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COMPLETE SPECIFICATION.

3424

Improvements in or relating to Processes for Carrying Out Exothermic Catalytic 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 13, 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 wherein the gases prior to reaction circulate in indirect contact with the catalysing material during which they become progressively heated whilst any additional heat they may require is supplied from an external source.

It is known that in processes of this type, in order to obtain normal working, the surfaces through which the exchange of heat takes place should be adapted to the quantity of heat evolved in the tube containing the catalysing material per unit of time, and once the correct adjustment in this respect has been made for definite working conditions, these conditions can only be varied to a slight degree limited by the variations which may be obtained by varying the amount of heating from an external source or the temperature of the gases which are circulated in indirect contact with the catalysing material.

It has been recognised that under industrial conditions it was necessary to obtain a greater latitude in the operation of the catalysis tubes, and the present invention has for its object to provide a means which will permit of ensuring at all times the normal working of catalysis tubes in which there may occur important variations in the quantity of heat evolved by the reaction per unit of time, such variations taking place during the working and arising from variations in the concentration of certain reacting substances in the gases subjected to the process, variations in the quantity of the gases treated, variations in pressure etc.

Such means is afforded according to the present invention by providing the receptacle or tube containing the catalysing material with two or more distinct surfaces or groups of surfaces for the transmission of heat, circulating the gases which are to react over the surfaces or groups of surfaces, and effecting during the working a variation in the transmission of heat through the said surfaces or groups of surfaces by varying the quantities of gases which respectively circulate, either before they react or during the time they react, over the different surfaces or groups of surfaces.

Whilst the total superficial area of the whole of the surfaces or groups of surfaces and their heat conductivity will be determined by the necessity of effecting a sufficiently efficacious exchange of heat, the relative importance of these different surfaces or groups of surfaces and of these different heat conductivities will be conditioned by the extent of the zone of regulation that it is desired to obtain.

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 in which:—

Figures 1 and 2 illustrate diagrammatically by way of example two constructions of apparatus for carrying out the present invention, and

Figure 3 is a plan view of the receptacle containing the catalysing material shown in Figure 2.

In the apparatus shown in Figure 1 the receptacle or tube A containing the catalysing material is arranged as usual within a receptacle B, which if necessary is made pressure-resisting; the tube A comprises an outer surface C as well as an inner nest of tubes D. The surfaces C and of the nest of tubes D may be partially heat insulated. The gases which are to react, which may if necessary be previously heated, enter the apparatus through the pipe G and can be separated into two parts, the one of which passes through the pipe H provided with the

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regulating valve S and enters the interior of the receptacle B and circulates round the surface C, whilst the other passes through the pipe F provided with the regulating valve R and enters a chamber E which distributes it into the nest of tubes D. These two gaseous streams again mix at the top of the receptacle B and then pass over the catalysing material where the reaction takes place, finally leaving the apparatus through the pipe K. By way of example there may be considered as a particular case for the application of the apparatus shown in Figure 1 that of a variation in the amount of the gaseous substances which are to react. If the quantity of the gases treated is low, the valve R is closed and there is only utilised the exterior surface C of the tube A containing the catalysing material. If the quantity of the gases treated increases, then the valve R is opened and the distribution of the gases passing through the valves R and S is regulated in such manner that the two streams of gas, after having circulated respectively around the surface C and through the nest of tubes D and then become mixed, are at a temperature suitable for the reaction.

In the apparatus shown in Figures 2 and 3 the catalysing material is disposed in a central tube L the surface of which may be partially heat insulated and in a nest of tubes M exterior to said central tube. The central tube L is connected with a gas outlet pipe F provided with a valve R and the nest of tubes M is connected through an intervening chamber E with an outlet pipe H provided with a valve S. The pipes F and H are finally connected by one common outlet pipe K. The gases which are to react enter the apparatus through the pipe G and by means of baffle plates suitably arranged are made to circulate simultaneously in contact with the surface of the nest of tubes M and the surface of the central tube L. Having reached the end of their course the gases divide into two parts regulated by means of the valves R and S, the one passing over the catalysing material in the tube L, and the other over the catalysing material contained in the tubes of the nest M. After reaction the gases leave the apparatus through the pipes H and F and become reunited in the pipe K. By way of example there may be considered as a particular case for the application of the apparatus shown in Figure 2 that of a variation of the concentration of certain reacting substances in the gases which may occur in the conversion into methane of carbon monoxide present in the gaseous mixture $N+3H$, it being assumed that the surface of the nest of tubes M permits, owing to a larger surface area or a higher heat conductivity or to the simultaneous effect of both these factors, of an exchange of heat notably greater than that afforded by the surface of the central tube L. If the concentration of carbon monoxide is high, say 2 to 3 per cent. for example, then the valve S will be closed and the valve R will be opened; if, on the contrary, it is low, of the order of one thousandth for example, then the valve R will be closed and the valve S will be opened. For intermediate concentrations the valves R and S will be regulated in a suitable manner.

Instead of the single central tube L mentioned above, there may also be employed a group of tubes connected to the same outlet pipe F.

It will of course be understood that the process according to the present invention may be employed with a regulable externally or internally arranged electric heating arrangement, and that means may also be provided for varying the temperature at which the gases reach the apparatus; the object of such arrangements will of course be to still further increase the extent of the zone of regulation afforded by the process forming the subject of the present invention. For example if it be assumed that in the case of the apparatus shown in Figure 1 these heat exchanges take place more actively at the contact with the walls of the nest of tubes D than at the contact with the outer wall C, whilst in the case of the apparatus shown in Figure 2, as has been assumed in the example given above, the heat exchanges are effected more actively at the walls of the nest of tubes M than at those of the central tube L, the temperature of the gases in the pipe F in the case of the apparatus shown in Figure 1 and that of the gases in the pipe G in the case of the apparatus shown in Figure 2 can be altered by a partial exchange of heat with the gases which leave the apparatus through the pipes K and H respectively.

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 gaseous reactions of the kind in which the gases which are to react are heated prior to reaction by circulating them in indirect contact with the catalysing material, which process consists in circulating the said gases, immediately before they are brought into contact with the catalysing material, simultaneously over two or more distinct surfaces or groups of surfaces enclosing the space

occupied by the catalysing material and through which the heat exchange takes place, the transmission of heat through the said surfaces being regulated during the operation of the process by regulating the quantities of gases which respectively circulate, before or while they react over the different surfaces or groups of surfaces.

2. An apparatus for carrying out the process according to Claim 1, in which the receptacle containing the catalysing material wherein the reaction takes place is provided with two or more distinct surfaces or groups of surfaces, a regulable inlet corresponding to each surface or group of surfaces being provided for the admission of the gases prior to reaction on to each surface or group of surfaces through their respective inlets.

3. An apparatus for carrying out the process according to Claim 1, in which the receptacle containing the catalysing material wherein the reaction takes place is provided with two or more distinct surfaces or groups of surfaces, a regulable outlet corresponding to each surface or group of surfaces being provided for the discharge of the gases after reaction.

4. An apparatus as in Claim 2, in which the reaction receptacle containing the catalysing material is provided with an external surface on to which the gases prior to reaction are admitted through a regulable inlet, and with a group of inner surfaces on to which the gases prior to reaction are admitted through another regulable inlet.

5. An apparatus as in Claim 3, in which one or more centrally arranged reaction receptacles are connected to a regulable outlet for the discharge of the gases after reaction and one or more reaction receptacles are arranged round the centrally disposed reaction receptacle or receptacles and connected to another regulable outlet for the discharge of the gases after reaction.

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

7. A process as in Claim 1 or 6 for use in combining the carbon monoxide contained as an impurity in a mixture of nitrogen and hydrogen with part of said hydrogen, prior to subjecting said mixture to ammonia synthesis.

8. An apparatus for carrying out the process as claimed in Claim 1, 6 or 7, having its parts constructed, arranged and adapted to operate substantially as described with reference to one or other of the constructions illustrated in the accompanying drawings.

Dated this 18th day of July, 1927.

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