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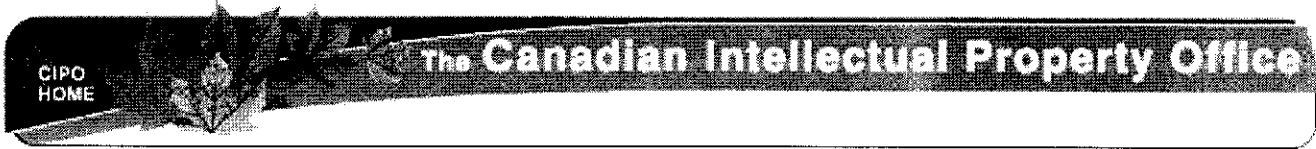
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(54) REACTION APPARATUS FOR CARRYING OUT THE HYDROGENATION OF CARBON MONOXIDE

(54) APPAREIL DE REACTION POUR L'HYDROGENATION D'OXYDE DE CARBONE

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The present invention relates to a proposed construction of the sump below the shafts and the gas intake of the reaction apparatus for the catalytic hydrogenation of carbon monoxide in the presence of finely divided catalysts which are suspended in a liquid medium, described in Federal German Patent Specification No. 948,781 which apparatus is divided into shafts over substantially the whole of its cross-section.

In the apparatus described in Federal German Patent Specification No. 948,781 substantially vertically directed streams of circulating liquid are prevented from forming when the gas is passed through, since the greater part of the length (including the upper area of contact with the gas space) of a stationary, that is to say, non-rotating liquid column having a diameter of over 30 cm., 100 cm. or more, is divided into separate individual columns which have a common sump and a common upper gas space, so that as a result the distribution of gas according to the size, quantity and speed of ascent of the gas bubbles, is substantially equalized over the whole cross-sectional area of the reaction space, even with a gas throughput of below 30 working litres per square centimetre of cross-section of the reaction space per hour. It has been considered to be advantageous to charge each individual shaft forming the individual liquid columns, with its own quantity of gas, which may even be individually controlled, and which was to be introduced direct into the shaft, or at least vertically below each shaft in

the common liquid sump. It thus became necessary to provide a gas intake or feed which was uniformly distributed over the whole cross-sectional area of the reaction space, and which had at least one gas outlet to each shaft.

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It has now been found that with such tall cylindrical reaction spaces which are divided into shafts, a substantially uniform gas distribution within the shafts may also be attained by providing a reaction space which converges below the shafts to a single, axially disposed, gas inlet opening, 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.

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This 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.

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It is to be assumed that with central gas intake, this internal rotation of the liquid would still be furthered. It has now been found that, when the liquid column is divided in the manner of shafts, the upper position of reversal of the liquid eddy is situated just below the lower edge of the bundle of shafts. As on or adjacent to the surface of the

liquid, the distribution of gas below the lower edge of the bundle of shafts is almost uniform over the cross-section. 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.

According to the invention, the shape in which the reaction space converges below the shafts may vary; for example, it may be conical, bulging, or the like as illustrated in Figures 1 and 2.

It is advantageous to provide a large distance between the gas inlet and the lower edge of the bundle of shafts, that is to say, the distance should be as large, or several times as large, as the diameter of the reactor so that the jet of gas is torn by the liquid column to a sufficient extent. It is also possible to provide baffle plates above the gas inlet. However, this function is adequately carried out by the horizontal feed pipes for the coolant which lead to the cooling system which is advantageously extended to a position as low as possible below the bundle of shafts, because the heat of reaction evolved in the sump below the bundle of shafts has likewise to be dissipated.

In the reaction apparatus according to the invention, the ratio of the free cross-sectional area of the gas intake nozzle to that of the reaction space at the level of the cylinder may range between 10 : 10,000 and 1 : 10,000 which is determined by the absolute size of the reaction apparatus 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 nozzle to the cross-section of the reaction space is so selected that in the gas inlet (nozzle) 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 distance between the nozzle and the lower edge of the bundle of shafts being increased as according to the diameter of the reaction space, 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 have a height of up to 5 metres.

Moreover, it is advantageous for the height of

reaction spaces of a 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 through 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 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 accordance with the pending Patent Application No. 663,966, filed March 31st 1954 in Canada in the name of Rheinpreussen Aktiengesellschaft fuer Bergbau und Chemie.

Due to the vigorous rotating movement of the liquid medium which, according to the invention, involves the total content of the sump, the suspended catalyst is uniformly distributed over the total content of 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 taper cones as illustrated in Figure 1, the tapering or manner of restricting the cross-section illustrated in Figure 2, which is
5 adapted to the rotating flow of the liquid, may also advantageously be used, and the rotating movement may effectively be furthered by extending the nozzle to project into the space.

It is also possible to coordinate with the nozzle
10 extended into the reaction space, concentric guide members for the liquid medium, as illustrated respectively 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
15 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
20 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
25 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 rotates at an increased rate. A forced rotation of the
30 catalyst suspension, which forces rotation 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
5 of from 0.2-1 atmosphere or more before and after the nozzle- to increase the rotational speed 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,
10 the gas in the zone below the shafts is already almost homogeneously divided into small bubbles.

As illustrated in Figure 6, the gas distribution according to the invention can be further improved by providing the top of the funnel-shaped extension
15 of the guide member with a perforated plate through the openings of which gas and rotating liquid together are forced. The openings in the perforated plate may be relatively wide; they have advantageously a diameter of over 10 mm. and may be of circular
20 cross-section. The openings may be distributed uniformly over the whole plate, or they may advantageously be increased in number towards the edge. However, the total of cross-sections of all the openings should be at least twice and up to twenty times as great as
25 the total 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.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:-

1. A reaction apparatus for the hydrogenation of carbon monoxide in the presence of a catalyst suspended in a liquid medium, comprising a relatively tall cylindrical vessel provided internally with a cooling system for the dissipation of reaction heat, a series of vertical shafts which are in liquid-tight connection with each other and which extend over the greater part of the length of the vessel, the shafts being open at the top and at the bottom, a common space below the lower edges of the shafts to form a sump for the liquid medium, a common gas space for the tail gas above the upper edges of the shafts, an axially provided gas inlet at the bottom of the reaction space, the reaction space being restricted below the lower edge of the series of shafts and converging to the gas inlet, the distance between the lower edge of the series of shafts and the gas inlet being at least as great as the diameter of the cylindrical part of the reaction space.

2. 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 space, 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 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, 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 pipe or nozzle, 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.

3. A reaction apparatus according to claim 1 or claim 2, in which the ratio of the free cross-section of the gas inlet nozzle to the cross section of the cylindrical 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.

4. A reaction apparatus according to claim 1 or claim 2, in which the ratio of the free cross section of the gas inlet nozzle to the cross section of the cylindrical 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, the linear gas velocity at the gas inlet nozzle is within the range 5 - 200 metres per second.

5. A reaction apparatus according to claim 1 or claim 2, in which the gas inlet pipe projects into the reaction space.

6. A reaction apparatus according to claim 1 or claim 2, in which the gas inlet pipe extends into the reaction space, and a concentric guide member is disposed between the gas inlet pipe and the inner wall of the vessel and is spaced from the gas 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 at its narrowest at a position which is approximately at the level of the upper edge of the gas inlet pipe or nozzle.

7. A reaction apparatus according to claim 1 or claim 2, in which the gas inlet pipe extends into the reaction space, and a concentric guide member is disposed between the gas inlet pipe and the inner wall of the vessel and is spaced from the gas 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 at its narrowest at a position which is approximately at the level of the upper edge of the gas inlet pipe or nozzle, the guide member opening upwardly and outwardly above the upper edge of the gas inlet pipe or nozzle.

8. A reaction apparatus according to claim 1 or claim 2, in which the gas inlet pipe extends into the reaction space, and a concentric guide member is disposed between the gas inlet pipe and the inner wall

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of the vessel and is spaced from the gas 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 at its narrowest at a position which is approximately at the level of the upper edge of the gas inlet pipe or nozzle, the guide member opening 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° .

9. A reaction apparatus according to claim 1 or claim 2, in which the gas inlet pipe extends into the reaction space, and a concentric guide member is disposed between the gas inlet pipe and the inner wall of the vessel and is spaced from the gas 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 at its narrowest at a position which is approximately at the level of the upper edge of the gas inlet pipe or nozzle, the guide member opening 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° , the upper end of the funnel of the guide member being closed by a perforated plate, the total area of the perforations in which plate is from two to twenty times as great as the smallest free cross-sections of

the gas inlet nozzle and of the annular space between
the nozzle and the guide member.

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Fig.1

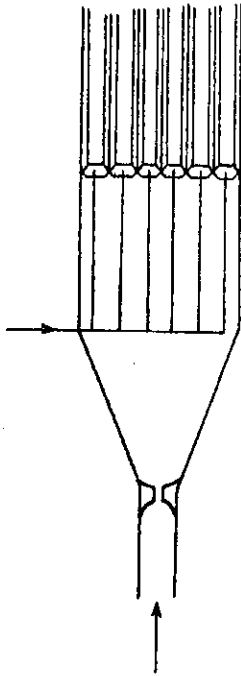


Fig.2

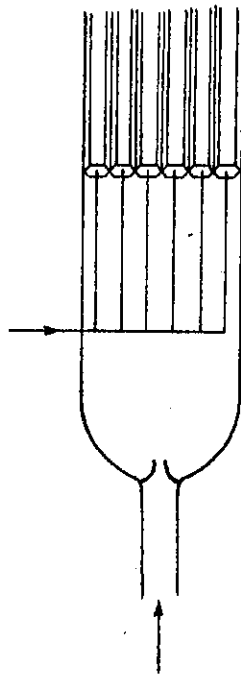


Fig.3

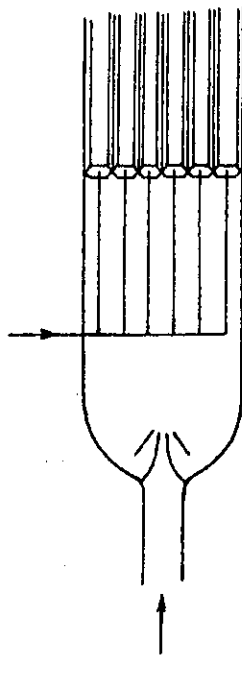


Fig.4

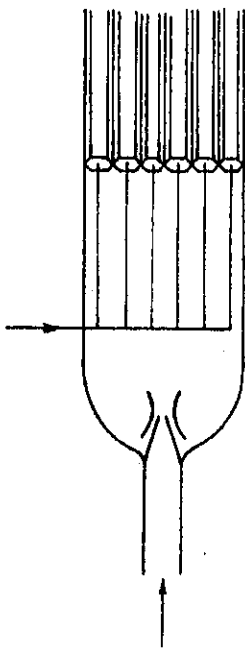


Fig.5

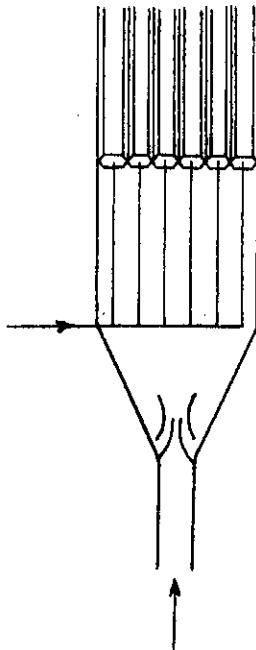
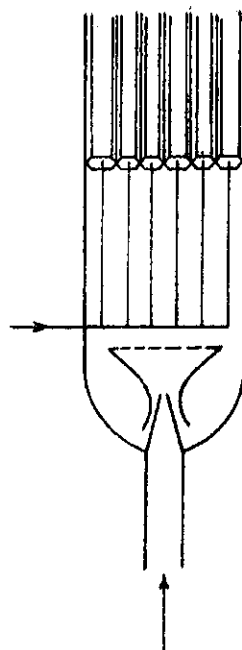


Fig.6



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