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DEVICE FOR PRODUCING EXOTHERMIC REACTIONS BETWEEN

GASES BY CONTACT WITH A SOLID CATALYZER

Filed Nov. 7, 1929

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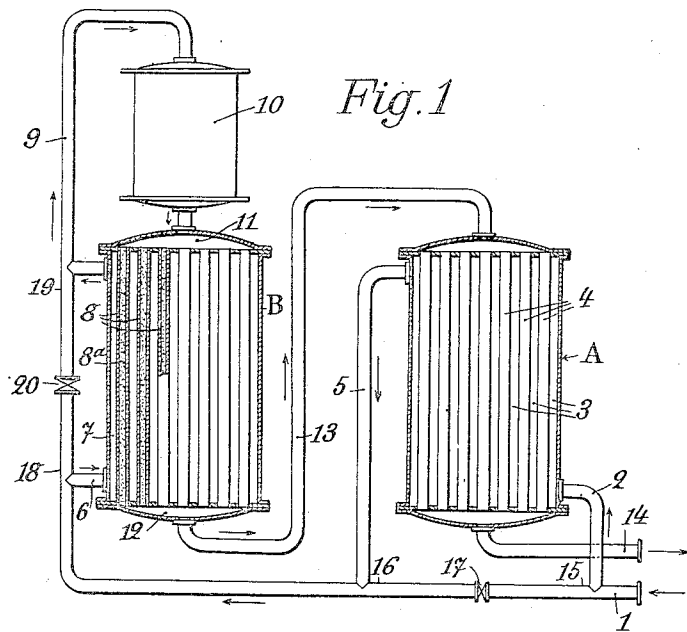


Fig. 1

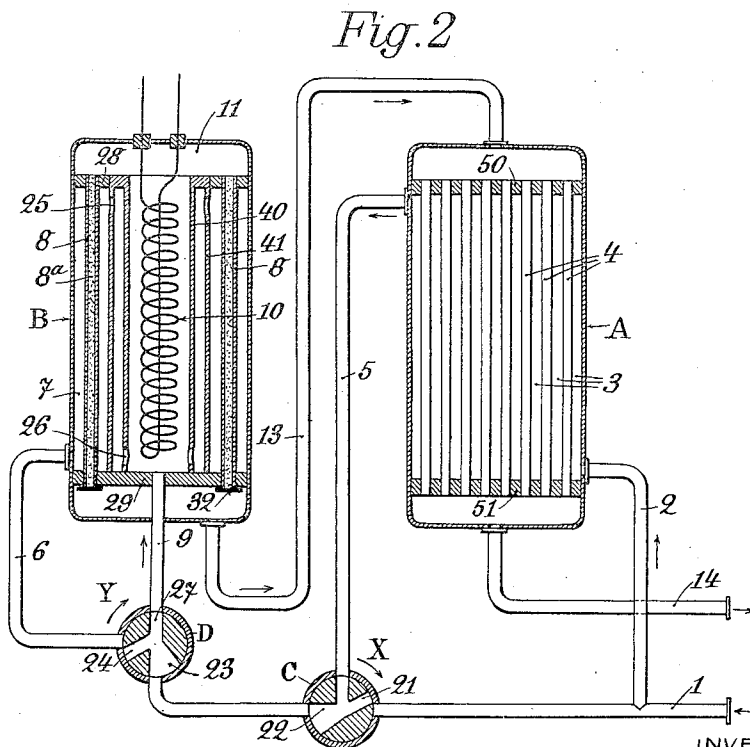


Fig. 2

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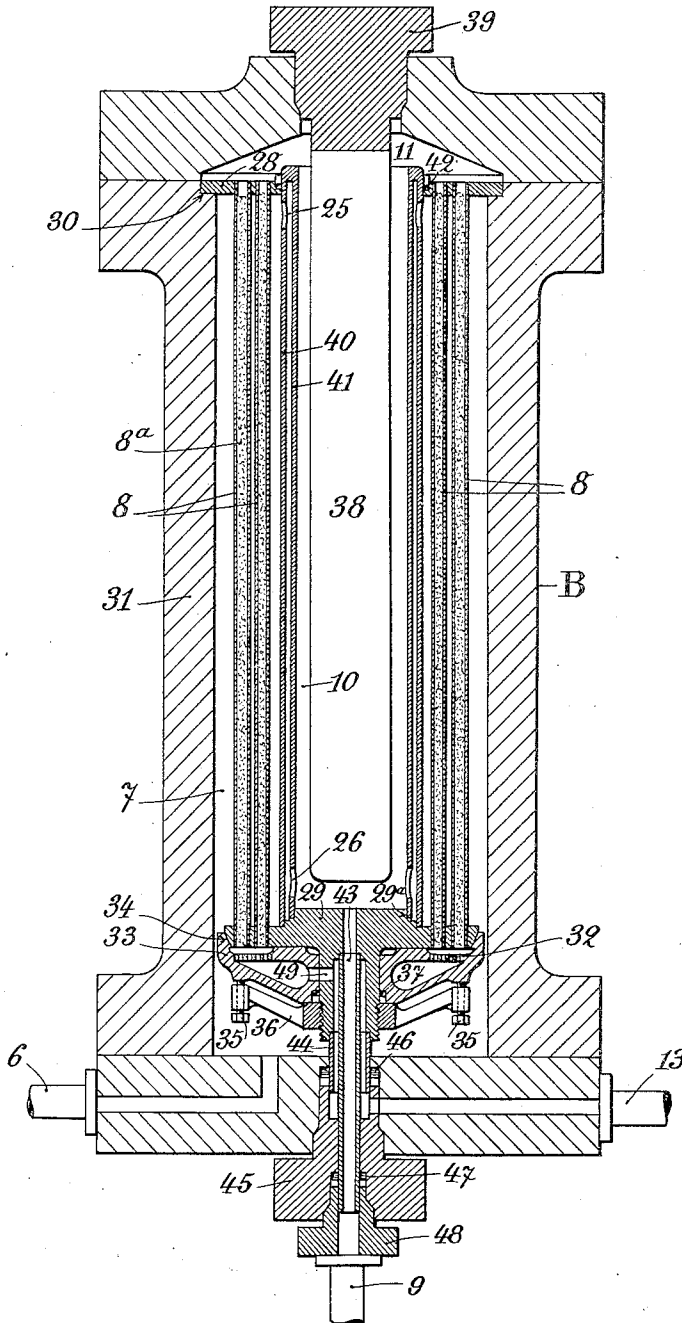
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Fig. 3



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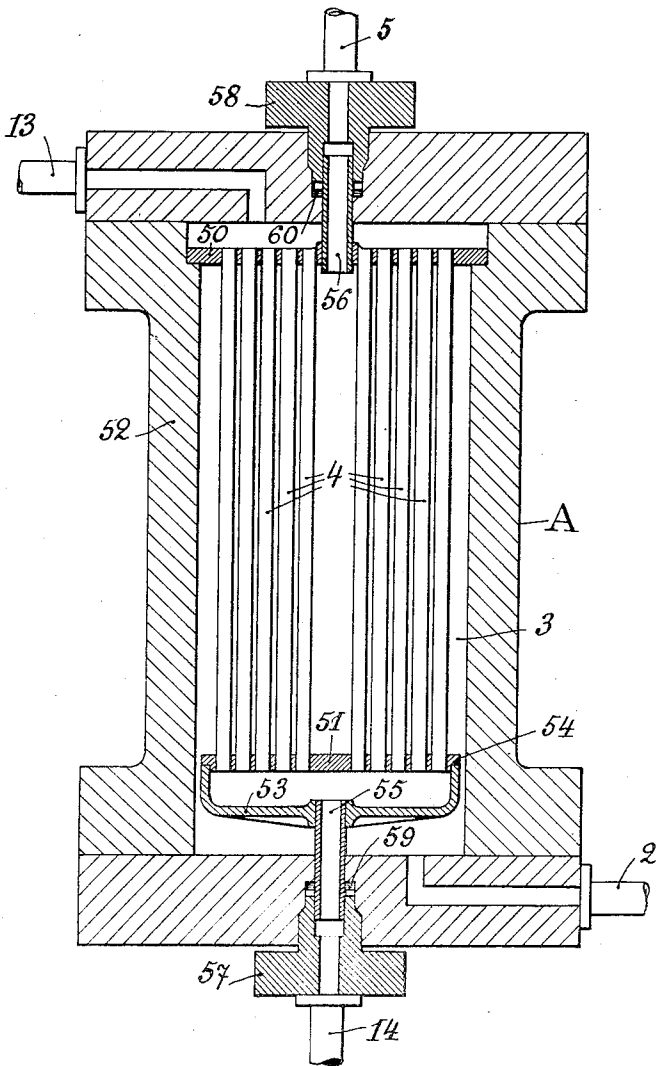
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3 Sheets-Sheet 3

Fig. 4



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DEVICE FOR PRODUCING EXOTHERMIC REACTIONS BETWEEN GASES BY CONTACT WITH A SOLID CATALYZER

Application filed November 7, 1929, Serial No. 405,495, and in France October 21, 1929.

The present invention has for an object a device for producing exothermic reactions between gases by contact with a solid catalyzer, and particularly for the synthetic manufacture either of ammonia gas or mixtures of hydrogen and nitrogen, or of methanol or of any other organic compounds of mixtures of hydrogen and carbon monoxide.

A device in accordance with this invention, for producing exothermic reactions between gases by contact with a solid catalyzer, comprises three elements: a heat exchanger, a catalyzing tube and an apparatus for heating the fresh gaseous mixture, which can be, according to circumstances, placed in one, two or three distinct casings.

Two forms of construction of a device in accordance with this invention are illustrated, by way of example, in the accompanying drawings, in which:

Fig. 1 is a diagrammatic view of a form of construction in which the three above mentioned elements are separated from each other.

Fig. 2 diagrammatically illustrates a second form of construction in which the catalyzing tube and the heating apparatus are connected in one and the same casing.

Figs. 3 and 4 illustrate in vertical section and in a detailed manner, the essential elements of this second form of construction.

As shown in Fig. 1, the fresh gaseous mixture is admitted, through 1, 2, into the heat exchanger A, it circulates in the space 3 included between the tubes 4, issues through 5, enters through 6 into the space 7 included between the tubes 8 of the catalyzing apparatus B, issues through 9 and is collected, after having passed through the heating device 10, in the chamber 11, from which it passes into the tubes 8 containing the contact mass 8a; the products of the reaction are fed, through 12 and 13, to the heat exchanger, from which they issue through 14.

One of the features of the device forming the subject-matter of the present invention consists in short-circuiting, on the one hand, the heat exchanger A, on the other hand, the intertubular space of the catalyzing apparatus B, for instance by means:

Of a by-pass 15, 16, provided with a cock 17, which puts the piping 1 feeding the fresh mixture to the heat exchanger, in direct communication with the outlet piping 5 of this apparatus;

And of a by-pass 18—19, provided with a cock 20, which connects the piping 6, feeding the gas to the intertubular space of the catalyzing apparatus, with the piping 9 feeding the gas to the heating apparatus.

Another mode of application of this principle consists, as shown by way of example in Fig. 2, in arranging on the pipings two identical apparatus C and D which will be designated hereinafter under the name of "distributors"; these apparatus are shown in the form of cocks, the plug of which is provided with three channels and a notch, but they can be constituted in any other manner capable of ensuring the following operation:

The distributor C serves to divide the stream of gaseous mixture used as raw material into two branches, the relation in the outflows of which can be controlled at will by the said distributor C, only one of these branches passing through the heat exchanger A; with the arrangement shown in Fig. 2, for instance, a rotation of the plug in the cock body, in the direction of the arrow X, has for effect to diminish the outflow through the heat exchanger A, by correspondingly increasing the outflow of the stream which, through 1, 21 and 22, is directly conveyed to the distributor D.

Similarly, the distributor D serves to divide the gaseous stream issuing from the distributor C into two branches, the relation of the outflows of which can be controlled at will by this distributor D; one of these branches is then directed towards the intertubular space 7 of the catalyzing apparatus B and subsequently towards the heating device 10, the other branch being sent directly towards this heating device. In the case of Fig. 2, for instance, the first branch of the stream passes through 23, 24, 6, 7, 25 and 26, and the second branch through 23, 27 and 9; a rotation of the cock plug in the direction of the arrow Y increases the outflow of the

first branch at the expense of the outflow of the second branch.

The use of the two distributors which has just been described allows of suitably controlling the temperature of the contact mass and, consequently, on the one hand, of overcoming the development of certain parasitic reactions of the main reaction as well as the prejudicious effects resulting, for the activity of a large number of catalyzers, from the fact that they are maintained at a too high temperature; on the other hand, of rendering the hourly production per litre of catalytic volume independent from the variations of activity of the contact mass.

In Fig. 2, both distributors are illustrated in the position which corresponds, for the tubes of the bundle, to the catalyzing apparatus B when it is at its highest working temperature; a rotation of the plugs of these distributors in the direction of the arrows X and Y allows of modifying this working temperature, owing to the putting in circulation in the intertubular space of a gaseous stream, the outflow of which is adjustable, and the initial temperature of which can moreover be varied between wide limits.

The catalyzing tube B, diagrammatically illustrated in Fig. 3, presents five essential features, viz:

1. The contact mass is introduced in a series of vertical tubes 8, secured at both ends on two horizontal metal plates 28, 29; the upper plate 28 is secured, by screws or bolts, on a ledge 30 of the steel casing 31 which supports the working pressure, whilst the lower plate 29 is free to move; the whole of the bundle is thus suspended within the casing.

2. The contact mass is retained within the tubes 8 by a perforated plate 32 rigid with a metal member 33 in the shape of a box assembled with the bottom plate by a fluid-tight packing 34. The clamping of 33 on 29 is ensured by clamping screws 35 carried by a member 36, which bears on a boss 37 of the bottom plate 29 through a screw-threaded portion having interrupted threads, or in any other manner allowing the device to be rapidly taken to pieces.

3. A chamber 10 is provided according to the axis of the catalyzing device and encloses the device for heating the fresh gaseous mixture; this heating device, diagrammatically indicated at 38 and suspended from an auxiliary plug 39, is completely independent from the remainder of the apparatus.

4. The gas contained in the intertubular space is brought in contact with the device 38 by a member constituted by two coaxial tubes 40 and 41, rigidly assembled together at their upper part on one and the same annular cover; at the lower part, one of these tubes is permanently secured on the lower plate 29, whilst the other can slide on a

shoulder 29a of this plate 29. On the other hand, these two tubes can freely slide, at the upper part, through a circular orifice provided in the plate 28, the fluid-tightness of the joint being ensured by a plastic packing 42.

5. Two co-axial tubes of small diameter, 43 and 44, secured on the lower plate 29, are inserted, with slight friction in corresponding bores of the lower plug 45 of the steel casing 41, the fluid-tightness of the assemblages being ensured by plastic packings 46 and 47 clamped, one by the plug 45, the other by a stuffing-box 48.

The tube 43 serves for the direct introduction into the heating chamber 10 of the fresh gaseous mixture admitted through the piping 9.

The tube 44 serves for evacuating outside the casing the products of the reaction which, when issuing from the bundle of tubes, are collected in the metal member 33 and flow through an orifice 49 into the annular space between 43 and 44.

The operation of the catalyzing device which has just been described is as follows: Both streams of fresh gaseous mixture separated by the distributor D, enter the catalyzing apparatus, one through 6, the other through 9. The gas introduced through 6 passes upwardly through the intertubular space 7, enters, through the orifices 25, into the annular space comprised between the tubes 40 and 41, circulates downwardly between these tubes, and, through orifices 26, enters the heating chamber 10, where it mixes with the gas admitted through 9. The mixture rises along the heating device 38 to the upper chamber 11, where it passes into the tubes 8 containing the contact mass; it passes downwardly through these tubes and is collected in the metal member 33, being finally evacuated through the orifice 49, the annular space between 43 and 44, and the piping 13.

The characteristic properties of the apparatus thus devised are the following:

a. The fluid-tightness of the device controlling the circulation of the gas is ensured by means of a small number of plastic packings, which latter have, moreover, a small diameter.

b. The various elements of this device can, however, freely expand; this allows, when choosing the metals used for constructing the said elements, of not taking into account the values of the coefficients of expansion and to exclusively take into consideration the mechanical and chemical conditions to be satisfied.

c. The contact mass inserted in the tubes 8 of the bundle can be rapidly evacuated; for that purpose, it suffices to loosen the screws 35, to release the member 36 from the boss 37 by rotation according to a fraction of a revolution, and to lower the members 36 and 33.

d. The heating device is completely inde-

pendent from the device used for producing catalysis and ensuring the circulation of the gas; this allows, on the one hand, to use the most varied heating sources; electricity, gaseous or liquid fluid, internal combustion, etc., and, on the other hand, to replace the heating apparatus, by simply unscrewing the plug 39, without having to touch any other member.

e. The device for producing the catalysis and ensuring the circulation of the gases is in contact with the steel casing only at very few points; this particularly facilitates the application, on the inner face of the said casing, of coatings which may be advantageously used for preventing the gaseous mixture from coming in contact with this casing.

f. The assemblage and taking to pieces of the catalyzing tube is effected with great facility.

20 The heat exchanger A, illustrated in detail in Fig. 4, presents three essential features, viz:

1. The vertical tubes 4 of the bundle constituting one of the essential elements of this heat exchanger, are secured at their ends on two horizontal metal plates 50 and 51; the upper plate 50 is secured, by screws, on an inner ledge of the steel casing which supports the working pressure, whilst the lower plate 51 is free to move. The bundle of tubes is thus suspended with the casing.

2. The lower plate 51 is completed by a member 53, in the shape of a box, with which it is assembled by a fluid-tight packing 54.

35 3. Two pipes of small diameter, 55 and 56, are in communication with the exterior: the pipe 55 being secured on the member 53 while the interior of this member 53 is in communication with the tubes 4 of the bundle. The other pipe 56 being secured on the upper plate 50 and in communication with the intertubular space 3.

45 These two tubes pass through the lower plug 57 and the upper plug 58 of the casing, through orifices in which they can freely slide, the fluid-tightness of the joints being ensured by plastic packings 59 and 60.

50 The operation of the apparatus which has just been described is as follows; the fraction of the fresh gaseous mixture directed by the distributor C towards the heat exchanger, enters therein through the piping 2, circulates in the intertubular space 3, and issues through the tube 56 and the piping 5. The products of the reaction coming from the catalyzing device enter the heat exchanger through the piping 13, circulate downwardly in the tubes 4, they are collected in the hollow member 53 and issue through 55 and 14.

60 The characteristic feature of this heat exchanging apparatus resides in the fact that the fluid-tightness of the device ensuring the circulation of the gases is obtained without hindering the free expansion of its elements.

65 Claims:

1. In a device for producing reactions between gases by contact with a catalyzer, the combination of a casing, a perforated plate secured in the upper part of this casing, a plurality of tubes secured in the holes of the said plate, a second perforated plate loosely placed in the lower portion of the said casing, the lower ends of the tubes being secured in the holes of this second plate, a catalyzing material in the said tubes, a hollow member detachably secured under the said second plate, this member having perforated portions extending under the said tubes and adapted to support the said catalyzing material.

2. In a device for producing reactions between gases by contact with a catalyzer, the combination of a casing, a perforated plate secured in the upper part of this casing, a plurality of tubes secured in the holes of the said plate, a second perforated plate loosely placed in the lower portion of the said casing, the lower ends of the tubes being secured in the holes of this second plate, a catalyzing material in the said tubes, a hollow member detachably secured under the said second plate, this member having perforated portions extending under the said tubes and adapted to support the said catalyzing material, a plug in the bottom of the said casing, this plug presenting a vertical conduit wide at the top and narrow at the bottom, the second perforated plate having two concentric tubular extensions downwardly directed and adapted to slide with slight friction respectively in the wide and narrow portions of the said conduit, the said hollow member having an annular shape and surrounding the outer tubular extension, this last tubular extension and the said hollow member having registering orifices, and inlet and outlet conduits respectively communicating with the said tubular extensions.

3. In a device for producing reactions between gases by contact with a catalyzer, the combination of a casing, a perforated plate secured in the upper part of this casing, a plurality of tubes secured in the holes of the said plate, a second perforated plate loosely placed in the lower portion of the said casing, the lower ends of the tubes being secured in the holes of this second plate, a catalyzing material in the said tubes, a hollow member detachably secured under the said second plate, this member having perforated portions extending under the said tubes and adapted to support the said catalyzing material, two concentric sleeves secured at their lower part on the second perforated plate, the top of the outer sleeve being connected in a fluid-tight manner to the top of the inner sleeve and being adjusted with slight friction in an opening of the said upper perforated plate, the said sleeves having side orifices, one near their upper end the other near their lower

end, and a heating device within the inner sleeve.

4. In a device for producing reactions between gases, a heat exchanger comprising the combination of a casing, an upper perforated plate secured in the upper portion of the said casing, a plurality of tubes secured in the holes of the said upper plate, a lower perforated plate loosely placed in the lower part of the casing, the lower ends of the said tubes being secured in the holes of the said lower plate, a hollow member secured under the said lower plate, perforated plugs secured in the top and in the bottom of the said casing, tubes respectively secured to the center of the upper tubular plate and to the center of the said hollow member, these tubes being fitted with slight friction in the openings of the said plugs, and conduits respectively leading to the top and to the bottom of the said casing for the admission and evacuation of the gases.

5. Device for exothermic reactions between gases by contact with a solid catalyzer, comprising a heat exchanger having two spaces separated in a fluid-tight manner, a catalyzing apparatus having two chambers, a plurality of tubes connecting these chambers, a solid catalyzing material in these tubes and a casing enclosing the said tubes, a heating device, pipings for supplying the gases to be treated and leading to one of the spaces of the heat exchanger, pipings between the said space of the exchanger and the casing of the catalyzing apparatus, between this casing and the heating device, between this heating device and one of the chambers of the catalyzing apparatus, between the other chamber of the catalyzing apparatus and the second space of the heat exchanger, and a piping for the exhaust of the gases of the reaction, starting from the second space of the exchanger.

6. Device for exothermic reactions between gases by contact with a solid catalyzer, comprising a heat exchanger having two spaces separated in a fluid-tight manner, a catalyzing apparatus having two chambers, a plurality of tubes connecting these chambers, a solid catalyzing material in these tubes and a casing enclosing the said tubes, a heating device, and pipings for supplying the gases to be treated and leading to one of the spaces of the heat exchanger, pipings between this space of the exchanger and the casing of the catalyzing apparatus, between the said casing and the heating device, between this heating device and one of the chambers of the catalyzing apparatus, between the other chamber of the catalyzing apparatus and the second space of the heat exchanger, an exhaust piping for the gases of the reaction starting from the second space of the exchanger, a by-pass between the inlet and the outlet of the casing of the catalyzing apparatus, and control means on these by-passes

for controlling the passage of the gases through these latter.

7. In a device for reactions between gases by contact with a catalyzer, the combination of a casing, a perforated plate secured in the top of the said casing, a plurality of tubes secured in the holes of the said plates, a second perforated plate loosely placed in the lower portion of the said casing, the lower ends of the tubes being secured in the holes of the second plate, a catalyzing material in the said tubes, and means for supporting the said catalyzing material in the said tubes.

8. In a device for producing reactions between gases by contact with a catalyst, the combination of a casing, a perforated plate secured in the upper part of this casing, a plurality of tubes secured in the holes of the said plate, a second perforated plate loosely placed in the lower portion of the said casing, the lower ends of the tubes being secured in the holes of this second plate, a catalyzing material in the said tubes, a cylinder having perforated walls, secured axially to the second plate, the first plate having an orifice in which the said cylinder freely slides, a fluid-tight packing between the said cylinder and the first plate, and a heating device hung from the casing of the catalyzing apparatus and arranged in the axis of the cylinder.

In testimony whereof I have hereunto affixed my signature.

ETIENNE AUDIBERT.

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