

Dec. 24, 1940.

E. J. HOUDRY

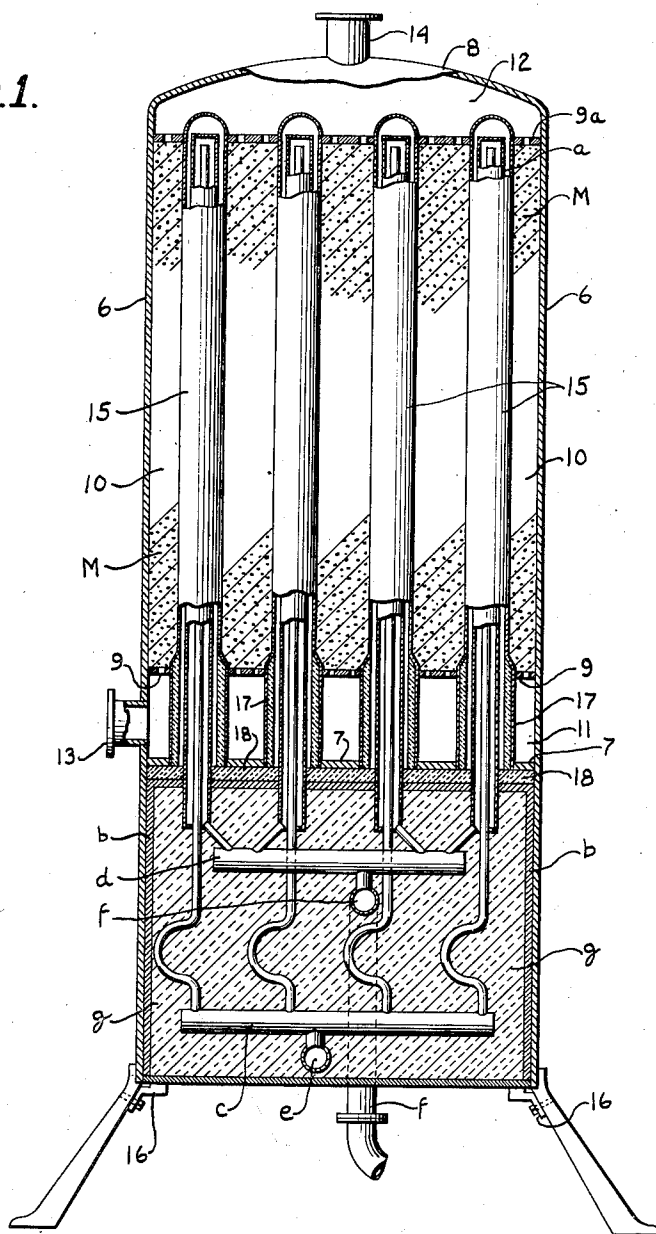
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REGULATION OF TEMPERATURE OF CHEMICAL REACTIONS

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Fig. 1.



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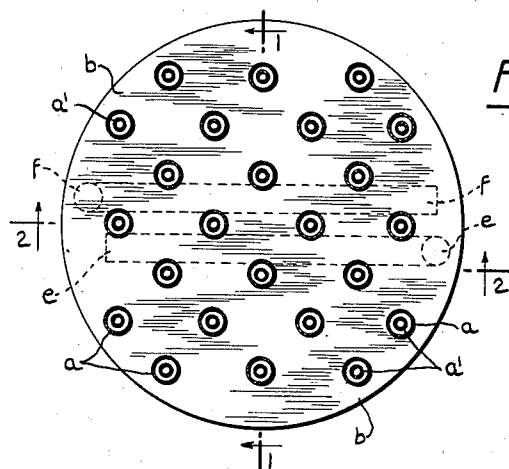


Fig. 3.

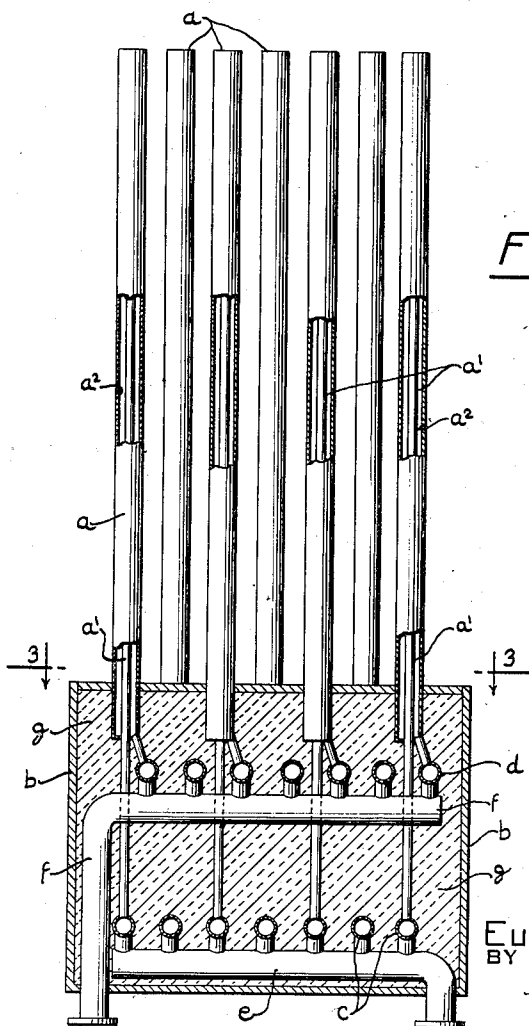


Fig. 2.

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REGULATION OF TEMPERATURE OF CHEMICAL REACTIONS

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4 Claims. (Cl. 23-288)

This invention relates to chemical reactions and their control. More particularly, it relates to the regulation of temperature of such reactions especially when effected in the presence of or by the aid of solid contact materials.

One object of the invention is to improve the temperature regulation of chemical converters. Another object is to restrict heat exchange to the reaction chamber or to a definite portion thereof. Another object is to avoid heat exchange with reactants entering a converter and/or with reaction products about to leave a converter. Another object is to devise improved forms of converters and of heat exchange apparatus therefor. Still other objects will be apparent from the detailed description which follows.

The present invention involves the utilization of cooperative but independent forms of converting and heat exchange apparatus. The entire heat exchange means is arranged to be applied to or removed from the converting means as a unit and to have extending members or elements received by and slidable within sockets or passageways which extend within or through the reaction chamber of the converting means. The heat exchange unit and the converter are suitably insulated from one another so that heat exchange is effected solely within the reaction zone. The transfer there is primarily by radiation. Hence the extent of heat exchange is governed by the temperature difference between the walls of the sockets or passageways and the heat exchange members or elements inserted therein. Accordingly, the maintenance of static or varying temperature conditions in the reaction zone is readily attained by suitable adjustment of the temperature and of the flow conditions of the heat exchange medium in the heat exchange unit. The extending members or elements on the latter are preferably of the nested conduit or reverse flow type to effect substantially uniform supply or removal of heat throughout the portions of the reaction zone traversed thereby.

In order to illustrate the invention, concrete embodiments of apparatus for practicing the same are shown in the accompanying drawings, in which:

Fig. 1 is a vertical, sectional view of a converter and its heat exchange unit in operative position, the section so far as the heat exchange unit is concerned being substantially on the line 1-1 of Fig. 3;

Fig. 2 is a vertical, sectional view of the heat exchange unit alone, but taken substantially at right angles to its position in Fig. 1, namely, on

the line 2-2 of Fig. 3, but on a broken line, so far as its base is concerned, in order to show both the inlet and outlet mains in elevation;

Fig. 3 is a transverse sectional view on the line 3-3 of Fig. 2, showing the base of the heat exchange member in elevation;

Fig. 4 is a vertical, sectional view similar to Fig. 1, but showing a different type of converter and the heat exchange unit mounted in a different position from Fig. 1, the section being taken substantially on the line 4-4 of Fig. 5; and

Fig. 5 is a right end elevational view of the converter shown in Fig. 4, with the heat exchange unit removed and the covering of heat insulating material stripped away from the converter.

In Fig. 1, the converter comprises an annular casing 6, closed at its lower end by a bottom plate 7 and at its upper end by a cover 8. The interior of the converter has perforated partitions 9 and 9a adjacent its lower and upper ends, respectively, dividing the interior into a large central reaction chamber 10 adapted to be filled with solid contact or catalytic material M, and end manifolding chambers 11 and 12. A conduit 13 gives access to manifolding chamber 11 and a conduit 14 to manifolding chamber 12. Reactants may enter the converter by either conduit, as desired, and the resulting reaction products may leave by the other conduit. For temperature regulation of the reactions taking place in chamber 10, the converter is provided with members 15 which provide recesses within the reaction chamber opening through a wall of the converter but which are sealed against all communication with the interior of the converter. In the form shown in Fig. 1, these members 15 consist of conduits sealed at one end, having their open ends secured to the bottom wall or plate 7 of the converter in fluid-tight relation, the members 15 extending through suitable openings therefor in partition 9 and into the reaction chamber 10 to any desired extent. By preference, members 15 project through suitable openings in upper partition 9a, so that members 15 are maintained in fixed position within the converter as well as in desired symmetrical pattern and in predetermined spaced relation with one another throughout the reaction chamber. The portions of members 15 which lie within reaction chamber 10 are entirely surrounded by the contact mass M.

Within the recesses or passageways provided by hollow members 15 extend heat exchange elements a of an independent and removable heat exchange unit, which is shown by itself in Figs. 2 and 3. This unit consists of a base b, from

which elements *a* extend, the base being adapted to fit in a chamber adjacent the converter. In Fig. 1, the chamber is beneath the converter and formed by an extension of the annular wall 6 of the converter, the base being held in place by suitable detachable supports indicated at 16 in Fig. 1. It will be understood that the converter is either in an elevated position or has a pit therebeneath of sufficient size to accommodate the heat exchange unit and to permit its convenient mounting in or removal from operative relation with the converter. Elements *a* conform in number and in arrangement to the socket members 15 of the converter with which they cooperate, and, in order to insure a substantially uniform rate of heat exchange with all parts of the reaction chamber adjacent thereto, elements *a* are preferably of the nested conduit type. Hence each heat exchange element *a* consists of a small inlet pipe *a*¹ which extends centrally of and substantially the full length of outer pipe *a*². Each inner pipe *a*¹ connects with an inlet manifold *c* and the connection thereto may be bent or looped, as indicated in Fig. 1, to take care of expansions and contractions due to changes in temperature. Each outer pipe *a*² is closed, save for an outlet connection to an outlet manifold *d*. There may be an inlet manifold *c* and an outlet manifold *d* for each row of heat exchange elements *a* mounted upon base *b*, seven manifolds of each kind being shown in Fig. 2, the inlet manifolds connecting with inlet main *e* and the outlet manifolds with outlet main *f*. The mains project through the bottom of base *b* and have flanged ends for convenient connection to conduits leading to the rest of the heat exchange circuit. The interior of base *b* not occupied by the manifolds, mains, etc., is filled with heat insulating material *g* to avoid or to restrict heat exchange between the various manifolds and mains. To simplify the showing in Fig. 3, only inlet main *e* and outlet main *f* are indicated in broken outline in the base *b* of the heat exchange unit.

When the heat exchange elements *a* extend through a manifolding chamber of a converter as shown in Fig. 1, it may be and frequently is desirable or important to prevent heat exchange with the reactants entering the converter or the reaction products leaving the converter, depending upon whether the manifolding chamber is used as an entrance or as an exit for fluid. This can be accomplished by an intervening wall of heat insulating material applied to the exterior of elements *a* or to the interior of members 15, as may be most desirable or convenient. One way to handle the difficulty is indicated in Fig. 1, wherein the lower ends of members 15, at least as to the parts traversing manifolding chamber 11, are expanded or enlarged, and this enlarged area is filled with heat insulating material 17. As a further protection against undesirable heat exchange, a thick layer of heat insulating material 18 may be applied to the exterior of bottom plate 7 to be thus interposed between base *b* of the heat exchange unit and the converter.

Figs. 4 and 5 show the invention applied to a converter of rectangular form, made up of end plates 21, side plates 22, and top and bottom plates 23 and 24, respectively. The interior of the converter has upper and lower perforated partitions 25 and 26, which divide the interior into a large central reaction chamber 27 adapted to be filled with solid contact or catalytic material *M*, and end manifolding chambers 28 and 29, to which fluids have access by conduits 30 and 31,

respectively. The entire reaction chamber 27 is traversed by a plurality of members 32 in the form of conduits which are mounted in a fluid-tight manner in the opposed end plates 21. Hence members 32 serve to tie opposite walls of the converter together in a rigid manner and to provide passageways or recesses communicating with the exterior of the converter for the convenient insertion of heat exchange elements, after the manner already illustrated in connection with Fig. 1. In this instance, however, the heat exchange unit is brought into operative relation with the converter by horizontal movement rather than vertical, as indicated in connection with Fig. 1.

The entire converter shown in Fig. 4 is enclosed by a thick layer 33 of heat insulating material, save for openings therethrough registering with one end of members 32 for the insertion of heat exchange elements *a* of the heat exchange unit. The unit shown in Fig. 4 has a base *b*¹, from which the heat exchange elements *a* extend, the base *b*¹ being of less depth or thickness than that shown in Figs. 1 and 2, and with its inlet manifolds *c* closer to the outlet manifolds *d* due to the omission of expansion bends in the inlet pipes for the heat exchange elements. This is possible when the entering temperature of the heat exchange fluid is close to its outlet temperature, the adequacy of heat removal or heat supply being then a function of the rapidity of circulation of the heat exchange medium. It will be understood that base *b*¹ is substantially rectangular in form and has the proper number of projecting heat exchange elements *a* and in proper relation to one another to cooperate with all the recesses or passageways provided by elements 32 in the converter. The support 45 for base *b*¹ may be associated with or independent of the supporting means for the converter with which the heat exchange unit cooperates.

Fig. 4 also indicates diagrammatically the parts of the exterior circuit for the heat exchange fluid. These parts comprise a conduit 40 connected to outlet main *f*, a heat exchanger 41 for removal or supply of heat to the heat exchange medium, and a conduit 42 conducting the fluid from heat exchanger 41 to the inlet main *e*, suitable impelling means, such as a pump 43, being provided in line 42, or, alternatively, in line 40, to insure and to control the circulation of the heat exchange medium.

The invention is adapted for the temperature regulation of chemical reactions of all types, endothermic or exothermic, such as those involving synthesis, metathesis, decomposition, hydrogenation, etc. Any form of solid contact material capable of promoting or in any way assisting in the reaction may be utilized. In certain reactions, such as those involving the treatment or conversion of hydrocarbons derived from deposits of petroleum, coal, lignite, shale, etc., the solid contact material may be silicious in character, including silica-alumina blends, with or without small amounts of other active components, such as metals or metallic compounds, the catalyst being in the form of bits, fragments, or molded pieces to facilitate regeneration. The catalyst, during the on-stream reaction, accumulates a deposit of a coky, sulphurous or resinous nature which impairs the activity of the catalyst and necessitates the removal of the deposit for the reaction to be continued. Hence, in treating or converting such starting materials, it is customary to conduct regenerating reactions for the

removal of contaminants following the on-stream reactions, and the regenerating reaction is usually different from the on-stream reaction, as exothermic instead of endothermic, or of the same nature but more intense.

Since the present invention seals off the heat exchange fluid from any possibility of admixture, through leakage or otherwise, with the reactants or with the contact mass within the reaction chamber, any desired fluid may be used as the temperature controlling medium. Hence the medium may be gaseous or liquid, single phase or two phase, under low pressure or high pressure. If the fluid selected is of the single phase type, such as a gas or a liquid which does not vaporize or is not permitted to vaporize, under the conditions of operation, the speed of circulation must be relied upon to remove or to supply the necessary quantity of heat. Liquids which may be used for this purpose include fused salts, certain metals and metal alloys, and water under pressure when the temperature does not exceed its critical temperature. Two phase fluids, such as water, mercury, diphenyl, etc., are useful when very large amounts of heat are to be supplied or removed. Elements *a* of the heat exchange unit may be provided with suitable guide or spacing means (not shown) to facilitate their insertion into the recesses or passageways formed by members 15 or 32 and to maintain them suitably centered therein.

Any of the above mentioned heat exchange fluids may be utilized when the reactions are mild, so far as heat liberated or heat required is concerned. A reaction typical of those most difficult to control is the regeneration of contact material which has been used for the conversion or treatment of hydrocarbons. Since this reaction is not productive, it is usually desirable to complete the reaction as soon as possible, in order that the contact material may be put back on stream and again used for a productive operation, as in the conversion or cracking of higher boiling hydrocarbons to produce lower boiling hydrocarbons including gasoline. The on-stream temperature may be in the range of 775° to 925° F., while the regeneration by oxidation of contaminants will produce a large amount of heat at a rapid rate, and, when the deposit is considerable, may carry the catalyst to such a temperature that its adsorptive capacity may be impaired or destroyed. Hence it is desirable to restrain the catalyst from exceeding a temperature of 1200° F. during the regenerating period and to bring the catalyst back quickly to the on-stream temperature after regeneration is completed. For the control of such a regeneration, water offers a convenient and cheap solution, especially when utilized in accordance with the present invention. The water can be sent into the inlet manifolds *e* of a heat exchange unit of the types shown at a low temperature as soon as the regeneration or burning starts or is well under way. It will remove heat rapidly and in large quantities from the reaction chamber both as sensible heat and as heat of vaporization, the vaporized water or steam being withdrawn through the outlet manifolds *f*. Insulation within the base *b* or *b'* of the heat exchange unit and between the latter and the converter serves to confine the heat exchange to the reaction chamber of the converter, and, since the heat exchange elements *a* are of the nested conduit type, and, since the heat exchange between the unit and the socket members 15 or 32 is primarily by radiation, heat is withdrawn uni-

formly from all parts of the contact mass, and excessive cooling of localized portions of the same is avoided. By continuing to supply water up to the end of the regeneration and even somewhat thereafter, the reaction chamber can be quickly cooled down to on-stream temperature. The circulation of water can then be stopped, and the heat exchange unit can be completely emptied of all heat exchange fluid, since the mains *e* and *f* represent the low points in the heat exchange unit and any liquid remaining will drain thereto by gravity.

When more time is available for the regeneration, as several hours instead of an hour or less, and especially when the on-stream reaction is conducted at a lower temperature, such as in the refining and stabilizing of light hydrocarbons, including motor fuels of the gasoline type, which may be effected in the range of 400° to 700° F. and usually between 500° and 600° F., the regenerating reaction may be conducted somewhat differently. However, in such a case the catalyst needs to reach temperatures as high or nearly as high as in the regeneration previously described, in order to be sure that the catalyst is properly freed of contaminants. In this instance, the reaction is not so intense or extreme, because more time is given to completing it. Accordingly a single phase heat exchange fluid, such as fused salts, can now be used to advantage. The burning or regeneration is initiated, after preheating the catalyst if necessary, and the salt is circulated at such a rate that it rises in temperature following the increase in temperature of the catalyst during regeneration. As the burning becomes less intense and finally ceases, heat is removed from the circulated salt, and the reaction chamber and the catalyst then follow the salt down in temperature. By suitable adjustment of the rate of circulating the salt, heat can be recovered at a constant rate by such means as heat exchanger 41 during the entire regenerating operation. Thus, if water is used as the cooling medium in heat exchanger 41, steam can be produced at a constant rate during the entire regenerating period. Of course, in a commercial installation two or more converters will be utilized, so that at least one is always on stream while at least one other is always in regeneration, thus effecting continuity of both reactions and permitting an economical operation of the heat exchange circuit.

From the above it will be apparent that the present invention has important advantages and wide flexibility in the control of chemical reactions. For example, the heat exchange medium may be utilized only during a portion of the cycle of operation, as when there is large evolution of or demand for heat, since thereafter the medium can be entirely removed or drained from the heat exchange unit. On the other hand, when heat rise or loss is gradual and continuous, heat exchange is accurately controlled by adjusting the temperature of the heat exchange medium and/or its rate of flow. Hence the movement of heat to or from the reaction chamber may be made as gradual or as abrupt as desired, while effecting the same with a high degree of uniformity throughout the reaction chamber and with an avoidance of chilling or over heating of localized portions of the contact mass. Moreover, since the heat exchange unit is separate and distinct from the converter and is readily withdrawn from cooperative relation therewith, all danger of leakage of the heat exchange medium into the re-

action chamber of the converter is avoided. This gives complete freedom of choice of heat exchange media, and facilitates inspection and repair of both the converter and the heat exchange unit.

It is to be understood that converters need not be mounted in a vertical position, such as shown in the drawings. Some, if not all, of the advantages of the present invention are available when converters are mounted in other positions. Such minor changes, modifications and variations are intended to be within the scope of the appended claims.

I claim as my invention:

1. A casing having side and end walls and partitions extending thereacross providing a reaction chamber containing contact material and adjacent manifolds for supplying fluid to and removing fluid from the contact material, apertures in an outer wall of the casing, conduits secured in said apertures and extending through the contact material providing passageways therein, in combination with a heat exchange unit, said unit comprising a container having an apertured end wall, means for detachably securing the container to the casing with the container end wall apertures in alignment with the casing conduits, tubes for heat exchange fluid disposed within the casing conduits and extending substantially the length of said conduits, said tubes having inlet and outlet end portions extending through the apertures in the container end wall and into said container, means for securing said tube end portions in the container and positioning the tubes within said casing conduits, manifolds within said container, at least one manifold in communication with tube inlet end portions and at least one other manifold in communication with the tube outlet end portions, means for supplying fluid to one manifold for passage through the tubes and means for removing fluid from another manifold after passage through the tubes, said means being so constructed and arranged with respect to the casing that the heat exchange unit is readily detachable from the casing.

2. A casing having side and end walls and partitions extending thereacross providing a reaction chamber containing contact material and adjacent manifolds for supplying fluid to and removing fluid from the contact material, apertures in an outer wall of the casing, conduits secured in said apertures and extending through the contact material providing passageways therein, in combination with a heat exchange unit, said unit comprising a container having an apertured end wall, means for detachably securing the container to the casing with the container end wall apertures in alignment with the casing conduits, sets of inner and outer nested tubes for passage of heat exchange fluid disposed within the casing conduits and extending substantially the length of said conduits, each of said tubes being secured in the container and having an end portion passing through the container end wall and into the container, said outer tubes having closed ends disposed within the casing conduits, said inner tubes having an open end positioned adjacent the closed ends of the outer tubes, manifolds within said container, at least one manifold in communication with the end portions of the inner tubes and at least one other manifold in communication with the end

portions of the outer tubes, means for supplying fluid to one manifold for passage through the tubes and means for removing fluid from another manifold after passage through the tubes, said means being so constructed and arranged with respect to the casing that the heat exchange unit is readily detachable from the casing.

3. A casing having side and end walls and partitions extending thereacross providing a reaction chamber containing contact material and adjacent manifolds for supplying fluid to and removing fluid from the contact material, apertures in an outer wall of the casing, conduits secured in said apertures and extending through the contact material providing passageways therein, in combination with a heat exchange unit, said unit comprising a container having an apertured end wall, means for detachably securing the container to the casing with the container end wall apertures in alignment with the casing conduits, sets of inner and outer nested tubes for passage of heat exchange fluid disposed within the casing conduits and extending substantially the length of said conduits, each of said tubes having an end portion passing through the container end wall and into the container, said outer tubes being secured to the container end wall and having closed ends disposed within the casing conduits and apertured end walls within the container, said inner tubes being secured in the apertured end walls of the outer tubes, said inner tubes having an open end positioned adjacent the closed ends of the outer tubes, manifolds within said container, at least one manifold in communication with the end portions of the inner tubes and at least one other manifold in communication with the end portions of the outer tubes, fluid connections secured to said manifolds and opening outwardly of the container for supplying fluid to and removing fluid from the manifolds.

4. A casing having side and end walls and partitions extending thereacross providing a reaction chamber containing contact material and adjacent manifolds for supplying fluid to and removing fluid from the contact material, apertures in an outer wall of the casing, conduits secured in said apertures and extending through the contact material providing passageways therein, in combination with a heat exchange unit, said unit comprising a container having an apertured end wall, means for detachably securing the container to the casing with the container end wall apertures in alignment with the casing conduits, tubes for heat exchange fluid disposed within the casing conduits and extending substantially the length of said conduits, said tubes having inlet and outlet end portions extending through the apertures in the container end wall and into said container, means for securing said tube end portions in the container and positioning the tubes within said casing conduits, manifolds within said container, at least one manifold in communication with tube inlet end portions and at least one other manifold in communication with the tube outlet end portions, insulating material within said container and surrounding said tube ends and manifolds, fluid connections secured to said manifolds and opening outwardly of the container for supplying fluid to and removing fluid from the manifolds.

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