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(54) REACTION APPARATUS FOR CARRYING OUT EXOTHERMIC AND ENDOTHERMIC GAS REACTIONS

(54) APPAREIL A REACTION POUR ACCOMPLIR DES REACTIONS EXOTHERMIQUES ET ENDOTHERMIQUES DE GAZ

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The invention relates to a development of the apparatus described in Federal German Patent Specification No. 948,781 for carrying out exothermic catalytic gas reactions in the presence of catalyst dissolved or suspended in a non-circulating liquid medium, in which the internally cooled reaction chamber, being relatively tall as compared with its diameter, is divided into vertical shafts. The vertical shafts are open at the top and at the bottom, and the lower ends of the shafts end freely above the bottom of the reaction chamber and within the liquid medium, whilst their top ends extend above the liquid medium into the gas space. The vertical shafts are formed by heat-conducting plates connected to cooling tubes or pipes, the coolant flowing upward through the cooling tubes in the same direction as the stream of gas.

In catalytic gas reactions, particularly in the hydrogenation of carbon monoxide in accordance with the Fischer-Tropsch process, and particularly 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 an increased or higher gas conversion obtained, but moreover undesirable side-reactions such as the formation of methane or the separation of carbon, are suppressed, while the quality of the synthesis products is at the same time improved.

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A number of proposals have been made to obtain the rise of temperature, that is, a temperature gradient, in reaction chambers provided with internal cooling. Thus, for example, a number of cooling elements are provided one above the other through each of which flows a separate stream of coolant and to which a separate vapour collector or drum is connected if a vaporizing coolant, such as water, is used. It is also possible to conduct a non-vaporizing coolant in cooling tubes at a controlled rate through the total height of the reaction chamber in such manner that it is heated to the desired degree and the reaction heat can be transferred to water in an external heat exchanger, thereby generating steam. Both proposals necessitate additional devices or means which complicate the cooling operation. It is also possible to attain a rise of temperature or 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 bring about disadvantageous effect on the reaction.

It has now been found that, when reactors for catalytic exothermic gas reactions in a liquid medium are used, in which the excess reaction heat is dissipated from the reaction chamber by allowing a substantially uniformly boiling cooling medium to vaporize, and the cooling medium flows upwardly through the cooling tubes in the direction of flow of the stream of synthesis gas, the vertical cooling tubes being interconnected by vertical heat-conducting plates in such manner that the reaction chamber is divided into a number of shafts of a small diameter which are open at the bottom and at the top and in liquid-tight connection, and below which the liquid catalyst suspension can freely communicate or flow over the entire bottom surface, and which end above the liquid medium in a common gas space, a rise of the reaction temperature, from the upper end of the main reaction zone upwardly, which is substantially uniform over the total cross-sectional area of the reaction chamber, and which being rapid at first dies slowly away in the upward direction, may be obtained if the path for the thermal flux or flow of heat is caused to grow within the cooling plates towards the cooling tubes by means of having the cooling tubes extend from the bottom only as far as up to the level of the main reaction zone without, however, reducing the cooling area from the main reaction zone upwards. The length of the main reaction zone may be assumed to occupy approximately one-third to at most two-thirds of the height of the reaction chamber, in dependence upon or according to the type and method of operation of the reaction to be carried out.

In the hydrogenation of carbon monoxide in a liquid medium, it appeared that approximately 70% of the conversion occurred relatively rapidly already in the lower third up to half the height of 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 consumed or used in the zone above for the required increase of temperature. The weak thermal flux to the cooling tube zone, which moreover decreases in the upward direction within the plates by which the shafts are bounded, renders it possible for a second reaction wave of the gas, already relatively impoverished in CO and H_2, to be started in the upper reaction zone, which is free from cooling tubes, of the reactor according to the invention, so that the gas is almost completely converted under a substantial rise of temperature and almost without any disturbing side-reactions.

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

The invention is illustrated by way of example in the accompanying drawings. Figures 1 and 2 show two constructions, by way of example, of the set or bundle of shafts at the level of the collecting pipes for the discharge or carrying away of the cooling medium. In both cases, the collecting pipes are horizontal. However, to avoid vapour or steam pockets, they may also be provided at a slight inclination so that they rise gently. Figure 1 shows the cross-section and the longitudinal section of part of a bundle of shafts and cooling tubes, with horizontal collecting pipes the levels of which are alternately staggered relatively to each other.

All of the collecting pipes illustrated in Figure 2 are on the same level.

Thus, for example, the linear collecting pipes may be introduced into a main collecting pipe which is formed as a circular arc and bears on the inner wall of the reaction chamber and, to provide a resilient equalization of differences in thermal expansion of the cooling system and the wall of the reactor, the pipe connecting the main collecting pipe to the vapour or steam collector 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 feed pipes leading to the vapour or steam collector or drum may also extend at right angles, advantageously through glands or stuffing boxes, through the head or top of the reaction chamber to the outside.

The grouping of the feed pipes for the cooling medium below the shafts, as illustrated by way of example in Figure 3 which shows a part of the view from below, corresponds to the grouping of the linear collecting pipes above the cooling tubes. The horizontally extending feed pipes do not impede or interfere with the distribution of the gas in the liquid medium. The gas distributing jets or nozzles may be provided uniformly over the whole bottom surface below the shafts without it being necessary to have regard to the transverse tubes or pipes. The feed pipes may also rest directly on the gas distributing plate, in which case the gas-distributing jets or nozzles are advantageously provided adjacent to the cooling tubes.

In reactors, the reaction chambers or spaces in which have total heights of up to approximately 5 to 12 metres, a single cooling tube unit or system 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 operational litres of synthesis gas per square centimetre of cross-section of the reaction space per hour.

However, when reactors are used in which the total or overall height of the reaction chamber or space exceeds 12 metres, and is for example 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 within the range or region of the main reaction zone, into two or, in given cases, 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, so that already 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 high 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 operational 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. However , already in the main reaction zone the percentage of converted gas or gas conversion per unit of length is correspondingly less than in low reactors so that 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 installation, the subsequent reaction also occurs in the manner hereinbefore described at the level of the shaft extensions which are free from cooling tubes.

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

1. A reactor for carrying out 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 or 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 tubes and forming a series of vertical shafts which extend substantially over the whole cross-section of the reaction chamber and which are open at their top and bottom ends but are otherwise substantially liquid-tight, each vertical shaft being of relatively small cross-sectional area, the lower ends of the heat-conducting plates being clear of the bottom of the reaction chamber to permit free flow of the liquid medium, and the upper ends of the heat-conducting plates extending above the upper level of the 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 for carrying out exothermic gas reactions in the presence of catalysts which are dissolved or suspended in a non-circulating

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liquid medium, including an internally cooled reaction chamber divided into vertical shafts open at the bottom and at the top, the vertical shafts ending freely at their lower ends in the liquid medium and with their upper ends extending above the liquid medium into the free gas space, which vertical shafts are formed by heat-conducting plates connected to cooling tubes through which a cooling medium to be vaporised and having a uniform boiling point flows upwardly in the direction of flow of the gaseous reactants, characterised in that, without changing the size of the cooling surface in the direction of flow of the gaseous reactants, the middle path for the thermal flux within the heat-conducting plates towards the cooling tubes is extended from the level of the main reaction zone to the upper end of the reaction chamber.

3. A reactor according to claim 2, in which in the upward direction, the cooling tubes are only connected to the heat-conducting plates as far as up to from one-third to two-thirds of the total height of the reaction chamber so that the upper ends of the heat-conducting plates are free from cooling tubes.

4. A reactor according to any one of claims 1, 2 and 3, in which the number of cooling tubes connected to the heat-conducting plates is reduced from approximately a mid-position of to the upper end of the main reaction zone.

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5. A reactor according to any one of claims 1, 2 and 3, in which 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.

6. A reactor according to any one of claims 1, 2 and 3, in which 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.

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Fig. 1

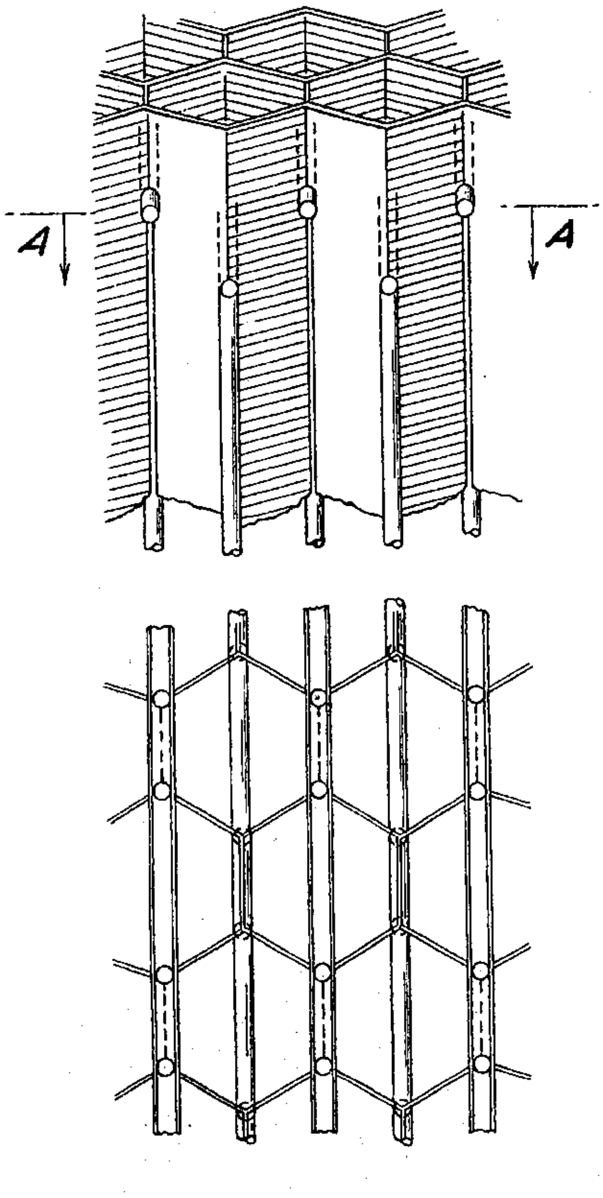

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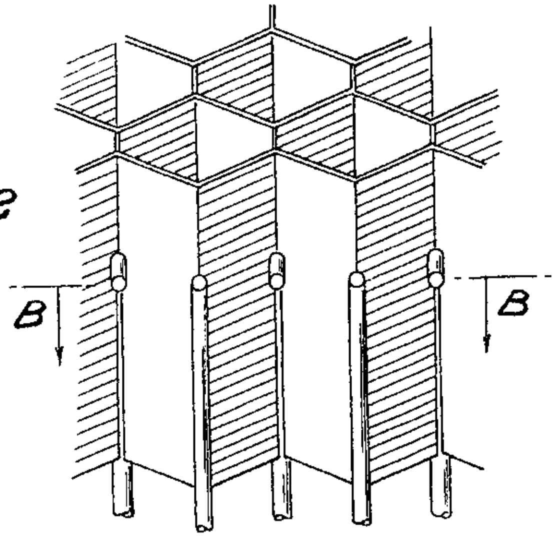
Fig. 2



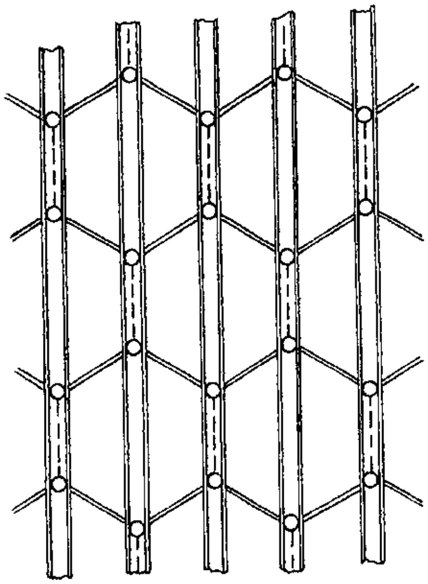
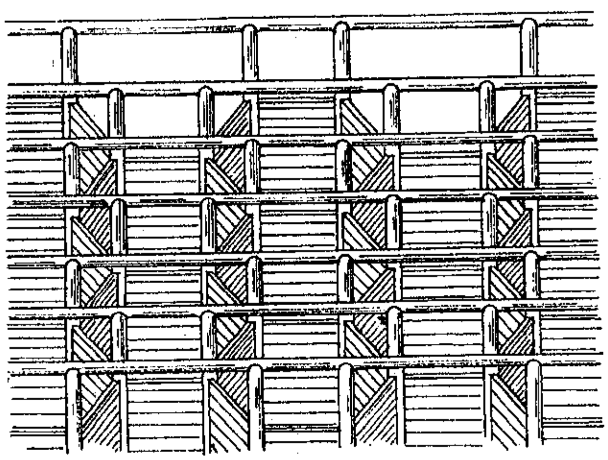


Fig. 3



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