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(54) APPARATUS FOR THE CATALYTIC GAS REACTIONS IN LIQUID MEDIA

(54) APPAREIL POUR DES REACTIONS CATALYTIQUES DE GAZ DANS DES MILIEUX LIQUIDES

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The invention relates to apparatus for carrying out gas reactions in the presence of liquid or solid catalysts which are dissolved or maintained in finely divided suspension in a liquid medium, and particularly to such apparatus for carrying out the hydrogenation of the oxides of carbon under elevated gas pressure to form hydrocarbons or organic compounds which contain oxygen or nitrogen.

The invention relates specifically to reaction apparatus in which the liquid medium is maintained stationary during the gas reaction, that is to say in which it is not subjected to any circulation or recycling inside or outside the reaction space.

The invention is described with reference to the accompanying diagrammatic drawings, in which,

Figure 1 shows the distribution of gas and liquid in a reactor of the prior art;

Figure 2 shows the distribution of gas and liquid in another reactor of the prior art;

Figure 3 shows the distribution of gas and liquid in a reactor embodying the features of the invention;

Figures 4, 5 and 6 are transverse sections through part of three constructions of the vertical shafts and heat-exchange tubes for use in a reactor provided according to the invention;

Figure 7 is a longitudinal section, on the line X-X of Figure 8, of a reactor provided according to the invention;

Figure 8 is a transverse section on the line Y-Y of Figure 7;

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Figure 9 is a transverse section through part of a further construction of vertical shafts and heat-exchange tubes;

Figure 10 is a transverse section through part of another construction of vertical shafts and heat-exchange tubes;

Figure 11 is a longitudinal section of the base of a reactor showing one means of feeding gas into the reactor;

Figure 12 is a plan view of a further means of feeding gas into the reactor;

Figure 13 is a plan view of yet another means of feeding gas into the reactor;

Figure 14 shows, in longitudinal section, four constructions of nozzles suitable for introducing the gas centrally at the base of the reactor;

Figure 15 shows means for controlling the rate of heat transfer in the reactor, and

Figure 16 shows another means for controlling the rate of heat transfer in the reactor.

It is known that in gas reactions which are carried out in the presence of catalysts in a liquid medium, and in which the gaseous phase does not disappear completely when passing through the liquid medium, being rather preserved as a separate phase on the whole of its path through the liquid medium due to the incomplete solubility of the gaseous starting materials and end products, the degree of gas conversion is proportional to the size of the boundary surface between the gas and the liquid. In such a system of distribution of gas in

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liquid, the size of the boundary surface is determined by the size of the gas bubbles.

For certain processes including the Fischer-Tropsch synthesis, it has been found to be sufficient to operate with the size of gas bubble which forms almost independently of the manner in which the gas is introduced into the liquid medium, by virtue of the surface tension and viscosity of the liquid under the operating conditions. It is thus unnecessary to provide for a specific kind of gas distribution. However, to obtain a complete gas conversion, a determined residence time, which should be as uniform as possible for the individual gas bubbles, is necessary.

These requirements are met by passing the synthesis gas upwardly into a narrow and relatively tall column of liquid at elevated pressure and at elevated temperature without using special means for distributing the gas, the quantity of gas being such that the space load, measured at the pressure and temperature of the synthesis, is between about 5 and 100 litres of gas per litre of liquid medium. At a certain distance above the position at which the gas is introduced, there develops a state in which the mixture of gas and liquid is such that the gas bubbles are of almost uniform size and are distributed substantially uniformly over the whole horizontal cross-section of the column and ascend at an almost uniform speed. The total volume of the liquid/gas system is directly proportional to the gas throughput, whereas the total contact time of the gas bubbles is almost independent of the gas throughput. As the gas

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throughput increases, the distance between the gas bubbles is reduced without coalescence of the gas bubbles taking place. The nature of this system, which we have named "liquid/gas suspension system" has not yet been fully explained. It has, however, been found that in this system the liquid is in a state of very fine turbulence in which the eddies are approximately only of the order of magnitude of the gas bubbles, that the bulk of the liquid remains stationary, that is to say, only a small fraction thereof travels upwards with the gas bubbles, and that, due to the friction between the gas bubbles and the liquid, a continual and rapid renewal or change of the intersurface occurs. It appears that through these factors, together with the fine turbulence, the exchange of substances between the gaseous phase, the liquid phase, and the surface of the catalyst is facilitated and improved to such a degree that the hydrogenation of carbon monoxide is no longer impeded by the liquid medium, but proceeds at much the same speed as in the known process using a dry catalyst whirled up in the manner of a fluid, that is to say, the so-called fluidized catalyst process (fluid bed). Thus, in the process carried out in the liquid phase with a stationary column of liquid, up to 15 litres (760 mm. Hg., $0^{o}C.$) of $CO + H_2$ may be converted per gram of catalyst per hour. This conversion may be obtained with a single passage of the gas through the liquid medium and at temperatures below $300^{o}C.$

By virtue of the high load capacity of the catalyst achieved in accordance with this process, combined with a high maximum content of catalyst in the suspension which may be up to 500 grams of the main or base catalyst metal per litre of non-inflated or gas-free suspension, without the suspension losing its property of ready fluidity or mobility, it is possible to obtain space-time yields of reaction products which are higher than those obtained by other known liquid phase processes.

In the Fischer-Tropsch synthesis carried out with a suspended iron catalyst, the particle size of which is advantageously maintained between 0.005 mm. and 1 mm. with a yield of 170 grams per normal cubic metre of $CO+H_2$ used and with a single passage of the gas at a temperature of approximately 275°C. a gas pressure of 25 atmospheres and an hourly throughput of up to 1250 litres of $CO + H_2$ (measured at 0°C. and at a pressure of 760 mm. Hg), approximately 5 kilograms of synthesis products containing more than 3 carbon atoms in the molecule may be produced within 24 hours per litre of reaction space. In accordance with the known process, this synthesis may be carried out smoothly in reaction spaces having a diameter of 20 centimetres. However, as the horizontal diameter of the reaction space increases, the degree of gas conversion decreases, and it becomes increasingly difficult to maintain a constant gas conversion. The greater the horizontal diameter of the reaction space, the more the body of liquid will tend to pass from the stationary state into a state of vertical rotation or circulation. The liquid then flows downwardly along the surfaces of the walls, and at the bottom flows towards

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the centre of the reaction space while entraining all the gas bubbles issuing from the centre of the base. The compressed, central gas stream drags the liquid upwardly, while the compressed gas bubbles coalesce to form giant and elongated gas bubbles. It is only at the upper position of reversal of the flow of the liquid, adjacent to the surface of the column of liquid, that the gas spreads out horizontally over the cross-section of the column of liquid and the large gas bubbles partially disintegrate. This movement of the gas and liquid is diagrammatically illustrated in Figure 1 of the accompanying drawings. The synthesis gas is passed through a line 2 upwardly into a reactor 1 within which the catalyst suspension 3 is contained, the gas stream being distributed by the nozzles or perforations 6 of the gas distributing plate 4. The movement and circulation of the liquid in the reaction space is shown by the arrows 5. It will be understood in this respect that the hydrodynamic pressure exerted on the gas distributing plate 4 at positions close to the wall of the reactor 1 may be higher than in the centre of the plate 4, whereby the passage of gas through the outer nozzles or perforations 6 is completely suppressed, so that the bulk of the gas enters the reaction space solely through the central nozzles of the plate 4. In addition, the rate of flow of the eddying or circulating liquid increases with the absolute height of the column of liquid. There is no purpose in trying to overcome this disadvantage by allowing the gases to pass into the catalyst suspension only at peripheral portions of the distributing plate 4, as diagrammatically

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illustrated in Figure 2 of the accompanying drawings. In this case, a similar state of affairs will develop, but with the difference that in addition to the tall eddy of liquid, a small eddy of liquid also develops at 7 below the funnel formed by the gas stream.

In a distributing system of this kind, the time of contact of the individual batches of gas is extremely irregular due to the irregular size of the gas bubbles and because part of the gas bubbles is incorporated in the cycle of the liquid. Thus, while on the one hand some of the gas does not contact the catalyst at all, that is to say, it slips through the suspension without contacting the suspended catalyst, on the other hand some of the gas, together with the reaction products contained therein, is repeatedly brought into contact with the catalyst for a longer time than is desirable. Moreover, the fresh gas is diluted by the tail gas entrained by the vertical cycle.

Thus in this known kind of reactor, these liquid eddies not only result in a lower gas conversion, but moreover cause undesirable secondary reactions. Over and beyond this, in the Fischer-Tropsch synthesis the catalyst which then circulates rapidly together with the liquid in such a reactor is substantially damaged by the rapid change in the surrounding conditions, so that its efficiency is reduced, its activity decreasing more rapidly then in the synthesis with a stationary column of liquid, in which the individual catalyst particles reside for a comparatively long time in a determined zone in which the composition of the

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gas is substantially constant.

Various proposals for improving the distribution of the gas in liquids in large reaction spaces have been made. These include the use of rotating bodies, the so-called turbo-mixers, or the introduction of gas by means of the Segner wheel. It has also been proposed to fix or determine the direction of the swarms of gas bubbles, which enter at the bottom of the reaction space through porous plates or individual nozzles, by means of guide members, such as tubes or plates. These guide members are, however, only provided at or close to the bottom or base of the reaction space and are shorter in height than the column of liquid.

The individual installations or members provided in the reactor are, therefore, not provided with their own surfaces of liquid and act in the manner of air-lift pumps, so that in reaction apparatus thus equipped, the circulation of the liquid, while being controlled, is in any case intensified.

By these known proposals, the disadvantageous consequences with respect to the gas conversion, the properties of the product and the efficiency of the catalyst caused by the circulation of the liquid, are not removed. Apparatus of this kind may be sufficient and adequate for gas reactions in which the tail gas is chemically identical with the primary or initial gas, such as is the case, for example, with the hardening of fats. None of these known proposals is sufficient or adequate with gas reactions in which the composition of the gas changes; in which reaction products have to be discharged together with the tail gas, and in which the gas conversion should be as complete as possible with a single pass of the gas.

In none of the known apparatus is the object particularly essential for the Fischer-Tropsch synthesis realised, namely that of maintaining the liquid medium and the suspended catalyst substantially stationary, whilst nevertheless allowing the gas bubbles to travel in substantially uniform size and substantially uniform distribution and with substantially uniform speed through the liquid medium.

It has now been found, according to the invention, that with gas reactions carried out in the presence of a liquid or solid catalyst dissolved or maintained in finely divided suspension in a liquid medium, in reaction apparatus of relatively large horizontal diameter, the disadvantages hereinbefore described may be removed or substantially avoided by dividing the reaction space (which consists of an upright cylinder having a horizontal diameter of more than approximately 30 centimetres and up to 3 metres or more, and having a height of more than 1.5 metres approximately), into a number of substantially uniform vertical shafts, as from a level above the gas inlet which equals or is greater than the diameter, and over the greater part of the cross-sectional area, the outer surfaces of which shafts are impervious to liquid, the shafts being open at both ends, having a diameter of a least 5 centimetres, and extending into the free gas space below the highest level of the liquid when the liquid is in the condition of operation and inflated by gas bubbles.

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Surprisingly, there develops the state of distribution of gas and liquid diagrammatically illustrated in Figure 3 of the accompanying drawings. In the sump 15 disposed below the shafts 14 of the reactor 11, the distribution of the gas, which is introduced into the reactor through the inlet 12, is coarse and irregular. Here also in the sump 15, eddies of liquid, though of a small vertical dimension, develop. The lower edge of the bundle of shafts 14 effects the movement of gas and liquid in the sump 15 in the manner of the surface of a liquid, in that the eddies of liquid have their upper point of reversal at a substantial distance therefrom, thus drawing the generally somewhat compressed stream of gas bubbles apart in the direction of the periphery. The shafts 14 disposed further away from the axis are thereby supplied with substantially the same amount of gas as the central shafts. In contrast with the sump 15, substantially stationary columns of liquid, the degree of inflation of which is determined by the quantity of the gas, are developed in the shafts 14. The tail gas leaves the reactor 11 through the outlet 13.

Due to the fact that each shaft has its own surface relatively to the common upper gas space 16, the supply of gas to the shafts is equalized to such a degree that the reaction apparatus according to the invention shows exactly the same results of operation as a reaction apparatus, the dimensions of which correspond to those of a single shaft having a diameter of less than 20 centimetres, and which, moreover, is provided with its own gas supply. Furthermore, the hydrostatic pressure of the columns of liquid at the base of the shafts is practically the same with all the shafts, independently of small or periodical differences in the passage of the gas.

The surprising effect of the division of the reaction apparatus into shafts according to the invention may be summed up as follows:-

(1) The equalizing or balancing effect produced by the shafts on the supply of gas to each individual shaft is almost independent of the processes taking place in the sump.

(2) The formation of a stable liquid/gas suspension system in each shaft with a substantially stationary column of liquid and substantially uniform size and substantially uniform velocity of ascent of the gas bubbles.

(3) The automatic supply to each individual shaft of an amount of suspendedcatalyst proportional to the quantity of gas entering the shaft. When, for example, gas is prevented from entering a shaft by a baffle plate, the liquid in that shaft is substantially free from suspended catalyst.

(4) The gas reaction in the shafts proceeds absolutely identically with the results obtained in the laboratory under similar operating conditions and with the same efficiency. The reaction is substantially independent of the manner in which the gas is introduced or distributed at the bottom of the sump below the shafts.

A selection of specific constructions of reaction apparatus according to the invention is hereinafter

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described with reference to Figures 4 to 16 of the accompanying drawings.

The diameter or width of the shafts should be at least 5 centimetres and may be up to 30 centimetres or even more. The diameter or width of the shafts is determined by the height of the apparatus or shafts and by the cross-section of heat-exchange tubes which may be provided in the shafts.

The height or length of the shafts is determined by the time provided for contact of the gas bubbles with the catalyst suspension; this is advantageously determined experimentally by means of tests on a small or pilot scale.

According to the invention the height/diameter ratio of the shafts may range between 10 and 200 and is preferably between 20 and 100.

The manufacture of the reaction apparatus provided according to the invention is substantially facilitated in that each individual shaft is formed by a separate casing. This casing must be impervious to liquid; its walls may be as thin as the technique of manufacture permits. If desired, plates or sheets having a thickness of up to 0. 5mm. may be used for the walls, more particularly when it is thereby possible to prevent corrosion.

Shafts of circular cross-section, that is to say, ordinary tubes the walls of which are as thin as possible, may be used, these tubes being mounted so as to be packed together as close as possible (triangular or delta formation), or so as to be connected for formed in squares. In Figures 4, 5 and 6, the different arrangements of circular shafts a are shown, with heat-exchange tubes b mounted within and/or externally of the shafts a.

A particularly efficient utilization of the reaction space is obtained by using shafts formed as the casings of regular hexagonal prisms, because in this case clearances or dead spaces between the shafts may be avoided. Such an arrangement is illustrated in Figures 9 and 10, the shafts d being of hexagonal cross-section and having heat-exchange tubes e mounted within them.

To prevent the gas bubbles from entering the dead spaces between the circular shafts or the spaces, hereinafter referred to as segmental spaces, between the bundle of shafts and the inner wall of the reactor, baffle plates are provided directly below the bottom edge of the bundle of shafts. The lower surface of the baffle plates inclines upwardly in the direction of the shafts and the baffle plates are so provided to leave sufficient space to permit the liquid in the dead spaces and in the segmental spaces to communicate with the liquid in the sump.

One construction of apparatus according to the invention is diagrammatically illustrated by way of example in Figures 7 and 8, Figure 7 being a longitudinal section of the apparatus along the line X-X of Figure 8, and Figure 8 being a transverse section along the line Y-Y of Figure 7. The cylindrical reactor 21, the lower end of which is tapered to form a cone, is provided at the bottom with a central gas inlet 22, the inlet being provided with a constriction or nozzle 23, a check valve 24, and a valve conduit 25 through which a liquid may be passed for rinsing or clearing the nozzle 23. An outlet 26 for the tail gas is provided at the top of the reactor. A bundle of shafts 27 is disposed within and longitudinally of the reactor 21. A governor or regulator 28 for the charge in the reactor 21 is provided in one of the segmental spaces 38 disposed between the bundle of shafts 27 and the inner wall of the reactor 21 and serves to control the respective valves for the supply and discharge pipes, according to whether the contents of the reactor 21 increases or decreases during the reaction. Baffle plates 29 are provided to prevent the admission of gas into the segmental spaces. A system of tubes 30 which pass longitudinally through the shafts 27, comprises a heat exchange system which is supported on members 31 within the reactor 21. In operation, a heat transfer medium is passed into the tubes 30 through a line 32 and headers 33 disposed in the sump 39 of the reactor 21, and is withdrawn from the tubes 30 through headers 34 and line 35, the line 35 passing out of the reactor 21 through a gland 36 provided in the reactor wall. If necessary, liquid medium free of catalyst may be withdrawn through valved conduits 37, for example, when it is desired to obtain reaction products of high molecule weight. The exchange of liquid between the shafts 27, the sump 39, and the segmental space 38 is generally sufficient for this purpose in spite of the almost stationary condition in the shafts 27. A valved tube 40 is advantageously provided for charging the reactor with catalyst suspension and for removing catalyst suspension from the reactor.

The tubes 30 of the heat exchange system must extend as low as possible into the sump 39, the position of the inlet headers 33 being advantageously approximately 100 centimetres or more below the lower edge of the bundle of shafts 27.

The heat exchange tubes 30 are passed vertically through the reactor. In the construction illustrated in Figures 7 and 8, one heat exchange tube is provided within each of the shafts; a similar arrangement is also illustrated in Figure 4. More than one heat exchange tube may, however, be provided within each shaft, as illustrated in Figures 9 and 10. Some of the heat exchange tubes may also be provided externally of the shafts as shown in Figure 6, or all of them may be passed through the spaces between the shafts, for example, as illustrated in Figure 5.

With the heat exchange tubes extending through the interior of the shafts, the smallest horizontal distance between them and from the inner wall of the shaft should exceed 3 centimetres.

The distribution of gas at the bottom of the sump may also be effected over the whole cross-

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section, and in cases where a suspended, solid catalyst is being used, the openings or mouths of the gas inlets or nozzles may advantageously be directed downwardly towards the bottom of the reactor. Furthermore, it is advantageous to wet or moisten the gas inlet nozzles continuously from the gas side by means of a liquid medium.

Suitable means for carrying out one or more of these methods of feeding the gas into the reactor, are diagrammatically illustrated by way of example in Figures 11, 12 and 13. Figure 11 is a longitudinal section of the sump of a reactor into which the gas is fed through a series of nozzles or jets 41 provided in lines 42, the nozzles or jets 41 being directed downwardly towards the bottom of the reactor. Scavenging or cleaning oil for the nozzles or jets 41 is fed into the lines 42 through one or more lines 43. Figures 12 and 13 are plan views of two different means of feeding and distributing the gas. In the means illustrated in Figure 12, the gas is fed into the reactor through four pipes 44 disposed 90° apart, the gas passing from the pipes 44 into a series of concentrically disposed pipes 45 provided with perforations, nozzles or jets through which the gas passes directly into the reaction space. The means illustrated in Figure 13 comprises a series of gas inlet tubes 46 disposed in parallel relationship, the tubes being provided with nozzles, jets or the like through which the gas passes directly into the reaction space.

The division of the gas feed into determined zones over the cross-section of the lower part of the reactor, as illustrated in Figures 11, 12 and 13, renders possible the control of the quantity of gas admitted to each zone. In the reaction apparatus according to the invention, such control or regulation is only necessary in special cases, for example for whirling up settled catalyst after a stoppage or interruption of operation, or when it is necessary to operate occasionally with quantities of gas which are so low that the cross-sectional load is substantially below approximately 10 - 15 litres (at the pressure and temperature of operation) per square centimetre of free reactor cross-section per hour.

Four possible constructions 47, 48, 49 and 50 of nozzles suitable for introducing the gas centrally at the tapered bottom part of the reactor, are diagrammatically illustrated by way of example in Figure 14.

It has been found that, when gas reactions are carried out in the reaction apparatus according to the invention, cross-sectional loads in the range of approximately from 3 to 200 working litres of gas (at the pressure and temperature of operation) per square centimetre of free reactor cross-section per hour, are suitable for the liquid-gas suspension system. The term 'free reactor cross-section' denotes the full internal cross-sectional area of the reactor minus the cross-sectional area occupied by the walls of the shafts and by the heat-exchange or cooling tubes.

The mobility of the liquid medium in the operating condition is of extreme importance for the liquid gas suspension system, and the viscosity of the liquid medium containing the suspended catalyst should preferably be below 3° Engler approximately, which corresponds approximately to 21 cs. (centistokes). In the Fischer-Tropsch synthesis, the reaction proceeds particularly rapidly when the viscosity of the suspension lies below approximately 1.4° Engler or 5.2 cs.

The heat of condensation of steam may be used in known manner for heating the reaction apparatus and for supplying the heat of reaction in endothermic reactions. Similarly, for indirect heat exchange, water may be vapourised in the same tube system to dissipate heat of reaction.

Due to the intense, fine turbulence of the liquid in the liquid-gas suspension system in the shafts, the transmission of heat to the cooling surfaces is accelerated to such a degree that, even when hydrocarbons are used as the liquid medium, the transmission of heat using vertical, smooth steel tubes through which cooling water flows, is at least 300 kilocalories per sq. metre of cooling surface per degree centigrade per hour.

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With certain gas reactions, for example, with the Fischer-Tropsch synthesis, it may be advantageous, in view of the gradual decrease in the content of the effective or reactive constituents of the gas, to allow the temperature to increase in the upward direction, that is to say, to provide a positive temperature gradient in the direction of flow of the gas through the liquid medium. This may be attained either by lining the cooling surfaces, or by creating or providing a vapour or gas cushion between the tube, through which the coolant flows, and the reaction medium.

The cooling surface may be made smaller in known manner by reducing the diameter of the cooling tubes. Similarly, in the vertical direction, the number of cooling tubes may be reduced relatively to the horizontal cross-section by introducing from below more than one cooling tube into each shaft. In accordance with the number of cooling tubes introduced from below into the shaft, two or more cooling tubes may be merged into one another at one or more levels in such a manner that only a single cooling tube issues from the top/end of the shaft.

It has also been found to be advantageous to permit a steam cushion of variable depth to develop in the upper parts of the cooling tubes. Means to provide such a cushion of steam, or a cushion of the vapour of another liquid coolant,

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is diagrammatically illustrated by way of example in Figure 15. For the sake of clarity, the shafts have been omitted from the Figure. Immersion tubes 60 are provided in the cooling tubes 61. The cooling water which circulates through the steam drum 62 and the pump 63 may be passed or discharged wholly through the header 64, or, in dependence, upon the degree to which the temperature in the reaction space is increased, the cooling water may be passed or discharged more or less completely through the header 65; this may be controlled by adjusting the valves 66 and 67 accordingly. It is thus possible to maintain a cushion of steam or vapour in the intermediate space 68, with the length of the cushion or space being adjustable.

Alternative means for the adjustable reduction or regulation of the transmission of heat in the direction of flow of the reaction gases, is diagrammatically illustrated by way of example in Figure 16. The shafts are omitted from the Figure for the sake of clarity. The cooling tubes 71 are in their upper parts surrounded by tubes 72 which are open at their lower ends, each tube 72 being substantially concentric with the cooling tubes 71 on which it is mounted. The tubes 72 are connected through a header 73 and a valve 74 to the gas space above the upper ends of the shafts (not shown). The reaction gases can enter and flow through the annular space 75 disposed between the cooling tubes 71 and the tubes 72. The length of the cushion of gas which forms in the annular space 75 by removal of gas from the liquid medium which enters the space, is determined by opening the valve 74 to a greater or lesser extent.

In the reaction apparatus according to the invention, instead of using a heat-exchange system with a volatile coolant, it is also possible to use a non-volatile coolant. In this case the coolant is recycled upwardly through the cooling system of the reactor and through an external heat exchanger, in which, for example, the reaction heat withdrawn from the reactor is utilized for the generation of steam. The vertical temperature gradient in the shafts is then obtained by controlling the rate at which the coolant is circulated.

An additional advantage of operation with the reaction apparatus according to the invention lies in the fact that, in contrast with reaction apparatus in which the catalyst suspension circulates, deposits of catalyst on the walls are avoided. This appears to be due to the fine, intense turbulence of the liquid medium in the stationary distributing system.

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

1. A reaction apparatus for carrying out reactions between gaseous reactants in the presence of a catalyst dissolved or suspended in a liquid medium through which a mixture of the gaseous reactants is passed upwardly in the form of bubbles, comprising a vertical reactor having a diameter or width of not less than 30 centimetres and a length of not less than 1.5 metres, a number of substantially uniform and vertical members within the reactor dividing the major part of the cross-section of the reactor into a number of vertical shafts which are open at both ends and which have a diameter or width of at least 5 centimetres, the walls of the vertical shafts being substantially liquid-tight with their lower ends disposed at a distance above the inlet or inlets for the gaseous reactants not substantially less than the diameter or width of the reactor and with their upper ends terminating in the upper part of the reactor at one or more levels such that the surface of the liquid medium in each shaft is disposed within the shaft when the reactor is in operation with the liquid medium inflated by the bubbles of the gaseous reactants.

2. A reaction apparatus for carrying out gas reactions on an industrial scale in the presence of a liquid or solid catalyst which is dissolved or suspended in a finely distributed state in a liquid medium through which the gas is passed upwardly in the form of bubbles, comprising a vertical,

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cylindrical reactor having a diameter of more than 30 centimetres and a length of more than 1.5 metres which, as from a level above the gas intake which is at least as great as or greater than the diameter of the reactor, is divided over the greater part of its cross-section into substantially uniform vertical shafts having a diameter or width of at least 5 centimetres and the outer surfaces of which are impervious to liquid, the shafts being open at both ends with their upper ends terminating in the free gas space above the highest level which the liquid medium assumes when in the operating condition and inflated by the gas bubbles.

3. A reaction apparatus for carrying out reactions between gaseous reactants in the presence of a catalyst dissolved or suspended in a liquid medium through which a mixture of the gaseous reactants is passed upwardly in the form of bubbles, comprising a vertical, cylindrical reactor having a diameter of not less than 30 centimetres and a length of not less than 1.5 metres, a number of substantially uniform, open-ended shafts disposed within and extending longitudinally of the reactor, the members forming the walls of the shafts being liquid-tight, each shaft having an average diameter or width which is not less than 5 centimetres and being of a length which is from 10 to 200 times the average diameter or width of the shaft, the lower ends of the shafts being disposed at a

distance above the gas inlet or inlets which is not less than the diameter of the reactor whilst the upper ends of the shaft are disposed at a level above the highest level which the liquid medium assumes when inflated by the gas bubbles under the conditions of the reaction, and means for guiding the gaseous reactants into the lower ends of the shafts.

4. A reaction apparatus according to any one of claims 1, 2 and 3, in which the length of the shafts is from 20 to 100 times the average diameter or width of the shafts.

5. A reaction apparatus according to any one of claims 1, 2 and 3, in which each individual shaft is formed by its own separate casing.

6. A reaction apparatus according to any one of claims 1, 2 and 3, in which tubes of circular cross-section are used to form the shafts.

7. A reaction apparatus according to any one of claims 1, 2 and 3, in which the shafts are formed by regular hexagonal hollow prisms which are open at both ends.

8. A reaction apparatus according to any one of claims 1, 2 and 3, in which the lower ends of the shafts are disposed in the same horizontal plane.

9. A reaction apparatus according to claim 1 or claim 2, including baffle plates which are provided at a small distance below the lower edge of the bundle of shafts to prevent

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substantial amounts of gas from entering the space between the shafts and/or the space between the bundle of shafts and the inner wall of the reactor.

10. A reaction apparatus according to any one of claims 1, 2 and 3, including a member for controlling the liquid contents of the reactor, the said member being provided in the space disposed between the bundle of shafts and the inner wall of the reactor, said space being substantially unaffected by the passage of gas.

11. A reaction apparatus according to any one of claims 1, 2 and 3, including one or more means for the continuous or periodical withdrawal, from the space between the bundle of shafts and the inner wall of the reactor, of liquid reaction products of high molecular weight which are almost or substantially freed from catalyst and which are not discharged with the tail gas from the reaction space.

12. A reaction apparatus according to any one of claims 1, 2 and 3, including heat-exchange tubes for the indirect supply or removal of heat, the heat-exchange tubes being provided coaxially or otherwise parallel with the shafts and being of a length greater than the length of the shafts so that the position at which the heat-exchange tubes enter their headers is disposed outside the bundle of shafts.

13. A reaction apparatus according to any of claims 1, 2 and 3, including heat-exchange tubes

fcr the supply or removal of heat, the heat-exchange tubes being provided substantially parallel with the shafts and of a length greater than that of the shafts, the lower header or headers of the hext-exchange tubes being disposed at least 100 centimetres from the lower edge of the bundle of shafts.

14. A reaction apparatus according to any one of claims 1, 2 and 3, including heat-exchange tubes which pass longitudinally through the shafts and extend beyond the ends of the shafts, the minimum distance of the tubes from each other or from the inner wall of the shaft being more than 3 centimetres.

15. A reaction apparatus according to any one of claims 1, 2 and 3, including heat-exchange tubes which are provided substantially parallel with the shafts and of a length greater than the length of the shafts, at least some of the heat-exchange tubes being provided outside the shafts.

16. A reaction apparatus according to any one of claims 1, 2 and 3, including heat-exchange tubes which are provided substantially parallel with the shafts and of a length greater than the length of the shafts, all of the heat-exchange tubes being provided outside the shafts and in direct contact with the outer surfaces of the shafts.

17. A reaction apparatus according to any one of claims 1, 2 and 3, including gas inlet means

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through which the gas is introduced in such manner as to be substantially uniformly distributed over the total cross-section of the reaction space.

18. A reaction apparatus according to any one of claims 1, 2 and 3, including gas inlet means through which the gas is introduced in such manner as to be substantially uniformly distributed over the total cross-section of the reaction space, the nozzles, jets, perforations or like members or openings through which the gas passes into the reactor being so provided as to direct the entrant gas downwardly towards the bottom of the reactor.

19. A reaction apparatus according to any one of claims 1, 2 and 3, in which gas inlet means are provided in such manner as to permit the entrant gas to be directed towards the bottom of the reactor, particularly when a solid, suspended catalyst is used.

20. A reaction apparatus according to any one of claims 1, 2 and 3, in which the reaction space is tapered at a position below the lower edge of the bundle of shafts, to form or to locate a single, axial gas inlet.

21. A reaction apparatus according to any one of claims 1, 2 and 3, including means for supplying the gas inlet line, preferably at a position directly before entry into the reactor, with a medium which is liquid under the reaction

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conditions and by which the gas inlet openings are substantially continuously wetted.

22. A reaction apparatus according to any one of claims 1, 2 and 3, including means for the utilization of the latent heat of steam for heating the reaction space and/or for supplying the reaction heat in endothermic reactions, and for dissipating the heat of reaction in exothermic reactions.

23. A reaction apparatus according to any one of claims 1, 2 and 3, including means for providing a positive temperature gradient in the liquid medium along at least a part of the path of the gaseous reactants through the liquid medium.

24. A reaction apparatus according to any one of claims 1, 2 and 3, in which means are provided to vary the rate of heat transmission between the liquid medium and the coolant in a system of heat-exchange tubes provided longitudinally within the reactor, said means comprising a casing provided about each of the majority of the heat-exchange tubes or about each of the heat-exchange tubes, and means for maintaining a cushion of gas or vapour within at least a part of each casing.

25. A reaction apparatus according to any one of claims 1, 2 and 3, in which, for carrying out exothermic gas reactions in which it is desirable to provide a reaction temperature increase in the upward direction, the heat transmission in the

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cooling system from the liquid medium to the coolant is reduced in stages upwardly by using a system of cooling tubes, in which the tube through which the coolant flows is isolated from direct contact with the liquid medium by means of a casing, a gas or steam cushion of variable height being maintained between the tube for the coolant and the casing.

26. A reaction apparatus according to any one of claims 1, 2 and 3, in which, with exothermic gas reactions, the area of the cooling surface of the cooling system is reduced in stages in the upward direction by reducing the diameter of the cooling tubes in the upward direction and/or by reducing the number of cooling tubes in the upward direction.

27. A reaction apparatus according to any one of claims 1, 2 and 3, including a heat-transmitting system in the reactor and a heat exchanger provided outside the reaction space in series with the heat-transmitting system, through which system and heat-exchanger a liquid which does not vaporise under the reaction conditions can be circulated.

28. A reaction apparatus according to any one of claims 1, 2 and 3, including a heat-transmitting system in the reactor and a heat exchanger provided outside the reaction space in series with the heat-transmitting system, and positive means for controlling the rate of circu-

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lation of a liquid, which does not vaporise under the reaction conditions, through the heat-transmitting system and the heat exchanger, whereby a vertical temperature gradient in the reaction space may be maintained and varied.

29. A reaction apparatus according to any one of claims 1, 2 and 3, in which the diameter or width of each of the shafts is not greater than 30 centimetres.

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FIG. 1

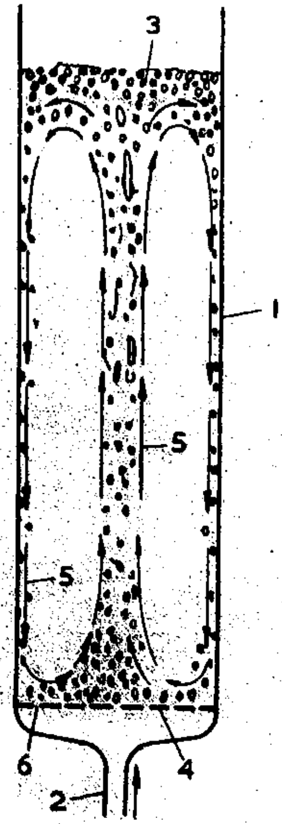


FIG. 2

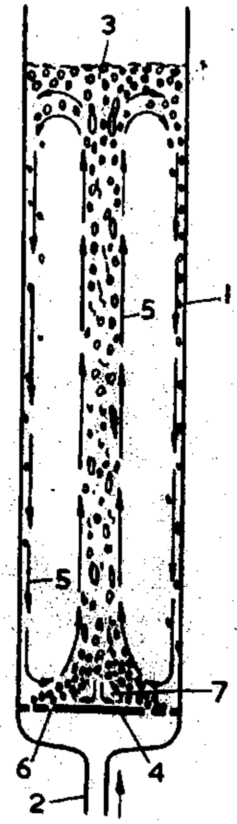
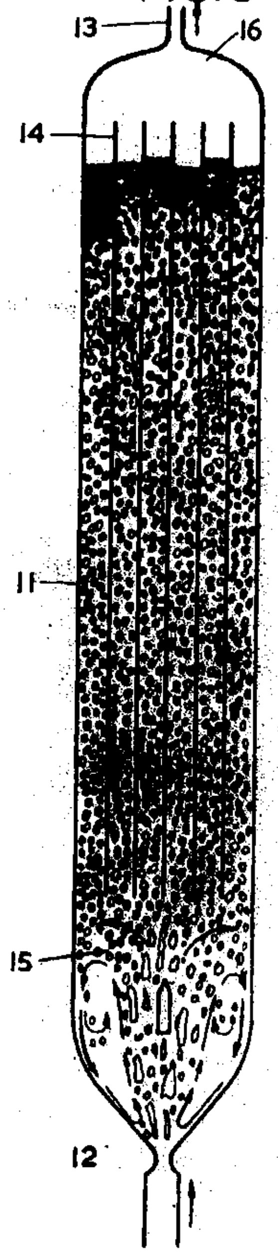


FIG. 3



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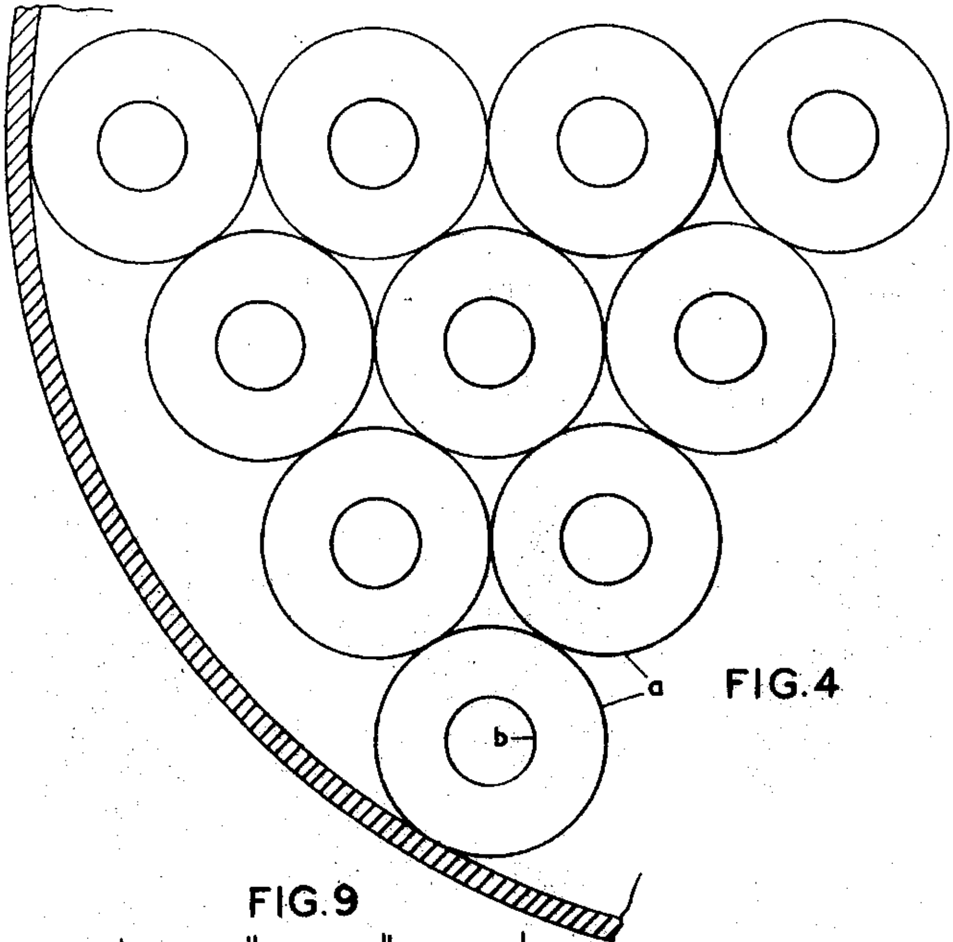


FIG.4

FIG.9

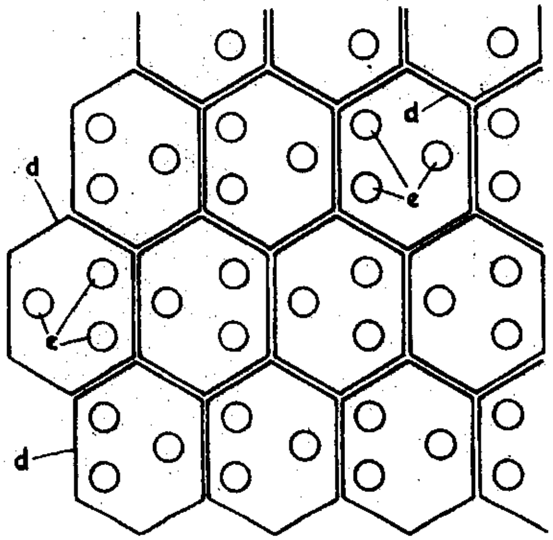
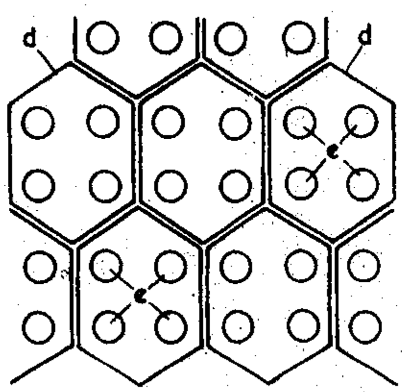


FIG 10



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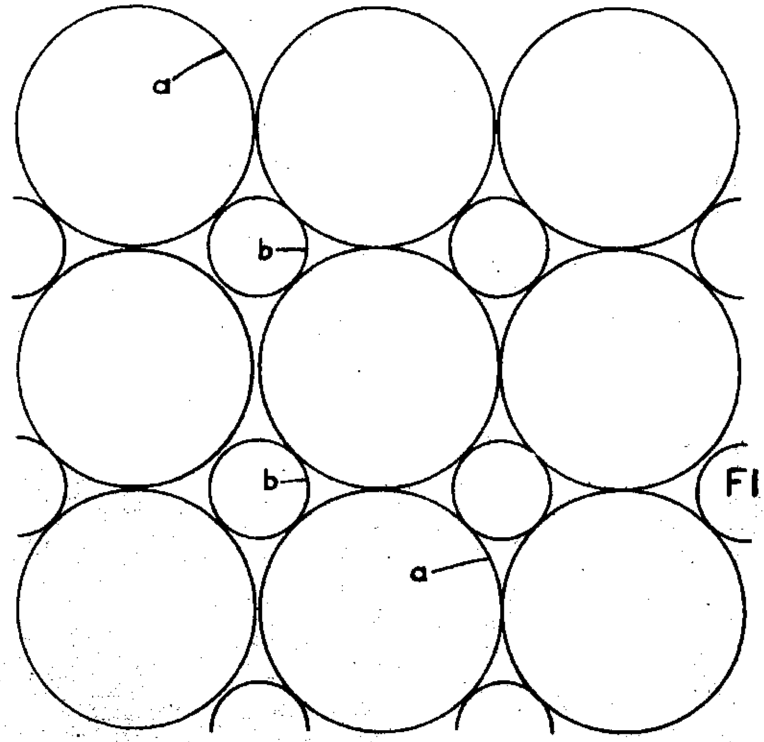

FIG.5

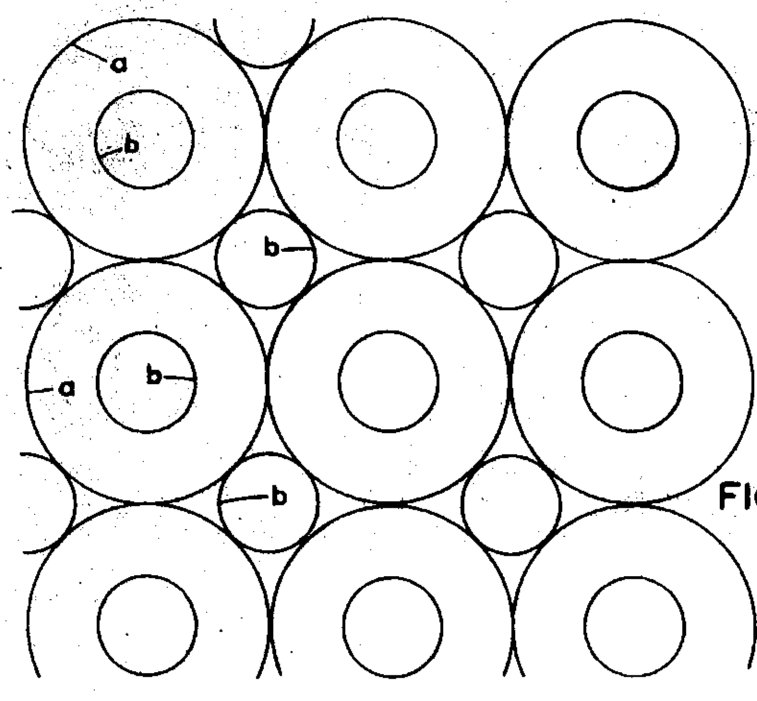

FIG.6

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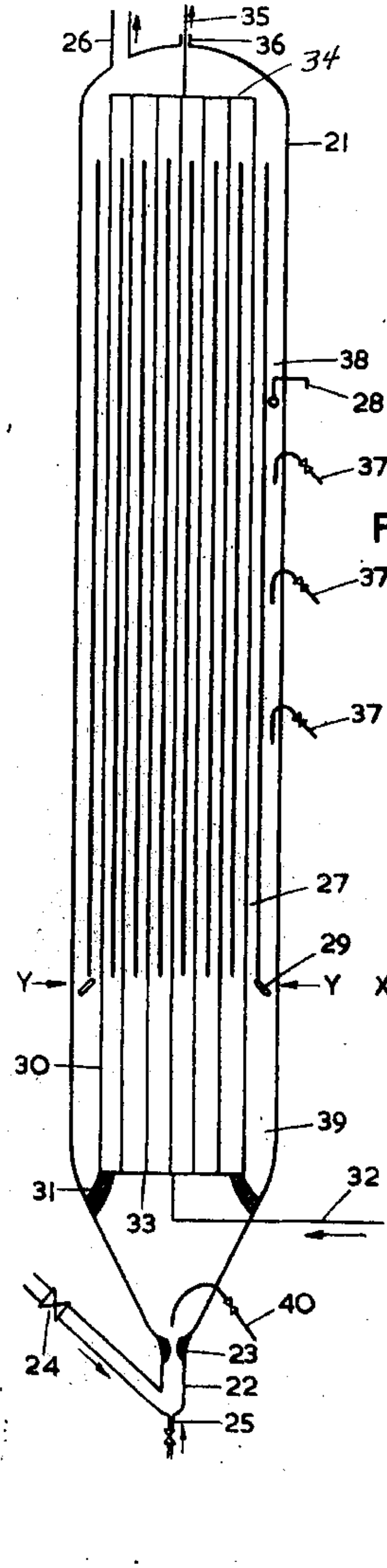

FIG. 7

FIG. 8

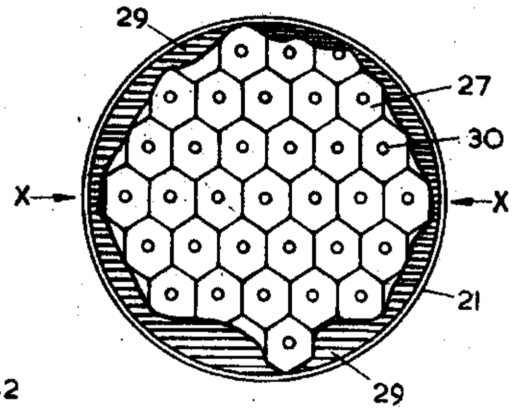

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FIG. 11

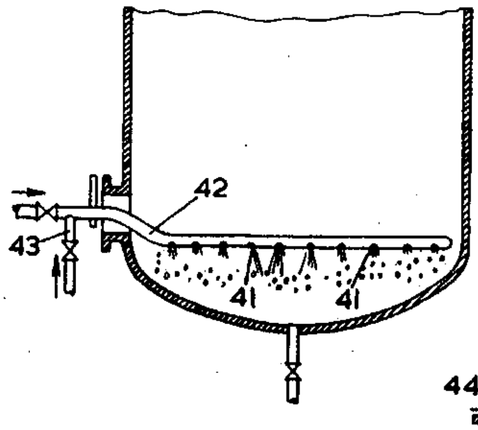


FIG. 13

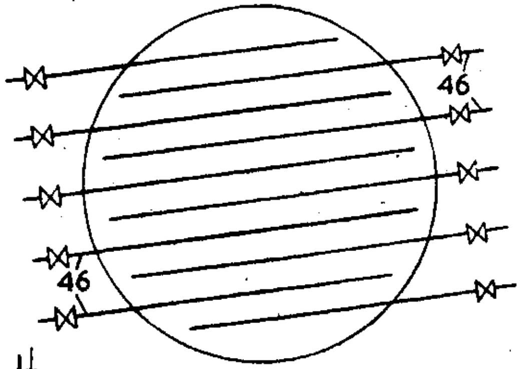


FIG. 12

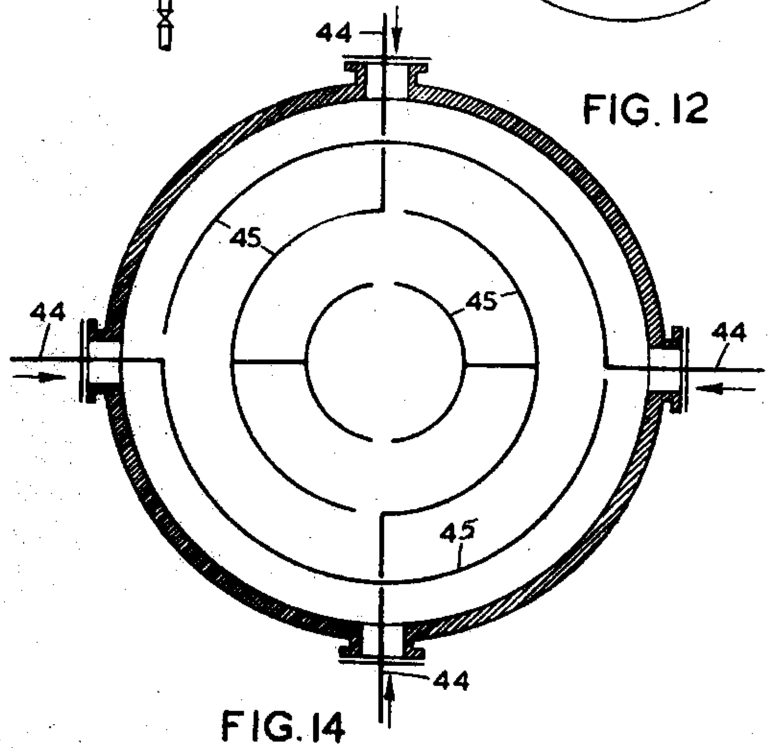
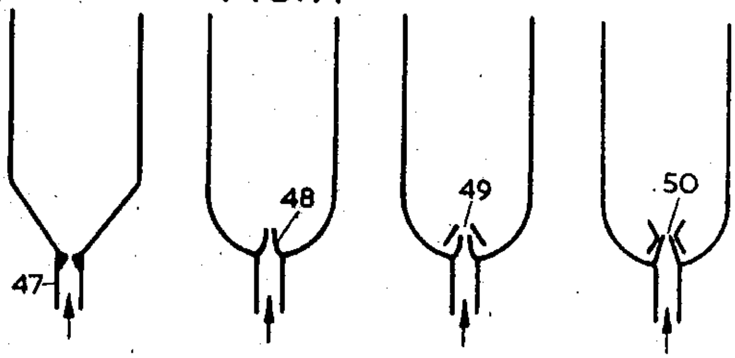


FIG. 14



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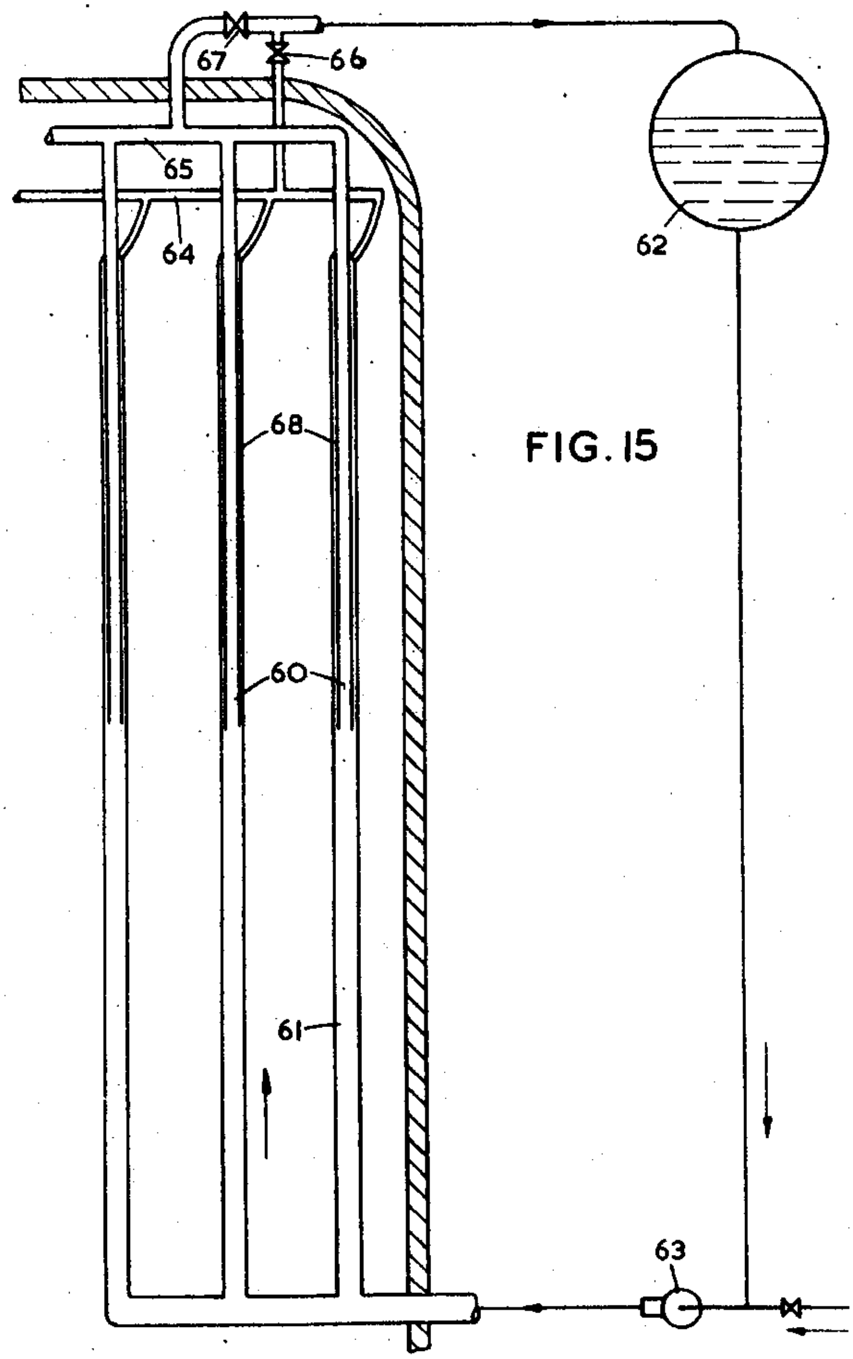

FIG. 15

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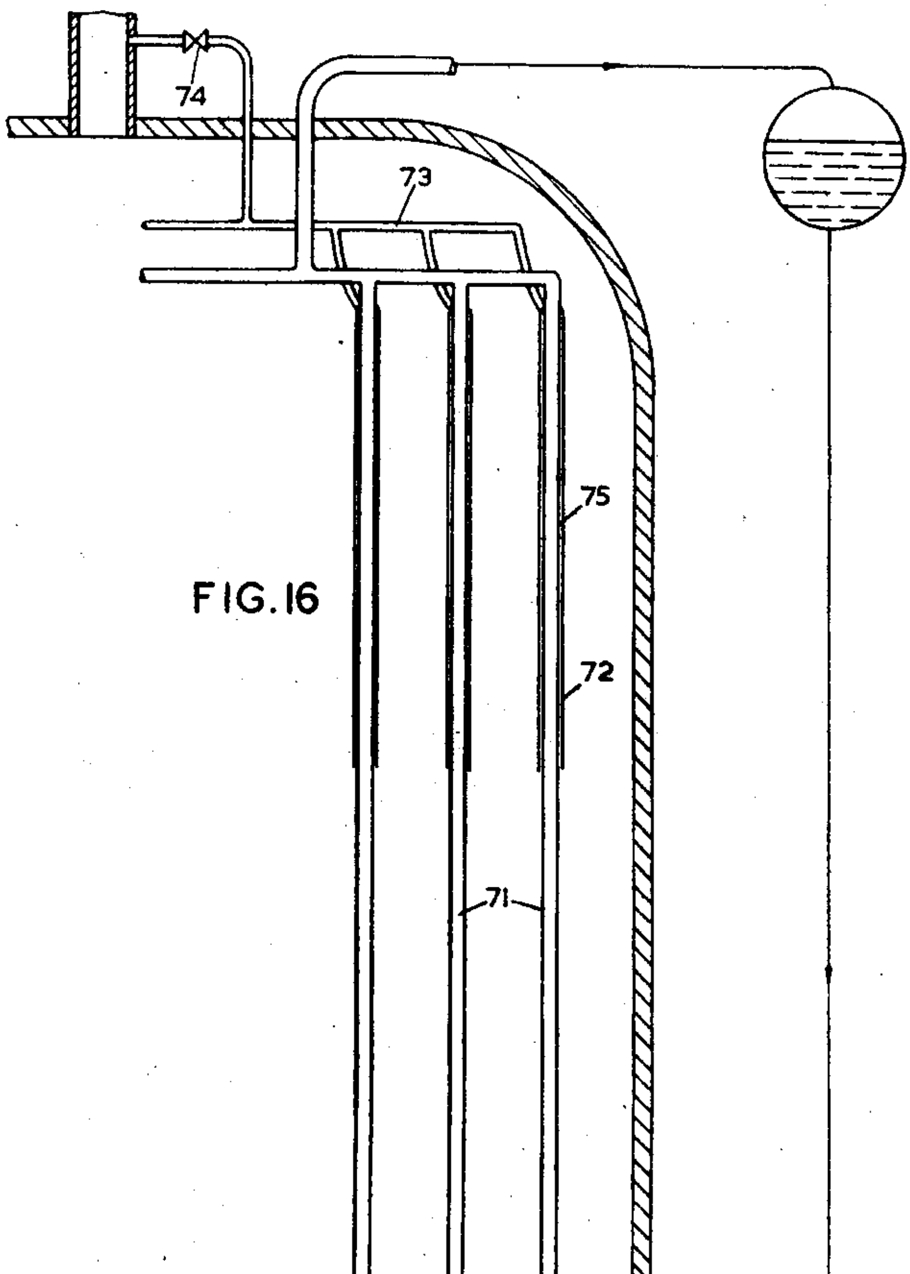

FIG. 16

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