PATENT SPECIFICATION



Date of Application and filing Complete Specification: June 2, 1948.

Application made in Netherlands on June 6, 1947.

Complete Specification Published: May 24, 1950.

Index at acceptance:—Class 1(i), F3c4.

COMPLETE SPECIFICATION

A Process for the Catalytic Production of Carbon Monoxide and Hydrogen from Hydrocarbons and Steam

We, N. V. DE BATAAFSCHE PETROLEUM MAATSCHAPPIJ, a Body Corporate organised under the laws of The Netherlands, of 30 Carel van Bylandtlaan, The Hague, 5 Holland, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the

following statement:-This invention relates to a process for the catalytic production of carbon monoxide and hydrogen from hydrocarbons

and steam.

Hitherto the conversion of methane with 15 steam into carbon monoxide and hydrogen has been carried out in practice in a tube reactor containing the catalyst in a fixed bed. The catalyst used is, for example, nickel on magnesite. The reaction is pre20 ferably carried out at atmospheric pressure. The temperature at which the conversion takes place amounts to about 600 to 700° C.

The reaction is strongly endothermic, so 25 that 55 kg cal. of heat per mol. of methane

have to be supplied.

In order to prevent superheating of the wall of the reactor it is necessary—in view of the relatively low heat transmis-30 sion co-efficient between reactor wall and gas flowing through—to use a tube reactor. In addition the temperature changes vary considerably in an axial direction in the reactor tubes. Depending upon the con-struction of the oven this change is from 300 to 700—750° C., which is to be con-sidered unfavourable for the reactor from the point of view of construction. These two drawbacks can be met to a

40 considerable extent according to the invention by carrying out the conversion in the presence of a catalyst in the fluidised state.

By this is understood a catalyst which is kept by the ascending gas stream in such 45 a state of suspension that a dense phase is obtained with a density of the order of 400 to 800 kg per cubic metre, depending

upon the specific gravity of the solid mass and on the linear gas velocity. The particle size used will preferably be of 10 to 50 200 microns.

When applying the process according to the invention the temperature change in the axial direction is neutralised practically completely. Moreover, the heat trans- 56 mission co-efficient between reactor wall and gas is raised considerably, so that the tube reactor can be replaced by a single converter of substantially larger diameter.

The process according to the invention 60 may be carried out in various ways. The catalyst may, for example, be applied in the form of a stationary or substantially stationary bed of fluidised particles. When the activity of the catalyst has 65 decreased down to a given point, it can be restored by a regeneration with steam, which can also be carried out while keeping the particles in a fluidised state.

It is also possible continuously to withdraw a portion of the catalyst mass from the reactor and to recycle it after heating to the desired temperature. The heat required for the reaction is thus trans- 75 mitted directly to the reacting gases. With this embodiment the indirect supply of heat may be omitted. It will then at most be necessary to take measures in order to prevent radiation losses, for 80 example by insulating the wall of the converter or by passing the hot gases discharged from the oven in which the catalyst is heated round the walls of the converter.

A special advantage of the process according to the invention consists in the possibility of converting with steam not only methane, but also higher paraffin hydrocarbons, such as those with at least 90 four carbon atoms in the molecule, further olefinic hydrocarbons and in principle hydrocarbons generally. In the known process these base materials cannot

be applied, because when the temperature rises from room temperature to a reaction temperature of 700° C. or higher, the material passes through a range in which considerable deposition of carbonaceous material takes place and in which the temperature is not yet high enough to convert this carbonaceous material with steam. With the process of the present invention,

10 on the contrary, the base material need not be heated to within this critical range, but the final heating may be effected in the fluidised bed itself. Owing to the uniformly high temperature in the fluid-15 ised bed no carbon deposition will occur

and/or any carbon deposits will at once

react with the steam present.

Having now particularly described and ascertained the nature of our said inven-20 tion and in what manner the same is to be performed, we declare that what we claim is:-

1. A process for the production of carbon monoxide and hydrogen by the conversion of hydrocarbons with the aid of steam in the presence of catalysts, wherein the conversion takes place in the presence

of a finely divided catalyst in the. fluidised state.

2. A process as claimed in claim 1, 30 wherein the catalyst is employed in the form of a stationary or substantially stationary bed of fluidised particles and wherein heat is supplied from the outside.

3. A process as claimed in claim 1, 35 wherein a portion of the catalyst mass is continuously withdrawn from the reactor and recycled after heating to the desired temperature.

4. A process as claimed in claim 3, 40 wherein the gases which have served to heat the catalyst mass are passed round the walls of the reactor in order to minimise radiation losses.

5. Carbon monoxide and hyrogen when 45 produced by the process claimed in any one of the preceding claims.

Dated the 2nd day of June, 1948. ELKINGTON & FIFE, Consulting Chemist and Chartered Patent Agents, Bank Chambers, 329 High Holborn, London, W.C.1, Agents for the Applicants.

THE RESERVE OF THE PARTY OF THE

Learnington Spa: Printed for His Majesty's Stationery Office by the Courier Press .-- 1950 Published at The Patent Office, 25, Southampton Buildings, London, W.C.2, from which copies, price 2s. 0d. each (inland) 2s. 1d. (abroad) may be obtained