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(54) **REFINING PROCESS AND APPARATUS**

4,304,948 12/1981 Vora et al. .
4,393,259 7/1983 Ward et al. .
4,403,999 9/1983 Bezman .
4,444,988 4/1984 Capsuto et al. .

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(List continued on next page.)

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(21) Appl. No.: **09/387,431**

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Related U.S. Patent Documents

Reissue of:

(64) Patent No.: **5,679,117**
Issued: **Oct. 21, 1997**
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Filed: **Oct. 21, 1996**

(57) **ABSTRACT**

A process of producing high octane hydrocarbons includes the steps of preparing a mixture of substantially ethanol and butane or natural gasoline, or low octane gasoline, the mixture having room temperature and atmospheric pressure, adjusting the pressure of the mixture to a magnitude within the range of 10 to 50 pounds per square inch, adjusting the temperature of the mixture to a magnitude within the range of 100 to 460 degrees Fahrenheit, adjusting the pressure of the mixture to a pressure within the range of 500 to 1000 [hydrocarbons] pounds per square inch, catalyzing the mixture with a platinum catalyst, lowering the temperature of the mixture to a magnitude within a range of 90 to 190 degrees Fahrenheit, and separating out liquid product and gas from the mixture. An apparatus for producing high octane alcohols includes a starting tank for retaining a mixture of substantially ethanol and butane or natural gasoline, a heat exchanger for raising the temperature of the mixture, a first high pressure conduit extending from the starting tank to the heat exchanger, a catalyzing chamber, second and third high pressure conduits extending from the heat exchanger to the catalyzing chamber, a nozzle interconnecting the second and third high pressure conduits, high pressure pumps for extracting the heated mixture from the heat exchanger and delivering the mixture to the catalyzing chamber through the second and third high pressure conduits, and a separator for precipitating liquid product out of the mixture.

U.S. Applications:

(63) Continuation-in-part of application No. 08/605,282, filed on Feb. 8, 1996, now abandoned, which is a continuation-in-part of application No. 08/430,275, filed on Apr. 28, 1995, now abandoned, which is a continuation-in-part of application No. 08/385,466, filed on Feb. 8, 1995, now abandoned.

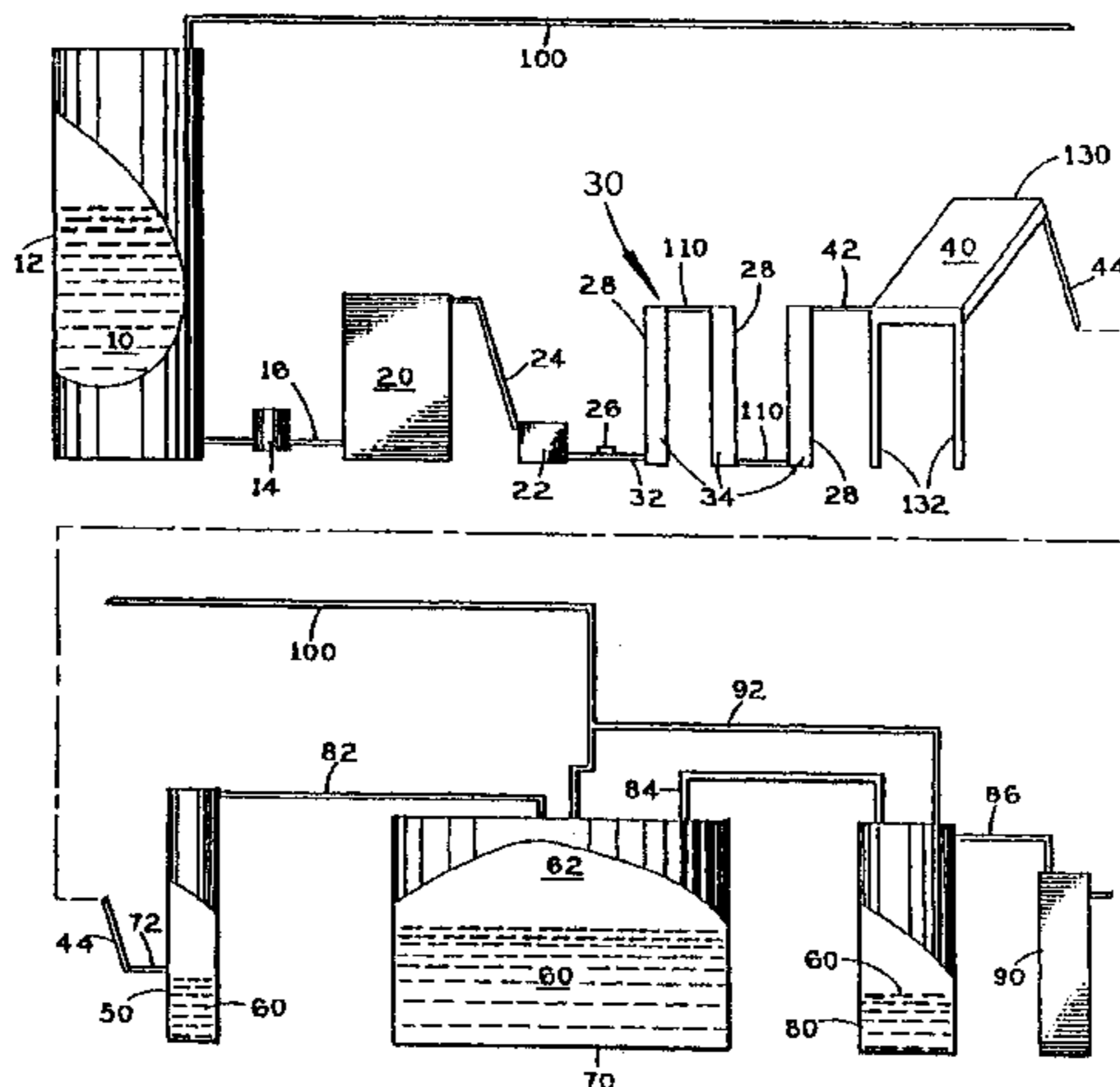
(51) **Int. Cl.**⁷ **C10L 1/18**
(52) **U.S. Cl.** **44/451; 585/1; 585/302**
(58) **Field of Search** **44/451; 585/1, 585/302**

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26 Claims, 1 Drawing Sheet



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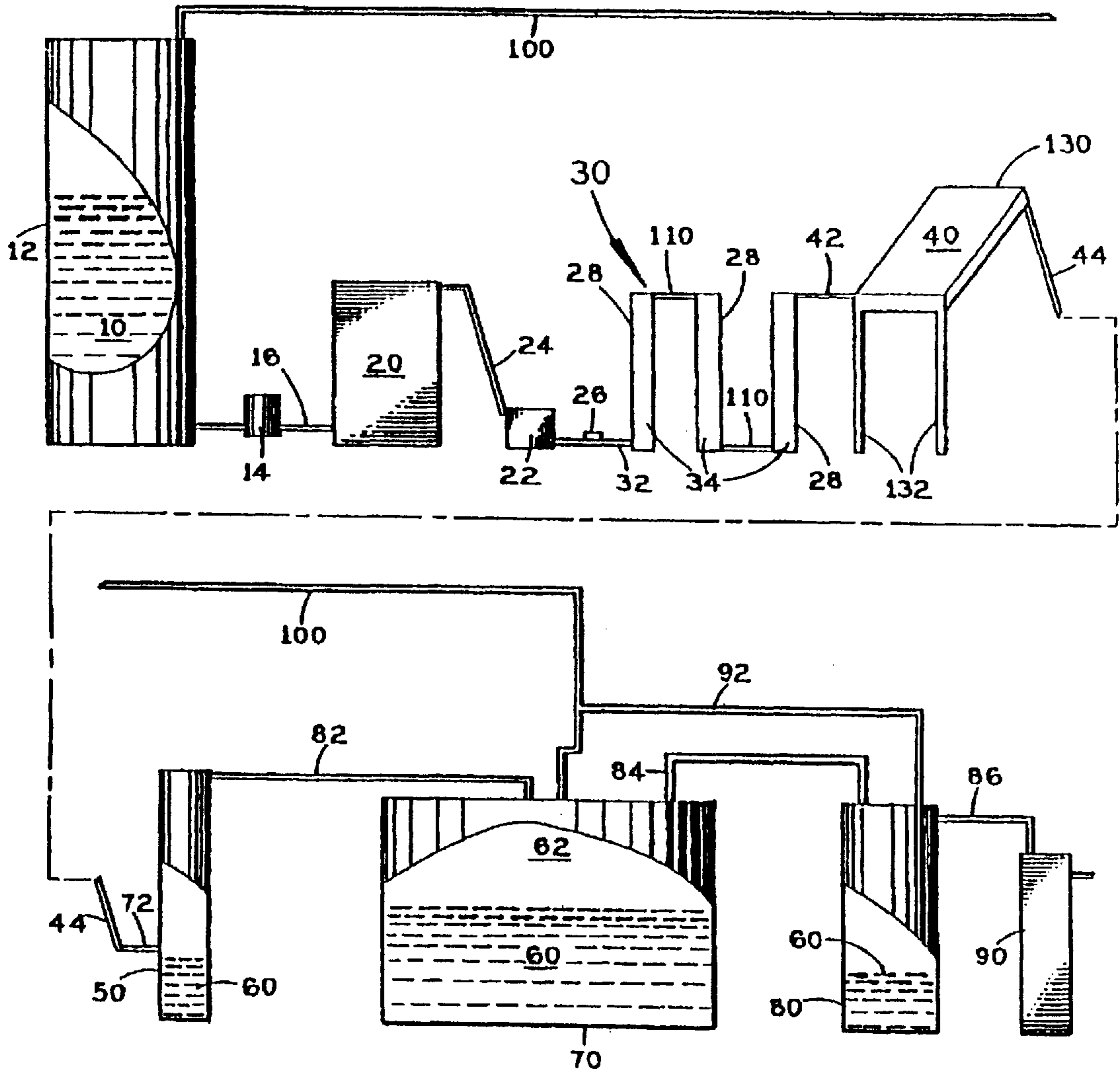


FIG. 1

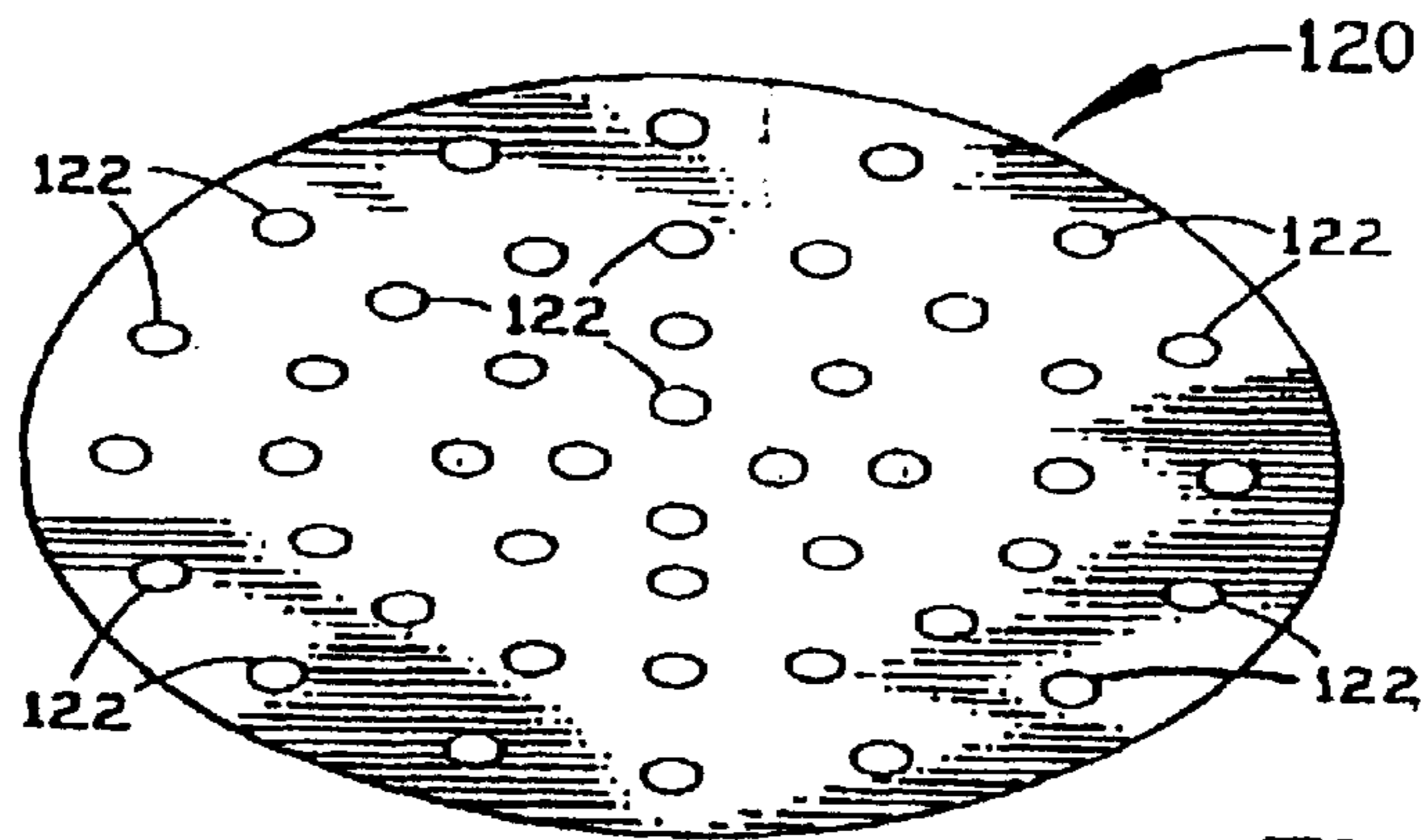


FIG. 2

REFINING PROCESS AND APPARATUS

Matter enclosed in heavy brackets [] appears in the original patent but forms no part of this reissue specification; matter printed in italics indicates the additions made by reissue.

The present invention is a *reissue of Ser. No. 08/734,891 filed Oct. 21, 1996, U.S. Pat. No. 5,679,117 which is a continuation-in-part of Ser. No. 08/605,282, filed Feb. 8, 1996, now abandoned, which is a continuation-in-part of Ser. No. 08/430,275 filed Apr. 28, 1995, now abandoned, which is a continuation-in-part of Ser. No. 08/385,466, Feb. 8, 1995, now abandoned.*

FIELD OF THE INVENTION

The present invention relates generally to the field of fuel forming processes. More specifically it relates to a process of producing high octane alcohols, or pump gasoline, including the steps of placing a pre-mixed mixture of ethanol [and] and other alcohols, and butane or natural gasoline or straight run gasoline in a starting tank, raising the pressure of gases above the surface of the mixture to fifty pounds per square inch, pumping the mixture from the bottom of the starting tank through a first high pressure conduit into a heat exchanger where the temperature of the mixture is raised to a magnitude within the range of 100 to 460 degrees Fahrenheit, extracting the heated mixture from the heat exchanger with high pressure pumps which raise the mixture pressure to 500 to 1000 pounds per square inch, and feeding the heated and pressurized mixture through a second high pressure conduit through a nozzle and through a third high pressure conduit into an elongate catalyzing chamber containing a platinum catalyst. Additional steps include delivering the catalyzed mixture through a fourth high pressure conduit into a cooler for lowering the temperature to a magnitude within a range of 90 to 190 degrees Fahrenheit, feeding the cooled mixture through a fifth high pressure conduit into a series of separator tanks in which liquid final product collects in the tank bottoms and gas rises within the tanks above the surface of the liquid, and the liquid is drained off as the final product. The final product is 120 to 160 research octane, 110 to 129 motor octane, and [R & M] (R+M)/2 about 148. In the case where low octane gasoline (straight run gasoline) is used as the starting material, the final product is a substantially higher octane gasoline called pump gasoline. In general, the starting material may be low octane hydrocarbon material, and the final product is higher octane hydrocarbon material.

BACKGROUND OF THE INVENTION

There have long been various chemical processes for producing gasoline and other fuels. A problem with these prior processes has been that they either fail to produce high octane gasoline, or they fail to do so efficiently.

These prior processes include that of Harandi, U.S. Pat. No. 5,171,912, issued on Dec. 15, 1992. Harandi discloses a process for the production of C+ gasoline from n-butane and propane. The Harandi process includes the steps of contacting a fresh feedstream including normal butane with shape selective medium pore zeolite catalyst particles under conditions sufficient to convert n-butane to an effluent stream including C+ alkanes; separating the effluent stream in a fractionator to recover an overhead stream including propane; contacting the propane stream and a fresh propane feedstream with shape selective, medium pore zeolite catalyst particles under conversion conditions sufficient to con-

vert propane to a mixture including C+ alkanes; deethanizing the mixture and passing the deethanized product including C+ alkanes to the fractionator for separation concurrent with the effluent stream; recovering a bottom stream including C+ gasoline from the fractionator; preferably, distilling an intermediate stream including C alkanes from the fractionator and recovering a stream including isobutane and a stream including unconverted normal butane; and recycling the unconverted normal butane to the normal butane feedstream to the integrated process.

Ward, et al., U.S. Pat. No. 4,393,259, issued on Jul. 12, 1983, reveals a process for converting propane or butane to gasoline. The Ward, et al. process includes the steps of passing feed hydrocarbon into a dehydrogenation zone; passing the entire dehydrogenation zone effluent including hydrogen and light by-products into a catalytic condensation zone where the resulting olefins are converted into dimers and trimers; passing the condensation zone effluent stream into a separation zone in which the dimers and trimers are concentrated into a product stream, with unconverted feed hydrocarbon and hydrogen being recycled to the dehydrogenation zone.

Vora, et al., U.S. Pat. No. 4,304,948, issued on Dec. 8, 1981, teaches a multi-step hydrocarbon conversion process for converting butane to gasoline. The process includes the steps of passing butane into a dehydrogenation zone and the entire dehydrogenation zone effluent is then passed into a catalytic condensation zone where butylene is converted into C and C hydrocarbons; commingling and separating the condensation zone effluent, a stripper overhead stream and an absorber bottoms stream into vapor and liquid portions; passing the liquid into the stripper and contacting the vapor portion with stripper bottoms liquid in an absorber; contacting the absorber overhead stream with liquid butane in a second absorber to remove C hydrocarbons and recycling the dehydrogenation zone; and debutanizing a portion of the stripper bottoms to yield the liquid butane and a gasoline product.

Capsuto, et al., U.S. Pat. No. 4,444,988, issued on Apr. 24, 1984, discloses the use of liquefied propane and butane or butane recycled to control the heat of reaction of converting olefins to gasoline and distillate. The Capsuto, et al. process uses beds and separates the effluent product from the beds into a gas in a liquid phase, cools the gas phase to form additional liquid and heat exchanges the liquid with the overhead gas from the separator.

Wilson, U.S. Pat. No. 5,093,533, issued on Mar. 3, 1992, reveals blended gasolines and a process for making the blended gasolines. The Wilson process involves mixing of a butane-pentane rich component, and natural gasoline component, and at least one octane-enhancing component. The mix is weathered during the blending operation to remove light-weight hydrocarbons including two, three and four-carbon components.

Hiles, et al., U.S. Pat. No. 5,310,954, issued on May 10, 1994, discloses a process for preparing tetrahydrofuran. The Hiles et al. process separates tetrahydrofuran from a feed mixture containing water, lower alkanol and tetrahydrofuran, which includes distilling the mixture in a first distillation zone at a first pressure; recovering from an upper part of the distillation zone a first vaporous mixture including water, lower alkanol and tetrahydrofuran; subjecting the material from the first vaporous mixture to condensation conditions in a condensation zone; passing condensate from the condensation zone to a second distillation zone operated at a second pressure higher than the first pressure;

recovering from an upper part of the second distillation zone a second vaporous mixture including water, lower alkanol and tetrahydrofuran that has a lower concentration of tetrahydrofuran than the first vaporous mixture; and recovering from a lower part of the second distillation zone a stream including substantially pure tetrahydrofuran.

It is thus an object of the present invention to provide a process of producing a very high octane alcohol product efficiently.

It is another object of the present invention to provide such a process which can be practiced with conventional heat exchanger and separator tank equipment.

It is still another object of the present invention to provide such a process which is safe to practice.

It is finally an object of the present invention to provide such a process which is inexpensive to practice.

SUMMARY OF THE INVENTION

The present invention accomplishes the above-stated objectives, as well as others, as may be determined by a fair reading and interpretation of the entire specification.

A process of producing high octane hydrocarbon material is provided, including the steps of preparing a mixture of substantially ethanol and butane or natural gasoline, or straight run gasoline, the mixture having a room temperature and [a] an atmospheric pressure, adjusting the pressure of the mixture to a magnitude within the range of 10 to 50 pounds per square inch, adjusting the temperature of the mixture to a magnitude within the range of 100 to 460 degrees Fahrenheit, adjusting the pressure of the mixture to a pressure within the range of 500 to 1000 pounds per square inch, catalyzing the mixture with a platinum catalyst, lowering the temperature of the mixture to a magnitude within a range of and gas from the mixture, and separating out liquid product and gas from the mixture. The process preferably includes the additional steps of delivering a quantity of gas separated from the liquid product into a furnace to supply heat required for the process, and the further additional steps of delivering a quantity of the gas separated from the liquid product into the mixture at the initial step of the process. The separating step preferably includes several separation steps of separating the mixture into liquid product and gas.

An apparatus for producing high octane alcohols is also provided, including a starting tank for retaining a mixture of substantially ethanol and butane or natural gasoline, a heat exchanger for raising the temperature of the mixture, a first high pressure conduit extending from the starting tank to the heat exchanger, a catalyzing chamber, second and third high pressure conduits extending from the heat exchanger to the catalyzing chamber, a nozzle interconnecting the second and third high pressure conduits, high pressure pumps for extracting the heated mixture from the heat exchanger and delivering the mixture to the catalyzing chamber through the second and third high pressure conduits, and a separator for precipitating liquid product out of the mixture.

The catalyzing chamber preferably includes several upright tubular segments, each tubular segment having a top portion and a bottom portion and containing the platinum catalyst, interconnection conduits interconnecting the tubular segments alternately across the top and bottom portions of the tubular segments, a baffle plate within at least one of the tubular segments, the baffle plate having several plate ports. The tubular segments each preferably include one baffle plate positioned within and across the top portion and the bottom portion of the tubular segment.

BRIEF DESCRIPTION OF THE DRAWINGS

Various other objects, advantages and features of the invention will become apparent to those skilled in the art from the following discussion taken in conjunction with the following drawings, in which:

FIG. 1 is a semi-schematic view of the preferred apparatus for carrying out each step of the inventive process.

FIG. 2 is a perspective view of the baffle plate for use in the catalyzing chamber tubular segments.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

As required, detailed embodiments of the present invention are disclosed herein; however, it is to be understood that the disclosed embodiments are merely exemplary of the invention which may be embodied in various forms. Therefore, specific structural and functional details disclosed herein are not to be interpreted as limiting, but merely as a basis for the claims and as a representative basis for teaching one skilled in the art to variously employ the present invention in virtually any appropriately detailed structure.

Reference is now made to the drawings, wherein like characteristics and features of the present invention shown in the various FIGURES are designated by the same reference numerals.

Process

Referring to FIG. 1, a process of producing high octane alcohols is disclosed, including the following steps. A pre-mixed mixture **10** of one third ethanol and two thirds butane at room temperature and atmospheric pressure is placed in a starting tank **12**. The pressure of gases above the surface of the mixture **10** is raised to fifty pounds per square inch. The mixture **10** is pumped with pumps **14** from the bottom of starting tank **12** through a first high pressure conduit **16** into a heat exchanger **20**, where the temperature of mixture **10** is raised to a level within the range of 100 to 460 degrees Fahrenheit. The preferred temperature is 225 degrees Fahrenheit. The heated mixture **10** is extracted from the heat exchanger **20** with high pressure pumps **22**, which raise mixture **10** pressure to a level within the range of 500 to 1000 pounds per square inch. The preferred pressure is 600 pounds per square inch. The heated and pressurized mixture **10** is fed through a second high pressure conduit **24**, through a nozzle **26** and through a third high pressure conduit **32** into an elongated catalyzing chamber **30** containing a platinum catalyst **34**. Chamber **30** includes three interconnected upright segments **28**. The catalyzed mixture **10** is delivered through a fourth high pressure conduit **42** into a cooler **40** for lowering the mixture **10** temperature to a level within a range of 90 to 190 degrees Fahrenheit.

The cooled mixture **10** is fed through a fifth high pressure conduit **44** into a first separator tank **50** in which final liquid product **60** collects in the bottom of first separator tank **50** and gas **62** rises to fill a space within tank **50** above the surface of liquid product **60**. The liquid product **60** is fed through a first separated liquid conduit **72** at the bottom of tank **50** and the gas **62** is drained off through a first separated gas conduit **82** at the top of tank **50**. Both liquid product **60** and gas **62** are delivered into a second separator tank **70**, in which more liquid product **60** is separated. Some of gas **62** within second separator tank **70** is delivered back through a feedback conduit **100** into the top of starting tank **12**. Some of gas **62** within the second separator tank **70** is simulta-

neously delivered through a second separated gas conduit **84** into a third separator tank **80** where still more liquid product **60** precipitates out and gathers in the bottom of third separator tank **80**. Some of gas **62** within third separator tank **80** is drained into a feedback conduit branch **92**. Some of gas **62** within third separator tank **80** is delivered through a third separated gas conduit **86** into a furnace **90**, where gas **62** is burned as fuel to supply heat to the process where needed.

Final liquid product **60** is within the range of 120 to 160 research octane, 110 to 129 motor octane, and about 148 [R and M] $(R+M)/2$. Other final product **60** test data are as follows:

Oxygenates	I. V. %	42.75
MTBE	I. V. %	<0.1
TAME	I. V. %	<0.1
Alcohols (Ethanol)	I. V. %	42.75
<hr/>		
G. C. Breakdown	Wt %	Vol %
<hr/>		
N. Butane	45.60	53.03
ISO Pentane	1.42	1.55
N. Pentane	1.02	1.10
Toluene	2.02	1.57
Ethanol	49.94	42.75
<hr/>		
PONA	Vol %	
<hr/>		
Paraffins	55.68	
Olefins	0.01	
Naphthenes	<0.01	
Aromatics	1.57	
<hr/>		

To produce high octane gasoline, add 20% by volume of the new product to 80 octane gasoline. The resulting mixture is 92.8 octane, with a vapor pressure in the range of 4 to 19 pounds per square inch.

Preferred Embodiments of Apparatus

Referring to FIG. 1, a preferred apparatus is disclosed for practicing the above-described process of producing high octane alcohols. This apparatus is merely exemplary and other forms of apparatus are contemplated.

Starting tank **12** is a vertical cylindrical drum. Heat exchanger **20** and pumps **14** and **22** are of any suitable conventional design. Nozzle **26** is preferably about three eights inches diameter. Catalyzing chamber **30** includes three elongate, upright tubular segments **28**, each containing platinum catalyst **34**. Segments **28** are interconnected by interconnection conduits **110**, across the tops of the first and second segments **28** and across the bottoms of second and third segments **28**. A baffle plate **120** having a plurality of ports **122** is positioned across the top and bottom of each segment **28**. See FIG. 2. Cooler **40** preferably includes a substantially horizontal tray **130** elevated on legs **132**. Separator tanks **50**, **70** and **80** are vertical cylindrical drums. Tank **70** is preferably of substantially larger diameter than tanks **50** and **80**.

Another embodiment of the invention uses as a starting material approximately one third ethanol mixed with two thirds natural gasoline. The process and apparatus for treating this mixture is the same as that previously described and this explanation will not be repeated herin. Natural gasoline is essentially a mixture of butanes and pentanes plus other hydrocarbon materials. Natural gasoline is derived from wet gas by stripping it. An example of natural gasoline is as follows:

C6+ . . . 53.871% by liquid volume
 Butane . . . 3.03% by liquid volume
 Neo-pentane . . . 0.697% of liquid volume
 Iso-pentane . . . 26.046% by liquid volume
 Normal pentane . . . 16.349% by liquid volume.

The resulting product is substantially one half natural gasoline and one half ethanol. It has a vapor pressure of 1.5 to 8.0 psi and an octane rating of 108 to 160.

A further embodiment uses as a starting material a mixture of 10% ethanol and 90% natural gasoline. The process steps and apparatus remain the same. The resulting product showed an increase in octane rating from 72 to 80–100.

It has been found that the starting material may contain 5% to 50% ethanol, and 50% to 95% natural gasoline. It is possible to add to the mixture 3% to 40% butane. The resulting product contains 5–50% ethanol, 50–90% natural gasoline including 3% to 50% hydrocarbons, and a trace of aromatics. The resulting product has a higher octane rating than the starting material. The product has an acceptable vapor pressure. This product appears to be a gasoline grade product. The ethanol can be removed without harming the product.

In the first embodiment, pentane, including iso-pentane, may be substituted for butane in the starting material. In another embodiment, the [starting] hydrocarbon material [is] contains a low octane hydrocarbon material known as light gasoline or straight run gasoline having an octane rating in the vicinity of 65 to 70. This material is processed through the apparatus described above and in the same way as described in connection with the first and further embodiments. One additional option is to inject a small amount of hydrogen in the catalyst bed. It has been found that the process increases the octane rating of the hydrocarbon material to a level in the vicinity of 87, such that the final product is pump gasoline. The final product has a vapor pressure in the range from 6 to 8 psi which is an acceptable range.

While the invention has been described, disclosed, illustrated and shown in various terms or certain embodiments or modifications which it has assumed in practice, the scope of the invention is not intended to be, nor should it be deemed to be, limited thereby and such other modifications or embodiments as may be suggested by the teachings herein are particularly reserved especially as they fall within the breadth and scope of the claims here appended.

We claim:

1. A process of producing a high octane [alcohols] alcohol composition, comprising the steps of:

preparing a mixture of ethanol and low octane hydrocarbon material, [having an octane rating in the vicinity of 65 and 70, said mixture having room temperature and atmospheric pressure,

adjusting said pressure of said mixture to magnitude within the range of 10 to 50 pounds per square inch,

adjusting said temperature of said mixture to a magnitude within the range of 100 to 460 degrees Fahrenheit,

adjusting the pressure of said mixture to a pressure within the range of 500 to 1000 pounds per square inch] said hydrocarbon material being selected from the group consisting of butane, natural gasoline, straight run gasoline, light gasoline, pentane, iso-pentane and a mixture of two or more of the foregoing,

catalyzing said mixture with a platinum catalyst by conducting said mixture through a catalyst bed at an elevated pressure and elevated temperature which are sufficient to produce a catalyzed mixture containing the high octane alcohol composition,

- lowering the temperature of said *catalyzed* mixture to a [magnitude] *temperature* within a range of *from about 90 to about 190 degrees Fahrenheit*,
- separating [out liquid product and] gas from said *catalyzed* mixture to provide the *high octane alcohol composition*.
2. A process according to claim 1, comprising the additional step of:
- delivering a quantity of said gas separated from said [liquid product] *catalyzed mixture* into furnace means to supply heat required for said process.
3. A process according to claim 1, comprising the additional steps of:
- delivering a quantity of said gas separated from said [liquid product] *catalyzed mixture* into said mixture [at the initial step of said process] *before the catalyzing step of said process*.
4. A process according to claim 1, wherein said separating step comprises a plurality of separation steps of separating said *catalyzed* mixture into liquid product and gas.
5. A [method] *process* according to claim 1, wherein said [hydrocarbons comprise] *hydrocarbon material* comprises butane.
6. A [method] *process* according to claim 1, wherein said [hydrocarbons comprise] *hydrocarbon material* comprises natural gasoline.
7. A [method] *process* according to claim 1, wherein said [hydrocarbons comprise] *hydrocarbon material* comprises straight run gasoline.
8. A *process* according to claim 1, wherein said mixture comprises *from about 5 to about 50% ethanol and from about 50 to about 95% said low octane hydrocarbon material*.
9. A *process* according to claim 8, wherein said *hydrocarbon material* comprises *natural gasoline*.
10. A *process* according to claim 9, wherein said *hydrocarbon material* further comprises *from about 3 to about 40% butane*.
11. A *process* according to claim 1, wherein said mixture comprises *about one third ethanol and about two thirds natural gasoline*.
12. A *process* according to claim 1, wherein said mixture comprises *about 10% ethanol and 90% natural gasoline*.
13. A *process* according to claim 1, wherein said *catalyzing step* is conducted at a *temperature ranging from about 100° to about 460° F*.
14. A *process* according to claim 13, wherein said *catalyzing step* is conducted at a *pressure ranging from about 500 to about 1000 pounds per square inch*.

15. A *process* according to claim 1, further comprising *injecting hydrogen into said catalyst bed during said catalyzing step*.
16. A *process* according to claim 1, wherein said *high octane alcohol composition* has a *research octane rating ranging from about 108 to about 160 and a vapor pressure ranging from about 1.5 to about 8 psi*.
17. A *high octane fuel composition* comprising a mixture of *relatively low octane fuel and relatively high octane alcohol composition*, said *relatively high octane alcohol composition* being made by a *process* comprising the steps of:
- mixing ethanol and a hydrocarbon material selected from the group consisting of butane, natural gasoline, straight run gasoline, light gasoline, pentane, isopentane and a mixture of two or more of the foregoing, catalyzing said mixture at a temperature of at least about 100° F. and a pressure of at least about 500 pounds per square inch to produce a catalyzed mixture containing a relatively high octane alcohol composition,*
- lowering the temperature of said catalyzed mixture to a temperature within a range of from about 90° to about 190° F.,*
- separating gas from said catalyzed mixture to provide the high octane fuel composition.*
18. The *fuel composition* of claim 17, wherein said *hydrocarbon material* comprises *butane*.
19. The *fuel composition* of claim 17, wherein said *hydrocarbon material* comprises *natural gasoline*.
20. The *fuel composition* of claim 17, wherein said *hydrocarbon material* comprises *straight run gasoline*.
21. The *fuel composition* of claim 17, wherein said *mixture* comprises *from about 5 to about 50% ethanol and from about 50 to about 95% hydrocarbon material*.
22. The *fuel composition* of claim 17, wherein said *catalyzing step* is conducted at a *temperature ranging from about 100° to about 460° F*.
23. The *fuel composition* of claim 17, wherein said *catalyzing step* is conducted at a *pressure ranging from about 500 to about 1000 pounds per square inch*.
24. The *fuel composition* of claim 17 wherein *hydrogen* is *injected into said catalyst bed during said catalyzing step*.
25. The *fuel composition* of claim 17 wherein said *fuel composition* has a *vapor pressure ranging from about 4 to about 19 psi*.
26. The *fuel composition* of claim 17 wherein said *high octane alcohol composition* has a *research octane rating ranging from about 108 to about 160 and a vapor pressure ranging from about 1.5 to about 8 psi*.

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