[Second Edition.]

PATENT SPECIFICATION

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268,721

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Complete Accepted: July 7, 1927.

3428

COMPLETE SPECIFICATION.

Improvements in or relating to Processes for Carrying out Exothermic Chemical Reactions under Pressure and at a High Temperature.

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 which take place under pressure and at a high temperature, and in particular to processes for the synthesis of ammonia from its elements. The principal object of the invention is to protect the metal wall of the pressure-resisting receptacle, which contains the chamber wherein the reaction is carried out, against the detrimental effect on its resistance due to the temperature of the reaction and the gases either before or after reaction, and at the same time properly to ensure the removal of the heat liberated in the inside of the aforesaid chamber.

It has already been mentioned in various of our prior specifications that the protection above referred to may be brought about by circulating between the chamber in which the reaction takes place and the outer pressure-resisting receptacle or tube the whole or a part of the gases which are to react and which are previously either unheated or only slightly heated, the said gases afterwards passing into the chamber wherein they finally combine in contact with the catalysing material. A method of this kind is indicated, for example, in our prior Specification No. 150,744 in which a part of the gases before reaction and whilst still cold form a protective screen for the external walls of the apparatus by circulating them between the said walls and the tube containing the catalysing material. In the specification just mentioned and also in certain of our other prior specifications there have also been described various means for absorbing the heat liberated by the reaction, including the utilization of the capacity of the gases, before they react, of being heated by absorption of heat up to their reaction temperature.

Now if, for example, in a catalysing apparatus of given production a more active catalysing material be used, it may happen that the means for the absorption of the heat hereinbefore indicated will not suffice to absorb the surplus heat so that the working pressure would have to be lowered.

The present invention provides another means for eliminating the large quantity of heat produced in such cases, whilst insuring under suitable conditions the protection of the pressure-resisting walls.

According to the present invention a process for carrying out exothermic chemical reactions under pressure and at a high temperature of the type in which a part or practically the whole of the heat disengaged by the reaction is caused to be absorbed by the gases which are to react immediately before the combination of the said gases consists in circulating between the outer pressure-resisting receptacle or tube and the inner device comprising the receptacle or tube containing the catalysing material a suitable gas under conditions so regulated that the said gas forms a protective screen for the walls, that is so that the part of the heat of reaction which reaches the said gas heats it at the utmost to the highest degree permissible for the resistance of

the outer receptacle or tube, and then discharging to the outside of the pressure resisting receptacle or tube the gas which has been thus circulated; the said gas may thereafter be cooled if necessary and the cooled gas utilised under the same conditions as it was previously used in a repetition of the process.

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

Figure 1 shows the whole of the tube containing the catalysing material and the outer receptacle the walls of which are protected by the gases before their reaction; Figure 2 shows a modification, and Figure 3 a further modification in which the protection of the walls is effected by the gases which have reacted.

In the apparatus shown in Figure 1 the tube or chamber containing the catalysing material consists essentially of a cylindrical chamber A the walls of which are of bad thermal conductivity; for this purpose the walls may be made for example of ordinary metal covered with a badly conducting material B. The outer pressure-resisting receptacle or tube is designated by C. The chamber A is entirely closed except for an inlet for the gases which are to react, an outlet for the gases which have reacted, and also small openings for the passage of the wires for the interior electric heating that may be required. In the space between the outer wall of the chamber A and the inner wall of the pressure-resisting tube C there is circulated a gas which in this particular case comprises the mixture of gases that are to react, the said mixture being admitted through the conduit D and leaving through the conduit E. During this circulation this gaseous mixture is heated to an extent which depends upon the surface, the thickness and the conductivity of the material B, and also upon the velocity of the current, by the part of the heat of reaction which passes through the wall of the chamber A. The extent to which the gaseous mixture is heated must however always be less than that at which it would no longer serve as a protective screen for the wall of the pressure-resisting tube C, that is to say it must not be heated to a temperature at which the said wall commences to lose its power of resistance. It should be noted that this heating of the gaseous mixture may be very small and reduced practically to the inevitable losses of heat consequent upon the imperfection of the heat insulation of the chamber A. The gaseous mixture which leaves through the conduit E is then cooled if necessary in the cooler F, whence it passes through the tube G into the interior of the chamber A where it circulates through a group of tubes H at the same time becoming heated by absorption of the heat of reaction. After being heated in this manner, and also additionally heated if necessary by means of an electric heating coil I, the gaseous mixture traverses the catalysing material where combination occurs and finally passes out through the conduit K whence it is delivered to the apparatus for collecting the product resulting from the combination.

As has been described in our prior Specification No. 171,970, the thermal conductivity of the outer wall of the chamber A may, if necessary, not be the same along the whole length of this wall; for example the heat-insulating material B may be of greater thickness towards the end of the chamber A close to the inlet G than towards the other end. In the same way the inner group of tubes H may have a thermal conductivity which varies at different places, or the surface of the said group of tubes may be greater in the hottest zone of the catalysing chamber, or both these features may be simultaneously used.

Alternatively the circulation of the gas in the space between the chamber A and the pressure-resisting tube C could take place in a reverse direction from that shown.

A by-pass M permits of regulating at will the quantity of gas circulating around the heat-insulated chamber A.

If necessary the chamber A may contain a heat exchanger through which there is circulated on the one hand the gases which are to react and which are admitted to the chamber A through the tube G and on the other hand the gases immediately upon their leaving the catalysing material.

In the arrangement shown in Figure 2 the inner tube A containing the catalysing material is enclosed by a jacket N which may be made of a material of bad thermal conductivity, as is necessary in the case of the synthesis of ammonia. The protecting gases which are admitted through the conduit D circulate between the jacket N and the outer tube C, passing off through the conduit E, whilst another part of the gases which are to react is admitted through the tube G, circulates in the interior of the jacket N

around the chamber A, becomes heated, and afterwards traverses the catalysing material where combination occurs, finally leaving by the conduit X. After suitable cooling and separation of the compounds formed in the separator L, this part of the gases may rejoin the other part of the gases which have served as the protective screen and which pass off through the tube E and are thereafter cooled if necessary; the two gaseous portions thus united may be sent to the following tube or may be returned by a compressor to the chamber A.

If the two gaseous portions which are admitted at D and G are at the same pressure and the same temperature, they may be admitted through one single tube and be divided afterwards in the apparatus by means of orifices in the lower part of the jacket N.

In the arrangement shown in Figure 3 the gases which are to react are admitted through the conduit G directly into the chamber A where they circulate in the interior of the group of tubes H; they then pass in the reverse direction round the exterior of this group of tubes and in contact with the catalysing material where they combine, afterwards leaving the chamber A through the conduit K, the product of the combination being separated either wholly or partially in the separator L after recovery if desired of the heat of the gases. The residual gases from this separation which are already cooled or which are then cooled if necessary return through the conduit D into the space between the outer wall of the chamber A and the inner wall of the pressure-resisting tube C, leaving the said space through the conduit E and being afterwards cooled to a suitable extent before being returned to the following catalysing tube.

A by-pass M permits of regulating the circulation of the gases round the chamber A.

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 chemical reactions under pressure and at a high temperature of the type in which a part or practically the whole of the heat disengaged by the reaction is caused to be absorbed by the gases which are to react immediately before the combination of the said gases, which process consists in circulating between the outer pressure-resisting receptacle or tube and the inner device comprising the receptacle or tube containing the catalysing material a suitable gas under conditions so regulated that the said gas forms a protective screen for the walls, that is so that the part of the heat of reaction which reaches the said gas heats it at the utmost to the highest degree permissible for the resistance of the outer receptacle or tube, and then discharging to the outside of the pressure-resisting receptacle or tube the gas which has been thus circulated.

2. A process as in Claim 1, in which the gas which has been circulated between the outer pressure-resisting receptacle or tube and the inner device comprising the receptacle or tube containing the catalysing material is thereafter cooled if necessary and the cooled gas is utilised under the same conditions at it was previously used in a repetition of the process.

3. A method of carrying out the process according to Claim 1 or 2, in which the gases which are to react are utilised for circulation between the outer pressure-resisting receptacle or tube and the inner device, the said gases after being thus circulated being cooled if necessary and thereafter combined.

4. A modification of the method of carrying out the process according to Claim 3, in which a whole or part of the gases which are to react circulate, immediately before their combination, around the receptacle or tube containing the catalysing material and are separated by a wall from the part of the gases which are to react which circulate round the exterior of said wall and between the latter and the outer pressure-resisting receptacle or tube.

5. A method of carrying out the process according to Claim 1 or 2, in which gases which have reacted are utilised for the circulation between the outer pressure-resisting receptacle or tube and the inner device.

6. A process as in any of the preceding claims adapted to the synthesis of ammonia from its elements.

7. Apparatus for carrying out the process as claimed in any of the preceding claims constructed, arranged and adapted to operate substantially as described with reference to any of the arrangements illustrated in the accompanying drawings.

Dated this 25th day of October, 1926.

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2nd Edition

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Fig. 1.

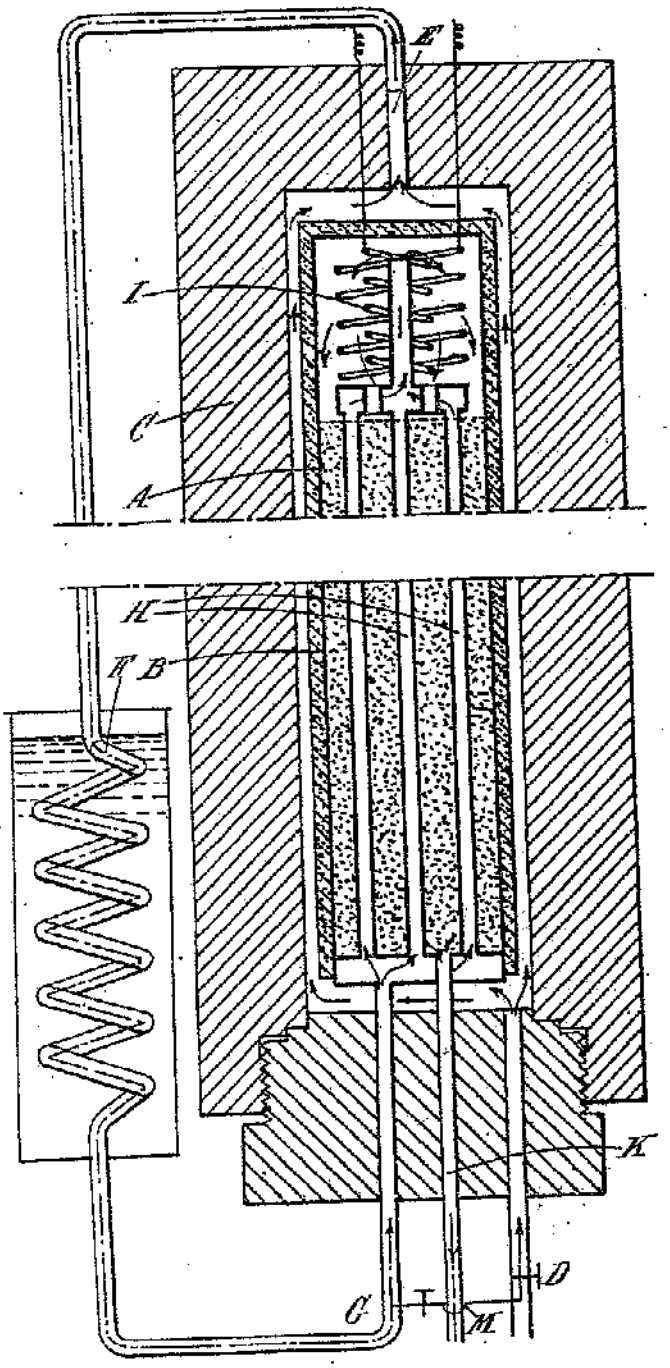
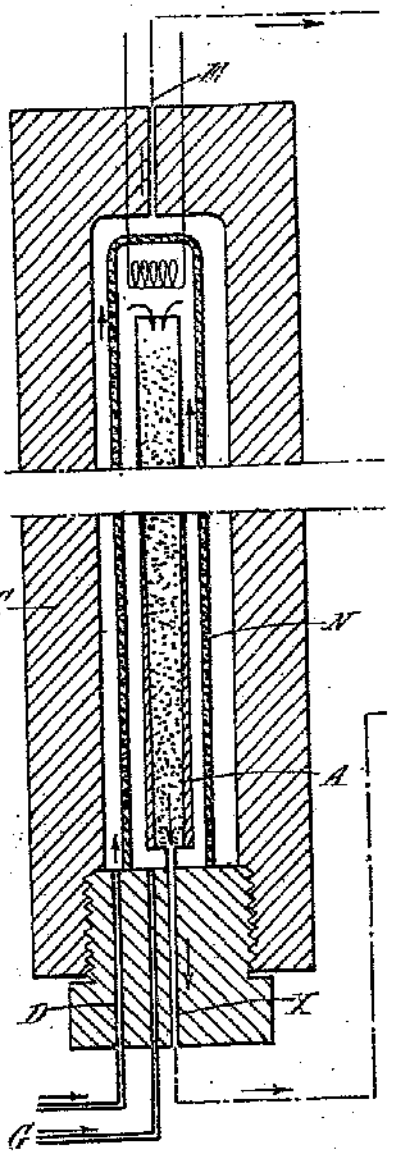


Fig. 2.



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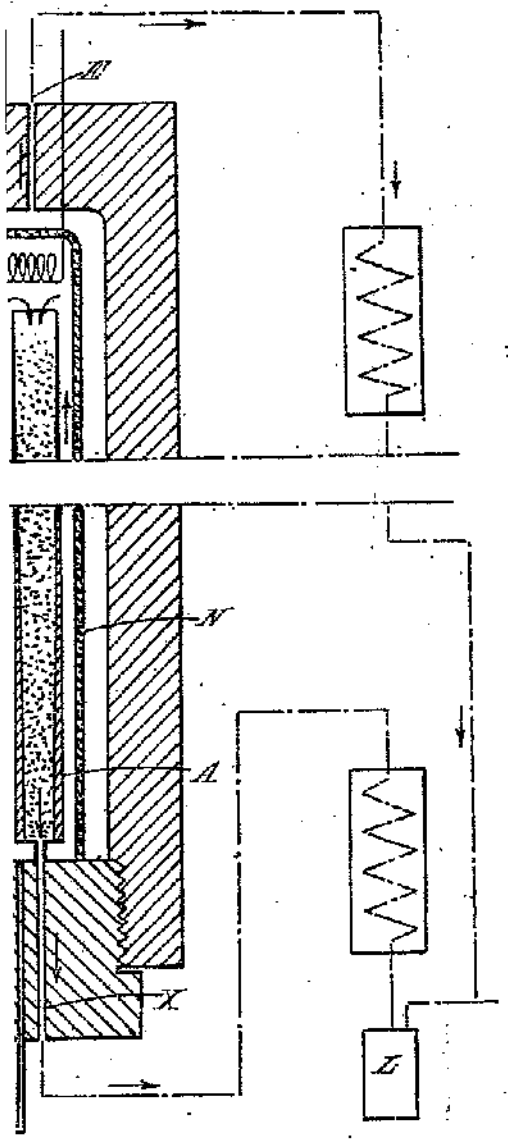
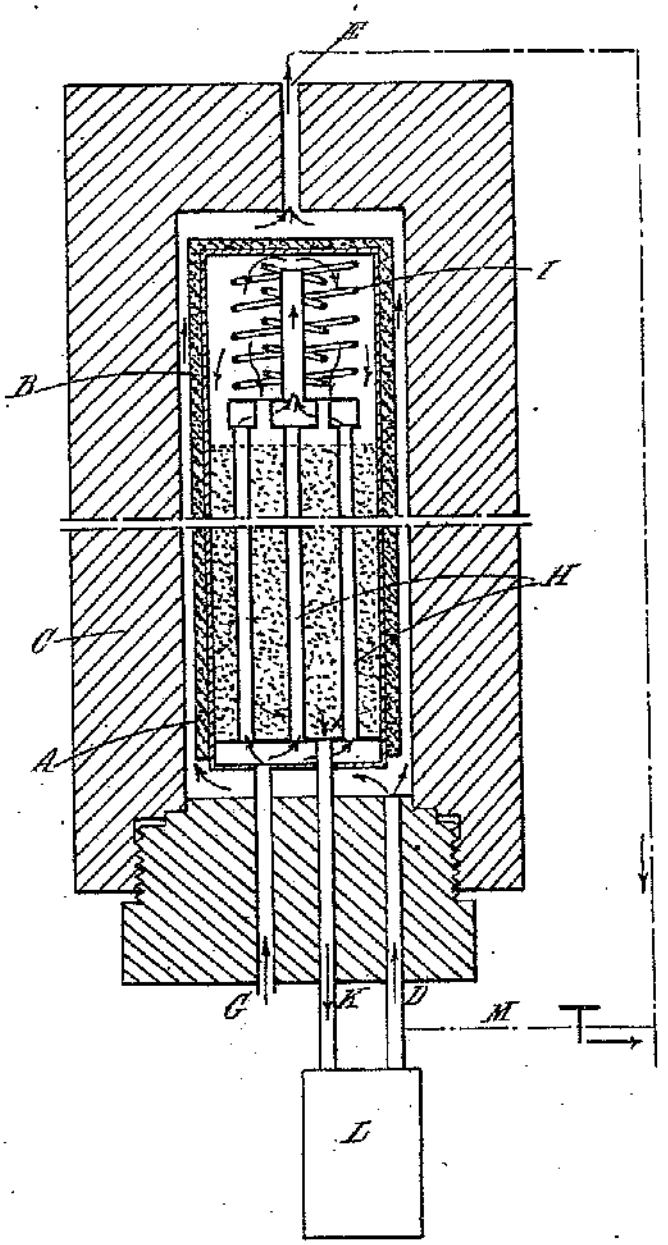


Fig. 3.



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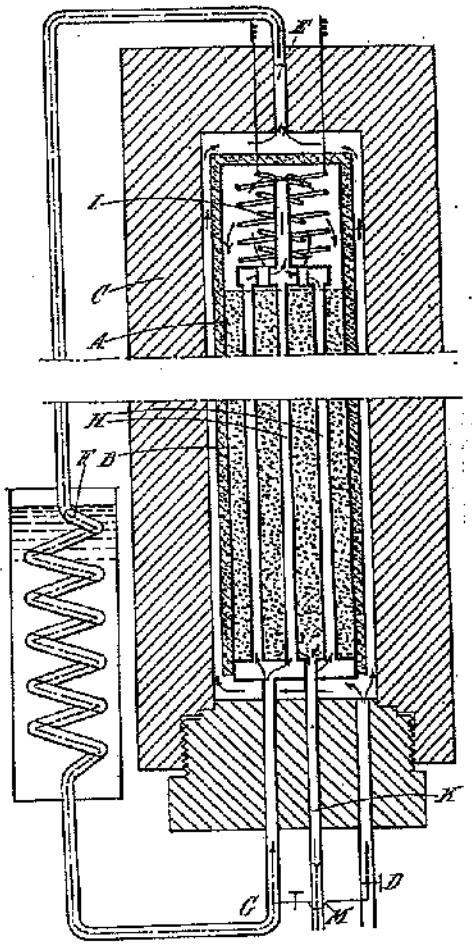


Fig. 2.

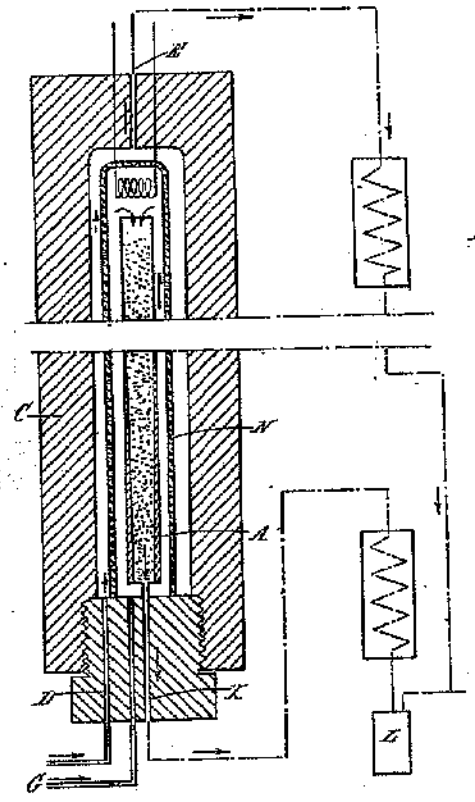
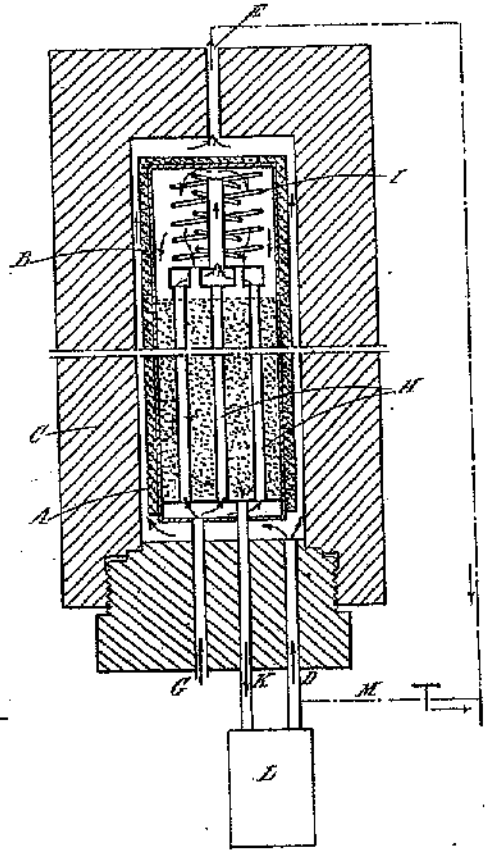


Fig. 3.



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