PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Synthesis Reactor for the Hydrogenation of Carbon Monoxide in a Liquid Medium

We, RHEINPREUSSEN AKTIENGESELLSCHAFT FUER BERGBAU UND CHEMIE, of Homberg, Niederrhein, Germany, a German Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention relates to a reactor for the synthesis of hydrocarbons by the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, and to a process for the production of hydrocarbons by the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium.

The dissipation of the heat evolved in the catalytic hydrogenation of carbon monoxide has probably always been the most difficult technical problem in this synthesis. Of all the known methods of carrying out the hydrogenation of carbon monoxide, the most favourable conditions for dissipation of the heat of reaction are probably provided when the synthesis is carried out in a liquid medium. However, even in this case, overheating may occur when very active catalysts and high space velocities of the synthesis gas are used. The increase in temperature generally results in a large increase in the formation of methane and the separation of carbon, with consequent reduction in the yield of C_2 and higher hydrocarbons and a shortening of the active life of the catalyst.

It has now been found that this overheating and, more particularly, the results thereof may be avoided if the cooling area of the cooling elements is reduced, continuously or in stages, in the upward direction.

According to the invention, a process for the production of hydrocarbons by the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium comprises cooling the catalyst suspension by means of cooling surfaces disposed within the suspension, the area of the cooling surfaces being reduced in the direction of flow of the synthesis gas. The area of the cooling surfaces may be reduced continuously or in stages.

According to the invention furthermore, a synthesis reactor for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium through which the reactants are passed in upward flow, includes a series of tubes disposed within the reactor through which tubes a cooling medium may be passed, the tubes being decreased in number in the upward direction whereby the total outer surface area of the tubes decreases in the same direction. The total outer surface area of the tubes may decrease continuously or in stages.

The tubes may be provided as a number of short cooling sets or bundles. It is preferred, however, to provide the tubes substantially vertically in the reactor and to combine together two or more of the tubes into a smaller number of tubes whereby the total outer surface of the tubes decreases in the upward direction.

In the accompanying drawings, two reactors are diagrammatically illustrated by way of example, Figure 1 showing a vertical section through a known kind of reactor, and Figure 2 showing a vertical section through a reactor according to the invention.

The two reactors, except for their cooling systems, are similar and comprise a shell 10, an inlet tube 11 for the synthesis gas, an outlet tube 12 for the gaseous products of the synthesis, and an inlet pipe 13 through which a catalyst and a liquid medium are introduced into the reaction space 14. The lower boundary of the reaction space 14 is defined by a diffusion plate 15 or the like through which the synthesis gas passes from the inlet tube 11 into contact with the catalyst in the liquid medium in the reaction space 14. The

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catalyst is finely-divided so that it is readily maintained in suspension in the liquid medium during the course of the synthesis.

The known form of cooling system illustrated in the reactor in Figure 1, comprises three bundles of cooling tubes 16. The lower ends of each bundle of cooling tubes are interconnected by a lower header 17 whilst the upper ends of the cooling tubes in a bundle are interconnected by an upper header 18. Each of the three headers 18 is connected by a tube 19 to a vapour drum 20. Each of the cooling tubes 16 extends from its lower header 17 to its upper header 18.

The cooling system provided according to the invention in the reactor illustrated in Figure 2, also comprises three bundles of cooling tubes 21, the lower ends of the cooling tubes in one bundle being interconnected by a lower header 22, whilst the upper ends of those tubes in each bundle which extend into the upper part of the reaction space 14, are interconnected by an upper header 23. Each of the three upper headers 23 is connected through a pipe 24 with a vapour drum 25.

In an upward direction, the area of the cooling surfaces provided by the cooling tubes 21 is reduced in three stages by combining a number of the tubes into a smaller number of tubes. Thus, for example, the number of cooling tubes in one bundle connected to the corresponding lower header 22 may be 120. At the level A, the 120 cooling tubes are reduced to 90 for example, by merging 60 of the 120 cooling tubes into 30 cooling tubes. At the level B, the number of the cooling tubes in each bundle is again reduced, for example, from 90 to 60, by merging 60 of the 90 cooling tubes into 30 cooling tubes. The 60 cooling tubes present in each bundle above the level B are reduced in number at the level C by merging together the 60 tubes to form 30 tubes. Thus the area of the cooling surfaces provided by the cooling tubes 21 is reduced in an upward direction to provide four cooling zones, the area of the cooling surfaces being different in each zone.

The invention is illustrated with reference to the two following comparative experiments, in which a known reactor was employed in the first experiment (Run A) and a reactor according to the invention was used in the second experiment (Run B).

RUN A.

A vertical synthesis reactor of the kind illustrated in Figure 1, had a height of 12 metres and a diameter of 1.40 metres, and contained 360 water-cooled tubes, and was used in the hydrogenation of carbon monoxide with a synthesis gas rich in carbon monoxide, the composition of which is set out in the Table hereinafter given. The catalyst employed was an iron catalyst used as a 10% suspension (10% Fe) in a hydrocarbon fraction having an initial boiling point higher than 250° C. The iron catalyst was of the kind which yields predominantly low-boiling products, 80% to 85% of the C_{3+} hydrocarbons (that is, hydrocarbons containing not less than 3 carbon atoms in the molecule), boiling below 200° C. A CO conversion of more than 90% with a temperature of 267° C in the cooling system was obtained, as will be noted from the end gas analysis A given in the Table. The yield was approximately 400 kilograms of hydrocarbons per kilogram Fe in the catalyst.

RUN B.

The synthesis in this run was effected in accordance with the invention. The reactor was of the same height and diameter as that employed in Run A, but the cooling system was provided in the manner illustrated in Figure 2. The number of water-cooled tubes in the lower part of the reactor was 360. At a level approximately 3 metres above the entry of the synthesis gas into the catalyst suspension, the number of cooling tubes was reduced to 270, after a further 3 metres the number of tubes was reduced to 180, and finally after a further 3 metres the number of tubes was reduced to 90. The synthesis gas and the catalyst suspension were of precisely the same composition as in Run A, and the space velocity was the same as that used in Run A. It will be noted from the end gas analysis B in the Table, that the results obtained were appreciably better than those obtained in Run A. With a lower temperature of 262° C in the cooling system, the CO conversion obtained was higher and, more particularly, considerably less methane was formed. Moreover, the amount of the $C_2 + C_3$ olefins was appreciably higher. Furthermore, the yield increased by approximately 50% to 600 kilograms of hydrocarbons per kilogram Fe in the catalyst, due to the reduced formation of methane and the longer active life of the catalyst.

TABLE

	Temper. °C.	CO_2 vol. %	C_3+C_4-olefins vol. %	C_2 vol. %	CO vol. %	H_2 vol. %	Hydrocarbons vol. %	N_2 vol. %	Co conversion %	grams CH_4 per normal cbm. $CO+H_2$	grams C_3+ per normal cubic metre $CO+H_2$
Synthesis gas analysis		4.0	—	—	56.3	34.5	0.2	5.0	—	—	—
End gas analysis A	267	59.4	3.1	0.7	9.4	11.2	6.2	10.0	91.8	21.2	157.8
End gas analysis B	262	61.4	4.7	0.5	7.7	11.0	4.2	10.5	93.6	12.2	172.3

What we claim is:—

1. A synthesis reactor for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium through which the reactants are passed in upward flow, including a series of tubes disposed within the reactor through which tubes a cooling medium may be passed, the tubes being decreased in number in the upward direction whereby the total outer surface area of the tubes decreases in the same direction.

2. A synthesis reactor for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium through which the reactants are passed in upward flow, including a series of tubes disposed substantially vertically within the reactor, through which tubes a cooling medium may be passed, two or more of the tubes being combined to form a smaller number of tubes whereby the total outer surface area of the tubes decreases in the upward direction.

3. A process for the production of hydrocarbons by the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, including the step of cooling the catalyst suspension by means of cooling surfaces disposed within the suspension, the area of the cooling surfaces being reduced in the direction of flow of the synthesis gas.

4. A process according to claim 3, in which the catalyst is an iron catalyst.

5. An improved process for the hydrogenation of carbon monoxide, substantially as hereinbefore described.

6. A synthesis reactor for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, the reactor being provided with cooling tubes which are disposed in the synthesis reactor

substantially as hereinbefore described with reference to Figure 2 of the accompanying drawings.

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1 SHEET *This drawing is a reproduction of the Original on a reduced scale*

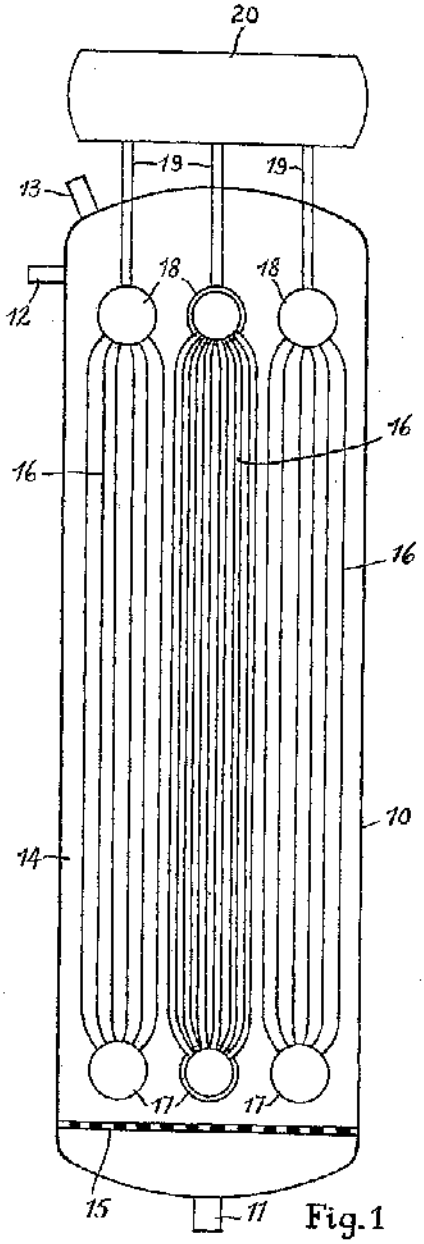

Fig. 1

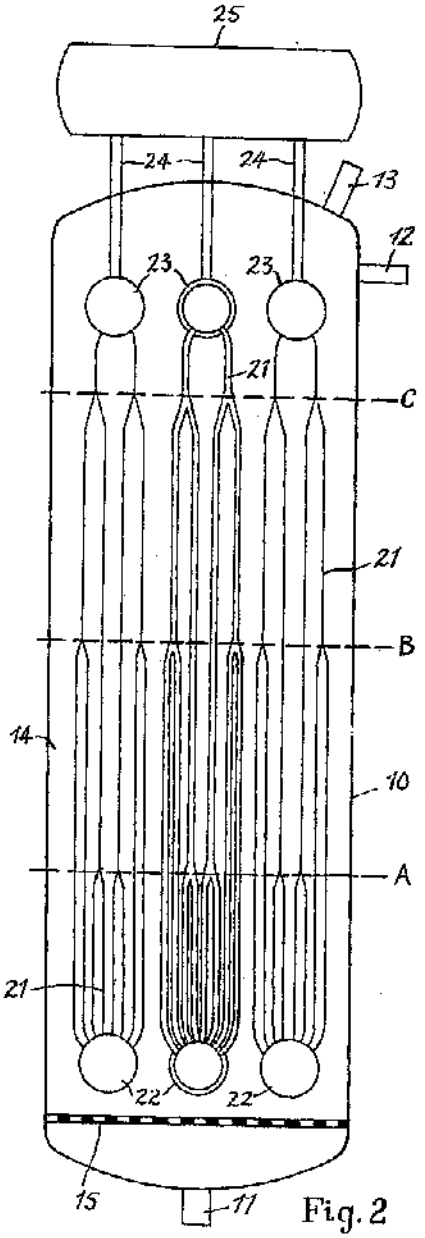

Fig. 2