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PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Process and Apparatus for the Production of Hydrocarbons by Reduction of Carbon Monoxide with Hydrogen

I, HAROLD EDWIN POTTS, Chartered Patent Agent, of 12, Church Street, Liverpool, in the County of Lancaster, Subject of the King of Great Britain, do hereby declare the nature of this invention, which has been communicated to me by N. V. INTERNATIONALE KOOLWATERSTOFFEN SYNTHESE MAATSCHAPPIJ (INTERNATIONAL HYDROCARBON SYNTHESIS COMPANY), of 34, Raamweg, The Hague, Holland, a Dutch Company, and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The present invention relates to a process and apparatus for the production of hydrocarbons with more than one carbon atom in the molecule by reduction of carbon monoxide with hydrogen.

In known processes and apparatus of this type the initial gases are passed over a catalyst, for example arranged in tubes, at elevated temperatures and it is advantageous to maintain the temperature within narrow limits. In the industrial operation of the process the heat generated by the reaction is generally removed from the reaction space through metal walls having a good heat conductivity, by indirect heat exchange with water boiling under a suitably elevated pressure.

It has now been found that the said process can be carried out in an advantageous manner when employing reaction tubes having a length of more than 5 meters, preferably a length of between 6 and 10 metres surrounded by a boiling cooling medium, and especially in tubes in which there is a vertical distance of at least 5 metres between inlet and outlet.

When compared with the method of working in shorter tubes at equal specific throughput, i.e. at equal time of contact of the reaction gases, the present invention enables the gases to be passed through the catalyst tubes at a higher linear velocity, thereby effecting a better transmission of the reaction heat to the cooling wall. The process according to the present invention also has a beneficial effect on the lifetime and the production capacity of the catalyst.

The apparatus may be constructed in various manners and the present invention is not limited to any particular type of apparatus. Preferably a plurality of tubes are combined in one apparatus immersed in the same cooling medium. Each tube may also be surrounded by a separate cooling space or contain an internal cooling space at its inner side, for example, in the form of an internal concentric tube opening into the cooling medium leaving the catalyst space as an annular column between the two tubes.

It is particularly advantageous to work in tubes which have been vertically arranged and to pass the gas to be converted in downward direction through the tubes. In this case the boiling cooling medium will show a small and gradual increase in the temperature in the direction of flow of the gas to be converted. This increase in temperature is caused by the fact that the cooling medium in the lower part of the cooling space is under a higher pressure as a result of the pressure exercised by the column of liquid. This increase in temperature compensates for the decrease in reaction velocity caused by the dilution of the gases to be converted with the desired final products as well as with water vapour, methane and carbon dioxide.

When employing, for example, water as the cooling medium and maintaining a pressure of 9 atmospheres in the vapour space of the cooling chamber, then the temperature of the boiling water at the upper end of the catalyst space will be 174.4° Centigrade if the water level in the cooling space is not substantially thereabove. When working in a reaction tube of a vertical height of 5 metres the pressure at the lower end of the cooling space will be 9.5 atmospheres and the temperature 176.7° Centigrade, thus 2.3° Centigrade higher than at the upper end. If a reaction tube of a vertical height of 10 metres is used, then at a pressure of 9 atmospheres in the vapour space of the cooling medium, the temperature at the exit of the reaction tube will be 178.9° Centigrade, i.e. 4.5° C. higher than at the upper end.

Instead of water also diphenyl, glyc-

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Price 3s

erine, paraffin-oil and tricresyl phosphate may be used as cooling medium. In this way it is possible to obtain a temperature gradient which (considering all other operating conditions as pressure, nature of the catalyst, velocity, heat transfer through the tube walls, etc.) in spite of increasing concentration of the inert material is most favourable for the maintenance of a constant conversion. The boiling point of diphenyl under ordinary pressure is 255° Centigrade and under a pressure of 6 atmospheres 340° Centigrade. An increase in pressure of 5 atmospheres therefore corresponds to an increase in boiling temperature of 85°C. so that when employing a reaction tube of a vertical height of, for instance, 10 metres, a difference in temperature exists between the upper and lower end of the reaction vessel of about 20°C. The difference in temperature between inlet and exit of the reaction space can thus be varied by the choice of a suitable cooling medium.

The temperatures at which the reduction of carbon monoxide with hydrogen is carried out lie in general between 160 and 220° Centigrade, preferable between 175° and 200° C., however, also higher temperatures up to 350°C. or thereabove may be employed. Preferably pressures above 5 atmospheres and up to 20 atmospheres are employed, although also higher or lower pressures may be employed.

The following Example will further illustrate how the invention is carried out in practice but it should be understood that the invention is not limited to the said Example.

EXAMPLE

When passing a mixture of carbon monoxide and hydrogen at a rate of 100 cubic metres per hour per cubic metre of catalyst space and at a temperature of 195°C. through a tube having a length of 2 metres, the yield drops from an initial value of 93 grams of liquid hydrocarbons per cubic metre of fresh gas to 80 grams after 30 days.

When working on the other hand in a tube having a length of 10 metres under otherwise similar conditions then the yield which at the beginning is also 93 grams of liquid hydrocarbons decreases to a lesser extent and amounts of 85 grams of liquid hydrocarbons per cubic metre of fresh gas after 30 days. In both cases a catalyst containing 46 per cent of cobalt, 8 per cent of thorium oxide and 46 per cent of kieselguhr is employed.

Having now particularly described and ascertained the nature of the said invention and in what manner the same is to be performed, as communicated to me by my foreign correspondents, I declare that what I claim is:—

1. Process for the production of hydrocarbons with more than one carbon atom in the molecule by reduction of carbon monoxide with hydrogen in the presence of catalysts contained in tubes which are surrounded by a boiling cooling medium in which the tubes have a length of more than 5 metres.

2. Process as claimed in claim 1 wherein the tubes in which the process is carried out are so arranged that the vertical distance between the inlet and outlet openings is at least five metres and the gases to be converted are passed in downward direction through the tubes.

3. A process as claimed in claim 2 wherein the tubes are disposed vertically and have a length of more than five metres.

4. Process as claimed in claims 1 and 2, in which tubes are employed having a length of between 6 and 10 metres.

5. Process as claimed in claims 1 and 2 in which boiling water is employed as cooling medium.

6. Apparatus for the production of hydrocarbons with more than one carbon atom in the molecule comprising tubes to contain catalysts and provided with means to surround them by a boiling cooling medium in which the tubes have a length of more than 5 metres as particularly described.

7. Hydrocarbons with more than one carbon atom in the molecule when obtained in a process in accordance with any of claims 1 to 4.

Dated this 6th day of May, 1941.

W. H. THOMPSON & CO.,
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12, Church Street, Liverpool, 1.

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