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**(54) A METHOD FOR PRODUCING OIL-BASED COMPONENTS**

VERFAHREN ZUR HERSTELLUNG VON KOMPONENTEN AUF ÖLBASIS

PROCÉDÉ DE PRODUCTION DE COMPOSANTS À BASE D'HUILE

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(73) Proprietor: **Neste Corporation**  
**02150 Espoo (FI)**

(72) Inventors:  
• **Makkonen, Jaana**  
**02150 Espoo (FI)**

• **Kettunen, Mika P.**  
**02150 Espoo (FI)**

(74) Representative: **Kolster Oy Ab**  
**(Salmisaarenaukio 1)**  
**P.O. Box 204**  
**00181 Helsinki (FI)**

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## Description

### FIELD OF THE INVENTION

**[0001]** The present invention relates to the field of producing traffic fuels and base oil components. More particularly, the invention relates to a method including hydrocracking and dewaxing of waxy feedstock for producing middle distillate fuel and lubricant base oil.

### BACKGROUND OF THE INVENTION

**[0002]** Cracking, and especially hydrocracking, is a well-known process in refinery used for cleaving larger hydrocarbon components into smaller short-chain hydrocarbons which are usable as traffic fuel components. Cracking is achieved by breaking the carbon-carbon bonds in the hydrocarbon chain of C15 to C45 typically in the presence of a cracking catalyst. The nature of the end products is dependent on the nature of the feed and on the process conditions under which the process is carried out, such as temperature, pressure and the nature of the catalyst.

**[0003]** A widely used method for carrying out catalytic cracking of high-boiling, high-molecular weight hydrocarbon fractions of petroleum crude oils is a fluid catalytic cracking (FCC) process, in which a powdered catalyst is employed. The catalyst particles are suspended in a rising flow of a heavy gas oil feed to form a fluidized bed. The feed is typically pre-heated and then sprayed into the base of the riser via feed nozzles to bring the feed in contact with the hot fluidized catalyst. The temperature of a FCC cracker is typically between 500°C and 800°C.

**[0004]** Hydrocracking of heavy hydrocarbons can also be achieved in a fixed bed hydrocracker. In this hydrocracker, the feedstock is fed downward through the catalyst bed together with hydrogen gas. In a fixed bed reactor, the catalyst is fixed onto particles, which can be of various sizes and forms providing an even and homogeneous distribution of the feedstock and hydrogen gas and, further, maximum contact with the active catalyst. The catalyst containing particles are fixed in layer or bed and the feedstock and hydrogen gas run through the layer. Typically, fixed bed reactors contain several catalyst layers.

**[0005]** A commonly used feedstock for hydrocracking is vacuum gas oil (VGO) which is heavy oils left-over from petroleum distillation. VGO contains a large quantity of cyclic and aromatic compounds as well as heteroatoms, such as sulphur and nitrogen, and other heavier compounds, depending on the crude source and VGO cut.

**[0006]** Hydrocarbon cracking is usually accompanied by a hydrogenation process, also called as hydrotreatment process. The purpose of the hydrogenation is to remove heteroatoms, such as oxygen, sulphur and/or nitrogen, and also to saturate aromatics typically present in the hydrocarbon feed. The hydrogenation process is typically performed before the actual hydrocracking process

in order to ensure removal of heteroatoms and saturation of aromatics. In some cases hydrogenation also includes ring-opening functions. However, no substantial cleavage of hydrocarbons takes place in the hydrogenation step. The preceding hydrogenation of the feedstock before hydrocracking ensures good hydrocracking performance and improves the yield of the products produced in the hydrocracking.

**[0007]** Waxes and slack waxes is a general term for feedstock comprising mainly paraffins. The paraffins in waxes are typically saturated C15-C45 linear hydrocarbons (normal or n-paraffins). Waxes may also contain to some extent branched (isomerized or i-paraffins) and cyclic hydrocarbons. Slack waxes are typically formed by a solvent dewaxing process of a waxy petroleum feed, such as a VGO stream. The solvent dewaxing process is typically performed by mixing a suitable solvent with the waxy petroleum feed, then cooling the mixture and separating the waxes as solid wax crystals, for example by filtration. Waxes can also be produced by Fischer-Tropsch (F-T) process, in which gas containing carbon monoxide and hydrogen is reacted to form hydrocarbon of various chain lengths. F-T process is widely used in gas-to-liquid (GTL) technologies where natural gas is used as a carbon source. Biomass can also be used as the carbon source in the F-T process and it is then generally called biomass-to-liquid (BTL) technology. Slack wax produced by solvent dewaxing contains varied amounts of impurities which makes the slack wax a low value product.

**[0008]** A catalytic dewaxing (cat-dewaxing) unit is a device where hydrocarbon feedstock is catalytically dewaxed, i.e. selectively cracked and isomerized. A cat-dewaxing unit is generally called as VHVI (Very High Viscosity Index) unit, since base oils with very high viscosity index are obtained in said unit. In the VHVI unit, in order to improve the cold properties of a base oil, linear paraffins are isomerized to form branched paraffins (iso-paraffins). Also, cracking of hydrocarbons takes place to some extent.

**[0009]** There is an ever increasing demand for high quality traffic fuels and base oil components in terms of purity, emissions, cold flow properties and lubricity of petroleum products. For example, the demand for middle distillates used as basic components in diesel fuel production is increasing in terms of cold flow properties and cetane number. Cetane number is an indicator of combustion speed of diesel fuel. The most important characteristic for a base oil component is the viscosity index, i.e. the measure for the change of viscosity with variations of temperature. High viscosity index of base oil component indicates that its lubricating ability is maintained over a large temperature range.

**[0010]** Various new technological developments have tried to meet the increasing demand for improved petroleum based product. US 2004/0256287 A1 discloses a process of hydrocracking a waxy hydrocarbon feedstock, such as Fischer-Tropsch waxy hydrocarbons, in the pres-

ence of a hydrocracking catalyst. The hydrocracking effluent is hydroisomerized in the presence of a hydroisomerization catalyst. The hydroisomerization effluent is fractionated to provide a heavy fraction and a middle distillate fuel, and the heavy fraction is dewaxed to provide a lubricant base oil having a viscosity index greater than 130.

**[0011]** US 2009/0065393 A1 discloses a method for obtaining a petroleum distillate in which a paraffin-based wax such Fischer-Tropsch wax and/or slack wax, is subjected to a cracking process in the presence of a catalyst, followed by hydrogenating the olefinic intermediate obtained in the presence of a catalyst, and recovering the distillate.

**[0012]** US2007/175794 and US6315891 disclose methods of producing base oils by hydrocracking a feedstock comprising a mixture of VGO and Fischer-Tropsch wax. US8404912 discloses a process of producing base oils by hydrocracking a mixture of VGO and a heavy wax derived from pyrolysis of plastic.

**[0013]** There remains a need for an efficient, simple and economic method of producing high quality middle distillates and very high viscosity index lubricant base oils in high yields from available feasible feeds and yet upgrading the product quality. The middle distillates and base oils desirably have high cetane number and high viscosity index, respectively.

#### BRIEF DESCRIPTION OF THE INVENTION

**[0014]** The present invention provides a method according to claim 1.

**[0015]** Vacuum gas oil is a petroleum based heavy hydrocarbon distillate which is widely used as a feedstock in fluid catalytic crackers to produce a wide range of valuable products for various applications, such as gasoline, diesel fuel and base oil. VGO contains a large quantity of cyclic and aromatic compounds as well as sulphur and nitrogen which impede its conversion to high quality valuable end products. As a rule, VGO quality influences on the quality of the end products. High quality Group III base oils with high viscosity index can be produced from suitable VGO in good yields. However, further increase of the viscosity index inevitably results in undesired lower yields.

**[0016]** It was surprisingly found in the present invention that high quality middle distillates and lubricant base oils in high yields can be produced when a feedstock for the overall refining process of hydrocracking and catalytic dewaxing is composed of VGO and wax. Also, it was surprisingly found that the viscosity index of a base oil was increased without compromising the yield, when wax in combination with VGO was used in the production of the base oil, compared to the viscosity index of a base oil produced from VGO only. It was further found that by introducing wax together with VGO to a hydrocracker, the performance of the hydrocracker catalyst was improved. In addition, the life time of the hydrocracking cat-

alyst was prolonged. Further, middle distillate with improved cetane number and base oil with increased viscosity index were produced in good yields.

**[0017]** A beneficial advantage of the method of the invention is that strict quality requirements of VGO as an adequate feedstock can be bargained when wax is introduced to the feedstock. In general, the adequate quality of the feedstock is adjusted by means of the VGO and wax. This means in practice that lower quality VGO with higher quality wax including a minor amount of impurities, and vice versa, can be used in the method of the invention and still high quality products are produced. This feature allows to employ VGO and waxes in a larger quality range and provides an economic advantage.

**[0018]** The method is efficient, simple and economic.

#### BRIEF DESCRIPTION OF THE DRAWINGS

**[0019]**

Figure 1 shows schematically an embodiment of the method of the invention.

Figure 2 shows an effect of slack wax to the viscosity index (VI) of base oil as a function of kinematic viscosity (KV).

#### DETAILED DESCRIPTION OF THE INVENTION

**[0020]** In an aspect, the present invention provides a method according to claim 1.

**[0021]** In the present invention, the term "slack wax" means a crude wax produced by chilling and solvent filter-pressing wax distillate; the term "vacuum gas oil (VGO)" means a hydrocarbon product obtained from crude oil vacuum distillation; the term "Group III Base oil" means paraffinic base stock which contains sulphur  $\leq 0.03$  wt.%, saturates  $\geq 90$  wt.% and has a viscosity index of  $\geq 120$ .

**[0022]** The feedstock suitable for use in the method of the invention typically comprises at most about 0.18 wt.% nitrogen, at most about 2 wt.% sulphur, and about 45 wt.% aromatic compounds.

**[0023]** The wax used is slack wax. The slack wax is provided as unpurified slack wax obtained from solvent dewaxing of a waxy petroleum feed.

**[0024]** Wax is combined with VGO to provide a feedstock comprising the VGO a major component and the wax as a minor component. In an embodiment, the feedstock comprises at most about 30 wt.% wax, the balance being the VGO. In another embodiment, the feedstock comprises at most about 25 wt.% wax. In still another embodiment, the feedstock comprises at most about 20 wt.% wax. In an embodiment, the feedstock comprises 1-25 wt.% wax. In another embodiment, the feedstock comprises 10-20 wt.% wax. In each embodiment, the balance is the VGO.

**[0025]** The feedstock comprising wax is subjected to a hydrocracking step. In the hydrocracking step, heter-

oatoms such as N and S are removed, larger long-chain hydrocarbons are cleaved into smaller short-chain hydrocarbons and/or some cyclic hydrocarbons are ring-opened to form linear and/or branched hydrocarbons. Also, dearomatization of the aromatic compounds and isomerization of the hydrocracked molecules may also occur to some extent in the process. However, the cleavage of the hydrocarbons of the wax in the hydrocracking step is not as extensive as that of VGO.

[0026] The hydrocracking is typically performed in the presence of a hydrocracking catalyst. Hydrocracking catalysts suitable for use in this step are well known to a skilled person in the art. The hydrocracking catalyst may be arranged in one or more layers in a fixed bed. The catalyst may also be arranged in graded catalyst bed. Alternatives for suitable arrangement of the catalyst are well known to a skilled person in the art.

[0027] In an embodiment, the feedstock is first subjected to a hydrotreatment step before the hydrocracking step. The hydrotreatment step is carried out under conditions where any heteroatoms, such as oxygen, sulphur and/or nitrogen present in the feedstock are removed. Also aromatic compounds are typically saturated in this step. No substantial cracking of the hydrocarbons in the feedstock takes place in this step. The hydrotreatment step before hydrocracking ensures good hydrocracking performance and improves the yield of the products produced in the subsequent hydrocracking. The hydrotreatment is typically performed in the presence of a catalyst. Catalysts suitable for use in this step are well known to a skilled person in the art. The catalyst may be arranged in one or more layers in a fixed bed. The catalyst may also be arranged in graded catalyst bed. Alternatives for suitable arrangement of the catalyst are well known to a skilled person in the art.

[0028] The hydrotreatment and hydrocracking steps can be conducted in a single reactor or separate reactors. When the two steps are conducted in separate reactors, the hydrotreatment reactor is arranged upstream of the hydrocracking reactor.

[0029] The feed rate of hydrogen per feedstock in the hydrocracking step, optionally including a hydrotreatment step, is about more than  $1000 \text{ Nm}^3/\text{m}^3$ . The hydrocracking is typically carried out at a pressure in the range from about 120 to about 170 bar. In an embodiment, the pressure is about 150 bar. The temperature is in the range of about  $350^\circ\text{C}$  to about  $450^\circ\text{C}$ .

[0030] The VGO and the wax forming a feedstock can be fed to the hydrocracking reactor as separate streams or combined into a single stream which is fed to the reactor.

[0031] The first effluent obtained from the hydrocracking step is fractionated by distillation whereby at least a bottom fraction and a middle distillate fraction are obtained. Adequate fuel oil quality for the middle distillate fraction is achieved and it is suitable for use as a component in diesel fuel. The bottom fraction is mainly composed of linear paraffins and naphtenes. Fractionation

of the first effluent also produces a quantity of light gaseous hydrocarbons.

[0032] In an embodiment, for further refining, the bottom fraction is subjected to a dewaxing step. In the dewaxing step, waxy n-paraffins are isomerized to provide branched iso-paraffins. Isomerization of hydrocarbons is desired and generally improves the cold flow properties of a base oil. The dewaxing is typically performed in the presence of a dewaxing catalyst. Dewaxing catalysts suitable for use in this step are well known to a skilled person in the art.

[0033] The dewaxing step provides a second effluent which is fractionated in a distiller to high quality middle distillate and base oil. Also lighter gaseous hydrocarbons are obtained. Group III Base oil having desirably a high viscosity index of  $>130$  and middle distillate with high cetane number and improved cold flow properties are achieved. The middle distillate obtained from the dewaxing step can be used as diesel fuel as such or as a blending component.

[0034] The present invention can be a batch process or a continuous process.

[0035] An example of an arrangement which can be used for conducting the method of the invention is shown in Figure 1. Referring to the figure, the arrangement for producing oil-based components comprises a hydrocracking reactor A for cracking a waxy feed to provide a first effluent 4, a first distiller B, a dewaxing unit C and a second distiller D. The hydrocracking reactor A comprises at least one inlet for supplying a feedstock into the hydrocracking reactor A and an outlet for discharging the first effluent 4 from the hydrocracking reactor A. In an embodiment of the invention, the hydrocracking reactor A comprises at least two inlets, one for wax 2 and one for combined hydrogen 1 and VGO 3. In the figure, the hydrocracking reactor A comprises a catalytic hydrotreatment zone HT and a catalytic hydrocracking zone HC, the hydrotreatment zone lying upstream of the hydrocracking zone. The catalysts in each of the two zones can be arranged in one or more beds. Further, the beds can be graded in respect to the amount of the catalyst employed. A suitable arrangement and grading of the catalyst are well known to a skilled person in the art. Alternatively, the hydrotreatment zone and the hydrocracking zone can be arranged in individual reactors, the hydrotreatment reactor lying upstream of the hydrocracking reactor (not shown in Fig. 1).

[0036] The first distiller B is in a flow connection with the hydrocracking reactor A for fractionating the first effluent 4 to provide at least a bottom fraction HCB and a middle distillate fraction MD. The first distiller B comprises an inlet for supplying the first effluent 4 into the first distiller B and at least one outlet for discharging the bottom fraction HCB and a middle distillate fraction MD. The flow connection between the hydrocracking reactor A and the first distiller B is arranged through a pipe connection between the outlet of the hydrocracking reactor A and the inlet of the first distiller B. The first distiller B is ar-

ranged downstream of the hydrocracking reactor A. The dewaxing unit C is in a flow connection with the first distiller B for dewaxing the bottom fraction HCB to provide a second effluent 6. In an embodiment, unit C is a solvent dewaxing reactor. The dewaxing unit C comprises an inlet for supplying the bottom fraction HCB into the dewaxing unit C and an outlet for discharging the second effluent 6. The flow connection between the first distiller B and the dewaxing unit C is arranged through a pipe connection between the outlet of the first distiller B discharging the bottom fraction HCB and the inlet of the dewaxing unit C. The dewaxing unit C is arranged downstream of the first distiller B. The second distiller D is in a flow connection with the dewaxing unit C for fractionating the second effluent 6 to provide at least a middle distillate fraction MD and base oil. The second distiller D comprises an inlet for supplying second effluent 6 into the second distiller D and at least one outlet, but preferably two outlets, for discharging the middle distillate MD and base oil. The flow connection between the dewaxing unit C and the second distiller D is arranged through a pipe connection between the outlet of dewaxing unit C and the inlet of the second distiller D. The second distiller D is arranged downstream of the dewaxing unit C and comprises at least one outlet for discharging the middle distillate MD and base oil.

**[0037]** In a further aspect, the disclosure provides an arrangement for producing oil-based components, the apparatus comprising:

- a hydrocracking reactor A for cracking a waxy feed to provide a first effluent 4, said hydrocracking reactor A comprising at least one inlet for supplying a feedstock to the hydrocracking reactor A and an outlet for discharging the first effluent 4 from the hydrocracking reactor A;
- a first distiller B in a flow connection with the hydrocracking reactor A for fractionating the first effluent 4 to provide at least a bottom fraction HCB and a middle distillate fraction MD, the first distiller B being arranged downstream of the hydrocracking reactor A;
- a dewaxing unit C in a flow connection with the first distiller B for dewaxing the bottom fraction HCB to provide a second effluent 6, the dewaxing unit C being arranged downstream of the first distiller B; and
- a second distiller D in a flow connection with the dewaxing unit C for fractionating the second effluent 6 to provide at least a middle distillate MD and base oil, the second distiller D being arranged downstream of the dewaxing unit C and comprises at least one outlet for discharging the middle distillate MD and the base oil.

**[0038]** The following examples are given for further illustration of the invention without limiting the invention thereto.

## Example

**[0039]** Feedstock comprising 20 wt.% slack wax and a balance of VGO was introduced to a hydrocracking reactor comprising a hydrotreatment zone and a hydrocracking zone. The content of sulphur and nitrogen of the feedstock was 1.3 wt.% and 0.079 wt.%, respectively. The viscosity index of the feedstock was 106.

**[0040]** The feedstock was introduced to the hydrocracking reactor at a temperature of about 400°C and at a pressure of about 132 bar. The feed of hydrogen per feedstock was 1.1 L/L.

**[0041]** As a reference, 100% VGO feedstock was introduced to the hydrocracking reactor under the same conditions.

**[0042]** The effluent from the hydrocracking reactor was fed to a distiller. A bottom fraction (HCB) and a middle distillate fraction were obtained. The yield of the bottom fraction with 20 wt.% slack wax addition to VGO was increased from 31% to 35% as compared to a VGO feed without slack wax. The cetane number of a middle distillate fraction was increased from 60 (100% VGO) to 64 (80% VGO/20% slack wax).

**[0043]** The bottom fraction was solvent dewaxed (SDW) to provide base oil and additional middle distillate. Catalytic dewaxing processes are especially developed to improve the cold properties of the base oil products without sacrificing the viscosity index. Therefore, SDW can be used as an indicator of the viscosity index also of the catalytically dewaxed product. SDW is also a generally accepted and used method in the field and correlates well with catalytic dewaxing.

**[0044]** Figure 2 shows that the viscosity index of base oil was remarkable higher with 20 wt.% slack wax addition than that of pure VGO feed. Base oil obtained from the solvent dewaxing, having a viscosity index of at least 130 was achieved even with 4 cSt product. Figure 2 further shows that the viscosity index of the base oil is increased by more than about 10 units compared to that of base oil which is prepared from VGO without wax.

**[0045]** It will be obvious to a person skilled in the art that, as the technology advances, the inventive concept can be implemented in various ways. The invention and its embodiments are not limited to the examples described above but may vary within the scope of the claims.

## Claims

1. A method of producing oil-based components, comprising the steps of:
  - providing VGO and slack wax;
  - combining the VGO as a major component and the slack wax as a minor component to provide a feedstock;
  - subjecting the feedstock to a hydrocracking

step to provide a first effluent;  
 - fractionating the first effluent to provide at least a bottom fraction and a middle distillate fraction;  
 - recovering the bottom fraction and the middle distillate fraction, wherein the feedstock comprises at most 30 wt. % slack wax and the slack wax is provided as unpurified slack wax obtained from solvent dewaxing of a waxy petroleum feed.

## 2. The method of claim 1, wherein

- the bottom fraction is subjected to a dewaxing step to provide a second effluent;  
 - the second effluent is fractionated to provide at least a middle distillate and base oil;  
 - recovering the middle distillate and the base oil.

## 3. The method of any one of the preceding claims, wherein the feedstock comprises at most 25 wt. % slack wax, more specifically at most 20 wt. %, even more specifically 1-25 wt. %, still even more specifically 10-20 wt. %, the balance being the VGO.

## 4. The method of any one of the preceding claims, wherein the feedstock comprises at most 0.18 wt. % nitrogen, at most 2 wt. % sulphur, and about 45 wt. % aromatic compounds.

## 5. The method of any one of the preceding claims, wherein the feedstock is subjected to a hydrotreatment step before the hydrocracking step.

## Patentansprüche

### 1. Verfahren zum Herstellen von Komponenten auf Öl-basis, das die folgenden Schritte umfasst:

- Bereitstellen von VGO (Vakuumgasölen) und Gatsch;  
 - Kombinieren des VGO als Hauptkomponente und Gatsch als Nebenkomponekte, um ein Ausgangsmaterial bereitzustellen;  
 - Unterziehen des Ausgangsmaterials einem Hydrocracking-Schritt, um einen ersten Abfluss bereitzustellen;  
 - Fraktionieren des ersten Abflusses, um mindestens eine Bodenfraktion und eine mittlere Destillatfraktion bereitzustellen;  
 - Rückgewinnen der Bodenfraktion und der mittleren Destillatfraktion, wobei das Ausgangsmaterial höchstens 30 Gew.-% Gatsch enthält, und Gatsch wird als ungereinigtes Gatsch bereitgestellt, das aus dem Lösungsmittel-Entwachsen einer wachshaltigen Petroleum-Zufuhr gewonnen wird.

### 2. Verfahren nach Anspruch 1, wobei

- die Bodenfraktion einem Entwachsungsschritt unterzogen wird, um für einen zweiten Abfluss bereitzustellen;  
 - der zweite Abfluss wird fraktioniert, um zumindest ein mittleres Destillat und Basisöl bereitzustellen;  
 - Rückgewinnung des mittleren Destillats und des Basisöls.

### 3. Verfahren nach einem der vorherigen Ansprüche, wobei das Ausgangsmaterial höchstens 25 Gew.-% Gatsch, spezieller höchstens 20 Gew.-%, noch spezieller 1-25 Gew.-%, noch spezieller 10-20 Gew.-% umfasst, wobei der Rest das VGO ist.

### 4. Verfahren nach einem der vorherigen Ansprüche, wobei das Ausgangsmaterial höchstens 0,18 Gew.-% Stickstoff, höchstens 2 Gew.-% Schwefel und etwa 45 Gew.-% aromatische Verbindungen enthält.

### 5. Verfahren nach einem der vorherigen Ansprüche, wobei das Ausgangsmaterial einem Hydrobehandlungsschritt vor dem Hydrocracking-Schritt unterzogen wird.

## Revendications

### 1. Procédé pour produire des composants à base d'huile, comprenant les étapes suivantes :

- obtention de VGO (gazole sous vide) et de gatsch ;  
 - combinaison du VGO en tant que composant majeur et du gatsch en tant que composant mineur pour former une charge ;  
 - soumission de la charge à une étape d'hydrocraquage pour former un premier effluent ;  
 - fractionnement du premier effluent pour former au moins une fraction de fond et une fraction de distillat moyen ;  
 - récupération de la fraction de fond et de la fraction de distillat moyen, dans lequel la charge comprend au plus 30 % en poids de gatsch et le gatsch est fourni sous la forme de gatsch non purifié obtenu à partir d'un déparaffinage au solvant d'une charge de pétrole paraffineux.

### 2. Procédé selon la revendication 1, dans lequel

- la fraction de fond est soumise à une opération de déparaffinage pour former un deuxième effluent ;  
 - le deuxième effluent est fractionné pour former au moins un distillat moyen et une huile de base ;  
 - le distillat moyen et l'huile de base sont récu-

pérés.

3. Procédé selon l'une quelconque des revendications précédentes, dans lequel la charge comprend au plus 25 % en poids de gatsch, plus spécifiquement au plus 20 % en poids, encore plus spécifiquement 1 à 25 % en poids, plus spécifiquement encore 10 à 20 % en poids, le reste étant le VGO. 5
4. Procédé selon l'une quelconque des revendications précédentes, dans lequel la charge comprend au plus 0,18 % en poids d'azote, au plus 2 % en poids de soufre, et environ 45 % en poids de composés aromatiques. 10  
15
5. Procédé selon l'une quelconque des revendications précédentes, dans lequel la charge est soumise à une étape d'hydrotraitement avant l'étape d'hydrocraquage. 20  
25  
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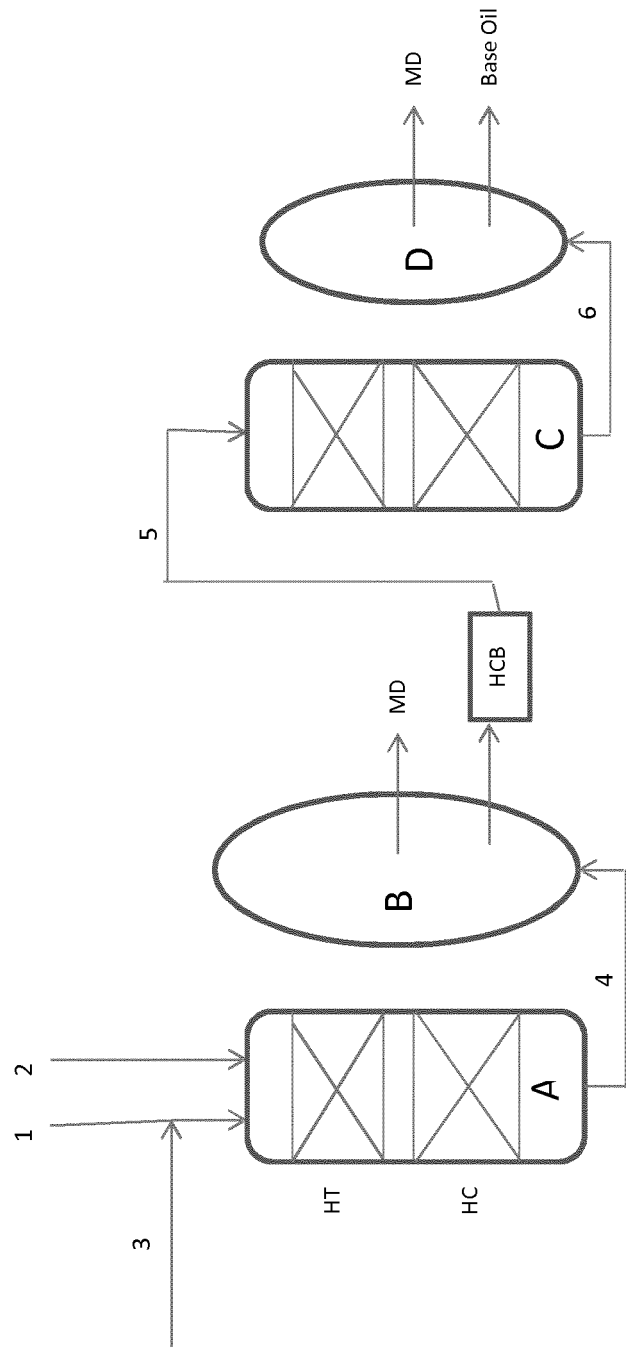


Fig. 1



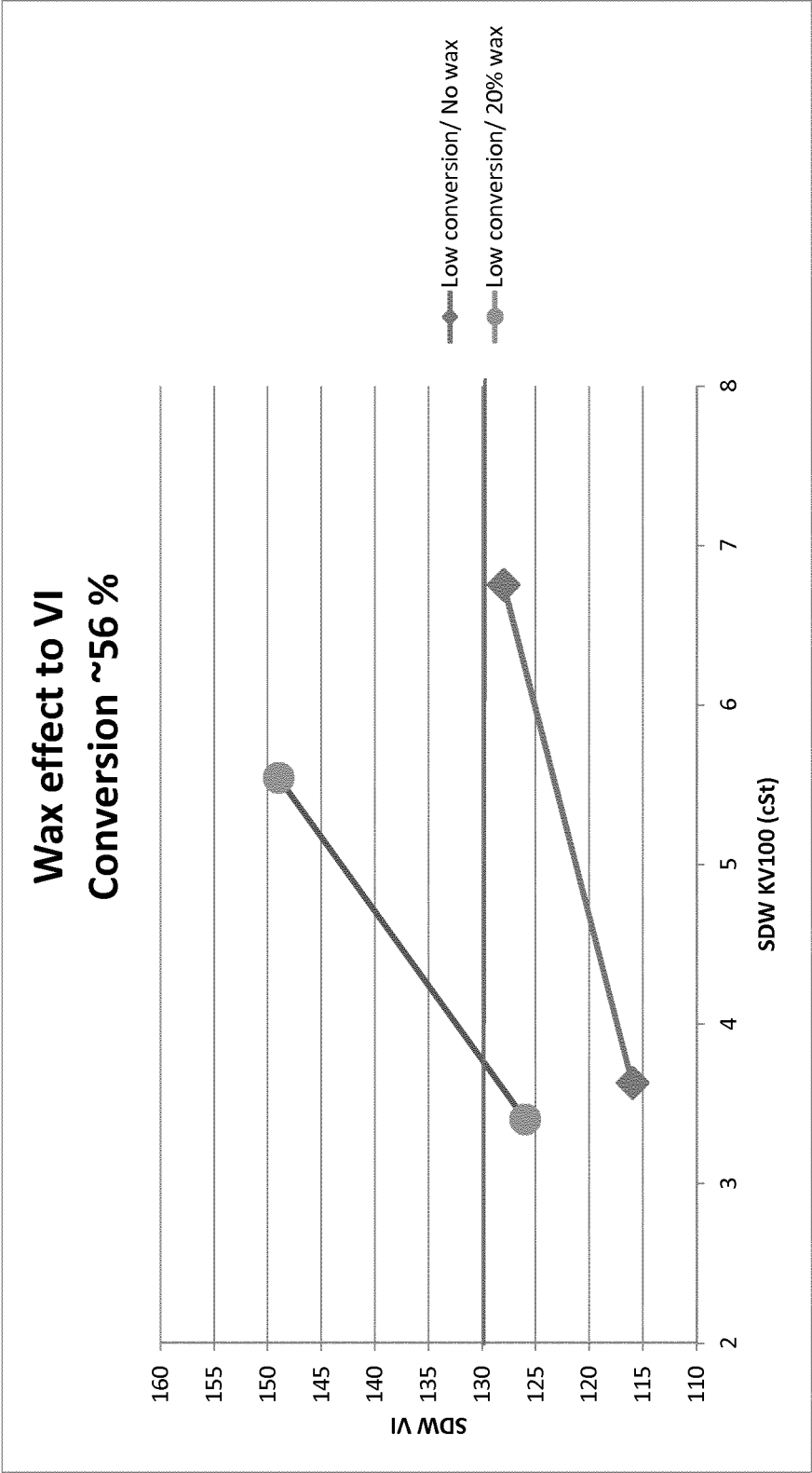


Fig. 2

**REFERENCES CITED IN THE DESCRIPTION**

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