[Second Edition.]

PATENT SPECIFICATION

303,192

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PROVISIONAL SPECIFICATION.

Improvements in Carrying Out Exothermic Catalytic Gas Reactions and in Apparatus therefor.

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 is recognised as useful, and sometimes even necessary, that exothermic catalytic gas reactions be carried out either in several stages, intermediate coolers being used, or that the heat of the reaction be carried away in some way or other as soon as generated, thereby regulating the reaction temperature and maintaining it at such a level that a maximum output is obtained for a given velocity of the reaction and the corresponding gaseous equilibrium. In order to effect this it is essential that the temperature in the chamber containing the catalyst should be approximately constant throughout the reaction, or slowly decline, in the catalyst layer, in the direction taken by the gases.

Vessels have already been designed in which for the purpose above mentioned, the chamber containing the catalyst is divided into separate parallel tubes, charged with the catalyst, the said tubes being secured in a tube plate at both ends and bathed by the entering cold fresh gas before the latter passes over or through the catalyst. In this arrangement, however, owing to the distance at which the tubes have to be spaced in order to bring the gas into contact with the whole surface of the tubes, the entire vessel space is very imperfectly utilised, and for the same reason the transmission of heat from the catalytic mass to the fresh gas is very incomplete.

It has also been proposed to lay the catalytic mass round the tubes, in which case the latter must be set still further apart. In this case, the fresh gas first passes through the tubes, from below upwards, and then through the catalytic mass from above downwards. In this case also, the exchange of heat is naturally poor, owing to the small superficial area of the tubes.

My foreign correspondents have now found that particularly good regeneration and distribution of the heat, coupled with a favourable utilisation of space, and consequently a substantial technical effect is attained by passing the gas through tubes containing the catalyst, which tubes may be fastened at only one end, if so desired, to a tube plate, and which are closely packed together to a bundle so that the fresh gas led past the tubes, on the outside of the same, travels at higher velocity than it does in the tubes containing the catalyst. In this manner the fresh gas is first led through the spaces between the tubes, for the purpose of an exchange of heat with the catalyst, and then preferably in the reverse direction through the tubes containing the catalyst. It is advisable to taper down the tubes to a small diameter at the end at which they are connected to the tube plate the said tapering affording the possibility of distributing the entering fresh gas uniformly over the spaces between the tubes. Under certain circumstances, the effect of the arrangement may be substantially increased by making the tubes of polygonal shape in cross-section such as square or hexagonal, instead of circular or oval, in order to utilise their superficial area more completely.

The warming up of the vessel to the reaction temperature is preferably effected by means of electric heating devices.

In a preferred construction of reaction vessel for carrying the invention into practical effect the cold fresh gas enters the vessel at the upper end thereof and flows in the first place along the inside of the shell. On reaching the end of a guide tube the fresh gas first passes into the intermediate spaces between a sheaf of tubes, ascending therethrough and passing, as required, over an electric heating device and then enters the said tubes, that is into the layer of the catalyst, at their upper end. In traversing this layer, the heat of the reaction is transmitted to the fresh gas flowing in the opposite direction on the outside. The

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gaseous mixture issues from the furnace at the lower part thereof. It is unnecessary to fill the tubes completely with the catalytic material; and they may for example be filled with the said material in their upper portion alone, according as is necessary for the reaction, the remaining portion of the tubes serving merely as a heat regenerator.

The larger intermediate spaces between the tubes sometimes unavoidable (for example when round tubes are used) and which are unfavourable to heat regeneration, may be charged with inert fillers, such as inserted iron rods, and thereby considerably reduced in dimensions. Large intermediate spaces may be avoided in a technically very simple manner, by making the individual catalyst tubes of polygonal shape in cross-section, for example, square or hexagonal, as aforesaid so that the said tubes will lie still more closely together, the chamber containing the catalyst will be larger in proportion to the total space, and the gas to be regenerated will flow at increased velocity between the parallel surfaces in close juxtaposition, the most favourable conditions for heat transmission being thereby established. In the case of round tubes, for example, the space surrounding the tubes occupies 22 per cent. of the total space taken up by the sheaf of tubes whereas, in the case of hexagonal tubes, it occupies only 3 to 5 per cent. The sheaf of tubes may be arranged in the form of a unit of any desired cross-sectional shape; and, in some cases, it is also advantageous to insert intermediate pieces between the parallel walls.

The tubes, especially those of regular hexagonal cross-section, may also be disposed with their longitudinal sides in contact so as to avoid interstices entirely, the whole then representing a chamber subdivided into numerous compartments by separating walls. In this case some of the tubes are filled with the catalyst, the remainder being left empty, or charged with fillers, to serve as regenerative spaces. In this arrangement it is advisable that the walls of the regenerative spaces should be in common with those of the adjoining spaces containing the catalyst; and the other separating walls in contact with each other then act solely to conduct heat to the regenerative spaces in which the heat is taken up by the fresh gas.

In this type of arrangement a substantial improvement in the utilisation of the vessel space and also in the regeneration of heat may be effected by inserting into the empty tubes, or in the tube-like intermediate spaces formed by a plurality of tubes, tubes of smaller diameter, preferably coaxially and in such a manner as to leave only a small space for the passage of the fresh gas between the outer and the inner tubes.

Dated this 23rd day of September, 1927.

JOHNSONS & WILLCOX,
47, Lincoln's Inn Fields, London, W.C. 2,
Agents.

COMPLETE SPECIFICATION.

Improvements in Carrying Out Exothermic Catalytic Gas Reactions and in Apparatus therefor.

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) and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

This invention relates to improvements in carrying out exothermic catalytic gas reactions and in apparatus therefor.

It is recognised as useful, and sometimes even necessary, that exothermic catalytic gas reactions be carried out either in several stages, intermediate coolers being used, or that the heat of the reaction be carried away in some way or other as soon as generated, thereby regulating the reaction temperature and maintaining it at such a level that a maximum output is obtained for a given velocity of the reaction and the corresponding gaseous equilibrium. In order to effect this it is essential that the temperature in the chamber containing the catalyst should be approximately constant throughout the reaction, or slowly decline, in the catalyst layer, in the direction taken by the gases.

Vessels have already been designed in which for the purpose above mentioned, the chamber containing the catalyst is divided into separate parallel tubes, charged with the catalyst, the said tubes being secured in a tube plate at both ends and bathed by the entering cold

fresh gas before the latter passes over or through the catalyst. In this arrangement, however, owing to the distance at which the tubes have to be spaced in order to bring the gas into contact with the whole surface of the tubes, the entire vessel space is very imperfectly utilised, and for the same reason the transmission of heat from the catalytic mass to the fresh gas is very incomplete.

It has also been proposed to lay the catalytic mass round the tubes, in which case the latter must be set still further apart. In this case, the fresh gas first passes through the tubes, from below upwards, and then through the catalytic mass from above downwards. In this case also, the exchange of heat is poor, owing to the small superficial area of the tubes.

My foreign correspondents have now found that particularly good regeneration and distribution of the heat, coupled with a favourable utilisation of space, and consequently a considerable improvement is attained by passing the gas through tubes containing the catalyst, which tubes may be fastened at only one end, if so desired, to a tube plate, and which are so closely packed together to a bundle so that the fresh gas led past the tubes, on the outside of the same, travels at higher velocity than it does in the tubes containing the catalyst. In arranging the tubes in the said manner regard must be had to the packing of the catalyst or other solid material within the tubes in order to ensure a higher velocity of the gases outside the tubes. In this manner the fresh gas is first led through the spaces between the tubes, for the purpose of an exchange of heat with the catalyst, and then preferably in the reverse direction through the tubes containing the catalyst. It is advisable to taper down the tubes to a small diameter at the end at which they are connected to the tube plate, the said tapering affording the possibility of distributing the entering fresh gas uniformly over the spaces between the tubes. Under certain circumstances the effect of the arrangement may be substantially increased by making the tubes of polygonal shape in cross-section such as square or hexagonal, instead of circular or oval, in order to utilise their superficial area more completely.

The warming up of the vessel to the reaction temperature is preferably effected by means of electric heating devices.

The invention will be further described with reference to the accompanying drawing in which a preferred construction of reaction vessel for carrying the invention into practical effect is shewn. Figure 1 shews such a reaction vessel in vertical section and Figure 2 in horizontal section. The cold fresh gas enters the vessel at the upper end thereof at *a* and flows in the first place along the inside of the shell. On reaching the end of a guide tube *b*, the fresh gas first passes into the intermediate spaces between a sheaf of tubes, ascending therethrough and passing, if required, over an electric heating device *c* then entering the said tubes, and passing into the layers of the catalyst, at their upper ends. In traversing a layer, the heat of the reaction is transmitted to the fresh gas flowing in the opposite direction on the outside. The gaseous mixture issues from the furnace at the lower part *d* thereof. It is unnecessary to fill the tubes completely with the catalytic material, and they may for example be filled with the said material in their upper portions only, according to what is necessary for the reaction, the remaining portions of the tubes serving merely as a heat regenerator.

The larger intermediate spaces between the tubes sometimes unavoidable (for example when round tubes are used) and which are unfavourable to heat regeneration, may be charged with inert fillers, such as inserted iron rods, and thereby considerably reduced in dimensions. Large intermediate spaces may be avoided in a very simple manner, by making the individual catalyst tubes of polygonal shape in cross-section, for example square or hexagonal (see Figure 2) so that the said tubes will lie still more closely together, the chamber containing the catalyst will be larger in proportion to the total space, and the gas to be regenerated will flow at increased velocity between the parallel surfaces in close juxtaposition, the most favourable conditions for heat transmission being thereby established. In the case of round tubes, for example, the space surrounding the tubes occupies 22 per cent. of the total space taken up by the sheaf of tubes whereas, in the case of hexagonal tubes, it occupies only 3 to 5 per cent. The sheaf of tubes may be arranged in the form of a unit of any desired cross-sectional shape, and, in some cases, it is also advantageous to insert intermediate pieces between the parallel walls.

The tubes, especially those of regular hexagonal cross-section, may also be disposed with their longitudinal sides in contact so as to avoid interstices entirely, the whole then representing a chamber subdivided into numerous compartments by separating walls. In this case some of the tubes are filled with the catalyst, the remainder being left empty, or

charged with fillers, these being hereinafter referred to as regenerative spaces. In this arrangement it is advisable that the walls of the regenerative spaces should be in common with those of the adjoining spaces containing the catalyst, and the other separating walls in contact with each other then act solely to conduct heat to the regenerative spaces in which the heat is taken up by the fresh gas, the gas being preferably passed in an opposite direction to that in the said regenerative spaces.

In this type of arrangement a substantial improvement in the utilisation of the vessel space and also in the regeneration of heat may be effected by inserting into the empty tubes, or in the tube-like intermediate spaces formed by a plurality of tubes, tubes of smaller diameter, preferably coaxially and in such a manner as to leave only a small space for the passage of the fresh gas between the outer and the inner tubes.

A further modification for carrying out the process of the present invention is shewn in section in Figure 3 in which the fresh gases introduced and the gases passing over the catalyst both flow in the same direction.

The cold gas entering into the reaction vessel at *a* passes downwards between the hot bundles of tubes *b* which are filled with the catalyst and then passes upwards through the central tube *z*, then enters into the catalyst tubes through which the gas passes in a downward direction and then leaves the reaction vessel at *c*.

This arrangement is particularly advantageous for carrying out reactions which are accompanied by strong evolution of heat, which evolution does not, however, take place uniformly over the whole length of the catalyst. In this manner it is secured at that point of the reaction vessel where the evolution of heat is strongest, namely at the beginning of the catalyst layer, the amount of heat taken off by the cold gases is greatest, since the difference of temperature between the two gases is at the maximum at this point. In the case of some reactions, it may be advantageous to heat up the fresh gas to a certain extent in order to avoid cooling the catalyst tubes too strongly. This preheating may take place either outside or within the reaction vessel and may be carried out by any suitable heating device or the heat contained in the gases issuing from the bundles of tubes may be wholly or in part employed to supply heat to the fresh gas by a heat exchanger operated on a counter current system, which heat exchanger may be of any known construction.

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

1. Apparatus for carrying out exothermic catalytic gas reactions, comprising a reaction vessel, tubes therein, the said tubes containing a catalyst, and being so closely packed together to a bundle having regard to the packing of solid material within said tubes that the fresh gas led past the tubes on the outside of the same, travels at higher velocity than it does in said tubes containing the catalyst.

2. In apparatus as claimed in the aforesaid first claiming clause the provision of tubes formed to taper down to a small diameter at the end at which they are connected to the tube plate.

3. In an apparatus as claimed in the foregoing first or second claiming clause, the provision of tubes made polygonal in cross-section.

4. In an apparatus as claimed in the foregoing first, second or third claiming clause, the arrangement of inert filling material in the larger intermediate spaces between the said tubes.

5. An apparatus as claimed in each of the foregoing claiming clauses, comprising a reaction vessel, tubes therein which are so closely packed together to a bundle that interstices are entirely avoided, the whole then representing a chamber subdivided into numerous compartments by separating walls, of which compartments part constitutes regenerative spaces and the other part is filled with a catalyst, the fresh gas being first passed through the regenerative spaces, so as to take up the heat of the catalyst and then through the compartments containing the catalyst.

6. Apparatus for carrying out exothermic catalytic gas reactions, comprising a reaction vessel, tubes therein which are closely packed together to a bundle that interstices are entirely avoided, the whole then representing a chamber sub-divided into numerous compartments by separating walls, of which compartments part constitutes regenerative spaces and the other part is filled with a catalyst, the said regenerative spaces containing coaxially arranged bodies of smaller diameter leaving small interstices for the passage of fresh gas, which after having absorbed heat of the catalyst passes through the compartments containing the catalyst.

7. Apparatus as claimed in the first claiming clause, comprising a reaction vessel, tubes therein which contain a catalyst and are so closely packed

together to a bundle that the walls of the said tubes are partly in contact with each other, the fresh gas being led past the tubes on the outside of the same travelling at a higher velocity than it does in the said tubes containing the catalyst.

8. Apparatus for carrying out exothermic catalytic gas reactions, comprising a reaction vessel, a bundle of tubes containing a catalyst arranged therein, the said tubes having part of their walls in common so that interstices are formed between the tubes through which the fresh gas is first passed so as to take up the heat of the catalyst and then passes through the tubes containing the catalyst.

9. Apparatus substantially as hereinbefore described and illustrated in Figures 1 and 2, and in Figure 3 of the accompanying drawings.

Dated this 14th day of June, 1928.

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Agents.

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2nd Edition

Fig. 1

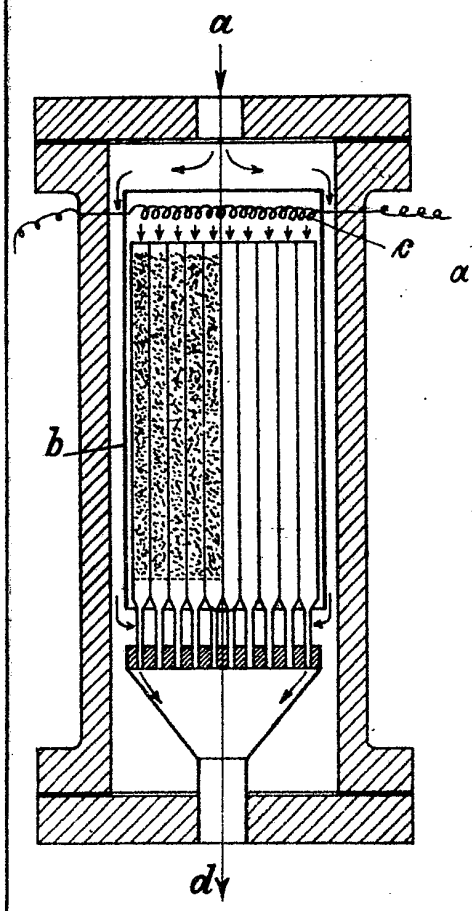


Fig. 2

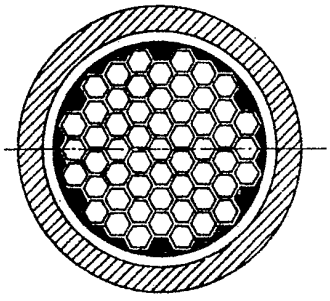
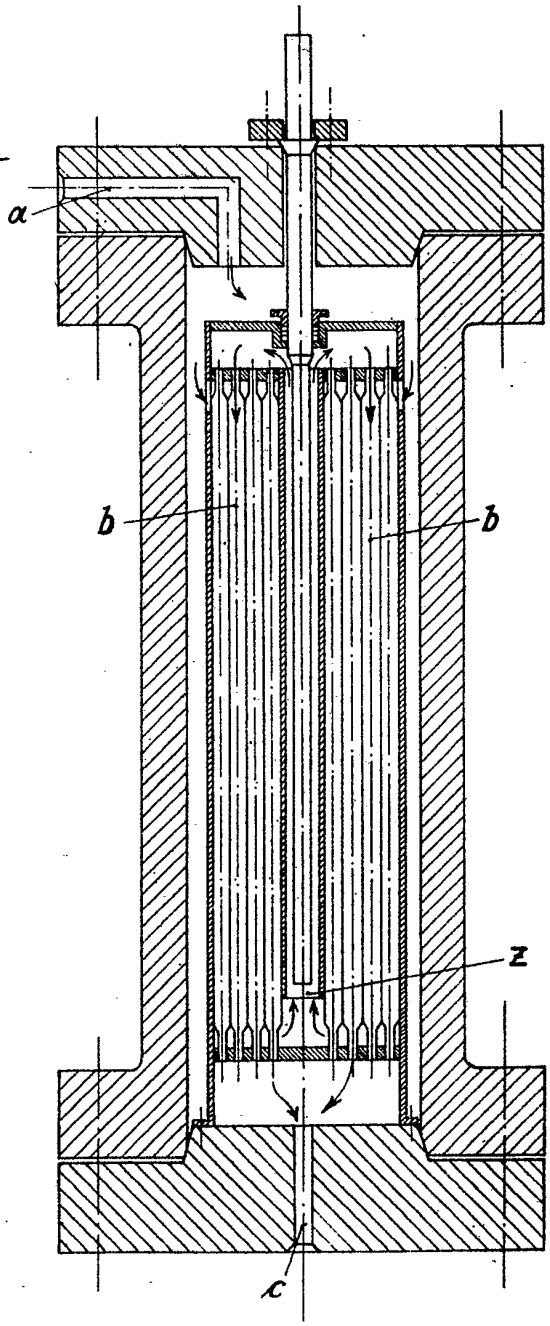


Fig. 3



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

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