PROCESS FOR OXIDATIVE DESULFURIZATION AND DENITROGENATION USING OXIDATION, FOLLOWED BY SOLVENT EXTRACTION AND A FLUID CATALYTIC CRACKING (FCC) PROCESS

Figure 1

A method and apparatus for recovering components from a hydrocarbon feedstock is provided. The method includes the steps of (a) supplying a hydrocarbon feedstock to an oxidation reactor, wherein the hydrocarbon feedstock is oxidized in the presence of a catalyst under conditions sufficient to selectively oxidize sulfur compounds and nitrogen compounds present in the hydrocarbon feedstock; (b) separating the hydrocarbons, the oxidized sulfur compounds, and the oxidized nitrogen compounds by solvent extraction; (c) collecting a residue stream that includes the oxidized sulfur compounds and the oxidized nitrogen compounds; and (d) supplying the residue stream to a fluid catalytic cracking unit.
**UTILITY PATENT APPLICATION**

PROCESS FOR OXIDATIVE DESULFURIZATION AND DENITROGENATION USING OXIDATION, FOLLOWED BY SOLVENT EXTRACTION AND A FLUID CATALYTIC CRACKING (FCC) PROCESS

INVENTORS: Abdennour Bourane  
OMer Refa Koseoglu  
Stephane Kressmann

ATTORNEY DOCKET: 004159.001749 (SA649 PCT)

**Field of the Invention**

[0001] This invention relates to a method and apparatus for recovering sulfur and nitrogen from a hydrocarbon feedstock. More specifically, the present invention relates to a method and apparatus for oxidative desulfurization and denitrogenation of a hydrocarbon stream and the subsequent disposal of resulting oxidized sulfur and nitrogen compounds.

**Background of the Invention**

[0002] Crude oil is the world's main source of hydrocarbons used as fuel and petrochemical feedstock. At the same time, petroleum and petroleum based products are also a major source for air and water pollution today. To address growing concerns surrounding pollution caused by petroleum and petroleum based products, many countries have implemented strict regulations on petroleum products, particularly on petroleum refining operations and the allowable concentrations of specific pollutants in fuels, such as the allowable sulfur and nitrogen content in gasoline fuels. While the exact compositions of natural petroleum or crude oils vary significantly, all crude oils contain some measurable amount of sulfur compounds and most crude oils also contain some measurable amount of nitrogen compounds. In addition, crude oils
may also contain oxygen, but oxygen content of most crude is low. Generally, sulfur concentrations in crude oils are less than about 5 percent by weight, with most crude oils having sulfur concentrations in the range from about 0.5 to about 1.5 percent by weight. Nitrogen concentrations of most crude oils are usually less than 0.2 percent by weight, but can be as high as 1.6 percent by weight. In the United States, motor gasoline fuel is regulated to have a maximum total sulfur content of less than 10 ppm sulfur.

[0003] Crude oils are refined in oil refineries to produce transportation fuels and petrochemical feedstocks. Typically fuels for transportation are produced by processing and blending of distilled fractions from the crude oil to meet the particular end use specifications. Because most of the crudes generally available today have high concentrations of sulfur, the distilled fractions typically requires desulfurization to yield products which meet various performance specifications and/or environmental standards.

[0004] The sulfur-containing organic compounds present in crude oils and resulting refined fuels can be a major source of environmental pollution. The sulfur compounds are typically converted to sulfur oxides during the combustion process, which in turn can produce sulfur oxyacids and contribute to particulate emissions.

[0005] One method for reducing particulate emissions includes the addition of various oxygenated fuel blending compounds and/or compounds that contain few or no carbon-to-carbon chemical bonds, such as methanol and dimethyl ether. Most of these compounds, however, suffer in that they can have high vapor pressures, be nearly insoluble in diesel fuel, and/or have poor ignition quality, as indicated by their cetane numbers.
Diesel fuels that have been treated by chemical hydrotreating and/or hydrogenation to reduce their sulfur and aromatics contents can have a reduced fuel lubricity, which in turn can cause excessive wear of fuel pumps, injectors and other moving parts that come in contact with the fuel under high pressures.

For example, middle distillates (a distillate fraction that nominally boils in the range of about 180-370°C) can be used as a fuel, or alternatively can be used as a blending component of fuel for use in compression ignition internal combustion engines (i.e., diesel engines). The middle distillate fraction typically include between about 1 and 3% by weight sulfur. Allowable sulfur concentration in middle distillate fractions were reduced to 5-50 part per million weight (ppmw) levels from 3000 ppmw level since 1993 in Europe and United States.

In order to comply with the increasingly stringent regulations for ultra-low sulfur content fuels, refiners must make fuels having even lower sulfur levels at the refinery gate so that they can meet the specifications after blending.

Low pressure conventional hydrodesulfurization (HDS) processes can be used to remove a major portion of the sulfur from petroleum distillates for the blending of refinery transportation fuels. These units, however, are not efficient to remove sulfur from compounds at mild conditions (i.e., 30 bar pressure) when the sulfur atom is sterically hindered as in multi-ring aromatic sulfur compounds. This is particularly true where the sulfur heteroatom is hindered by two alkyl groups (e.g., 4,6-dimethyldibenzothiophene). Because of the difficulty in the removal the hindered dibenzothiophenes predominate at low sulfur levels such as 50 ppmw to 100 ppmw. Severe operating conditions (i.e., high hydrogen partial pressure, high temperature, high catalyst volume) must be utilized in order to remove the sulfur from these refractory sulfur compounds.
Increasing the hydrogen partial pressure can only be achieved by increasing the recycle gas purity, or new grassroots units must be designed, which can be a very a costly option. The use of severe operating conditions typically results in decreased yield, lower catalyst life cycle, and product quality deterioration (e.g., color), and therefore are typically sought to be avoided.

[0010] Conventional methods for petroleum upgrading, however, suffer from various limitations and drawbacks. For example, hydrogenative methods typically require large amounts of hydrogen gas to be supplied from an external source to attain desired upgrading and conversion. These methods can also suffer from premature or rapid deactivation of catalyst, as is typically the case during hydrotreatment of a heavy feedstock and/or hydrotreatment under harsh conditions, thus requiring regeneration of the catalyst and/or addition of new catalyst, which in turn can lead to process unit downtime. Thermal methods frequently suffer from the production of large amounts of coke as a byproduct and a limited ability to remove impurities, such as, sulfur and nitrogen. Additionally, thermal methods require specialized equipment suitable for severe conditions (high temperature and high pressure), and require the input of significant energy, thereby resulting in increased complexity and cost.

[0011] Thus, there exists a need to provide a process for the upgrading of hydrocarbon feedstocks, particularly processes for the desulfurization and denitrogenation of hydrocarbons that use low severity conditions that can also provide means for the recovery and disposal of usable sulfur and nitrogen compounds.
SUMMARY OF THE INVENTION

[0012] The current invention provides a method and apparatus for the upgrading of a hydrocarbon feedstock that removes a major portion of the sulfur and nitrogen present and in turn utilizes these compounds in an associated process.

[0013] In one aspect, a method of upgrading a hydrocarbon feedstock is provided. The method includes the steps of (a) supplying a hydrocarbon feedstock to an oxidation reactor, wherein the hydrocarbon feedstock includes sulfur compounds and nitrogen compounds; (b) contacting the hydrocarbon feedstock with an oxidant in the presence of a catalyst in the oxidation reactor under conditions sufficient to selectively oxidize sulfur compounds and nitrogen compounds present in the hydrocarbon feedstock to produce an oxidized hydrocarbon stream that includes hydrocarbons, oxidized sulfur compounds, and oxidized nitrogen compounds; (c) separating the hydrocarbons and the oxidized sulfur and nitrogen compounds by solvent extraction with a polar solvent to produce an extracted hydrocarbon stream and a mixed stream, wherein the mixed stream includes the polar solvent, the oxidized sulfur compounds, and the oxidized nitrogen compounds, and wherein the extracted hydrocarbon stream has a lower concentration of sulfur compounds and nitrogen compounds than the hydrocarbon feedstock; (d) separating the mixed stream into a first recovered polar solvent stream and a first residue stream; and (e) supplying the first residue stream to a fluid catalytic cracking (FCC) unit to catalytically crack the oxidized sulfur and the oxidized nitrogen to recover hydrocarbons from the first residue stream.

[0014] In certain embodiments, the oxidants are selected from the group consisting of air, oxygen, peroxides, hydroperoxides, organic peracids, ozone, and combinations thereof. In
certain embodiments, the catalyst is a metal oxide having the formula \( M_xO_y \), wherein \( M \) is an element selected from Groups IVB, VB, and VIB of the periodic table. In certain embodiments, the polar solvent has a Hildebrandt value of greater than about 19.

[0015] In another embodiment, the present invention provides a method of upgrading a hydrocarbon feedstock. The method includes the steps of supplying a hydrocarbon feedstock to an oxidation reactor, wherein the hydrocarbon feedstock includes sulfur compounds and nitrogen compounds. Then catalytically oxidizing the sulfur compounds and the nitrogen compounds in the hydrocarbon feedstock in the oxidation reactor with an oxidant in the presence of a catalyst under conditions sufficient to selectively oxidize the sulfur compounds present in the hydrocarbon feedstock to sulfones, oxidize the nitrogen compounds present in the hydrocarbon feedstock, and produce an oxidized hydrocarbon stream that includes hydrocarbons, sulfones, and oxidized nitrogen compounds. Then extracting the oxidized hydrocarbon stream with a polar organic solvent to produce an extracted hydrocarbon stream and a mixed stream, wherein the mixed stream includes the polar organic solvent, the sulfones, and the oxidized nitrogen compounds and wherein the extracted hydrocarbon stream has a lower sulfur concentration and nitrogen concentration than the hydrocarbon feedstock. The mixed stream is separated into a first recovered polar solvent stream and a first residue stream that includes the sulfones and the oxidized nitrogen compounds. The extracted hydrocarbon stream is supplied to a stripper, wherein the stripper is operable to separate the extracted hydrocarbon stream into a stripped oil stream and a second recovered polar solvent stream. The first recovered polar solvent stream and second recovered polar solvent stream are supplied to the extraction step. The residue stream which includes sulfones and the oxidized nitrogen compounds is supplied to a fluid
catalytic cracking (FCC) unit where the sulfones and the oxidized nitrogen compounds are catalytically cracked to allow for recovery of hydrocarbons from the residue stream.

**BRIEF DESCRIPTION OF THE DRAWINGS**

[0016] So that the manner in which the above-recited features, aspects and advantages of the invention, as well as others that will become apparent, are attained and can be understood in detail, more particular description of the invention briefly summarized above can be had by reference to the embodiments thereof that are illustrated in the drawings that form a part of this specification. It is to be noted, however, that the appended drawings illustrate some embodiments of the invention and are, therefore, not to be considered limiting of the invention's scope, for the invention can admit to other equally effective embodiments.

[0017] Figure 1 provides a schematic diagram of one embodiment of the method of upgrading a hydrocarbon feedstock according to the present invention.

[0018] Figure 2 provides a schematic diagram of one embodiment of the method of upgrading a hydrocarbon feedstock according to the present invention.

[0019] Figure 3 provides a schematic diagram of the process described in the example in accordance with an embodiment of the present invention.

**DETAILED DESCRIPTION OF THE INVENTION**

[0020] Although the following detailed description contains many specific details for purposes of illustration, it is understood that one of ordinary skill in the art will appreciate that many examples, variations and alterations to the following details are within the scope and spirit
of the invention. Accordingly, the exemplary embodiments of the invention described herein and provided in the appended figures are set forth without any loss of generality, and without imposing limitations, relating to the claimed invention.

[0021] The present invention addresses problems associated with prior art methods of upgrading and recovering compounds from a hydrocarbon feedstock, particularly the desulfurization and denitrogenation of hydrocarbon feedstocks, and the subsequent removal and recovery of usable hydrocarbons. In one aspect, the present invention provides a method for the removal of sulfur and nitrogen compounds from a hydrocarbon feedstock and the use of oxidized sulfur species and oxidized nitrogen species in a fluid catalytic cracking process.

[0022] As used herein, the terms "upgrading" or "upgraded", with respect to petroleum or hydrocarbons refers to a petroleum or hydrocarbon product that is lighter (i.e., has fewer carbon atoms, such as methane, ethane, and propane), has at least one of a higher API gravity, higher middle distillate yield, lower sulfur content, lower nitrogen content, or lower metal content, than does the original petroleum or hydrocarbon feedstock.

[0023] Figure 1 provides one embodiment of the present invention for the recovery of hydrocarbons. Hydrocarbon recovery system 100 includes oxidation reactor 104, extraction vessel 112, solvent regeneration column 116, stripper 120, and fluid catalytic cracking unit 130.

[0024] In one aspect, the present invention provides a method for the recovery of components from a hydrocarbon feedstock, particularly a hydrocarbon feedstock that includes sulfur and nitrogen containing compounds. The method includes supplying hydrocarbon feedstock 102 to oxidation reactor 104, where the hydrocarbon feedstock is contacted with an oxidant and a
catalyst. The oxidant can be supplied to oxidation reactor 104 via oxidant feed line 106 and fresh catalyst can be supplied to the reactor via catalyst feed line 108.

[0025] Hydrocarbon feedstock 102 can be any petroleum based hydrocarbon, and can include various impurities, such as elemental sulfur, and/or compounds that include sulfur and/or nitrogen. In certain embodiments, hydrocarbon feedstock 102 can be a diesel oil having a boiling point between about 150°C and about 400°C. Alternatively, hydrocarbon feedstock 102 can have a boiling point up to about 450°C, alternatively up to about 500°C. Alternatively, hydrocarbon feedstock 102 can have a boiling point between about 100°C and about 500°C. Optionally, hydrocarbon feedstock 102 can have a boiling point up to about 600°C, alternatively up to about 700°C, or, in certain embodiments, greater than about 700°C. In an aspect, the feedstock exists in a solid state after distillation called residue. In certain embodiments, hydrocarbon feedstock 102 can include heavy hydrocarbons. As used herein, heavy hydrocarbons refer to hydrocarbons having a boiling point of greater than about 360°C, and can include aromatic hydrocarbons, as well as alkanes and alkenes. Generally, in certain embodiments, hydrocarbon feedstock 102 can be selected from whole range crude oil, topped crude oil, product streams from oil refineries, product streams from refinery steam cracking processes, liquefied coals, liquid products recovered from oil or tar sand, bitumen, oil shale, asphaltene, hydrocarbon fractions such as diesel and vacuum gas oil boiling in the range of 180-370°C and 370-520°C, respectively, and the like, and mixtures thereof.

[0026] Exemplary sulfur compounds present in hydrocarbon feedstock 102 can include sulfides, disulfides, and mercaptans, as well as aromatic molecules such as thiophenes, benzo thiophenes, dibenzothiophenes, and alkyl dibenzothiophenes, such as 4,6-dimethyl-
dibenzothiophene. Aromatic compounds are typically more abundant in higher boiling fractions, than is typically found in the lower boiling fractions.

[0027] Exemplary nitrogen containing compounds present in hydrocarbon feedstock 102 can include compounds having the following structures:

Neutral

Indoles
Carbazoles

Basic

Anilines
Quinolines
Acridines

Please note that the sulfur oxidation is the limiting targeted reaction, during which nitrogen oxidation occurs. Two types could be considered basic and neutral nitrogen.

[0028] Oxidation reactor 104 can be operated at mild conditions. More specifically, in certain embodiments, oxidation reactor 104 can be maintained at a temperature of between about 30°C and about 350°C, or alternatively, between about 45°C and about 60°C. The operating pressure of oxidation reactor 104 can be between about 1 bar and about 30 bars, alternatively between about 1 bar and about 15 bars, alternatively between about 1 bar and about 10 bars, or alternatively between about 2 bars and about 3 bars. The residence time of the hydrocarbon feedstock within oxidation reactor 104 can be between about 1 minutes and about 120 minutes, alternatively between about 15 minutes and about 90 minutes, alternatively between about 5 minutes and about 90 minutes, alternatively between about 5 minutes and about 30 minutes,
alternatively between about 30 minutes and about 60 minutes, and is preferably for a sufficient amount of time for the oxidation of any sulfur or nitrogen compounds present in the hydrocarbon feedstock. In one embodiment, the residence time of the hydrocarbon feedstock within oxidation reactor 104 is between about 15 minutes and about 90 minutes.

[0029] Oxidation reactor 104 can be any reactor suitably configured to ensure sufficient contacting between hydrocarbon feedstock 102 and the oxidant, in the presence of a catalyst, for the oxidation of the sulfur and nitrogen containing compounds. Sulfur and nitrogen compounds present in hydrocarbon feedstock 102 are oxidized in oxidation reactor 104 to sulfones, sulfoxides, and oxidized nitrogen compounds, which can be subsequently removed by extraction and/or adsorption. Various types of reactors can be used in embodiments of the present invention. For example, the reactor can be a batch reactor, a fixed bed reactor, an ebullated bed reactor, lifted reactor, a fluidized bed reactor, a slurry bed reactor, or combinations thereof. Other types of suitable reactors that can be used will be apparent to those of skill in the art and are to be considered within the scope of the present invention. Examples of suitable oxidized nitrogen compounds can include pyridine-based compounds and pyrrole-based compounds. It is believed that the nitrogen atom is not directly oxidized, rather it is the carbon atom(s) next to the nitrogen that is actually oxidized. A few examples of oxidized nitrogen compounds can include the following compounds:
The oxidant is supplied to oxidation reactor 104 via oxidant feed stream 106. Suitable oxidants can include air, oxygen, hydrogen peroxide, organic peroxides, hydroperoxides, organic peracids, peroxy acids, oxides of nitrogen, ozone, and the like, and combinations thereof. Exemplary peroxides can be selected from hydrogen peroxide and the like. Exemplary hydroperoxides can be selected from t-butyl hydroperoxide and the like. Exemplary organic peracids can be selected from peracetic acid and the like.

The mole ratio of oxidant to sulfur present in the hydrocarbon feedstock can be from about 1:1 to about 50:1, preferably between about 2:1 and about 20:1, more preferably between...
about 4:1 and about 10:1. In an aspect, the molar feed ratio of oxidant to sulfur can range from about 1:1 to about 30:1. In an aspect, the molar feed ratio of oxidant to nitrogen compounds can be from about 4:1 to about 10:1. In an aspect, the feedstock can contain more nitrogen compounds than sulfur, such as, for instance, South American Crude oils, Africa crude oils, Russian crude oils, China crude oils, or intermediate refinery streams such as coker, thermal cracking, visbreaking, gas oils, fluid catalytic cracking (FCC) cycle oils, and the like.

[0032] The catalyst can be supplied to oxidation reactor 104 via catalyst feed stream 108. The catalyst can be a homogeneous catalyst. The catalyst can include at least one metal oxide having the chemical formula MₓOᵧ, wherein M is a metal selected from groups IVB, VB, or VIB of the periodic table. Exemplary metals can include titanium, vanadium, chromium, molybdenum, and tungsten. Molybdenum and tungsten are two particularly effective catalysts that can be used in embodiments of the present invention. In an aspect, the spent catalyst can be rejected from the system with the aqueous phase (e.g., when using an aqueous oxidant) after the oxidation vessel.

[0033] The ratio of catalyst to oil is between about 0.1% by weight and about 10% by weight, preferably between about 0.5% by weight and about 5% by weight. In certain embodiments, the ratio is between about 0.5% by weight and about 2.5% by weight. Alternatively, the ratio is between about 2.5% by weight and about 5% by weight. Other suitable weight ratios of catalyst to oil will be apparent to those of skill in the art and are to be considered within the scope of the present invention.

[0034] Catalyst present in oxidation reactor 104 can increase the rate of oxidation of the various sulfur and nitrogen containing compounds in hydrocarbon feedstock 102, and/or reduce
the amount of oxidant necessary for the oxidation reaction. In certain embodiments, the catalyst can be selective toward the oxidation of sulfur species. In another aspect, the catalyst can be selective toward the oxidation of nitrogen species.

[0035] Oxidation reactor 104 produces oxidized hydrocarbon stream 110, which can include hydrocarbons and oxidized sulfur and oxidized nitrogen containing species. Oxidized hydrocarbon stream 110 is supplied to extraction vessel 112 where the oxidized hydrocarbon stream and oxidized sulfur and oxidized nitrogen containing species are contacted with extraction solvent stream 132. The extraction solvent can be a polar solvent, and in certain embodiments, can have a Hildebrandt solubility value of greater than about 19. In certain embodiments, when selecting the particular polar solvent for use in extracting oxidized sulfur and oxidized nitrogen containing species, selection can be based upon, in part, solvent density, boiling point, freezing point, viscosity, and surface tension. Exemplary polar solvents suitable for use in the extraction step can include acetone (Hildebrand value of 19.7), carbon disulfide (20.5), pyridine (21.7), dimethyl sulfoxide (DMSO) (26.4), n-propanol (24.9), ethanol (26.2), n-butyl alcohol (28.7), propylene glycol (30.7), ethylene glycol (34.9), dimethylformamide (DMF) (24.7), acetonitrile (30), methanol (29.7), and the like. In certain embodiments, acetonitrile and methanol, due to their low cost, volatility, and polarity, are preferred. Methanol is a particularly suitable solvent for use in embodiments of the present invention. In certain embodiments, solvents that include sulfur, nitrogen, or phosphorous, preferably have a relatively high volatility to ensure adequate stripping of the solvent from the hydrocarbon feedstock.

[0036] In preferred embodiments, the extraction solvent is non-acidic. The use of acids is typically avoided due to the corrosive nature of acids, and the requirement that all equipment be
specifically designed for a corrosive environment. In addition, acids, such as acetic acid, can present difficulties in separation due to the formation of emulsions.

[0037] Extraction vessel 112 can be operated at a temperature of between about 20°C and about 60°C, preferably between about 25°C and about 45°C, even more preferably between about 25°C and about 35°C. Extraction vessel 112 can operate at a pressure of between about 1 and about 10 bars, preferably between about 1 and about 5 bars, more preferably between about 1 and about 2 bars. In certain embodiments, extraction vessel 112 operates at a pressure of between about 2 and about 6 bars.

[0038] The ratio of the extraction solvent to hydrocarbon feedstock can be between about 1:3 and about 3:1, preferably between about 1:2 and about 2:1, more preferably about 1:1. Contact time between the extraction solvent and oxidized hydrocarbon stream 110 can be between about 1 second and about 60 minutes, preferably between about 1 second and about 10 minutes. In certain preferred embodiments, the contact time between the extraction solvent and oxidized hydrocarbon stream 110 is less than about 15 minutes. In certain embodiments, extraction vessel 112 can include various means for increasing the contact time between the extraction solvent and oxidized hydrocarbon stream 110, or for increasing the degree of mixing of the two solvents. Means for mixing can include mechanical stirrers or agitators, trays, or like means.

[0039] The extraction vessel produces mixed stream 114 that can include extraction solvent, oxidized species (e.g., the oxidized sulfur and nitrogen species that were originally present in hydrocarbon feedstock 102), and the hydrocarbon feedstock, and extracted hydrocarbon stream 118, which can include the hydrocarbon feedstock having a reduced sulfur and low nitrogen
content, relative to hydrocarbon feedstock 102. Typically, the hydrocarbon feedstock is only present in mixed stream 114 in trace amounts.

[0040] Mixed stream 114 is supplied to solvent regeneration column 116 where extraction solvent can be recovered as first recovered solvent stream 117 and separated from first residue stream 123, which includes oxidized sulfur and oxidized nitrogen compounds. Optionally, mixed stream 114 can be separated in solvent regeneration column 116 into a recovered hydrocarbon stream 124, which can include hydrocarbons present in mixed stream 114 from hydrocarbon feedstock 102. Solvent regeneration column 116 can be a distillation column that is configured to separate mixed stream 114 into first recovered solvent stream 117, first residue stream 123, and recovered hydrocarbon stream 124.

[0041] Extracted hydrocarbon stream 118 can be supplied to stripper 120, which can be a distillation column or like vessel designed to separate a hydrocarbon product stream from residual extraction solvent. In certain embodiments, a portion of mixed stream 114 can be supplied to stripper 120 via line 122, and may optionally be combined with extracted hydrocarbon stream 118. In certain embodiments, solvent regeneration column 116 can produce recovered hydrocarbon stream 124, which can be supplied to stripper 120. Where the recovered hydrocarbon stream can be contacted with extracted hydrocarbon stream 118 and/or a portion of mixed stream 114, which can be supplied to the stripper via line 122.

[0042] Stripper 120 separates the various streams supplied thereto into stripped oil stream 126, which has a reduced sulfur and nitrogen content relative to hydrocarbon feedstock 102, and second recovered solvent stream 128.
In certain embodiments, first recovered solvent stream 117 can be combined with second recovered solvent stream 128 and recycled to extraction vessel 112. Optionally, make-up solvent stream 132, which can include fresh solvent, can be combined with first recovered solvent stream 117 and/or second recovered solvent stream 128 and supplied to extraction vessel 112.

First residue stream 123, which includes oxidized compounds, such as oxidized sulfur and nitrogen compounds, and can also include some hydrocarbonaceous material, can be supplied to fluid catalytic cracking unit 130 where the hydrocarbons 136 are recovered. In one embodiment of the present invention, oxidized sulfur compounds, such as sulfones, and oxidized nitrogen compounds are embedded in heavy hydrocarbons, such as hydrocarbons having a boiling point in a range of about 343°C to about 524°C; or alternatively, in a range of about 360°C to about 550°C.

In aspects of the present invention in which the first residue stream 123 is sent to the fluid catalytic cracking unit 130, the first residue stream 123 is contacted with a fluid catalytic cracking feedstream 134 in the presence of a catalyst to catalytically crack the fluid catalytic cracking feedstream 134 to recover hydrocarbons 136 from the first residue stream 123. In an aspect, the catalyst can include hot solid zeolitic active catalyst particles. In an aspect, the weight ratio of catalyst to the fluid catalytic cracking feedstream 134 is within a range of between about 1 and about 15 with a pressure ranging from about 1 bar g to about 200 bar g to form a suspension. Other suitable ratios of catalyst and fluid catalytic cracking feedstream 134 and operating conditions will be apparent to those of skill in the art and are to be considered within the scope of the present invention.
The suspension is then passed through a riser reaction zone or downer at a temperature between about 300 °C and less than about 650 °C to catalytically crack the fluid catalytic cracking feedstream 134 while avoiding thermal conversion of said feedstream 134 and providing a hydrocarbon residence time between about 1 second and about 10 minutes.

The lower boiling components and said solid catalyst particles are then separated and recovered. At least a portion of the separated solid catalyst particles is regenerated with a water-free oxygen-containing gas in a fluidized bed operated at conditions to produce regenerated catalyst 140 and gaseous products 138 consisting essentially of carbon monoxide and carbon dioxide. At least a portion of the regenerated catalyst is returned and combined with the fluid catalytic cracking feedstream 134.

The types of components contained in the fluid catalytic cracking feedstream 134 can vary. In an aspect, the fluid catalytic cracking feedstream 134 can include vacuum gas oil, reduced crude, demetalized oil, whole crude, cracked shale oil, liquefied coal, cracked bitumens, heavy coker gas oils, and FCC heavy products such as LCO, HCO and CSO. Table 1 shows the typical yield from a FCC unit. As another example, the fluid catalytic cracking feedstream 134 sent to the FCC unit 130 can have the properties shown in Table 2. Other suitable compounds that can be used in the fluid catalytic cracking feedstream 134 being sent to the FCC unit 130 will be apparent to those of skill in the art and are to be considered within the scope of the present invention.
Various types of catalysts can be used in the FCC unit 130. In an aspect, the FCC catalyst particles comprise a zeolitic matrix with metals selected from Groups IVB, VI, VII, VIIIB, IB, IIB or a compound thereof and with catalyst particles less than 200 microns in nominal diameter. Other suitable types of catalysts that can be used in the FCC unit 130 will be apparent to those of skill in the art and are to be considered within the scope of the present invention.

The operating parameters for the FCC unit 130 can be varied depending upon the type of fluid catalytic cracking feedstream 134 that is sent to the FCC unit 130. In an aspect, the FCC

<table>
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<th>Table 1</th>
<th>Products</th>
<th>Yields</th>
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<td>Evaporation Point (EP)</td>
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[0049] Various types of catalysts can be used in the FCC unit 130. In an aspect, the FCC catalyst particles comprise a zeolitic matrix with metals selected from Groups IVB, VI, VII, VIIIB, IB, IIB or a compound thereof and with catalyst particles less than 200 microns in nominal diameter. Other suitable types of catalysts that can be used in the FCC unit 130 will be apparent to those of skill in the art and are to be considered within the scope of the present invention.

[0050] The operating parameters for the FCC unit 130 can be varied depending upon the type of fluid catalytic cracking feedstream 134 that is sent to the FCC unit 130. In an aspect, the FCC
unit 130 is conducted in the temperature range of about 400°C to about 850 °C. In another aspect, the FCC unit 130 can be operated at a pressure ranging from about 1 bar g to about 200 bar g. In another aspect, the FCC unit 130 can be operated for a residence time ranging from about 1 second to about 3600 seconds. Other suitable operating parameters for the FCC unit 130 will be apparent to those of skill in the art and are to be considered within the scope of the present invention.

[0051] The properties of the components recovered from the FCC unit 130 will vary depending upon the composition of the hydrocarbon fluid catalytic cracking feedstream 134.

[0052] Figure 2 provides one embodiment of the present invention for the recovery of hydrocarbons from a feedstream. Hydrocarbon recovery system 100 includes oxidation reactor 104, extraction vessel 112, solvent regeneration column 116, stripper 120, fluid catalytic cracking unit 130, and adsorption column 202.

[0053] As shown in Figure 2, in certain embodiments of the invention, stripped oil stream 126 can be supplied to adsorption column 202, where the stream can be contacted with one or more adsorbent designed to remove one or more of various impurities, such as sulfur containing compounds, oxidized sulfur compounds, nitrogen containing compounds, oxidized nitrogen compounds, and metals remaining in the hydrocarbon product stream after oxidation and solvent extraction steps.

[0054] Exemplary adsorbents can include activated carbon; silica gel; alumina; natural clays; silica-alumina; zeolites; fresh, used, regenerated or rejuvenated catalysts having affinity to oxidized sulfur and nitrogen compounds and other inorganic adsorbents. In certain preferred embodiments, the adsorbent can include polar polymers that have been applied to or that coat
various high surface area support materials, such as silica gel, alumina, and activated carbon. Exemplary polar polymers for use in coating various support materials can include polysulfones, polyacrylonitrile, polystyrene, polyester terephthalate, polyurethane, other like polymer species that exhibit an affinity for oxidized sulfur species, and combinations thereof.

[0055] The adsorption column can be operated at a temperature of between about 20°C and about 60°C, preferably between about 25°C and about 40°C, even more preferably between about 25°C and about 35°C. In certain embodiments, the adsorption column can be operated at a temperature of between about 10°C and about 40°C. In certain embodiments, the adsorption column can be operated at temperatures of greater than about 20°C, or alternatively at temperatures less than about 60°C. The adsorption column can be operated at a pressure of up to about 15 bars, preferably up to about 10 bars, even more preferably between about 1 and about 2 bars. In certain embodiments, the adsorption column can be operated at a pressure of between about 2 and about 5 bar. In an exemplary embodiment, the adsorption column can be operated at a temperature of between about 25°C and about 35°C and a pressure of between about 1 and about 2 bars. The weight ratio of the stripped oil stream to the adsorbent is between about 1:1 to about 20:1; or alternatively, about 10:1.

[0056] Adsorption column 202 separates the feed into extracted hydrocarbon product stream 204 having very low sulfur and very low nitrogen content and second residue stream 206. Second residue stream 206 includes oxidized sulfur and oxidized nitrogen compounds, and can be combined with first residue stream 123 and supplied to FCC Unit 130 and processed as noted above. The adsorbent can be regenerated. Use of a polar solvent for removal of adsorbed molecules such as methanol or acetonitrile can be used to desorb the adsorbed oxidized compounds from the adsorbent. Heat and gas stripping can also be used to remove the adsorbed
compounds from the adsorbent. Other suitable methods for removing the absorbed compounds will be apparent to those of skill in the art and are to be considered within the scope of the present invention.

EXAMPLE

[0057] Figure 3 shows the process flow diagram for the oxidative desulfurization (oxidation and extraction steps) and FCC Unit. The vessels 10, 16, 20 and 24 are oxidation, extraction, solvent recovery and fluid catalytic cracking vessels, respectively.

[0058] A hydrotreated straight run diesel containing 500 ppmw of elemental sulfur, 0.28 wt. % of organic sulfur, density of 0.85 Kg/l was oxidatively desulfurized. The reaction conditions were as follows:

- Hydrogen peroxide: sulfur mol ratio: 4:1
- Catalyst: Molybdenum based Mo(VT)
- Reaction time: 30 minutes
- Temperature: 80 °C
- Pressure: 1 Kg/cm²

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<th>Oxidation</th>
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<th>12</th>
<th>13</th>
<th>14</th>
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<td>Component\stream</td>
<td>Diesel</td>
<td>H₂O₂</td>
<td>Catalyst</td>
<td>Catalyst Waste</td>
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<td></td>
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<td>Acetic Acid</td>
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<td>H₂O₂</td>
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<td>Na₂WO₄ (kg)</td>
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<td>Total Kg/h</td>
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<td>15,435</td>
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The fluid catalytic unit was operated at 518 °C with a catalyst to oil ratio of 5, which resulted in 67 wt. % conversion of the feedstock. In addition to the sulfones produced in the oxidative step, straight run vacuum gas oil derived from Arabian crude oils was used as a blending component. The feedstock contained 2.65 wt. % sulfur and 0.13 wt. % of micro carbon residue. The mid and 95 wt. % boiling points for the feedstocks were 408 °C and 455 °C, respectively.

The FCC conversion of the feedstock was calculated as:

\[
\text{Conversion} = \text{Dry Gas} + \text{LPG} + \text{Gasoline} + \text{Coke}
\]

The catalyst used was an equilibrium catalyst and used as is without any treatment. The catalyst has 131 m²/g surface area and 0.1878 cm³/g pore volume. The nickel and vanadium contents are 96 and 407 ppmw, respectively. The FCC process yielded the following products and deposited coke on the catalysts.
Dry gas  \[ \text{CH}_2, \text{C}_2\text{H}_6, \text{C}_2\text{H}_4 \]
Wet gas  \[ \text{C}_3, \text{C}_4 \text{ compounds (LPG)} \]
Gasoline  Liquid product containing \( \text{C}_5 \text{ to C}_1 \) hydrocarbons; typical end boiling point 221 °C
LCO  Light cycle oil containing \( \text{C}_{15}-\text{C}_{20} \) hydrocarbons; typical boiling point 221-343°C
HCO  Heavy cycle oil containing \( \text{C}_{20}+ \) hydrocarbons with a minimum boiling point of 343°C
Coke  Solid carbonaceous deposit on the catalyst; typical C-H ratio = 1

[0062] The coke produced in the FCC process was 2.5 wt. % of the feedstock processed. The product yields are given below:

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<th>Vacuum Gas Oil</th>
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<td>2.67</td>
<td>2.5</td>
<td>0.27</td>
<td>2.72</td>
<td>4.82</td>
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<td>Vacuum Gas Oil</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<tr>
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<td></td>
<td>1822</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gasoline</td>
<td></td>
<td></td>
<td></td>
<td>4957</td>
<td>1707</td>
<td>1764</td>
<td></td>
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<tr>
<td>LCO</td>
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<tr>
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<td></td>
<td></td>
<td>1764</td>
</tr>
<tr>
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<td>10000</td>
<td>10507</td>
<td>1822</td>
<td>4957</td>
<td>1707</td>
<td>1764</td>
</tr>
</tbody>
</table>

[0063] It is believed that the methods and systems described herein will increase the amount of liquid hydrocarbons from aromatic sulfur, nitrogen compounds, and aromatic streams by linking an oxidative desulfurization and denitrogenation process with a fluid catalytic cracking unit. Furthermore, it is believed that there are not any efficient methods for disposing of the oxidation reaction byproducts, i.e., the oxidized sulfur and nitrogen compounds. Embodiments
of the present invention provide a way of disposing of the oxidized sulfur and nitrogen compounds without having to dispose of the compounds.

[0064] Although the present invention has been described in detail, it should be understood that various changes, substitutions, and alterations can be made hereupon without departing from the principle and scope of the invention. Accordingly, the scope of the present invention should be determined by the following claims and their appropriate legal equivalents.

[0065] The singular forms "a", "an" and "the" include plural referents, unless the context clearly dictates otherwise.

[0066] Optional or optionally means that the subsequently described event or circumstances may or may not occur. The description includes instances where the event or circumstance occurs and instances where it does not occur.

[0067] Ranges may be expressed herein as from about one particular value, and/or to about another particular value. When such a range is expressed, it is to be understood that another embodiment is from the one particular value and/or to the other particular value, along with all combinations within said range.

[0068] Throughout this application, where patents or publications are referenced, the disclosures of these references in their entireties are intended to be incorporated by reference into this application, in order to more fully describe the state of the art to which the invention pertains, except when these reference contradict the statements made herein.
That which is claimed is:

1. A method of recovering components from a hydrocarbon feedstock, the method comprising the steps of:

   supplying the hydrocarbon feedstock to an oxidation reactor, the hydrocarbon feedstock comprising sulfur compounds and nitrogen compounds;

   contacting the hydrocarbon feedstock with an oxidizing agent in the oxidation reactor under conditions sufficient to selectively oxidize sulfur compounds and nitrogen compounds present in the hydrocarbon feedstock to produce an oxidized hydrocarbon stream that comprises hydrocarbons, oxidized sulfur compounds, and oxidized nitrogen compounds;

   separating the hydrocarbons, the oxidized sulfur compounds, and the oxidized nitrogen compounds in the oxidized hydrocarbon stream by solvent extraction with a polar solvent to produce an extracted hydrocarbon stream and a mixed stream, the mixed stream comprising the polar solvent, the oxidized sulfur compounds, and the oxidized nitrogen compounds, wherein the extracted hydrocarbon stream has a lower concentration of sulfur compounds and nitrogen compounds than the hydrocarbon feedstock;

   separating the mixed stream into a first recovered polar solvent stream and a first residue stream, the first residue stream comprising the oxidized sulfur compounds and the oxidized nitrogen compounds; and

   supplying the first residue stream to a fluid catalytic cracking unit, the fluid catalytic cracking unit being operative to catalytically crack the oxidized sulfur and the oxidized nitrogen and allow for recovery of hydrocarbons from the first residue stream.
2. The method of claim 1, wherein the oxidant is selected from the group consisting of air, oxygen, peroxides, hydroperoxides, organic peracids, ozone, nitrogen oxides compounds, and combinations thereof.

3. The method of any of claims 1 or 2, wherein the step of contacting the hydrocarbon feedstock with an oxidizing agent occurs in the presence of a catalyst comprising a metal oxide having the formula \( \text{M}_x\text{O}_y \), wherein M is an element selected from Groups IVB, VB, and VIB of the periodic table.

4. The method of any of claims 1 - 3, wherein the sulfur compounds comprise sulfides, disulfides, mercaptans, thiophene, benzothiophene, dibenzothiophene, alkyl derivatives of dibenzothiophene, or combinations thereof.

5. The method of any of claims 1 - 4, wherein the oxidation reactor is maintained at a temperature of between about 20 and about 350°C and at a pressure of between about 1 and about 10 bars.

6. The method of any of claims 1 - 5, wherein the ratio of the oxidant to sulfur compounds present in the hydrocarbon feedstock is between about 4:1 and about 10:1.

7. The method of any of claims 1 - 6, wherein the polar solvent has a Hildebrandt value of greater than about 19.

8. The method of any of claims 1 - 7, wherein the polar solvent is selected from the group consisting of acetone, carbon disulfide, pyridine, dimethyl sulfoxide, n-propanol, ethanol, n-butanol, propylene glycol, ethylene glycol, dimethylformamide, acetonitrile, methanol and combinations thereof.

9. The method of any of claims 1 - 7, wherein the polar solvent is acetonitrile.
10. The method of any of claims 1-7, wherein the polar solvent is methanol.

11. The method of any of claims 1-10, wherein the solvent extraction is conducted at a temperature of between about 20°C and about 60°C and at a pressure of between about 1 and about 10 bars.

12. The method of any of claims 1-11, further comprising the step of supplying the extracted hydrocarbon stream to a stripper to produce a second recovered polar solvent stream and a stripped hydrocarbon stream.

13. The method of any of claims 1-12, further comprising the step of supplying the extracted hydrocarbon stream to an adsorption column, the adsorption column being charged with an adsorbent suitable for the removal of oxidized compounds present in the extracted hydrocarbon stream, the adsorption column producing a high purity hydrocarbon product stream and a second residue stream, the second residue stream including a portion of the oxidized compounds

14. The method of claim 13, further comprising supplying the second residue stream to the fluid catalytic cracking unit.

15. The method of any of claims 1-14, wherein the adsorbent is selected from the group consisting of activated carbon; silica gel; alumina; natural clays; silica-alumina; zeolites; fresh, used; and combinations of the same.

16. The method of any of claims 1-14, wherein the adsorbent is a polymer coated support, wherein the support has a high surface area and is selected from the group consisting of silica gel; alumina; silica-alumina; zeolites; fresh, used; and activated carbon, and the polymer is
selected from the group consisting of polysulfone, polyacrylonitrile, polystyrene, polyester terephthalate, polyurethane, and combinations of the same.

17. The method of any of claims 1 - 16, wherein the step of supplying the first residue stream to the fluid catalytic cracking unit further comprises contacting the first residue stream with a fluid catalytic cracking feedstream in the presence of a catalyst to catalytically crack the fluid catalytic cracking feedstream to recover hydrocarbons from the first residue stream.

18. The method of claim 17, wherein the fluid catalytic cracking feedstream comprises vacuum gas oil, reduced crude, demetalized oil, whole crude, cracked shale oil, liquefied coal, cracked bitumen, heavy coker gas oils, light cycle oil (LCO), heavy cycle oil (HCO), clarified slurry oil (CSO), or combinations thereof.

19. A method of recovering components from a hydrocarbon feedstock, the method comprising the steps of:

   supplying the hydrocarbon feedstock to an oxidation reactor, the hydrocarbon feedstock comprising sulfur compounds and nitrogen compounds;

   contacting the hydrocarbon feedstock with an oxidizing agent in the oxidation reactor under conditions sufficient to selectively oxidize sulfur compounds and nitrogen compounds present in the hydrocarbon feedstock to produce an oxidized hydrocarbon stream that comprises hydrocarbons, oxidized sulfur compounds, and oxidized nitrogen compounds;

   separating the hydrocarbons, the oxidized sulfur compounds, and the oxidized nitrogen compounds in the oxidized hydrocarbon stream by solvent extraction with a polar solvent to produce an extracted hydrocarbon stream and a mixed stream, the mixed stream comprising the polar solvent, the oxidized sulfur compounds, and the oxidized nitrogen compounds, wherein the
extracted hydrocarbon stream has a lower concentration of sulfur compounds and nitrogen compounds than the hydrocarbon feedstock;

separating the mixed stream into a first recovered polar solvent stream and a first residue stream, the first residue stream comprising the oxidized sulfur compounds and the oxidized nitrogen compounds;

supplying the first residue stream to a fluid catalytic cracking unit, the fluid catalytic cracking unit being operative to catalytically crack the oxidized sulfur and the oxidized nitrogen and allow for recovery of hydrocarbons from the first residue stream;

contacting the first residue stream with a fluid catalytic cracking feedstream in the presence of a catalyst to catalytically crack the fluid catalytic cracking feedstream to recover hydrocarbons from the first residue stream.

20. The method of claim 19, wherein the oxidant is selected from the group consisting of air, oxygen, peroxides, hydroperoxides, organic peracids, ozone, nitrogen oxides compounds, and combinations thereof.

21. The method of any of claims 19 - 20, wherein the step of contacting the hydrocarbon feedstock with an oxidizing agent occurs in the presence of a catalyst comprising a metal oxide having the formula \( \text{M}_x\text{O}_y \), wherein \( \text{M} \) is an element selected from Groups IVB, VB, and VIB of the periodic table.

22. The method of any of claims 19 - 21, wherein the sulfur compounds comprise sulfides, disulfides, mercaptans, thiophene, benzothiophene, dibenzothiophene, alkyl derivatives of dibenzothiophene, or combinations thereof.
23. The method of any of claims 19 - 22, wherein the oxidation reactor is maintained at a temperature of between about 20 and about 350°C and at a pressure of between about 1 and about 10 bars.

24. The method of any of claims 19 - 23, wherein the ratio of the oxidant to sulfur compounds present in the hydrocarbon feedstock is between about 4:1 and about 10:1.

25. The method of any of claims 19 - 24, wherein the polar solvent has a Hildebrandt value of greater than about 19.

26. The method of any of claims 19 - 25, wherein the polar solvent is selected from the group consisting of acetone, carbon disulfide, pyridine, dimethyl sulfoxide, n-propanol, ethanol, n-butanol, propylene glycol, ethylene glycol, dimethylformamide, acetonitrile, methanol and combinations thereof.

27. The method of any of claims 19 - 25, wherein the polar solvent is acetonitrile.

28. The method of any of claims 19 - 25, wherein the polar solvent is methanol.

29. The method of any of claims 19 - 28, wherein the solvent extraction is conducted at a temperature of between about 20°C and about 60°C and at a pressure of between about 1 bar and about 10 bars.

30. The method of any of claims 19 - 29, further comprising the step of supplying the extracted hydrocarbon stream to a stripper to produce a second recovered polar solvent stream and a stripped hydrocarbon stream.

31. The method of any of claims 19 - 30, further comprising the step of supplying the extracted hydrocarbon stream to an adsorption column, the adsorption column being charged with an adsorbent suitable for the removal of oxidized compounds present in the extracted
hydrocarbon stream, the absorption column producing a high purity hydrocarbon product stream and a second residue stream, the second residue stream including a portion of the oxidized compounds.

32. The method of claim 31, further comprising supplying the second residue stream to the fluid catalytic cracking unit.

33. The method of any of claims 31 or 32, wherein the adsorbent is selected from the group consisting of activated carbon, silica gel, alumina, natural clays, silica-alumina, zeolites, and combinations of the same.

34. The method of any of claims 31 - 33, wherein the adsorbent is a polymer coated support, wherein the support has a high surface area and is selected from the group consisting of silica gel, alumina, and activated carbon, and the polymer is selected from the group consisting of polysulfone, polyacrylonitrile, polystyrene, polyester terephthalate, polyurethane, silica-alumina, zeolites, and combinations of the same.

35. The method of any of claims 19 - 34, wherein the first residue stream and the fluid catalytic cracking feedstream are present in a weight ratio of the catalyst to the first residue stream and the fluid catalytic cracking feedstream ranges from about 1 to about 15.

36. The method of any of claims 19 - 35, wherein the fluid catalytic cracking feedstream comprises vacuum gas oil, reduced crude, demetalized oil, whole crude, cracked shale oil, liquefied coal, cracked bitumen, heavy coker gas oils, light cycle oil (LCO), heavy cycle oil (HCO), clarified slurry oil (CSO), or combinations thereof.
37. The method of any of claims 19 - 36, wherein the step of contacting the first residue stream with a fluid catalytic cracking feedstream in the presence of a catalyst occurs in a temperature range of about 300°C to about 650°C.

38. The method of any of claims 19 - 37, wherein the step of contacting the first residue stream with a fluid catalytic cracking feedstream in the presence of a catalyst occurs in a residence time of about 1 second to about 10 minutes.

39. The method of any of claims 19 - 38, further comprising the steps of:

   a. separating lower boiling components and catalyst particles from the first residue stream and the fluid catalytic cracking feedstream; and

   b. regenerating at least a portion of the catalyst particles.

40. The method of claim 39, wherein the step of regenerating at least a portion of the catalyst particles includes contacting the portion of the catalyst particles with a water-free oxygen-containing gas in a fluidized bed operated at conditions to produce regenerated catalyst and gaseous products comprising carbon monoxide and carbon dioxide.

41. The method of any of claims 39 or 40, further comprising the step of adding at least a portion of the regenerated catalyst to the fluid catalytic cracking feedstream.
## A. CLASSIFICATION OF SUBJECT MATTER

**INV. Cl0Gll/00**

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
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<td>SEE FIGURES 1-3; SEE TABLES 1-5 SEE &quot;BYPRODUCTS&quot; AT PAGE 4.</td>
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<td>Y</td>
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Further documents are listed in the continuation of Box C. See patent family annex.

* Special categories of cited documents:
- **A**: document defining the general state of the art which is not considered to be of particular relevance
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- **O**: document referring to an oral disclosure, use, exhibition or other means
- **P**: document published prior to the international filing date but later than the priority date claimed

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- **X**: document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- **Y**: document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- **S**: member of the same patent family

## Date of the actual completion of the international search

10 November 2011

## Date of mailing of the international search report

22/11/2011

Name and mailing address of the ISA/Authorized officer

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016
Rumbo, Angel
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