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METHOD AND MEANS FOR CONVERTING HYDROCARBON IN THE PRESENCE
OF HYDROGEN IN THE CONVERSION AND REGENERATION ZONES

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FIG. I

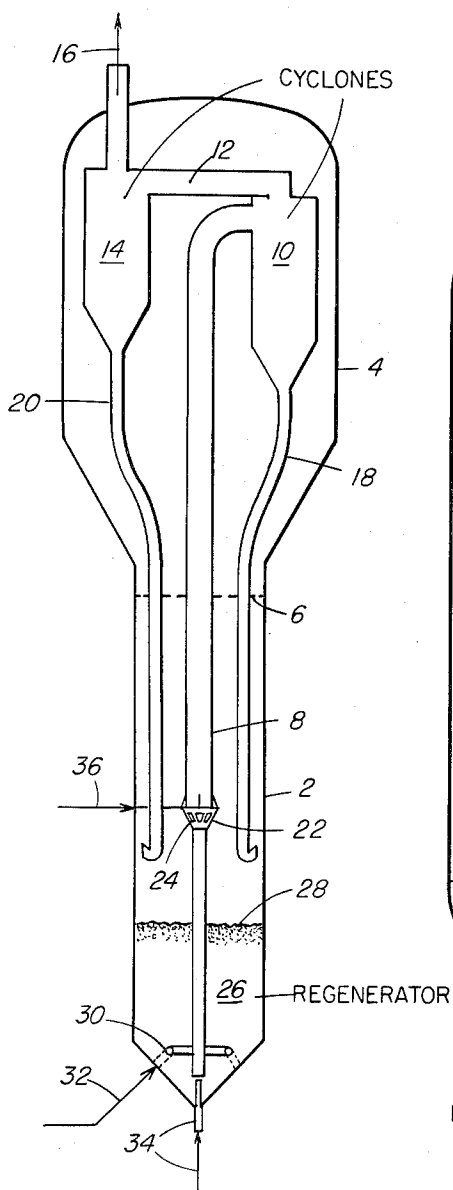
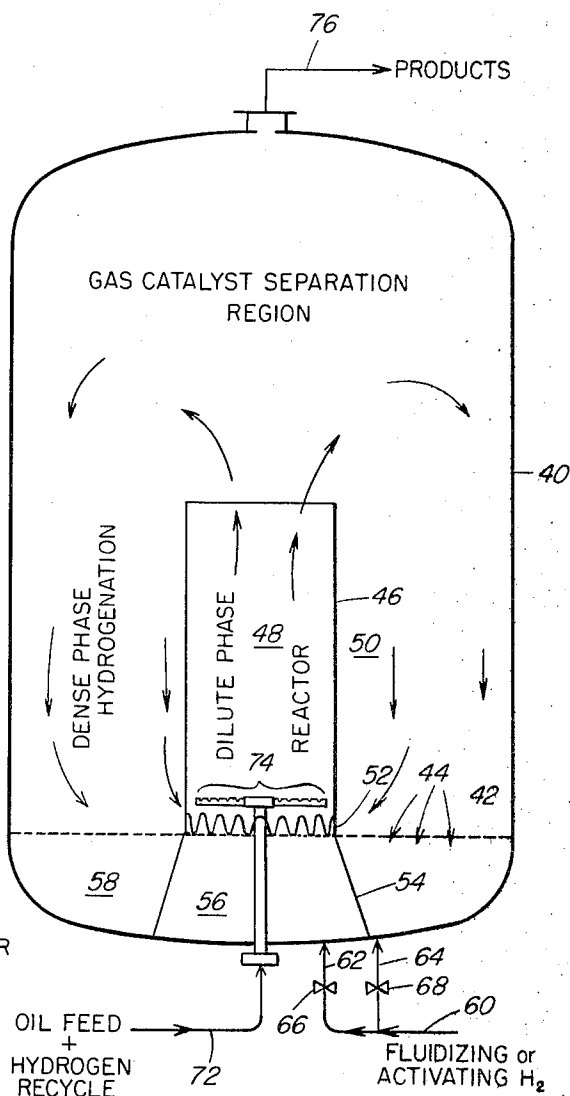


FIG. II



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METHOD AND MEANS FOR CONVERTING HYDROCARBON IN THE PRESENCE OF HYDROGEN IN THE CONVERSION AND REGENERATION ZONES

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This invention relates to the conversion of hydrocarbonaceous material and in a particular aspect relates to the hydrogenative conversion of hydrocarbonaceous material in the presence of a hydrocracking catalyst.

Various processes have been proposed for converting hydrocarbons to desired boiling range hydrocarbon products including gasoline, fuel oils and other suitable saleable product materials. A large portion of these processes involve the conversion of the hydrocarbon in the presence of particulate contact material on which a hydrocarbonaceous deposit is formed by the hydrocarbon undergoing conversion and which must be removed from time to time to permit continued use of the contact material. Generally speaking, the heavier or higher boiling the feed hydrocarbons are, the greater is the amount of carbonaceous deposits formed on the contact material; this deposition is also influenced by the severity of the conversion process employed. The cost of regenerating the contact material is an important factor in the operating costs of a hydrocarbon conversion process and unless the contact material passed thereto is properly stripped to recover available entrained hydrocarbonaceous material, desired economies of the process are difficult to maintain.

An object of this invention is to convert hydrocarbonaceous material to desired product material in the presence of hydrogen-rich gaseous material.

A further object of this invention is to provide an improved method and arrangement of process steps for catalytically converting hydrocarbonaceous material under destructive hydrogenation conditions.

Other objects and advantages of this invention will become more apparent from the following description.

In accordance with this invention, finely divided solid particulate material is caused to circulate in a unitary system comprising a reaction zone and a regeneration zone in the presence of a hydrogen containing gaseous material and under operating conditions sufficient to convert and recover hydrocarbonaceous product material therefrom. In a more particular aspect, a method and arrangement of contact steps is provided for effecting the catalytic hydrocracking or destructive hydrogenation of a hydrocarbon feed material under desired conversion conditions in conjunction with hydrogenative treatment of the catalyst to remove hydrocarbonaceous material deposited thereon sufficiently to permit reuse of the catalyst in the process. Accordingly, the method of this invention relates to a hydrogenative-hydrocracking process which is characterized by generally lower operating pressures, relatively short hydrocarbon feed contact times and optimized temperature conversion condition. Hydrogenative treatment to effect catalyst regeneration is accomplished by passing hydrogen-rich gas in contact with the catalyst at a pressure equal to or above reaction pressure and generally at temperatures higher than reaction temperatures. Accordingly, the method of this invention may be broadly construed as including a two-page hydrocracking process or hydrogenative process wherein catalyst is circulated between zones of substantially different conversion severity wherein hydrogen-rich gas may be introduced separately

to each zone or passed first to the zone of maximum severity for sequential flow through the zones and generally countercurrent to the flow of catalyst bearing hydrocarbonaceous material passing through the contact zones. In another aspect, the process of this invention may be viewed as one in which catalyst and hydrocarbonaceous material is caused to flow through a plurality of hydrogenative and/or hydrocracking zones maintained under conditions of desired severity and in at least one embodiment thereof the catalyst flows generally countercurrent to the flow of hydrogen-rich gases passing through the zones with the catalyst leaving the zone maintained under maximum severity conditions being suitable for returning or recycling directly to the initial hydrocarbon feed contact zone in the series of zones.

In the dense fluid bed—dilute phase catalyst system of this invention approximately constant temperatures may be maintained between zones or one zone may be maintained at a substantially higher temperature than the other zone. Furthermore, allowing a temperature rise of about 100° F. or more in the dilute phase reactor due to heat of reaction is possible depending on the conversion conditions desired and/or employed therein and the hydrocarbon feed material being treated. Generally speaking, a desired product distribution of the hydrocarbon feed being converted will at least in part dictate the temperatures employed in the method and means of this invention. In addition, the activity and intrinsic selectivity of the catalyst bears importantly on the temperatures employed.

In the fluid system and method of operation herein described, the heat of reaction may be easily controlled and relatively quickly absorbed by adjusting the temperature, for example, of the hydrocarbon feed and hydrogen-rich streams introduced thereto. Accordingly, when practicing the method of this invention, the conversion of the hydrocarbon feed material to more valuable products is carried out on the one hand at a relatively low hydrogen partial pressure consistent with maintaining relatively mild hydrocracking conditions in the fresh feed hydrocarbon reaction zone and thereafter the circulating catalyst is contacted under generally more severe hydrogen partial pressure conditions in the absence of added fresh feed and sufficient to effect hydrogen regeneration of adsorbed hydrocarbonaceous material retained with the catalyst. Accordingly, hydrogen partial pressures in the range of from about 300 p.s.i. to about 2,000 p.s.i., preferably from 500 to about 1,000 p.s.i. are maintained in the unitary system encompassing the hydrogen treating steps of this invention.

As suggested herein, the initial time of contact of the fresh hydrocarbon reactant with the catalyst in the dilute phase conversion zone, the temperature employed, and hydrogen consumption rate will vary considerably dependent upon the hydrocarbon feed being introduced, the extent of hydrocarbon conversion desired and the activity of the catalyst being employed therein. This invention contemplates employing catalysts varying considerably in their activity, either alone or in admixture with one another, as separate and/or individually discrete particles or intimately combined physical mixtures thereof. Furthermore, the dilute phase contact time of the hydrocarbon with catalyst in the riser reactor or dilute phase reactor may be a small fraction of a second less than 1/2 second and of the order of about .01–0.5 second; or the contact time therein may be higher, up to a time of about 2 and as high as about 6 seconds. In addition, it is contemplated employing temperatures in the dilute phase hydrocracking step in the range of from about 400° F. to about 1,000° F., preferably from about 550 to about 800° F. with the higher temperatures being preferably employed with catalyst of relatively low activity. In the course of effecting the dilute

phase hydrocracking step, hydrocarbonaceous material incompletely converted is adsorbed on the catalyst and this adsorbed material herein referred to as hydrocarbonaceous material is removed by the method of this invention comprising hydroregenerative treatment of the catalyst in a relatively dense fluid bed condition maintained at a desired elevated temperature. That is, the catalyst is regenerated under what may be considered as destructive hydrogenation or hydrocracking conditions at an elevated temperature up to about 1,600° F. with hydrogen-rich gases. In the unitary system of FIGURE I the hot products of the hydrogen regeneration step are caused to flow with the catalyst and reactants passing through the dilute phase hydrocracking step and the temperature of this mixture may be adjusted as desired by the temperature of the makeup hydrogen-rich gas and hydrocarbon feed introduced to the riser conversion zone.

It is clear from the above that the method and sequence of hydrogenative process steps desired to be effected in the unitary system of this invention presents a most unusual arrangement and method of operation of maximum flexibility permitting the use of substantially any pressure including elevated pressure in addition to a wide variety of processing conditions including temperatures for contacting different feed materials with catalyst of greatly differing activity and selectivity levels. That is, when employing what may be referred to as superactive catalyst, more fully described hereinafter in the system of this invention, space velocities far exceeding that ever considered feasible heretofore are permitted. In this respect, space velocities in the range of from about 10 to about 10,000 w./w./hour are contemplated with a space velocity above 1,000 w./w./hour being employed with superactive catalyst comprising crystalline aluminosilicate. Of course, when employing catalyst compositions of substantially lower hydrocracking activity and selectivity comprising silica-metal oxide carrier materials either with or without a base or rare earth exchanged crystalline aluminosilicate and promoted with a Group VIII metal hydrogenating promoter, lower space velocities in the range of from about 0.5 to about 100 w./w./hour may be employed in conjunction with relatively high temperatures and pressures to obtain the desired degree of conversion or hydrocracking severity in the individual steps of the process.

Other catalysts which may be employed in the method and system of this invention include those comprising one or more hydrogenation components combined with a cracking component particularly a silica-metal oxide cracking component and which are sufficiently active to permit conversion of the hydrocarbon feed at temperatures in the range of from about 500° F. to about 900° F. and preferably below about 700° F. The hydrogenation component may include metal oxides and sulfides of metals of Group VI including chromium, molybdenum, tungsten and/or one or more elements of Group VIII including Co, Ni, Pt, Pd, Rh.

That is, one or more metal components comprising the sulfides and oxides or mixtures thereof of the metals of Group VI and VIII may be employed such as nickel-tungsten sulfide, cobalt oxide-molybdenum oxide. Other suitable hydrocracking catalysts include a Group VIII hydrogenating component deposited on a suitable carrier material including composites such as silica-alumina, silica-titania, silica-zirconia, silica-magnesia and alumina boria either with or without a halogen promoter.

A preferred constituent of the hydrocracking catalyst employed in the method of this invention whether used alone or combined with silicon containing cracking components and/or hydrogenating components comprises a crystalline aluminosilicate, preferably a base or rare earth exchanged crystalline aluminosilicate having an alkali metal content of less than about three weight percent and preferably less than 1.0 weight percent and having a struc-

ture of rigid three-dimensional networks characterized by a substantially uniform pore diameter selected from within the range of from about 6 to 15 Angstrom units. One method of preparing such a crystalline aluminosilicate material is described in U.S. patent specification 2,882,244. This material is treated with a fluid containing an ion capable of replacing the alkali metal constituent thereof. As the replacing ion, alkaline earth metals and ammonia, either alone or mixtures thereof may be employed. The alkali metal content of the finished product should be less than about 3 and preferably less than about 0.5 percent by weight. The exchange media used may be employed in the form of a solution for contacting the crystalline aluminosilicate particles while in the form of a fine powder, pellets, beads or other suitable particle shape. Accordingly, the hydrocracking catalyst comprising an ion exchanged crystalline aluminosilicate combined with a hydrogenating-hydrocracking promoter may be formed under conditions to produce a particle of substantially any size and preferably a size less than about 1/4" diameter particle size, more usually less than about 1/8" particle size and of a size which may be easily fluidized. Generally, any fluidizable particle size may be employed and this includes particles of from about 60 to about 200 mesh size or the particles may be less than 100 microns and in the range of from about 20 to about 80, for example, from 40 to about 60 micron particle size. Under some conditions, it may be preferred to employ the active ion exchanged crystalline aluminosilicate alone and of a particle size within the range above described.

As indicated above, other suitable catalyst may be used in the method and unitary pressure systems of this invention which include a hydrogenating metal component combined with one or more oxides selected from the group consisting of clays and inorganic oxide gels having dispersed therein in finely divided form, a catalytically active crystalline aluminosilicate.

The hydrocarbon feed materials which may be processed by the method and means of this invention include substantially any hydrocarbon feed material which one desires to treat under hydrocracking conditions including those materials which are lower boiling than gas oil boiling range materials. Generally speaking, however, it is preferred to hydrocrack high boiling range materials including heavy gas oils, residual stocks, cycle stocks, topped crudes, reduced crudes and those relatively high boiling hydrocarbon fractions derived from coal, tars, pitches, asphalts, shale oils, etc.

In the method of this invention, the removal of hydrocarbonaceous material accumulated on and retained within the catalyst during the conversion is carried out in the presence of hydrogen-rich gases under stripping and reactivating conditions. Accordingly, the hydrogen regeneration step comprises a combination of steps comprising stripping and destructive hydrogenation of retained and/or unconverted hydrocarbonaceous material under conditions generally more severe than employed in the dilute phase fresh hydrocarbon feed conversion step. It has been found that by the proper selection of hydrocracking conditions in the dilute phase conversion step which will vary considerably with the type of hydrocarbon feed being converted and catalyst employed that the residual unconverted hydrocarbonaceous material deposited on the catalyst will be a relatively soft, hydrogen deficient carbonaceous material, that may be converted by hydrocracking and removed from the catalyst. Accordingly, the method and system of this invention accomplishes the removal of deposited hydrocarbonaceous material from the catalyst in the presence of hydrogen and under conditions which permits passing the regenerated catalyst either with or without the regeneration product gases directly to the dilute phase contact zone for effecting hydrocracking and/or hydrogenation of additional fresh hydrocarbon feed material.

On the other hand, the gaseous or vaporous products of the separate contact steps may be combined and passed to a suitable product recovery zone such as a distillation or fractionation zone.

The conversion of the fresh hydrocarbon feed material to lower boiling range products may vary from about 20 to about 100 volume percent conversion depending on the conversion conditions employed for the particular hydrocarbon feed being treated. In this respect, a gas oil boiling between about 400° F. and 600° F. may be hydrocracked without difficulty to products boiling below about 400° F. at conversion levels in the range of from about 40 to about 80 and as high as 100 volume percent conversion with recycle of unconverted material. On the other hand, hydrocracking higher boiling materials including residual oil fractions boiling in excess of 600° F. to lower boiling range products generally requires conversion levels in the range of about 50-70 volume percent to products boiling below about 400° F. and utilizing hydrocracking temperature sufficiently low to avoid permanent damage to the catalyst.

A particularly desirable feature of the method of this invention resides in eliminating the production of water vapor or moisture resulting from oxidation of hydrogen containing deposits during regeneration or stripping with steam and the adverse effects of this moisture on very high activity moisture sensitive catalyst. Accordingly, the hydroregenerative systems proposed herein may be operated under dry or moisture free conditions, especially if means are provided to remove moisture constituents from the input streams (hydrocarbon feed and hydrogen-rich gaseous material). A further desirable feature of the systems of this invention resides in the confinement of the individual hydrocracking and hydroregeneration steps within a single or unitary vessel arrangement which permits maintaining substantially any desired operating pressure and in the event that minor pressure leak should develop between zones and process steps within the unitary vessel arrangements, it is of no great importance and concern in view of the compatibility of the reactions taken place therein, the reactant materials and the products produced. Furthermore, since the reaction and regeneration zones are operated under conditions which permit commingling of reaction products, no harmful effects will accrue by using a relatively large hydroregeneration zone and/or catalyst separation zone for the purpose of providing one or more catalyst inventory chambers in the system. This feature is particularly desirable when using very high activity catalyst and one or more relatively small dilute phase reaction zones in association therewith. Although it has not been specifically shown in the drawings, it is to be understood that more than one, for example, a plurality of dilute phase riser reactor conversion zones may be employed and associated with a single hydroregeneration zone.

In the proposed arrangements herein described, it is significant to note that the hydrocarbon conversion reaction temperature level can be controlled by:

- (1) The temperature level of the entire mixture of the hydrocarbon feed plus recycle gas;
- (2) By the temperature of the fluidizing hydrogen-rich gas flowing into the reactor;
- (3) By the temperature of the activating hydrogen-rich gas flowing to the hydroregeneration zone; and
- (4) By the temperature of the hydrogen employed for the hydroregeneration step.

In addition to the above, it is contemplated providing heat exchange means for adding or extracting heat as desired either directly or indirectly to the catalyst passing through the hydroregeneration step of the process. Accordingly, one of the important aspects of the method and means of this invention resides in the addition of heat to the hydroregeneration zone to provide a suitable heat well or heat source for initiating and carrying out the hydroregeneration step of the process. In this re-

spect, heating coils may be maintained, for example, in the bed of catalyst in the regeneration zone or entering the regeneration zone or heat may be supplied by preheating the regeneration hydrogen containing gas to an elevated temperature above the hydrocarbon feed cracking temperature, for example, in the range of from about 1,000° F. to about 1,600° F. more preferably at least about 1,150° F. before introducing the hydrogen-rich gas to the regeneration step of the process. With the introduction of hydrogen-rich gas at an elevated temperature above about 1,000° F. either with or without the presence of heat exchange coils, one does not need a particularly long catalyst residence time in the regenerator and, therefore, the catalyst circulation rate and inventory may be kept at a desired low minimum. The catalyst heated as hereinbefore described to effect hydrogenative treatment of hydrocarbon material, can be cooled to the temperature desired for the hydrocracking operation substantially instantly with the reactant materials entering the reaction zone. That is, it is contemplated effecting the desired catalyst cooling either with hydrogen-rich gas alone or combined with hydrocarbon feed material upon introduction of the catalyst into the dilute phase conversion zone of the process. In this respect, it is contemplated employing hydrogen-rich gases at a desired temperature to convey the catalyst upwardly through a portion of the riser contact zone prior to the introduction of hydrocarbon reactant material at a desired temperature thereto so that the temperature and time of contact of the hydrocarbon reactant with the catalyst in the riser may be controlled by the temperature of the reactants and point of hydrocarbon reactant inlet to the riser.

Having thus given a general description of the method and means of this invention, reference is now had to the drawings by way of example which show in diagrammatic elevation arrangements of means which permit effecting the methods and sequence of contact steps contemplated by this invention.

FIGURE I presents in diagrammatic elevation one arrangement of unitary apparatus permitting the products obtained from the catalyst hydrogen regeneration step to be passed with fresh hydrocarbon reactant material and regenerated catalyst through a dilute phase hydrocracking step of the process.

FIGURE II, on the other hand, presents in diagrammatic arrangement a unitary apparatus wherein the products of the hydrogen regeneration step and hydrocracking steps of the process are commingled at the outlet of the separate zones and the dilute phase hydrocracking step of the process is surrounded by an annular dense phase hydrogen regeneration zone which is in open communication at the top and bottom thereof with the dilute phase contact zone for circulation of catalyst between zones.

Referring now to FIGURE I by way of example, a unitary substantially vertical vessel arrangement is shown comprising a lower relatively small cylindrical chamber 2 separated from an upper larger cylindrical chamber 4 by a common support baffle member 6 or other suitable structural means for aligning the riser and standpipes provided in the vessel. Chamber 2 may be of a size equal to or larger than chamber 4 rather than smaller as shown and in open communication with chamber 4. A riser conduit 8 of smaller diameter in the lower portion than in the upper portion extends substantially vertically upwardly from the lower portion of the lower chamber 2 through baffle 6 into the upper portion of chamber 4. As indicated herein, a plurality of riser conduits may be employed. The upper end of riser conduit 8 is connected to and in open communication with a first separation zone 10 shown to be a cyclone separation zone. Separation zone 10 is sequentially connected by passageway 12 to a second separation zone 14 also confined within the upper portion of chamber 4 and

shown to be a cyclone separation zone. A conduit 16 connected to cyclone 14 is provided for withdrawing gasiform material from the unitary apparatus of FIGURE I.

A dipleg or standpipe 18 communicating with the bottom portion of cyclone separation zone 10 is provided for returning separated finely divided solid contact material from the separation zone 10 to chamber 2 therebelow for discharge therein beneath the upper portion thereof. A dipleg or standpipe 20 similar to dipleg 18 communicates with the lower portion of separation zone or chamber 14 for the purpose of passing finely divided solids separated therein to said chamber 2 for discharge therein beneath the upper portion thereof. In the specific embodiment of FIGURE I the transition area of riser conduit 8 from a relatively small diameter conduit to a larger diameter conduit is formed by a frustoconical baffle member 22 provided with a plurality of openings or passageways 24 therein.

In the method and circulating catalyst system of FIGURE I, briefly described above, a relatively dense fluid bed of catalyst particulate material 26 having an upper level 28 is maintained in the lower portion of chamber 2 or hydrogen regeneration zone at an elevated temperature. Hydrogen-rich regeneration gas is introduced to the lower portion of the dense fluid bed of catalyst 26 at an elevated pressure sufficient to effect the desired hydrogenation steps by a distributor manifold 30 supplied by conduit 32. In the method of this invention, hydrocarbonaceous material adsorbed on the catalyst during the dilute phase hydrocracking step of the process is removed in the presence of hydrogen-rich regeneration gas while maintaining the catalyst in a relatively dense fluid bed condition. The thus treated catalyst at an elevated temperature and pressure is caused to flow at least in part to the bottom open end of the riser conduit 8 wherein it is combined with a hydrogen-rich lift gas introduced by conduit 34 to form a suspension thereof at a desired hydrocracking temperature which will pass upwardly through the riser conduit and eventually discharged into separator 10. The transition area of riser 8 is at an elevation above the upper level 28 of the dense fluid bed of catalyst 26 in the regeneration zone so that regeneration product gases either with or without entrained catalyst will pass through the passageways 24 opening into the enlarged portion of the riser. Conduit 36 communicating with riser conduit 8 adjacent to the riser transition area and preferably above the passageways 24 is provided for introducing the fresh hydrocarbon feed material to be cracked in an atomized or vaporous condition. Although not specifically shown, hydrogen-rich gas may also be combined with the hydrocarbon feed introduced by conduit 36. It is contemplated in an embodiment of this invention of providing a plurality of hydrocarbon feed inlet conduits, for example, 3 or 4 inlet conduits to the riser and spaced vertically apart from one another between the bottom of the riser and the separator 10 so that the dilute phase residence time of the introduced hydrocarbon feed in contact with the catalyst may be controlled as a function of its length of travel in the riser reactor in terms of time. In addition, the catalyst-hydrocarbon mixture residence time is controlled by the mixture space velocity so that when employing space velocities of about 1,000 and as high as 10,000 w./w./hour in a reactor length up to about 50 ft. and most usually less than about 40 ft., the catalyst-hydrocarbon residence time may be controlled over a wide contact time range generally less than one second. The catalyst-hydrogen-hydrocarbon mixture passes through the riser, maintained in a specific embodiment at a temperature of about 500-800° F., to a separation zone 10 wherein vaporous hydrocarbon constituents are initially separated from the catalyst. The vaporous hydrocarbon constituents entraining catalytic material are passed by conduit 12 to a second separation zone 14

wherein additional catalyst is separated from the vaporous hydrocarbon constituents. The catalyst separated in zones 10 and 14 is passed by diplegs 18 and 20 respectively to the regeneration zone 2 as relatively dense columns of catalyst developing pressure by the head of catalyst therein sufficient to discharge the catalyst into the regenerator section either above or below the dense fluid bed of catalyst maintained therein. The bottom discharge end of the diplegs may be provided with a suitable flap valve or other arrangement which will open only after developing a predetermined pressure head of catalyst in the leg so that the leg acts as a seal leg excluding the flow of gaseous material from the regenerator to the separator by the dipleg.

FIGURE II presents diagrammatically an embodiment wherein the regeneration product gases bypass the dilute phase reaction zone and are combined with the products obtained therefrom above the open discharge or upper end thereof. In this embodiment, a unitary chamber 40 is provided with a perforated grid means 42 containing openings or passageways 44 across its lower cross section. A first cylindrical baffle 46 open at its upper end extends upwardly from said grid means to provide a dilute phase reaction zone or chamber 48 confined within an annular reaction zone or chamber 50. Openings or passageways 52 in the wall of baffle 46 adjacent grid means 42 provide communicating passageways between the lower portion of the annular chamber and the cylindrical chamber. A second cylindrical baffle means 54 communicating between grid means 42 and the bottom of chamber 40 forms a separate plenum chamber 56 beneath chamber 48 and surrounded by an annular plenum chamber 58 beneath annular chamber 50. Baffle means 54 may be perforated for restricted flow of gaseous material if desired, however, it is preferred to employ a non-perforated baffle means for reasons more fully explained hereinafter. Conduit means 60 is provided for supplying fluidizing gas, such as hydrogen-rich gas to the plenum chambers 56 and 58 by branch conduits 62 and 64 provided with valve means 66 and 68. Conduit means 72 connected to a distributor means 74 positioned in the lower portion of dilute phase reaction chamber 48 and above grid 42 is provided for passing hydrocarbon reactant with or without hydrogen containing gas there-through. Conduit 76 is provided for withdrawing product material from the unitary chamber 40. It is to be understood that suitable cyclone separator equipment not shown but well known in the art, may be positioned in the upper portion of chamber 40 to remove and recover entrained catalyst material from vaporous product material prior to removing the vaporous products from the vessel. The separated and recovered catalyst is thereafter returned to the annular regeneration section of the apparatus.

When employing the above apparatus to effect the hydrogenative process of this invention, the catalyst is regenerated with hydrogen containing gases in the annular portion of the reactor while maintaining the catalyst in a fluidized condition moving generally downwardly therethrough. In addition to supplying heat in the regeneration step with the hydrogen-rich regenerating gases by providing suitable heaters not shown in conduit 62, heat coils may also be employed in the dense fluid bed to supply heat thereto. The regenerated catalyst is caused to move to and through passageways 52 into the lower portion of the dilute phase contacting section 48 wherein the catalyst combined with hydrogen-rich gas at a desired temperature passing upwardly through grid 42 from plenum chamber 56 is combined with the hydrocarbon feed material discharge therein by distributor manifold 74 to form a suspension. The suspension thus formed and at an elevated temperature and pressure passes upwardly through the cylindrical reactor under desired conversion conditions for discharge into an enlarged dis-

engaging area or catalyst separation section above the upper open end of the reactor. In the separator section, the catalyst separates from gasiform and/or vaporous material due to a substantial change in its vertical velocity component and this may be generally aided by the use of suitable cyclone separator equipment not shown. The vaporous conversion products including gasiform product material of the catalyst regeneration step is withdrawn as a combined stream through conduit 76 for passage to suitable product recovery equipment not shown. The separated catalyst is caused to flow to the annular regeneration section of the process maintained at an elevated temperature equal to or higher than the temperature in the dilute phase reaction zone and a pressure greater than that employed in the cylindrical reactor section. In the annular regeneration section, the catalyst entraining hydrocarbonaceous material is contacted with hydrogen-rich gas introduced to the lower portion of the annular section by grid 42 from annular plenum section 58 at such a rate to maintain the catalyst therein in a relatively dense fluid bed condition moving generally downwardly therethrough countercurrent to the regenerating gas. By establishing desired and proper flow of hydrogen-rich gas into the separate plenum chambers and out thereof through grid 42, the catalyst may be made to circulate through the system as briefly described above.

Particular advantages of the method and means of FIGURE II resides in the substantial compactness of the reactor-regenerator system thereof, its simplified flow arrangement, the compatibility of reactions taking place therein and the fact that relatively high pressures may be handled in a fluidized solids contact system without encountering many of the problems of prior art processes and particularly those employing adjacent reactions which are not compatible with one another.

Having thus given a general description of the method and means of this invention and provided by way of example specific embodiments thereof, it is to be understood that no undue restrictions are to be imposed by reasons thereof and minor modifications may be made thereto without departing from the scope thereof.

We claim:

1. A method for converting hydrocarbon which comprises circulating a catalytic material sequentially through a dilute phase contact zone and a dense phase contact zone, passing a hydrocarbon reactant material together with hydrogen in contact with the catalytic material in the dilute phase contact zone, passing H_2 rich gas first through said dense phase contact zone and then in contact with catalytic material used in said dilute phase contact zone and maintaining conversion conditions in said zones sufficient to convert hydrocarbonaceous material in the presence of hydrogen-rich gases to lower boiling range products.

2. A method for converting hydrocarbonaceous material in the presence of hydrogen-rich gases to lower boiling range products which comprises circulating a hydrocracking catalyst through a plurality of contact zones comprising a dilute phase catalyst contact zones and a dense phase catalyst contact zone, passing hydrogen-rich gaseous material upwardly through said zones, passing hydrocarbonaceous material into said dilute phase contact zone with the result that a hydrocarbonaceous material is deposited on the catalyst and is carried with the catalyst into said dense phase contact zone wherein it is contacted under more severe hydrogenating conditions, and thereafter passing the resultant regenerated catalyst and hydrogenated products from said dense phase contact zone separately upwardly through said dilute phase contact zone.

3. A method for converting hydrocarbons which comprises circulating a hydrocracking catalyst first through a dilute phase catalyst cracking zone with hydrogen and then with hydrogen through a dense phase catalyst crack-

ing zone, passing a hydrocarbon reactant material in contact with the catalyst in the dilute phase cracking zone combined with hydrogen containing gaseous material recovered from said dense phase catalyst cracking zone, passing any insufficiently converted hydrocarbon reactant from the dilute phase contact zone to the dense phase contact zone and maintaining hydrocracking severity conditions in said dense phase contact zone generally more severe than the conversion conditions maintained in said dilute phase contact zone.

4. A method for hydrocracking gas oil and higher boiling hydrocarbons with a catalyst comprising crystalline aluminosilicate which comprises circulating a hydrocracking catalyst comprising catalytically active crystalline aluminosilicate through a system comprising a dilute catalyst phase contact zone and a dense catalyst phase contacting zone; maintaining conversion severity conditions in said dense phase contacting zone at least equal to the conversion severity conditions of said dilute phase contacting zone, introducing the hydrocarbon to be converted to the dilute phase contacting zone, introducing hydrogen to be used in the process initially in part to said dense phase contacting zone and passing gaseous products obtained from said dense phase contacting zone rich in hydrogen through a substantial portion of said dilute phase contacting step to supply a portion of the hydrogen requirements thereof.

5. A method for converting hydrocarbons in the presence of hydrogen which comprises, passing a suspension of a hydrocracking catalyst containing rare earth exchanged crystalline aluminosilicate in a hydrogen-rich gas stream through an elongated reaction zone in dilute phase condition, introducing hydrocarbon to be converted to said suspension under conditions to obtain a contact time of not more than a fraction of a second, passing catalyst and any retained hydrocarbonaceous material from said dilute phase contact step to a dense phase contact step, regenerating the catalyst with hydrogen in said dense phase contact step, said dilute phase contact step and said dense phase contact step being in open communication with one another in at least the lower portion thereof and withdrawing the products of said dilute phase and dense phase contact steps as a combined product from above said dilute phase contact step.

6. A method for converting hydrocarbonaceous material comprising gas oil and higher boiling material and boiling above gas oil boiling range in the presence of hydrogen-rich gases and a hydrocracking catalyst containing a Group VIII metal hydrogenating component which comprises passing a Group VIII metal promoted hydrocracking catalyst sequentially through a rising dilute phase catalyst reaction zone and then into a dense phase catalyst reaction zone about at least the lower portion of said dilute phase reaction zone, causing at least a portion of the hydrocarbonaceous material deposited on the catalyst during its passage through the dilute phase contact zone to pass with the catalyst to the dense phase contact zone, passing hydrogen-rich gases in parallel flow arrangement to said dilute and dense phase contacting zones and passing catalyst from which hydrocarbonaceous material has been removed in said dense phase contact zone and gaseous products thereof to said dilute phase contact zone.

7. A method for converting hydrocarbonaceous material comprising gas oil and higher boiling material and boiling above gas oil boiling range in the presence of hydrogen-rich gases and a hydrocracking catalyst containing a Group VIII metal hydrogenating component which comprises passing a Group VIII metal promoted hydrocracking catalyst sequentially through a rising dilute phase catalyst reaction zone and then into a dense phase catalyst reaction zone about at least the lower portion of said dilute phase reaction zone, causing at least a portion of the hydrocarbonaceous material deposited on the catalyst during its passage through the dilute phase contact zone to pass with the catalyst to the dense phase

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contact zone, passing hydrogen-rich gases in parallel flow arrangement to said dilute and dense phase contacting zones and passing catalyst from which hydrocarbonaceous material has been removed from said dense phase contact zone to said dilute phase contact zone separately with gaseous products of said dense phase contacting step.

8. A process for hydrocracking a residual oil fraction with a hydrocracking catalyst comprising rare earth exchanged crystalline aluminosilicate which comprises passing the catalyst sequentially through a dilute phase contact step and a dense phase contact step maintained under conditions of increasing conversion severity, passing catalyst from said dense phase contacting step to said dilute phase contacting step, passing hydrogen-rich gas in parallel flow arrangement to said dilute and dense phase contacting steps under conditions to effect conversion of hydrocarbonaceous materials flowing thereto to lower boiling range products therein, passing gasiform product material obtained from said dense phase contact step through at least a portion of said dilute phase step and recovering hydrocarbon product of said dilute and dense phase conversion steps from the upper portion of said dilute phase conversion step.

9. A method for converting hydrocarbons which comprises passing a hydrocarbon feed combined with hydro-

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gen in contact with a catalyst having cracking activity comprising a catalytically active crystalline aluminosilicate through a dilute phase catalyst contact zone wherein hydrocarbonaceous deposits are formed on the catalyst, thereafter passing catalyst with formed hydrocarbonaceous deposits in contact with hydrogen rich gaseous material through a dense fluid catalyst bed contact zone; passing catalyst from a lower portion of said dense catalyst bed to the inlet of said dilute phase contact zone and passing vaporous material recovered from above the dense fluid bed of catalyst to said dilute phase contact zone above the inlet of catalyst thereto.

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