

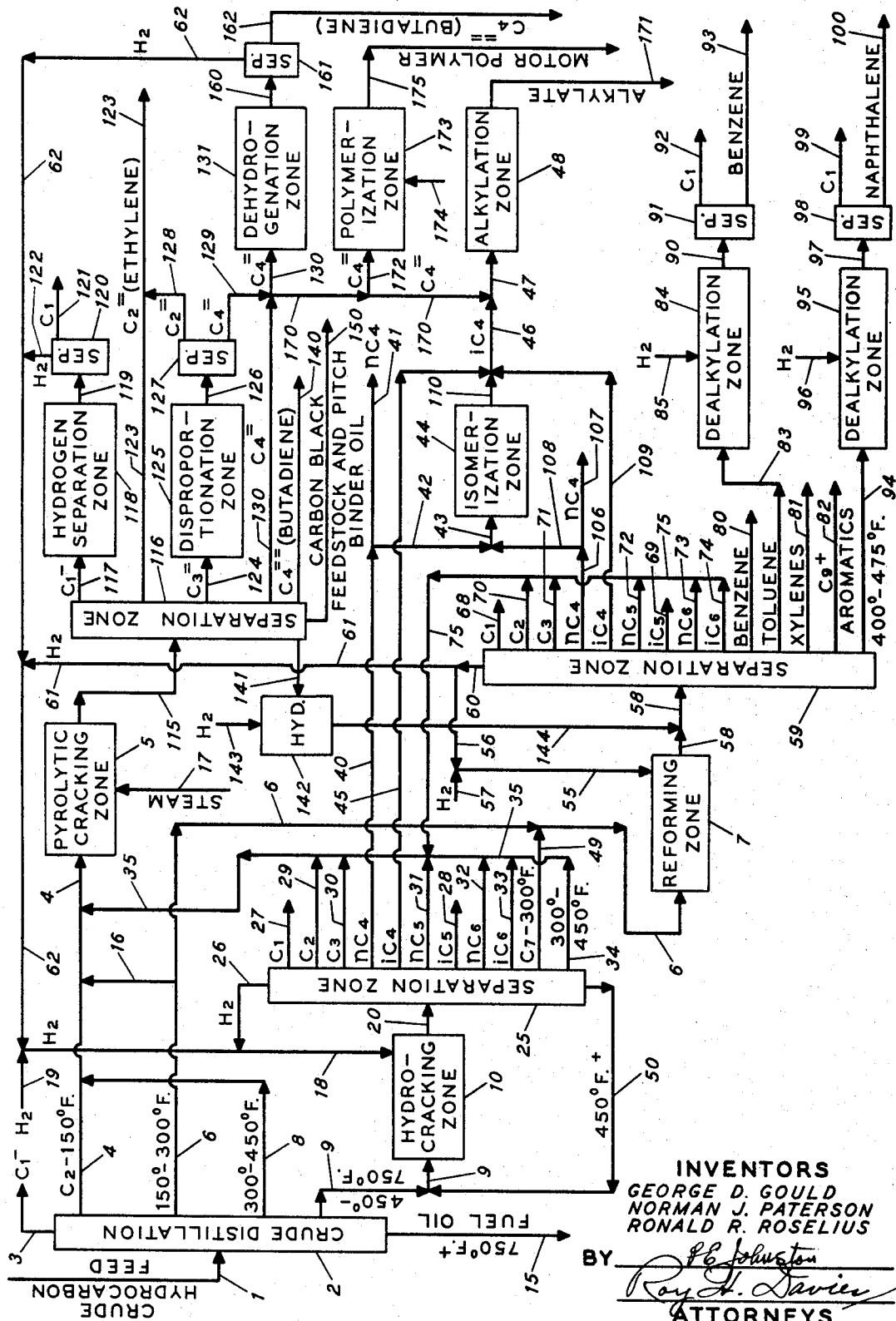
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3,409,540

COMBINATION CATALYTIC HYDROCRACKING, PYROLYTIC CRACKING AND
CATALYTIC REFORMING PROCESS FOR CONVERTING A WIDE
BOILING RANGE CRUDE HYDROCARBON FEEDSTOCK
INTO VARIOUS VALUABLE PRODUCTS
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COMBINATION CATALYTIC HYDROCRACKING, PYROLYTIC CRACKING AND CATALYTIC REFORMING PROCESS FOR CONVERTING A WIDE BOILING RANGE CRUDE HYDROCARBON FEEDSTOCK INTO VARIOUS VALUABLE PRODUCTS

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ABSTRACT OF THE DISCLOSURE

A process for converting a wide boiling range crude petroleum feedstock into various valuable products including ethylene and aromatic hydrocarbons, comprising fractionating the crude, catalytically reforming a heavy straight run fraction thereof, catalytically hydrocracking a fraction thereof boiling in the range 350° to 800° F., and pyrolytically cracking a light straight run fraction thereof together with a portion of the effluent from the hydrocracking zone.

Background of invention

(1) Field of invention.—The invention claimed herein is a combination process for converting a wide boiling range crude petroleum feedstock into various products including ethylene and aromatic hydrocarbons, using steps including catalytic reforming, pyrolytic cracking and catalytic hydrocracking.

(2) Description of prior art.—Heretofore it has been known to convert a naphtha feedstock pyrolytically to various products including ethylene. Such a process is disclosed in U.S. Patent 3,281,351. Heretofore it also has been known to fractionate a wide boiling range crude petroleum stock and to pyrolytically convert a lighter fraction thereof to various products including ethylene. Such a process is disclosed in U.S. Patent 3,060,116. In the former process the boiling range of the feedstock is limited. In the latter process the feedstock boiling range limitation is removed, and a wide boiling range crude petroleum stock is processed; however, the straight run gas oil and residual portions of the feedstock are converted by catalytic cracking and thermal cracking, respectively. Catalytic cracking of the gas oil results in a catalytically cracked gasoline that would be a generally unsatisfactory feed for a pyrolytic cracking zone because of its high content of isoparaffins. This catalytically cracked gasoline also would be an unsatisfactory feed for a catalytic reformer because of its high olefin content. Thermal cracking of the residual portion produces a light thermally cracked gasoline and a heavy thermally cracked gasoline. The heavy thermally cracked gasoline, while suitable as a feed for a catalytic reformer, is produced in low yields.

By-product hydrogen produced in the foregoing types of prior art ethylene manufacturing processes is largely wasted by being dissipated as a fuel gas component.

In view of the foregoing, a useful technological improvement over the process of U.S. Patent 3,060,116, which by utilizing a full boiling range crude feedstock is itself an improvement over the process of U.S. Patent 3,281,351, would be contributed to the art by a process that would accomplish the following purposes without increasing the throughput of the crude feedstock:

(a) Decrease production of less valuable fuel oil, fuel gas and naphtha;

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- (b) Increase yield of more valuable ethylene and butadiene;
- (c) Increase yield of manufactured reformer feedstock;
- (d) Increase quality of manufactured reformer feedstock;
- (e) Produce other more valuable products including benzene and xylenes;
- (f) By increasing yield of manufactured reformer feedstock, release some straight run naphtha reformer feedstock for use as pyrolytic cracking zone feedstock, thereby increasing production of low boiling olefins, pyrolysis naphtha and carbon black feedstock and pitch binder oil;
- (g) Incorporate into higher value liquid fuel products the hydrogen dissipated as a fuel gas component in prior art processes, thereby increasing both yield and quality of said liquid fuel products;
- (h) Permit operation of entire system in hydrogen balance, that is, with the hydrogen requirements of the entire system supplied by hydrogen produced in the system.

It is an object of the present invention to provide a process that will accomplish the foregoing purposes.

Statement of invention

In accordance with one embodiment of the present invention there is provided an integrated hydrocarbon conversion process for converting a crude hydrocarbon feedstock, boiling in the range C₅ to 1400° F., containing substantial quantities of materials boiling in the range C₅ to 430° F. and substantial quantities of materials boiling in the range 350 to 800° F., and having an API gravity in the range 20 to 60, into various valuable products including ethylene and aromatic hydrocarbons, which comprises:

- (a) separating said feedstock into fractions, including a first fraction boiling below 400° F., a second fraction boiling below 430° F., and a third fraction boiling in the range 350° to 800° F., a substantial portion of the materials in said first fraction boiling below the boiling range of said second fraction, and a substantial portion of the materials in said second fraction boiling above the boiling range of said first fraction;
- (b) catalytically hydrocracking said third fraction in a catalytic hydrocracking zone;
- (c) catalytically reforming in a catalytic reforming zone said second fraction and a portion of the effluent from said hydrocracking zone;
- (d) pyrolytically cracking in a pyrolytic cracking zone said first fraction and a portion of the effluent from said hydrocracking zone; and
- (e) recovering as products ethylene from the effluent from said pyrolytic cracking zone and aromatic hydrocarbons from the effluent from said catalytic reforming zone.

In accordance with another embodiment of the present invention said catalytic hydrocracking zone is supplied with hydrogen from said catalytic reforming zone and with hydrogen separated from the effluent from said pyrolytic cracking zone.

In accordance with another embodiment of the present invention there is recycled to said hydrocracking zone from the effluent thereof a fraction boiling in the range 350° to 800° F.

In accordance with another embodiment of the present invention said third fraction boils in the range 450° to 750° F., the portion of the effluent from said hydrocracking zone that is pyrolytically cracked in said pyrolytic cracking zone boils below 450° F. and the portion of the effluent from said catalytic hydrocracking zone that is reformed in said catalytic reforming zone boils below 430° F.

Advantages of the invention

The advantages of the process of the present invention, compared with prior art processes such as the process of U.S. Patent 3,060,116, include accomplishment of the purposes set forth above in connection with "Description of Prior Art." It will be noted that, in accomplishing those purposes, advantage is taken of these facts and discoveries:

(a) As the end point of the feed to the pyrolytic cracking zone rises, the yield of ethylene, based on the feed, decreases. In the process of the present invention the permissible end point of the feed to the pyrolytic cracking zone is higher than in the process of U.S. Patent 3,281,351 or 3,060,116. Accordingly, a smaller yield of ethylene is obtained as a result of pyrolytically cracking the heavier materials not previously sent to the pyrolytic cracking zone, based on the total feed to that zone. However, because those heavier materials are sent to that zone in addition to the lighter straight run materials heretofore cracked in that zone, that is, because the throughput of those lighter straight run materials being fed to that zone is not decreased, there is a greater production of ethylene from that zone on an absolute basis.

(b) The heavier straight run materials that may be sent to the pyrolytic cracking zone in the process of the present invention were sent in previous processes to the reforming zone. These heavier straight run materials have a low naphthene content, and therefore are not an ideal reformer feed. They have a high normal paraffin content, however, and therefore constitute an excellent feed for the pyrolytic cracking zone.

(c) The heavier straight run materials diverted from the reforming zone and sent to the pyrolytic cracking zone in the process of the present invention are replaced with a manufactured reformer feedstock, namely a portion of the effluent from the hydrocracking zone. This portion of the effluent from the hydrocracking zone has a higher naphthene content than the straight run materials it replaces as reformer feedstock, and therefore is a more desirable reformer feedstock.

(d) The catalytic reforming zone in the process of the present invention, supplied with a higher quality reformer feedstock than in processes such as that of U.S. Patent 3,060,116, produces a reformate from which valuable aromatic hydrocarbons can be extracted. From the remaining portion of the reformate, additional quantities of pyrolytic cracking zone feedstock, desirably rich in normal paraffins, is obtained.

Further advantages of the process of the present invention will be apparent from the following description.

Drawing

The accompanying drawing illustrates an embodiment of process units and flow paths suitable for carrying out the process of the present invention.

Description of a preferred embodiment

The preferred embodiment of the process of the present invention that is illustrated in the accompanying drawing will now be discussed.

Referring now to the drawing, a crude hydrocarbon feed is passed through line 1 to crude distillation column 2. The crude hydrocarbon feed may have an extremely wide boiling range, for example C_1 to 1200° F. or wider. It contains substantial quantities of materials boiling in the ranges C_5 to 430° F. and 350° to 800° F. It has an API gravity in the range 20 to 60, preferably 35 to 45. Examples of suitable crude feeds are Middle East, African, North American and South American petroleum crude oils having the foregoing characteristics.

In crude distillation column 2, the crude feed is separated into: a C_1 - fraction, withdrawn through line 3; a C_2 to 150° F. fraction, passed through line 4 to pyrolytic cracking zone 5; a 150° to 300° F. fraction, passed through line 6 to reforming zone 7; a 300° to 450° F.

fraction, passed through lines 8 and 4 to pyrolytic cracking zone 5; a 450° to 750° F. fraction, passed through line 9 to hydrocracking zone 10; and a 750° F.+fuel oil fraction, withdrawn through line 15. The fuel oil withdrawn through line 15 may be burned to supply all or a portion of the thermal energy requirements of the system. All or a portion of that fuel oil may be passed to a conventional thermal cracker or to another hydrocracking zone for further processing. However, it is preferred to minimize the production of the fuel oil and to use it as such rather than subjecting it to further processing.

A portion of the 150° to 300° F. fraction in line 6 may be passed through lines 16 and 4 to pyrolytic cracking zone 5 if desired.

Pyrolytic cracking zone 5 is a conventional high temperature cracking zone, operating at temperatures in the range of about 1200° to 1800° F. or higher and preferably 1400° to 1600° F., at pressures of 0 to 50 p.s.i.g. and residence periods at reaction temperatures in the range of 0.1 to 3 seconds and preferably 0.6 to 1.3 seconds. The pyrolytic cracking zone preferably is operated as a thermal cracker, that is, in the absence of catalysts. However, it is within the purview of the process of the present invention for the pyrolytic cracking zone to contain suitable cracking catalysts, for example, silica-alumina catalysts or mixtures of cracking and dehydrogenation catalysts. When operated in the absence of catalysts the pyrolytic cracking zone preferably is operated with the addition of steam to the zone, the amount of steam added ranging broadly from 30 to 150 percent and preferably from 75 to 125 percent by weight of the hydrocarbon feed. Alternatively, the pyrolytic cracking zone may be operated in accordance with the conventional Wulff process, that is, at the aforesaid high temperatures and low pressures but in the absence of steam. In any event, the pyrolytic cracking zone effects conversion of the feed thereto to a cracked product rich in gaseous olefins and also containing diolefins, aromatics and other products. When pyrolytic cracking zone 5 is operated as a steam cracker, steam may be introduced thereto through line 17.

Hydrocracking zone 10 is a conventional hydrocracking zone supplied with hydrogen through line 18.

The catalyst employed in hydrocracking zone 10 comprises an active cracking catalyst component and at least one hydrogenating-dehydrogenating component. The cracking component may comprise any one or more of such acidic materials as silica-alumina, silica-magnesia, silica-alumina-zirconia, alumina-boria, various acid-treated clays and similar materials. Particularly preferred catalyst cracking components are synthetically prepared silica-alumina compositions having a silica content in the range of from about 15 to 99 percent by weight and an alumina content in the range of from about 1 to 85 percent by weight. The hydrogenating-dehydrogenating components of the catalyst can be selected from any one or more of the various Groups V, VI, VII and VIII metals, as well as from the oxides and sulfides thereof alone or together with promoters or stabilizers that may have by themselves small catalytic effect. Especially suitable catalysts include:

(a) Nickel or a compound thereof in association with silica-alumina;

(b) Nickel or a compound thereof and either tungsten or a compound thereof or molybdenum or a compound thereof in association with silica-alumina.

The amount of hydrogenating-dehydrogenating components present may be varied within relatively wide limits of from about 0.5 to 30 percent based on the weight of the entire catalyst.

Hydrocracking zone 10 is supplied with at least 1,500 s.c.f. of hydrogen per barrel of feed thereto. At least 500, and normally from about 1,000 to 2,000 s.c.f. of hydrogen are consumed in zone 10 per barrel of feed thereto that is converted to synthetic products—i.e., to products boiling below the initial boiling point of said feed. While operation of the process of the present invention in the inte-

grated manner disclosed herein can readily be accomplished in hydrogen balance so that hydrogen supplied from extraneous sources need not be resorted to, extraneous hydrogen nevertheless may be supplied to zone 10 through line 19 if desired.

Hydrocracking zone 10 is operated at a temperature of 400° to 900° F., preferably 550° to 750° F., a pressure of 500 to 3,500 p.s.i.g., preferably 1,000 to 1,500 p.s.i.g., and an LHSV from 0.1 to 15, preferably 0.5 to 5.0. Under these conditions, the feed is converted in amounts exceeding 20 percent per pass to synthetic materials—i.e., materials boiling below the initial boiling point of the feed. The reactions that occur in zone 10 include hydrocracking and isomerization and result in the production of significant quantities of low-boiling, normal paraffins, valuable as feedstock components for pyrolytic cracking zone 5, significant quantities of normal butane and isobutane, valuable for purposes hereinafter discussed, significant quantities of a valuable feedstock for catalytic reforming zone 7 and a normally liquid fraction boiling below 450° F., valuable as an additional feedstock for pyrolytic cracking zone 5.

The effluent from hydrocracking zone 10 is passed through line 20 to separation zone 25. From separation zone 25, a hydrogen stream is recycled to hydrocracking zone 10 through lines 26 and 18, and a fuel gas fraction and an isopentane fraction are withdrawn as products through lines 27 and 28, respectively. From separation zone 25, an ethane fraction, a propane fraction, a normal propane fraction, a normal hexane fraction, a normal isohexane fraction and a fraction boiling in the range 300° to 450° F. are passed through lines 29 to 34, respectively, and thence through lines 35 and 4 to pyrolytic cracking zone 5.

From separation zone 25, a normal butane fraction is passed through line 40 and thence through line 41 for recovery as a product. Alternatively, all or a portion of the normal butane fraction in line 40 may be passed through lines 42 and 43 to isomerization zone 44. From separation zone 25, an isobutane fraction is passed through lines 45, 46 and 47 to alkylation zone 48. From separation zone 25, a fraction boiling in the range C₇ to 300° F. is passed through lines 49 and 6 to reforming zone 7. From separation zone 25, a bottoms fraction boiling above 450° F. is recycled through lines 50 and 9 to hydrocracking zone 10.

Reforming zone 7 is a conventional catalytic reforming zone containing a suitable reforming catalyst having dehydrogenation or aromatization activity, particularly an activity for promoting aromatization of naphthenes to aromatics and desirably for aromatization for paraffinic hydrocarbons to aromatics through a dehydrocyclization reaction. Examples of suitable catalysts are Group VI metal sulfides or oxides, such as the oxides or sulfides of molybdenum, chromium or tungsten, or mixtures thereof, on suitable carriers, such as activated alumina, bauxite, zinc, aluminate or the like. A preferred reforming catalyst comprises about 0.3 to 1.5 percent by weight platinum or palladium associated with a carrier material, such as alumina or silica-alumina. Reforming zone 7 is operated at a temperature of 850° to 1,050° F., preferably 875 to 1,000 F., a pressure of 50 to 1,500 p.s.i.g., preferably 100 to 700 p.s.i.g., a space velocity of 0.5 to 10, preferably 0.5 to 3, volumes of liquid oil feed per hour per volume of catalyst, and a hydrogen-to-hydrocarbon mol ratio of 2 to 20, preferably 4 to 10. Hydrogen may be supplied to reforming zone 7 through line 55 and may comprise hydrogen recycled through line 56, hydrogen supplied through line 57, or both.

The effluent from reforming zone 7 is passed through line 58 to separation zone 59. From separation zone 59, hydrogen is recycled to reforming zone 7 through lines 60, 56 and 55 and also is passed through lines 60, 61, 62 and 18 to hydrocracking zone 10. From separation zone 59 a fuel gas fraction and an isopentane fraction are withdrawn as products through lines 68 and 69, respectively.

From separation zone 59, an ethane fraction, a propane fraction, a normal pentane fraction, a normal hexane fraction and an isohexane fraction are passed through lines 70 to 74, respectively, and thence through lines 75, 35 and 4 to pyrolytic cracking zone 5.

From separation zone 59, aromatic product fractions—namely, benzene, xylenes and C₉+ aromatics—are withdrawn through lines 80 to 82, respectively. From separation zone 59, a toluene fraction is passed through line 83 to conventional dealkylation zone 84 where it is dealkylated, in the presence of hydrogen supplied through line 85, to produce benzene. The effluent from dealkylation zone 85 is passed through line 90 to separation zone 91 from which fuel gas and benzene fractions are withdrawn through lines 92 and 93, respectively. From separation zone 59, a fraction boiling in the range of 400° to 475° F., comprising methyl naphthalenes, is passed through line 94 to conventional dealkylation zone 95 where it is subjected to dealkylation conditions in the presence of hydrogen supplied through line 96. The effluent from dealkylation zone 95 is passed through line 97 to separation zone 98 from which a fuel gas fraction and naphthalene fraction are withdrawn through lines 99 and 100, respectively.

From separation zone 59, a normal butane fraction is passed through line 106 and thence through line 107 as a product. Alternatively, all or a portion thereof may be passed through lines 108 and 43 to isomerization zone 44. From separation zone 59, an isobutane fraction is passed through lines 109, 46 and 47 to alkylation zone 48.

Isomerization zone 44 is a conventional isomerization zone in which the normal butane supplied thereto through line 43 is isomerized to isobutane which is passed through lines 110, 46 and 47 to alkylation zone 48.

The effluent from pyrolytic cracking zone 5 is passed through line 115 to separation zone 116. From separation zone 116, a C₁- fraction is passed through line 117 to conventional hydrogen separation zone 118. The effluent from separation zone 118 is passed through line 119 to separation zone 120 from which a fuel gas fraction is withdrawn through line 121 and from which hydrogen is passed to hydrocracking zone 10 through lines 122, 62 and 18. From separation zone 116, an ethylene fraction is withdrawn as a product through line 123. From separation zone 116, a propylene fraction is passed through line 124 to conventional disproportionation zone 125. The effluent from zone 125 is passed through line 126 to separation zone 127 from which an ethylene fraction is passed through line 128 to line 123 and from which a butylene fraction is passed through lines 129 and 130 to dehydrogenation zone 131. From separation zone 116, a butylene fraction is passed through line 130 to dehydrogenation zone 131. From separation zone 116, a butadiene product fraction is withdrawn through line 140. From separation zone 116, a pyrolysis naphtha fraction, boiling generally in the range C₈ to 450° F. and containing a high percentage of aromatics, is passed through line 141 to conventional hydrogenation zone 142. In zone 142, C₆ and C₇ diolefins in the pyrolysis naphtha are selectively hydrogenated in the presence of hydrogen supplied to zone 142 through line 143. The effluent from hydrogenation zone 142, still containing a high percentage of aromatics, is passed through lines 144 and 58 to separation zone 59 where it is separated into fractions previously discussed which are withdrawn from separation zone 59 as previously discussed. From separation zone 116, a bottoms product fraction useful as a carbon black feedstock and pitch binder oil is withdrawn through line 150.

Dehydrogenation zone 131 is a conventional dehydrogenation zone employing any of a wide variety of known dehydrogenation catalysts of which the commercially available chromia-on-alumina catalyst containing about 20 weight percent Cr₂O₃ is a typical and suitable embodiment. Operating conditions may include temperatures from about 950° to 1,200° F., preferably 1,000° to 1,100° F., relatively low pressures, generally atmospheric or subatmos-

pheric, and 0.5 to 2.1 volumes of charge per volume of catalyst per hour. In dehydrogenation zone 131, the butylene feed is dehydrogenated to produce butadiene. The effluent from dehydrogenation zone 131 is passed through line 160 to separation zone 161 from which hydrogen is recycled to hydrocracking zone 10 through lines 62 and 18 and from which butadiene is recovered through line 162.

A portion of the butylene in line 130 is passed through lines 170 and 47 to alkylation zone 48. Alkylation zone 48 is a conventional alkylation zone in which the isobutane and butylene feeds thereto are reacted together under conventional alkylation conditions to produce to alkylate product which is withdrawn from zone 48 through line 171.

If desired, a portion of the butylene in line 170 may be passed through line 172 to conventional polymerization zone 173. This is a convenient manner of utilizing butylenes which are produced in excess of the amount for which there is isobutane available for alkylation and, therefore, is a convenient way from maintaining isobutane balance in the system. Conventional polymerization zone 173 may be, for example, a bulk liquid acid polymerization zone supplied with liquid phosphoric acid through line 174. The acid may have a concentration of from about 117 to 122 percent. Polymerization zone 173 may be operated at a temperature of from about 175° to 300° F., a pressure of from about 200 to 1,800 p.s.i.g. and a space velocity in excess of about 0.2 volume of hydrocarbon per volume of acid per hour. From zone 173, a motor polymer product is withdrawn through line 175. Acid from the effluent in line 175 is recycled to zone 173 after having been separated in an acid-hydrocarbon settler, which is not shown.

The foregoing detailed description will be further appreciated from a consideration of the following additional points:

- (1) The isopentane that is recovered from separation zones 25 and 59 may be used as a gasoline blend stock;
- (2) The C_9+ aromatic fraction recovered from separation zone 59 may be used to make various valuable products, such as duren or pseudocumene or used as a gasoline blend stock;
- (3) If desired, LPG ($C_3+nC_4+iC_4$) may be recovered as a product from separation zone 25 or separation zone 59, or both;
- (4) If desired, 5 to 15 weight percent of colloidal carbon black may be added to the carbon black feedstock and pitch binder oil recovered from separation zones 59 and 116 to aid the recovered oil in meeting the specifications for pitch binder oil; and
- (5) The separation of aromatics from the other components of the effluent from reforming zone 7, that is accomplished in separation zone 59, may be accomplished by steps including a conventional aromatic extraction step.

From the foregoing, it may be seen that with the process of the present invention there may be produced from a barrel of crude a variety of valuable products, including ethylene, butadiene, carbon black and pitch binder oil, benzene and xylenes. It may be seen that the process enables these results to be obtained with a self-contained operation in which all necessary hydrogen for the operation is produced internally, thereby obviating the need for an external source of hydrogen. The process results in the production, from a low-grade crude fraction, of a high-quality reformer feedstock having a high aromatic and isoparaffin content. The 450° to 750° F. portion of the original crude feedstock, that heretofore has been of little use except as furnace oil or a bunker fuel blend stock, is converted in the hydrocracking zone to lower

boiling and more valuable products and to quality feedstocks for the other conversion units in the combined processing scheme.

Although only specific arrangements and modes of operation of the present invention have been described and illustrated, numerous changes can be made in those arrangements and modes without departing from the spirit of the invention, and all such changes that fall within the scope of the appended claims are intended to be embraced thereby.

What is claimed is:

1. An integrated hydrocarbon conversion process for converting a crude hydrocarbon feedstock, boiling in the range C_5 to 1400° F., containing substantial quantities of materials boiling in the range C_5 to 430° F. and substantial quantities of materials boiling in the range 350° to 800° F., and having an API gravity in the range 20 to 60, into various valuable products including ethylene and aromatic hydrocarbons, which comprises:

- (a) separating said feedstock into fractions, including a first fraction boiling below 400° F., a second fraction boiling below 430° F., and a third fraction boiling in the range 350° to 800° F., a substantial portion of the materials in said first fraction boiling below the boiling range of said second fraction, and a substantial portion of the materials in said second fraction boiling above the boiling range of said first fraction;
- (b) catalytically hydrocracking said third fraction in a catalytic hydrocracking zone;
- (c) catalytically reforming in a catalytic reforming zone said second fraction and a portion of the effluent from said hydrocracking zone;
- (d) pyrolytically cracking in a pyrolytic cracking zone said first fraction and a portion of the effluent from said hydrocracking zone; and
- (e) recovering as products ethylene from the effluent from said pyrolytic cracking zone and aromatic hydrocarbons from the effluent from said catalytic reforming zone.

2. A process as in claim 1, wherein said catalytic hydrocracking zone is supplied with hydrogen from said catalytic reforming zone and with hydrogen separated from the effluent from said pyrolytic cracking zone.

3. A process as in claim 1, wherein there is recycled to said catalytic hydrocracking zone from the effluent thereof a fraction boiling in the range 350° to 800° F.

4. A process as in claim 1, wherein said third fraction boils in the range 450° to 750° F., wherein the portion of the effluent from said catalytic hydrocracking zone that is pyrolytically cracked in said pyrolytic cracking zone boils below 450° F., and wherein the portion of the effluent from said catalytic hydrocracking zone that is reformed in said catalytic reforming zone boils below 430° F.

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