Jan. 10, 1956 E. J. HOUDRY 2,730,434

CATALYTIC CONTACTING UNIT

Filed May 1, 1950 3 Sheets-Sheet 1

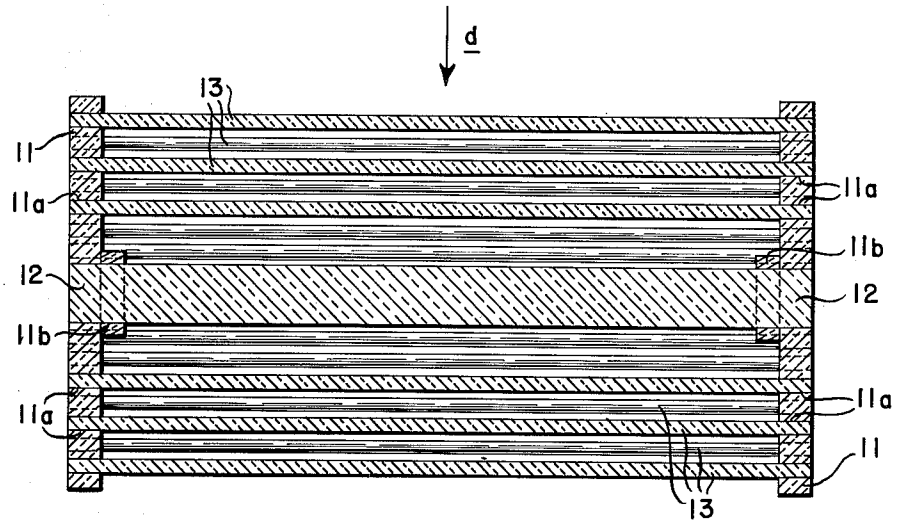

FIG. 1.

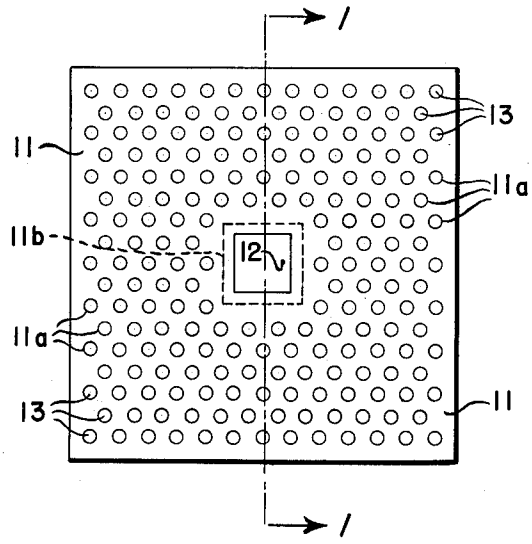

FIG. 2.

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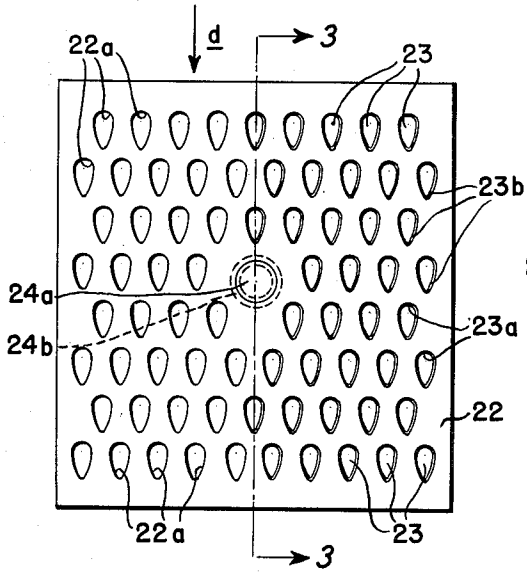

FIG. 4.

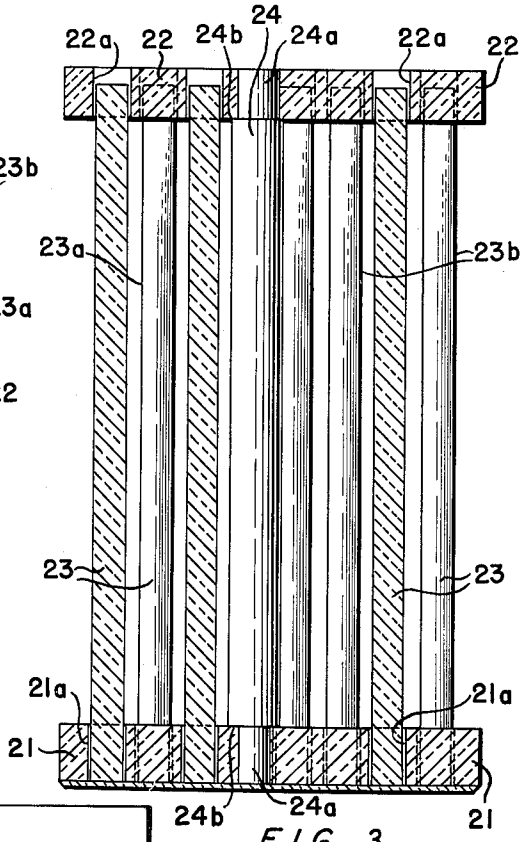

FIG. 3.

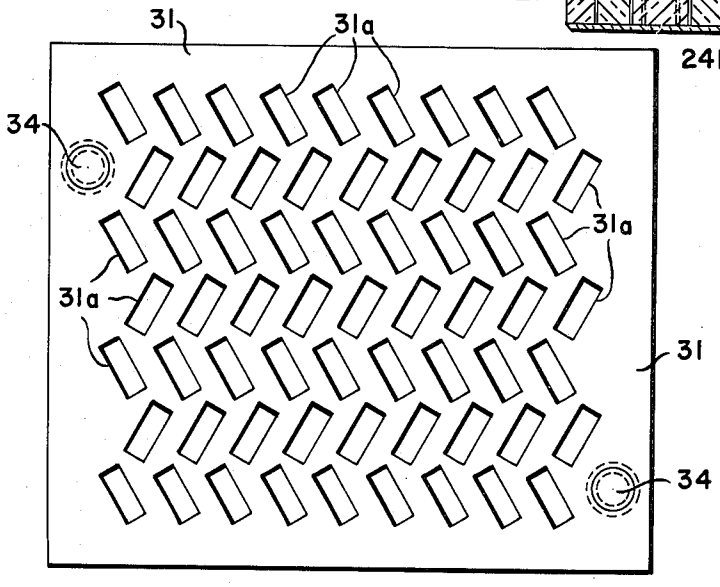

FIG. 7.

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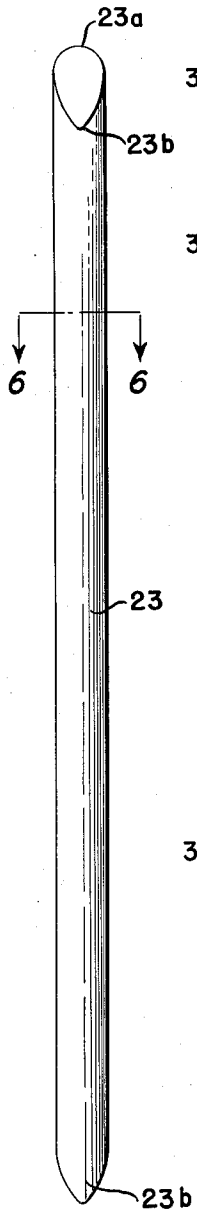

FIG. 5.

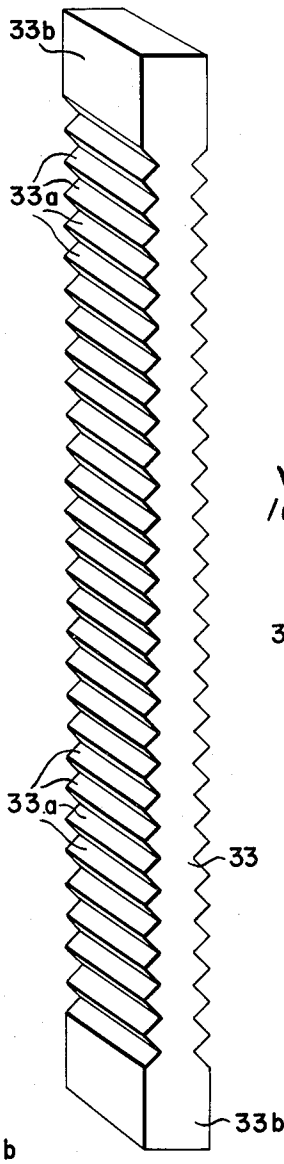

FIG. 8.

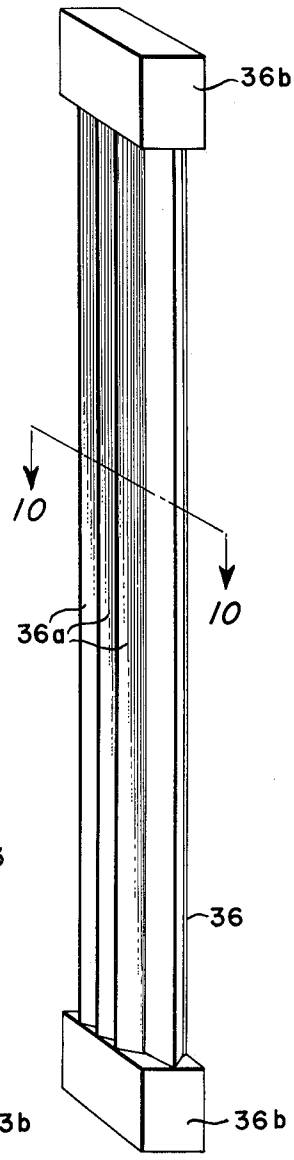

FIG. 9.

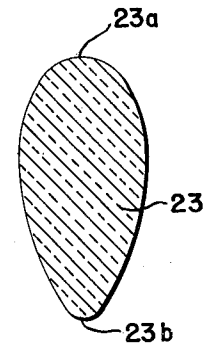

FIG. 6.

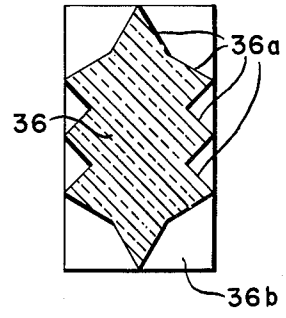

FIG. 10.

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United States Patent Office 2,730,434 Patented Jan. 10, 1956

2,730,434

CATALYTIC CONTACTING UNIT

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Application May 1, 1950, Serial No. 159,191

15 Claims. (Cl. 23—288)

This invention relates to contacting operations generally and in particular to such operations when promoted by catalysts. More particularly it has to do with the effective control of contacting and catalytic operations and reactions and involves both process and apparatus aspects.

A principal object of the invention is to establish control of temperature, pressure, and flow conditions in contacting operations.

Another object is to solve the problem of excessive temperature and pressure changes.

Another object is to provide for independent mounting and support of contacting and catalytic elements.

Another object is to support the contacting and catalytic elements in such a manner that friction and attrition are avoided thereby to prolong the life and effective use of such elements.

Another object is to provide the contacting elements in units for convenient handling, assembly, impregnation with catalysts, and mounting, while furnishing a desired degree of flexibility as to number, disposition, and arrangement of elements for control of temperature, pressure, and time of contact.

Other objects and advantages will be apparent from the detailed description which follows.

In accordance with the present invention the fluid contacting or catalytic elements are formed in units or assemblies which may be utilized individually or stacked adjacent to or upon one another like bricks. By preference, the parts of these units or assemblies are of dense hard material having any suitable or desired degree of porosity or absorptivity, usually slight or low, such as ceramic material including porcelain, and the latter may be of the type used for the cores of spark plugs, and which has been heated to high temperatures of the order of 2000° and above. The unitary structure comprises upper and lower apertured members or plates maintained in fixed spaced relation by one or more posts or spacer bars, the apertures in the members or plates being disposed in alignment for the individual insertion of elongate elements in the form of rods or sticks. Each element is by preference secured at one end to one of the end members or plates and is endwise movable in the aperture therefor in the other member or plate so as to avoid breakage from expansion and contraction during temperature changes. Time and extent of contact of fluids is determined in part by the number of elements mounted in the apertures in the end members of each unit and by the size, shape, arrangement and cross sectional contour of the elements.

In order to illustrate the invention concrete embodiments of apparatus suitable for carrying out the process of the invention are shown in the accompanying drawings in which:

Fig. 1 is a longitudinal sectional view of one form of unit, the section being taken in the line 1—1 of Fig. 2;

Fig. 2 is an end elevational view of the unit shown in Fig. 1;

Fig. 3 is a longitudinal sectional view, taken on the line 3—3 of Fig. 4, showing a modification of the invention;

Fig. 4 is an end elevational or top plan view of the unit shown in Fig. 3, the left half of unit being without elements;

Fig. 5 is a perspective view on an enlarged scale of one form of element for mounting in the unit shown in Figs. 3 and 4;

Fig. 6 is a transverse sectional view on a still larger scale, the section being taken on the line 6—6 of Fig. 5;

Fig. 7 is a plan or end elevational view similar to Fig. 4 of a modified form of unit, elements to fit the apertures being omitted;

Figs. 8 and 9 are perspective views on an enlarged scale of elements to be mounted in the unit of Fig. 7; and

Fig. 10 is a transverse sectional view on a still larger scale on the line 10—10 of Fig. 9.

As shown in Figs. 1 and 2 the contacting or catalytic unit comprises a pair of apertured end members or plates 11 in spaced relation to each other and secured to a central spacer or post 12 to form a frame, adapted to support a multiplicity of contacting or catalytic elements 13. As indicated in Fig. 2 elements 13 are round in cross section and extend between end members 11 and have their ends seated in aligned pairs of apertures 11*a*. Spacer or post 12 is square in cross section and extends through openings of the same configuration in end members or plates 11, inwardly directing bosses 11*b* on the latter insuring secure attachment between post 12 and members 11 and imparting strength and rigidity to the frame. Elements 13 are in the form of rods which are inserted endwise through aligned apertures 11*a* to be supported individually and independently of one another. By preference elements or rods 13 are cemented, glazed or otherwise anchored in the apertures of one of end members 11 while care is taken to see that their other ends are freely slidable in the apertures of the other end member 11. This arrangement prevents breakage of elements 13 or of end members 11 when temperature differences occur in the parts of the unit.

As indicated by Fig. 2 apertures 11*a* in members or plates 11 dispose elements 13 in rows with adjacent elements in staggered arrangement so that fluids sent through this unit from the side, as in the direction of the arrow *d* on Fig. 1, are caused to flow over and around each element.

In the modification of the invention shown in Figs. 3–6 the lower end member or plate 21 of the frame has its apertures 21*a* extending only partly therethrough to provide a positive stop for the lower ends of elements 23 which are mounted in the frame by inserting them through the apertures 22*a* in upper end member or plate 22. A central spacer or post 24 is attached at its ends to members or plates 21 and 22 to form the frame of the unit, the ends 24*a* of the spacer 24 being reduced in size to provide stop shoulders 24*b* to engage the inner faces of members 21 and 22 as indicated in Fig. 3. As clearly shown by Figs. 4, 5 and 6 elements 23 to be mounted in the frame of Fig. 3 are of teardrop or streamlined design in cross section and the apertures 21*a* and 22*a* in end members or plates 21 and 22 are of complementary shape and are all disposed in the same direction. Since the elements or rods, 23, are rounded at one vertical edge 23*a* and tapered toward the opposite vertical edge 23*b* (as shown in Figs. 4, 5 and 6) to produce a streamlined effect, they offer the possibility of a minimum pressure drop in reactants passing thereover in the direction of movement indicated by arrow *d* of Fig. 4 and at the same time give the best contact of the reactants with the surfaces of elements 23 while producing a minimum of

turbulence. The lower ends of elements **23** will be secured in apertures **21***a* of lower member or plate **21** by cementing, glazing or otherwise while their upper ends will be freely slidable in apertures **22***a* of upper member or plate **22**.

Figs. 7, 8, 9 and 10 illustrate further modifications of the contacting unit and of the elements supported thereby, in this instance, for creating turbulence and a greater period and extent of contact of reactants with the elements. In the form shown in Fig. 7 there are two spacers or posts **34** (similar to spacer **24** of Figs. 3 and 4) to support and hold end members or plates **31** (only one of which is shown) in spaced relation. Apertures **31***a* to receive elements **33** (Fig. 8) or elements **36** (Fig. 9 and Fig. 10) are rectangular in shape and are not only staggered relative to the apertures in adjacent rows (as in the modifications of Figs. 1 and 2 and of Figs. 3 and 4) but are also zig-zagged to baffle and to create turbulence and frequent changes in direction in reactants sent through the unit from any side. In order to effect further turbulence and extended contact with the elements mounted in apertures **31***a* the surfaces of the elements are made irregular or of increased extent in any desired manner. One way to accomplish this is to form ribs on the elements. Fig. 8 shows element **33** provided with a series of cross or transverse ribs **33***a* on its widest sides while element **36** of Figs. 9 and 10 has longitudinally extending or vertical ribs **36***a* on all its sides.

Element **33** is shown in Fig. 8 as having rectangular ends **33***b* loosely to fit apertures **31***a* in end members or plates **31** of Fig. 7. However, it is obvious that, if desired, the transverse ribs **33***a* may be formed throughout the length of element **33** without interference with its mounting in the frame of the unit or its use when so mounted. In a similar manner the longitudinal or vertical ribs **36***a* of element **36** of Figs. 9 and 10 may be extended to the very ends of the element, thus eliminating the rectangular ends **36***b* shown thereon, in which case the apertures **31***a* in members **31** could be made, if desired, to conform to the cross section of element **36** shown in Fig. 10. By providing rectangular apertures **31***a* in end members or plates **31** both elements **33** and **36** in any desired distribution may be mounted in the same unit. As in the previously described forms of the invention the elements **33** and **36** will be secured at one end to one of the end members or plates **31** while their other ends are freely slidable in the apertures **31***a* of the other end member or plate.

The various parts making up the herein described units may be of any suitable or desired materials, catalytically active or inactive, and if absorbent, of any suitable or desired degree of porosity, so as to assist or to promote any desired type of operation or reaction. One kind of material particularly adapted to units of the present invention is ceramic material, such as porcelain. The latter can be readily molded, cast or extruded into the forms illustrated and can by composition and subsequent firing be made of any desired degree of porosity in the range of substantially no or slight porosity to about 60% porosity. By firing into the range of 2300 to 3900° F. it can be made stable both physically and chemically as well as catalytically inert. It is then useful as a support, in or on which catalytic materials can be incorporated in any known or suitable manner as by dipping, immersing, spraying, etc. Bonding of the elements such as **13**, **23**, **33** and **36** to one of the end members or plates of the frame is frequently incidental to incorporating or applying catalytic materials to the assembled unit, as by dipping the latter up to but not including the top end member or plate (as **22** of Fig. 3), followed by heating step to decompose the solution or mixture so as to deposit the catalytic material in active form in or on the frame and elements mounted therein. On the other hand, the individual parts making up the units may have catalytic material incorporated therein or deposited thereon prior to assembly of the unit.

The unit can be made of any desired size and the end members or plates may be shaped as required to fit, or for use in, a particular reaction chamber. The elements may be in any desired number, form and arrangement. A square or rectangular shape for the end members or plates, as shown in Figs. 2, 4 and 7, is desirable for stacking or side-by-side disposition of units. Of the units shown on the drawings, that of Figs. 1 and 2 makes provision for **196** round elements or rods **13**, the modification of Figs. 3 and 4 provides for **73** streamlined elements or rods **23**, while the modification of Fig. 7 will hold **63** elements or rods such as **33** and **36**. One use of units of the type of Figs. 1 and 2 is set forth in my copending application Serial No. 117,490 filed September 23, 1949.

In general in utilizing the unit as a support for catalysts, a firmly adherent base or film of active alumina (or alternatively of magnesia, beryllia or thoria) may be applied in any suitable or desired manner to the unit or its parts followed by impregnating such base or film with other and additional finely divided metals or oxides. Such a combination and arrangement of catalytic materials has been found to be characterized in many instances by exceptionally high activity for the promotion of many reactions, both endothermic and exothermic, and to be further characterized by long life. For example, for catalytic reactions of the oxidation type, such as oxidation of CO to CO_2, the base film of alumina or the like should be impregnated with platinum, or silver, or copper; for oxidation of SO_2 to SO_3, platinum or vanadium oxide should be deposited on the base film; for synthesis of methane by the reaction of $CO+3H_2=CH_4+H_2O$, cobalt or nickel should be added to the base film of active alumina or the like.

If the unit is to be used to promote a hydrogenation reaction, such as the hydrogenation of fats, a nickel catalyst should be deposited on the base film of active alumina. When the nickel hydrogenation of fats is in the liquid phase, the parts of the unit should be formed of porcelain or the like which is practically impervious or of extremely low porosity so that only the catalytically active material deposited on the surface of the parts of the unit will contact the oil or fats undergoing hydrogenation. Separation of reactants can then be easily effected with obvious economies and advantages.

Both oxidizing and hydrogenating reactions being exothermic, the liberation of heat diminishes as the reactants progress through the reaction zone. Consequently to effect a uniform operation the rate of reaction should be adjusted as the reaction progresses. Control of the rate of reaction per unit volume of the reaction zone is readily accomplished by adjusting the number of elements in the catalytic units in accordance with the location of the units within the reaction zone. Thus the first unit, or first group of units, to be contacted by the reactants in an exothermic operation would be provided with fewer elements and/or in a more open arrangement than subsequent units so as to adjust the extent and rate of the reaction.

For catalytic cracking of petroleum, active alumina is first deposited on the parts of the unit followed by a deposition of silica; and for aromatization of naphtha, finely divided molybdenum oxide is deposited upon a base film of alumina. In these operations which are both endothermic the rate of reaction changes as the hydrocarbon vapors flow through the catalytic zone, with the result that coke is not evenly deposited on the catalyst, with the further result that during regeneration, effected by burning off the coke, heat is liberated unevenly, thus rendering control of temperature of the catalyst difficult. Here again adjustment of the number and arrangement of elements mounted in the contacting units of the present invention will be effective in establishing proper time and extent of contact to control the rate of reaction.

The above examples indicate some of the catalytic reactions for which the contacting unit may be employed; others are synthesis of alcohol, oxidation of acetylene to acetone, dehydration of ethyl alcohol, hydrogenation of fuels, destructive hydrogenation of mineral oils, desulphurization of crude oils in liquid phase, chlorination reactions, etc., and still others which will occur to those skilled in the art of catalysis.

From the above it will be apparent that the present invention provides a contacting or catalytic unit in which contacting or catalytic elements may be fixedly mounted so that each element is independent of the other, thereby avoiding friction and consequent damage to the elements and partial or complete destruction of catalysts impregnated therein or applied thereto in the form of films, that the elements are not in compact relationship with one another but are spaced from one another so that low pressure drops through the units are easily obtained and the problem of excessive temperatures can be solved by adjusting the number and distribution of elements in each unit, and that the units and elements therefor may be formed and arranged to produce a minimum of turbulence and contact or great turbulence and extensive contact thereby effectively to control the rate and extent of a catalytic or contacting reaction.

While the invention has been herein shown and described in connection with certain preferred forms of apparatus, it is to be understood that the invention is not restricted to the specific features thereof but covers all changes, modifications and adaptations within the scope of the appended claims.

I claim as my invention:

1. A catalytic unit through which a fluid may flow comprising a pair of end members secured to one another in spaced relation by a single post extending between said members and to which said members are rigidly fastened, and a plurality of elongated elements extending between said end members and supported at both ends thereby, each of said elements providing a surface of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

2. A catalytic unit according to claim 1 in which said elements are of ceramic material.

3. A catalytic unit according to claim 1 in which the surfaces of catalytic material are provided by coatings on said elements.

4. A catalytic unit through which a fluid may flow comprising a pair of end members secured to one another in spaced relation by a single post extending between said members and to which said members are rigidly fastened, and a plurality of elongated elements extending between said end members and supported at both ends thereby, said elements being disposed in rows with the elements in each row in staggered relation with the elements in adjacent rows, and each of said elements providing a surface of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

5. A catalytic unit through which a fluid may flow comprising a pair of end members secured to one another in spaced relation by a single post extending between said members and to which said members are rigidly fastened, and a plurality of elongated elements extending between said end members and supported at both ends thereby, each of said elements having at least one end free to move longitudinally with respect to an end member, each of said elements providing a surface of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

6. A catalytic unit through which a fluid may flow comprising a pair of end members secured to one another in spaced relation by a single post extending between said members and to which said members are rigidly fastened, and a plurality of elongated elements extending between said end members and supported at both ends thereby, each of said elements having in transverse section rounded and tapered portions forming a streamlined contour to minimize turbulence in the flowing fluid, and each of said elements providing a surface of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

7. A catalytic unit through which fluid may flow which comprises a pair of platelike end members, a single post for rigidly positioning said end members in parallel, spaced relaionship, said end members being centrally fastened to said post at either end thereof, a plurality of elongated elements extending between said end members and supported at both ends thereby, at least one end of each of said elements being free to move longitudinally with respect to one of said end members, each of said elements providing a surface of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

8. A multi-unit catalytic assembly for providing an extended area of catalytic surface, each unit of said assembly comprising a pair of end members secured to one another in spaced relation by a single post extending between said members and to which said members are rigidly fastened, and a plurality of elongated elements extending between said end members and supported at both ends thereby, each of said elements providing a surface of catalytic material exposed to the flow of a fluid passing through the unit, said units being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

9. A catalytic unit for carrying out high temperature catalytic reactions which comprises a pair of plate-like end members composed of ceramic material, a single post composed of ceramic material for rigidly positioning said end members in parallel, spaced relationship, said end members being centrally fastened to said post at either end thereof, a plurality of elongated, rod-like, elements composed of ceramic material extending between said end members and supported at both ends thereby, at least one end of each of said elements being free to move longitudinally with respect to one of said end members, each of said rod-like elements being provided with a relatively thin film of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

10. A unit in accordance with claim 9 in which said end members, said post, and said rod-like elements are composed of a dense, relatively impervious porcelain.

11. A ceramic unit for use in carrying out high temperature reactions which comprises a pair of plate-like end members of ceramic material, a single post of ceramic material for rigidly positioning said end members in parallel, spaced relationship, said end members being centrally fastened to said post at either end thereof, a plurality of elongated rod-like elements composed of ceramic material extending between said end members and supported at both ends thereby, at least one end of each element being free to move longitudinally with respect to one of said end members, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

12. A unit constructed in accordance with claim 11 in which said end members, said post, and said rod-like elements are composed of a relatively dense and impervious porcelain.

13. A unit constructed in accordance with claim 11 in which said rod-like elements have in transverse section rounded and tapered portions forming a streamlined contour to minimize turbulence in the fluid flowing through said unit.

14. A catalytic unit adapted for high temperature catalytic reactions which comprises a pair of plate-like end members composed of ceramic material, a single post composed of ceramic material for rigidly positioning said end members in parallel, spaced relationship, said end members being centrally fastened to said post at either end thereof, a plurality of elongated rod-like elements extending between said end members and supported at both ends thereby, said elements being disposed in rows with the elements in each row in staggered relation with respect to the elements in adjacent rows, each of said elements having in transverse section rounded and tapered portions forming a streamlined contour to minimize turbulence in the flowing fluid, at least one end of each of said elements being free to move longitudinally with respect to one of said end members, each of said rod-like elements being provided with a relatively thin film of catalytic material exposed to the flow of fluid passing through the unit, said unit being open laterally for the passage of fluid therethrough in a direction transverse to said elongated elements.

15. A unit constructed in accordance with claim 14 in which said elongated rod-like elements are supported in apertures provided in said plate-like end members.

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