PATENT SPECIFICATION

Application Date: Sept. 28, 1934. No. 27839/34. **444,898**

Complete Specification Left: Sept. 16, 1935.

Complete Specification Accepted: March 30, 1936.

PROVISIONAL SPECIFICATION

Improvements in and Apparatus for Carrying out Exothermic Catalytic Gas Reactions

I, JAMES YATE JOHNSON, a British Subject, of 47, Lincoln's Inn Fields, in the County of London, Gentleman, do hereby declare the nature of this invention (which has been communicated to me from abroad by I. G. Farbenindustrie Aktiengesellschaft, of Frankfort-on-Main, Germany, a Joint Stock Company organized under the Laws of Germany), to be as follows:—

It has already been proposed to carry out exothermic catalytic gas reactions in tubular reaction chambers while employing cooling means. For the said purpose tubes filled with catalyst are usually employed, the said tubes being combined in the form of a bundle, the gas to be reacted flowing along the hot external walls and thus entering the tubes in a preheated condition. When working with gases which have a very high or greatly fluctuating content of constituents to be reacted, such as is the case for example with the content of sulphur dioxide in the catalytic production of sulphuric acid from the same, there is the difficulty that the cooling means hitherto proposed and their arrangement do not always exert a satisfactory effect. When employing highly concentrated gases and carrying out strongly exothermic catalyses, injury to the upper parts of the reaction chamber easily occurs; this may become evident by the burning of the wrought iron tubes. Furthermore the efficiency of the apparatus is reduced by the overheating of the catalyst.

My foreign correspondents have now found that exothermic catalytic reactions can be carried out in tubular reaction chambers without the said drawbacks and in particular can be controlled without difficulty in the case of a high or fluctuating concentration of the initial gases, by subdividing the layer of catalyst and by flushing the tubes before such places at which overheating would otherwise occur with an additional current of fresh gas, preferably at places at which the tubes are free from catalyst. In this manner it is possible for example in the catalytic conversion of gases obtained by roasting pyrites into sulphur trioxide, to completely avoid the hitherto usual formation of laminated iron cinder which becomes detached when the reaction chamber is cooled and thus causes stoppage of the spaces between the tubes as well as of supply openings and distribution openings for the fresh gas. Furthermore a reaction chamber arranged according to this invention works with a smaller gas resistance because the catalyst is absent from those places in the tubes where otherwise there is only a small conversion, the temperature being too high; for example it is possible to reduce the length of the layer of catalyst, and consequently the gas resistance, by 25 per cent. or more without impairing the degree of conversion. Furthermore it is also possible to employ strongly active, but heat-sensitive catalysts which cannot resist overheating.

The process according to this invention may be employed with special advantage in conjunction with a recently proposed process for carrying out exothermic catalytic reactions; in the latter process, the catalyst is arranged partly within the tubes and partly in the spaces between the tubes and the reaction gas, after partial conversion in one or both parts of the catalyst, passes into a collecting chamber for the purpose of temperature equalisation, where if desired a lowering or raising of the equalised temperature may be carried out before the further conversion, then the gas is led into the other part of the catalyst after reversing the direction of flow. According to another process already proposed, a chamber filled with catalyst is attached to the bundle of tubes and the current of gas coming from the single tubes flows through the said chamber for the purpose of completing the reaction. In order to prevent too great an accumulation of the heat of reaction, the further precaution may be employed that one reaction component is at first only used in a deficient amount, the amount necessary to complete the reaction being added later at one or more places to the current of gas.

The nature of the invention will be further described with reference to the

[*Price* 1/-]

accompanying drawings which illustrate arrangements of apparatus suitable for carrying out the process according to this invention but the invention is not restricted to the particular arrangements shewn.

Referring to Figure 1, the gases to be converted enter the reaction chamber through the attachment 1 and are distributed through holes 2 in the space surrounding the reaction chamber and through the pipes 3 and 4 in the space in the middle of the reaction chamber. They flow past the outer walls of the tubes charged with catalyst A—B and are heated by the heat of reaction developed in the tubes. Between B and C, an additional current of fresh gas is supplied; this may be regulated by regulating members, such as valves, and passes through the attachment 5, holes 6 and pipes 7 and 8. The gas mixture then flows past the outer walls of that portion of the tubes C—D wherein the first step of the catalysis takes place, and cools the partially converted gases therein, it then enters the catalyst at the temperature necessary for ignition. If the temperature of the gas mixture entering the tubes is substantially above that sufficient for ignition, further amounts of fresh gas may be introduced into the upper chamber through the inlet 9. Outside the apparatus is a heat-exchanger for transferring the heat of the gases leaving the apparatus to the inflowing fresh gas. The manner in which this heat-exchanger operates is evident from the drawing. This heat-exchanger serves the purpose, in the case of any dilution of the initial gas, of lessening the cooling of the partially converted gas flowing in the tubes between C and B, by correspondingly preheating by means of the heat-exchanger the cooling gas which is to be supplied between C and B.

Referring to Figure 2, the portion of the catalyst in the upper part of the tubes has been removed and arranged in the space between the tubes. The first step of the catalysis is carried out in this layer of catalyst which is exposed to heat radiation from the upper cover of the apparatus.

Figure 3 illustrates an apparatus the efficiency of which is increased by the arrangement in the last part of the path of the gas mixture of a layer of catalyst G in which the last traces of the constituents to be converted are caused to react.

The arrangement shewn in Figure 4 is intended for exothermic reactions in which the evolution of heat is especially strong. In this case the layer of catalyst is subdivided into three portions and a further means (9, 10, 11 and 12) for the supply of fresh gas is provided. The apparatus shewn also renders it possible to carry out the catalysis with a deficiency of one of the components until the gas mixture reaches the layer of catalyst G, the gas necessary for completing the conversion being supplied through the member 14.

Dated this 28th day of September, 1934.

J. Y. & G. W. JOHNSON,
47, Lincoln's Inn Fields, London, W.C.2,
Agents.

COMPLETE SPECIFICATION

Improvements in and Apparatus for Carrying out Exothermic Catalytic Gas Reactions

We, COUTTS & COMPANY, a Company with unlimited liability, incorporated under the Companies Acts, of 440, Strand, in the County of London, and FREDERICK JOHNSON, a British Subject, of 218, Victoria Drive, Eastbourne, in the County of Sussex, legal representatives of JAMES YATE JOHNSON, deceased, late of 47, Lincoln's Inn Fields, in the County of London, do hereby declare the nature of this invention (which has been communicated from abroad by I. G. Farbenindustrie Aktiengesellschaft, of Frankfort-on-Main, Germany, a Joint Stock Company organized under the Laws of Germany), and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

It has already been proposed to carry out exothermic catalytic gas reactions in tubular reaction chambers while employing cooling means. For the said purpose tubes filled with catalyst are usually employed, the said tubes being combined in the form of a bundle, the gas to be reacted flowing along the hot external walls and thus entering the tubes in a preheated condition. When working with gases which have a very high or greatly fluctuating content of constituents to be reacted, such as is the case for example with the content of sulphur dioxide in the catalytic production of sulphuric acid from the same, there is the difficulty that the cooling means hitherto proposed and their arrangement do not always exert a satisfactory effect. When employing

highly concentrated gases and carrying out strongly exothermic catalyses, injury to the upper parts of the reaction chamber easily occurs; this may become evident by the burning of the wrought iron tubes. Furthermore the efficiency of the apparatus is reduced by the overheating of the catalyst.

Our foreign correspondents have now found that exothermic catalytic reactions can be carried out in tubular reaction chambers without the said drawbacks and in particular can be controlled without difficulty in the case of a high or fluctuating concentration of the initial gases, by subdividing the layer of catalyst and by cooling the gases within the tubes before such places at which overheating would otherwise occur by flushing the exterior of the tubes with an additional current of cool fresh gas, preferably at places at which the tubes are free from catalyst. In this manner it is possible for example in the catalytic conversion of gases obtained by roasting pyrites into sulphur trioxide, completely to avoid the hitherto usual formation of laminated iron cinder which becomes detached when the reaction chamber is cooled and thus causes stoppage of the spaces between the tubes as well as of supply openings and distribution openings for the fresh gas. Furthermore a reaction chamber arranged according to this invention works with a smaller gas resistance because the catalyst is absent from those places in the tubes where otherwise there is only a small conversion, the temperature being too high; for example it is possible to reduce the length of the layer of catalyst, and consequently the gas resistance, by 25 per cent. or more without impairing the degree of conversion. Furthermore it is also possible to employ strongly active, but heat-sensitive catalysts which cannot resist overheating. Our invention can be applied with great usefulness also to the catalytic oxidation of organic compounds such as naphthalene, anthracene, acenaphthene, and the like.

The process according to this invention may be employed with special advantage in conjunction with two recently proposed processes for carrying out exothermic catalytic reactions; in one of these, the catalyst is arranged partly within the tubes and partly in the spaces between the tubes and the reaction gas, after partial conversion in one or both parts of the catalyst, passes into a collecting chamber for the purpose of temperature equalisation, where if desired a lowering or raising of the equalised temperature may be carried out before the further conversion, then the gas is led into the other part of the catalyst after reversing the direction of flow. In the other process recently proposed, a chamber filled with catalyst is attached to the bundle of tubes and the current of gas coming from the single tubes flows through the said chamber for the purpose of completing the reaction. In order to prevent too great an accumulation of the heat of reaction, the further precaution may be employed that one reaction component is at first only used in a deficient amount, the amount necessary to complete the reaction being added later at one or more places to the current of gas.

The nature of the invention will be further described with reference to the accompanying drawings which illustrate arrangements of apparatus suitable for carrying out the process according to this invention but the invention is not restricted to the particular arrangements shown.

Referring to Figure 1, the gases to be converted enter the reaction chamber through the attachment 1 and are distributed through holes 2 in the space surrounding the reaction chamber and through the pipes 3 and 4 in the space in the middle of the reaction chamber. They flow past the outer walls of the tubes charged with catalyst A—B and are heated by the heat of reaction developed in the tubes. Between B and C, an additional current of fresh gas is supplied; this may be regulated by regulating members, such as valves, and passes through the attachment 5, holes 6 and pipes 7 and 8. The gas mixture then flows past the outer walls of that portion of the tubes C—D wherein the first step of the catalysis takes place, and cools the partially converted gases therein, it then enters the catalyst at the temperature necessary for ignition. If the temperature of the gas mixture entering the tubes is substantially above that sufficient for ignition, further amounts of fresh gas may be introduced into the upper chamber through the inlet 9. Outside the apparatus is a heat-exchanger for transferring the heat of the gases leaving the apparatus to the inflowing fresh gas. The manner in which this heat-exchanger operates is evident from the drawing. This heat-exchanger serves the purpose, in the case of any dilution of the initial gas, of lessening the cooling of the partially converted gas flowing in the tubes between C and B, by correspondingly preheating by means of the heat-exchanger the cooling gas which is to be supplied between C and B.

Referring to Figure 2, the portion of the catalyst in the upper part of the tubes has been removed and arranged in the

space between the tubes. The first step of the catalysis is carried out in this layer of catalyst which radiates to the upper cover of the apparatus.

Figure 3 illustrates an apparatus the efficiency of which is increased by the arrangement in the last part of the path of the gas mixture of a layer of catalyst G in which the last traces of the constituents to be converted are caused to react.

The arrangement shown in Figure 4 is intended for exothermic reactions in which the evolution of heat is especially strong. In this case the layer of catalyst is subdivided into three portions and a further means (9, 10, 11 and 12) for the supply of fresh gas is provided. The apparatus shown also renders it possible to carry out the catalysis with a deficiency of one of the components until the gas mixture reaches the layer of catalyst G, the gas necessary for completing the conversion being supplied through the member 14.

Figure 5 illustrates an arrangement for supplying the additional cool fresh gas to the catalyst tubes, in which the zones of each catalyst tube to be cooled are of less diameter than the other parts of the tube and in which these narrower parts of the tubes are surrounded by a ring-shaped fresh gas supply pipe, the outer diameter of this ring not exceeding that of the non-narrowed parts of the catalyst tube. The small tubes delivering the cool fresh gas to the rings are preferably arranged in parallel to the catalyst tubes and in grooves on the surface of these in order not to broaden the total external cross-section of each catalyst tube. Such catalyst tubes with fresh gas supply rings attached at narrowed zones offer the advantage that no lateral openings of the furnace are necessary for the insertion of the additional cooling device, as this latter can easily be mounted together with each single catalyst tube.

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. A process for carrying out exothermic catalytic gas reactions which comprises passing the gas to be reacted upon through a plurality of layers of catalytic material which are contained in a bundle of tubular reaction chambers and, if desired, in the spaces between said tubes, first along the outer walls of the tubes in heat exchange relation to the contents of the tubes and then through the said tubes, and cooling the gases within the tubes before such places at which overheating would otherwise occur, by flushing the exterior of the tubes with an additional current of cool fresh gas.

2. A process as claimed in claim 1, in which the flushing of the exterior of the tubes is carried out at places at which the tubes are free from catalyst and at which the greater part of the reaction has already been performed.

3. The process for carrying out exothermic catalytic gas reactions substantially as herein described with reference to the accompanying drawings.

4. Apparatus for carrying out exothermic catalytic gas reactions substantially as herein described or shown in the accompanying drawings.

Dated the 16th day of September, 1935.

J. Y. & G. W. JOHNSON,
47, Lincoln's Inn Fields, London, W.C.2,
Agents.

Leamington Spa: Printed for His Majesty's Stationery Office, by the Courier Press.—1936.

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

Fig. 1.

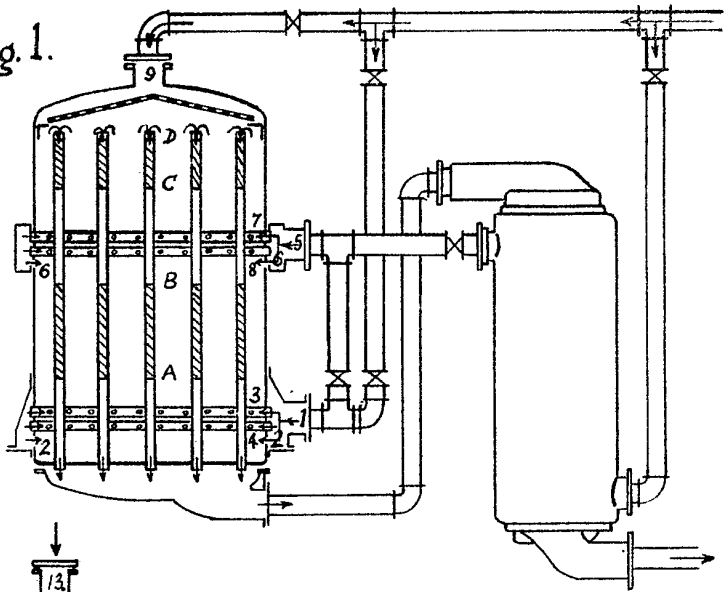

Fig. 4.

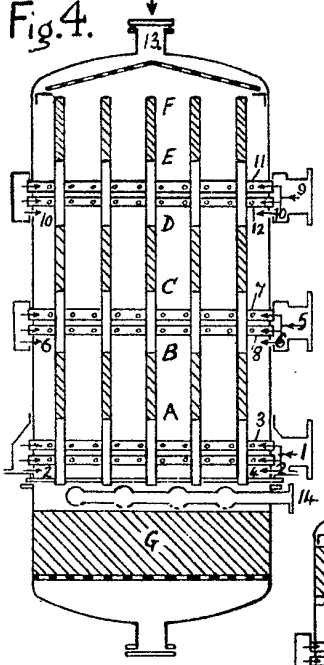

Fig. 3.

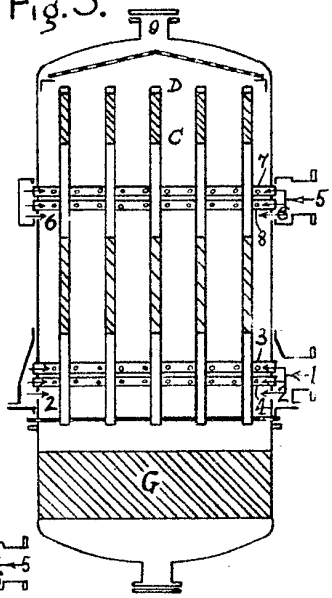

Fig. 2.

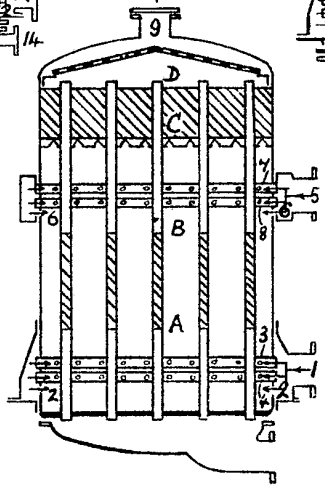

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Fig. 1.

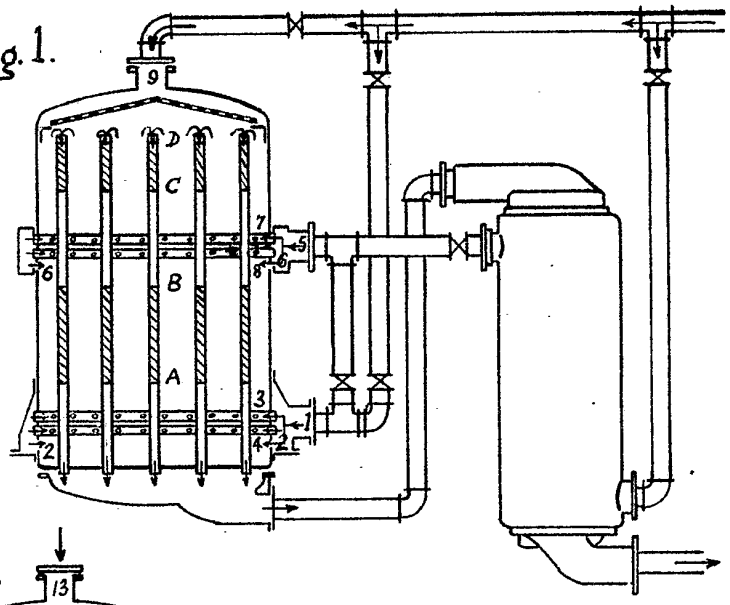


Fig. 4.

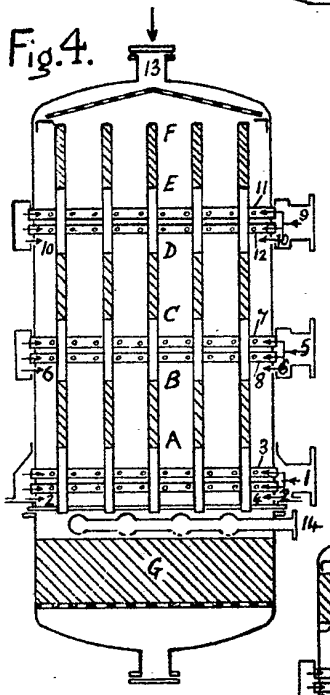


Fig. 3.

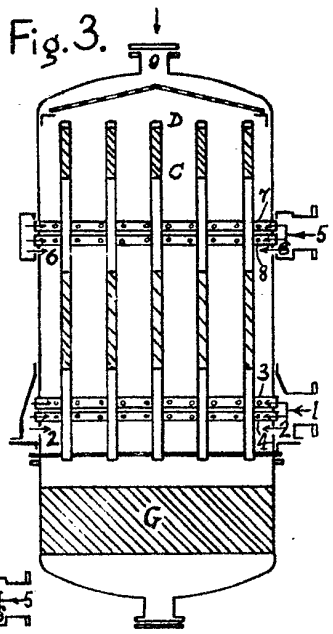


Fig. 2.

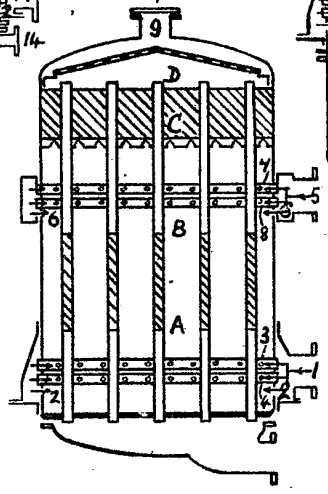
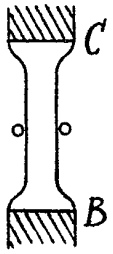


Fig. 5.



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