PATENT SPECIFICATION

786,888

Date of Application and filing Complete Specification May 11, 1954.

No. 13793/54.

Application made in Germany on May 26, 1953.

Complete Specification Published Nov. 27, 1957.

Index at Acceptance:—**Class 1(1), A3A2.**

International Classification: —**B01j.**

COMPLETE SPECIFICATION

Reaction Apparatus for carrying out the Hydrogenation of Carbon Monoxide

We, RHEINPREUSSEN AKTIENGESELLSCHAFT FUER BERGBAU UND CHEMIE, 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 reaction apparatus for carrying out the hydrogenation of carbon monoxide in the presence of a finely-divided catalyst suspended in a liquid medium, in which the reaction chamber which is relatively tall compared to its width or diameter is divided over the greater part of its cross-section into a series of vertical shafts which are open at both ends and which are formed by heat-conducting members which are secured in a liquid-tight manner to each other and to cooling tubes disposed within the reaction chamber. The lower ends of the vertical shafts terminate at a position above the level at which the synthesis gas is fed upwardly into the reaction chamber, whilst the upper ends of the vertical shafts are disposed in the gas space in the upper part of the reaction chamber, that is to say, the upper ends of the vertical shafts are disposed at a level which is above the level which the liquid medium assumes when the apparatus is on stream.

The diameter of the reaction chamber is more than 30 cm. and may, for example, be 100 cm. or more. In spite of this, vertically circulating streams do not occur within the main body of the liquid medium when the synthesis gas is passed upwardly through the liquid medium, the formation of such vertically directed streams being prevented by the fact that the liquid medium is divided, by the vertical shafts, over the greater part of its depth into a number of separate individual columns, which are in communication with each other only at their lower ends in the sump of the reaction chamber. With such an apparatus, the distribution of the synthesis gas can be made substantially uniform over the whole cross-sectional area of the reaction chamber, even with a gas throughput of below 30 working litres per square centimetre of cross-section of the reaction chamber per hour. For this purpose, it has been considered to be necessary to charge each individual shaft, that is to say, each individual column of liquid, with its own quantity of gas, and even advantageous to control each of such charges individually, each charge being introduced directly into the shaft or at least vertically below the shaft in the common liquid sump. It was thus necessary to provide a series of gas intakes which were uniformly distributed over the whole cross-sectional area of the reaction chamber, with the provision of at least one gas intake or inlet to each shaft.

It has now been found that with such tall cylindrical reaction chambers which are divided into shafts, a substantially uniform gas distribution within the shafts may also be attained by providing a reaction chamber which converges below the shafts to a single, axially disposed, gas inlet, the distance of which from the lower edge of the bundle or group of shafts is at least as great as, or greater than, the diameter of the cylindrical reaction space.

According to the invention, therefore, a reaction apparatus for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, comprises a cylindrical vessel relatively tall compared to its diameter and forming a series of liquid-tight vertical shafts which extend substantially over the whole of the cross-section of the vessel, each vertical shaft being of relatively small cross-sectional area, being open at both ends and extending over the greater part of the length of the vessel, the lower edges of the series of vertical shafts ending short of the base of the vessel in a common space for the liquid medium, a common gas space for the tail gas above the upper edges of the series of vertical shafts, and a single, axially-provided gas inlet at the bottom of the vessel, the cross-sec-

[*Price 3s. 6d.*]

tion of the vessel being decreased below the lower edges of the vertical shafts to converge on to the gas inlet, the distance between the lower edge of the series of vertical shafts and the gas inlet being at least as great as the diameter of the cylindrical part of the vessel.

The construction according to the invention has an unexpected effect since with cylindrical liquid columns having a diameter of from 30 cm. upwards, strong liquid eddies are formed which extend vertically over the total height of the column and which prevent a uniform distribution of the gas bubbles over the total cross-sectional area at every level except in the vicinity of the surface of the liquid, even if the gas is introduced in small bubbles which are uniformly distributed over the whole of the base. It would be assumed that with a single, centrally disposed gas inlet, this internal rotation of the liquid would be increased still further. It has been found however, that, when the liquid column is divided by a series of shafts, the upper position of reversal of the liquid eddy is situated just below the lower edge of the bundle of shafts, and the distribution of gas immediately below the lower edge of the bundle of shafts is almost uniform over the whole of the cross-section of the reaction chamber. Moreover, the regulating action of the liquid-gas bubble suspension system in the shafts on the hydrostatic equilibrium at the base of the bundle of shafts results in an almost uniform distribution of the gas over all the shafts.

The shape or form in which the reaction chamber or space converges below the shafts may vary; for example, it may be conical or arcuate as illustrated in Figures 1 and 2 respectively of the accompanying drawings.

The distance between the gas inlet and the lower edge of the bundle of shafts is at least as great as the diameter of the reactor to permit the jet of gas to be torn apart or subdivided to a sufficient extent by the liquid in the sump. It is also possible to provide baffle plates above the gas inlet. However, the function of such baffle plates is adequately carried out by the horizontal pipes through which the coolant is fed into the tubes of the cooling system. The cooling system is advantageously extended to a position as low as possible below the lower edge of the bundle of shafts, in order that the heat of reaction evolved in the sump below the shafts may also be dissipated.

In the reaction apparatus according to the invention, the ratio of the free cross-sectional area of the gas inlet nozzle to the free cross-sectional area of the reaction space in the cylindrical part of the vessel, may range between 10:10,000 and 1:10,000, according to the absolute size of the cylindrical vessel and the cross-sectional load in working units by volume of synthesis gas provided, relative to the free cross-section of the reaction space. In practice, the cross-sectional load used in the hydrogenation of carbon monoxide carried out with a suspended iron catalyst at synthesis pressures in the approximate range of from 5 to 25 atmospheres, ranges advantageously between 5 and 200 working litres of synthesis gas per hour relative to each square centimetre of cross-section of the reaction space.

In the whole range of the cross-sectional load, the ratio of the cross-section of the gas inlet nozzle to the cross-section of the reaction space is so selected that in the nozzle of the gas inlet the linear gas velocity ranges between 2 metres per second and 400 metres per second, and preferably between 5 and 200 metres per second. In the direction in which the diameter of the reaction space increases, it is advantageous to provide nozzles the cross-section of which is smaller relatively to the cross-section of the reaction space, so that with equal cross-sectional load of synthesis gas, the linear gas intake velocity is correspondingly increased. According to the invention, in view of the fact that the distance between the nozzle and the lower edge of the bundle of shafts is increased according to the diameter of the cylindrical part of the reaction vessel, a correspondingly increased gas intake velocity is desirable since it is necessary for a vigorous turbulence of the liquid to be maintained in the sump which may be of a depth of up to 5 metres.

Moreover, it is advantageous for the height of reaction spaces of large diameter to be correspondingly increased; thus when the diameter is 2.5 metres, the height may, for example, be 25 metres. In order to obtain the same space load of synthesis gas, and consequently the same space-time yield of reaction products, it is possible to operate these tall reaction spaces with higher cross-sectional loads than is possible in lower reaction spaces, without increasing the catalyst load though the concentration remains the same.

By increasing the cross-sectional load, it is possible for the reaction zone which, for example, is only approximately 2 metres in length at a cross-sectional load of 10 working litres per sq. cm. per hour, to be drawn or spread substantially uniformly over the total height of the reaction space. Equally advantageous effects are obtained when a temperature rise in the upward direction is maintained, while the temperature in the sump of the reactor is relatively low, which may, for example, be attained in the manner described in the pending Patent Application No. 9234/54 (Serial No. 785,752).

Due to the vigorous rotating movement of substantially the whole of the liquid medium in the sump, the suspended catalyst is uniformly distributed in the liquid medium in the sump thereby ensuring that the individual shafts are uniformly provided with catalyst. More particularly, dead spaces with little movement and, therefore, deposits of catalyst are avoided, since in accordance with the invention the lower part of the reactor is tapered or otherwise constricted.

Instead of providing the lower part of the reaction chamber of conical form as illustrated in Figure 1, the cross-section may advantageously be tapered or restricted, as illustrated in Figure 2, in a manner adapted to assist the rotating flow of the liquid in the sump, and the rotating movement of the liquid in the sump may effectively be further assisted by extending the gas inlet nozzle so that it projects into the reaction space.

When the gas inlet nozzle extends into the reaction space, an annular, concentric guide member for the liquid medium may also be provided, as illustrated in Figures 3, 4 and 5. The annular guide member is advantageously so provided that a free annular space for the liquid medium is formed between the guide member and the inner wall of the reaction vessel and between the guide member and the gas inlet pipe, which annular space converges upwardly between the gas inlet pipe and the guide member, being narrowest at about the level of the upper edge of the gas inlet pipe. As from this position, the guide member may be provided with a head-piece which opens upwardly and outwardly in the manner of a funnel, as illustrated by way of example in Figures 4, 5 and 6. The aperture angle of the head-piece advantageously lies between 50° and 120°.

In this construction, the gas stream has a vigorous suction effect so that the liquid in the sump rotates or circulates at an increased rate. A forced circulation of the catalyst suspension, which forced circulation may be varied over an appreciable range, is thereby obtained. Since the energy required for the purpose is supplied to the system solely by the synthesis gas, it is possible by increasing the gas velocity—it being necessary to accept a pressure drop of a magnitude of from 0.2—1 atmosphere or more before and after the nozzle —to increase the speed of circulation of the liquid to such a degree that a substantial part of the gas, which is mixed with liquid above the nozzle, circulates together with the liquid past the nozzle. As a result, the gas in the zone below the shafts, that is, in the sump, is already almost homogeneously divided into small bubbles.

The gas distribution in the reaction apparatus according to the invention can be further improved by providing the top of the funnel-shaped extension of the guide member with perforated plate, as illustrated in Figure 6, through the openings of which gas and rotating liquid together are forced. The openings in the perforated plate may be relatively wide; advantageously the openings, which may be of circular cross-section, have a diameter greater than 10 mm. The openings may be distributed uniformly over the whole plate, or they may, with advantage, be increased in number towards the edge of the plate. However, the total of the cross-sectional areas of all the openings should be at least twice and up to twenty times as great as the sum of the smallest free cross-sections of the gas inlet nozzle and of the annular space between the gas inlet nozzle and the guide member.

What we claim is:—

1. A reaction apparatus for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, comprising a cylindrical vessel relatively tall compared to its diameter and forming the reaction chamber, one or more bundles of tubes disposed in the vessel and through which a cooling medium may be passed, a series of heat-conducting plates connected to the tubes and forming a series of liquid-tight vertical shafts which extend substantially over the whole of the cross-section of the vessel, each vertical shaft being of relatively small cross-sectional area, being open at both ends and extending over the greater part of the length of the vessel, the lower edges of the series of vertical shafts ending short of the base of the vessel in a common space for the liquid medium, a common gas space for the tail gas above the upper edges of the series of vertical shafts, and a single, axially-provided gas inlet at the bottom of the vessel, the cross-section of the vessel being decreased below the lower edges of the vertical shafts to converge on to the gas inlet, the distance between the lower edge of the series of vertical shafts and the gas inlet being at least as great as the diameter of the cylindrical part of the vessel.

2. A reaction apparatus according to Claim 1, in which the ratio of the free cross section of the gas inlet nozzle to the cross section of the reaction space is so selected that with a cross-sectional load of from 5 to 200 working litres of synthesis gas per hour per square centimetre of free cross section of reaction space, there is obtained at the gas inlet nozzle a linear gas velocity of from 2 to 400 metres per second.

3. A reaction apparatus according to Claim 2, in which the ratio is so selected that the linear gas velocity at the gas inlet nozzle is within the range 5—200 metres per second.

4. A reaction apparatus according to any one of the preceding claims, in which the gas inlet nozzle projects into the reaction space.

5. A reaction apparatus according to any one of the preceding claims, including a gas inlet pipe extending into the reaction space, and a concentric guide member disposed between the gas inlet pipe and the inner wall of the vessel and spaced from the inlet pipe and from the wall and from the base of the vessel, the guide member being such that the space between it and the gas inlet pipe converges upwardly so that the said space is narrowest at a position which is approximately at the level of the upper edge of the gas inlet pipe or nozzle.

6. A reaction apparatus according to Claim 5, in which the guide member opens upwardly and outwardly above the upper edge of the gas inlet pipe or nozzle.

7. A reaction apparatus according to Claim 5, in which the guide member opens upwardly and outwardly in the manner of a funnel above the upper edge of the gas inlet pipe or nozzle, the aperture angle of the funnel being not less than 50° and not substantially greater than 120°.

8. A reaction apparatus according to Claim 7, in which the upper edge of the guide member is closed by a perforated plate, the total area of the perforations in which is from twice to twenty times as great as the sum of the smallest free cross-sections of the gas inlet nozzle and of the annular space between the nozzle and the guide member.

9. A reaction apparatus for carrying out the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, substantially as hereinbefore described.

10. A reactor for carrying out the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, substantially as hereinbefore described with reference to any one of Figures 1 to 6.

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Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press.—1957.
Published at the Patent Office, 25, Southampton Buildings, London, W.C.2, from which copies may be obtained.

786,888 COMPLETE SPECIFICATION

1 SHEET

This drawing is a reproduction of the Original on a reduced scale.

Fig. 1

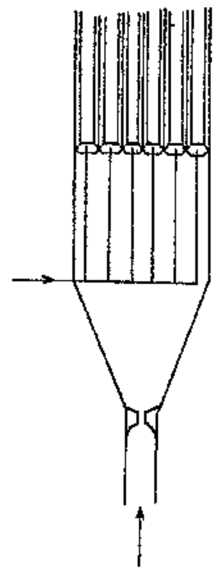

Fig. 2

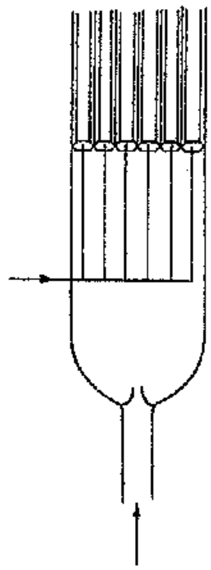

Fig. 3

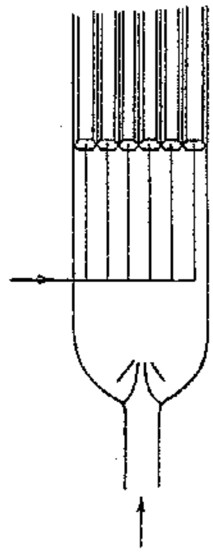

Fig. 4

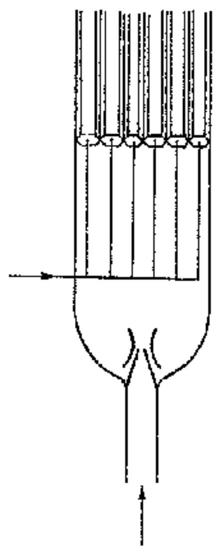

Fig. 5

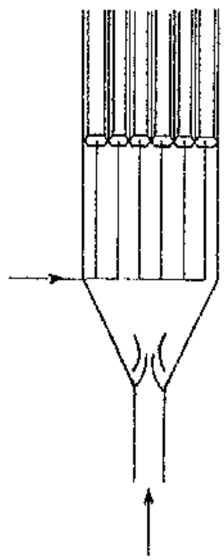

Fig. 6