A combination process for obtaining motor fuel from a mixture of hydrocarbons comprising isobutane and normal butane involving alkylation and paraffin disproportionation and preferably also light hydrocarbon dehydrogenation.
3,657,109

1

MOTOR FUEL PRODUCTION

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to the production of valuable hydrocarbons such as high-octane fuels by a combination process. More particularly, the present invention relates to the production of motor fuels by a combination process involving alkylation and paraffin disproportionation, and preferably also paraffin dehydrogenation.

2. Prior Art

There are a number of known processes to produce motor fuels. Alkylation is one of the well-known methods for producing relatively high-octane motor fuel.

The basic elements of most alkylation units are the same. The heart of an alkylation process is the reactor, in which the fresh feed of olefin and isoparaffin (usually isobutane) together with recycle isoparaffin is contacted in the presence of an acid catalyst to form alkylate. Facilities are provided to settle the catalyst from the product alkylate and to recycle the catalyst, to remove traces of acid catalyst from the product alkylate and to fractionate recycled isobutane from the product.

To prevent propane from building up in the recycle isobutane, a small depropanizer is usually provided to remove propane from a side stream. Also, a debutanizer is usually provided to separate normal butane from the product alkylate.

Although disproportionation of hydrocarbons is not normally used to produce motor fuels, disproportionation is an important reaction in the motor fuel production process of the present invention.

The term "disproportionation" is used in the present specification to mean the conversion of a hydrocarbon of a given molecular weight to new hydrocarbons of both higher and lower molecular weight. For example, butane may be disproportionated according to the reaction:

\[ 2C_2H_6 = C_3H_8 + C_3H_12 \]

It is known that paraffins can be disproportionated. For example, U.S. Pat. No. 3,445,541 discloses a high-temperature process which may be applied to the disproportionation of either paraffins or olefins having from three to six carbon atoms per molecule. According to the process of U.S. Pat. No. 3,445,541, the hydrocarbon feed is contacted with a combined dehydrogenation-disproportionation catalyst at conditions of temperature and pressure to promote appreciable amounts of both dehydrogenation and disproportionation. According to the disclosure of U.S. Pat. No. 3,445,541, a temperature between 800° and 1,200° F. can be used for the disproportionation-dehydrogenation reaction. The lowest temperature used for processing a paraffin in accordance with any of the examples of the patent is 800° F. and, typically, the temperatures used for paraffin disproportionation according to U.S. Pat. No. 3,445,541 are between 1,040° and 1,125° F.

According to laboratory research, we have found that C₄ paraffins can be disproportionated to form propane and C₅+ paraffins at temperatures below 950° F., and that the paraffin disproportionation catalysts are more stable at these lower temperatures and that the paraffin disproportionation process shows a higher selectivity for, and ultimate yield of, C₅+ paraffins at temperatures below 950° F., and preferably below 800° F.

SUMMARY OF THE INVENTION

According to the present invention a combination process involving alkylation and paraffin disproportionation is provided for producing motor fuel from a mixture of hydrocarbons comprising isobutane and normal butane, which process comprises:

a. Feeding the mixture of hydrocarbons and a heavier hydrocarbon fraction to a fractionation zone and therein

b. Fractionating the feed to the fractionation zone to obtain at least an isobutane-rich stream, a normal butane-rich stream and said motor fuel,

c. Reacting at least a portion of the isobutane-rich stream with an olefin in an alkylation zone to form raw alkylate,
d. Disproportionating at least a portion of the normal butane-rich stream in a disproportionation zone to form raw disproporionated, and

e. Feeding the raw alkylate and the raw disproportionated to the fractionation zone as the aforesaid heavier hydrocarbon fraction.

In the process of the present invention it is particularly preferred to generate the olefin feed for the alkylation zone by dehydrogenation of at least a portion of the propane which is generated by the disproportionation of the normal butane feed to the disproportionation zone. Preferably the propane is converted to the olefin feed for the alkylation zone by catalytic dehydrogenation or by thermal cracking. Because propane is essentially always generated in the disproportionation of normal butane, it is very advantageous to use the disproportionation-generated propane in the overall process to produce motor fuel. Ordinarily, the propane might be burned as a fuel gas, but in accordance with a preferred embodiment of the present invention the propane is converted to an olefin such as ethylene or propylene, and this olefin is used as at least part of an olefinic feed for reaction with the isoparaffin in the alkylation zone.

The alkylation step of the combination process of the present invention can employ a number of known alkylation processes, including HF and H₂SO₄ alkylation. The alkylation step is operated at conditions of temperature and pressure known according to the prior art.

Similarly, in those preferred embodiments of the present invention wherein propane is converted to an olefin feed for the alkylation zone, the conversion is carried out as, for example, by catalytic dehydrogenation or by thermal cracking in accordance with operating conditions known according to the prior art.

However, in the present process combination invention it is preferred to carry out the disproportionation of the normal butane feed at relatively low temperatures compared to temperatures previously indicated to be desirable for paraffin disproportionation using a dehydrogenation-disproportionation type catalyst. It is preferred to carry out the normal butane disproportionation at temperatures below 950° F., and still more preferably, below 800° F. Temperatures in excess of 800° F. and particularly in excess of 950° F. result in the production of appreciable amounts of olefins in the disproportionation zone, and in most instances it is not desired to produce a motor fuel which has a particularly high olefin content. Catalysts such as disclosed in the two patent applications by T. R. Hughes entitled "Saturated Hydrocarbon Averaging," Ser. No. 864,870, and "Saturated Hydrocarbon Conversion," Ser. No. 864,871, both filed Oct. 8, 1969 and now abandoned, are well suited for disproportionation at temperatures below 950° F. and particularly below 800° F. The above mentioned patent applications are incorporated by reference in their entirety into the present patent application.

A particularly preferred hydrocarbon feed source for the process of the present invention is a natural gas plant, or a gas recovery plant, C₅+ hydrocarbon fraction. The notation C₅+ indicates propane and heavier hydrocarbons. The C₅+ fraction may contain a substantial amount of hydrocarbons which are in the C₅+ boiling range. Normally, the feed consists mostly of propane, C₃ and C₅ hydrocarbons, with the C₃ hydrocarbon content being relatively low (1-10 volume percent C₃'s) compared to the propane and C₅'s. Propane is normally present to the extent of about 5 to 60 volume percent in the net feed.

In accordance with the preferred embodiment of the present invention wherein the feed is dehydrogenated to form at least a part of the olefinic feed for the alkylation zone, a portion of the propane feed to the dehydrogenation step is usually obtained from the fractionation zone C₅+ fractionation of propane from the net feed. Thus, the portion of the net feed usually increases the propane fractionated from the raw
disproportionate fed to the fractionation zone. The olefins which are formed in the dehydrogenation zone can be used as chemical feedstock, for example, polyethylene or polypropylene chemical plant, in addition to their preferred use in the alkylation step of the present invention.

One of the important advantages of the present invention is that the fractionation zone provides fractionation for several processing steps in the overall process combination. In the preferred embodiment just referred to, the fractionation zone provides separation of propane from the net feed to the inverted process as well as providing for separation of propane generated in the disproportionation step and propane which is generated due to side reactions in the alkylation step.

The fractionation zone, in accordance with the process of the present invention, also serves to fractionate isobutane from other constituents (C₆ and possibly C₇+) in the net fed to the process, as well as fractionating isobutane from the raw alkylate from the alkylation step, and, if necessary, fractionating isobutane from the raw disproportionate formed in the disproportionation step of the present invention.

Therefore, the fractionation zone in the process of the present invention serves as a common fractionation zone for a number of integrated processing steps. The economic attractiveness of the process of the present invention is increased because of the fact that common fractionation facilities may be used for several of the processing steps of the present invention.

Preferably the fractionation zone consists essentially of a "deisobutanizer" distillation column with associated equipment (reboiler, overhead system, etc.) and a debutanizer column with associated equipment. The feed to the deisobutanizer preferably includes the net feed and the raw alkylate and the raw disproportionate. The bottoms from the deisobutanizer are preferably fed to the debutanizer for fractionation into a normal butane-rich stream and the product motor fuel. The normal butane-rich stream from the debutanizer is fed to the disproportionation step of the present invention. An isobutane-rich stream is removed from the upper part of the deisobutanizer and fed to the alkylation step of the present invention. A propane-rich stream is preferably withdrawn as an overhead vapor from the deisobutanizer. Thus the deisobutanizer could be referred to as a depropanizer, but because isobutane is a major stream withdrawn from the upper part of the column, the column is referred to as a deisobutanizer. In certain instances, depending on overall material balance, it is economically preferable to use a deproparanizer column to remove propane from the combined net feed, raw alkylate and raw disproportionate prior to feeding these combined streams to the deisobutanizer. Therefore, it is to be understood that the deisobutanizer zone can include a depropanizer distillation column located ahead of a deisobutanizer distillation column.

Thus, according to a preferred embodiment of the present invention, a motor fuel is produced from a mixture of hydrocarbons comprising at least isobutane and normal butane using a common deisobutanizer fractionation zone and a debutanizer fractionation zone in accordance with the following steps:

a. Feeding the mixture of hydrocarbons to a deisobutanizer zone together with a heavier hydrocarbon fraction,

b. Fractionating isobutane from the feed to the deisobutanizer zone to obtain an isobutane-rich stream and a deisobutanizer heavy stream.

c. Feeding the isobutane-rich stream with an olefin to an alkylation zone wherein they are reacted together to form a raw alkylate,

d. Feeding the deisobutanizer heavy stream to a debutanizer zone and therein

e. Fractionating normal butane from the feed to the debutanizer zone to obtain a normal butane-rich stream and said motor fuel,

f. Feeding the normal butane-rich stream to a disproportionation zone wherein the normal butane is disproportionate to form raw disproportionate, and

g. Feeding the raw alkylate and the raw disproportionate to the deisobutanizer zone as the aforesaid heavier hydrocarbon fraction to a polyethylene or polypropylene chemical plant, in addition to their preferred use in the alkylation step of the present invention.

Other preferred embodiments for the fractionation zone include:

1. Just one fractionation column receiving a total feed comprising the net feed, raw disproportionate, and raw alkylate, and separating this total feed into a propane-rich stream, an isobutane-rich stream, a normal butane-rich stream and the product motor fuel, or
2. Two fractionation columns, the first fractionation column receiving the total feed as above and splitting the feed into a propane, isobutane-rich stream; and a normal butane, C₇+ rich stream; the second fractionation column receiving the propane, isobutane-rich stream and splitting it into a propane-rich stream and an isobutane-rich stream for feed to the alkylation step; and the third fractionation column receiving the normal butane, C₇+ stream and splitting it into a normal butane-rich feed to the disproportionation step and a product C₇+ stream.

As indicated previously, in one preferred embodiment of the present invention, propane is dehydrogenated to form an olefinic feed for the alkylation step of the present invention. In accordance with that concept of the present invention wherein propane is dehydrogenated to form olefinic feed for the alkylation step, a particularly advantageous combination process, which typically would not utilize a common fractionation zone, contains the following steps:

a. Disproportionation of a normal butane-rich stream to obtain propane and C₂+ hydrocarbons,

b. Separation of the propane from the C₂+ hydrocarbons to obtain a propane-rich stream and a disproportionate C₇+ rich stream,

c. Dehydrogenation of the propane-rich stream to obtain olefins,

               d. Alkylation of an isoparaffin by reacting the isoparaffin with said olefins to thereby form an alkylate.

In accordance with a particularly preferred embodiment of the above steps (a) through (d) combination process, at least a portion of the alkylate is blended with at least a portion of the C₂+ disproportionate to form a valuable motor fuel. The octane, for example, of the motor fuel can be regulated by blending more of less alkylate into the C₂+ disproportionate. The raw alkylate formed in accordance with the step (d) alkylation is, of course, usually subjected to fractionation to remove at least a large part of the C₆ and lighter hydrocarbons before the alkylate is blended with the C₇+ disproportionate.

In accordance with a particularly preferred embodiment of the present invention, at least a portion of the C₇+ disproportionate is isomerized, for example in a conventional pen-hex isomerization unit. The C₇+ isomerized product is then preferably blended with the alkylate. When an isomerization step is included, common fractionation facilities are preferably used for the net feed (e.g., a C₇+ feed stream from a gas plant) and the alkylation step effluent. The disproportionation step preferably is followed by a separate fractionation step to prepare feed for the isomerization step.

BRIEF DESCRIPTION OF THE DRAWING

The drawing is a schematic process flow diagram illustrating a preferred embodiment of the present invention.

DESCRIPTION OF THE DRAWING

Referring now in more detail to the drawing, a C₂+ stream comprising propane, isobutane, and normal butane is fed via line 1 together with recycle raw alkylate and raw disproportionate in line 2 to fractionation zone 21. Fractionation zone 21 separates the total feed stream into a propane-rich stream withdrawn in line 16, an isobutane-rich stream withdrawn in line 17, a normal butane-rich stream withdrawn in line 7 and the product motor fuel stream withdrawn in line 20. The term "rich" is used in the specification to mean a stream containing at least 50 volume percent of the specified component.

It is to be understood that the fractionation zone may be comprised of a number of separate distillation columns and
appurtenant equipment. However, in any case there will be at least one distillation column which is common to at least two streams of the process of the present invention.

Particularly preferred combined streams for fractionation in a common distillation column or columns in the fractionation zone of the present invention include: the raw alkylate and the raw disproportionation, the raw alkylate and the net feed, the raw disproportionation and the net feed, or the raw alkylate, the raw disproportionation, and the net feed. The term "raw alky-

late" is used herein to mean alkylation product before final separation of isobutane (including recycle isobutane) from the alkylate; the term "raw disproportionation" is used to mean disproportionation product at least before final separation of butanes, and normal butane (including recycle butanes) from the disproportionation and usually also before final separation of propane from the disproportionate; and the term "net feed" refers to the net input to the process of the invention which is a stream containing at least isobutane and normal butane.

For purposes of simplified illustration, the process of the present invention may be considered in one broad embodiment. At least one large distillation zone with the total feed to the fractionation zone including raw alkylate transferred via lines 12 and 2, raw disproportionation transferred via lines 9 and 2, and net feed via line 1. This total feed to the large common fractionation zone is then split into the product motor fuel withdrawn via line 20, a normal butane-rich stream withdrawn via line 7 and fed to the disproportionation zone, an isobutane-rich stream withdrawn via line 10a and fed to the alkylation zone, and a propane-rich stream withdrawn from the large central fractionation zone via line 16 and burned as fuel, or in a preferred embodiment fed to the dehydrogenation zone for conversion to olefin which is used in the alkylation zone.

The fractionation zone 21, however, can consist of a desisobutanizer zone and a debutanizer zone as is indicated in the drawing. In most instances, the propane stream which is obtained by fractionation in the desisobutanizer zone preferably is further fractionated into a depropanizer to separate isobutane from the propane. The desisobutanizer zone (which is within fractionation zone 21) preferably consists essentially of a "desisobutanizer" distillation column, although the desisobutanizer zone may be split into two or more columns, particularly in view of the fact that more than two streams are fractionated or separated from one another in the desisobutanizer zone. In certain instances it is preferred to deprotoanize both isobutane zones.

Referring to the particular schematic embodiment shown in the drawing, a C4+ stream is withdrawn in line 5 from the lower part of the deisobutanizer zone. If the majority of the normal butane is removed from the heavy fraction withdrawn from the bottom of the deisobutanizer zone, then the C4+ fraction from the bottom of the deisobutanizer zone is withdrawn in line 5 and fed to the process as product motor fuel via lines 24 and 20.

The majority of the normal butane, in accordance with one preferred embodiment of the present invention, is separated from the C4+ fraction by withdrawing the normal butane as a vapor sidestream from the deisobutanizer fractionation column and is withdrawn via line 15. This sidestream is indicated schematically by line 15. In the typical case, however, normal butane is not withdrawn as a vapor sidestream but is instead contained in the bottom stream withdrawn via line 5 from the deisobutanizer zone.

In this typical case the normal butane and C4+ material withdrawn via line 5 is fed to debutanizer zone 6 for separation of normal butane from C4+ material. The C4+ material is withdrawn from the bottom of debutanizer zone 6 via line 25 and is withdrawn from the process as product motor fuel via line 20. A normal butane-rich stream is withdrawn from the upper part of debutanizer zone 6 via line 22 and is fed to disproportionation zone 8 via line 7. The debutanizer preferably consists primarily of a debutanizer fractionation column. The debutanizer fractionation column will, of course, have associated equipment such as a reboiler and an overhead cooling and refluxing system.

The normal butane-rich stream, which is withdrawn from the upper part of the debutanizer zone as schematically indicated by line 22, generally contains considerable amounts of isobutane. The isobutane content can be controlled to less than a few tenth volume percent in the normal butane stream, withdrawn via line 22, depending upon the fractionation obtained prior to the debutanizer zone. However, in the overall process of the present invention usually it is economically preferable in terms of capital investment, operating expense and product value to allow the isobutane content of the normal butane stream withdrawn via line 22 to be in the range of 1 to 10 volume percent. This has the advantage of resulting in a more branched-chain disproportionated and, therefore, higher octane disproportionate product from disproportionation zone 8. Allowing the isobutane content of stream 22 to be between 1 and 10 volume percent also is advantageous in that the operating expense and capital investment for fractionation is less than if the isobutane is maintained at a very low content in the normal butane stream withdrawn from debutanizer zone 6 via line 22.

In the alternate schematic embodiment indicated in the drawing wherein normal butane is withdrawn as a vapor stream via line 23 from the desisobutanizer zone, there will usually be between about 1 and 10 volume percent isobutane in the normal butane vapor stream. Also, the normal butane vapor stream will typically contain several volume percent C4+ hydrocarbons. Because the C4+ material may be disproportionated to valuable higher normal butanes and isobutanes in disproportionation zone 8, in many instances it is very advantageous to omit debutanizer zone 6 to thus realize fractionation operating cost savings as well as investment savings.

The normal butane-rich stream, which is withdrawn either as a vapor via line 23 from the desisobutanizer zone or withdrawn via line 22 from the debutanizer zone 6, is fed to disproportionation zone 8 via line 7. Therein the normal butane is disproportionated, preferably at a temperature below 950°F. and still more preferably at a temperature below 800°F., to form raw disproportionate, which is withdrawn via line 9. The raw disproportionate, comprising propane, unconverted butanes, and C4+ hydrocarbons, is transferred via line 2 to desisobutanizer zone 4.

An isobutane-rich stream is withdrawn from desisobutanizer zone 4 via line 10 and from depropanizer column 14 via line 15 or in general from the fractionation zone via line 10a, and fed to alkylation zone 11. In most instances the propane stream withdrawn via line 22 from the debutanizer zone 6 is fed to disproportionation zone 8 via line 7. In general, as is schematically indicated in the drawing, to fractionation zone 21. The raw alkylation stream contains propane, isobutane, and C4+ hydrocarbons. The propane is formed by side reactions in the alkylation reaction, the isobutane is the reactant which is present in large excess for the alkylation reaction (the olefins being very reactive), and the C4+ hydrocarbons are the high-octane motor fuel formed in the alkylation reaction.

The propane-rich stream which is obtained by fractionation of the feed streams to the fractionation zone is fed via line 16 to dehydrogenation zone 17. Dehydrogenation zone 17 can consist of a thermal cracking reactor and associated facilities or a catalytic debutanizer reactor and associated facilities. In any case, the propane is dehydrogenated to form propylene and/or ethylene, which is withdrawn from dehydrogenation zone 17 via line 19 and passed at least in part to alkylation zone 11 via line 18. Excess olefins formed in the
3,657,109

dehydrogenation zone are withdrawn via line 19b and passed to further processing or used, for example, in chemical manufacture.

EXAMPLE

Table 1 below shows a material balance keyed to the schematic process flow diagram. The material balance illustrates the production of valuable motor fuel from a C₅⁺ feed which is rich in normal butane. The C₅⁺ feed is a typical gas recovery plant C₅⁺ stream.

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Although various specific embodiments of the invention have been described and shown, it is to be understood they are meant to be illustrative only and not limiting. Certain features may be changed without departing from the spirit or essence of the invention. It is apparent that the present invention has broad application to the production of motor fuel in a combination process involving alkylation and paraffin disproportionation. Accordingly, the invention is not to be construed as limited to the specific embodiments illustrated, but only as defined in the appended claims.

What is claimed is:

1. A combination process for obtaining motor fuel from a mixture of hydrocarbons comprising isobutane and normal butane which comprises:
   a. feeding the mixture of hydrocarbons to a fractionation zone together with a heavier hydrocarbon fraction and therein
   b. fractionating the feed to the fractionation zone to obtain at least an isobutane-rich stream, a normal butene-rich stream and said motor fuel,
   c. reacting at least a portion of the isobutane-rich stream with an olefin in an alkylation zone to form raw alkylate,
   d. disproportionating at least a portion of the normal butene-rich stream in a disproportionation zone to form raw disproportionate, and
   e. feeding at least a portion of the raw alkylate to the fractionation zone as the aforesaid heavier hydrocarbon fraction.

2. A process in accordance with claim 1 wherein propane is generated by the disproportionation of said normal butane-rich stream and at least a portion of said propane is converted to olefins, which are reacted with isobutane in the alkylation zone.

3. A process in accordance with claim 2 wherein the disproportionation is carried out at temperatures below 950°F.

4. A process in accordance with claim 2 wherein the disproportionation is carried out at temperatures below 950°F.

5. A combination process for obtaining motor fuel from a mixture of hydrocarbons comprising isobutane and normal butane which comprises:
   a. feeding the mixture of hydrocarbons to a deisobutanizer zone together with a heavier hydrocarbon fraction,
   b. fractionating an isobutane-rich stream from the feed to the deisobutanizer zone to obtain an isobutane-rich stream and a deisobutanizer heavy stream,
   c. feeding the isobutane-rich stream with an olefin to an alkylation zone wherein they are reacted together to form a raw alkylate,
   d. feeding the deisobutanizer heavy stream to a debutanizer zone,
   e. fractionating a normal butane-rich stream from the feed to the debutanizer zone to obtain a normal butane-rich stream and said motor fuel,
   f. feeding the normal butane-rich stream to a disproportionation zone wherein the normal butane is disproportionated to form raw disproportionate, and
   g. feeding the raw alkylate and the raw disproportionate to the deisobutanizer zone as the aforesaid heavier hydrocarbon fraction.

6. A process in accordance with claim 5 wherein the disproportionation is carried out at temperatures below 950°F.

7. A process in accordance with claim 5 wherein the disproportionation is carried out at temperatures below 800°F.

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