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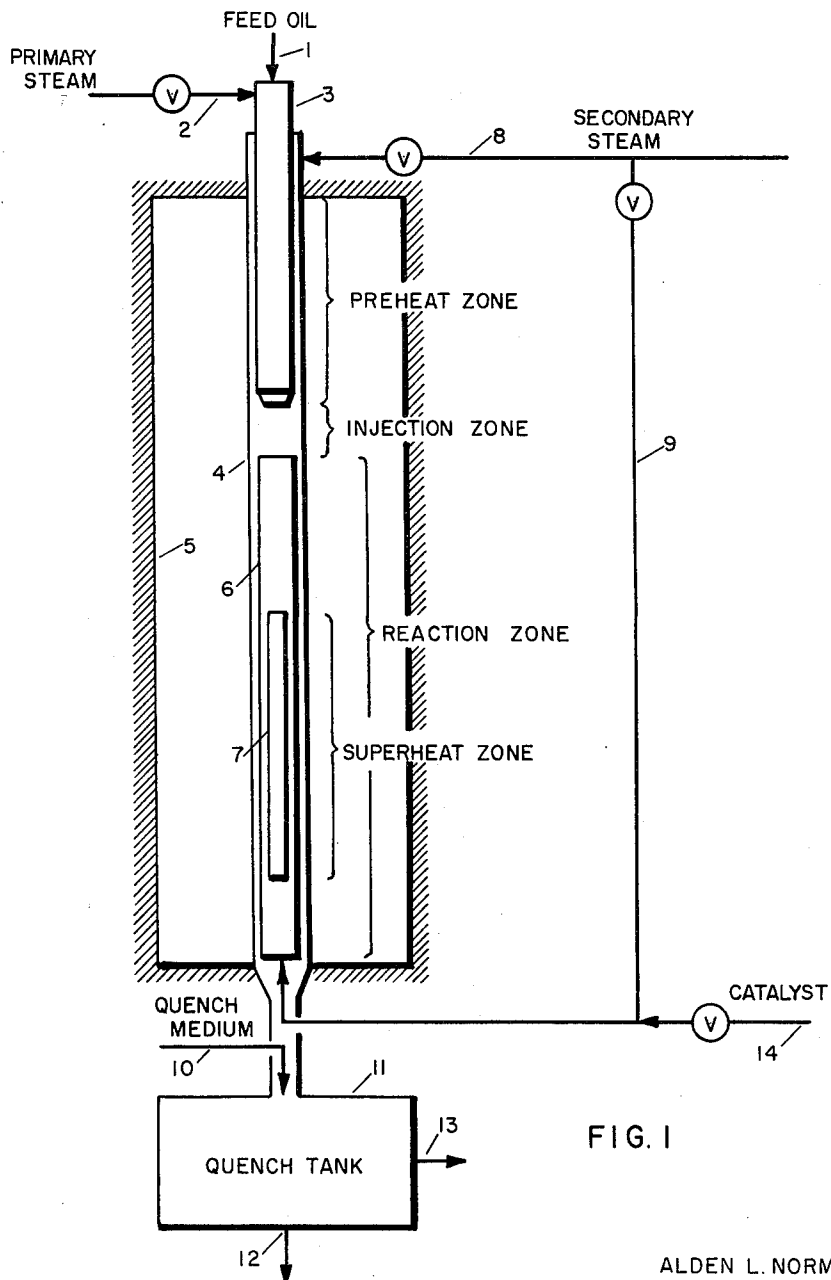
J. K. JACOBS ETAL

3,154,385

APPARATUS FOR PYROLYSIS OF HYDROCARBONS

Filed Oct. 11, 1960

2 Sheets-Sheet 1



ALDEN L. NORMAN, JR.
JOHN K. JACOBS
INVENTORS

BY

Ernest G. Peterson

AGENT

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2 Sheets-Sheet 2

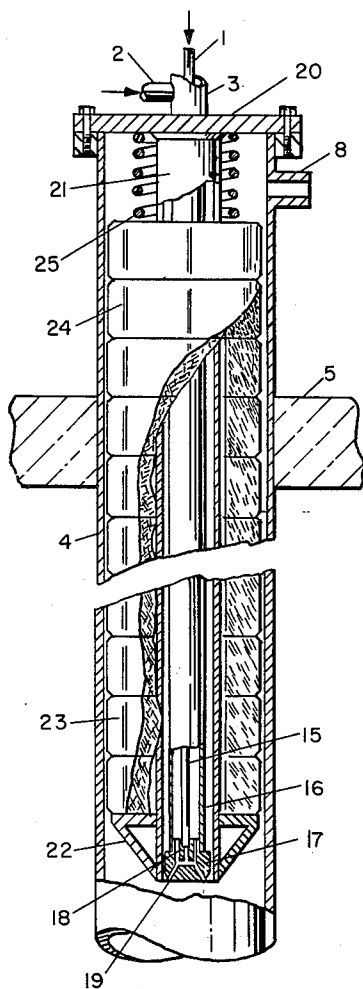


FIG. 2

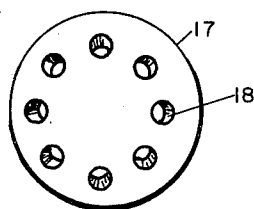


FIG. 3

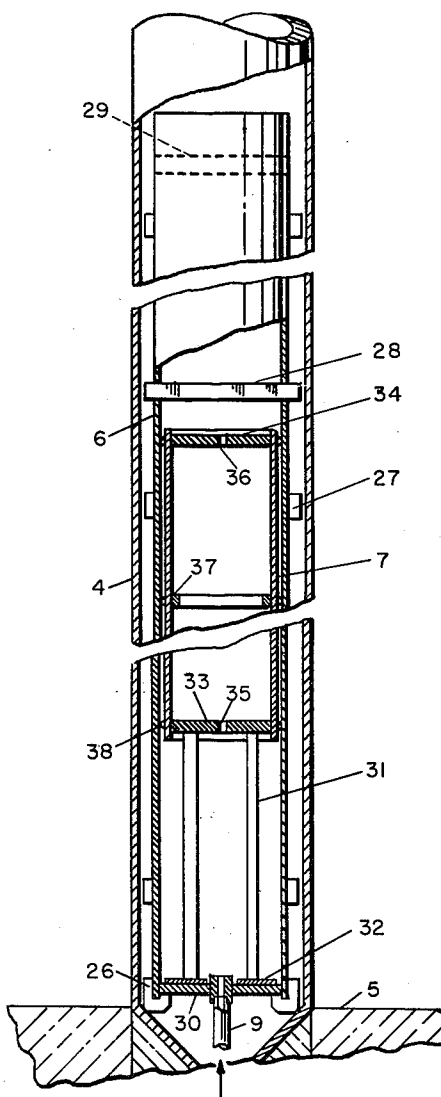


FIG. 4

ALDEN L. NORMAN, JR.
JOHN K. JACOBS
INVENTORS

BY *Ernest G. Peterson*

AGENT

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APPARATUS FOR PYROLYSIS OF
HYDROCARBONS

John K. Jacobs, Boston, and Alden L. Norman, Jr.,
Topsfield, Mass., assignors to Hercules Powder Com-
pany, Wilmington, Del., a corporation of Delaware
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4 Claims. (Cl. 23-284)

This invention relates to the pyrolysis of hydrocarbons and particularly to apparatus for the thermal cracking of normally liquid, as well as normally gaseous, hydrocarbons in tubular furnaces and more particularly to the introduction of superheated fluid medium thereinto.

The art of high temperature cracking in tubular furnaces has reached a high degree of proficiency in more recent years. This is particularly evident in that the utilization of tubular cracking furnaces has provided practical flexible processes for the production of hydrogen, olefins, and other valuable gaseous and liquid products. Furthermore, the versatility of these furnaces is most readily appreciated in considering the variety of charging stocks which may be used including normally gaseous as well as normally liquid hydrocarbons and the excellent reaction control possible to obtain predetermined, desired end products. Still further, advances in the art of tubular cracking have afforded yields of cracked products and throughput of charging stock which is attractive to large-scale handling of hydrocarbon stock such as is encountered in the refining of petroleum and other large-scale operations. These advances in the art may be more explicitly appreciated with reference to, for example, Patent No. 2,914,386 which discloses a tubular reaction furnace fired at a plurality of spaced points to give flexible and high temperature control of the reacting fluids and extended tube life; Patent No. 2,525,276 which discloses a process for the cracking of hydrocarbon oils with a minimum of carbon deposition for the substantial production of olefins and including injection means for normally gaseous as well as normally liquid hydrocarbon feed stock; and Patent No. 2,904,502 which discloses a process of converting hydrocarbons to obtain high capacity throughput in a tubular furnace with a high yield of liquid products.

Although the above patents are exemplary of the proficiency presently existing in the tubular furnace art, still further advances are being sought and particularly in respect to capital investment, processing variables, continuity of operation, and the quantity and quality of desired end products.

Accordingly, a principal object of this invention is to provide fundamental improvements in the art which pertain to the factors heretofore set forth and particularly to apparatus for increasing the heat input for the injection and reaction zones of a pyrolysis tube. The accomplishment of this object and others will appear hereinafter, the novel features and combinations being set forth in the appended claims.

Generally described, the present invention provides apparatus for the pyrolysis of hydrocarbons in an exteriorly-heated pyrolysis tube having a reaction zone therein formed by an elongated main core means disposed within the pyrolysis tube to form an annular passage and reaction zone therebetween for a mixture of hydrocarbon and diluent gas downwardly passed therethrough, in which the improvement comprises an elongated, substantially impervious inner core means disposed and annularly spaced within the main core means and spaced from the top and bottom thereof to form an annular passage and superheat zone therebetween for diluent gas upwardly passed therethrough and into the mixture, after which

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the mixture thus formed is downwardly passed through the reaction zone.

A preferred embodiment of the invention has been chosen for purposes of illustration and description and is shown in the accompanying drawings wherein reference symbols refer to like parts wherever they occur.

FIG. 1 is a diagrammatic, vertical, sectional view of one form of apparatus for carrying the invention into effect;

FIG. 2 is a fragmentary, enlarged, vertical, part elevational and part sectional view depicting the injection and preheat apparatus diagrammatically shown in FIG. 1;

FIG. 3 is an enlarged plan view of the bottom of the atomizing nozzle depicted in FIG. 2; and

FIG. 4 is a fragmentary, enlarged, vertical, part elevational and part sectional view depicting the superheat and reaction apparatus diagrammatically shown in FIG. 1.

Referring more particularly to FIG. 1, suitably preheated and pressurized oil is passed through a conduit 1 and primary steam is admitted through a conduit 2 into an injection tube 3 whereby the oil is atomized and discharged therefrom. The injection tube 3 extends into a pyrolysis tube 4 which extends through a refractory walled furnace 5 which is preferably fired at a plurality of spaced points. Within the pyrolysis tube 4 there is deposited a first internal or main core 6 of extended length which in combination with the pyrolysis tube 4 defines an annular reaction zone. Within the first internal core 6 there is intermediately disposed a second internal or inner core 7 which is spaced from the bottom and from the top of the first internal core 6 and which in combination therewith defines an annular superheat zone. Similarly, the extension of the injection tube 3 into the pyrolysis tube 4 forms an annular preheat zone. The atomized mixture of oil and primary steam is injected in the form of a conical spray pattern from the injection tube 3 into a concurrently moving, concentric body of secondary steam which is fed through a valved conduit 8 into the pyrolysis tube 4 and flows downwardly around the injection tube into an injection zone. A portion of the secondary steam is passed through a valved conduit 9 into the bottom of the first internal core 6 and passes upwardly through the annular space between the first internal core 6 and the second internal core 7 which provide the superheat zone. This steam, referred to as secondary core steam, is brought to a high temperature in the superheat zone in this manner and emerges from the top of the core 6, which is open, into the injection zone wherein it is intimately mixed with the gaseous streams of oil and primary steam emerging from the injection tube 3 and the secondary steam which flows downwardly around the injection tube. The gaseous mixture thus formed is quickly passed downwardly through the annular reaction zone which is formed between the first internal core 6 and the pyrolysis tube 4. In the reaction zone, the reactants are thermally cracked and the cracked products are quenched as they leave the pyrolysis tube 4 by quenching medium. The quenching medium is passed through line 10 and is preferably hydrocarbons produced as the result of quenching in a quench tank 11. Liquid products and gaseous products are passed from the quench tank 11 through conduits 12 and 13, respectively, to suitable subsequent processing equipment.

Referring now particularly to FIGS. 2 and 3 which depict in detail preferred injection and preheat apparatus for use in conjunction with practicing the invention, the pyrolysis tube 4 is suspended by suitable means (not shown) through the arch of the furnace 5. Projecting concentrically within the pyrolysis tube 4 is the injection tube 3 which is supplied with feed oil and primary steam through the conduits 1 and 2, respectively. The injection

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tube 3 has a concentric internal oil delivery tube 15 which is surrounded by a steam delivery tube 16. An atomizing nozzle 17 is affixed to the bottom of the tubes. The atomizing nozzle is of the shear type, having its atomizing head provided with eight channels 18 in communication with the steam tube 16. The channels 18 toward the egress end thereof diverge at an angle of 11 degrees from the vertical axis to thus form a conical spray pattern. Oil delivery from the oil delivery tube 15 is at approximately 90 degrees to the steam flow and eight oil delivery ports 19 are provided in communication with the channels 18. This combination results in high shearing forces on the oil stream which in turn is conducive to produce very fine atomization of the oil which is important for aiding the vaporization of the oil. Additionally, the ability to utilize nozzles having various degrees of egress divergency results in the obtention of an optimum distribution of atomized oil in the injection zone of the pyrolysis tube under considerable variation in process and apparatus conditions and sizes.

The preferred preheat apparatus in combination with the above-described injection apparatus has a cover plate 20 which is secured to the top of the pyrolysis tube 4. The cover plate 20 supports the injection tube 3 and also supports a girth tube 21, both being affixed to the cover plate in fluid impervious relationship to the atmosphere. The lower end of the girth tube 21 has a truncated conical support member 22 affixed thereto. The egress end of the nozzle 17 is approximately flush with the bottom of the girth tube 21 and is rotatably spaced therein. A plurality of stacked refractory discs or elements 23 supported by the support member 22 is concentric with the girth tube 21 and is in spaced relationship thereto providing about a 1/4 inch wide annulus. Each of the refractory discs 23 is provided with six equally spaced ribs or protrusions 24 extending about 1/8 inch radially to provide a slip fit and annulus between the discs and the interior wall of the pyrolysis tube 4 to thereby form the preheat zone annulus. A compression spring 25 interposed between the cover plate 20 and the top refractory disc maintains the plurality of stacked refractory discs 23 in tight compressive relationship and yet permits individual lateral displacement thereof for facile insertion or removal of the entire injection apparatus. Heretofore it was found that injection apparatus utilizing such close tolerance as utilized here could only be removed from the pyrolysis tube with great difficulty and in many instances causing costly damage to the injection apparatus or to the pyrolysis tube or to both of them. However, the injection apparatus hereinbefore described is free from such deficiency since the refractory discs which are slidable relative to each other move to accommodate any irregularities of the pyrolysis tube such as caused by warpage, distortion and the like due to the high heats involved. Additionally, in service, the refractory discs accommodate any movement of the pyrolysis tube longitudinally and transversely to maintain distinctly optimum conditions in the preheat zone and proper injection into the injection zone.

Referring now particularly to FIG. 4 which depicts in detail a preferred form of superheat and reaction apparatus for practicing this invention, the pyrolysis tube 4 which is suspended extends to the hearth of the furnace 5 where it is necked down and extends through the hearth, being insulated relative thereto, and accommodates the quench tank conduit. The first internal core 6 is supported at its base by a plurality of support lugs 26 which rests on the necked-down portion at the bottom of the pyrolysis tube 4. A plurality of centering lugs 27 which are spaced circumferentially and longitudinally is affixed to the first internal core 6 to keep it properly centered with a slip fit within the pyrolysis tube 4 and to provide an annulus between the outer wall of the core 6 and the inner wall of the pyrolysis tube 4 to thereby form the reaction zone annulus. A stiffening bar 28 which extends transversely across the core 6 and is rigidly secured thereto is provided

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at near the top of the second internal core 7 and a lift bar 29 is provided near the top of the core 6 which in conjunction with suitable lifting equipment provides a means for removing the entire assembly of cores from the pyrolysis tube. A sealing plate 30 is secured to the bottom of the first internal core 6, and secondary core steam through valved conduit 9 is admitted through the sealing plate into the interior of the core 6, and after passing around the core 7 eventually emits from the top of core 6 which is open. In further detail, the second internal core 7 is provided with a plurality of supporting legs 31 which in turn are secured to an annular base plate 32 which rests upon the sealing plate 30. The legs 31 are secured at their other end to a bottom plate 33 which in turn is secured to the core 7. A top plate 34 is secured to the core 7 and the plates 33 and 34 provide a gas-tight seal for the interior of the core 7 with the exception of two small vent holes 35 and 36 of 1/4 inch diameter which extend through the plates 33 and 34, respectively, and permit the escape of any entrapped gas. Thus, the inner core 7 is substantially impervious and forces fluid medium passed into the core 6 to flow thereabout. A plurality of stiffening rings as represented by 37 is spaced along the length of the core 7 to insure stability of the structure throughout its length. Additionally, a plurality of spacer buttons or elements 38 is spaced circumferentially about and along the length of the core 7 to insure proper centering of the core 7 within the core 6. The spacer buttons 38 extend about 1/8 inch radially and provide a slip fit and centering for the core 7 within the core 6 and, accordingly, an annulus between the cores to thereby form the superheat zone annulus.

Examples of the operation of the invention are given in which the length of the pyrolysis tube from the bottom of the furnace arch to the top of the hearth was 28 feet. The pyrolysis tube was stabilized stainless steel and was 8 inches in inside diameter and had a wall thickness of 0.25 inch. The injection and preheat apparatus extended into the furnace 7.5 feet below the arch and the preheat zone had an annulus 0.25 inch wide. The first internal core was Inconel, which is classed as a nonferrous alloy, and had an outside diameter of 6.5 inches and an inside diameter of 6 inches, thus forming an annulus 0.75 inch wide for the reaction zone. The first internal core was 17.75 feet long extending upwardly from the hearth. The second internal core was also Inconel and had an outside diameter of 5.5 inches and an inside diameter of 5.12 inches, thus forming an annulus 0.25 inch wide for the superheat zone. The second internal core was 9 feet long and was spaced approximately 3 feet above the hearth by the supporting legs, leaving a distance of 5.75 feet between the top ends of the respective cores and leaving a distance of 2.75 feet for the injection zone.

Experiments were conducted using the processing conditions, pyrolysis tube and furnace as disclosed in U.S. Patent No. 2,904,502 to J. H. Shapeleigh. The first runs were made using the injection nozzle and perforated top core described in the patent. Later runs were made using the preferred atomizing nozzle and the open top core as hereinbefore described in combination with the superheat apparatus in accordance with this invention. The following examples are representative of the results obtained and demonstrate the improved operability obtained. The oil processed for all examples was a Gulf Coast crude oil (Texas) having a 22° A.P.I. gravity.

Example 1

This example is typical of results obtained with utilization of the injection nozzle and the perforated top core as disclosed in U.S. Patent No. 2,904,502. Oil was fed at a 400 lb./hr. rate with 800 lb./hr. of total steam for 7 hours before purging 13 lb. of carbon in 1 hour. In this case 0.46% of the oil fed was deposited as carbon in the cracking tube. Of the total steam used 200 lb./hr. was primary steam, 575 lb./hr. was secondary steam,

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and 25 lb./hr. was secondary core steam. The oil was cracked 57.8 weight percent to gaseous products. The following olefin yields (lb./100 lb. feed stock) were obtained: ethylene 20; propylene 5.3; 1,3-butadiene 2.8; and isobutylene 0.9.

Example 2

In this example the atomizing nozzle and open top core were used. Oil was fed at a 533 lb./hr. rate with 1000 lb./hr. of steam for 87.5 hours. There was no indication of fouling in the cracking tube by the end of the run. The purge gas obtained indicated the removal of 0.25 lb. of carbon. In this run 0.0005% of the oil fed was deposited as carbon in the cracking tube. Of the total steam used 75 lb./hr. was primary steam, 725 lb./hr. was secondary steam, and 200 lb./hr. was superheated secondary core steam generated in accordance with this invention. The oil was cracked 60.0 weight percent to gaseous products. The following olefin yields (lb./100 lb. feed stock) were obtained: ethylene 18.9; propylene 9.8; 1,3-butadiene 5.0; and isobutylene --.

Example 3

In this example the atomizing nozzle and open top core were used. Oil was fed at a 530 lb./hr. rate with 850 to 950 lb./hr. of steam for 88.4 hours before purging 12 lb. of carbon in 1 hour. In this case 0.026% of the oil fed was deposited as carbon in the cracking tube. Of the total steam used 90 lb./hr. was primary steam, 560 to 660 lb./hr. was secondary steam, and 200 lb./hr. was superheated secondary core steam generated in accordance with this invention. The oil was cracked 56.1 weight percent to gaseous products. The following olefin yields (lb./100 lb. feed stock) were obtained: ethylene 16.0; propylene 8.2; 1,3-butadiene 3.7; and isobutylene 3.3.

With reference to the above examples, the improvement afforded is readily appreciated. Although it will be further appreciated that the invention has been demonstrated in combination with improved atomizing and core apparatus wherein the atomizing nozzle imparts high shearing forces on the oil stream which is conducive to very fine atomization; wherein the use of high pressure drops (60 to 100 p.s.i.g.) through the nozzle also assures fine atomization; and wherein the use of an open top core eliminates the surface that is most likely to be fouled by carbon deposits, it will be still further appreciated that the superheated steam emitting from the top of the core prevents smaller liquid particles from entering and acts to more quickly and thoroughly evaporate larger particles that do enter. Furthermore, the overall combination of conical pattern atomization, secondary steam flow, and opposing superheated secondary core steam flow assures intimate mixing and improved vaporization while concomitantly tending to force the atomized oil stream against the hot tube wall of the injection zone, thus affording more rapid vaporization of the atomized oil.

In this respect it will be appreciated that the steam atomizing nozzle incorporates an atomizing head to which attaches a steam delivery tube of larger diameter than the concentric internal oil delivery tube. The atomizing head is drilled with a multiplicity of steam holes connecting with nozzle-shaped apertures. At the junction of the nozzle-shaped apertures and the steam holes are oil delivery ports connecting with the central oil supply. Oil delivery is at approximately 90° to the direction of the steam flow. The steam nozzles are drilled at the desired angle to obtain the desired spray pattern in the injection zone of the cracking tube. The atomizing assembly is sized to give a 40 to 100 p.s.i.g. pressure drop for the oil and a 60 to 120 p.s.i.g. pressure drop for the steam. The atomizing steam flow can be varied from 0.1 to 0.5 lb. of steam per lb. of oil with preferred operation at 0.15 lb. of steam per lb. of oil. Moreover, the top of the core does not have a top closure, conical

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or otherwise. The top of the core may be the same diameter as the main core or tapered to a smaller diameter; however, the top of the core is maintained completely open and the superheated core steam is passed therethrough at a temperature of about 1400° to 1800° F. Thus, in accordance with this invention, the provision of the inner core within the main core intermediate thereof to superheat the core steam or other fluid medium increases the heat load in the reaction zone of the pyrolysis tube in the region where the heat requirement for reaction is normally lightest. Accordingly, the total heat input to the injection zone of the pyrolysis tube is increased for a given tube wall temperature and the superheat is generated in an area where normally it can be given without deleteriously effecting the cracking efficiency required for the reaction zone. Normally, from 10 to 40 percent of the total steam flow may be passed through the core to obtain superheated steam for the injection zone. In terms relative to the apparatus described for demonstrating the invention, this amounts to from about 150 to about 250 pounds per hour of steam superheated to a temperature of from about 1400° to about 1650° F. Accordingly, it will be appreciated that a principal advantage of this invention resides in the fact that utilization of the inner core within the main core provides hotter core steam than could otherwise be obtained with absence of the inner core. For example, if a 6-inch I.D. main core and a 5.5-inch O.D. inner core are used as previously described, the convection heat transfer rate is raised by a factor of 6.48 as compared to a single 6-inch I.D. core at the same flow rate and core wall temperature and, further, as an additional advantage, the use of the intermediately disposed inner core increases the heat load on that section of the pyrolysis tube where the heat load requirement would normally be lowest.

From the foregoing, it will be apparent that the process and apparatus herein disclosed for diagrammatic presentation of the invention are susceptible to numerous other possible combinations and arrangements for operation of the invention including equipment other than that shown, such as waste heat boilers, heat exchangers, condensers, coolers, multiple fractionating columns with take-off, and the like. However, such means have not been shown or disclosed since they are well within the province of persons skilled in the art. Furthermore, it will be apparent that the invention herein disclosed may be operated with or without the use of catalysts. In using catalyst, the catalyst may be employed in a simple and expedient manner relative to the preferred embodiment of the invention illustrated in FIG. 1 wherein fluid catalyst is passed through valved conduit 14 into the conduit 9 which in turn feeds secondary steam and catalyst to the injection zone, after which the reactants and catalyst pass through the reaction zone from whence or after which the catalyst may be removed.

In cases where it is desired to utilize a minimum quantity of secondary core steam passed countercurrently to the reactants passing through the pyrolysis tube, there may be used only a small percentage of the total diluent passing from the tube; for example, when a 2 to 1 weight ratio of steam to hydrocarbon is used, the countercurrent steam passing through the core may amount to only about 5%. On the other hand, when the use is primarily that of obtaining superheat for the reactant mixture or, additionally, as a carrier for catalyst, the steam or other diluent being passed through the core may more substantially replace the other diluent being passed concurrently into the reaction zone, but preferably the countercurrent steam will not exceed from about 10 to about 40% by weight of the total. When fluid medium or diluents other than steam are used, there may be used materials which react with carbon and which are not solely diluents in that sense. In this category, hydrogen, and even air, may be used although with the latter, dilution with nitrogen, flue gas, etc., is safer and preferred.

The cores and appurtenances thereof may be type 310 stainless steel, stabilized or unstabilized, the nonferrous alloys such as Inconel and Incoloy, and the like. However, in cases where anticatalytic action is highly desirable, it is preferred to restrict metal contact through use of a nonmetallic exposed core surface, such as ceramic, refractory, or Carborundum types or to utilize refractory discs especially for the inner core, selected and assembled according to the reaction conditions to be employed.

In practicing this invention, the hydrocarbon feed stock may be a liquid product such as a fuel oil or crude oil and is initially introduced into the process with diluent. With steam, for example, as the diluent, the ratio of steam to hydrocarbon can be varied to give either high or low ratios as desired depending on the difficulty encountered in atomizing and vaporizing the feed stock. Normally, this ratio will be in the order of 1:10 to 5:1 with a preferred steam to hydrocarbon ratio of from about 1:1 to 3:1. The use of a diluent, such as steam, lowers the boiling point of the hydrocarbon and makes possible substantially complete vaporization of certain stocks, such as crude or topped crude, which are heavier than gas-oil, for example. Additionally, the diluent, such as steam, can be used to assist in control of the residence time which in conjunction with the control of the pyrolysis tube wall temperature makes it possible to control the effluent temperature of the reactants within a considerable range as desired.

In the reaction zone of the pyrolysis tube, the average metal wall temperature will be maintained at a temperature of at least 1685° F. with the average surface temperature of the refractory walled furnace maintained at a temperature of at least 1835° F. Higher temperatures may be employed with the upper limit primarily dictated by the materials of construction involved. Generally, it has been found that an average metal wall temperature of from about 1700° to about 2000° F. for the reaction zone of the pyrolysis tube gives very satisfactory results.

The cores in relationship to the annuli and to size of tube are a process variable. They are a factor to regulate time and play an important part because of core temperatures controlled on the one hand by radiant heat and on the other by extraction of heat by the reactants. Under normal operating conditions, the high surface temperatures of the tube and internal cores are not found to be conducive to formation of carbon or to the retention of carbon to any detrimental degree. The preferred vertical tube arrangement disclosed herein is conducive to cleanliness and freedom from operating trouble.

The use of catalysts to influence the purity or the composition of ultimate end products is within the scope of this invention and includes arrangements referred to herein along with such catalysts as pumice, clays, aluminum silicates, hydrosilicates, chromium, molybdenum, vanadium oxides, ferric oxides, magnesia, cupric oxide, zinc oxide, potassium oxide, plus other materials and modifiers therefor as are well known in the art. Dehydrogenation is one type of reaction taking place while cracking, and butadiene is a normal constituent of the product gas. Butadiene, however, can be produced by the well known dehydrogenation of butane or butene using well known catalysts, and such a reaction is illustrative of the known use of catalysts and of diluents other than steam such as flue gas, nitrogen, etc., where, for example, catalyst activity might be impaired by the presence of steam.

It will be understood that this invention is not limited to normally liquid feed stock and that regardless of whether the feed stock is normally liquid or normally gaseous, the reactants entering the reaction zone are intended to be gaseous, as effectively as practical possible within the objectives of the invention. Once in the gaseous state, the reaction zone processing is unresponsive to any characterization of the original feed stock state as it may have existed in storage. Butane, butene, propane, and hexane are examples of feed stocks differing from oils. Such stocks are processable to utilize the improved features of this invention.

In the practice of this invention it has been found particularly effective to employ the tubular furnace and process such as disclosed in Patent No. 2,914,386 and Patent No. 2,904,502, respectively. It will be evident, therefore, that this invention may be carried out by the use of various modifications and changes without departing from its spirit and scope.

What we claim and desire to protect by Letters Patent is:

1. In apparatus for the pyrolysis of hydrocarbons in an exteriorly heated pyrolysis tube having an elongated main core disposed therein forming an annular passage reaction zone between the interior of the pyrolysis tube and the exterior of the main core and having injection means for feeding a vaporized mixture of hydrocarbon and diluent gas into the pyrolysis tube and downwardly through the annular passage reaction zone and therefrom, the improvement comprising

(a) an elongated inner core intermediately disposed within the main core forming an annular passage superheat zone between the interior of the main core and the exterior of the inner core,

(b) the inner core being substantially impervious and of less length than the main core with the distance from the top of the inner core to the top of the main core being substantially greater than the distance from the bottom of the inner core to the bottom of the main core; and

(c) feed means at the bottom of the main core for passage of additional diluent gas thereinto and upwardly through the annular passage superheat zone and from the top of the main core into the vaporized mixture whereby the entire superheated mixture thus formed passes downwardly through the annular passage reaction zone and therefrom.

2. The improvement according to claim 1 in which the inner core means has a plurality of spacer elements, each of which radially extends therefrom in slidable relationship with the interior wall of the main core means.

3. The improvement according to claim 1 in which the substantially impervious inner core has vent means therefor.

4. The improvement according to claim 1 in which the inner core means has support legs at the bottom thereof to maintain said core in spaced relationship to the bottom of the main core means.

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