PATENT SPECIFICATION

Convention Date (United States): March 23, 1935. **471,930**

Application Date (in United Kingdom): March 13, 1936. No. 7685/36.

(Patent of Addition to No. 449,050: dated June 1, 1934.)

Complete Specification Accepted: Sept. 13, 1937.

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COMPLETE SPECIFICATION

Control of Chemical Reactions

We, HOUDRY PROCESS CORPORATION, of 19, Dover Green, Dover, Delaware, United States of America, a corporation duly organized and existing under the laws of the State of Delaware, United States of America, (assignees of EUGENE JULES HOUDRY) 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:—

The present addition has as object certain means for utilizing the invention described in the Specification of the principal patent No. 449,050 and in the Specifications of Applications for Patents of Addition, viz., Nos. 456,091, 5673/36, 470,976 and 5674/36 (Serial No. 470,977).

This invention relates to chemical reactions, both endothermic and exothermic, which involve the use of contact masses. The latter may be absorbent, adsorbent, mere spreading material, or enter chemically into the reaction. More particularly, the invention has to do with reactions which are strongly exothermic, such, for example, as the catalytic synthesis of ammonia, production of sulphur trioxide in the manufacture of sulphuric acid, the regeneration or reactivation by oxidation of contact masses contaminated by coky and tarry deposits after use in the treatment or conversion of mineral oils, etc. The invention involves both process and apparatus aspects.

One object of the invention is to control the temperature of the reaction in a uniform manner all through the contact mass so as to maintain the same within a predetermined temperature range. Another object is to remove the heat at a rapid rate and in large amount. Another object is to spread or disperse the reactant fluids all through the contact mass. Still another object is to provide suitable apparatus for realizing the above results.

The invention involves supplying the reactant fluids to the converter at a temperature or temperatures within or near the reaction range, so that the reaction will begin and be continued at practically the same rate without possibility of interruption, and passing a fluid in a plurality of streams through the mass in intimate but indirect heat exchange with the latter. By "indirect heat exchange" is meant an exchange effected through an intervening wall and across a gaseous space. The fluid may supply heat to the reaction if endothermic, and, when exothermic, it is utilized to remove the generated heat or any undesired or excess part of the same. By preference, distribution of reactants and removal of products is effected substantially uniformly all through the mass by two series of apertured elements embedded in the mass in symmetrical arrangement after the manner disclosed in our British Patent No. 414,779, dated May 14, 1932, and the independent heat exchange fluid may be circulated through the interior of the series of elements which remove the reaction products without interfering with such function, as disclosed in the Specification of Houdry Process Corporation, No. 449,050. Suitable heat conducting means are provided within the contact mass to effect rapid movement of heat between the mass and the circulated heat exchange medium, such means may be of the kind described in our prior Specification No. 430,174. Such means are preferably in the form of fins radiating from the elements of the outlet series. These fins control to a large extent the dispersion and movement of the reactant fluids and the reaction products as well as the movement of heat. In one modification of the invention, they even combine or cooperate to form elements of the other conduit series.

The apparatus of this invention and all embodiments thereof illustrated in the drawing, provide for conducting all or substantially all of the heat to be transferred to or from the channels or outlet conduits, which surround and are in spaced relation with the ducts for the passage of heat exchange fluid. This provision for transfer of all or substantially all of the heat to the conduits of

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one group only (or channel members equivalent thereto) so that no substantial or large amounts of heat are transferred from or to incoming reactants or regenerating medium, is an essential aspect of the apparatus of this invention. Also the construction which provides for circulating heat exchange fluid within and through the bed of catalyst or mass so that a free space is provided between the ducts carrying the same and the surrounding bed of mass, through which a fluid such as outgoing gaseous products of reaction may be passed or not, as desired, is also an important aspect of this invention.

In order to illustrate the invention and the manner of its use, reference will now be had to the accompanying drawings, in which:—

Fig. 1 is a vertical sectional view of a converter, the fins on one of the series of conduits being omitted for the sake of clearness;

Fig. 2 is a transverse sectional view on an enlarged scale of the interior of the converter substantially on the line 2—2 of Fig. 1;

Fig. 3 is a transverse sectional view similar to Fig. 2, omitting the contact mass and showing a modification in which the fins cooperate to form one of the series of conduits;

Fig. 4 is a greatly enlarged sectional view showing a sliding joint between the interengaging fins on two of the conduits shown in Fig. 3; and

Fig. 5 is a fragmentary vertical sectional view of the lower end of a converter such as shown in Fig. 1, illustrating the assembly of the modification shown in Figs. 3 and 4.

The converter shown in Fig. 1 is made up of an outer casing or shell 6 having a detachable cover 6*a*, the entire exterior being suitably protected against heat losses as by a thick layer of heat insulating material 7. The interior of the converter is divided by upper and lower partitions or flue sheets 8 and 9 into a large central reaction chamber A and upper and lower end or manifolding chambers B and C. Mounted upon the flue sheets so as to extend across reaction chamber A from opposite sides in parallelism and in symmetrical arrangement are two series of elements D and E adapted for uniform distribution of reactant fluids and removal of reaction products within and throughout a contact mass M substantially filling the reaction chamber A. In the form shown, perforated conduits D of one series are mounted in and extend somewhat above upper flue sheet 8 to effect communication between reaction chamber A and upper manifolding chamber B, while elements E of the other series are mounted in lower flue sheet 9 to effect communication between reaction chamber A and lower manifolding chamber C. Elements D are of the single conduit type, but are arranged to enclose heat exchange members as will be later described. Suitable means will be used to keep mass M from beneath elements D in order that they may be free to expand or lengthen under temperature changes but, for clearness, such means are not shown since they form no part of the present invention. Elements E are of the nested conduit type such as disclosed in the aforesaid British patent No. 414,779, and comprise an outer perforated member 10 having upper and lower closed ends contained wholly within reaction chamber A and an inner conduit 11 which extends through flue sheet 9 and to a point adjacent the farther closed end of conduit 10. Inner member 11 serves as the sole means of communication between the interior of member 10 and manifolding chamber C.

Into the upper open end of each of conduits D extends a heat exchange member in the form of an imperforate conduit 12 which encloses an inner supply conduit 13, the heat exchange member extending substantially the full length of member D in spaced telescoped relation. Each heat exchange member extends beyond its corresponding element D and conduits 12 connect with the outer member 14 of a nested manifold F, the inner or supply conduit 15 of which is connected to the inner supply conduits 13 of each of the heat exchange members. There may be a series of such nested manifolds F having a transverse supply main 16 connected to inner supply members 15 and an outlet main 17 connected with the outer members 14. The connections leading from mains 16 and 17, such as 17*a*, to the exterior of the converter are preferably arranged with a detachable joint such as 17*b* for convenient assembly and disassembly of the converter. The outer conduits 12 of the heat exchange members may have stops 18 thereon to serve as covers or closures for the upper open ends of elements D and to limit the projection of the heat exchange members within such elements. The details of the heat exchange system and the manifolding arrangement therefor is more fully disclosed in the copending British application of Houdry Process Corporation, No. 5673, filed 25th February, 1936 (Serial No. 470,976).

To insure the desired degree of heat transfer between elements D enclosing

the heat exchange members 12, 13 and contact mass M, heat conducting members or fins radiate from members D out into the mass, as clearly indicated in Fig. 2, to provide a multiplicity of heat conducting channels all through the mass, such fins being omitted from Fig. 1 for the sake of clearness. In the form shown, elements D are provided with four straight axially disposed fins 19 in symmetrical arrangement 90° apart. Alternating with fins 19 are other straight fins 20 having transverse outer ends 20*b* with diverging or flaring extensions or wings 20*c* so arranged as to substantially enclose, in a rather closely spaced manner, the plain or non-finned outer conduits 10 of elements D. As shown in Fig. 2, the fins form a uniform pattern of heat conducting elements radiating into mass M so as to conduct heat either toward or from elements D depending upon whether the fluid circulated in the heat exchange members 12, 13 is at a higher or lower temperature than that of the contact mass M. The form and arrangement of fins 20 is such as to make the exchange of heat very effective in those portions of the mass which immediately surround conduits 10 of elements E. By preference, the ports in conduits 10 are substantially opposite the center of outer portions 20*b*, 20*c* of fins 20, while the ports in elements D are intermediate the fins of the series 19 and 20 so that the movement of fluid between elements D and E is baffled and directed so as to be spread as uniformly as possible throughout contact mass M.

Figs. 3, 4 and 5 show a modification providing a simplified construction in which the fins radiating from elements D*a* are utilized to form the outer conduits of elements E*a* corresponding to conduits 10 of Figs. 1 and 2. As indicated in Fig. 3, the straight fins 19*a* and 20*a* are substantially the same as in Fig. 2, with the exception that they are slightly longer and the transverse outwardly flared portion 20*d* on fins 20*a* is circular, although this shape may be varied. Parts 20*d* cooperate to form the outer conduits of series E*a*. For this purpose, the ends of outwardly flared portions 20*d* may merely abut, or interfit, or interlock, as may be desired. One arrangement which is satisfactory is disclosed in enlarged detail in Fig. 4, where the reduced end 21 of one member 20*d* is shown as received within an elongate slot or recess formed by the forked end 22 of an adjacent part 20*d* on the next element D*a*. Any suitable porting arrangement may be provided. If parts 20*d* are in abutting relation, the ports may be provided by notches in one or both of the abutting edges. When interfitting or interlocking joints are provided, as shown in Figs. 3 and 4, ports may be provided in the web parts 20*d* preferably adjacent the joint and slanting in the direction of the same, as indicated at 23 in Fig. 4.

Inasmuch as the elements D of Fig. 1 and D*a* of Fig. 5 must not contact lower tube sheet 9 or 9*a* in order that there may be space for expansion, it is necessary that a somewhat different structural arrangement from that shown in Fig. 1 be provided for the members D*a* with interlocking fins disclosed in the modification in Figs. 3, 4 and 5. Such an arrangement is illustrated in Fig. 5, wherein inner conduits 11*a* are provided with or extend through conical members or bosses 25 mounted in or on lower tube sheets 9*a*, such bosses to telescope with a sliding fit within the lower ends of the interengaging fin parts 20*d* which make up the outer conduits of elements E*a*. This construction lends itself readily to assembly and disassembly of the converter. Inner conduits 11*a*, with bosses 25, are mounted on lower tube sheet 9 or 9*a*, while the elements D*a*, with interengaging fin members forming the outer conduits E*a*, will be mounted upon the upper tube sheet. The upper tube sheet, with the conduits D*a* assembled thereon, will then be lowered through the open top of the converter with the interengaging fin members D*a* telescoping over the inner conduits 11*a* and finally engaging conical bosses 25 when the upper tube sheet is in place. The heat exchange system will then be lowered through the open end of the converter with the members 12, 13 extending within the upper open ends of conduit series D or D*a*, and, after the connections have been made, cover 6*a* will thereafter be secured in place to complete the assembly of the converter.

When the reaction taking place within chamber A is strongly exothermic, the reactants will be admitted to manifolding chamber C at a temperature within the range of the reaction or slightly therebelow. The reactants pass up through inner tubes 11 or 11*a* without substantial heat exchange with contact mass M before passing into outer conduits 10 of Figs. 1 and 2, or E*a* of Figs. 3 and 5, to be uniformly distributed throughout the mass. The reaction products pass from the mass into the ports of the nearest elements D or D*a* and over the outer imperforate conduits 12 of the heat exchange elements therein, giving up some of the heat of the reaction to the latter. In the meantime, the fin series 19, 20 and 19*a*, 20*a* conduct heat from all

portions of the mass back to elements D or D*a*, whence it is transmitted largely by radiation and, to a less extent, by convection, to the heat exchange medium in members 12, 13. The circulated heat exchange medium may be a gas, but is preferably a liquid such as water, mercury, diphenyl, or the like, circulated under pressure. The disclosed arrangement of fins, in combination with the fluid heat exchange system, permits the removal of all heat in excess of that necessary to maintain the reaction. By adjusting the speed of the circulation and the entering temperature of the fluid, the temperature of the entire contact mass may be regulated to a nicety and transitions in temperature of the entire mass can be effected quickly and uniformly.

When the reaction is endothermic, the movement of reactants through the mass may be in the same direction as above described or in the opposite direction. If in the opposite direction, the reactants will be admitted to manifolding chamber B, be distributed by elements D or D*a* all through the mass, and leave through elements E or E*a*. The heat exchange medium can then be utilized in the same or in reverse flow to impart any desired degree of heat to the reactants. If the reactants pass, before entering the mass, over manifolds F and the connections thereto in manifolding chamber B and over outer imperforate conduits 12 of the heat exchange members which are disposed within elements D and D*a*, they will carry some of the heat from the heat exchange medium by convection into the mass. In either case, however, heat will be distributed or spread through the mass by radiation from conduits 12 to elements D and D*a* and thence conducted by fins 19, 20 and 19*a*, 20*a* uniformly all through the mass. Thus the requisite heat to hold the contact mass at the reaction temperature is supplied directly to the mass by the fins and the heat exchange system, and superheating of the reactants prior to admission to the converter with the attendant disadvantages can be greatly minimized, or entirely avoided when the reactants are charged into manifolding chamber C.

To reduce heat exchange by radiation between upper flue sheet 8 and manifolds F of the heat exchange system, suitable baffling means may be provided such as a removable baffle sheet 27 (Fig. 1). This and other baffling arrangements are disclosed and claimed in the copending British application of Houdry Process Corporation, No. 5674, filed February 25, 1936 (Serial No. 470,977).

The present invention differs in a number of respects from a companion invention disclosed in the British Specification of Houdry Process Corporation, No. 456,091, filed January 27, 1936. An important difference lies in supplying in the present case the reactants for an exothermic reaction at substantially the temperature of the reaction and in extracting the excess heat of the reaction by the independent heat exchange medium alone, and in apparatus for carrying out this process. The said copending application has fins on the conduits of both groups, arranged so as to withdraw a desired percentage of the heat of reaction by the incoming reactants or regenerating fluid and the balance by the heat exchange fluid; whereas the apparatus of the present application, as illustrated in Fig. 2 of the drawing, has fins only on the outlet conduits or channels within which, and out of heat conducting relationship therewith, an inner conduit is shown for passage of heat exchange medium. Figs. 3, 4 and 5 of the present invention also show a fin arrangement whereby all of the heat developed in the reaction may be conducted to the outlet conduits, the fins being so constructed that their outer extending portions form the outer duct or members of inlet conduits which are interspersed therewith.

The converter may be mounted, assembled and operated in any desired position, as vertically, horizontally or obliquely. To reduce damage to a contact mass from the movement of the conduits due to temperature changes, especially when the mass is in bits or molded pieces, the converter shown in Fig. 1 may be inverted. The invention is to be considered as covering the above and all other changes, modifications and adaptations within the scope of the appended claims.

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. Process of controlling chemical reactions in a converter having a large and deep mass of contact or catalytic material in the reaction chamber and a heat exchange fluid passed in a plurality of streams within and through the mass in indirect heat exchange with the same in accordance with the principal patent and said patents of addition, characterized by feeding reactant fluid to the mass at substantially the temperature of the reaction and utilizing heat conducting channels disposed in all parts of the mass

to disperse or collect heat for transfer by radiation to or from the heat exchange fluid.

2. Process according to claim 1, further characterized by directing a major part of the excess heat of an exothermic reaction by radiation from said channels and a minor part of such heat by convection to the stream of heat exchange fluid, the convection transfer being effected by passing the reaction products leaving the mass over the streams and between the latter and the mass.

3. Process according to claims 1 or 2, further characterized during the regeneration of a bed of catalytic material contaminated with combustible deposits by the use of an oxygen-containing gas or during an exothermic reaction in the presence of such a bed of material, by feeding reactant fluid or regenerating medium to the bed of material substantially at the desired reaction temperature and removing all or substantially all of the exothermic heat developed with a heat exchange or cooling fluid sent within and through the bed of material in a plurality of streams held in spaced relation with the bed to provide passages therearound, and circulating gaseous products through said passages or not, as desired, to effect heat transfer to said heat exchange fluid by radiation and convection or substantially completely by radiation.

4. Converter for effecting exothermic and endothermic reactions having a reaction chamber with a contact mass therein, one or more apertured inlet conduits for reactants and one or more apertured outlet conduits for reaction products embedded in the mass and means associated with the outlet conduits for passing an independent cooling or heating medium therethrough out of direct contact with the mass, according to the principal patent and said patents of addition and according to the preceding claims characterized by means for conducting heat between the said outlet conduit or conduits and all parts of the mass.

5. Converter according to claim 4, further characterized in that the heat conducting means are proportioned and arranged to conduct substantially all of the undesirable or excess heat of an exothermic reaction to the outlet conduits for absorption by the independent heat exchange medium.

6. Converter according to claim 4 and/or claim 5, further characterized in that the heat conducting means comprise fins radiating from the outlet conduits, the fins presenting a large surface area and at least certain of them partly surrounding the inlet conduits.

7. Converter according to claim 6, further characterized in that straight fins alternate with the larger fins having outwardly flaring portions utilized to baffle and disperse through the mass fluids issuing from the inlet conduits.

8. Converter according to claim 6, further characterized in that certain of the fins interconnect individual conduits of the inlet and outlet series.

9. Converter for chemical reactions providing a reaction chamber for containing a contact mass and perforated conduits extending into said chamber for admission or egress of fluid according to the principal patent and said patents of addition further characterized by means including elongate arcuate members with interengaging sides attached to said conduits to form other and independent conduits for egress or admission of fluid.

10. Converter according to claim 9, further characterized in that the conduits are symmetrically arranged in the reaction chamber with the elongate arcuate members attached to the first conduits by spacing webs or fins.

11. Converter according to claim 10, further characterized in that straight fins project from the first conduits in symmetrical alternation with those carrying the elongate arcuate members.

12. Converter according to the principal patent with disposition and arrangement of conduits, certain of which have fins, substantially as described and shown in Figs. 1 and 2.

13. Converter according to the principal patent with two series of interconnected conduits substantially as described and shown in Figs. 3 and 5.

14. Apertured conduits according to the principal patent with fins radiating therefrom and with flaring ends adapted to interengage to form other conduits substantially as described and shown in Figs. 3 and 4.

Dated the 13th day of March, 1936.
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London, W.C.2,
Chartered Patent Agents.

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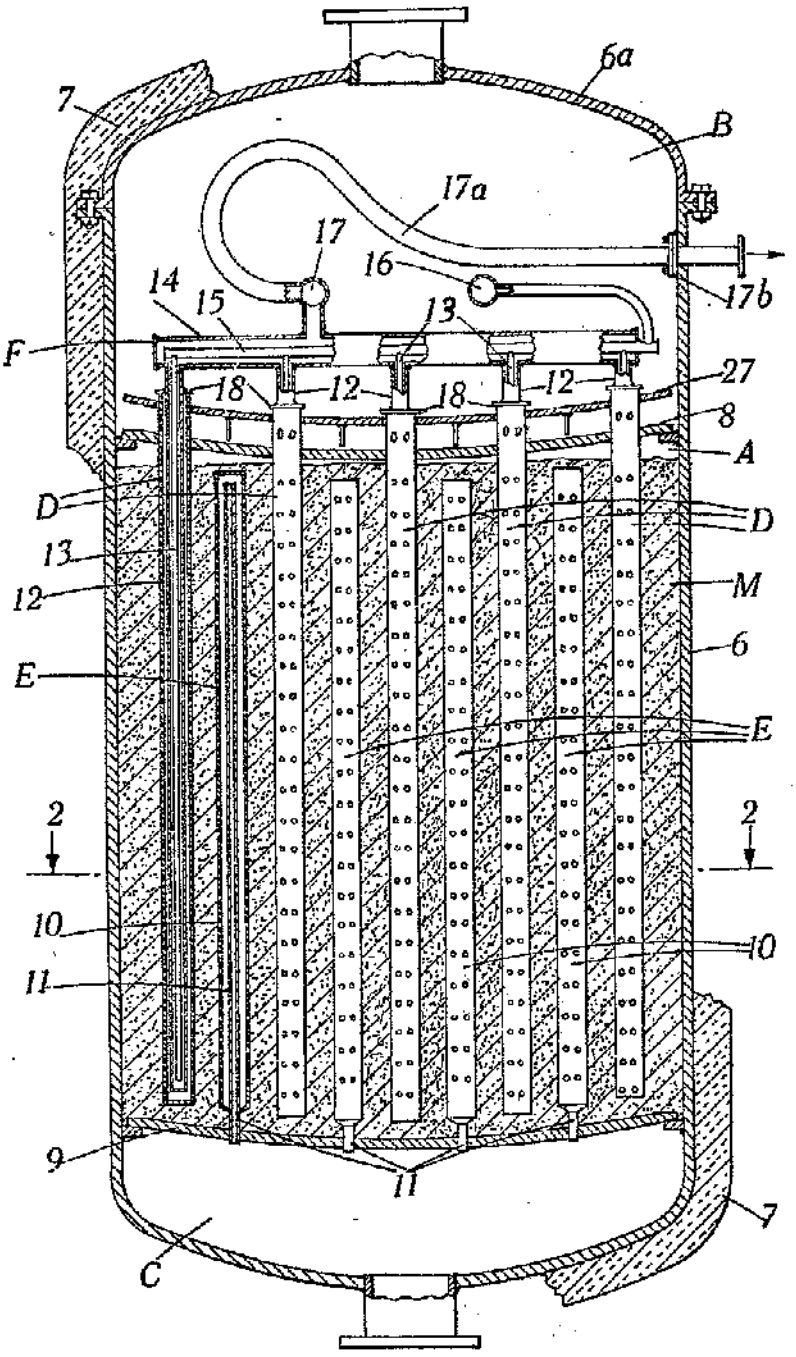
[This Drawing is a reproduction of the Original on a reduced scale.]

Fig. 1.



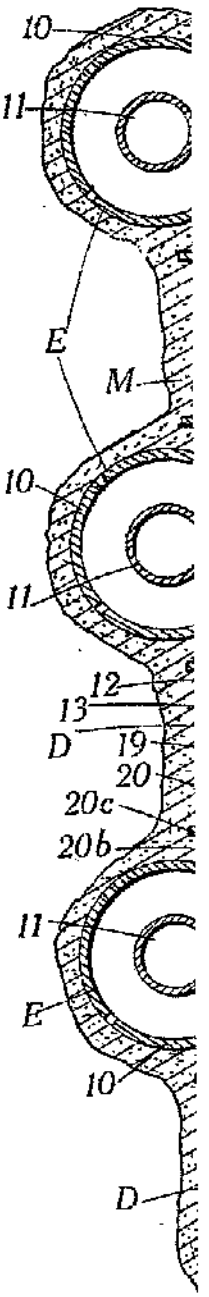
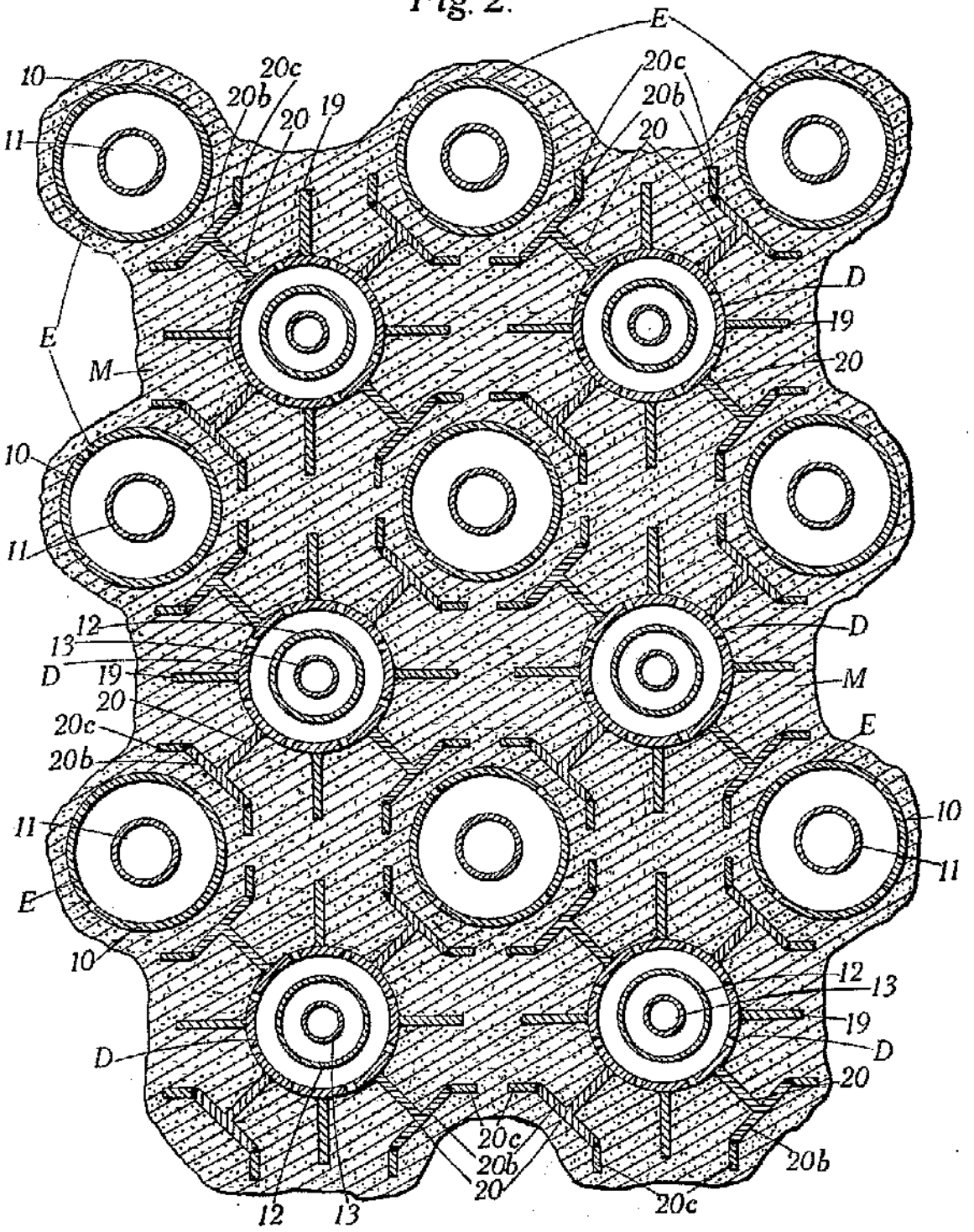


Fig. 2.



471,930 COMPLETE SPECIFICATION

3 SHEETS

SHEET 1

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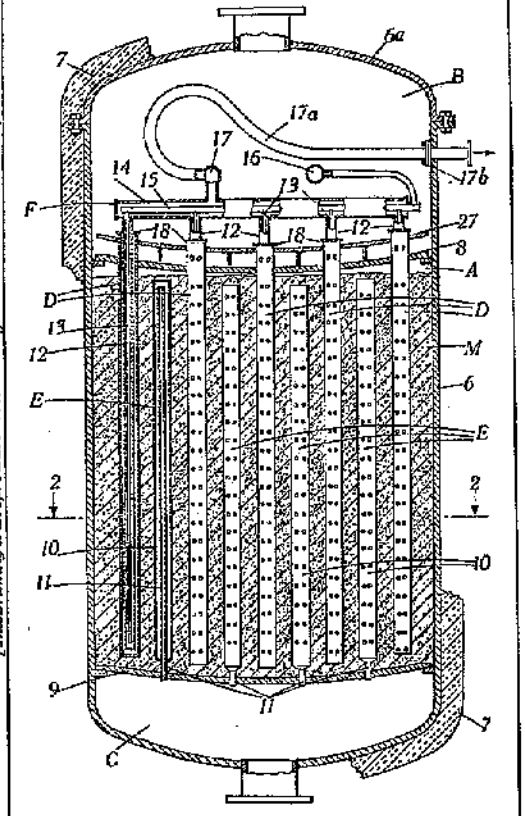
Fig. 1.



SHEET 2

Fig. 2.

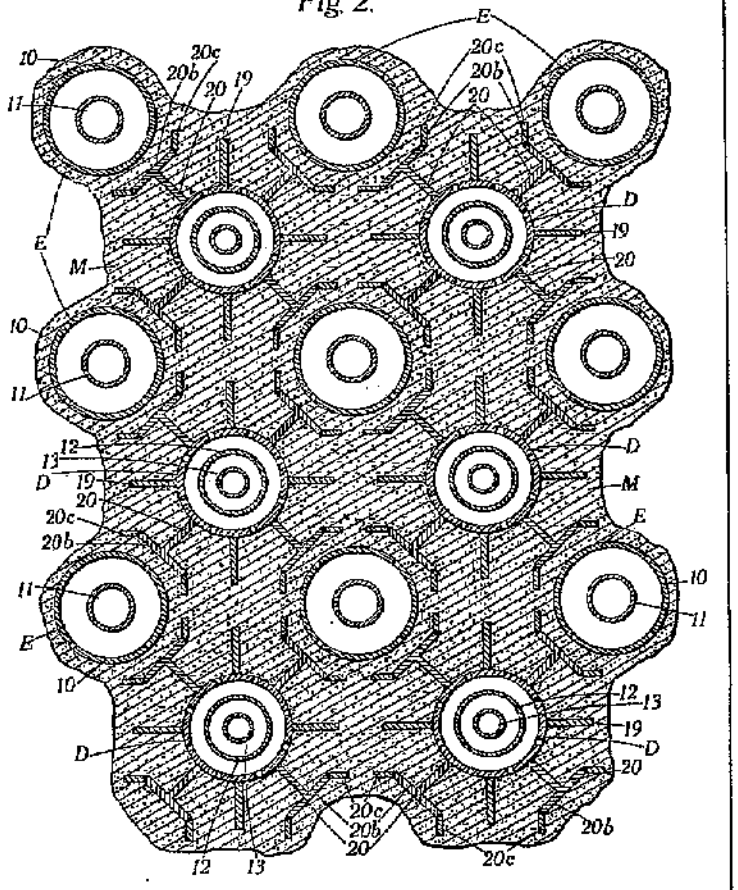


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Fig. 3.

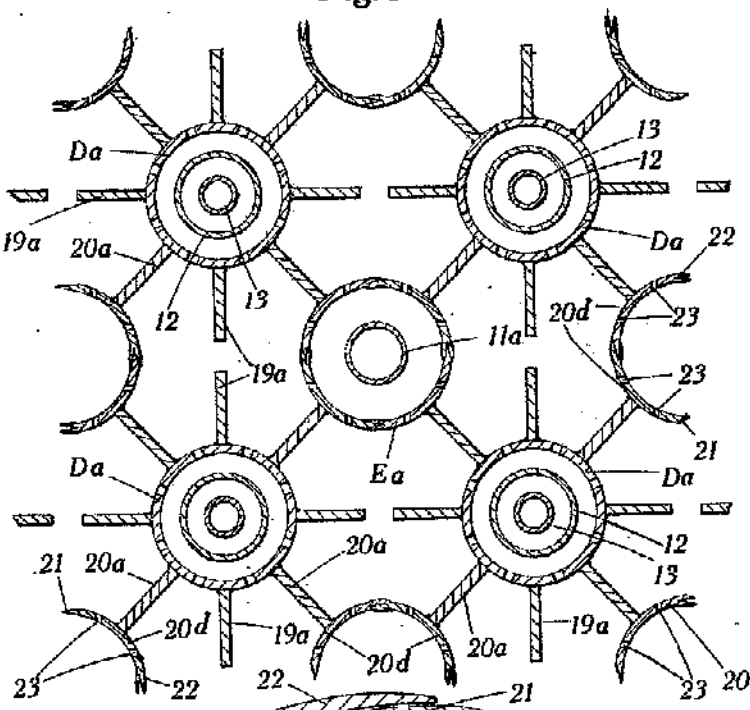

Fig. 4.

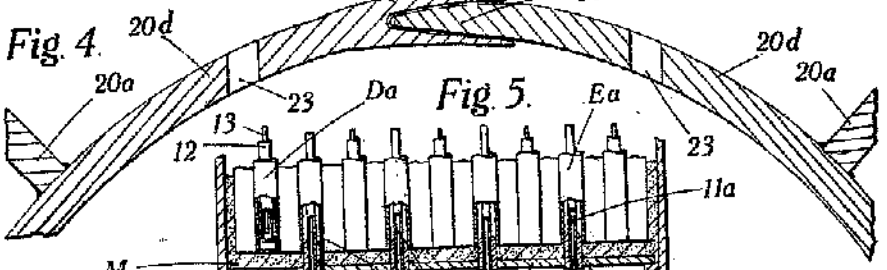

Fig. 5.

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