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2,956,005

COMBINATION REFORMING AND SOLVENT  
EXTRACTION PROCESS

Irvin H. Lutz and Herbert D. Holdridge, Jr., Texas City,  
Tex., assignors to The American Oil Company, Texas  
City, Tex., a corporation of Texas

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This invention relates to the production of high octane gasoline from relatively low octane naphthas by catalytic reforming. More particularly, it combines catalytic reforming with selective fractionation and extraction in a manner increasing the potential barrel-octanes of gasoline from a given feed stock, i.e. raising either the product volume or the product octane while holding the other constant or both. It provides lower incremental cost per incremental octane member, particularly at the higher octane levels, i.e. 95 CFR-R and higher. The process is also designed to provide greater flexibility with respect to feed stock availability, catalyst suitability and process design limitations.

Steadily increasing octane levels for both premium and regular gasoline have created serious refining problems. The problems are basically economic in nature because within the limits of high quality feed stock and catalyst availability, it is possible, theoretically at least, to produce gasoline of the present octane level and that projected for the near future. The increasing volume of the demand for high octane premium gasoline (i.e. about 98 CFR-R and higher) however is working a serious dislocation in the structure of the domestic refining industry. In general, refiners have been relying on the existing large capacity of catalytic cracking facilities to produce high octane base stocks for premium gasoline production. Reforming has been used in general to upgrade virgin and other low-grade naphthas to permit blending into the total gasoline pool (premium plus regular) without disturbing the weighted average octane number of the pool necessary to meet competition. In some instances where high quality naphthas containing relatively high percentages of naphthenes were available, reforming has also been used to produce special, high octane blending stocks. However, as premium requirements have reached the 98+ CFR-R level, the limit of feasibility of catalytic gasoline as the major premium base stock is reached because catalytic gasoline rarely exceeds about 92-93 CFR-R clear and is not high enough in blending octane to blend to above about the 97 level in leaded gasoline. Thus, a drastic shift in the function of catalytic reforming is demanded, but as the level of reforming severity is lifted to meet premium blending standards instead of regular or pool blending standards, costs are greatly magnified. There is a pronounced drop-off in recoverable liquid yield of reformate as reforming severity is increased. Hence, there is an effective decrease in reforming plant capacity. Reforming catalyst life also is sharply reduced as severity is increased. The operating costs per barrel are increased, and since catalyst cost is a major cost factor, the effect is a significantly increasing factor in the incremental cost of the higher octane gasoline. Space velocity (the rate of flow of feed in units of feed flow per unit of catalyst charge) must be reduced to achieve higher octane levels with the same catalyst so that again plant capacity is lost, or catalyst inventory must be significantly increased. Moreover, the availability of suitable feed stocks becomes more critical. In-

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stead of serving to dispose of low grade stocks by economical upgrading, the character of the feed and its availability become determinative factors in the cost and volume of premium gasoline production.

5 This summary of the economic situation affecting reforming indicates that conventional once-through catalytic reforming may suffer severe future limitations unless the catalyst available has unusually high activity which is maintained by frequent regeneration in an economic  
10 manner and unless a sufficient volume of suitable feed stock is available. As an alternative to once-through, regenerative reforming, several schemes involving extraction of reformates followed by recycle or separate reforming of the raffinate have been proposed. In a sense,  
15 these processes substitute selective extraction of reformate and raffinate rerunning for reforming severity and catalyst regeneration. They attempt to operate for relatively long periods without regeneration by using high pressures of hydrogen, e.g. 500-700 p.s.i.g. They are costly both  
20 investment-wise and in operation, and they encounter limitation in the potential barrel-octanes of high octane gasoline producible because, for example, of unfavorable equilibrium conditions encountered in the reforming step. Nevertheless, they have commanded attention because  
25 they have seemed the most logical, and perhaps only reasonable alternatives, to increasing once-through severity by catalyst and process design means.

The present invention applies a number of new operating factors. It has been found that there is a relationship between reforming severity, particularly with  
30 the use of a platinum type reforming catalyst in a low pressure operation, and the distribution of aromatics in the reformate. As the reforming severity reaches the level of about 90 CFR-R octane clear, and particularly  
35 as it exceeds 95 octane clear, the concentration of aromatics in the heavier portion of the reformate becomes such that upwards of 40 to 65 volume percent of a heavy fraction containing at least about 75 volume percent aromatics can be recovered by simple fractionation at a cut-  
40 point in the range of about 220° to 300° F. The resulting stocks have extrapolated octanes exceeding 100 CFR-R clear. At severity levels below 90 octane clear, there is considerable variation in the relationship of octane number vs. aromatics content with the source of the naphtha  
45 feed, that is whether the feed is a high naphthenic Mid-Continent naphtha or a paraffinic type Middle East naphtha, for example. Surprisingly, above 90 octane number it has been found that the nature of the feed source is immaterial so that a high octane heavy reformate can  
50 be produced from any feed of appropriate boiling range by correlation of reforming severity with reformate cut-point based on aromatics content. The light reformate, usually comprising about 35 to 60 volume percent of the total, is of relatively low octane number, e.g. about  
55 75 to 80 CFR-R clear, but at these reforming severities will contain a considerable amount of aromatics which can be selectively extracted at relatively low cost. It has also been found that as reforming severity is increased, particularly in the range above 95 octane clear,  
60 the volume of very high octane stock, i.e. 100+ clear, that can be produced is significantly increased. The present invention is directed to an integrated process which provides cost economies but with the capability and flexibility to accomplish variable refining objectives  
65 depending upon the refining limitations encountered. An object of the invention, for example, is to provide a process maximizing barrel-octanes at any level required for production of 100+ octane gasolines. Another object is to provide process flexibility with respect to feed,  
70 catalyst and plant design. A further object is to reduce the incremental cost for incremental increases in the octane level of premium gasoline and of the overall

gasoline pool. Alternatively, costs can be reduced for attaining given octane goals. A further object is to provide a feasible means for octane upgrading beyond design and/or catalyst limits in particular instances of installed capacity.

According to the invention, the naphtha charge, which may be any naphtha boiling in the range of say about 150° or 200° F. to 400° F. but advantageously is a C<sub>7</sub>+ naphtha, is contacted in a reforming zone with a reforming type catalyst in the presence of recycle hydrogen. The reforming is conducted under typical reforming conditions of temperature, space velocity and hydrogen to feed ratios. A relatively low total pressure, in the range of about 50 to 400 p.s.i.g., preferably about 200 to 300 p.s.i.g., is used. The reforming conditions are such as to provide a severity with the catalyst used producing a C<sub>5</sub>+ reformat of at least about 90 CFR-R clear. The reformat is fractionated at a cut-point in the range of about 220° to 300° F. so as to obtain a heavy fraction containing at least 75 volume percent aromatics and a lighter fraction. As the reforming severity is increased, the cut-point is at a lower temperature in the above range; thus for a severity giving 95+ octane reformat, the cut-point is in the range of about 220° to 270° F. The lighter reformat fraction is extracted by treatment with an extractive agent which is selective for aromatics to recover an aromatics rich extract fraction. The extractive agent preferably is a liquid solvent, e.g. diethylene or triethylene glycol, but may be one of the selective solid adsorbents, e.g. silica gel, known in the art. The extract is separated from the extractive agent and is blended with the heavy reformat fraction to provide a very high octane stock for gasoline blending.

The general flow of the process of the invention is illustrated in Figure 1 of the accompanying drawings which is a simplified process flow diagram. A more specific example of the invention is illustrated in the somewhat more detailed flow diagram of Figure 2 of the drawings.

In the drawings, the naphtha feed through line 10 is charged to a conventionally fired heater 11. The preheated feed in line 12 may be combined with hot recycle hydrogen gas from heater 13 and line 14. The charge mixture is contacted in reforming zone 15 with a reforming type catalyst in a manner and under reforming conditions described in further detail below. The reformed mixture passes via connection 16 to cooler 17 and thence via connection 18 to high pressure separator 19. Gas, predominantly hydrogen, is separated from vessel 19 through line 20 for recycle by means of a recycle compressor or blower system 21 and line 22. The net production of gas in the process may be withdrawn from the system as by connection 23. The liquid reformat separated in vessel 19 is withdrawn through line 24 and is charged via preheater 25 and line 26 to debutanizer or stabilizer tower 27. In typical operation, light ends are taken overhead from tower 27 through line 28 and cooler 29 to receiver 30. A portion of the liquid collected in receiver 30 is returned to tower 27 as reflux through line 31, pump 32 and connection 33. The remainder, usually comprising a mixed C<sub>3</sub>-C<sub>4</sub> light hydrocarbon fraction is withdrawn through line 34. An external steam reboiler 35 supplies heat to tower 27. Liquid from the tower is circulated by means of lines 36 and 37 through reboiler 35.

The stabilized reformat is withdrawn from tower 27 by line 38 and is charged to splitter tower 39. Heat is supplied to tower 39 as by steam reboiler 40. Liquid is withdrawn from the lower portion of the tower through line 41, is passed through reboiler 40 and returned to tower bottoms via line 42. The operation of splitting tower 39 is controlled to remove the heavy reformat fraction containing upwards of about 75 to 80 volume percent aromatics (100+ octane) through line 43. A lighter fraction is taken overhead through line 44 and cooler 45

to receiver 46. A portion of the liquid collected in receiver 46 is returned to the tower top by means of line 47, pump 48 and line 49. Net product is light reformat, containing about 15 to 50 volume percent aromatics, which is charged by means of line 50 to an aromatics extraction process conducted in zone 51. Extract enriched in aromatics and increased in octane to the 98+ CFR-R clear level is produced in zone 51 and is suitable for blending, as shown by means of line 52, with heavy reformat recovered in line 43. Therefore, as shown, the two aromatic rich products are passed, as by common line 53, to gasoline blending facilities. If desired, a portion of the light aromatic extract may be diverted as through connection 54 to fractionation facilities for recovering pure aromatics, e.g. benzene, toluene, and/or xylenes. The raffinate produced in zone 51 is of low octane. It can be recycled to reforming zone 15 via line 55, or it can be subjected to a wide variety of selective conversions to improve its quality. Alternatively, it can be blended off by mixing with other refinery streams.

As indicated above, it is essential that the reforming process conducted in zone 15 be conducted under conditions of severity such that a C<sub>7</sub>+ fraction of the feed would produce a C<sub>5</sub>+ reformat of at least about 90 octane number clear. Also, to obtain the desired distribution of aromatics in the reformat, the reforming should be conducted at relatively low pressure favoring dehydrocyclization rather than hydrocracking. Ordinarily, the average reaction temperature should be in the range of about 850° to 1000° F. at a pressure in the range of about 50 to 400 p.s.i.g., advantageously about 200 to 300 p.s.i.g. The recycle hydrogen rate should approximate 1,000 to 10,000 cubic feet per barrel. The space velocity will depend on the activity of the catalyst but ordinarily will be in the range of about 0.1 to 5.0 WHSV. The catalyst may comprise any of the active reforming catalysts such as platinum-on-alumina or platinum-on-silica alumina, molybdenum oxide-alumina or alkaliized chromium oxide-alumina. Advantageously, however, the catalyst is of the platinum-alumina type containing about 0.1 to 1.0 weight percent platinum, which affords higher yields of 100 octane gasoline at lower severities than can be obtained with other catalysts such as the molybdena type catalysts, for example. The catalyst can be employed in the form of a fixed bed of pelleted, beaded or extruded particles, or may be employed in the form of a moving bed, or in fluidized form as a finely divided powder. In fixed bed processes, a series of two or more, usually 3 to 5 reactors are used with interheaters between the reactors so that temperature drop due to the endothermic nature of the major reactions can be compensated for. Operating at the desired severity levels and at low pressure requires more or less frequent regeneration of the catalyst. This can be provided by a controlled process for burning off carbon deposits, advantageously followed by an elevated temperature "soak" with oxygen enriched gas. Advantageously, a "swing" reactor as used in the Ultraforming process is provided so that cyclic regeneration can be effected without shutting down the unit.

The fractionation incident to the invention can be conducted with considerable flexibility. For example, it may comprise separation of recycle gas after cooling at approximately reaction pressure followed by stabilization of the condensed reformat, as shown in the drawings, or it may comprise partial condensation of the heavier components of the reformed mixture followed by separation of recycle gas, stabilization and such further fractionation of the lighter components as is desired. In any event, the essential fractionation is splitting the reformat into a heavy reformat fraction containing at least about 75 to 80 volume percent aromatics and a lighter reformat fraction. The lighter fraction may comprise the overhead from the initial boiling point of the C<sub>5</sub>+ reformat to an end point in the range of 220 to 300° F. depending upon the reforming severity. Alternatively, the lighter

components, including  $C_5$ 's and  $C_6$ 's, may be separated so that the light reformate essentially comprises a  $C_6+$ ,  $C_7$  or  $C_7+$  fraction. The light reformate is charged to a selective extraction producing an aromatics rich extract suitable in octane for blending with the heavy reformate and a raffinate of low octane. If a  $C_5$  fraction rich in isopentane has been segregated from the lighter reformate by prefractionation it may have a blending octane high enough to permit reblending with the high octane heavy reformate. If the low octane raffinate is to be recycled to the reforming zone, or is to be separately reformed, it is usually desirable to separate  $C_6-$  hydrocarbons by prefractionization since the lighter hydrocarbons are difficult to convert under reforming conditions.

The initial boiling point of the heavy reformate fraction is correlated according to the invention, as noted above, with the reforming severity. For example, charging a  $C_7+$  heavy naphtha of 212° to 404° F. boiling range and 46.7 CFR-R octane (52% naphthenes), the octane of 220° to 430° F. heavy reformate was 96.4 CFR-R at 72 volume percent yield based on the  $C_5+$  reformate or 60.8 volume percent based on the naphtha feed. With a reformate severity of 99.5 CFR-R, the 220° to 430° F. fraction had an octane number of 102.9 CFR-R clear (extrapolated) with 70.2 and 54.5 volume percent yields on reformate and feed, respectively. Concomitantly, there was an increase in the octane of the 270 to 430° F. heavy reformate from 98.8 to 104.8. A particular factor appears to have been the large relative gain in quality of the 220° to 270° F. fraction as severity was increased. The octane gain of this fraction was from 77 CFR-R clear for a 93 octane reformate to 97 for a 100 octane reformate. Accordingly, it is preferable to include this material in the heavy reformate when the reforming severity is above 95 octane, but at lower severities, it is better to split at a higher cut-point nearer the 270° to 300° F. range.

It is an essential feature of the present invention that only the light reformate fraction is extracted for recovery of premium gasoline blending stock and separation of low grade raffinate for reprocessing. In this way, the cost burdens of the inherently expensive separation necessary to segregate hydrocarbons by type are greatly reduced. The size of the extraction equipment can be minimized, thus reducing investment cost and the expense of solvent inventory. Operating costs are greatly reduced because it is so much cheaper to strip the lower volume of light hydrocarbons from the solvent. This is particularly true in the case of higher boiling solvents such as glycols, phenol, cresols, nitrogen containing solvents such as substituted amines and the like. Both the thermal requirements and the cost of handling larger quantities of materials are greatly reduced. Also, greater flexibility is provided in the selection of solvents. For example, butyrolactone can be better utilized. Butyrolactone has remarkable capacity combined with selectivity for separating aromatics, but it is relatively low boiling and presents a problem when a full range gasoline stock is handled.

In a specific example of operation according to the invention following the flow of Figure 2 of the drawings, 15,000 barrels per day of Mid-Continent naphtha are reformed in reforming zone 15 in the presence of a 0.6% platinum-on-alumina catalyst at about 900° F., 300 p.s.i.g., 1-2 WHSV and 5,000 s.c.f. of hydrogen per barrel of feed. After cooling by passage through exchanger 17, 12,400 b./d. of 94.5 CFR-R product is recovered at 120° F. and 250 p.s.i.g. in high pressure separator 19. Operating at a tower top temperature of 185° F. in stabilizer 27, the net overhead of  $C_3-C_4$  is 1270 barrels per day recovered at 100° F. and 200 p.s.i.g. from overhead receiver 30. By appropriate adjustment of the fractionation conditions,  $C_5$ 's or  $C_6$ 's can be taken overhead from tower 27 is desired to reduce handling burdens in

subsequent operations. With a bottoms temperature of 470° F., 11,130 barrels per day of 95.2 octane stock is recovered for charging to the splitter tower 39. With a tower top temperature of 470° F., 3,500 barrels per day of 77 CFR-R light reformate is recovered at 300° F. and 155 p.s.i.g. from overhead receiver 46. The heavy reformate recovered as bottoms amounts to 7,630 barrels per day of 103 CFR-R octane boiling in the range of 270° F. to 400° F. The heavy product is removed from the system as shown in Figure 2 by lines 43 and 44 in a manner providing heat by exchange to the reformate passing through exchanger 25.

The light reformate from line 50 of the splitter 39 is heated in exchanger 150 and is charged via line 151 to extractor tower 152 where it is contacted countercurrently with 19,800 barrels per day of diethyleneglycol solvent charged through line 153. The extraction is conducted at 300° F. and 140 p.s.i.g. The raffinate is removed overhead through line 154 and amounts to about 2375 barrels per day of 61 CFR-R octane. The raffinate can be treated further to remove traces of solvent if desired as by water washing and is recycled to reforming zone 15 by means of lines 154 and 155 or may be diverted via line 156 to other operations. The rich solvent mixture amounting to 20,925 barrels per day is withdrawn from the bottom of column 152 by line 157 and is charged to solvent stripper tower 158 operated at 250° F. overhead and 300° F. (5 p.s.i.g.) bottoms temperature. Steam is used to strip the extract which is recovered overhead at the rate of 1140 barrels per day of 101 CFR-R aromatic extract through line 159. Solvent is recovered from the bottom of the tower through line 160 for recirculation to the extractor 152 via solvent accumulator 161 and line 162. The selectivity of the solvent is advantageously controlled by the concentration of water therein so that a portion of the solvent is treated continuously or intermittently, after withdrawal through connection 163, to maintain the desired water content. Regenerated or fresh solvent may be added by line 164.

The light aromatic extract in line 159 is blended with the heavy reformate in line 144 so as to produce a maximum yield of 100+ octane blending stock which may be passed via line 165 to facilities for blending with selected light components of high octane value, e.g. light catalytic gasoline, alkylate, tetraethyl lead, etc. It is possible of course to withdraw a portion of the aromatic extract, as indicated by line 166, and fractionate for the purpose of recovering benzene, toluene or xylenes.

The advantages of operation according to the invention may be perceived by comparing the barrel-octanes of gasoline recovered at a severity of 93.5 for the  $C_5+$  reformate, which is then treated by successive fractionation and extraction to recover 100 octane+ blending stock, with the barrel octanes recovered by once-through reforming to 100 octane severity. The total barrel-octanes recovered from the latter operation is 7,700 while the total recovered according to the invention is 8,895, of which 68 volume percent is 101 CFR-R clear octane value, with the balance raffinate suitable for recycle or blending into regular gasoline.

In the production of high octane gasoline blending stock, there is a strong incentive for going to severities above the 95 octane level because of the marked concentration of aromatics in the heavier fractions of the reformate and the increasing potential yield of 100+ octane stock by fractionation plus extraction. On the other hand, the penalties for operating at high severities rapidly become greater as the octane level of once-through reforming approaches 100. There is a pronounced increase in drop-off in yield above about the 95 level, and there is an increasing amount of light hydrocarbons in the  $C_5-$  range produced. The catalyst requires more frequent regeneration, and with stocks which are lean in naphthas or with cracked stocks which contain substantial quantities of olefins, the amount of coke formation is in-

creased to an extent requiring excessively frequent regeneration. Operating costs are increased thereby, and capacity is reduced. In addition, a substantially higher inventory of catalyst is required, and the effective life of the catalyst is significantly reduced. The order of the cost difference involved is indicated by typical savings that can be made by application of the invention. In a particular case it was calculated that blending 1000 barrels per day of aromatics from light reformat with  $C_8+$  heavy reformat would allow a reduction in reforming severity from 100 CFR-R to 98 CFR-R. After taking into account the need for purchasing additional butane and the cost of upgrading the raffinate to the quality level of regular gasoline, the net savings obtainable by reducing severity about 2 CFR-R units, from 100 to 98, with consequent increases in yields, approximates \$1,000 per day. The savings result primarily from improved catalyst life and reduced catalyst requirements.

In the practice of the invention, a variety of extractive agents can be used for treating the light reformat. Polyhydroxy solvents such as diethyleneglycol, dipropyleneglycol, triethyleneglycol, or mixtures thereof, advantageously promoted in selectivity by the addition of water, are particularly suitable. Other useful solvents are described in U.S. Patent 2,364,517. Sulfur dioxide also is feasible although it requires added facilities for refrigerated handling of the solvent. Usually, the feed and solvent will be contacted countercurrently in one or more extraction columns of the number of theoretical extraction stages required to effect the degree of separation desired. The conditions of extraction will be determined by the nature of the solvent and its selectivity for aromatics at various temperature conditions. Usually, selectivity is improved with decreasing temperatures, and temperatures in the range of say  $-40^\circ$  F. to  $300^\circ$  F. or more may be used, with adjustment of pressure to obtain the desired phase separation, at solvent to feed ratios advantageously in the range of 1/1 to 25/1. Various methods may be used to separate extract and solvent, but in general, distillation is most satisfactory. Traces of solvent can be removed from the separated raffinate and extract phases by washing with water or other solvents, or by stripping.

The use of solid adsorbents such as silica gel, activated carbon, activated alumina and the like also is feasible for extracting an aromatics rich product from the light reformat. If the objective is maximum octane, silica gel may possess advantages over many solvents. For example, in treating a 44-octane,  $156^\circ$  to  $271^\circ$  F. reformat fraction split from a 93.5 reformat, an extract having an unleaded octane equivalent to  $100+ 2.69$  cc. TEL in 11% yield on the total reformat was obtained. Using a selective solvent, the extract from the same light reformat had an octane of 99.3 CFR-R in 15% yield.

Contrary to the prevailing opinion that the heavy paraffins in heavy reformat are of such low octane value that their removal necessarily results in appreciable octane improvement, it has been found in the development of the invention that only small increases in the octane of heavy fractions are obtained by further extraction. Thus, extraction of a  $270^\circ$  F. + heavy reformat having an

octane of 102 CFR-R with silica gel improved the octane only from  $100+ 0.2$  cc. TEL to  $100+ 0.36$  at a percentage sacrifice in yield from 47.1 to 40.3, based on the total reformat.

We claim:

1. A process for producing high octane gasoline blending stock of at least 100 CFR-R clear from low octane naphtha charge stock by catalytic reforming at less than once-through reforming severity which comprises reforming a naphtha charge stock consisting of a  $C_7+$  naphtha boiling in the range of about  $200^\circ$  to  $400^\circ$  F. in the presence of a platinum-alumina type reforming catalyst and recycle hydrogen under a combination of conditions including a temperature in the range of  $850^\circ$  to  $1,000^\circ$  F., a pressure in the range of about 200 to 400 p.s.i.g., a weight hourly space velocity in the range of 0.1 to 5.0 and a hydrogen recycle rate in a range of 1,000 to 10,000 cubic feet per barrel providing a severity producing a  $C_5+$  reformat having an octane number of at least about 95 CFR-R clear whereby the reformat may be fractionated at a cut point in the range of about  $220^\circ$  to  $270^\circ$  F. to obtain a light fraction boiling in the range of  $C_5+$  to said cut point and comprising about 35 to 60 volume percent of the total  $C_5+$  reformat and having an octane number in the approximate range of 75 to 80 CFR-R clear and to obtain a heavy fraction boiling in the range of said cut point to about  $430^\circ$  F. and comprising about 40 to 65 volume percent of the total reformat and containing at least 75 volume percent aromatics, selectively extracting an aromatics-rich fraction of high octane number from said light reformat fraction and thereafter blending said aromatics-rich fraction with the said foregoing heavy reformat fraction so as to obtain a combined blending stock of at least 100 CFR-R clear octane number.

2. The process of claim 1 in which the light reformat is treated with a selective solvent to extract the aromatic rich fraction which is stripped from the solvent for blending with the heavy reformat.

3. The process of claim 1 in which the light fraction treated with a selective adsorbent for aromatics to extract the aromatics rich fraction which is recovered by desorption for blending with the heavy reformat.

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