PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Reaction apparatus for carrying out Exothermic Gas Reactions

We, RHEINPREUSSEN AKTIENGESELLSCHAFT FUER BERGBAU UND CHEMIE, a German Company, of Homberg, Niederrhein, Germany, 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 apparatus for carrying out exothermic catalytic gas reactions in the presence of catalysts dissolved or suspended in a non-circulating liquid medium, in which an internally cooled reaction chamber, being relatively tall as compared with the diameter, is divided into vertical shafts which are open at both ends with the lower ends of the shafts terminating above the base of the reaction chamber within the liquid medium and with the upper ends of the shafts extending above the liquid medium into the gas space, and which are formed by heat-conducting plates connected to cooling tubes or pipes through which the coolant flows upwardly in the same direction as the flow of gas through the reaction chamber.

In catalytic gas reactions, particularly in the hydrogenation of carbon monoxide in accordance with the Fischer-Tropsch process when it is desired to obtain a substantially complete conversion of the synthesis gas in one step and with a single pass of the gas, it has been found to be advantageous to allow the reaction temperature to increase in the direction of flow of the gas and in proportion to the impoverishment of the gas in initial reaction constituents, such as CO and H_2. By adjusting the reaction temperature to the partial pressure of the reactants, not only is higher gas conversion obtained, but undesirable side-reactions such as the formation of methane or the separation of carbon, are suppressed, whilst the quality of the synthesis products is at the same time improved.

A number of proposals have been made to obtain the rise of temperature, that is, a positive temperature gradient, in reaction chambers provided with internal cooling means. Thus, according to one proposal, a number of cooling elements are provided one above the other through each of which flows a separate stream of coolant and to each of which a separate vapour collector or drum is connected if a vaporizing coolant, such as water, is used. According to another proposal, a non-vaporizing coolant is passed at a controlled rate through tubes extending over the total height of the reaction chamber in such manner that the coolant is heated to the desired degree and the reaction heat is transferred to water in an external heat exchanger, thereby generating steam. Both proposals necessitate additional devices which complicate the cooling operation. It is also possible to obtain a rise of temperature or a positive temperature gradient by reducing the number or the cross-sections of the cooling tubes, so that the area of the cooling surfaces decreases in the direction of flow of the gas, and the distance between the cooling areas is increased. However, when the coolant used is water, and in view of the fact that the temperature of the water is uniform throughout, there will occur within the reaction chamber horizontal differences in temperature which increase in the direction of flow of the gas stream and which result in disadvantageous effects on the reaction.

It is an object of the invention to provide a reactor in which such disadvantages are avoided or mitigated.

According to the invention, a reactor for carrying out exothermic gas reactions in the presence of a catalyst dissolved or suspended in a liquid medium comprises a reaction chamber provided at its lower end with means whereby the gaseous reactants may be passed upwardly through the liquid medium in the reaction chamber, one or more bundles of cooling tubes through which a liquid heat transfer medium may be passed, the bundle or bundles of cooling tubes being disposed in the main reaction zone in the lower two-thirds of the reaction chamber, and a series of heat-conducting plates connected to the cooling tubes and forming a series of open-ended vertical shafts which extend substantially over the whole cross-section of the reaction chamber, each vertical shaft being of relatively small cross-sectional area, the lower ends of the heat-conducting plates being disposed above the bottom of the reaction chamber and within the liquid medium, and the upper ends of the heat-conducting plates extending above the upper level of the cooling tubes to a position adjacent to the top of the reaction chamber to form extensions of the vertical shafts above the surface of the liquid medium. The length of the main reaction zone is generally from about one-third to at most two-thirds of the height of the reaction chamber, the actual length of the main reaction zone depending on the specific reaction and on the conditions under which the reaction is carried out. In one construction the bundle or bundles of cooling tubes may be disposed wholly within the lower third of the total height of the reaction chamber. In another construction, the cooling tubes may be so provided that they are reduced in number in the upward direction from approximately a mid-position of the main reaction zone.

The vertical shafts may be circular in cross-section, and of a diameter in the range 5-30 centimetres depending on the length of the vertical shafts and the diameter or cross-sectional area of the reactor.

In the hydrogenation of carbon monoxide in a liquid medium, it appeared that approximately 70% of the conversion occurred relatively rapidly in the lower third or at most within the lower half of the layer required for a complete conversion. From this zone, there is obtained a larger proportion of the total reaction heat utilizable for the generation of steam or vapour than corresponds to the respective proportion of converted gas, because it has been found that a large portion of the free reaction heat is used in the upper zone for the required increase of temperature. The weak thermal flux to the cooling tube zone, which moreover decreases in the upward direction within the heat-conducting plates which forms the walls of the vertical shafts, permits a second wave of reaction to occur in synthesis gas in the upper zone which is free from cooling tubes in spite of the fact that the concentration of the reactive components in the synthesis gas is much lower in this zone due to the initial reaction in the main reaction zone. A substantial rise of temperature thus occurs in the upper part of the reactor with the result that the $CO + H_2$ in the gas can be converted almost completely and almost without any disturbing side-reactions.

It has also been found that the reactor is particularly suitable for the production, from CO and H_2, of low-boiling synthesis products which are rich in olefins and substantially enriched in iso-hydrocarbons. Due to the high temperature in the upper reaction zone, these products are obviously discharged more rapidly and more completely from the reactor. The temperature of the end gas escaping at the top of the reactor which is up to 50°C., or more higher than the mean temperature of the main reaction zone, renders it possible to preheat the synthesis gas, which is fed into the bottom of the reactor, in heat exchangers almost to the initial synthesis temperature. Any excess reaction heat of the second or upper reaction zone which cannot be dissipated through the heat-conducting plates to the cooling tubes, is, therefore, also utilized in the form of vapour for the preheating of the synthesis gas.

The invention is illustrated by way of example in the accompanying drawings, in which

Fig. 1 is a perspective view of the upper part of one construction of a set or bundle of shafts at the level of the collecting pipes through which the cooling medium passes out of the reaction chamber;

Fig. 1A is a transverse section on the line A—A of Fig. 1;

Fig. 2 is a perspective view of part of a further construction of a set or bundle of shafts;

Fig. 2A is a transverse section on the line B—B of Fig. 2; and

Fig. 3 is a perspective view, at the lower ends of the shafts, of some of the feed tubes for the cooling medium.

As illustrated in Figs. 1 and 2, a series of heat-conducting plates 11 are connected together in such manner as to form a series of liquid-tight, hexagonal, vertical shafts 10. The vertical shafts 10 are open at their upper and lower ends, the lower ends of the shafts terminating at a position above the base or gas distributing plate of the reactor, whereby the liquid medium, when the reactor is in use, may circulate below the lower ends of the vertical shafts. Cooling tubes 12 are provided to run vertically at the angles of the shafts 10, the heat-conducting plates 11 being joined to the tubes 12 in a manner which ensures that the joints are both liquid-tight and efficient conductors of heat. The tubes 12 extend from the lower ends of the shafts 10 to a position short of the upper ends of the shafts, such position being approximately at the upper end of the main reaction zone; above such position the reaction space is free from cooling tubes and the plates 11 are directly joined to one another to form the liquid-tight shafts 10. The tubes 12 are connected at their upper ends to horizontal, collecting pipes or tubes 13, the pipes 13 being alternately staggered in the construction illustrated in Fig. 1 and being all at the same level in the construction illustrated in Fig. 2. It will be understood that the collecting pipes 13 may be provided at a slight angle to the horizontal in order to avoid vapour or steam pockets.

When the reactor is in operation, the cooling medium flows upwardly through the tubes 12 and passes into the collecting tubes 13 through which it is led out of the reactor. The collecting pipes 13 may, for example, lead into a main collecting pipe or header which is formed to a circular arc and bears on the inner wall of the reaction chamber and, to provide a resilient equilization of differences in thermal expansion of the cooling tube system and the wall of the reactor, the pipe connecting the main collecting pipe to the vapour or steam drum is preferably of adequate length and leads from the reaction chamber to the outside only at the end of a sufficiently long pipe connection which is mounted on the outer wall.

Instead of providing only a single arcuate main collecting pipe, it is also possible to use two or more arcuate main collecting pipes which are provided opposite and/or above one another. The pipes leading to the vapour or steam collector or drum may also extend upwardly at right angles from the main collecting pipe or pipes to pass, advantageously through glands or stuffing boxes, through the head or top of the reaction chamber to the outside.

The cooling medium is passed into the cooling tubes 12 through feed pipes 14 which are provided below the lower ends of the vertical shafts 10, (Fig. 3).

The formation of the feed pipes 14 is similar to that of the collecting pipes 13 which are provided at the upper ends of the tubes 12. It will be appreciated that the feed pipes 14, which extend horizontally across the lower part of the reaction chamber, do not in any way impede or interfere with the distribution of the synthesis gas in the liquid medium in which the synthesis is effected. The gas distributing jets or nozzles may be provided uniformly over the whole of the base of the reaction chamber, without any regard to the position of the transverse feed pipes 14. The feed pipes 14 may themselves also rest directly on the gas distributing plate at the base of the reaction chamber, in which case the gas distributing nozzles may advantageously be provided adjacent to the feed pipes 14 and/or the cooling tubes 12.

The lower ends of the vertical shafts 10 are spaced from the base of the reaction chamber, so providing a path at the base of the reaction chamber along which the liquid medium, in which the synthesis or reaction is carried out, may circulate.

The diameter of the cooling tubes 12 is small relatively to the diameter or width of the shafts 10, being less than 1 :2.

In reactors, the reaction chambers or spaces of which have a total or overall height of from about 5 metres to about 12 metres, one cooling unit or bundle of cooling tubes is sufficient to dissipate all of the excess reaction heat, since in this case the process is only carried out with cross-sectional loads of up to approximately 40 working litres of synthesis gas per square centimetre of cross-section of the reaction space per hour. The term "working litre" denotes a litre of the gas under the temperature and pressure conditions of the reaction.

However, when reactors are used in which the total or overall height of the reaction chamber or space exceeds 12 metres, and which may, for example, be up to 25 metres, it has been found to be advantageous to divide the cooling tube system, which in accordance with the invention is provided in the main reaction zone, into two or more sets or bundles of cooling tubes disposed one above the other, each of which is advantageously connected to its own vapour or steam collector or drum, and each of which is preferably provided with its own temperature regulating means, so that even within the main reaction zone the synthesis temperature may be increased in stages in the upward direction.

In order to obtain a substantially constant space-time yield of reaction products, it is necessary for the cross-sectional load of the synthesis gas to be increased in accordance with the extent to which the height of the reaction chamber is increased. For example, for the same space-time yield, and with otherwise constant conditions of pressure and temperature, the cross-sectional load of synthesis gas required in a reaction chamber having a height of 25 metres. is five times as great as that required in a reaction chamber having a height of 5 metres. With tall reaction chambers, the cross-sectional load may be increased to 100 working litres per sq. cm., and the length of the main reaction zone cooled by a cooling medium is correspondingly extended in view of the high velocity of ascent of the gas bubbles. In view of this, the percentage gas conversion per unit of length in the main reaction zone is correspondingly less than in low reactors so that on this account and also in view of the more vigorous turbulence of the liquid medium, the reaction temperature can only rise by accumulation of heat to a correspondingly slight degree above the temperature of the cooling medium.

Therefore, with increasing height, the rise in the reaction temperature within the region of the main reaction zone necessary to maintain an adequate conversion of gas, is advantageously ensured by providing several cooling tube units as proposed according to the invention in the manner hereinbefore described. In this reactor, a subsequent or second wave of reaction also occurs in the manner hereinbefore described in the zone in which the vertical shafts are free from the cooling tubes.

What we claim is:—

1. A reactor for carrying out exothermic gaseous reactions in the presence of a catalyst dissolved or suspended in a liquid medium, comprising a reaction chamber provided at its lower end with means whereby the gaseous reactants may be passed upwardly through the liquid medium in the reaction chamber, one or more bundles of cooling tubes through which a liquid heat transfer medium may be passed, the bundle or bundles of cooling tubes being disposed in the main reaction zone in the lower two-thirds of the reaction chamber, and a series of heat-conducting plate sconnected to the cooling tubes and forming a series of open-ended vertical shafts which extend substantially over the whole cross-section of the reaction chamber, each vertical shaft being of relatively small cross-sectional area, the lower ends of the heat-conducting plates being disposed above the bottom of the reaction chamber and within the liquid medium, and the upper ends of the heat-conducting plates extending above the upper level of the cooling tubes to a position adjacent to the top of the reaction chamber to form extensions of the vertical shafts above the surface of the liquid medium.

2. A reactor according to Claim 1, in which the bundle or bundles of cooling tubes are disposed wholly within the lower third of the total height of the reaction chamber.

3. A reactor according to Claim 1, in which the cooling tubes are reduced in number in the upward direction from approximately a mid-position of the main reaction zone.

4. A reactor according to any one of the preceding claims, in which the collecting pipes through which the cooling medium flows out from the cooling tubes, extend horizontally, or upwardly at a slight angle to the horizontal, through the system of shafts.

5. A reactor according to any one of the preceding claims, in which with very tall reaction chambers and/or when the reaction requires a sharper rise in temperature in the direction of flow of the gas stream, two or more sets of cooling tubes are provided one above the other in the main reaction zone, each of which sets is provided with its own temperature regulating means.

6. A reactor for carrying out exothermic reactions with gaseous reactants, substantially as hereinbefore described with reference to Figs. 1 and 3 or to Fig. 2.

7. A process for the synthesis of hydrocarbons from carbon monoxide and hydrogen by the Fischer-Tropsch process, in which the synthesis is carried out in the presence of a catalyst in a liquid medium in the reactor claimed in any one of the preceding claims.

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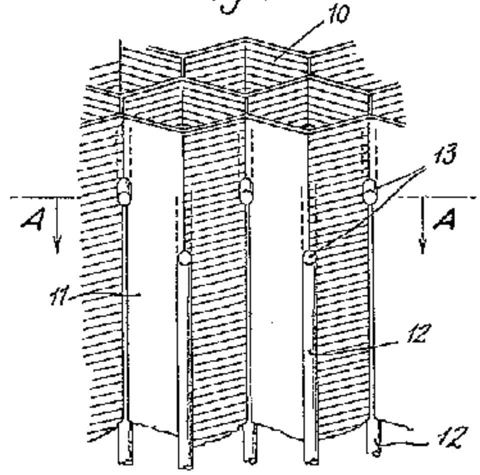
Fig. 1



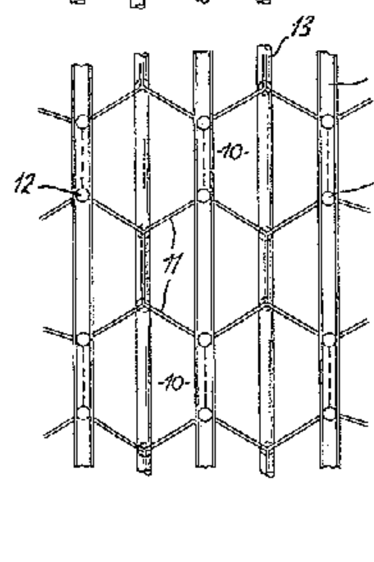


Fig. 1a

13
A
12
12
13
12

Fig. 2

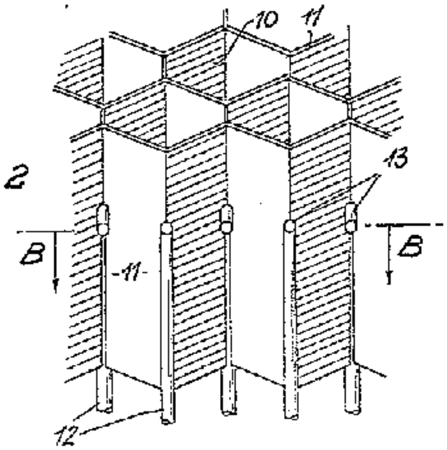


Fig. 2a

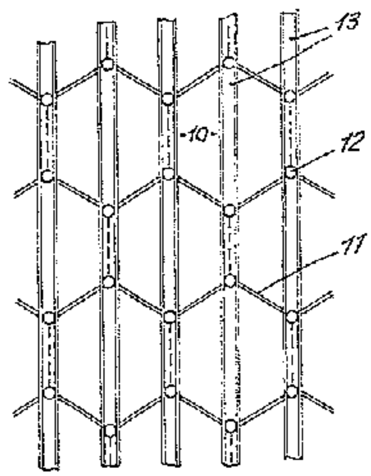
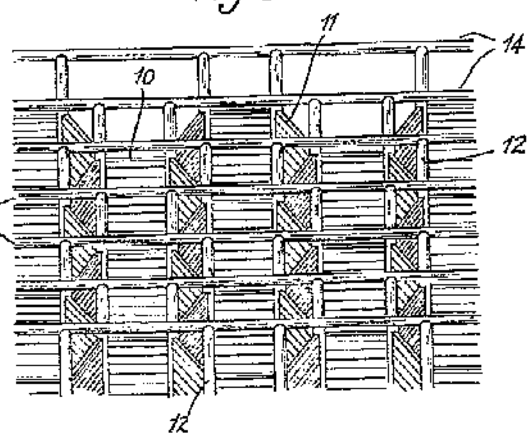


Fig. 3



785752 2 SHEETS

COMPLETE SPECIFICATION
This drawing is a reproduction of the Original on a reduced scale
Sheets 1 & 2

Fig. 2

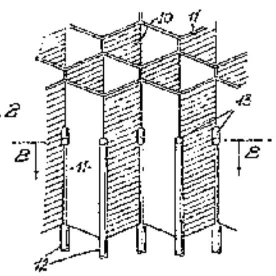


Fig. 2a

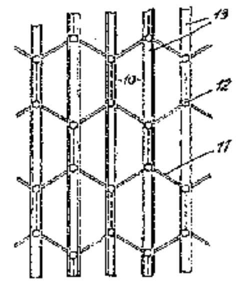


Fig. 3

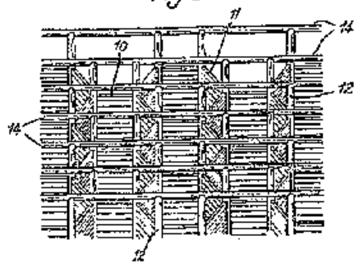


Fig. 1

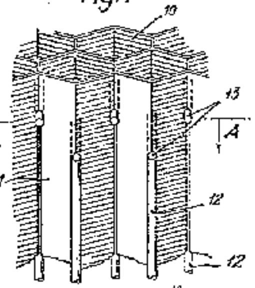


Fig. 1a