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PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Process of and Apparatus for Performing Reactions in the Gaseous or Vapour Phase in the Presence of Catalysts

We, METALLGESELLSCHAFT AKTIENGESELLSCHAFT, a Corporation organised under the Laws of Germany, of 45, Bockenheimer Anlage, Frankfurt-on-the-Main, Germany, 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 a process of and apparatus for performing reactions in the gaseous or vapour phase in the presence of catalysts.

It is known that in the catalytic conversion of gaseous and vapourous reaction mixtures, such as in the catalytic oxidation of SO_2 in gases containing same, into SO_3, the course of the reaction is critically dependent on the conversion temperatures, and that it is of prime importance, during the conversion process that the contact mass should be maintained over the longest possible portion of the path of the gases, at the temperature assuring maximum conversion.

The attainment of just this latter object, however, encounters great difficulties which—for example in the catalytic oxidation of SO_2 to SO_3—increases considerably with increasing concentration of the substance to be converted in the gases under treatment, so that it becomes more and more difficult, with an increasing content of SO_2 to maintain the temperature of the contact apparatus within the permissible limits. See Waeser, "Handbuch der Schwefelsäurefabrikation" (Handbook of Sulphuric Acid Manufacture), vol. 3 (1930), bottom of page 1566.

In many cases, these difficulties have led to the practice of adapting the working conditions in the contact apparatus to suit a gas concentration that is the optimum for the apparatus concerned because, in each case, it was only with these definite concentrations that constant temperature conditions and satisfactory yields would be obtained.

Apart from measures of a purely constructional character, directed to obtaining the maximum utilisation of the heat liberated by exothermic conversion, attempts have been primarily devoted to ensure the admittedly important maximum constancy of temperature in the contact apparatus, by employing the heat liberated during the conversion for preheating the reaction gases prior to their entry into the contact mass.

Attempts have been made to achieve additional control of the temperature by mixing an unheated gas—for example a portion of the reaction gas—with the reaction gas (preheated in the aforesaid manner) at one or more points before introduction into the contact mass. According to a recent suggestion, the introduction of this cold auxiliary gas into the flow of preheated reaction gas, should be effected—in apparatus in which, prior to its entry into the contact mass (contained for example in tubes) the reaction gas is led, for the purpose of equalising the temperature, along the outside of the spaces charged with the contact mass and in the opposite direction to that traversed by the gas in said spaces—at a point that is somewhat to the rear of the main reaction zone, in relation to the direction of flow in the contact mass, i.e. at a point which is somewhat further removed from the point of entry of the gas into the contact mass than the main reaction zone.

In all these cases it has hitherto been believed that optimum working conditions could be attained by endeavouring with the aid of thermometrical instruments disposed, for checking the temperatures, at various points in relation to the contact mass, to attain or maintain the temperatures or temperature differences regarded as best by controlling the quantities of the various currents of gas supplied to the apparatus, by means of valves, dampers and the like located in the gas supply pipes. However, the results hitherto obtained in this manner are so unsatisfactory that it has so far been found impracticable to obtain satisfactory yields in operating, for example, with a gas containing appreciably more than 7%—for example about 9%—by volume of SO_2.

Now, whereas it has long been the practice to employ thermometrical instru-

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ments for accurately determining the temperatures in the contact mass the variation of the quantitative supply of the various gas currents introduced has, in general, been a matter of guess work in the control members, such as valves, throttles, dampers and the like.

Applicant's experiments have shown that such a method of procedure is extremely misleading and that one of the chief, if not the chief cause of the failures that have occurred for example in the attempts to subject gases containing over 7%—for example 9%—of SO_2 to catalytic oxidation into SO_3, is the impossibility of controlling the quantities of gases supplied, by mere guess-work methods.

It has also been ascertained that the effect resulting from each adjustment, by guess-work alone, of the aperture of a control member interposed in a gas supply pipe, is entirely beyond control, primarily because such adjustment also affects the quantities introduced through all the other pipes, not only on account of the conditions of static pressure, and the extensively uncontrollable internal resistance of the apparatus, but also in an entirely unforshadowable and unanticipated manner, on account of the dynamic influences set up between the various currents of gas.

The experiments have also shown that, in contrast to the foregoing, highly unexpectedly favourable results and increased yields are directly attainable when in accordance with the present invention the controlling of the quantities of the various gases—which may differ either in respect of their composition or temperature, and also in both respects—admitted into the contact apparatus is effected, not merely by guess-work, but accurately by means of metering instruments.

It has transpired that by proceeding in this manner and by successive systematic adusiments of the apparatus to different and accurately determined relative proportions of the amount of gas admitted at the various points, results can be obtained which were quite unachievable without the aid of the accurate measuring of the gas, by means of metering instruments, already long known in other spheres of application but not previously employed in connection with the present problem, so that according to the present invention, it is now also possible, in this manner, to treat gas containing 9% and over of SO_2 for the production of SO_3 with the best results.

Another extremely important advantage is the very considerable latitude secured by the present invention in the composition of the gaseous or vaporous mixtures treated, and also in the construction of the apparatus at disposal for the time being, since the hereindescribed procedure enables the optimum working conditions to be most rapidly ascertained and continuously maintained in the simplest manner with any convenient reaction gas, and in any apparatus—whether the contact mass be arranged in or between tubes or in rack or stage apparatus.

It is also possible, on the occurrence of a change in the composition of the reaction gases to be treated, to adapt the working conditions accordingly and directly to the changed circumstances, for example in accordance with the current results of an analytic check.

The devices for measuring the quantities of gases introduced may be of very different types, such as known apparatus. Merely by way of example, mention may be made of baffle rims, Venturi tubes and other metering devices based on the same or similar principles.

Although, in suitable cases, satisfactory results can even be obtained by incorporating the gas metering appliances, provided in accordance with the present invention in a single one only of the conduits supplying gas to the contact apparatus—for example when only a single auxiliary gas (which may be unheated) is employed in addition to the actual reaction gas, preheated in a heat exchanger in known manner—it will nevertheless be preferable in general to equip all the conduits supplying the various gases introduced with metering appliances.

It will generally be advisable to have the metering appliances always in operation and keep a constant check on the amount of gas traversing the conduit concerned. Satisfactory results may, however, also be obtained if—other working conditions remaining unaltered—the checking is undertaken only at certain (for example, automatically fixed) time intervals, in which event one and the same metering instrument can, for example, be used, if desired, for measuring the amounts of gas supplied to one or more contact apparatus, for example, in automatically controlled sequence.

By proceeding in the hereindescribed manner, the resulting substantially improved regulation and assured control of the course of the temperature and reaction has enabled improved yields and the maintenance of optimum conditions in protracted working, to be obtained in all cases. Above all, in the catalytic oxidising gases containing SO_2, it has also rendered the operations independent of the SO_2 content of the gas, and enabled conversions of 98% and more to be obtained continuously in one and the same apparatus (of the most diverse types) irrespective of whether the gases coming for treatment contain 4% or, for example, 9.5% or more of SO_2 and indeed also when the amount of gas is raised to 250-300% of the minimum load.

According to the invention it is possible even for example in employing additional oxygen, or gases enriched therewith, in the production of the reaction gases containing SO_2 and/or in treating the same in the contact apparatus, to operate with SO_2 concentrations far exceeding the above-mentioned limits.

A further advantage of the present process consists in that, owing to the improved utilisation of the catalysts, the amount of the latter required for furnishing the same effects can be reduced down to a fraction of that hitherto needed in given cases.

Apart from the production of sulphuric acid, the present process can also be applied to various other reactions between mixtures in the gaseous or vaporous phase and in the presence of catalysts, and also in particular, to conversions such as the oxidation or reduction of organic substances, in which case it affords very special advantages in view of the particular susceptibility of such reactions to temperature conditions.

In order more clearly to understand the invention, reference is made to the accompanying drawing which illustrates diagrammatically and by way of example a typical embodiment of plant for the production of sulphuric acid by the catalytic oxidation of sulphur dioxide in gases containing same.

In said drawing, 1 represents the cylindrical contact apparatus, 2 the main pipe supplying the reacting gas and opening into the lower part of the apparatus. 3 are tubes, open at both ends, carrying the contact mass and fitting tightly, at their lower ends, in a tube plate 4. By means of other tubes 5, which project, in the intermediate spaces between the tubes 3, from above as far as about one quarter of the length of the tubes 3 and are mounted on the top in the cover 6 of the contact apparatus, unheated gas containing sulphur dioxide—of the same composition as the gas admitted through 2, if desired—is introduced from the pipe 8. The gas entering through 2 flows upward in the first place, into the spaces between the tubes 3, and on the one hand, in so doing exerts a cooling action, through the walls of said tubes, on the contact mass contained in the latter, whilst, on the other hand, it becomes preheated during its ascent. Mingled with the unheated gas issuing from the tubes 5, it then enters, by way of the chamber 7, into the top of the contact tubes 3. After passing downward through these tubes, the gas reaches the collecting chamber 10, from which it leaves the apparatus through 11. 12 are screens, provided at the lower ends of the contact tubes 3 and supporting the contact mass.

By means of the relatively cool gas, admitted, by the tubes 5 into the spaces between the contact tubes at a short distance in the rear of the main reaction zone in the contact tubes, a powerful cooling of the contact mass is obtained, with the arrangement herein described, directly in the rear of the reaction zone, thus assuring a more uniform course of the temperature, and one that increases the yield, in the lower layers of the contact mass.

The exhaust gas from the reaction, issuing at 11, first passes through the preheater 20, to be then led through the pipe 22 to the absorption apparatus. In this heat exchanger it parts with a portion of its heat to the fresh gas, containing SO_2, arriving from the pipe 21 by way of the throttle and control device 17 and issuing from the preheater through the tube 9, from which it can be admitted, through the tube 13, into the contact apparatus at 2. Fresh gas from the pipe 21 can also be passed direct through the tube 23 and the throttle and control member 19, into the tube 13 and, in this way, admitted, together with the preheated fresh gas from 9, into the contact apparatus at 2. 18 is another throttle and control member for regulating the amount of unheated fresh gas passed through the pipe 8 and tube 5 into the contact apparatus. The throttle and control members employed may consist, in known manner, of valves, throttles and the like.

14, 15 and 16 are gas-control apparatus for determining and controlling the quantities of gas supplied through 21 and also through the pipes 13 and 8.

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 performing reactions in the gaseous or vaporus phase in the presence of catalysts, for example in the production of sulphuric acid by the so-called contact process, with the simultaneous introduction of different currents of gas into the contact apparatus, in which the amount of at least one of the

gases introduced per unit of time is continuously or periodically controlled by the aid of gas metering instruments, in accordance with the active constituents present in the reaction gas or gases.

2. Process as set forth in Claim 1, in which, in the case of variations in the content of active constituents—such as SO_2—in the reaction gas or gases, said content is continuously or periodically determined and the quantity of the gases admitted is regulated accordingly.

3. The process for performing reactions in the gaseous or vapour phase, substantially as described.

4. Apparatus for carrying out the process set forth in the preceding claims, in which a gas metering instrument for the continuous or periodic control of the gas supplied in unit time, is provided in at least one of the feed pipes for the separately introduced gases.

5. The apparatus for performing reactions in the gaseous or vapour phase, constructed, arranged and adapted to operate substantially as described, with reference to the accompanying drawing.

Dated this 5th day of September, 1934.

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19, Southampton Buildings,
Chancery Lane, London, W.C.2.

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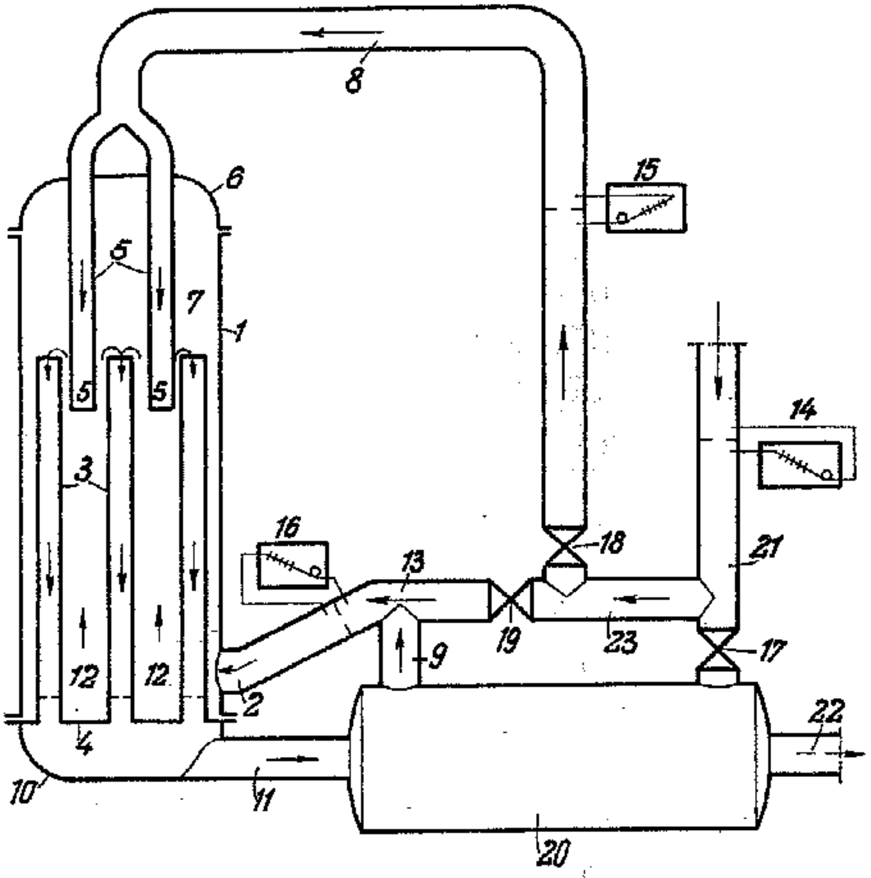
[This Drawing is a reproduction of the Original on a reduced scale.]



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