[Second Edition.]

PATENT SPECIFICATION

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268,722

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Complete Accepted: Aug. 11, 1927.

3429

COMPLETE SPECIFICATION.

Improvements in or relating to Processes for Carrying Out Exothermic Chemical Reactions.

We, L'AIR LIQUIDE SOCIETE ANONYME POUR L'ETUDE ET L'EXPLOITATION DES PROCEDES GEORGES CLAUDE, a French company, of 48, rue St. Lazare, Paris (Seine), in the Republic of France, Assignees of SOCIETE CHIMIQUE DE LA GRANDE PAROISSE (Azote & Produits Chimiques), of 18, rue des Saussaies, Paris, in the Republic of France, a French 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:—

This invention relates to processes for carrying out exothermic catalytic chemical reactions and in particular to processes for the synthesis of ammonia from its elements. The principal object of the invention is to render the temperature as uniform as possible over the whole length of the catalysing material in contact with which the reaction is effected.

It is known that, in the case for example of the synthesis of ammonia, the first layers of the catalysing material which are traversed by the gases are those upon contact with which the largest quantity of heat is disengaged, and in various of our prior specifications there have been described processes which permit of the elimination along the whole length of the catalysing material of the heat disengaged by the reaction to the extent of and in proportion to its production.

The object of the present invention is to improve still further the results obtained, especially when the catalysing material is enclosed in a receptacle or chamber of fairly large diameter.

For this purpose according to the present invention the process for carrying out catalytic exothermic gaseous reactions consists in circulating the gases which are to react in such a manner that they first absorb heat from the catalysing material, then give up part of their heat to the coldest portions of the catalysing material, and finally absorb anew heat from the catalysing material before their admission on to it.

In order that the said invention may be clearly understood and readily carried into effect, the same will now be more fully described with reference to the accompanying drawings which show diagrammatically, by way of example, various methods of carrying out the present process, and in which:—

Figure 1 represents an apparatus in which the gases which are to react are admitted through a central tube, Figure 2 represents a modification, and Figure 3 represents an apparatus in which the gases are admitted through a group of tubes.

In the apparatus shown in Figure 1 the gases which are to react, either in the cold state or slightly heated, are admitted through the inlet A, pass through the central tube B, and then return through the **U**-shaped tubes C, C^1 finally impinging at the ends of the tubes C^1 upon the catalysing material. During this circulation the gases are heated to a lesser or greater extent in the interior of the tube B according, for example, as to whether the walls of the tube B possess a more or less bad thermal conductivity, and then they continue to absorb heat in the portions of the tubes C adjacent to the end of the catalysing material opposite to the inlet A, that is towards the end where the gases finally impinge upon the catalysing material; the gases thus heated thereafter yield up their heat in the portions of the tubes C,

C^1 which are adjacent to the inlet A, and become heated afresh during their passage through the parts of the tubes C^1 which are adjacent to the place where they are admitted on to the catalysing material. The gases traverse this material and pass away through the outlet D.

The transmission of heat between the gases passing through the interior of the tubes C, C^1 and the gases exterior thereto can be regulated by modifying the velocity of the interior gaseous current, and the nature, the surface, and the thickness of the transmitting walls. For example it can be arranged that the gases leaving the very hot regions of the catalysing material lose as little heat as possible in their passage through the tubes C before reaching the less hot regions which are situated at the opposite end of the catalysing chamber to the very hot regions.

Figure 2 represents an arrangement which is suitable for use in this latter case just mentioned; here the apparatus shown in Figure 1 is modified by placing the tubes C inside the tubes C^1. The tube C is of small cross section, and can be made heat-insulating if desired. The hot gases after having circulated in the region F where the reaction is most intense, traverse the tubes C and arrive, without having lost any sensible amount of heat, at the other end of the catalysing material in the region G where the reaction is less intense, and in this latter region they give up a part of their heat. Then these gases become heated again and up to the necessary temperature during their passage through the tubes C^1.

Thus this method of operation of the apparatus illustrated in Figure 2 which has for its object, in common with the other methods of operation according to the present invention, to prevent a local cooling of the catalysing material, is quite different from the operation of a device in which, for the opposite purpose of obviating a superheating of the catalyst at high concentrations of ammonia, the reacting gaseous mixture is conveyed through a plurality of double pipes, arranged in the contact chamber, in a double countercurrent in such manner that the entering fresh gases first pass through the inner pipes.

The arrangement of the tubes C, C^1 according to Figures 1 and 2 furthermore affords the advantage of avoiding or diminishing the difference in temperature between the gases impinging upon the catalyst material and the latter, when for any reason whatsoever the temperature at which the gases arrive at the apparatus tends to fall, the result of which might be to diminish or even stop the reaction altogether.

It will be understood that any suitable method of circulating and heating the gases prior to their passage through the said tubes C, C^1 may be employed; for example instead of the central inlet tube B there may be several similar gas inlet tubes distributed throughout the catalysing material.

It should also be observed that the arrangement of the U-shaped tubes shown in Figures 1 and 2 lends itself to a certain extent to the expansion of these tubes under the influence of heat, and that instead of being made as straight branches they could be made in the form of a coil. Moreover the resistance offered by the catalysing material to the expansion of the tubes may be diminished by providing their ends with pointed or tapered parts or by the use of other suitable devices.

In the case of the synthesis of ammonia and in order to avoid too high a temperature at the wall of the external pressure resisting tube H, use may be made, as indicated in Figure 1, of the method referred to in the Specification of our prior Patent No. 150,744. For this purpose a part of the gases which are to react, either slightly heated or unheated, is admitted through the inlet E and circulates in the space between the chamber J containing the catalysing material and the outer pressure resisting tube H; the chamber J is preferably covered on its exterior with a heat insulating material X. The part of the gas slightly heated by this circulation then passes through the orifices K on to the catalysing material.

Figure 3 shows a modified apparatus in which for the protection of the wall H there is used the process described in the Specification of our co-pending Application No. 268,721. In this figure, which is given solely by way of example, the gases which are to react are admitted through the inlet A and ascend through the outer tubes F of the group of tubes F, F^1, B, which outer tubes are more or less adjacent to the walls of the reaction chamber J. Upon arrival at the ends of the tubes F the gases then descend through the inner tubes F^1 of the group of tubes and finally reascend through a central tube B from which they pass out to the catalysing material in contact with which their combination takes place. After passing through the catalysing material the gaseous mixture leaves the chamber J through the outlet

D. The reaction chamber J is located inside a pressure resisting container H and in the space between the chamber J and the pressure resisting container H there is circulatd a gas which enters through the inlet E and passes off through the outlet M.

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 catalytic exothermic gaseous reactions which consists in circulating the gases which are to react in such a manner that they first absorb heat from the catalysing material, then give up part of their heat to the coldest portions of the catalysing material, and finally absorb anew heat from the catalysing material before their admission on to it.

2. An apparatus for carrying out the process according to Claim 1, in which for the circulation of the aforementioned gases there are provided one or more tubes having one or more branches in one direction and one or more branches in the opposite direction so that the gases after having circulated in contact with the warmest regions of the catalysing material are returned to the colder regions of the catalysing material and from thence are again returned towards the warmest regions where admission of the gases on to the catalysing material and their resultant combination ensues.

3. An apparatus for carrying out the process according to Claim 1, in which the gases which are to react are first heated by their circulation in indirect contact with the catalysing material in countercurrent to the gases during reaction, then pass in the opposite direction from the warmest portions of the catalysing material to the coldest portions through one or more tubes in which there is practically no exchange of heat, and then, by passage through one or more outer tubes into which each of the aforesaid tubes opens, circulate through the coldest portions of the catalysing material to which they give up some of their heat and are returned towards the warmest regions whereby they are heated again before admission on to the catalysing material.

4. A process for carrying out catalytic exothermic gaseous reactions substantially as described.

5. A process as in Claim 1 or 4 adapted to the synthetic production of ammonia from its elements.

6. An apparatus for carrying out the process as claimed in Claim 1, 4 or 5 constructed and adapted to operate substantially as described with reference to any of the constructions illustrated in the accompanying drawings.

Dated this 26th day of October, 1926.

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Agents for the Applicants.

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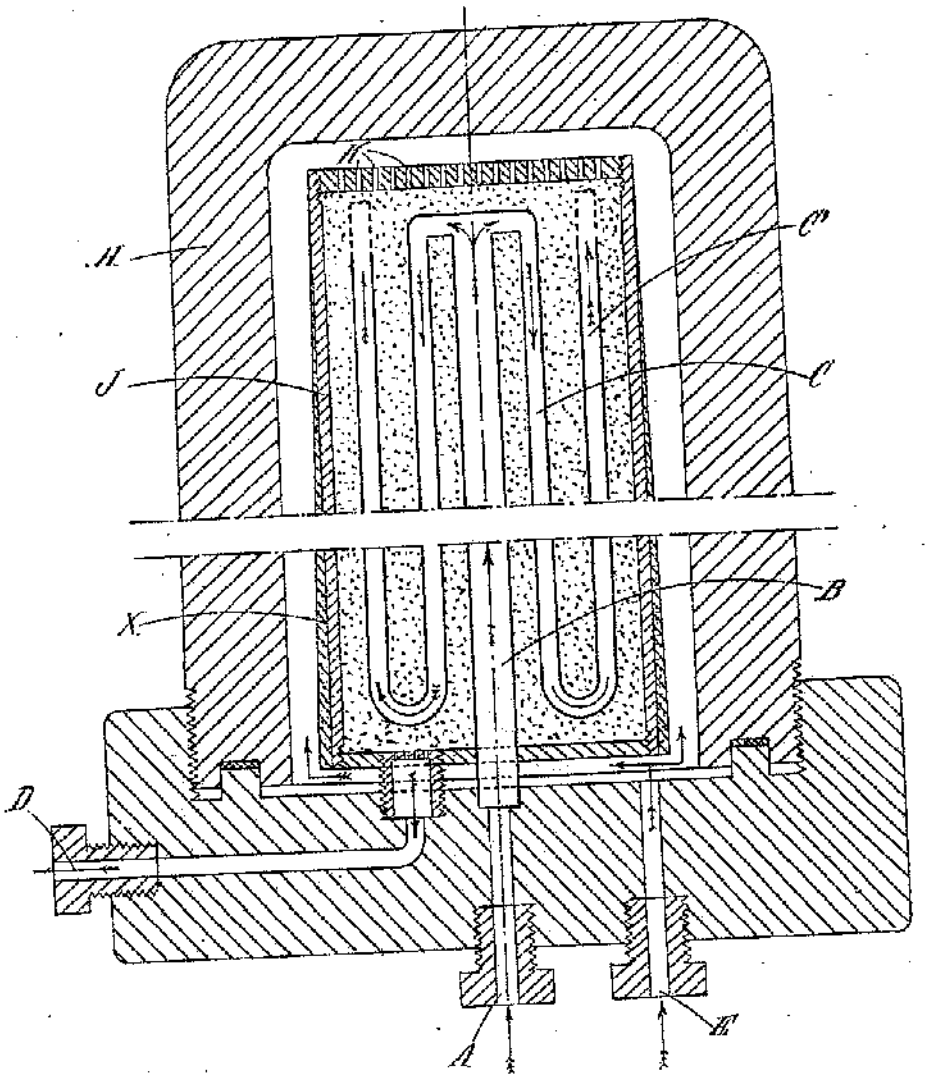
2nd Edition

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

Fig. 1.



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SHEET 2.

Fig. 2.

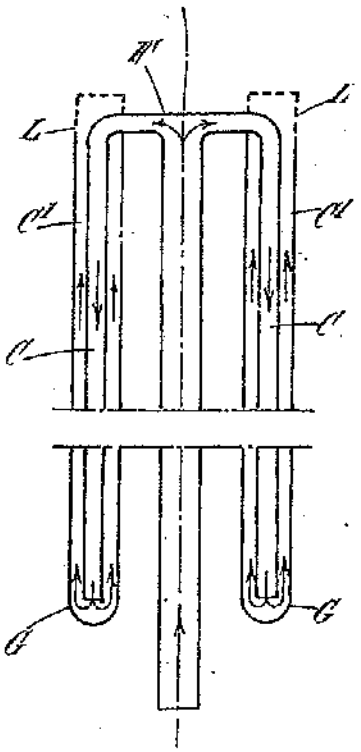

Fig. 3.

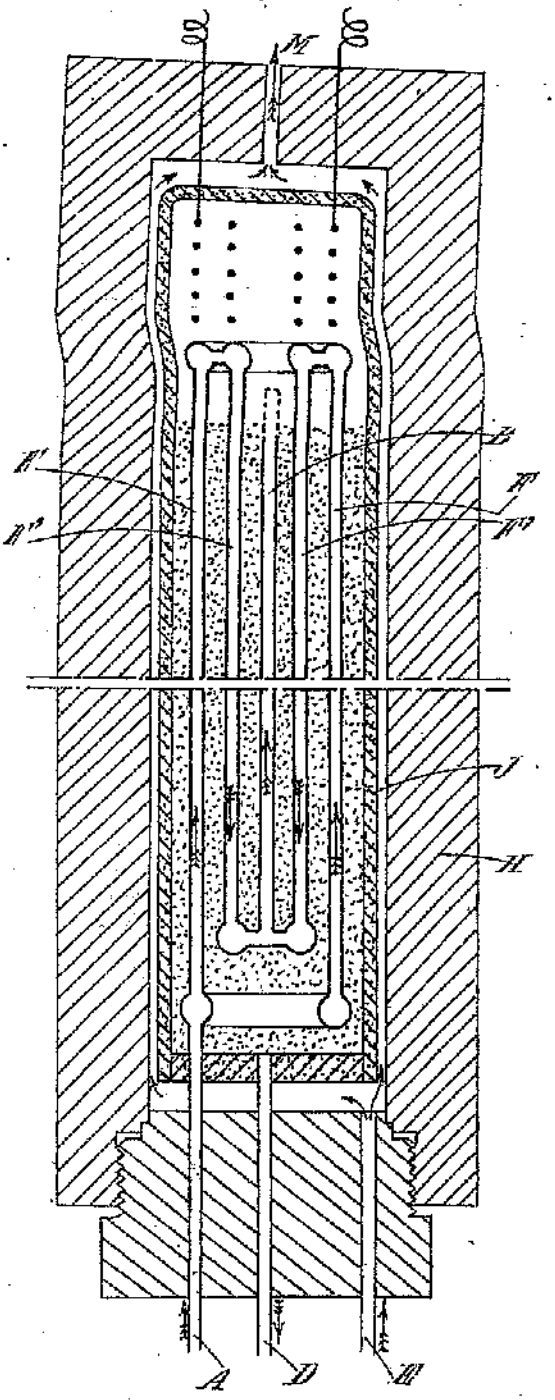

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Fig. 1.

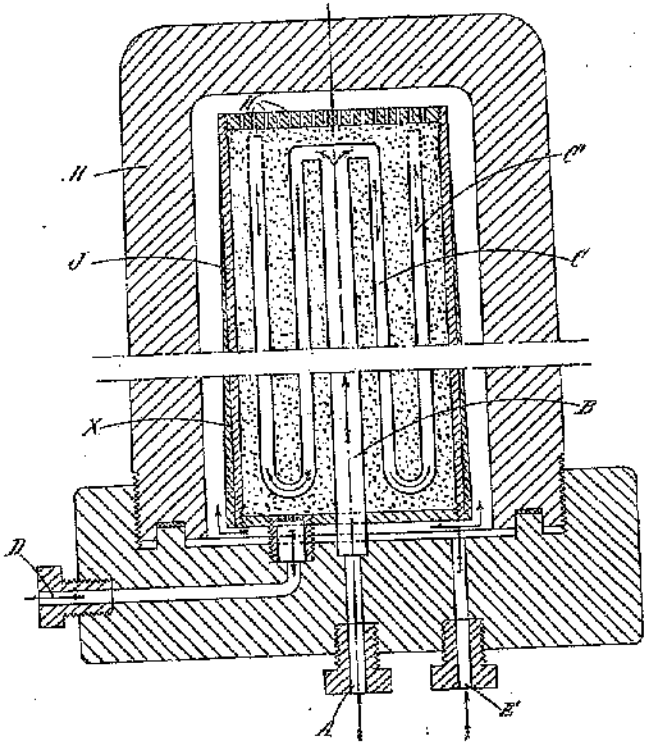


Fig. 2.

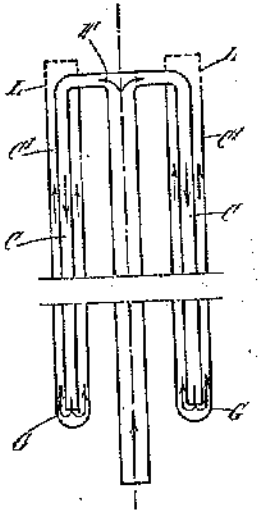
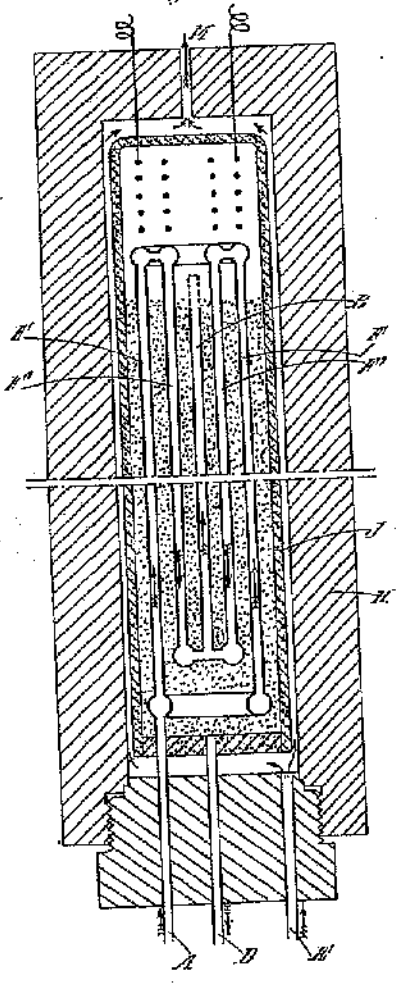


Fig. 3.



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