W. M. GROSVENOR.

METHOD OF CONDUCTING CATALYTIC REACTIONS.

APPLICATION FILED JAN. 20, 1908.

1,036,609. **Patented Aug. 27, 1912.**

3 SHEETS—SHEET 1.

Fig. 1,

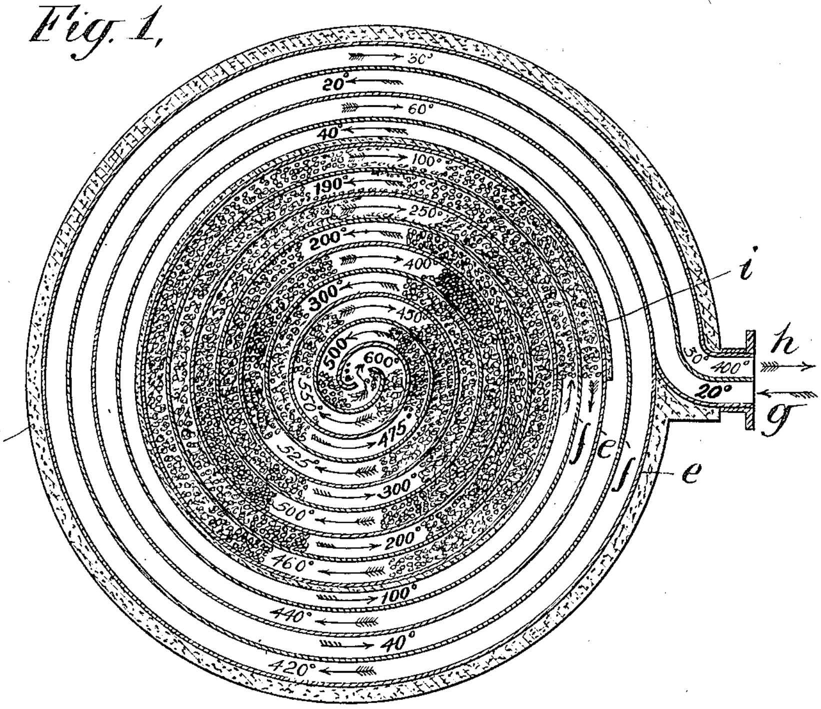

Fig. 2,

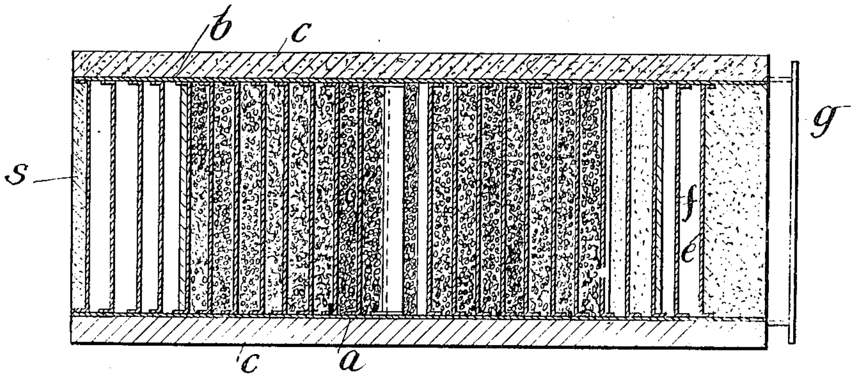

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Fig. 3.

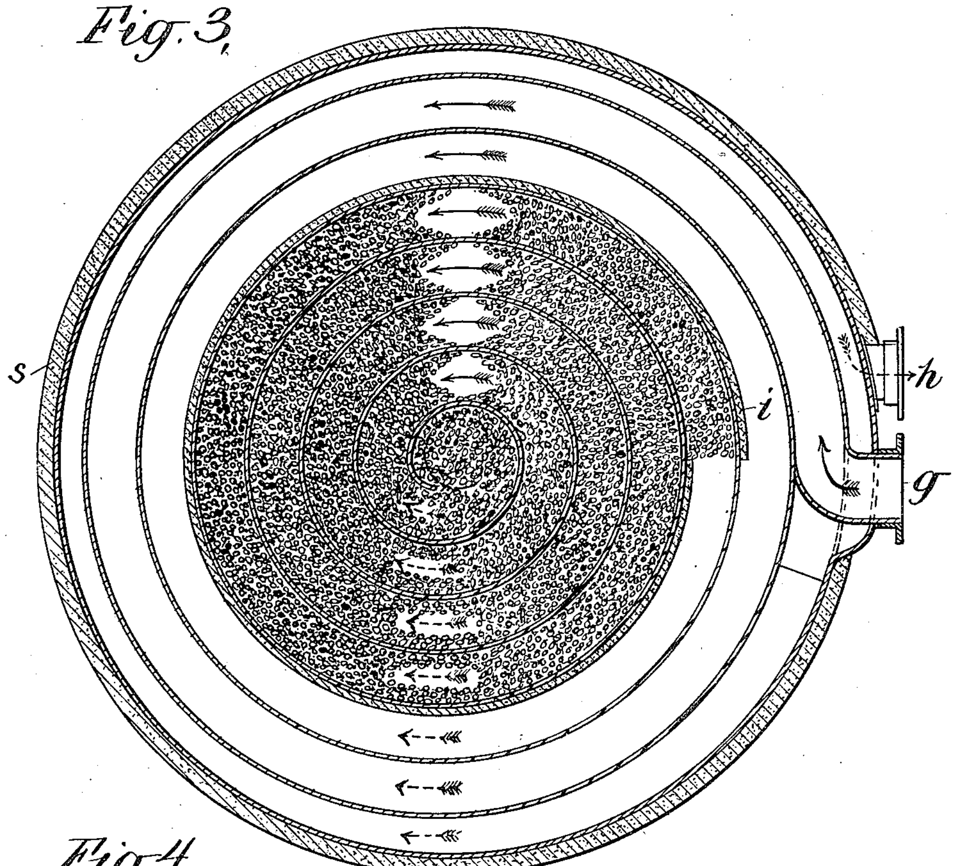
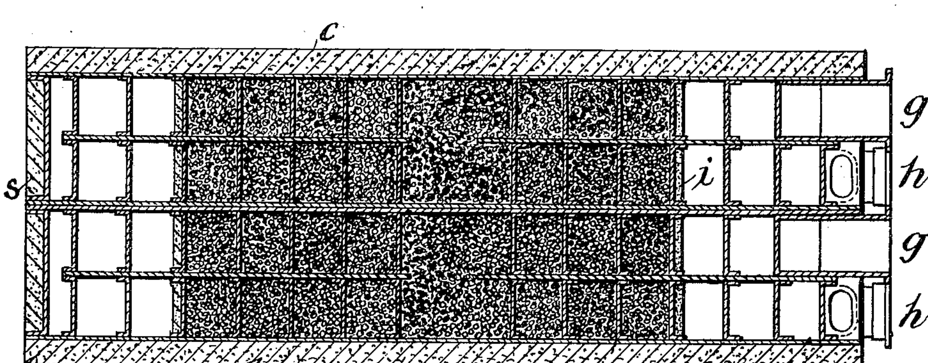


Fig. 4.



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3 SHEETS—SHEET 3.

Fig. 5,

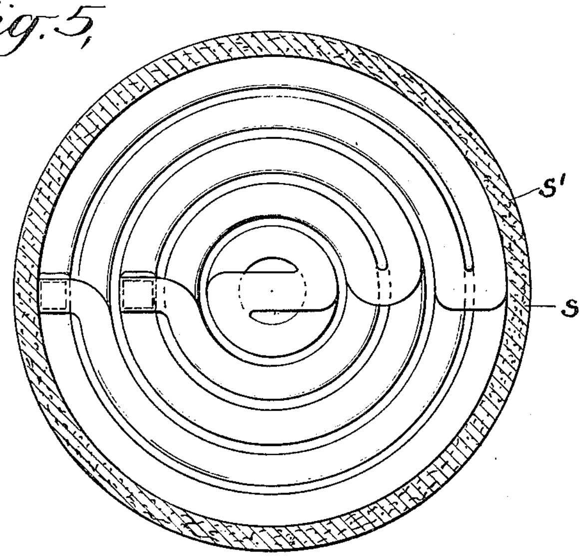
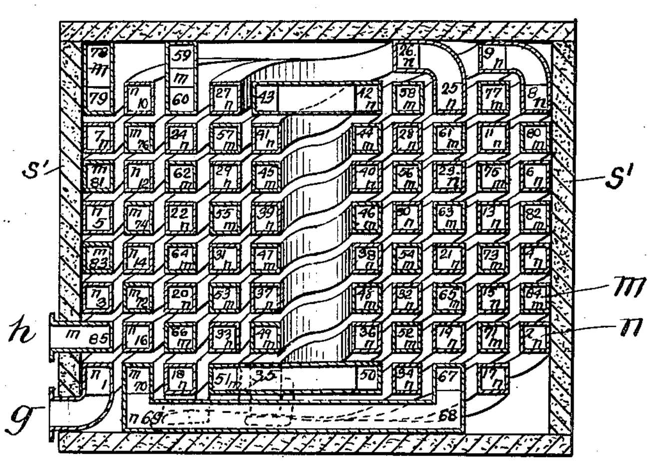


Fig. 6,



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UNITED STATES PATENT OFFICE.

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METHOD OF CONDUCTING CATALYTIC REACTIONS.

1,036,609. Specification of Letters Patent. Patented Aug. 27, 1912.

Application filed January 20, 1908. Serial No. 411,725.

To all whom it may concern:

Be it known that I, WILLIAM M. GROSVENOR, a citizen of the United States, residing at New York, in the county of New York and State of New York, have invented certain new and useful Improvements in Methods of Conducting Catalytic Reactions; and I do hereby declare the following to be a full, clear, and exact description of the invention, such as will enable others skilled in the art to which it appertains to make and use the same.

My invention relates to the production of catalytic reactions by means of contact materials as, for instance, in the conversion of a mixture containing SO_2 and O_2 into SO_3, by means of platinized substances, such as platinized asbestos, or other like catalytic material.

In carrying out my invention, I first raise the body of catalytic material employed in the converting apparatus to a temperature appropriate to the conversion required, and I thereafter maintain the necessary temperature of reaction within the apparatus by the heat developed by the reaction of fresh quantities of the gases to be converted, supplied in a relatively cool condition to the contact mass, the temperature of the inflowing gases being far below the reacting temperature thereof. This initial heating of the catalytic material may be accomplished in any convenient manner, as, for example, by means of a hot blast of air (or any other gas that will not "poison" the catalytic material) blown through the apparatus in any well-known way, followed by the action of the cool gases to be converted, which remove the heat from the outlying portions of the contact mass concentrating it toward the center. At the same time, the temperature within the contact mass itself is maintained at the desired elevation, by means of heat, transmitted from the outgoing converted gases to the initial portion of the contact mass through which the gases to be converted are entering, and the entire contact mass is so jacketed or insulated against radiation of heat that, commercially speaking, practically all of the heat of reaction developed in the contact mass is available for utilization within the contact mass itself.

By the practice of my invention, it is made possible, in the manufacture of sulfuric acid by the contact process, to supply cold gases varying anywhere from 3% SO_2 to 12% SO_2 directly to the contact chamber itself and to effect their complete commercial conversion therein, the apparatus automatically accommodating itself to the varying conditions; the size of the reaction zone changing and its position shifting, the temperature rising or falling, without personal control or supervision; thereby insuring at all times a practically complete reaction and the proper transfers of heat from one portion of the contact mass to another.

In the accompanying drawings, Figure 1 represents a plan view of one form of apparatus appropriate for the practice of my invention, the top cover of the apparatus being removed. Fig. 2 represents a vertical sectional view thereof. Figs. 3 and 4 represent like views, respectively, of a modified form of the apparatus. Figs. 5 and 6 represent like views, respectively, of a further modification.

Similar letters of reference indicate similar parts throughout the several views.

Referring to the drawings, and particularly to Figs. 1 and 2 thereof, *a* indicates the bottom plate of the apparatus and *b* the top plate thereof, said bottom and top plates being provided with a thick layer of insulating material *c*, so as to prevent loss of heat. Between the top and bottom plates is located a continuous passage involuted about its center and made up by spiral walls *e*, *f*, preferably of metal, said involuted passage communicating at the two outer ends thereof with an inlet *g* for the gases to be converted and an outlet *h* for the converted product, the involutions of the passage from its central portion to either end being substantially parallel and adjacent throughout. By this expedient, an involuted passage is provided leading spirally from the inlet *g* for the gases to be converted, to the center of the apparatus, and back again by a similar spiral to the final exit of the converted gases, and the first spiral is, throughout its whole extent, flanked on both sides by the involutions of the other spiral, which finally opens out into the exit port *h*, as described. The entire continuous passage may be filled with the contact material, which may be inserted therein in any suitable or convenient manner. A convenient method of supplying the contact material to the converter, would be, for instance, to mount the apparatus temporarily or permanently upon trunnions, and to feed the contact material into the involuted passage in the shape of small balls having a surface coating of the active mass; so, also, contact material of this type could be as readily removed from the apparatus by reversing its rotation upon the trunnions; or the apparatus may, if preferred, have doors in its top and bottom for the insertion and removal of the contact material.

Although, as I have just said, the entire continuous channel formed by the two in-going and out-going portions of the involute passage may be filled with contact material, when employing moderately weak gases, it is possible to economize the contact material by filling only the central zone of the apparatus therewith. In this case, however, I find it desirable to make the dividing walls between the zone containing the contact material and the outer zone of the contact chamber, of insulating material, as indicated at *i* in Fig. 1, the purpose of this insulating material being to prevent the cooling of the contact mass by the outlying portion of the chamber containing the entering gases, and preserving its heat, so that such heat may be utilized to the full extent practicable in maintaining the temperature of the inlet portions of the contact mass.

It will be apparent from what has been hereinbefore set forth, that the cold gases, introduced through the inlet *g* pass through the intake portion of the involuted passage to the axial center of the apparatus, side by side throughout with the gases passing away from the center and that they are everywhere flanked by said outgoing gases and insulated above and below by the insulating jackets *c*, and that the gases passing away from the center of the involution finally, as shown, envelop the entire periphery of the apparatus, thereby inclosing all portions of the spiral passages and maintaining, as far as possible, the temperature of the contact mass within. As an additional means for preventing radiation, the outer periphery of the apparatus may be protected against radiation by an additional jacketing of insulating material, a portion of which is indicated at *s* in Figs. 1 and 2. With richer gases, however, this precaution is not required, although it will in no way prevent the efficient conversion of said gases, inasmuch as the incoming gases always find themselves inclosed, within the contact chamber between layers of gases passing away from the center of involution and having a higher temperature than the entering gases.

It will be noted that colder portions of the contact mass, which colder portions are first encountered by the entering gases, are located within the succeeding or hotter portions of said mass, and it will be further noted that the length of the passage through the contact mass and the involution of the contact mass are sufficient to allow the zone of maximum temperature to shift and broaden out in order to accommodate its action to the nature of the gases supplied. This is illustrated by the upper and lower sets of arrows and the figures attached thereto. The upper set of arrows indicates the conditions to be expected in converting a moderately weak gas, say from three to four per cent., by volume, of SO_2. In such case, the zone of active reaction is quite narrow and is confined well within the contact mass, occupying only four or five of the convolutions of the spirals, and the maximum temperature is reached at a point near the center of involution. The lower set of arrows, and the figures attached thereto indicate the conditions to be expected when a large stream of rich gas is supplied to the apparatus. In such case the reaction zone broadens very greatly and a practically neutral area appears in its center wherein reaction ceases substantially, the equilibrium of the reaction having been reached, as determined by the temperature of said maximum zone.

For convenience of illustration, I have indicated by heavy-line (black face) numbers the temperatures of the incoming gas, and by light-line numbers the temperatures of the out-going gas.

In the form of apparatus shown in Figs. 3 and 4, another arrangement of the involuted contact chamber is shown, wherein, instead of being in the same plane and interwoven with each other, the two spirals into which the passage is involuted are superposed, so that the currents of gas traverse them in the same direction, the one over the other. An advantage possessed by the arrangement shown in Figs. 3 and 4 and by that shown in Figs. 1 and 2 is that units of very large capacity can be built up in practically the same floor space upon identical foundations, and that the addition of each integer of the larger plant will improve the heat economy of the whole. Furthermore, each individual unit or integer of the assembled apparatus may be removed by suitable machinery and another inserted in its stead without interrupting the general course of the operation. In said Figs. 3 and 4, two units or integers are shown, protected above and below by the insulating jackets *c*. As before, the involuted passage is preferably made of metal walls, except where it is desired to confine the contact material to the inner portion only of the spiral, in which event said inner portion or zone is provided with an insulating wall *i*. The entering gases are admitted through the intake g and the converted product issues through the out-take h.

The most marked advantage possessed by the form of apparatus shown in Figs. 3 and 4 is that the hottest particles of gas rising to the upper surface of the lower portion of the involuted passage of each unit or integer are brought immediately opposite the colder portions on the lower surface of the upper portion of the involuted passage of said unit or integer. The entering gases passing through the inlets g pass spirally within the contact chamber and either enter the contact mass immediately, provided that it fills the entire converter, or finally enter the contact mass when such mass fills less than the entire chamber. Upon reaching the center of involution the gases pass downward and thence outward through the contact mass (and through the unoccupied portion of the lower part of the passage, if any unoccupied portion exists) until they reach the outer turn of the lower part of the passage, which is made sufficiently high to entirely surround the converter considered as a whole, including the upper portion, as shown in Fig. 4.

The broad underlying or characteristic principle of the invention resides in the circumstance that before the entering gas has reached the temperature of active conversion, it is introduced directly into the initial portion of the contact mass, in which it is gradually raised to the temperature of complete conversion, partly by local reaction, and partly, by heat communicated to said initial portion of the mass from that portion of the mass beyond the zone of maximum temperature. After traversing the zone of maximum temperature, which may even be a bright red heat (and which zone I do not attempt to restrict in extent, position or temperature), it passes on into farther portions of the mass, until it has again fallen below the temperature at which dissociation is possible; for the duration of the passage of the gas through the contact mass must allow extra contact material at both ends to permit of the shifting and expansion of the maximum zone.

It will be understood that whereas the arrangement of the involute passage is not restricted to the substantially circular contour illustrated in Figs. 1 to 4, but may be elliptical or even rectangular, without departing from the spirit of the invention, and that it is likewise feasible to vary the cross section of the individual turns of the involute passage within wide limits. In Figs. 5 and 6, for instance, I have shown the cross section of the involute passage as rectangular, and its general arrangement as in the form of a helical spiral, whose in-going convolutions n, (communicating with the inlet g for the gases to be converted) alternate both vertically and horizontally with its out-going convolutions m; which latter communicate with the outlet port h for the converted product. In Fig. 6, the course of the in-going gas is indicated by the n passages numbered from 1 to 42 and the course of the outgoing gases is indicated by the m passages numbered from 43 to 85 in said figure. It is, of course, obvious that in order to insulate the apparatus shown in Figs. 5 and 6 against radiation of heat, it may conveniently be provided with an external jacket of asbestos, or the like, which jacket may completely envelop the involuted passage at top and bottom and at the center and circumference, leaving passage, however, for the inlet and outlet ends of the passage. A portion of said insulating cover is indicated at s' in Figs. 5 and 6.

What I claim is:—

1. The method of conducting catalytic conversion of gases, which consists in bringing an intermediate portion of the mass of contact material to a temperature suitable for the desired conversion, admitting the incoming gases into the presence of an initial portion of the mass at an entrance temperature below that of their reaction, and raising them to the temperature of completed conversion, partly by heat communicated to the initial portion of the mass from that portion of the mass beyond the zone of maximum temperature, and partly by the heat generated by local reaction; substantially as described.

2. The method of conducting catalytic conversion of gases, which consists in bringing an intermediate portion of the mass of contact material to a temperature suitable for the desired conversion, admitting the incoming gases into the presence of an initial portion of the mass at an entrance temperature below that of their reaction, passing them on through an intermediate or central zone of maximum temperature of the mass, and finally through a succeeding egress zone of decreasing temperature of the mass until they have fallen below the temperature of dissociation, and, during the travel of the gases through the mass, effecting a transfer of heat from the egress zone of the mass to the initial portion thereof; substantially as described.

3. The method of conducting the catalytic conversion of gases, which consists in arranging the contact mass in an involute path, the return convolutions of which are adjacent to the incoming convolutions, bringing an intermediate portion thereof to a temperature suitable for the desired conversion, admitting the incoming gases at an initial portion of the involute and at an entrance temperature below that of their reaction, passing them through the central

zone of maximum temperature, and finally through an egress zone of gradually decreasing temperature of the involute until they have fallen below the temperature of dissociation and effecting a transfer of heat from the return to the incoming convolutions of the involute; substantially as described.

4. The method of conducting the catalytic conversion of gases, which consists in conducting the gases by a circuitous route into the center of the contact mass initially heated to the temperature of reaction and thence returning them through the mass by a circuitous route adjacent to their ingoing path; substantially as described.

5. The method of conducting the catalytic conversion of gases, which consists in conducting the gases by a circuitous route into the center of the contact mass and thence returning them through the mass by a circuitous route adjacent to their ingoing path, said gases being at a lower temperature than that of reaction on entering the mass, and at a temperature lower than that of dissociation on leaving it; substantially as described.

6. The method of conducting the catalytic conversion of gases, which consists in conducting the gases by a circuitous route into the center of the contact mass and thence returning them through the mass by a circuitous route adjacent to their ingoing path, said gases being at a lower temperature than that of reaction on entering the mass, and at a temperature lower than that of dissociation on leaving it, and likewise conducting the said gases before entering the contact mass in a path adjacent to the converted product issuing from the contact mass; substantially as described.

7. The method of conducting the catalytic conversion of gases, which consists in continuously introducing said gases, at an entrance temperature below reaction into the presence of a preliminary portion of heated contact material, and elevating their temperature on the way to the zone of maximum temperature required for their conversion and cooling them as they leave said maximum zone by transference of heat to the preliminary portion of the contact material from the egress portion thereof while retaining the gases in the presence of the contact mass substantially throughout the operation; substantially as described.

8. The method of conducting the catalytic conversion of gases, which consists in introducing the gases below their reaction temperature into heated contact material, allowing the conversion thus initiated and the transfer of heat from other portions of the contact mass to raise the temperature to the maximum that may be generated by the reaction and of so involving the path of the gases through the contact material that their point of removal therefrom shall be adjacent to the point of entrance of said gases and shall be cooler than the temperature of dissociation; substantially as described.

In testimony whereof I affix my signature, in presence of two witnesses.

WILLIAM M. GROSVENOR.

Witnesses:
JOHN C. PENNIE,
LAURA B. PENFIELD.