PATENT SPECIFICATION

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451,177

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COMPLETE SPECIFICATION

3662

Improvements in Apparatus for Carrying out Catalytic Gas Reactions

We, STUDIEN-UND VERWERTUNGS-GESELLSCHAFT MIT BESCHRÄNKTER HAFTUNG, of Mülheim-Ruhr, Germany, a German Company, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

To carry out catalytic gas reactions by the use of a circulating liquid medium for maintaining the temperature, it is necessary to employ apparatus in which the contact composition is uniformly penetrated by conduits through which the circulating oil flows but which is completely liquid tight and which at the same time can be readily charged with and cleared or freed of the catalytic substances. Certain reactions particularly sensitive to the influence of temperature, such, for example, as the reduction of carbon monoxide to hydrocarbons which contain more than two carbon atoms in each molecule, can be carried out only if the mass of catalyst present between adjacent conduits is of small thickness, which on practical grounds may vary only within a few millimetres as, for example, between 10 and 15 millimetres. It has been found that this essential condition, that is to say, the exact maintenance of the thickness of the intervening masses of the catalyst in the case of very thin but widely distributed masses, as well as the necessary fluid tightness of the apparatus offer special difficulties when carrying out known processes on a large scale, for example, when the catalyst is disposed between oil-jacketed pocket-shaped cavities.

It has been found that these difficulties do not arise if conduits of non-circular section, that is to say, for example, conduits of elliptical, rhombic or rectangular section be used in the construction of the apparatus.

It has been proposed to make use of tubes which are circular in cross-section in the construction of such contact apparatus, as, for example, in boilers known as "tube boilers" the oil being inside and the catalyst substances outside—that is to say, between—the tubes, for the better utilisation of the available space. It is, however, easy to understand that a filling of the catalyst substances of very uneven and variable thickness only is possible between tubes which are circular in cross-section.

This is not the case if shallow non-circular conduits are provided instead of tubes which are circular in cross-section. For example, seamless drawn steel tubes of elliptical, rectangular, rhombic or similar shallow cross-sections may be provided side by side in parallel ranks, the tubes in adjacent ranks being advantageously arranged in such a manner that the tubes of the said ranks are mutually displaced through a distance of, for example, half the pitch of the tubes in a rank and disposed so that they partially overlap.

The invention consists in providing apparatus for carrying out catalytic gas reactions by the use of a circulating liquid medium for maintaining the temperature wherein the liquid medium is caused to pass through narrow conduits, the cross-section and disposition of which are such that the smallest distances between the conduits, taken from any particular point on the periphery of any particular conduit to the adjacent conduit nearest the said point, are approximately equal, so that the thickness of catalyst between any one conduit and the adjacent conduits is uniform or substantially so, and the catalyst can pass downwardly by gravity through the set of conduits when the apparatus is being filled or emptied.

Oil is advantageously used as the circulating liquid medium.

The conduits are advantageously disposed horizontally and have their ends, which are preferably circular, rolled into common end members or headers.

In the application of the apparatus for the catalytic reduction of carbon monoxide to produce hydrocarbons which contain more than two carbon atoms in each molecule all the particles of the catalyst are advantageously disposed less than 10 mms. distant from the wall of a conduit.

Constructions such as those described

are illustrated in Figures 1 and 2 of the accompanying diagrammatic drawings, which figures are cross-sectional views through groups of tubes of differing cross-sections. Figures 3 and 4 illustrate one example of the building of a complete apparatus from shallow non-circular conduits, Figure 3 being a view approximating to a perspective view of two vertical series of tubes, one being shown partly in section.

Figures 5 and 6 illustrate diagrammatically a contact apparatus in which non-circular conduits are connected at each end into a common tube plate, as by rolling, Figure 5 being a longitudinal section and Figure 6 a cross-section.

The interstitial spaces *a*, *b*, between the tubes or conduits *d*, *e* respectively in Figures 1 and 2 of the accompanying drawings leave a free path for the contact substance to travel downwardly during charging and clearing or withdrawing the catalyst by gravity, as will be clear from the drawings.

By selecting suitable cross-sectional shapes, such as the oval or diamond shapes of Figures 1 and 2 respectively, and proper dimensions for the cross-section and by suitably spacing-off the tubes or conduits from each other it is possible to adapt the apparatus to every requirement. It has been found that the conditions for the synthesis of benzine can be easily fulfilled with respect to the requirement for a sufficiently thin uniform layer between the adjacent tubes and about each tube of 10 to 15 millimetres.

It is known to roll non-circular tubes or conduits, such as *g* in Figure 3, into tubular base or end columns *h* after short lengths of the tube at the ends as at *i* have first been restored to the circular shape. It has been found that it is possible by this means to construct from non-circular conduits and tubular base or end columns contact apparatus which meets all the requirements.

The especial purpose of the apparatus hereinbefore described—namely, the formation between adjacent tubes and about each tube of uniformly thick intervening masses of the catalyst as thin, for example, as 10 to 15 millimetres—can be secured by staggering the tubes or conduits in the individual rows, that is to say, by arranging each conduit in a lower row below the space between two adjacent conduits in the next upper row. This offers no difficulty if all the conduits forming a set are rolled on each side into a common tubular base or end column.

This construction is particularly advantageous. If the non-circular conduits be horizontally disposed and the tubular base or end columns set vertically, the catalyst can be easily charged into the system of conduits at the top and withdrawn at the bottom. The reaction chamber is closed perfectly liquid-tight by the seamless tubes, the tubular base or end columns and the strong rolled parts without the uncertainty produced in the use of welded seams. Finally, by rigidly connecting a large number of tubes secured adjacent to each other to the tubular base or end columns the tubes are effectively held in fixed relation to almost within a millimetre, under all working conditions. It is known that the degree of security can be improved by the use of intermediate columns. Thus it is easily possible to maintain the thickness of the layers even in the case of large plants.

As illustrated in Figure 4, the several vertical series of tubes *g* with their end columns *h* are connected to a common oil supply or discharge pipe *j* and are enclosed within a casing *k* in which the contact substance is supported between the tubes on a grid *l*. The gas to be treated is passed through the casing from the top to the bottom.

In Figures 5 and 6 which illustrate an apparatus somewhat similar to that shown in Figure 4, the non-circular conduits A extend throughout the height of the apparatus, although for simplicity only three of them are shown. They are rolled on each end into a common tube plate B, short lengths of the ends of the tubes being reformed to a cylindrical shape for the purpose. The reaction gas enters the containing casing at C and passes out at D. At E the circulating oil flows into the oil distributing chamber F, thence through the conduits A into the discharge chamber G from which it passes out at H. J is an intermediate tube plate for more effectively securing the tubes or conduits. The contact composition K fills the space between the conduits A. The oils that can be used for the purpose of temperature regulation are natural oils or oils artificially produced and other organic liquids—and mixtures thereof—which have a high boiling point. Instead of such oils, superheated water or superheated aqueous solutions may be employed. In this case conduits employed must have their walls suitably strengthened in view of the pressures which arise. Furthermore organic liquids under high pressure may be used. The high pressure can be produced either by the vapour from the liquid or by means of compressed gases such as nitrogen, in each case the boiling point of the circulating liquids being raised. Salt melts and

metals of low melting point can be used, more especially mercury.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. Apparatus for carrying out catalytic gas reactions by the use of a circulating liquid medium for maintaining the temperature, characterised in that the liquid medium is caused to pass through narrow conduits the cross-section and disposition of which are such that the smallest distances between the conduits, taken from any particular point on the periphery of any particular conduit to the adjacent conduit nearest the said point, are approximately equal, so that the thickness of catalyst between any one conduit and the adjacent conduits is uniform or substantially so, and the catalyst can pass downwardly by gravity through the set of conduits when the apparatus is being filled or emptied.

2. Apparatus for carrying out catalytic gas reactions according to claim 1, in which oil is used as the circulating liquid medium.

3. Apparatus according to claim 1 or claim 2, characterised in that the conduits are disposed horizontally, and have their ends which are preferably circular, rolled into common end members.

4. Apparatus according to any one of the preceding claims, for the catalytic reduction of carbon monoxide to produce hydrocarbons which contain more than two carbon atoms in each molecule, characterised in that all the particles of the catalyst are less than 10 mms. distant from the wall of a conduit.

5. Apparatus for carrying out catalytic gas reactions substantially as hereinbefore described and illustrated.

Dated the 29th day of January, 1935.

EDWARD EVANS & Co.,
27, Chancery Lane, London, W.C.2,
Agents for the Applicants.

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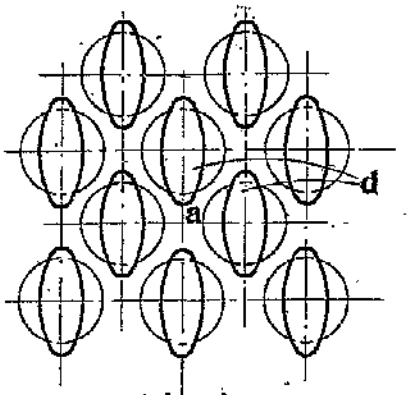

Fig. 1

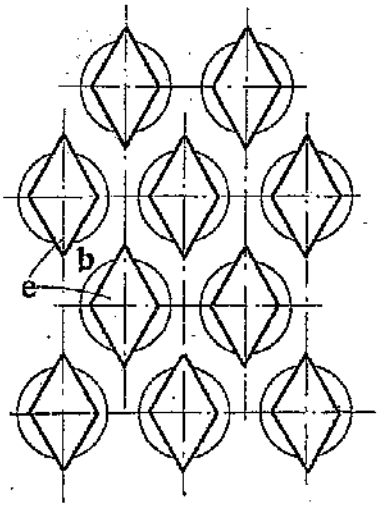

Fig. 2

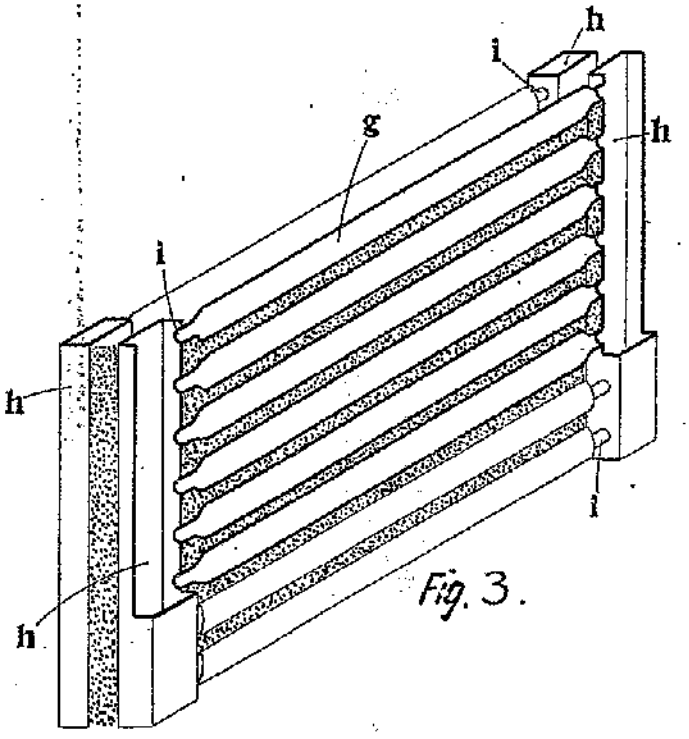

Fig. 3.

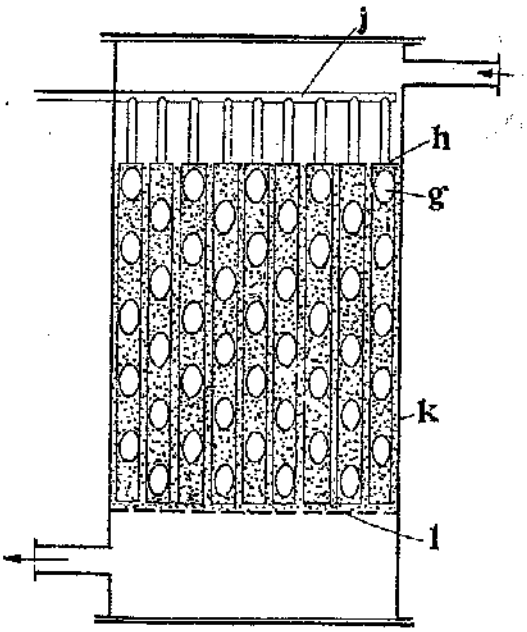

Fig. 4.

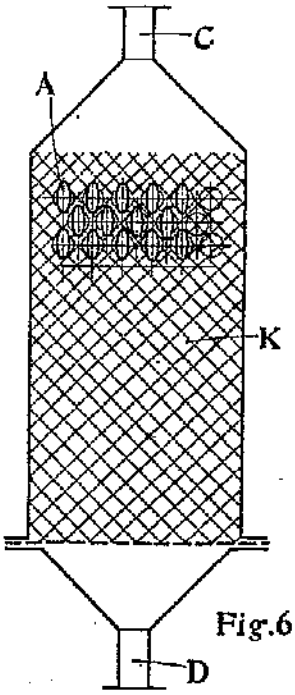

Fig.6.

h

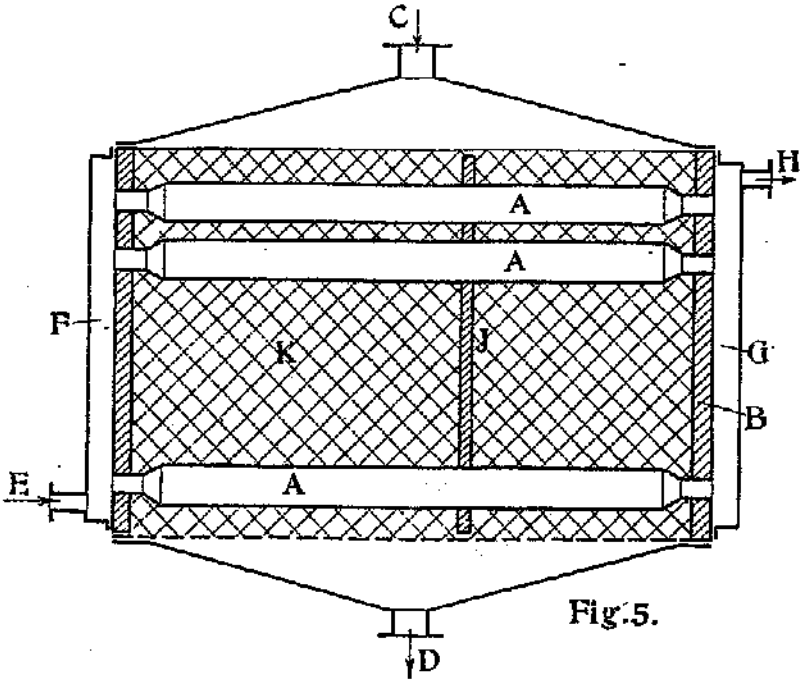

Fig.5.

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2 SHEETS
SHEET 2

SHEET 1

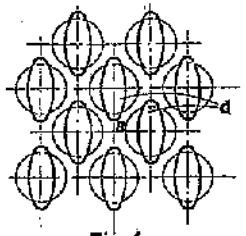

Fig. 1

Fig. 2

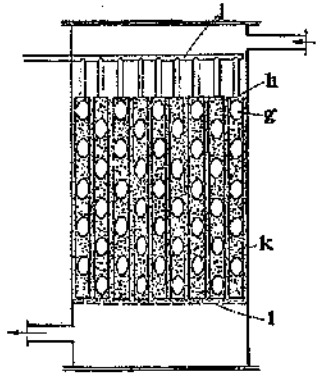

Fig. 4.

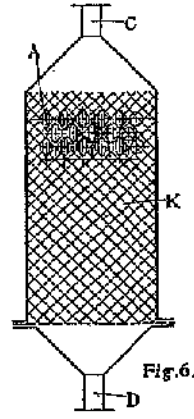

Fig.6.

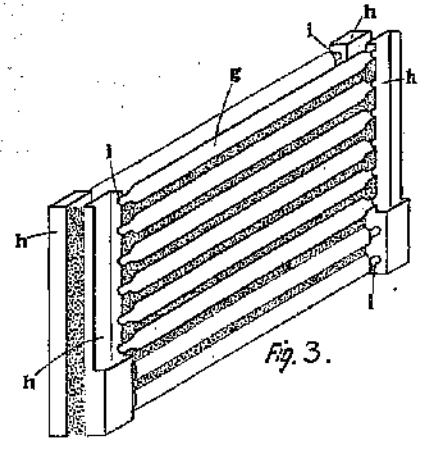

Fig. 3.

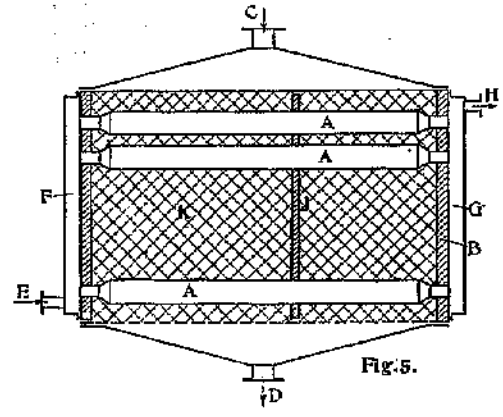

Fig.5.

[This Drawing is a reproduction of the Original on a reduced scale.]

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