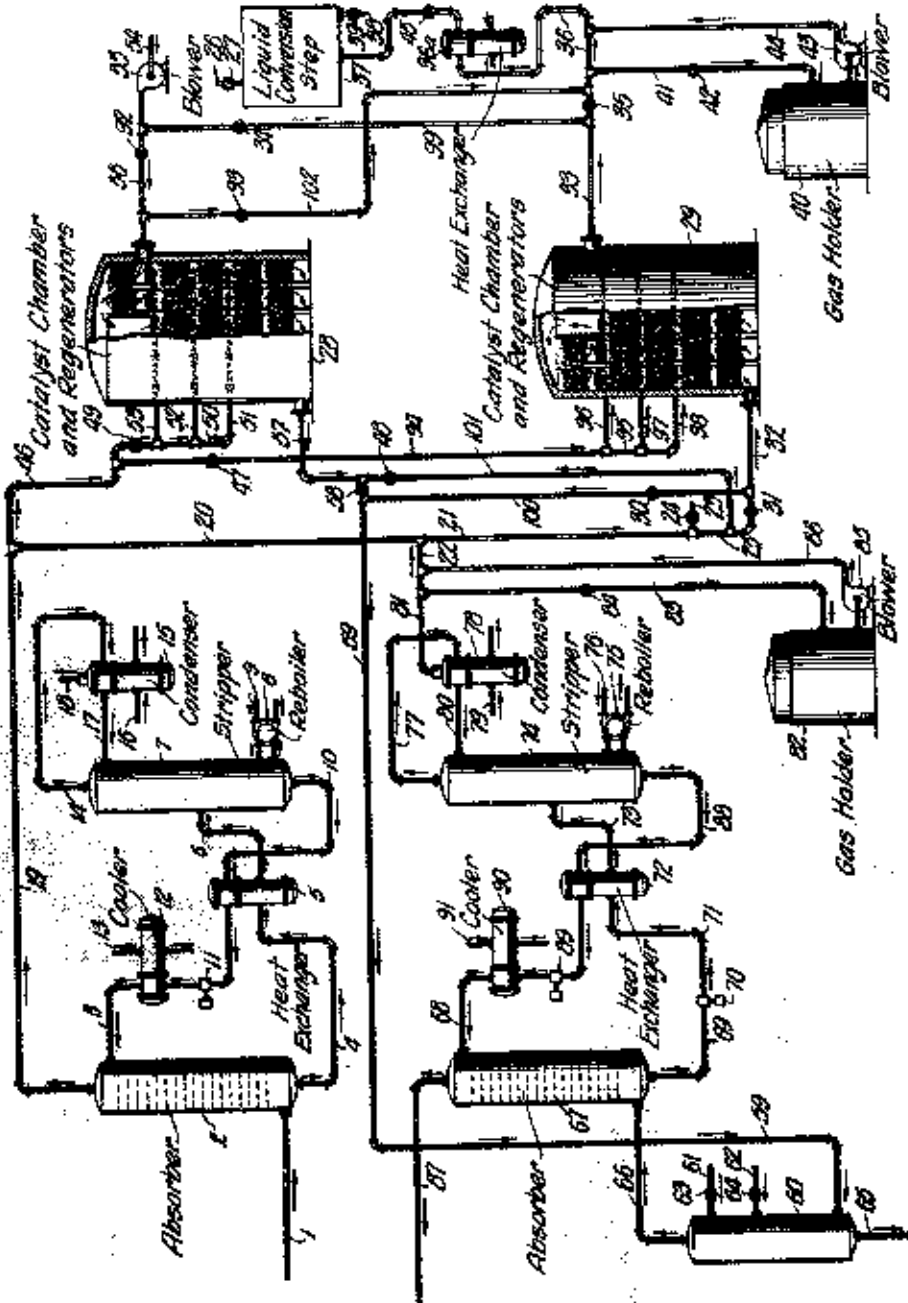
2. In a method of converting hydrocarbon gases into liquid hydrocarbons including the steps of burning hydrocarbons in a first catalyst contact zone to raise the same to a temperature between 1800° F. and 3200° F., recovering carbon dioxide from the products of combustion, forming a mixture of carbon dioxide, hydrocarbon gas and steam, subjecting said mixture to catalytic conversion into synthesis gas comprising carbon monoxide and hydrogen in a second catalytic contact zone between the temperatures of 2200° F. and 1800° F., periodically reversing the zones in which burning of hydrocarbons and conversion of hydrocarbon gas to synthesis gas takes place, and converting said synthesis gas into liquid hydrocarbons in the presence of a catalyst at elevated temperature.

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3. A method as in claim 2 in which said hydrocarbon gas is subjected to the action of a hydrogen sulphide selective solvent prior to use in the process.

4. A method as in claim 3 in which said carbon dioxide is recovered from the products of combustion by absorption with a selective solvent.

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 and Dustin W. Wilson.

INVENTOR

BY

Smart & Biggar
 ATTORNEY

Certified to be the drawing referred to
 in the specification hereunto annexed.

Ottawa, Ontario, Canada, October 25th, 1938.