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COMPLETE SPECIFICATION

Improvements relating to the Performance of Gaseous Exothermic Catalytic Reactions

We, MONTECATINI SOCIETA GENERALE PER L'INDUSTRIA MINERARIA E CHIMICA, a Company incorporated under the laws of Italy, of 18, Via Albania, Milan, Italy, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement :—

The present invention relates to improvements in the method of performing gaseous exothermic catalytic reactions taking place at high temperature and under pressure, and to apparatus for carrying out such improved methods. The invention has for an object to enable the removal of the heat generated by such reactions, thereby obtaining a more advantageous gradient of temperature in the reaction vessel, and for a further object the utilisation of said heat for the production of steam.

The invention is applicable more specifically to methods of synthesis for example of ammonia or methanol.

The quantity of heat generated in the formation of one Kg. of ammonia from its elements at the temperature of 500° C. is approximately 750 KCal. Therefore, as the reaction proceeds in contact with the catalyst, the temperature of the nitrogen-hydrogen mixture rises, and the chemical equilibrium is thereby unfavourably influenced. In order to obtain a high rate of formation of ammonia it is necessary to maintain the temperature of the catalyst at a suitable level.

In the synthesis of methanol from carbon monoxide and hydrogen, this requirement is even stricter than in the synthesis of ammonia. In this case abnormally high temperatures not only have an unfavourable influence on the rate of formation of methanol, according to the equilibrium laws, but they also have the effect of promoting the formation of undesired products such as carbon dioxide and methane, and of reducing the efficiency of the catalyst.

It is known in apparatus for carrying out gaseous exothermic catalytic reactions such as the production or conversion of hydrocarbons to provide tubular cooling water systems distributed throughout a catalytic mass.

The invention has for an object to provide a method of and means for enabling reactions of the aforesaid kind and particularly the synthesis of ammonia or methanol, to be carried out under optimum conditions of yield and heat balance.

Accordingly the invention provides the method of performing gaseous exothermic reactions over a catalyst, and particularly the synthesis of ammonia or methanol, at high temperature and under pressure in which the temperature is controlled by removing heat of reaction by circulating water through tubes positioned in the interior of the catalyst chamber, characterised in that heat is removed by evaporation at a plurality of zones, spaced between successively deeper catalyst layers in the direction of gas flow, of portions of a quantity of water in a common closed circuit, the quantities of water evaporated at each zone are independently controllable to produce steam at high pressure, the produced steam is condensed externally by heat exchange with water to evaporate the latter and form steam at lower pressure, and the condensate is recirculated to said zones.

Further, the invention provides apparatus for performing gaseous exothermic catalytic reactions, and particularly the synthesis of ammonia or methanol, at high temperature and under pressure, comprising a catalyst chamber adapted to enclose the catalyst in a plurality of successively deeper layers or masses in the direction of gas flow, a plurality of heat exchangers interposed one between each adjacent two of said layers or masses and spaced at successively greater intervals in the direction of gas flow, a pump for supplying water in common to said heat exchangers, valve means independently controllable for regulating the supply of water to each heat exchanger for the purpose of promoting evaporation of the water therein

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to produce steam at high pressure, a common header for the steam from the heat exchangers, and a closed external circuit comprising said header, said pump and a boiler, in which boiler said steam is condensed in indirect heat exchange with a separate water feed to produce from the latter steam at lower pressure, the condensate from the boiler being returned to the intake of said pump.

Advantageously, the method may comprise, and the apparatus may include means to enable pre-heating of the reactant gases entering the catalyst chamber by heat exchange with issuing product gases, and alternatively or additionally pre-heating of the feed water for low pressure steam production.

The foregoing and other features of the invention will be more clearly apparent from the following description which is given by way of illustration and explanation, and without limitation, of a mode of carrying out the invention and apparatus therefore, for the synthesis of ammonia, with reference to the accompanying diagrammatic drawing in which:

Figure 1 is a section through a catalytic apparatus,

Figure 2 is a curve of temperature gradient throughout such apparatus during the performance of the method, and

Figure 3 is a curve showing percentage of ammonia present during performance of the method.

Referring to Figure 1 of the drawings, the apparatus in the form of a converter includes at its lower end a branch 13 for the entry of a gaseous mixture $N_2 + 3H_2$ under pressure. The gases thus admitted circulate through a heat exchanger 7, wherein they are pre-heated by issuing product gases as will be described and then ascend in an outer jacket between the inner catalyst chamber and the outer shell of the converter. The gases then pass downwardly through the catalyst chamber which encloses a plurality of layers or masses 1–6 of catalyst, thereafter issuing through the tubes of the heat exchanger 7. Interposed between each adjacent two of the catalyst layers or masses are a plurality of heat exchangers 17, each fabricated in any suitable form, for example as a spiral coil, the said heat exchangers being provided individually with inlet control valves 9 associated with a common header and a cooling water supply pump 10, and with outlet branches 12 associated with a common outlet header.

By virtue of pre-heating in the heat exchanger 7 and the outer jacket of the converter the gaseous mixture arrives at the first layer 1 of catalyst at a temperature sufficiently high for the reaction to start, that is, approximately 400° C. In this catalyst layer by reason of the exothermic reaction the temperature of the gases then rises quickly, exceeding 500° C. as indicated in the curve, Figure 2. Below the catalyst layer 1 the gases are then cooled by the subjacent coil 17, supplied with a regulated flow of water. Similar increases of temperature occur in each of the succeeding catalyst layers 2–6, with intervening decreases effected by the passage of the gases across the heat exchangers immediately below said layers. In order to obtain optimum yield, the temperature of the gases must be progressively decreased with increase of the percentage of ammonia present. As indicated in Figure 3, the reaction takes place rapidly in catalyst layer 1, and progressively less rapidly in the successive layers, and the heat exchangers are therefore interposed between catalyst layers or masses of progressively greater depth or thickness in the direction of gas flow as indicated in Figure 1.

The quantities of water supplied to the coils 17 successively traversed by the gases are individually regulated by means of the valves 9 so as to achieve the aforesaid desideratum of progressive overall decrease of temperature as indicated in Figure 2.

Further, said quantities of water may be regulated so as to allow evaporation within the coils with advantageous absorbtion of large quantities of heat from the gases, and with the production of high pressure steam.

The high pressure steam issuing by way of the branches 12 is passed to a boiler 11, wherein it is condensed and thereafter returned to the coils 17 in the converter, by means of the pump 10, in a closed circuit, thereby avoiding the deposition of salts and impurities from the water in said coils.

An additional amount of heat can be recovered by pre-heating feed water for the boiler 11 in a pre-heater 8, in heat exchange with product gases issuing from the catalyst chamber, which gases have already been partly cooled in the heat exchanger 7 and are further cooled in the pre-heater 8 for issuing from the apparatus by way of branch 16. The feed water is supplied to pre-heater 8 by a branch 14 and steam is produced at branch 15 of the boiler 11.

What we claim is:—

1. The method of performing gaseous exothermic reactions over a catalyst, and particularly the synthesis of ammonia or methanol, at high temperature and under pressure, in which the temperature is controlled by removing heat of reaction by circulating water through tubes positioned in the interior of the catalyst chamber, characterised in that heat is removed by evaporation at a plurality of zones, spaced between successively deeper catalyst layers in the

direction of gas flow, of portions of a quantity of water in a common closed circuit, the quantities of water evaporated at each zone are independently controllable to produce steam at high pressure, the produced steam is condensed externally by heat exchange with water to evaporate the latter and form steam at lower pressure, and the condensate is recirculated to said zones.

2. The method according to claim 1, wherein the reactant gases entering the catalyst chamber are pre-heated by heat exchange with issuing product gases.

3. The method according to claim 1, wherein feed water for the production of steam at lower pressure by the external evaporation is pre-heated by heat exchange with product gases issuing from the catalyst chamber.

4. The method according to claim 1, wherein feed water for the production of steam at lower pressure by the external evaporation is pre-heated by heat exchange with product gases issuing from the catalyst chamber, said product gases having been partly cooled by preceding heat exchange with reactant gases entering said chamber.

5. Apparatus for performing gaseous exothermic catalytic reactions, and particularly the synthesis of ammonia or methanol, at high temperature and under pressure, comprising a catalyst chamber adapted to enclose the catalyst in a plurality of successively deeper layers or masses in the direction of gas flow, a plurality of heat exchangers interposed one between each adjacent two of said layers or masses and spaced at successively greater intervals in the direction of gas flow, a pump for supplying water in common to said heat exchangers, valve means independently controllable for regulating the supply of water to each heat exchanger for the purpose of promoting evaporation of the water therein to produce steam at high pressure, a common header for the steam from the heat exchangers, and a closed external circuit comprising said header, said pump and a boiler in which boiler said steam is condensed in indirect heat exchange with a separate water feed to produce from the latter steam at lower pressure, the condensate from the boiler being returned to the intake of said pump.

6. Apparatus according to claim 5, having a heat exchanger wherein reactant gases entering the catalyst chamber are pre-heated by product gases issuing from the catalyst mass.

7. Apparatus according to claim 6, having a jacket surrounding the catalyst chamber through which the pre-heated gases are passed prior to their entry into the catalyst chamber.

8. Apparatus as claimed in any of claims 5–7, having a pre-heater for boiler feed water by heat exchange with product gases issuing from the catalyst chamber.

9. Apparatus for performing gaseous exothermic catalytic reactions at high temperature and under pressure, constructed and adapted for operation substantially as hereinbefore described with reference to the accompanying drawing.

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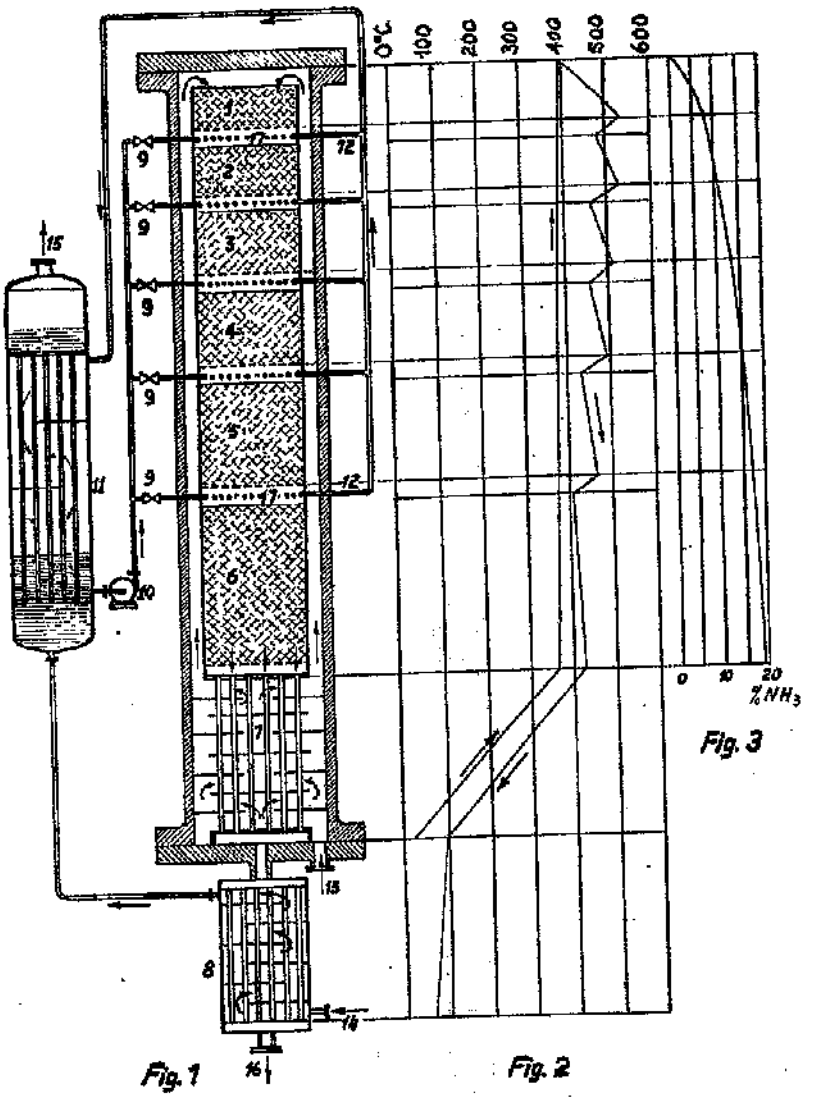

Fig. 1

Fig. 3

Fig. 2