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PATENT SPECIFICATION

728,521

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COMPLETE SPECIFICATION

Improvements in or relating to Reactors

We, RUHRCHEMIE AKTIENGESELLSCHAFT, of Oberhausen-Holten, Germany, a German Company, and LURGI GESELLSCHAFT FUER WAERMETECHNIK M.B.H., of Heddernheim, Frankfurt am Main, Germany, a German Company, 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 invention relates to reactors, particularly to reactors in which the reaction is effected in the gaseous phase in the presence of a catalyst.

In many reactions carried out in the gaseous phase and in the presence of a catalyst, large quantities of heat are often evolved, but in other gaseous phase reactions it is sometimes necessary to supply heat. In the development of chemical engineering, reaction vessels have been evolved for use in both exothermic and endothermic gaseous phase reactions in which particular attention has been given to the heat exchange and pressure problems which are generally involved. For example, in the synthesis of hydrocarbons on an industrial scale by the catalytic hydrogenation of carbon monoxide, which hydrogenation is a strongly exothermic reaction, the use of "lamellar" reactors has become common for the synthesis when effected at or about atmospheric pressure. The catalyst space in such "lamellar" reactors is traversed by a large number of horizontally disposed cooling tubes through which the heat transfer medium is circulated, and vertical, metal plates extending throughout the length of the reactor are secured in spaced relationship to the horizontal tubes, the catalyst being retained in the reactor between the metal plates. When the synthesis is effected at pressures which lie approximately within the range 5-15 atmospheres, reactors have been used in which the catalyst is disposed in the annular space between two concentric tubes, a series of such double or twin tube elements being connected to a pressure drum from which boiling water flows around the elements. Furthermore, it has also been proposed to carry out the hydrogenation of carbon monoxide under pressure in vertical reactors in which the catalyst is disposed between tubes though which the cooling medium is circulated. The heat evolved in the reaction is conducted through the walls of the tubes to the liquid cooling medium which may then be cooled outside the reaction space.

It is an object of the invention to provide a reactor for carrying out catalytic gaseous phase reactions, particularly for the catalytic hydrogenation of carbon monoxide, in which the heat exchange and pressure problems encountered in such reactions may be solved in a particularly advantageous manner.

According to the invention, a reactor for carrying out reactions in the gaseous phase in the presence of a catalyst, comprises a vessel containing a system of welded tubes for a heat transfer agent, the tubes being substantially uniformly disposed over the major part of the cross-section of the vessel and having their corresponding ends connected to an upper series of headers which are connected to a drum disposed in the upper part of the reactor, the other ends of the tubes being connected to a lower series of headers which are connected to one or more inlet mains, one or more pipes connecting the inlet main or mains to the drum, the vessel having an inlet for the gaseous reactants and an outlet for the reaction product. The catalyst is disposed between the tubes.

In a preferred form of reactor according to the invention, the tubes are provided in the form of **U**-tubes, several of the tubes, for example three, being grouped together and cradled one in the other. The legs of the **U**-tubes which carry the rising heating or cooling medium terminate at their upper ends in the upper series of headers which in turn are connected by rising conduits to the drum disposed within and at the top of the reactor. From the drum, steam under pressure may be withdrawn if, for example, water under pressure is used as the cooling agent. The legs of the **U**-tubes carrying the descending heating or cooling agent are connected at their upper ends with the lower series of headers which in turn are connected by tubes to two inlet mains which project through the wall of the reactor and which are disposed transversely to the headers. From the drum, the end of which preferably extends outside the reactor, the heating or cooling agent is passed by pumps into the two inlet mains which likewise extend through the wall of the reaction vessel.

By the use of a pump, the rate of circulation of the heat transfer medium may be adjusted to correspond to the particular requirements with respect to the removal or supply of heat. At the same time it is possible to choose the particular direction in which the heating or cooling agent is circulated through the tubes. By means of a suitable reversing device for the pump the direction of circulation of the cooling or heating agent within the reactor may be changed. Where heat is removed from the reaction space by the heat transfer medium, the heat may be used for other purposes, for example for the heating of gases, for the generation of steam, or vapour formed by the heating of the heat transfer medium itself may be used as the working medium for a turbine. Similarly, where the reaction is endothermic, the heat transfer medium cooled in its passage through the reaction space may be used to cool other media before it is returned to the reaction space.

It is also possible so to provide the reaction vessel that the inlet mains and the lower series of headers are located below or in the lower part of the catalyst bed. In this case, straight tubes instead of the **U**-tubes connect the lower headers to the upper headers. The upper headers are connected to the drum disposed in the upper part of the reactor, and the drum is connected to the inlet mains. Thus, the circulation of the cooling agent may proceed by natural recirculation, that is to say, by thermo-syphon action, as well as by a pump.

The welded connection between the **U**-tubes and the headers is most practicably effected by providing the headers with connections, the spacing of which is as close as possible and to which the cooling tubes are welded. Thus, there is everywhere the same space available for the catalyst mass, that is to say, the spacing between two cooling tubes is substantially the same at any point.

The advantages provided by a reactor constructed in accordance with the invention consist in the possibility of controlling the temperature within narrow limits and of maintaining a substantially uniform temperature level in all parts of the reactor. Moreover, the particular arrangement of the tube system permits control of high pressures of the heating or cooling media, which pressures may be considerably higher than the gas pressure in the catalyst space surrounding the tube system.

For technical reasons, water cooling is generally preferred. Boiling water, by virtue of its high latent heat of vaporisation, provides, as cooling agent, one of the surest means for the removal of the considerable heat of reaction which is produced in many gas reactions. Furthermore, the reaction heat removed in this manner is readily and simply utilised for the production of useful steam in the simplest manner.

Apparatus constructed according to the invention is diagrammatically illustrated by way of example in the accompanying drawings, in which:—

Fig. 1 is a longitudinal section through the reactor;

Fig. 2A is a longitudinal section through the upper part of the reactor at right angles to the section shown in Fig. 1;

Fig. 2B is a transverse, staggered section through the reactor;

Fig. 3A is a transverse section through a header;

Fig. 3B is a longitudinal section through a portion of a header;

Fig. 4 is a longitudinal section through another reactor; and

Fig. 5 is a longitudinal section at right angles to the section shown in Fig. 4.

The reactor shown in longitudinal section in Fig. 1 consists of a pressure-resistant, cylindrical reaction vessel 1, the lower part of which is conically restricted, whilst its upper part may be closed by a removable, convex cover 2. In the upper part of the cylindrical shell, a drum 3 is mounted, one end of which is located inside the vessel and the other end of which extends outside the vessel 1. Below the drum 3, two parallel inlet mains 4 of smaller diameter are provided, one end of each of the mains 4 being disposed externally of the vessel 1 in a manner similar to that of the drum 3. A row of upper headers 5 and a row of lower headers 6 are provided below the two inlet mains 4, but the ends of the headers do not extend outside the cylindrical reaction vessel 1. The upper headers 5 are directly connected to the drum 3 by pipes 5*a*. The lower headers 6 are similarly connected by pipes 6*a* to the inlet mains 4.

The headers 5 and 6 are interconnected through tubes 14 provided in the form of hair pins or **U**-tubes. The tubes 14 are provided in groups of three, the tubes of one group being cradled one within the other. The heat transfer medium passes downwardly from the lower headers 6 through one leg of each tube 14 and rises through the other leg of each tube 14 to pass into the upper headers 5. The rising heat transfer medium then passes from the headers 5 into the drum 3 through the pipes 5*a*. From the drum 3, the heating or cooling medium is drawn through a flanged connection member or suction nozzle 7 by a pump (not shown) and passed through a pipe connection 8 into the two inlet mains 4; the heating or cooling agent then passes through the tubes 6*a* into the lower row of headers 6. From the headers 6 the heat transfer medium again enters the tubes 14, which are bent into the shape of a hair pin or **U**, and is thus recirculated through the system. Vapour under pressure, for example, steam where water is used as the cooling agent, may be withdrawn from the drum 3, through a valve. The valve or other pressure release means may be provided to operate automatically when a determined pressure is attained in the drum 3.

The removal or supply of heat required for the control of the reaction may also be effected by means of a heat exchanger provided in the suction or discharge line of the circulating pump. The heat exchanger may be utilised for the production of cold or heat.

The hair pin or **U**-tubes 14 are provided substantially over the entire cross section below the two rows of headers 5 and 6, each group of three tubes being provided so that the axes of the legs which are connected to headers 6 are disposed at the apices of an equilateral triangle; the axes of the legs connected to the headers 5 are disposed in a similar manner.

Just below the **U**-tubes 14, hinged, perforated plates 9 are provided to serve as a support for the catalyst mass. The plates 9 may be operated by lever 10 from outside the reactor. A removable base member 11 provided with a central gas outlet pipe 12, is secured to lower, conical part of the reactor 1. The gaseous reactants are passed into the reactor through an inlet 13.

The lower half of the staggered section shown in Fig. 2B is taken transversely through half of the reactor above the row of headers 5, whilst the upper half of Fig. 2B is a similar section taken through half of the reactor below the headers 6. Each two adjacent headers of the rows of headers 5 and 6 form one pipe element in which according to the space available, several groups of hair pin or **U**-tubes 14 are bunched together. As may be seen from the section in the upper part of Fig. 2B, the cooling or heating tubes 14 are so disposed that between them substantially the same space is available for the catalyst mass. Thus, heat may be removed or supplied in a substantially uniform manner during the reaction. Between the outer tubes 14 and the shell of the reactor, an insulating layer 15 is provided which is adapted to prevent the catalyst mass from settling between the reactor shell and the tubes whilst simultaneously maintaining the outer layer of the catalyst mass within a determined distance of the cooling surface of an adjacent tube 14.

The manner in which the **U**-tubes 14 may be connected and welded to the headers 5 and 6, is illustrated in Figs. 3A and 3B.

A further reactor constructed according to the invention is illustrated in Figs. 4 and 5. In this case, the circulation of the cooling or heating agent may be effected not only by a pump but also by a thermosyphon action (natural circulation). In this construction, the inlet mains 4 and the lower headers 6 and the interconnecting pipes 6*a* are provided in the lower part of the catalyst bed, whilst the lower headers are connected to the upper headers 5 by straight tubes 16, and not by **U**-tubes as in the reactor illustrated in Fig. 1. The headers 5 are directly connected to the drum 3.

With natural circulation, the heat transfer agent flows from the outlet 7 to the pipe connection 8 if heat is to be removed from the reactor (exothermic reaction), or in reverse direction through a heating system if heat is to be supplied to the reactor (endothermic reaction). As in the reactor first described, vapour may be withdrawn in exothermic operation by expansion in the drum 3. Moreover, the necessary quantity of heat may be removed by inserting a heat exchanger in the pipe line connecting 7 to 8. The heat exchanger may be utilised for the production of either cold or heat.

What we claim is:—

1. A reactor for carrying out reactions in the gaseous phase in the presence of a catalyst, comprising a vessel containing

a system of welded tubes for a heat transfer agent, the tubes being substantially uniformly disposed over the major part of the cross section of the vessel and having their corresponding ends connected to an upper series of headers which are connected to a drum disposed in the upper part of the reactor, the other ends of the tubes being connected to a lower series of headers which are connected to one or more inlet mains, one or more pipes connecting the inlet main or mains to the drum, the vessel having an inlet for the gaseous reactants and an outlet for the reaction product.

2. A reactor according to Claim 1, in which the tubes are provided as **U**-tubes, the upper and lower series of headers being provided adjacent to each other in the upper part of the reactor.

3. A reactor according to Claim 1 or Claim 2, in which the upper and lower series of headers are provided in parallel formation and substantially at right angles to the drum and to the inlet main or mains.

4. A reactor according to Claim 1, in which the tubes are substantially straight, the lower series of headers and each inlet main being disposed in the lower part of the reactor.

5. A reactor according to any one of the preceding claims, in which the pipe or pipes connecting the drum to each inlet main is disposed wholly outside the reactor.

6. A reactor according to any one of the preceding claims, in which means for circulating a heat transfer medium is provided in one or more of the pipes connecting the drum to each inlet main.

7. A reactor according to any of the preceding claims, in which the tubes are similar one to the other and are equally spaced apart.

8. A reactor according to Claim 2, in which the **U**-tubes are provided in groups of three which are cradled one within the other.

9. A reactor according to any one of the preceding claims, provided with a shell adapted to withstand pressure and with a hinged perforated plate or plates in its lower part adapted to support a mass of catalyst.

10. A reactor according to any one of the preceding claims, in which the drum is provided with a pressure release means.

11. A reactor for carrying out gaseous phase reactions in the presence of a catalyst, substantially as hereinbefore described.

12. A reactor, substantially as hereinbefore described with reference to Figs. 1, 2A, 2B, 3A and 3B of the accompanying drawings.

13. A reactor, substantially as hereinbefore described with reference to Figs. 4 and 5 of the accompanying drawings.

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728,521 COMPLETE SPECIFICATION
5 SHEETS
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SHEET 1

Fig. 1

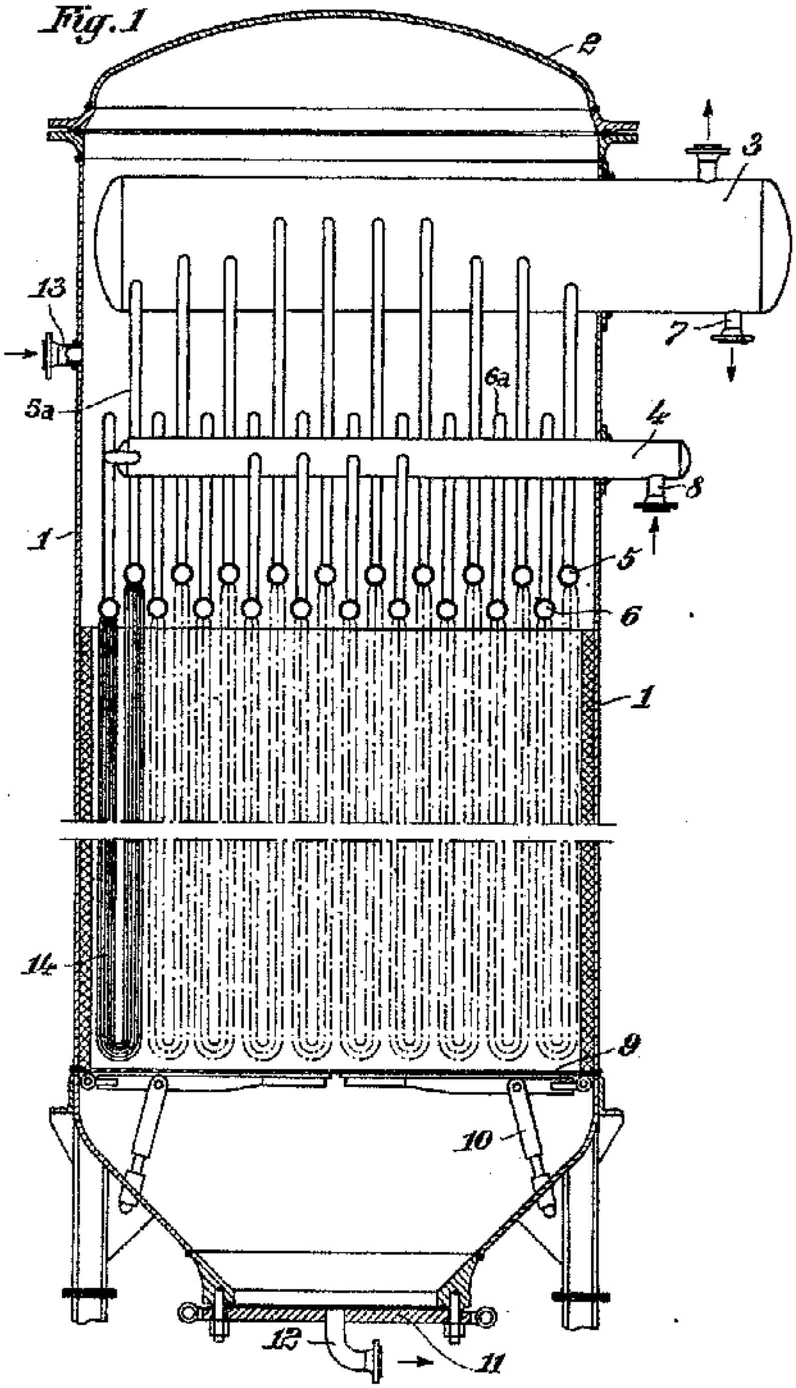


Fig. 2a

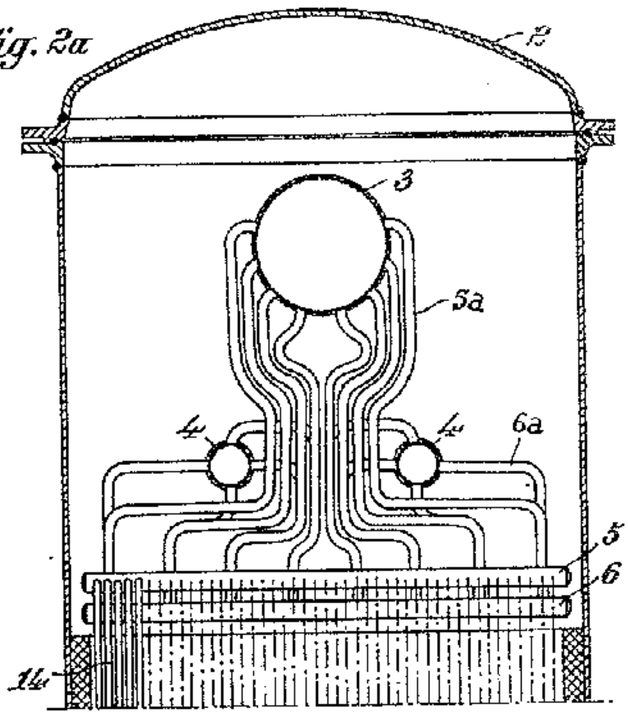
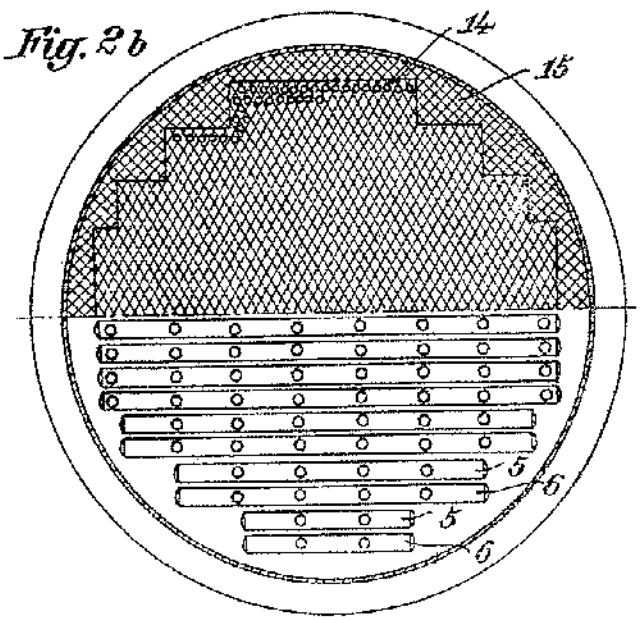


Fig. 2b



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SHEETS 2 & 3

Fig. 3a

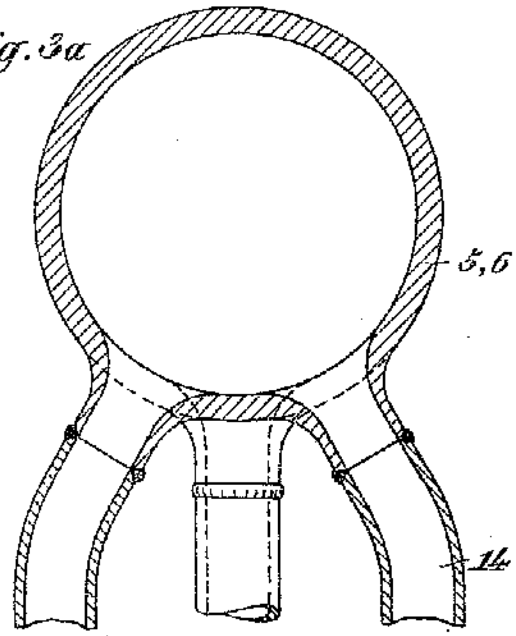
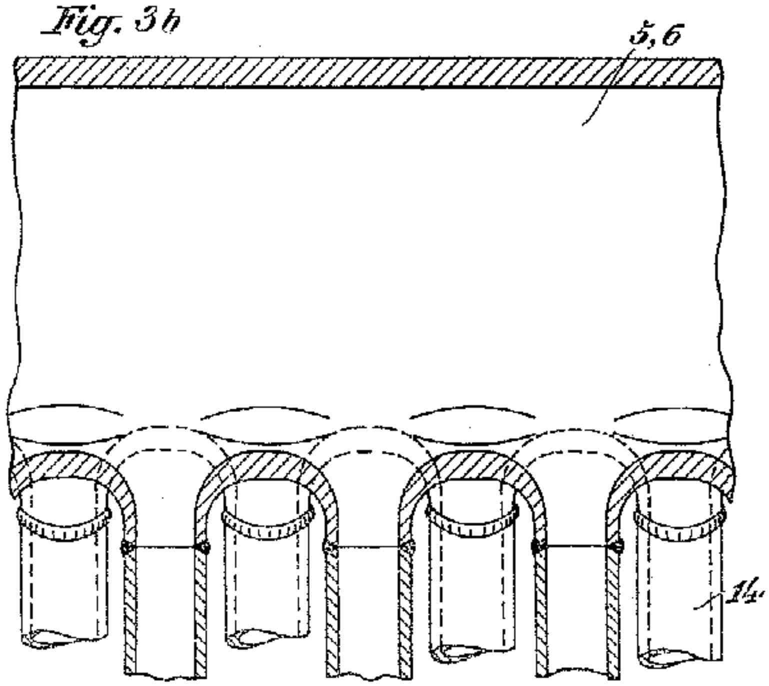


Fig. 3b



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SHEETS 2 & 3

Fig. 2a

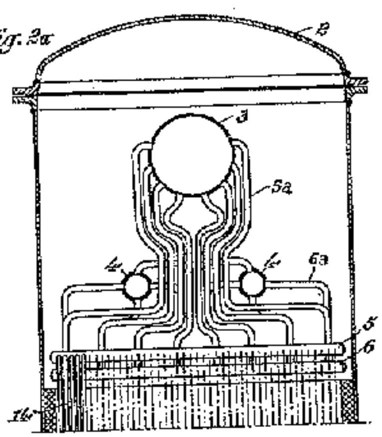


Fig. 2b

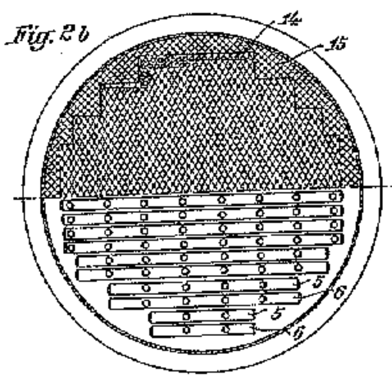


Fig. 3a

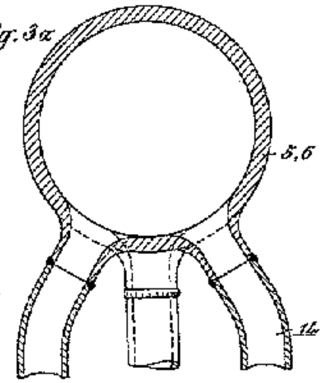


Fig. 3b

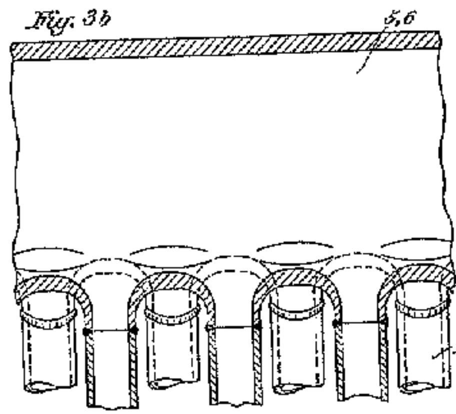
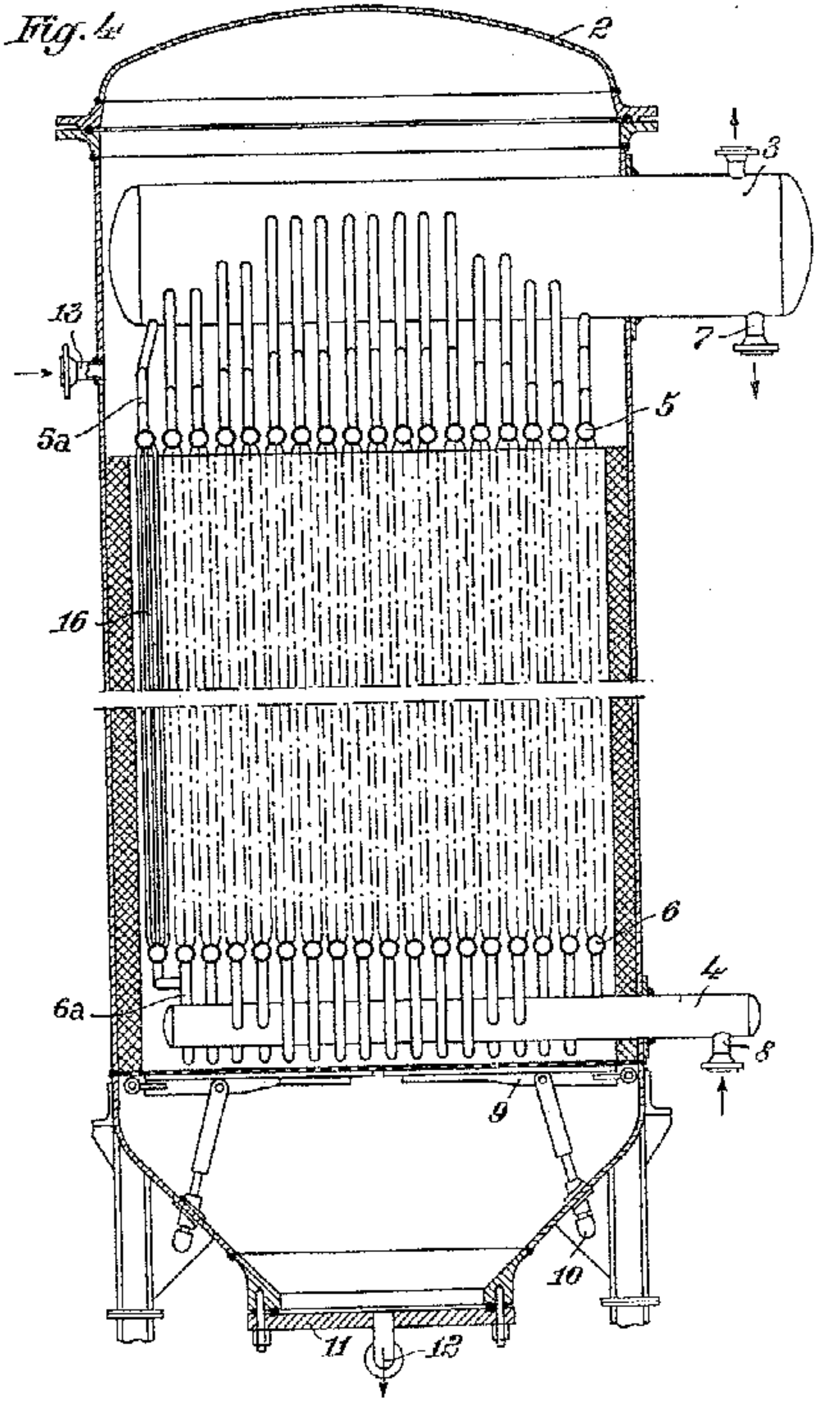


Fig. 4



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SHEETS 4 & 5

Fig: 5

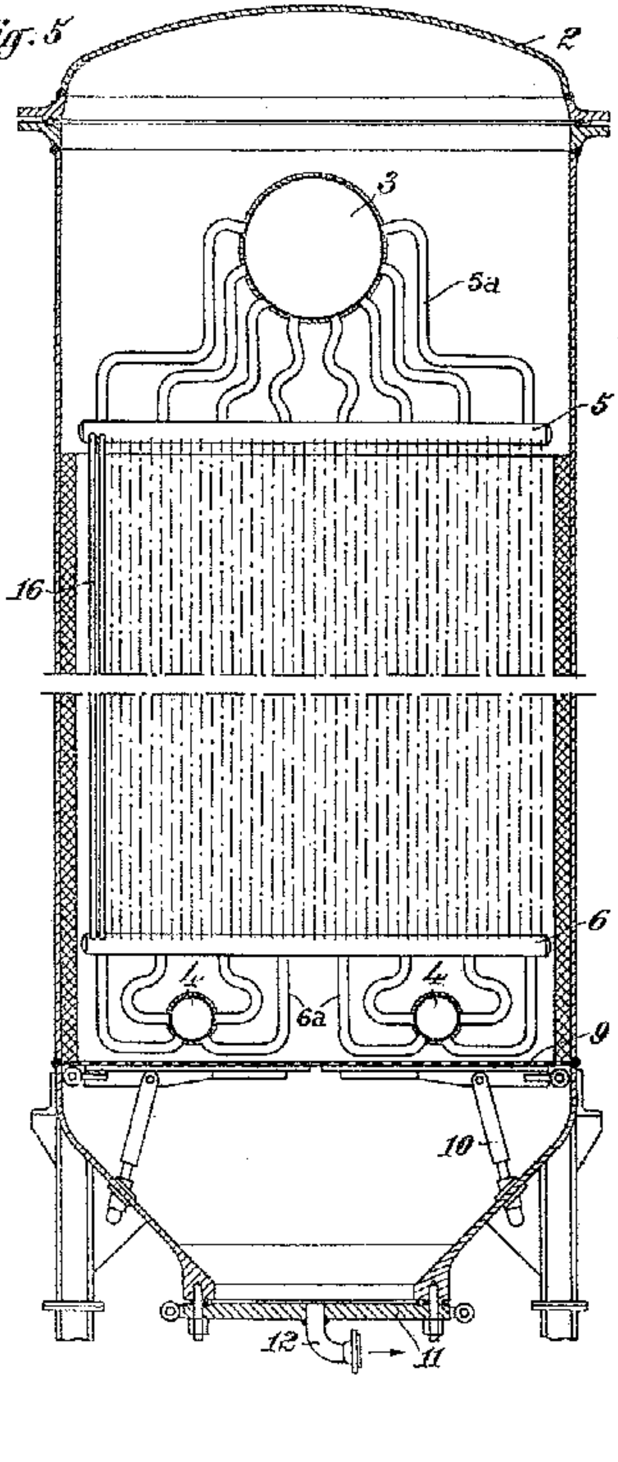

Fig. 4

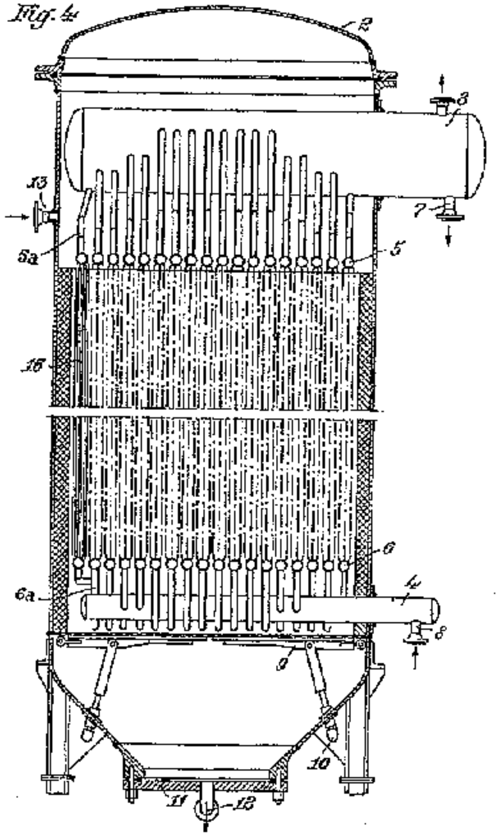

Fig. 5