

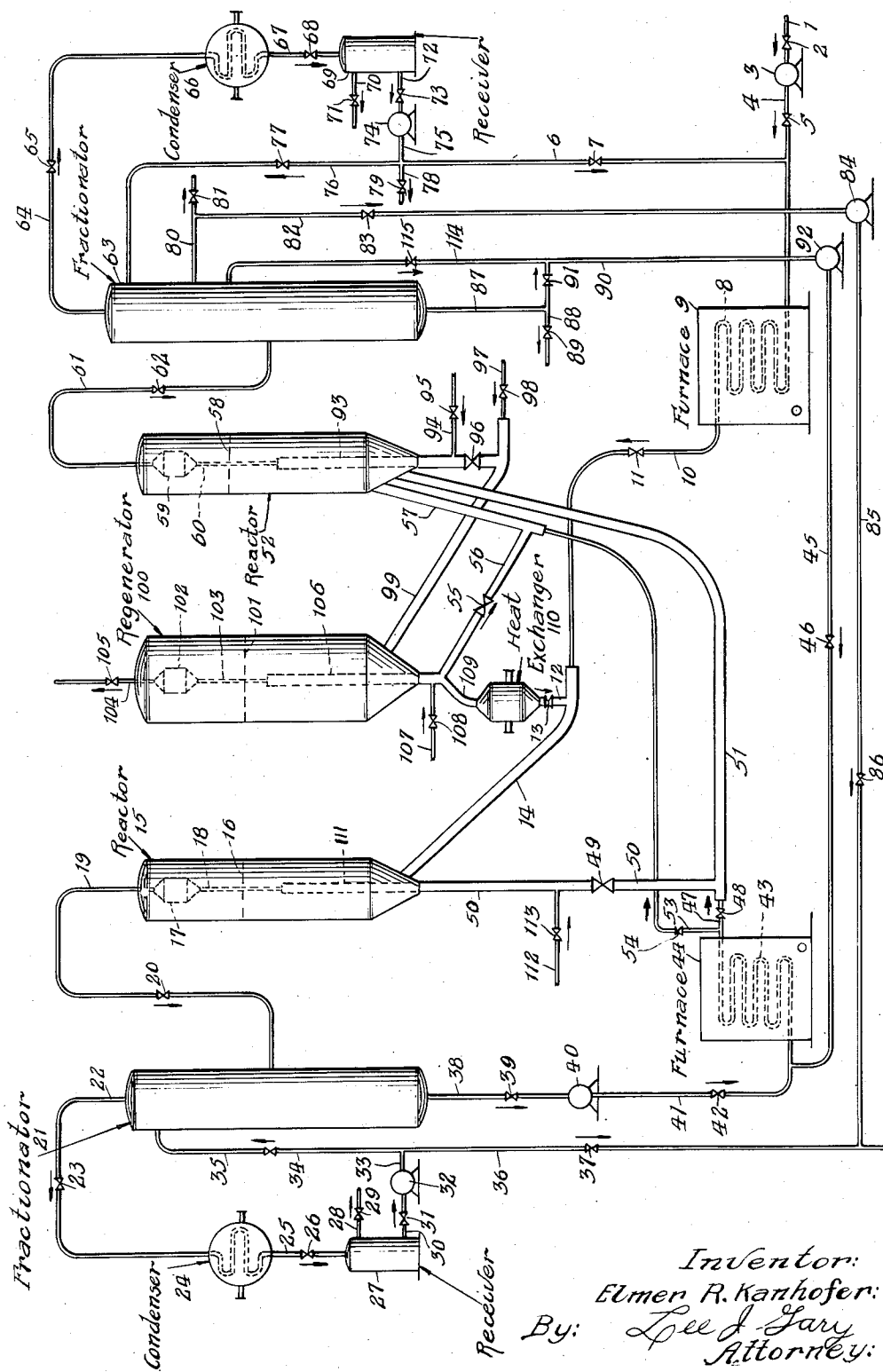
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E. R. KANHOFFER

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PROCESS FOR THE CATALYTIC CONVERSION OF HYDROCARBONS

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Inventor:
Elmer R. Kanhofer:
By: Lee J. Gary
Attorney:

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PROCESS FOR THE CATALYTIC CONVERSION
OF HYDROCARBONS

Elmer R. Kanhofer, Riverside, Ill., assignor to
Universal Oil Products Company, Chicago, Ill.,
a corporation of Delaware

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This invention relates particularly to a two-stage process for the catalytic conversion of hydrocarbon oils into gasoline fractions of high antiknock value. More specifically, it is concerned with a combination of a series of interdependent and cooperative steps for the production of a gasoline fraction low in olefins and suitable for use as an aviation base blending fuel.

Hydrocarbon oils heavier than gasoline, such as kerosene, gas oil, fuel oil, etc., have heretofore been subjected to cracking in the presence of siliceous-type catalysts at temperatures in the range of from about 900° to about 1200° F. to produce gasoline which has a high antiknock value, but which is generally unsuitable as an aviation base blending fuel because of its relatively high olefin content. The high olefin-content gasoline has likewise been heretofore subjected to treatment with a cracking catalyst at a temperature within the range of from about 600° to about 1000° F. under conditions to produce a gasoline fraction which not only retains the high antiknock characteristics of the olefin charge, but also has a relatively low olefin content and a resulting high lead susceptibility. It has also been heretofore proposed to subject a heavier oil in admixture with an olefinic gasoline to conversion in the presence of a cracking catalyst at a temperature within the range of from about 600° to about 1000° F. under conditions to effect cracking of the heavier oil into a gasoline fraction which is low in olefin content and at the same time converting the olefinic gasoline into a gasoline which is low in olefin content, but which retains its high antiknock characteristics. Such a combination process is of the greatest importance in the production of aviation base blending fuel and it is with an improvement in a combination process of this type that the present invention is directed to.

The catalytic masses employed in these reactions comprises particularly the so-called silica-alumina, silica-zirconia or silica-alumina-zirconia composites, made by the separate or simultaneous precipitation of silica and alumina and/or zirconia under conditions such that alkali metal ions are eliminated. Other cracking catalysts, such as acid treated clays may also be used, although not necessarily with equivalent results. Eventually the catalyst requires regeneration in order to restore its activity. In reactivating the catalytic masses, air or other oxygen-containing gases are contacted with the catalyst and regeneration is effected at a temperature which may be of the order of, or higher than, that employed

in the high temperature cracking step. The present invention is concerned with the process utilizing a low temperature cracking step followed by a high temperature cracking step and finally by reactivation of the catalyst.

The present invention is characterized by the following cooperative steps in order to produce high yields of gasoline of low olefin content and particularly suitable for use as aviation base blending fuel:

(1) An oil heavier than gasoline is subjected to treatment in commingled state with an olefinic gasoline in a first conversion step in the presence of a cracking catalyst at a temperature of from about 600° to about 1000° F. under conditions to produce a gasoline fraction of low olefin content. The products of this conversion step are separated into a low boiling gasoline fraction having an end boiling point of from about 200° to about 250° F., which in some cases may be as high as 300° or even up to 400° F., and higher boiling products.

(2) The higher boiling products from the first conversion step are subjected to cracking in a second conversion step in contact with a siliceous catalyst and particularly the spent catalyst from the first conversion step, the second conversion step being at a temperature of from about 900° to about 1200° F. and under conditions to produce an olefinic gasoline. The olefinic gasoline is then supplied to the first conversion step for treatment in the manner heretofore described.

In addition to the above, the present invention includes the following important features:

(1) The low boiling gasoline fraction recovered from the second conversion step of the process will have an end boiling point similar to or higher than the low boiling gasoline fraction recovered from the first conversion step of the process as heretofore described. Hydrocarbon oil boiling above this low boiling gasoline fraction may be recycled to the second conversion step in order to obtain increased yields or other improvements in the desired gasoline fraction.

(2) When desired, an intermediate naphtha fraction, such as one having an initial boiling point within the range of from about 200° to about 300° or 350° F., and an end boiling point within the range of from about 300° to about 400° F., may be separated and withdrawn from the products of the second conversion step and all or a portion of this naphtha may be blended with the low boiling low olefin content gasoline recovered as the final product of the process from the first conversion step. The naphtha fraction

will be low in olefin content and high in aromatic content so that it is particularly suitable for blending with the low boiling gasoline fraction for the production of aviation gasoline.

(3) Depending on the balance between the transfer of the catalyst in the various stages of the process, provision is also made for supplying a portion of the freshly regenerated catalyst to a second conversion step.

The present invention is particularly applicable to the treatment of oil in a process employing a moving bed of catalyst. The type of process now known as the "Fluidized operation" is especially preferred in the present invention. In this type of operation the catalyst powder is maintained in constant agitation by a rising stream of reactants and the net upward flow of the reactants is greater than that of the catalyst particles. In both the moving bed type of process and the fluidized operation there is usually a transfer of catalyst from one zone to another zone, the transfer being accomplished by any suitable means including, for example, suspension or mixture of the catalyst in the reactants or in any other suitable transfer medium, mechanical conveyors, etc.

In a broad aspect the present invention relates to a process for the manufacture of gasoline of low olefin content and of high antiknock value which comprises subjecting an oil heavier than gasoline in commingled state with an olefinic gasoline to contact in a first conversion step with a siliceous catalyst under conditions to produce a low boiling gasoline fraction of low olefin content, separating the same from heavier hydrocarbons and subjecting the latter to cracking in a second conversion step in contact with a siliceous catalyst under conditions to produce a low boiling olefinic gasoline, and supplying said low boiling olefinic gasoline to treatment in the first step as heretofore described.

The term olefinic gasoline or gasoline of high olefin content is used in the present specification and claims to mean the type of gasoline normally produced by cracking of heavier oil in contact with a cracking catalyst under temperatures of the order of about 900° to about 1200° F. at a weight hourly space velocity which is usually above 1. The term weight hourly space velocity is defined as the weight of oil per hour per weight of catalyst in the reaction zone. On the other hand, the gasoline of low olefin content is intended to mean the gasoline produced by treatment of a heavier oil in commingled state with a high olefin content gasoline in the presence of a cracking catalyst at temperatures of the order of about 600° to about 1000° F. but at a weight hourly space velocity which is usually below 1. However, it is understood that the temperature and space velocity may be correlated in order to produce either high olefin or low olefin content gasolines and also that these operating conditions will be dependent upon the activity of the particular catalyst employed. Thus in the first conversion step in which a low olefin content gasoline is produced, if the temperature employed is within the upper limits of the range heretofore given, the space velocity will be correspondingly increased. Likewise, if the temperature employed is within the lower limits of the range heretofore specified, the space velocity may be correspondingly reduced. A similar situation occurs in connection with the second conversion step for the production of high olefin content gasoline.

It is emphasized that the reaction in the first

conversion step of the process, in which the heavier oil and the olefinic gasoline are treated in commingled state, serves to increase the yield of the low boiling low olefin-content gasoline produced in the process. It is believed that hydrogen from the heavier oil is transferred to the low boiling olefinic hydrocarbons during the reaction and at the same time the heavier oil is cracked to produce additional low boiling olefins, which are likewise saturated by the hydrogen released from the heavier oil. Whatever portions of the heavier oil which are not converted under the relatively mild conversion conditions of the first step of the process are subsequently in the second step of the process converted under the more severe conditions employed therein.

In one specific embodiment the present invention relates to a process for the manufacture of aviation base blending fuel which comprises subjecting a mixture of raw oil heavier than gasoline and an olefinic gasoline formed in the process, in the manner to be hereinafter described, to contact in a first conversion step with an active cracking catalyst under fluidized conditions at a temperature within the range of from about 600° to about 1000° F. at a weight hourly space velocity of not greater than 1 to produce a low boiling gasoline fraction of low olefin content, separating the low boiling low olefin content gasoline from heavier hydrocarbons, subjecting said heavier hydrocarbons to cracking in a second conversion step in contact under fluidized conditions with at least a portion of the catalyst which has previously been employed in the first conversion step, the second conversion step being effected at a temperature of from about 900° to about 1200° F. at a weight hourly space velocity of greater than 1, separating the products of the second conversion step into a low boiling high olefin-content gasoline fraction and heavier hydrocarbons, supplying at least a portion of said low boiling high olefin content gasoline to treatment in the first conversion step as heretofore described, and recycling at least a portion of said heavier hydrocarbons to said second conversion step for further treatment therein.

The invention will be more fully explained in connection with the attached diagrammatic flow drawing which illustrates several specific embodiments of the invention. The embodiments in the invention illustrated describe a process using fluidized operation which, as heretofore stated, is preferred. However, the invention may be practiced in a process using other types of operation.

Referring to the drawing, a hydrocarbon charging oil such as kerosene, gas oil, or any other hydrocarbon oil heavier than gasoline, is supplied through line 1 and valve 2 to pump 3, by means of which it is directed through line 4 and valve 5 and is admixed with a low boiling high olefin content gasoline fraction formed in the process, in the manner to be hereinafter described in detail, and directed through line 6 and valve 7 to commingle with the charging oil in line 4. The mixture is then directed into and through heating coil 8 in furnace structure 9, wherein the mixture is heated to the desired temperature. Heated products are then directed through line 10 and valve 11, whereafter they are commingled with fresh or reactivated powdered catalyst introduced through standpipe 12 and valve 13 and obtained in the manner to be hereinafter described in detail. The mixture of oil and catalyst is then directed through line 14 into reactor 15

which comprises the first conversion step of the process for the production of a low boiling low olefin content gasoline fraction.

In reactor 15 a turbulent mass of catalyst powder is maintained in constant agitation by a rising stream of reactants introduced by means of line 14 and the conversion products formed during the reaction. The top of this turbulent bed is indicated diagrammatically by line 16. As previously mentioned, the temperature in this zone will ordinarily range from approximately 600° to approximately 1000° F. and preferably will be from about 850 to about 950° F. Pressure in this zone will ordinarily be slightly above atmospheric and may be of the order of 5 to 25 pounds per square inch gauge, although in some cases pressures as high as 100 pounds per square inch gauge or more may be employed. Weight hourly space velocities will usually not be greater than 1, although, as heretofore mentioned, higher space velocities may be employed depending upon the temperature utilized.

Vaporous conversion products containing some suspended catalyst are withdrawn from the top of reactor 15 through cyclone separator 17 wherein entrained catalyst is separated from the vaporous conversion products and returned through line 18 to the reactor. The conversion products are then withdrawn through line 19 and valve 20 and are supplied to fractionator 21 wherein a low olefin content gasoline fraction is separated from heavier hydrocarbons.

As heretofore mentioned, the low olefin low boiling gasoline fraction may have an end boiling point within the range of from about 200° to about 400° F. and more generally will have an end boiling point of from about 200° to about 250° F. This fraction is withdrawn from the upper portion of fractionator 21 through line 22 and valve 23 into and through condenser 24, through line run-down 25 and valve 26 into receiver 27. In receiver 27, a separation is effected between the gaseous phase and the liquid phase, the gaseous phase being withdrawn through line 28 and valve 29 to storage or elsewhere as desired. This gaseous phase will contain a relatively high proportion of isobutane and is particularly suitable for use in subsequent alkylation or other conversion treatments. The distillate collected in receiver 27 is withdrawn therefrom through line 30 and valve 31 to pump 32, by means of which it is directed through line 33 and a portion thereof recycled by way of line 34 and valve 35 to the upper portion of fractionator 21 to serve as a refluxing and cooling medium therein while the remaining portion of the distillate is directed through line 36 and valve 37 and is recovered as the final low boiling gasoline product of the process.

The heavier oil separated in fractionator 21 is withdrawn therefrom through line 38 and valve 39 to pump 40, by means of which all or a portion thereof is supplied through line 41 and valve 42 to heating coil 43 which is positioned in furnace structure 44. Prior to its introduction into heating coil 43, the heavier oil may be commingled with a recycle oil formed in the process, in the manner to be hereinafter described, and directed through line 45 and valve 46 into line 41. The heavier oil introduced into line 43 is heated therein to the desired temperature and is then directed through line 47 and valve 48, whereafter it is commingled with a partly spent powdered catalyst introduced through valve 49 and standpipe 50 and obtained in the manner to be hereinafter described in detail. The resulting mixture

of catalyst and oil is then directed through line 51 into reactor 52. When desired, a portion of the heavier oil may be diverted from line 47 through line 53 and valve 54, to commingle with a portion of the freshly regenerated catalyst withdrawn through valve 55 and standpipe 56, the resulting mixture being directed through line 57 into reactor 52.

Reactor 52 may be similar in design to reactor 15 and thus line 58 indicates the top of the turbulent bed, numeral 59 indicates the cyclone separator and line 60 is provided for the return of catalyst back to the reactor. However, as heretofore mentioned, reactor 52 will be operated at a temperature within the range of from about 900° to about 1200° F. at a weight hourly space velocity which will usually be greater than 1, these operating conditions being dependent upon the activity of the particular catalyst employed.

The vaporous conversion products containing some suspended catalyst are directed through cyclone separator 59 in which the catalyst is separated out and the hydrocarbon conversion products are then directed through line 61 and valve 62 into fractionator 63. In fractionator 63 a low boiling high olefin content gasoline fraction is separated and this fraction may have an end boiling point similar to or different than that heretofore described in connection with the overhead products withdrawn through line 22 from fractionator 21. Usually this fraction will have an end boiling point within the range of from about 200° to about 250° F. and is directed through line 64 and valve 65 into and through condenser 66 and run-down line 67 containing valve 68 into receiver 69. The gases are released from receiver 69 through line 70 and valve 71. These gases will be relatively rich in olefins and therefore are particularly suitable for use in either alkylation, polymerization, or other suitable conversion processes. The distillate collected in receiver 69 is withdrawn therefrom through line 72 and valve 73 to pump 74, by means of which it is supplied through line 75 and a portion thereof recycled by way of line 76 and valve 77 to the upper portion of fractionator 63 to serve as a cooling and refluxing medium therein. A portion of the distillate from receiver 69 may be withdrawn to storage or elsewhere as desired through line 78 and valve 79 but, in accordance with the present invention, at least a portion of the distillate is directed through line 80 and valve 81 to commingle with the hydrocarbon charging oil introduced through line 1 in the manner heretofore described.

In one embodiment of the invention an intermediate naphtha fraction having an initial boiling point within the range of from about 200° to about 350° F. and an end boiling point within the range of from about 250° to about 400° F., preferably one having an initial boiling point within the range of from about 200° to about 300° F. and an end boiling point within the range of from about 250° to about 350° F., may be separated from other products in fractionator 63 and withdrawn therefrom through line 80 and valve 81 to storage or elsewhere as desired. In the production of aviation gasoline, it is a particular feature of the present invention that a naphtha fraction having an initial boiling point from about 200° to 250° F. and an end boiling point from about 275° to about 325° F. is withdrawn through line 80 and is directed through line 82 containing valve 83 to pump 84, by means of which it is supplied through line 85 and valve

86 into line 36 or otherwise commingled with the low boiling low olefin gasoline fraction. The naphtha fraction will be low in olefin content and high in aromatic content so that it is particularly suitable for use as a blending agent in aviation gasoline.

The heavier oil separated in fractionator 83 may be withdrawn therefrom through line 87 and withdrawn from the process through line 88 and valve 89 to storage or elsewhere as desired. When desired, the heavier oil may be directed through line 90 and valve 91 to pump 92 by means of which it is recycled by way of line 45, valve 46 and line 41 to heating coil 43 for subsequent further treatment in reactor 52. It is also within the scope of the invention to separate a heavy naphtha fraction in fractionator 83 and withdraw the same through line 114 and valve 115. This heavy naphtha fraction will have an initial boiling point above the intermediate naphtha fraction withdrawn through line 80 and an end boiling point within the range of from about 400° to about 500° F. The heavy naphtha fraction may be recycled for further treatment by way of lines 90, 45, 41, 47 and 51 into reactor 52.

The spent catalyst in reactor 52 is directed into standpipe 93 wherein the catalyst is stripped of its volatile hydrocarbons absorbed therein by an upwardly moving stream of gas, such as steam, fluegas, etc., introduced through line 94 and valve 95. This gas not only serves to strip volatile hydrocarbons from the catalyst but also serves to maintain the powdered catalyst in a fluidized state so that the suspension will readily flow like a liquid. The spent catalyst is supplied through valve 96 and is picked up by a stream of air or other suitable oxidizing gas introduced through line 97 and valve 98 and the mixture is supplied through line 99 to regenerator 100 wherein the hydrocarbonaceous deposit formed on the catalyst is removed from the catalyst by a combustion reaction. In the regenerator, as in the reactors, a turbulent bed of powdered catalyst is maintained by means of an upwardly flowing stream of oxygen-containing gas. The upper limit of this bed is illustrated diagrammatically by line 101. Above this point, the concentration of the catalyst steadily diminishes, while below this point, a very high concentration of catalyst is maintained.

Spent reactivated gases are withdrawn from the regenerator through cyclone separator 102 wherein entrained catalyst is thrown out by centrifugal action and returned to the catalyst bed through line 103. Vent regenerating gases are removed through line 104 containing valve 105. It is usually necessary to remove heat from the regenerator in order that the regeneration temperature will not exceed a certain maximum, and any suitable means, not illustrated, may be employed for this purpose such as, for example, cooling and recycling a portion of the regenerated catalyst or the positioning of a closed cooling coil within the regeneration zone.

The regenerated catalyst is withdrawn from the regenerator through standpipe 106 wherein the catalyst is contacted with an upwardly moving stream of inert gases, preferably steam or flue gases, introduced through line 107 containing control valve 108, to desorb any oxygen which may be present with the catalyst. The catalyst withdrawn through standpipe 106 may be supplied in part through line 56 as heretofore mentioned, but at least a portion of the regenerated catalyst is directed through line 109 to be picked

up by the heated charging oil being supplied through line 10 as heretofore described. Heat exchanger 110 is provided in case it is desired to either cool or heat the catalyst prior to introduction to reactor 15. Usually the catalyst is at too high a temperature when withdrawn from the regenerating zone and heat exchanger 110 suitably comprises a cooling zone. In some cases it may be satisfactory to heat the charging oil in coil 8 to a lower temperature and then obtain additional heat from the hot regenerated catalyst, thus eliminating heat exchanger 110.

The catalyst in reactor 15, after it has served to effect conversion of the charging oil and low boiling high olefin gasoline, is withdrawn through standpipe 111 into line 50. Line 112 containing valve 113 is provided for the introduction of a suitable stripping medium to remove volatile hydrocarbons from the catalyst and to maintain the powdered catalyst in fluidized state. It is a particular feature of the present invention that this catalyst, after having been used in the first conversion step, will retain sufficient activity for use in the second conversion step of the process.

The following example is introduced for the purpose of further illustrating the novelty and utility of the present invention but not with the intention of unduly limiting the same.

A 32° A. P. I. gravity Mid-Continent gas oil may be commingled with an olefinic gasoline fraction having an end boiling point of about 250° F. and the mixture subjected to conversion in the presence of a silica-alumina catalyst at a temperature of about 850° F. and an hourly weight space velocity of .5. A low olefin, high antiknock gasoline fraction having an end boiling point of about 250° F. may be separated from the conversion products and the heavier hydrocarbon oil commingled with the spent catalyst from the first conversion step and subjected to cracking at a temperature of about 950° F. and at a weight hourly liquid space velocity of about 1.7.

On the basis of an operation such as described above, the following products are obtainable, these products being reported on the basis of weight per cent of the gas oil charging stock:

Table

	First step	Second step	Total
C ₁ and lighter.....	8.9	6.7	15.6
Butylenes.....	3.3	1.7	5.0
Isobutane.....	7.3	3.7	11.0
Normal butane.....	1.1	0.6	1.7
Amylenes.....	2.8	1.1	3.9
Isopentane.....	5.8	2.2	8.0
Normal pentane.....	0.8	0.3	1.1
C ₁ to 250° F.....	12.3	7.2	19.5
250° to 340° F.....		5.0	5.0
340° to 400° F.....		7.6	7.6
Recycle stock.....		11.4	11.4
Coke.....	6.4	3.8	10.2

It is to be noted from the above table that 19.5% of low boiling low olefin content gasoline, based upon the gas oil charge, is produced. This gasoline is of high antiknock value and is particularly suitable for use as aviation base blending fuel. In addition there is produced 5% of naphtha boiling between 250° and 340° F. and 7.6% of naphtha having an end point of 400° F. All or a portion of the 340° F. end boiling point naphtha may be blended with the low boiling gasoline fraction to increase the yield of aviation base gasoline stock.

It is to be further noted that there also is produced 11.0% of isobutane, 8.0% of isopentane

as well as 5% of butylenes. The isopentane may be blended with the aviation gasoline, while the isobutane and butylenes may be utilized in alkylation reactions for the production of an alkylate which may also be used in the aviation gasoline.

The above results are compared to a two-step process in which the charging stock is cracked at a temperature of about 950° F. in the first step and the gasoline product therefrom is subjected to treatment in a second stage at a temperature of about 900° F. In this operation there is produced 16% of a low olefin high antiknock gasoline containing hydrocarbons having 6 carbon atoms to the molecule up to 340° F. end boiling point. It is to be noted that the 16% of desired gasoline for aviation base blending fuel from the last mentioned treatment is lower than the 24.5% of similar low boiling high antiknock gasoline (19.5% C₆ to 250° F. + 5% 250° to 340° F.) produced in accordance with the present invention. Likewise, the percent of isobutane produced in the other type of operation is 8.5% which is lower than 11.0% produced in accordance with the present invention. Both of these operations are based upon a similar coke production of 10.2% by weight of the charging stock.

I claim as my invention:

1. A process for the manufacture of a gasoline of low olefin content, high antiknock value and suitable for use in aviation gasoline which comprises commingling an oil heavier than gasoline with a gasoline of higher olefin content than said first mentioned gasoline, and utilizing the commingled stream to convey freshly regenerated powdered cracking catalyst into a first reaction zone operated at a temperature within the range of from about 600° to about 1000° F. and under conditions whereby the net upward flow of hydrocarbons is greater than that of the catalyst and to produce a low olefin content gasoline, withdrawing the vaporous conversion products from said reaction zone and separating the same into a low olefin content gasoline having an end boiling point within the range of from about 200° to about 300° F., and heavier hydrocarbons, recovering said low olefin content gasoline as the product of the process, withdrawing the partly spent cracking catalyst from said reactor and utilizing said heavier hydrocarbons to convey said partly spent catalyst into a second reaction zone operated at a temperature within the range of from about 900° to about 1200° F. and under conditions whereby the net upward flow of hydrocarbons is greater than that of the catalyst and to produce a high olefin content gasoline, withdrawing the vaporous conversion products from the second reaction zone and separating the same into a high olefin content gasoline having an end boiling point within the range of from about 200° to about 300° F. and heavier hydrocarbons, commingling said high olefin content gasoline with said oil heavier than gasoline as aforesaid, withdrawing the spent catalyst from said second reaction zone and conveying the same by means of an oxygen-containing gas to

a regenerating zone in which the catalyst undergoes regeneration, withdrawing the regenerated catalyst from the regenerating zone and supplying the same to said first reaction zone in the manner heretofore described.

2. The process of claim 1 further characterized in that a portion of said regenerated catalyst is supplied to the second reaction zone by means of a portion of the heavier oil separated from products of the first reaction zone.

3. In the catalytic cracking of hydrocarbons wherein used cracking catalyst from the hydrocarbon conversion is regenerated in a regenerating zone, the process which comprises removing regenerated catalyst from said zone and introducing the same to a first conversion zone, simultaneously introducing to said conversion zone hydrocarbon oil heavier than gasoline and olefinic hydrocarbons boiling in the gasoline range, contacting the hydrocarbons with the catalyst in said conversion zone under conditions to produce gasoline of low olefin content therefrom, separately removing vapors and partially spent catalyst from the conversion zone, fractionating the vapors to separate fractions thereof heavier than gasoline, introducing at least a portion of said fractions and said partially spent catalyst to a second conversion zone maintained at higher temperature than said first conversion zone and therein cracking the heavier than gasoline fractions in the presence of the partially spent catalyst to produce olefinic gasoline therefrom, separately removing vapors and used catalyst from said second zone and introducing the latter to said regenerating zone, fractionating the last-mentioned vapors to separate olefinic gasoline therefrom, and supplying at least a portion of said olefinic gasoline to the first conversion zone.

4. The process as defined in claim 3 further characterized in that the first conversion zone is maintained at a temperature of from about 600° F. to about 1000° F. and the second conversion zone at a temperature of from about 900° F. to about 1200° F.

5. The process as defined in claim 3 further characterized in that the first conversion zone is maintained at a temperature of from about 600° to about 1000° F. and the second conversion zone at a temperature of from about 900° F. to about 1200° F., the hydrocarbons in said first zone being contacted with the catalyst therein at a weight hourly space velocity not greater than 1 and the hydrocarbon in said second zone being contacted with the catalyst therein at a weight hourly space velocity greater than 1.

6. The process as defined in claim 3 further characterized in that reflux condensate from the second-mentioned fractionating step is recycled to said second conversion zone.

7. The process as defined in claim 3 further characterized in that freshly regenerated catalyst is supplied from the regenerating zone to said second conversion zone.

ELMER R. KANHOFER.