Aug. 29, 1950 V. MILLS ET AL 2,520,424

CONTINUOUS HYDROGENATOR

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FIG. 1.

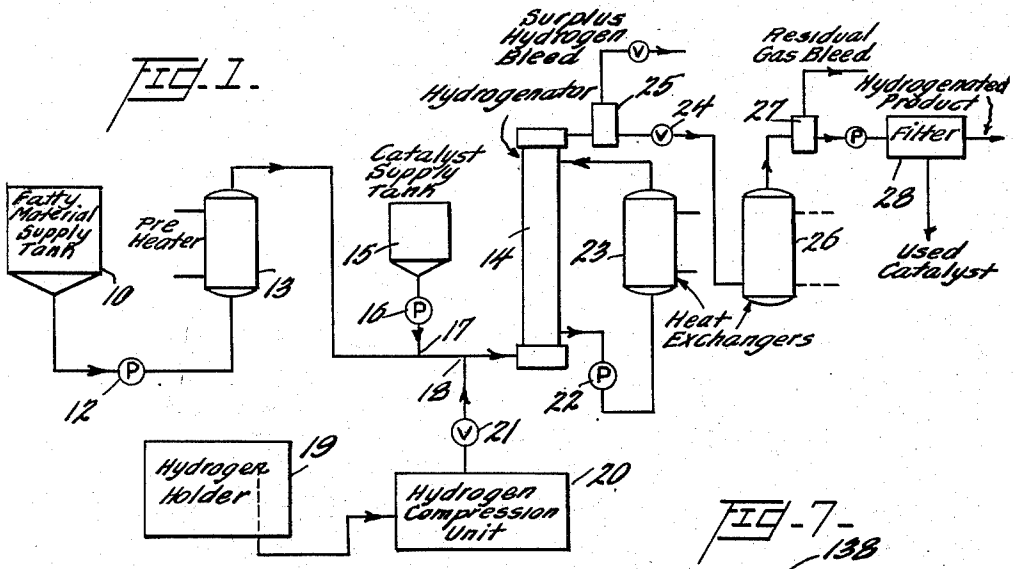


FIG. 4.

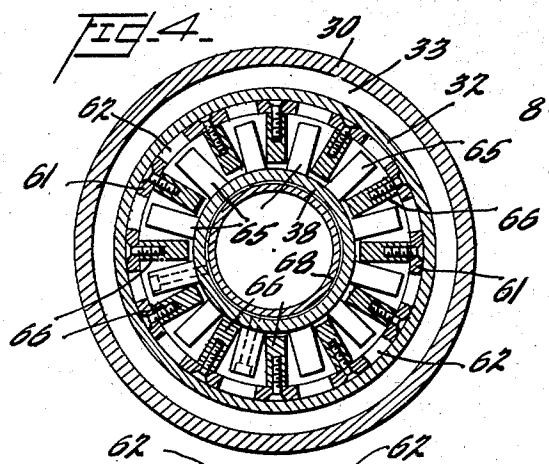


FIG. 7.

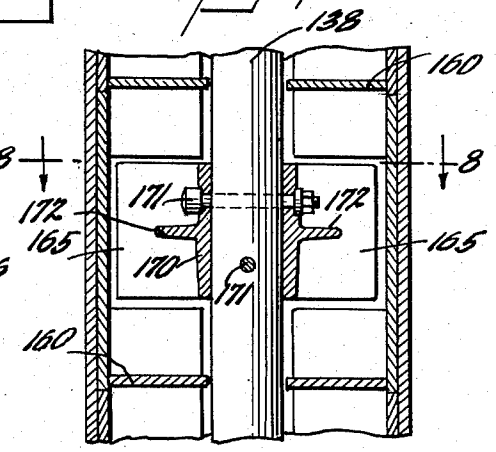


FIG. 5.

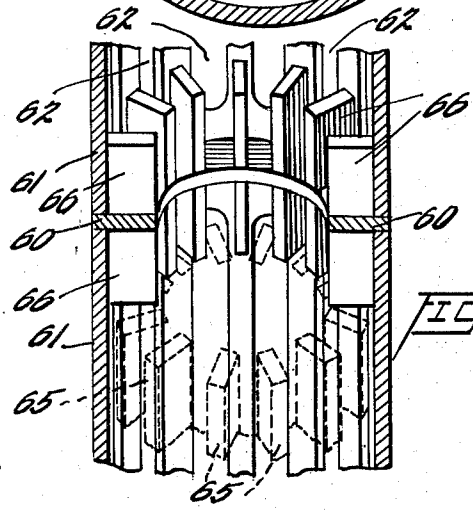
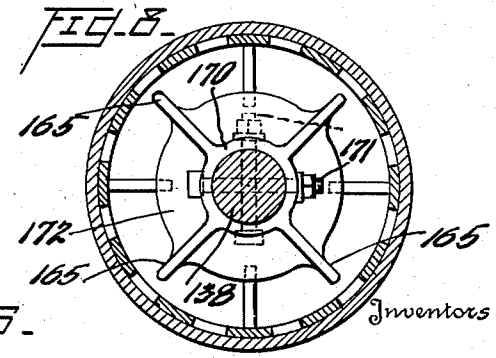


FIG. 8.



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FIG. 2.

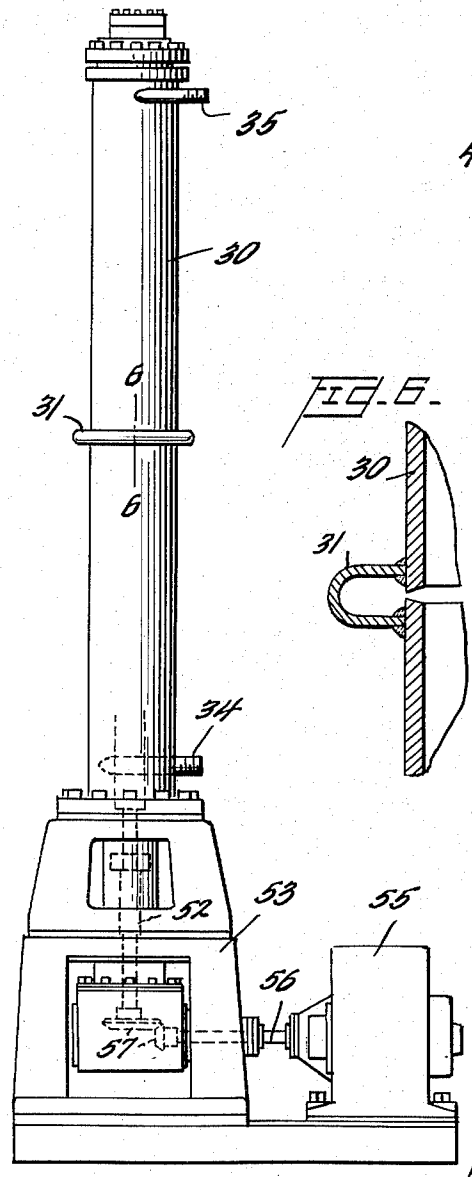


FIG. 6.

FIG. 3.

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CONTINUOUS HYDROGENATOR

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Application May 2, 1947, Serial No. 745,662

4 Claims. (Cl. 23—285)

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This invention relates to apparatus suitable for use in continuous hydrogenation of unsaturated liquid substances, i. e., for addition of hydrogen to carbon to carbon double bonds in a continuous process.

An object of the invention is to provide such intense agitation in the hydrogenation chamber of apparatus of this character as to permit the reaction to be carried out at an exceptionally rapid rate, thereby effecting economies in operation and improvements in the quality of the product.

Another object is to provide means of removing from the liquid undergoing hydrogenation the heat of reaction as fast as it is liberated, thereby enabling the reaction rate to be kept relatively constant as the liquid progresses through the hydrogenation chamber, thus facilitating control of the desired end point of the reaction.

Another object is to provide means of preventing "channeling" of unhydrogenated liquid to the outlet of the hydrogenation chamber, thereby avoiding contaminating the finished hydrogenated product with raw or incompletely processed material.

Another object is to provide a complete assembly of apparatus particularly adapted to continuous hydrogenation of unsaturated liquid materials in a practical and easily controllable manner.

Other objects are to provide the foregoing features in apparatus of sound engineering design, suitable for economical commercial use.

The apparatus of the present invention is designed for continuous hydrogenation of unsaturated materials such as higher fatty acids and their esters, particularly unsaturated natural glyceride oils, in that form of practice which employs a continuous liquid phase with dispersions of finely divided catalyst and of hydrogen gas throughout this liquid phase. Continuous hydrogenation processes which may be carried out in the apparatus of the present invention are described and claimed in applications Serial No. 745,660 and Ser. No. 745,661, filed concurrently herewith by Mills, Sanders and Hawley.

Two forms of hydrogenators which meet the requirements of our invention, together with a flow chart of a hydrogenation system employing such hydrogenators, are illustrated in the accompanying drawings, in which

Figure 1 is a schematic flow chart showing the principal elements of a typical continuous hydrogenation system;

Figure 2 is a side elevation of a mechanically agitated continuous hydrogenator vessel;

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Figure 3 is a vertical section of the vessel shown in Figure 2;

Figure 4 is a horizontal section of the vessel, taken on the line 4—4 of Figure 3;

Figure 5 is a fragmentary vertical section of the inner chamber of this same vessel, showing in perspective some of the hold-back baffles, or stators, and one of the horizontal baffles;

Figure 6 is a fragmentary sectional view taken on the line 6—6 of Figure 2;

Figure 7 is a fragmentary elevation, partly in section, illustrating a modification of the agitator shaft and blade structure; and

Figure 8 is a horizontal section taken on the line 8—8 of Figure 7.

The several elements of apparatus indicated schematically in Figure 1, and their respective purposes and functions as part of a complete continuous hydrogenation system, will be understood from the following description of a preferred mode of carrying out the process.

The unsaturated liquid material to be hydrogenated is delivered from supply tank 10 by means of pump 12 through a tubular preheater 13 to and through the hydrogenator 14. A suitable hydrogenation catalyst suspended in a small quantity of a suitable liquid (which may often be a portion of the material to be hydrogenated) is delivered from catalyst supply tank 15 by means of pump 16 either directly into the hydrogenator, or (as shown) into the pipeline which is delivering the main supply of material to be hydrogenated from the preheater to the hydrogenator, for example, at point 17. A continuous supply of hydrogen, or a suitable hydrogen-containing gas, is introduced into the same pipeline at another point ahead of the hydrogenator, for example at point 18, the hydrogen supply being drawn from a suitable reservoir or supply, as illustrated at 19, by means of compressor 20 through a pressure regulating valve 21. While flowing through the hydrogenator 14, the mixture of the liquid material to be hydrogenated, the catalyst, and the hydrogen is subjected to violent agitation to bring these three materials into intimate contact with one another and to bring about a rapid movement of the individual particles of each of the non-liquid phases in contact with the particles of the liquid phase, thus promoting a high velocity of the hydrogenation reactions which occur in this vessel. The heat of reaction which is liberated is preferably (especially when only partially hydrogenating an unsaturated material) completely, or at least for the most part, removed continuously by circulating a cooling medium through a jacket surround-

ing the reaction space in the hydrogenator, the cooling medium being circulated by means of pump 22, and the heat being removed from the cooling medium in heat exchanger 23. The reaction mixture passing through hydrogenator 14 is maintained at superatmospheric pressure, and this pressure may conveniently be regulated by means of the adjustable relief valve 24 in the outlet line leading from the hydrogenator. If a surplus of hydrogen is used, over that which reacts with the oil and that which remains dissolved in the liquid leaving the hydrogenator, this surplus may be separated and bled off through the top of small tank 25. The liquid outlet of this tank is provided with a float valve adapted to close this outlet against the discharge of gas. The hydrogenated material leaving the hydrogenator is cooled by means of heat exchanger 26, and any remaining gas which has come out of solution subsequent to the drop in pressure at valve 24 is then separated from the hydrogenated material in small tank 27, and the catalyst is removed from the oil, for example by means of the filter 28.

The hydrogenator 14 of the system just described may be constructed as shown more particularly in Figures 2 to 6, inclusive, in which is illustrated a preferred type of hydrogenating apparatus. Thus the hydrogenator may comprise an outer jacket 30, provided with an expansion coupling 31 to relieve strains caused by temperature changes, and an inner cylinder 32, the jacket and cylinder defining an annular space 33 within which is circulated a fluid coolant, the inlet and outlet conduits for the coolant being shown in Figure 2 at 34 and 35 respectively. A hollow shaft 38, of substantially less diameter than cylinder 32 is disposed coaxially within the cylinder and supported for rotation about its vertical axis, shaft 38 and cylinder 32 defining an annular reaction passage in which the mixture of the liquid fatty material, the hydrogen, and the catalytic agent, is intensely agitated while flowing in an upward direction, being introduced through an inlet passage 39 formed in an annular plate 40 at the lower end of cylinder 32, and discharging through an outlet passage 42 in an annular plate 43 at the upper end of cylinder 32.

The hydrogenator is closed at its upper end by a cap structure comprising plate 43 and closure members 45 and 46, the several parts being bolted together as shown more particularly in Figure 3. A radial thrust bearing 48, seated in member 45 and retained in position by member 46 engages and supports a shaft 49 which extends within and is secured to the hollow shaft 38, whereby the latter is journaled for rotation. Received within and secured to shaft 38 at its lower end is a coupling element 51; a drive shaft 52, disposed coaxially of shaft 38, extends within and is secured for rotation with coupling element 51. Drive shaft 52 is journaled for rotation in the supporting base structure indicated generally at 53, the hydrogenator being suitably mounted on this base structure. A motor 55 having a shaft 56 drives shaft 52 through bevel gearing 57, whereby the hollow shaft 38 is rotated rapidly; variable speed gearing may be incorporated in the motor housing in order that the speed of the shaft 38 may be appropriately selected. It will be appreciated that the details of this construction may be varied widely.

The annular reaction passage between shaft 38 and cylinder 32 is divided into a series of compartments or reaction zones by means of a plurality of horizontally disposed annular disks or baffles 60, the baffles being spaced longitudinally of the hydrogenator. The outer diameter of each baffle 60 is such that the baffles fit snugly within cylinder 32, the inner diameter being slightly larger than the outer diameter of shaft 38, so as to afford slight mechanical clearance therebetween. The reacting materials flowing upwardly are thus caused to flow through the restricted annular passages defined between shaft 38 and baffles 60 in moving from each compartment or reaction zone to the next higher zone, retention of the materials in each zone for a substantial length of time being assured. "Channeling," or too rapid movement of insufficiently reacted materials through and out of the hydrogenator, is thereby avoided. The baffles 60 may be retained in proper spaced relation by a series of spacing sleeves, the several sleeves fitting snugly within cylinder 32, so that each of the baffles is clamped between an adjacent pair of sleeves 61. Each of the sleeves 61 may be formed with longitudinally extending slots 62, as shown more particularly in Figures 3 and 5, to reduce the weight of the sleeves and to increase the heat transfer surface and the volume and capacity of the several reacting zones. An efficient hydrogenator may be provided with as many as 11 baffles, or even more, so as to provide 12 or more reacting zones, but the number of zones may be varied widely.

It will be appreciated that while baffles 60 are conveniently mounted on the cylinder 32, they may be mounted, if desired, on shaft 38, clearance for the passage of liquid being afforded between the periphery of the baffles 60 and the cylinder 32. The structure shown in the drawings is preferred, however, since the retarding effect on the axial flow of liquid is greater.

In order to effect intense agitation of the material, each zone may be provided with a series of agitator blades 65, and with cooperating stator blades or stationary baffles 66 located above and below the agitator blades. The agitator blades 65 are disposed radially of and are bolted securely to the hollow shaft 38 in circumferentially spaced relation, one such series of agitator blades being shown in dotted lines in Figure 5. In order to prevent leakage of the reacting materials past the securing bolts to the interior of the shaft 38, a sleeve 68 extending within and over the major portion of the length of shaft 38, is welded to the latter at each end. The circumferentially spaced stator blades 66 are likewise disposed radially of the axis of shaft 38, and are bolted or are otherwise secured to the sleeves 61 adjacent to each of the annular disks 60, preferably intermediate the slots 62 formed in sleeve 61, as shown in Fig. 5. It will be observed that each reaction zone is provided with one series of agitator blades 65 and two series of cooperating stator blades 66, the latter acting to resist continuous swirling movement of the reacting materials about shaft 38 and otherwise serving to increase the degree of agitation imparted to the materials. Preferably, blades 65 and 66 in adjacent series are so dimensioned as to afford only the necessary mechanical clearance between each other; similarly, only mechanical clearance is afforded between the stator blades 66 and the shaft 38 and between the agitator blades 65 and the sleeves 61. As a rule we prefer to employ stator blades projecting inwardly from the wall of the vessel at least 85

per cent of the distance from wall to shaft, and similarly to employ agitator blades extending outwardly from the shaft at least 85 per cent of the distance from shaft to wall.

In the particular hydrogenator illustrated, we effectively avoid undue contamination of the finished product with raw oil or with insufficiently hydrogenated oil by a combination of two provisions: First, the relatively long passage through which the reaction mixture passes on its way from the entrance to the exit of the hydrogenator, the flow through this passage being interrupted repeatedly by a series of transverse agitators separated by a corresponding series of stator blades tending to break up swirling induced by the agitators; and second, the horizontal circular baffles, elements **60**, which subdivide the reaction zone into a plurality of lesser zones each communicating with the adjoining one by means of a passage of greatly restricted cross-sectional area.

The horizontal circular baffles, elements **60**, although preferred features of the hydrogenation vessel, may if desired by omitted entirely. In this event "channeling" of insufficiently hydrogenated material to the outlet is to a large extent avoided, although not to the full extent preferred for some purposes, when the length of the chamber from inlet to outlet is such as to permit the employment of at least 10 to 15 separate sets of agitators, each set separated from the next by a cooperating set of stator blades.

By providing the mechanical agitation in a direction which is predominantly transverse (preferably perpendicular or even somewhat counter) to the main direction of flow of the liquid being hydrogenated, the agitator-impelled transfer of unprocessed raw material towards the outlet of the vessel is minimized. This may be accomplished by aligning the pressure resisting surfaces of the agitator blades and of the stator blades so that they face in a direction generally transverse to, and not tending towards, the general direction of liquid flow. While the blade surfaces thus define planes substantially parallel to the general direction of liquid flow, slight inclination is of course permissible, and inclination tending to retard axial flow may in some cases be desirable.

When the chief reliance for the avoidance of "channeling" is placed in horizontal baffles such as elements **60**, with a minimum of agitator blades—i. e. when each of the lesser zones between two horizontal baffles is provided with just one set of transverse agitators—"channeling" is to a large extent avoided, although perhaps not to the full extent preferred for some purposes, by thus subdividing the hydrogenation space into at least six lesser zones. The interconnecting passages between the lesser reaction zones of the hydrogenation chamber are preferably so greatly restricted in cross-sectional area that the agitation occurring in one of these zones is not felt to an appreciable extent in the adjoining zone. Our circular baffles, **60**, which in a preferred case leave an annular passage around the shaft just one eighth of an inch wide in a hydrogenator having an internal diameter of 8 inches and a shaft diameter of 4 inches, accomplish this purpose.

The hydrogenator used in our process is designed to provide for efficient removal from the reactants of large quantities of heat, since the heat generated by the reaction amounts to about 2 B. t. u. per pound of oil hydrogenated per unit of iodine value drop. In the hydrogenator illustrated in the drawings, the heat transfer surface is large in relation to the cubical contents of the vessel, and the heat transfer coefficient is high because of the violent agitation.

A very high degree of agitation is provided in the hydrogenator illustrated by the cooperation of the moving agitator blades and the fixed vertical baffles. Preferably the clearance between these blades and baffles does not exceed about three sixteenths to one fourth of an inch, and the preferred speed of the outer edges of the rotors is of the order of 6 feet per second. The use of fixed vertical baffles, or stators, to retard the swirling action of the liquid, which would otherwise be induced by the rotating agitators, is important particularly in order to avoid a centrifugal effect which would tend to move the suspended catalyst towards the outer walls of the vessel and would simultaneously tend to move the gas bubbles towards the center of the vessel.

In Figures 7 and 8 is shown a modified form of the invention in which the agitator shaft **138** is solid. This permits a decrease in the shaft diameter and a corresponding increase in the horizontal cross-sectional area of the free path traveled by the liquid in moving from the entrance to the exit of the hydrogenation chamber, without increase in the diameter of the vessel. In this modification the agitator blades **165** are formed integrally with a sleeve **170**, the sleeve being dimensioned to fit snugly on the shaft and being retained thereon by bolts **171**. Radial fins **172** are provided, these fins serving to retard axial flow of liquid and to reinforce and strengthen the agitator structure. It will be observed that four blades are shown; the same number of blades may be employed in each of the adjacent stator units, but the number of stator blades may be greater or less. When using this form of vessel "channeling" is quite effectively prevented when the hydrogenation zone is subdivided into 12 lesser zones by means of horizontal baffles **160**, although fewer than this number will suffice when substantially complete avoidance of contamination of the finished product is not required.

It will be appreciated that many variations from the design of apparatus illustrated in the accompanying drawings may be employed without departing from the spirit or scope of the invention. Thus, the oil and catalyst inlet may be placed near the top of the hydrogenator and the oil and catalyst outlet at the bottom, with the hydrogen inlet remaining near the bottom, a design which may be employed to advantage when the complete saturation of the oil is desired. Another modification is to mount the hydrogenator horizontally instead of substantially upright, in which case each of baffles **60** (now vertical) is provided with an opening along its upper edge to permit passage of hydrogen along the length of the vessel.

When employing the apparatus herein illustrated and described under typical hydrogenation conditions, with the flow of liquid, suspended catalyst, and hydrogen passing at uniform rates in contact with one another through the reaction vessel under conditions of extreme turbulence, the temperature of the liquid is established at a chosen value by adjusting the pressure of the steam or the temperature and rate of flow of other heating medium through the jacket of the liquid preheater, and this temperature is maintained with but little or no rise as the liquid passes through the hydrogenator by controlling the temperature or rate of flow of

the water or other cooling medium through the jacket surrounding the hydrogenator. When a relatively small overall iodine value drop is being brought about and consequently the heat liberation is small in amount, it may not be necessary or desirable to remove more than about two thirds of the heat of reaction, although when the total iodine value drop is 10 units or more we prefer to keep the temperature of the liquid practically constant as it progresses through the reaction vessel, with a preferred maximum temperature rise of not more than 10° to 15° C. An exception to this preferred temperature control is in the complete hydrogenation of a material, in which case the control of the end point is relatively simple and the temperature may be allowed to rise if desired.

The degree of reaction, or completeness of hydrogenation (as measured from time to time by determining the iodine value, or the refractive index, or the congeal point, or the titer, or other index, of samples of the hydrogenated liquid withdrawn from the system after the reaction has ceased) may conveniently be controlled by regulating the rate of introduction of the liquid to be hydrogenated. As this liquid rate is varied one may simultaneously and correspondingly vary the rates of feed of the catalyst slurry and the hydrogen, thus maintaining constant proportions of catalyst and hydrogen in relation to the liquid feed rate, or one may alternatively keep one or both of these secondary feed rates constant, or vary it independently of the liquid feed rate, thus gaining an independent or supplementary control over the extent of the hydrogenation which occurs. The amount of hydrogen which is supplied to the hydrogenator should of course be adequate for the degree of hydrogenation desired, and may conveniently be somewhat in excess of this amount in order to insure utilizing the maximum hydrogenating capacity of the hydrogenator.

Having thus described our invention, what we claim and desire to secure by Letters Patent is:

1. In apparatus for continuous hydrogenation of unsaturated liquids, the combination with an elongated generally cylindrical hydrogenation vessel member having inlet and outlet passages at opposite ends thereof, whereby hydrogen, a hydrogenation catalyst, and a liquid to be treated may be passed through said vessel member, said vessel being encircled by a cooling jacket, of an agitator shaft member extending centrally within and lengthwise of said vessel member and having an outside diameter not less than half the internal diameter of the vessel member, means journaling said shaft member for rotation, a plurality of outwardly directed, axially spaced agitator blades carried by said shaft member, a plurality of inwardly directed, axially spaced stator blades carried by said vessel member, said stator blades extending between adjacent, axially spaced agitator blades, said stator and agitator blades having the pressure surfaces thereof disposed generally axially of the vessel member and a plurality of transverse baffle plates carried by one of said members and extending transversely and substantially radially toward and into close proximity to the other of said members to afford restricted passages therebetween, said baffle plates being annular in form and being axially spaced to define with said members a plurality of annular zones in restricted communication, whereby channeling of unhydrogenated liquid to the outlet of said vessel member is prevented.

2. Apparatus as claimed in claim 1, in which said stator and agitator blades have an effective width, measured radially of the vessel member, which is at least 85% of the difference between the external radius of the agitator shaft and the internal radius of the vessel.

3. Apparatus as claimed in claim 1, in which the axial spacing between adjacent agitator and stator blades is not substantially in excess of that required for mechanical clearance.

4. Apparatus as claimed in claim 1, and comprising variable capacity pumping means adapted to force continuous controlled streams of unsaturated liquid, of hydrogenation catalyst slurry, and of hydrogen into and through said vessel member, and a heat exchanger for preheating said stream of liquid to hydrogenating temperature.

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