PATENT SPECIFICATION

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3430

COMPLETE SPECIFICATION.

Improvements in or relating to Effecting Reactions between Gases with the Aid of a Catalyst.

We, SOCIETE NATIONALE DE RECHERCHES SUR LE TRAITEMENT DES COMBUSTIBLES, a French Societe Anonyme, of 10, Rue St. Hilaire, Senlis (Oise Department), France, 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:—

The present invention relates to apparatus for effecting exothermic reactions between gases when in contact with a solid catalyst, and particularly for the synthetic manufacture, either of ammonia gas by means of mixtures of hydrogen and nitrogen, or of methanol or other organic compounds such as alcohols, homologues of methanol, acids, aldehydes, ketones and hydrocarbons by means of mixtures of hydrogen and oxides of carbon.

Apparatus of this type is already known comprising a heat exchanger, a heating apparatus and a catalytic apparatus in which the gaseous mixture to be treated flows successively through the heat exchanger and through the heating apparatus, a portion of this mixture coming into direct contact with the catalyst in the catalytic apparatus while the rest of the gaseous mixture comes into contact with tubes containing the catalyst.

It is also known, in apparatus of this kind, to conduct the gaseous mixture in heat-exchanging relation to the catalyst before passing through the catalyst, and to provide a by-pass for by-passing a proportion, or all, of the gaseous mixture past the heat exchanger.

Finally, apparatus is also known wherein the gaseous mixture to be treated enters from above into a distribution chamber, the bottom of which is formed by a perforated plate, from which are suspended tubes which descend into other tubes resting on a lower plate and extending freely upward. The lower plate is perforated outside the tubes. In apparatus of this kind, the gaseous mixture to be treated circulates in opposite directions in the descending and ascending tubes and then passes into the catalytic mass disposed between the ascending tubes, whereupon the gases leave by the perforations of the lower plate.

According to the present invention an apparatus comprising a heat exchanger, a heating apparatus and a catalytic apparatus in which the gaseous mixture to be treated flows successively through the heat exchanger, through the catalytic apparatus in contact with the exteriors of containers holding the catalyst, through the heating apparatus, through the catalytic apparatus in contact with the catalyst, and finally through the heat exchanger, is provided with a by-pass conduit for conducting, if desired, a proportion, or all, of the gaseous mixture from the heat exchanger to the heating apparatus without flowing through the catalytic apparatus in contact with the exteriors of the containers holding the catalyst.

Two forms of construction of an apparatus according to this invention are illustrated in the accompanying drawing by way of example:—

Figure 1 is a diagram of a form of construction in which the heat exchanger, catalytic apparatus and heating apparatus are separated from each other.

Figure 2 diagrammatically illustrates a second form in which the catalysing apparatus and the heating apparatus are assembled in one and the same casing.

Figures 3 and 4 illustrate in vertical section and in detail the essential parts of this second form of construction.

According to Figure 1, the fresh gaseous mixture is admitted through 1, 2 into the heat exchanger A, circulates in the space 3 comprised between the tubes 4, issues through 5, is admitted through 6 into the space 7 comprised between the tubes 8 of the bundle of catalyst tubes B, issues therefrom through 9 and is collected, after having passed through the heating apparatus 10, in the chamber 11, from which it passes through the tubes 8 containing the contact mass 8a. The reaction products are conveyed, through 12 and 13, to the heat exchanger, from which they issue through 14.

[illegible]

A feature of the apparatus forming the subject-matter of the present invention consists in short-circuiting, on the one hand, the heat exchanger A, and on the other hand, the intertubular space of the catalyser B, for instance by means of a by-pass, 15, 16, having a cock 17, which puts the piping 1 supplying 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, having a cock 20, which connects the piping 6 conveying the gas in the intertubular space of the catalysing bundle to the piping 9 feeding the gas to the heating apparatus.

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

The three-way cock C serves to divide the stream of gaseous mixture used as raw material into two branches, the outflow ratio of which can be controlled by this three-way cock, one of these branches only passing through the heat exchanger A; in the arrangement of Figure 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 increasing to the same amount the outflow of the stream which, through 1, 21 and 22 is directly conveyed to the three-way cock D.

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

The use of the two three-way cocks which have just been described allows of suitably regulating the temperature of the contact mass and, as a consequence, on the one hand, of checking the development of certain parasitic reactions of the main reaction as well as the prejudicious effects which result, for the activity of a great number of catalysts, from their being maintained at a too high temperature; on the other hand, of rendering the hourly production per litre of catalytic volume independent of the variations of activity of the contact mass.

In Figure 2, both three-way cocks, are illustrated in the position giving the highest operating temperature of the catalyst tubes of the catalyser B. Rotation of the plugs of the three-way cocks in the direction of the arrows X and Y allows the operating temperature to be modified owing to the fact that a gaseous stream is caused to circulate in the intertubular space, the volume of this gaseous stream being controllable and its initial temperature being moreover variable between wide limits.

The catalysing tube B, diagrammatically illustrated in Figure 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 operating pressure, whilst the lower plate 29 is left free to move; the entire bundle being thus hung within the casing;

2) The contact mass is retained within the tubes 8 by a perforated plate 32 which forms a part of a metal member 33, in the shape of a hollow box assembled with the bottom plate by a fluidtight 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 screw-threads, or in any other manner allowing rapid taking to pieces.

3) A chamber 10 is provided in the centre of the catalyser and encloses the device for heating the fresh gaseous mixture; this heating device, diagrammatically indicated at 38 and hung from an auxiliary plug 39, is completely independent from the remainder of the apparatus.

4) Gas from the intertubular space is brought into contact with the device 38 by a member composed of two concentric tubes 40 and 41, sealed together at their upper ends. At their lower ends, one of these tubes is permanently secured to the bottom plate 29, whilst the other is left free to slide on a shoulder 29*a* of this plate. The unit constituted by these two tubes moreover freely slides, at the upper end through a circular orifice provided

in the plate 28, the fluid-tightness of the joint being ensured by a plastic-packing 42.

b) Two concentric tubes 43 and 44, of small diameter and secured on the bottom plate, are fitted with slight friction in corresponding bores of the lower plug 45 of the steel casing 31, 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 in the heating chamber 10 of the fresh gaseous mixture supplied by the piping 9.

The tube 44 serves for removing the reaction products which, upon issuing from the tubes of the bundle, are collected in the box 33 and flow through an orifice 49 into the annular space comprised between 43 and 44.

The operation of the catalyser which has just been described is as follows: Both branches of the fresh gaseous mixture, which is separated by the three-way cock B, enter the catalyser, one through 6, the other through 9. The gas introduced through 6 passes upwardly through the intertubular space 7, penetrates, through the orifices 25, into the annular space comprised between the tubes 40 and 41, circulates downwardly between these tubes, and, through the orifices 26, enters the heating chamber 10, where it mixes with gas admitted through 9. The mixture rises along the heating device 38 up to the upper chamber 11, where it passes into the tubes 8 containing the contact mass after passing downwardly through these tubes the reaction products are collected in the box 33 to be finally evacuated through the orifice 49 and the annular space comprised 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 minimum number of plastic packings, which, moreover, have small diameters only.

b) The various elements of this device can nevertheless expand quite freely; this allows, in choosing the metals to be used for constructing the said elements, the relative values of the expansion coefficients to be ignored, the mechanical and chemical conditions to be satisfied being exclusively considered.

c) The contact mass inserted in the tubes 8 of the bundle can be rapidly removed; in fact, it suffices for this operation to loosen the screws 35, to release the member 36 from the boss 37 by rotation to the extent of a fraction of a revolution and to take down the members 36 and 33.

d) The heating device is completely independent of the catalysing and gas circulating device; this allows, on the one hand, the most varied sources of heat, for example, electricity, gaseous or liquid fluid, internal combustion, etc., to be used, and on the other hand, the heating apparatus to be replaced by simply unscrewing the plug 39, without having to touch any other part whatever.

e) The catalysing and gas circulating device is in contact with the steel casing only at a very small number of points; this feature facilitates the application on the inner face of this casing of the linings or coatings which it may be advantageous to use for preventing the gaseous mixture from coming into contact with the same.

f) The fitting up and removal of the tube are effected with the greatest care.

The heat exchanger A illustrated in detail in Figure 4, presents three essential features, viz:

1) The vertical tubes 4 of the bundle which constitutes one of the essential elements of the same are secured at their ends to 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 operating pressure, whilst the lower plate 51 is left free to move. The bundle is then hung within 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 means of a fluid-tight joint 54.

3) Two pipes 55 and 56, of small diameter, lead respectively from the exterior to the tubes and to the intertubular space. These two pipes pass respectively through the lower plug 57 and the upper plug 58 of the casing, through orifices in which they are free to slide, the fluid-tightness of the joints being ensured by plastic packings 59 and 60

The apparatus which has just been described operates as follows: The fraction of the fresh gaseous mixture which is directed by the three-way cock C towards the heat exchanger, enters therein through the piping 2, circulates in the intertubular space 3 and issues through the tube 56 and piping 5. The reaction products which come from the catalyser, enter the heat exchanger through the piping 13, circulate downwardly in the tubes 4, are collected in the hollow member 53 and issue through 55 and 14.

An advantage 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 prejudice to the free expansion of its members.

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 effecting exothermic reactions between gases in contact with a solid catalyst, comprising a heat exchanger, a heating apparatus and a catalytic apparatus, the gaseous mixture to be treated flowing successively through the heat exchanger, through the catalytic apparatus in contact with the exteriors of containers holding the catalyst, through the heating apparatus, through the catalytic apparatus in contact with the catalyst, and finally through the heat exchanger, characterised by the provision of a by-pass conduit for conducting if desired a proportion, or all, of the gaseous mixture from the heat exchanger to the heating apparatus without flowing through the catalytic apparatus in contact with the exteriors of the containers holding the catalyst.

2. Apparatus as claimed in claim 1, characterised by the provision of a by-pass for by-passing a proportion, or all, of the gaseous mixture to be treated past the heat exchanger.

3. Apparatus as claimed in claim 1, characterised by the provision of a three-way cock for regulating the relative proportions of gaseous mixture flowing through the catalytic apparatus in contact with the exteriors of the containers holding the catalyst and flowing direct to the heating apparatus.

4. Apparatus as claimed in claim 2, characterised by the provision of a three-way cock for regulating the relative proportion of gaseous mixture by-passed past the heat exchanger.

5. Apparatus as claimed in claim 1, characterised in that in the catalytic apparatus the catalyst is contained in tubes secured at their two ends to horizontal metal plates, the upper plate being secured to a ledge of the casing and the tubular bundle being suspended from the said plate so that it can freely expand in the casing.

6. Apparatus as claimed in claim 5, characterised in that the tubes containing the catalyst open at their lower end into a box-shaped member which is pressed in a fluid-tight manner against the lower plate by a member secured to the said plate so that the box-shaped member can be rapidly removed.

7. Apparatus as claimed in claim 6, characterised in that the heating apparatus is contained within the same casing as the catalytic apparatus and comprises a heating element projecting from a plug in the top of the casing through a hole in the upper tube-plate in the axis of the bundle of catalyst tubes.

8. Apparatus as claimed in claim 7, characterised in that the heating element is surrounded by two concentric tubes mounted in the upper tube-plate and enclosing a space between them, the outer tube being provided with orifices at its upper part, and the other with orifices at its lower part, while both tubes engage the lower tube-plate in a fluid-tight manner and are adapted to slide in one of the tube plates.

9. Apparatus as claimed in claim 8, characterised in that a conduit is provided in the bottom of the casing for the passage of gaseous mixture into the space between the catalyst tubes.

10. Apparatus as claimed in claim 6, characterised in that a conduit is provided in the box-shaped member secured to the lower plate for the passage of the reaction products from the catalyst apparatus.

11. Apparatus as claimed in claims 6 and 8, characterised in that a conduit is provided in the box-shaped member communicating with a conduit in the lower tube-plate leading to the space between the heating element and the inner of the two concentric tubes surrounding it.

12. Apparatus as claimed in claims 10 and 11, characterised in that the conduits in the box-shaped member communicate with the exterior of the casing by means of tubes which slide in a fluid-tight manner in the bottom of the casing.

13. Apparatus for effecting exothermic reactions between gases in contact with a solid catalyst, substantially as described or substantially as shown in Figures 1, 2 and 3 of the accompanying drawings.

Dated this 18th day of October, 1930.

SOCIETE NATIONALE DE RECHERCHES SUR LE TRAITEMENT DES COMBUSTIBLES.
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Fig. 1.

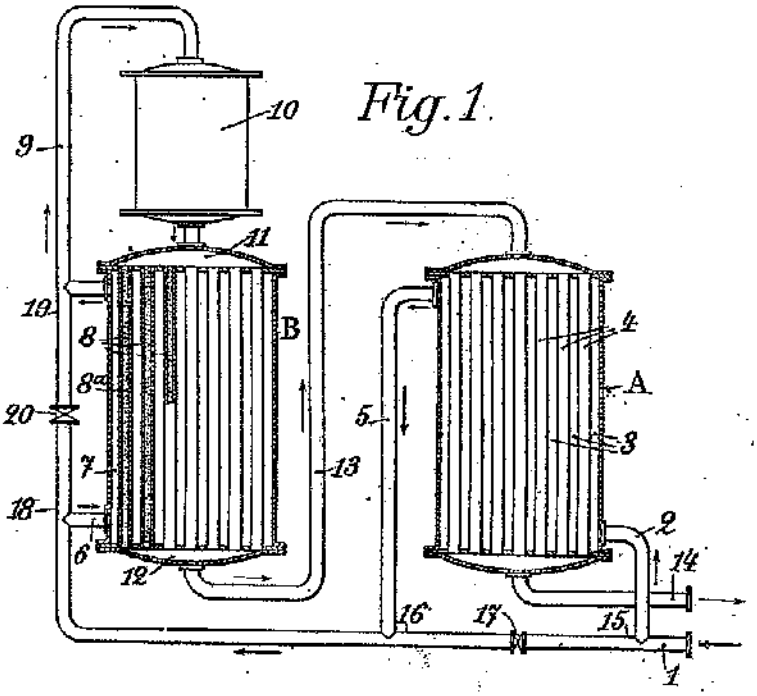

Fig. 2.

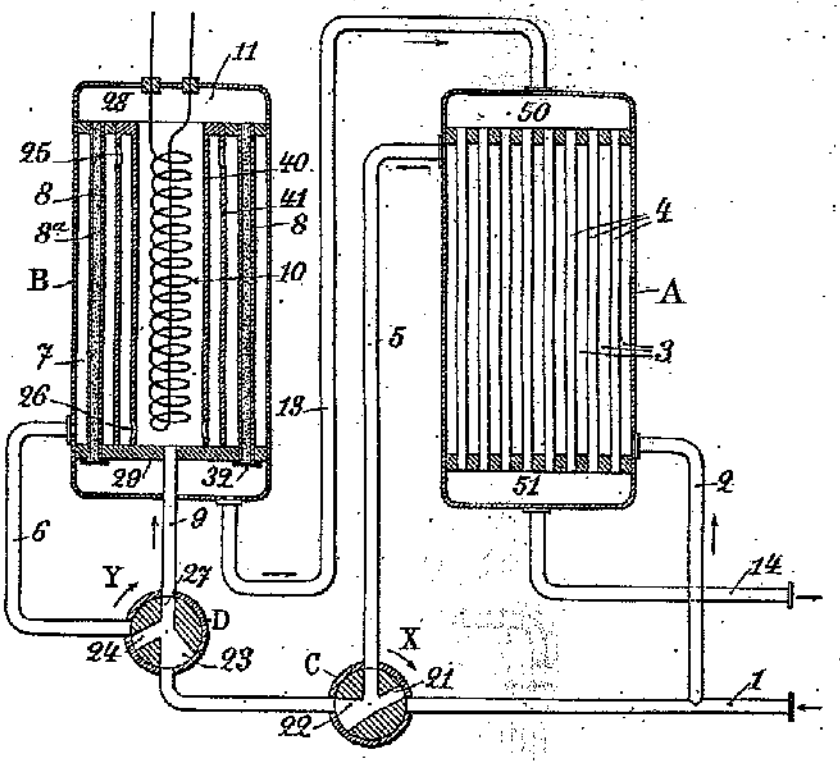

Fig.3

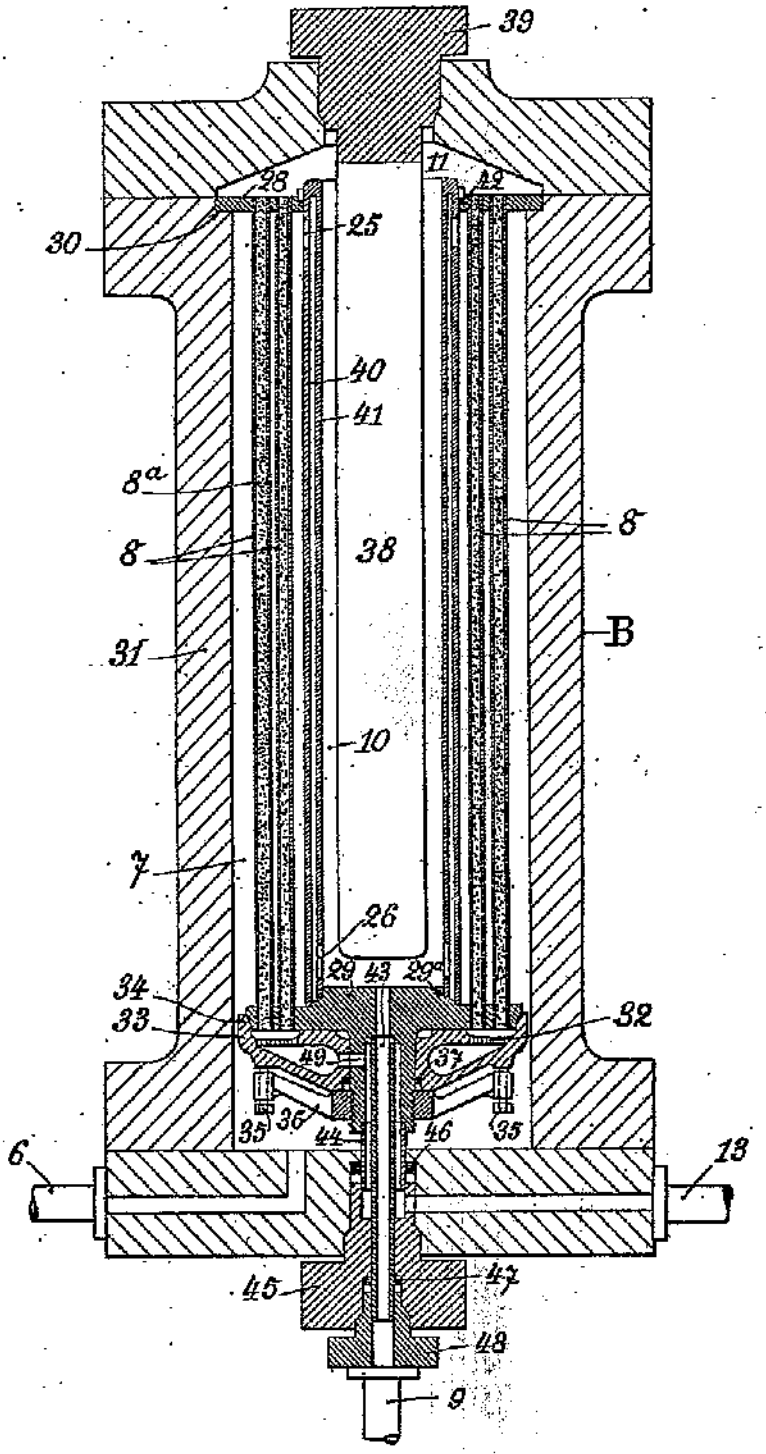

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Fig. 1

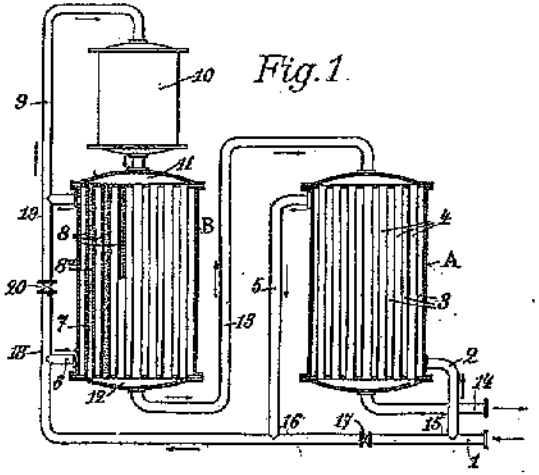


Fig. 2

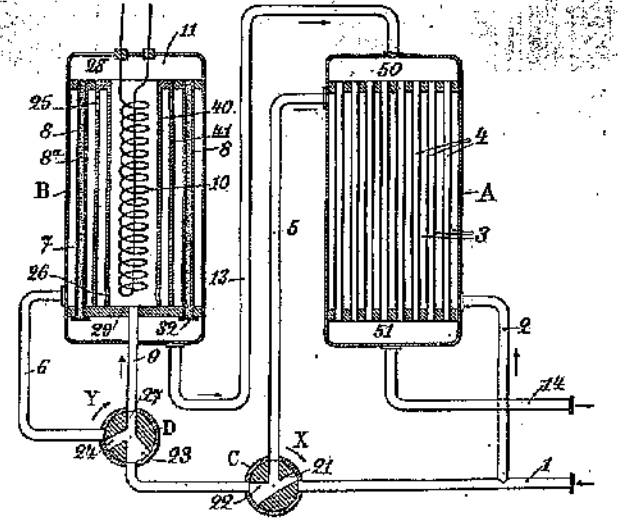


Fig. 3

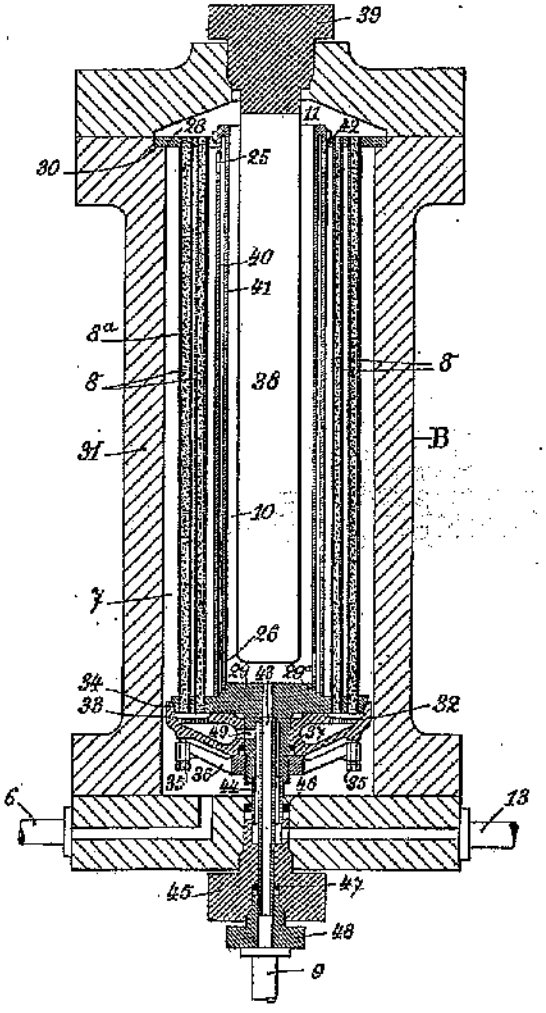
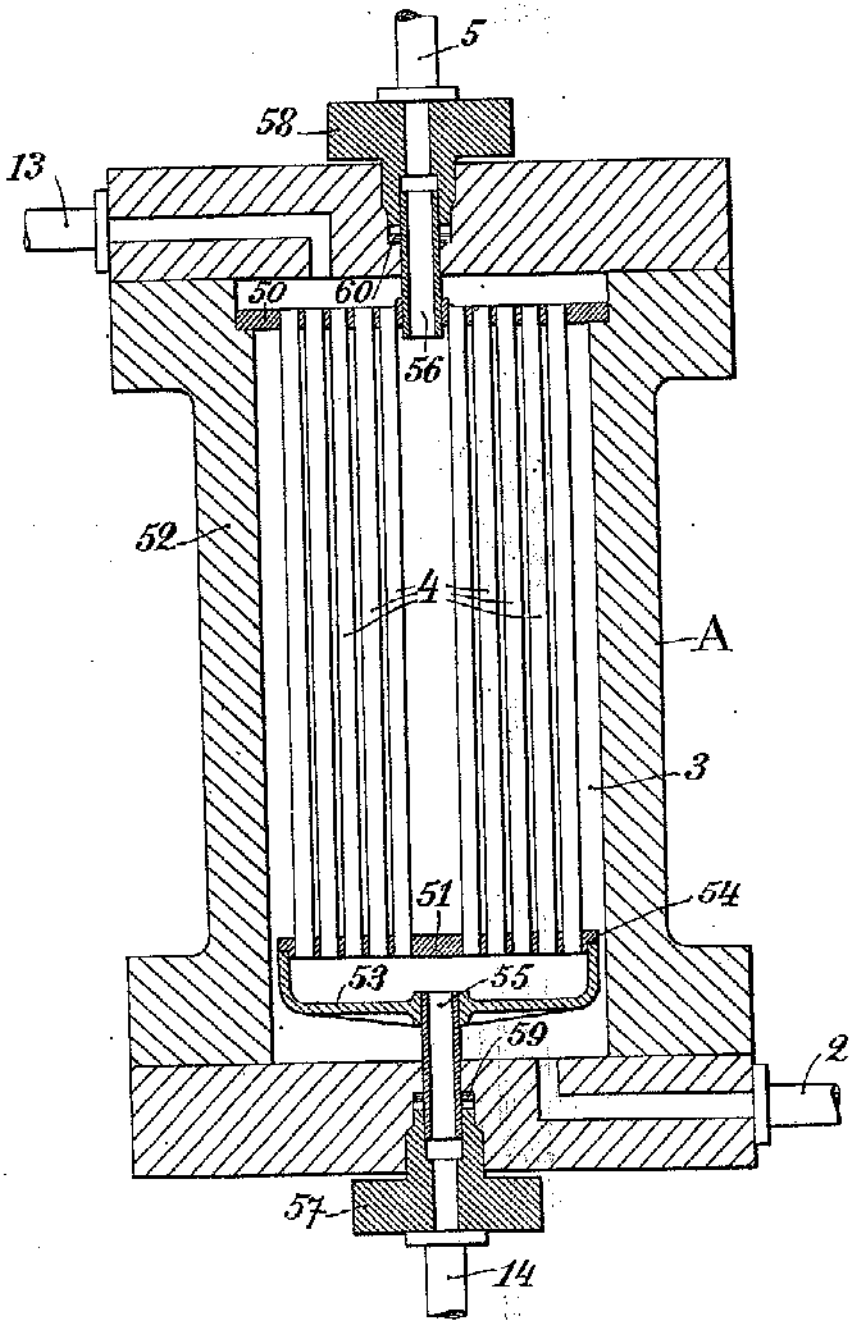


Fig. 4



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