



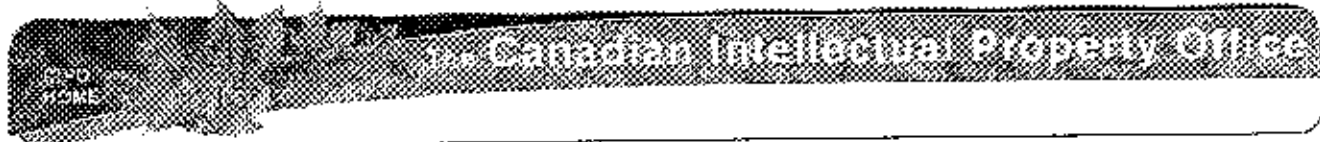
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(54) MANUFACTURE OF HYDROCARBON OILS

(54) FABRICATION D'HUILES D'HYDROCARBURES

Information Table

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ABSTRACT:

CLAIMS: Show all claims

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The present invention relates to the novel features disclosed in the following specification and claims considered in connection with the accompanying drawing.

The synthesis of hydrocarbons from carbon monoxide and hydrogen in the presence of a suitable catalyst is broadly old. However, a great deal of research work is now being directed to this field by many public and private interests for the purpose of developing a source of hydrocarbon oils which, by such treatment, may be converted or transformed into valuable commercial products such as motor gasoline, heating oil, and the like.

The gas used for the synthesis of hydrocarbons, according to the method above indicated, consists of a substantially equimolar mixture of carbon monoxide and hydrogen. Depending, however, somewhat on the conditions of temperature, pressure, contact time, catalyst, and the product desired, the ratio of these components may vary somewhat between 1:1 and 1:2 parts of CO per part of hydrogen by volume or mol. Such a gaseous mixture can be prepared from almost any carbonaceous material by suitable means; for example, coal, petroleum, coke, pitch, lignite, natural gas, or the gases from an oil refinery may be used as the source of carbon. Where the starting material is deficient in hydrogen, the gas is usually prepared by the action of steam on hot coke or other heated carbonaceous material, the steam thus supplying the necessary hydrogen. In the case of natural gas, however, which is largely methane, there is sufficient hydrogen present to form the synthesis mixture and consequently, in theory, no steam should be required if the hydrogen is not consumed in undesired side reactions.

One method of producing synthesis gas from natural gas which has been previously proposed consists of burning natural gas in the

presence of a limited supply of oxygen. The combustion supplies the necessary heat and the lack of sufficient oxygen to oxidize all of the carbon induces the formation of CO. Steam may be added as necessary to provide sufficient hydrogen. While such a method will produce CO and hydrogen, it has the disadvantage that where air is used the nitrogen content thereof dilutes the synthesis gas for of course there is about four times as much nitrogen as oxygen in ordinary air. The presence of the undesired nitrogen complicates the purification problem and increases the gas handling facilities required. In a number of cases where this problem has arisen it has been found more economical to use pure oxygen to burn the natural gas in place of air.

It is the main object of our invention to provide a stoichiometric mixture of carbon monoxide and hydrogen suitably proportioned to serve as a feed gas in the synthesis of hydrocarbons, in a manner which is cheaper and more expeditious than those previously employed.

A specific object of our invention is to produce a hydrocarbon synthesis feed gas from natural gas in a process in which the feed gas is obtained substantially undiluted with undesired components.

As a corollary to the next preceding object, it is a still further object of our invention to produce synthesis gas suitable for feed stock in the manufacture of hydrocarbons in a continuous operation which provides a mixture of CO and hydrogen substantially undiluted with undesired components and, at the same time, a substantially pure nitrogen gas.

In the accompanying drawing, we have shown diagrammatically the essential apparatus elements which illustrate a preferred embodiment of our invention.

Referring in detail to the drawing, in carrying out invention into effect we provide a natural gas burning reactor (1) and a catalyst regenerator (3). In manufacturing the CO-hydrogen mixture from natural

gas we cause the oxidation of the natural gas by means of a reducible oxide which releases its oxygen for that purpose. We prefer to carry out this reaction while the reducible oxide in the form of a powder is suspended in a dense phase suspension in the gas being oxidized or burned. Thus, to reactor (1) we charge natural gas via line (10) containing suspended therein powdered iron oxide, for example, Fe_2O_3 , into the bottom of reactor (1), thence the suspension passes upwardly through a grid (6) into the main body of the reactor (1). The iron oxide is in subdivided form consisting of particles smaller than about 100 mesh, preferably around 400 mesh, and for each cubic foot of methane or natural gas or equivalent converted in the reactor (1) (measured under standard conditions of temperature and pressure) we charge suspended in the gas from 0.10 to 0.20 lbs. of iron equivalent. Within the reactor (1), the gas velocity is controlled within the limits of from 1/4 to 5 ft. per second, preferably 1 to 3 ft. per second whereby we form a dense, turbulent, fluidized mass of iron oxide in gas. By maintaining a temperature within the range of 1600° to 2000°F., the iron oxide releases its oxygen to the methane or natural gas causing the production of CO and hydrogen with a consequent reduction of the iron oxide. The carbon monoxide and hydrogen is withdrawn overhead through line (15) and delivered to a storage (not shown). The reduced iron, on the other hand, is withdrawn through a standpipe (16) carrying a plurality of taps (17) into which may be injected steam or methane or natural gas for the purpose of increasing the fluidity of the iron in the standpipe thus obviating bridging or plugging of the said pipe. The flow of iron in the standpipe is controlled by a valve (18). The reduced iron discharges into an air line (20) where it is picked up by a current of air to form a suspension and transported to oxidation zone (3). The suspension passes upwardly through the grid (6) therein disposed in the main body of the oxidation or burning zone where the flow conditions of the gasiform material are fixed within the limits set forth in reactor (1), namely, from 1/4 to

5 ft. per second, preferably 1-3 ft. per second. This rate of flow causes the formation of a dense, turbulent suspension or mass of catalyst in gasiform material. By carefully controlling the quantity of air, the iron may be oxidized, for example, to Fe_2O_3 , with substantially complete consumption of the oxygen contained in the air so that the gasiform material exiting via line (25) is substantially pure nitrogen. Of course this nitrogen in pure state is a valuable material for it may be used with hydrogen in the synthesis of ammonia from its elements.

The nitrogen gas is delivered to storage (not shown). The temperature prevailing in the burning zone or reoxidizing zone (3) is of the order of 1200°F. or higher with a temperature of, say, 1600°F. being preferred.

We have thus described the essential features of our invention, omitting, however, for clarity a description of conventional means and methods for improving the process, such as the use of flow meters, pumps, and the like. It should be pointed out, however, that the net overall heat of reaction, which is exothermic in character in the reoxidation zone and endothermic in the reaction zone, is about 66 Btu's per cubic foot of methane or its equivalent. This quantity of heat is not sufficient for heating the air and gas to reaction temperatures, and therefore it is necessary to supply this by heat exchange with the product gases in lines (15) and (25) and/or by burning gas in the reoxidation chamber (3) or by both or any of the well-known means of supplying heat. This, however, is an expedient which the skilled engineer can be relied upon to include in his commercial design, and it is not necessary to illustrate and describe these known engineering principles herein for they do not go to the heart of this invention. However, it is pointed out that where iron oxide is used as the oxidizing material, it is advisable to add or include therein 1-20% nickel oxide. Also, instead of using iron oxide, we may use molybdenum oxide, chromium oxide, tungsten oxide, cobalt oxide, or nickel oxide, but

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as indicated we prefer to use iron oxide promoted with nickel oxide. For the purpose of simplicity and explanation, we have stated that the iron withdrawn from the burning zone (1) is reoxidized in zone (3) to Fe_2O_4 . Actually the iron oxide withdrawn via standpipe (30) may be a mixture of several oxides. In whatever state of oxidation the oxide may exist as it is withdrawn through line (30) it is discharged with natural gas and conveyed to the burning zone (1) as previously described. Standpipe (30) carries a plurality of taps (32) into which natural gas may be discharged for the purpose of increasing the fluidity of the iron passing therethrough. Also, standpipe (30) is provided with a flow control valve (33) by manipulation of which the quantity of iron oxide discharged into the gas stream may be controlled.

Numerous modifications of our invention falling within the scope thereof may be made by those familiar with this art without departing from the spirit thereof.

What we claim is:

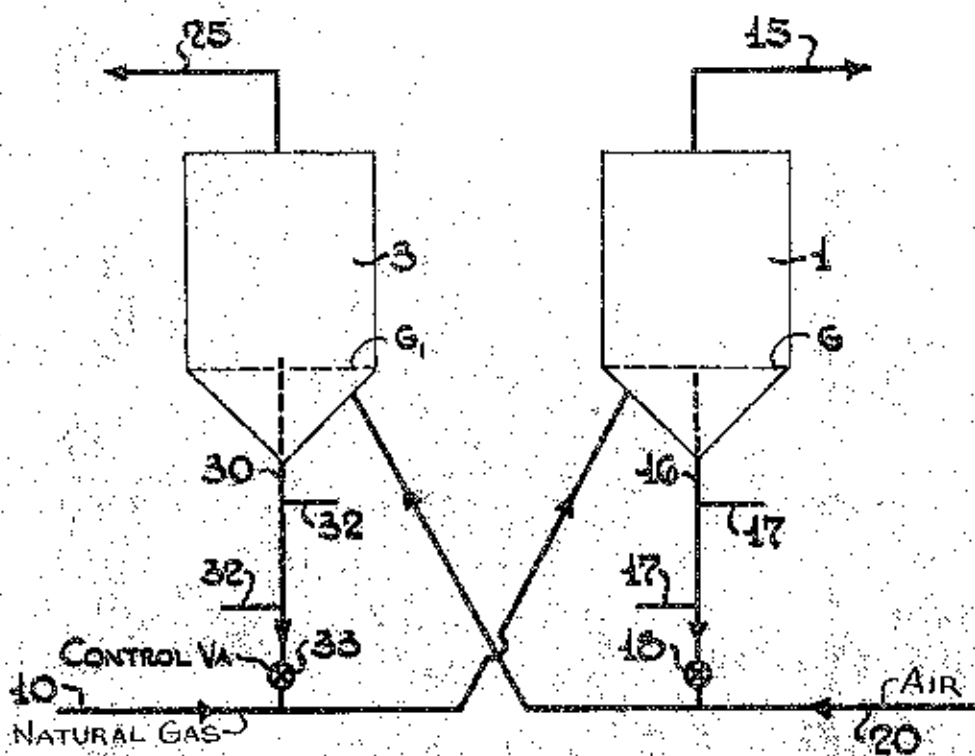
1. A continuous method for producing a mixture of substantially undiluted CO and hydrogen which comprises suspending in a gas predominantly, methane, a quantity of a heavy metal reducible oxide in powdered form, transporting the suspension to a reaction zone, causing the gas to flow upwardly in this reaction zone at a rate of from about 1/2 to 5 ft. per second thereby causing the formation of a dense, turbulent, fluidized mass of a heavy metal oxide in gas, fixing the temperature within the zone within the limits of 1200°F. to 2000°F., permitting the gas to remain resident in the burning zone for a sufficient period of time to cause substantial quantities of its methane content to be converted to CO and hydrogen, recovering a mixture of CO and hydrogen from the said burning zone as product, withdrawing the reducible oxide by gravity in reduced condition from the burning zone, suspending the reduced oxide in an oxygen-containing gas, transporting the suspension to a reoxidizing zone, causing the oxygen-containing gas to flow upwardly in the reoxidizing zone at a rate of from about 1/2 to 5 ft. per second thereby causing the formation of a dense, turbulent, fluidized mass of heavy metal in oxygen-containing gas, fixing the temperature in the reoxidizing zone sufficiently high to cause reoxidation of the heavy metal, withdrawing the heavy metal in reoxidized condition from the reoxidizing zone, and resuspending the reoxidized metal in a further quantity of gas, predominantly methane, for transport to the said burning zone.

2. The method of claim 1 in which the heavy metal oxide is iron oxide.

3. The method of claim 1 in which the heavy metal oxide is iron oxide admixed with nickel oxide.

4. The method of claim 1 in which the feed gas to the burning zone is natural gas.

5. The method of claim 1 in which a quantity of combustible gas is fed to the recirculating zone in order to add available heat to the system.



Certified to be the drawings referred to
in the specification herewith annexed.

Ottawa, Jan. 23, 1946

Standard Oil Development
Company

Thorp & Selck

ATTORNEYS