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(54) Title: PROCESS FOR THE CONVERSION OF BIOMASS-DERIVED VAPOURS AND PROCESS FOR THE CONVERSION
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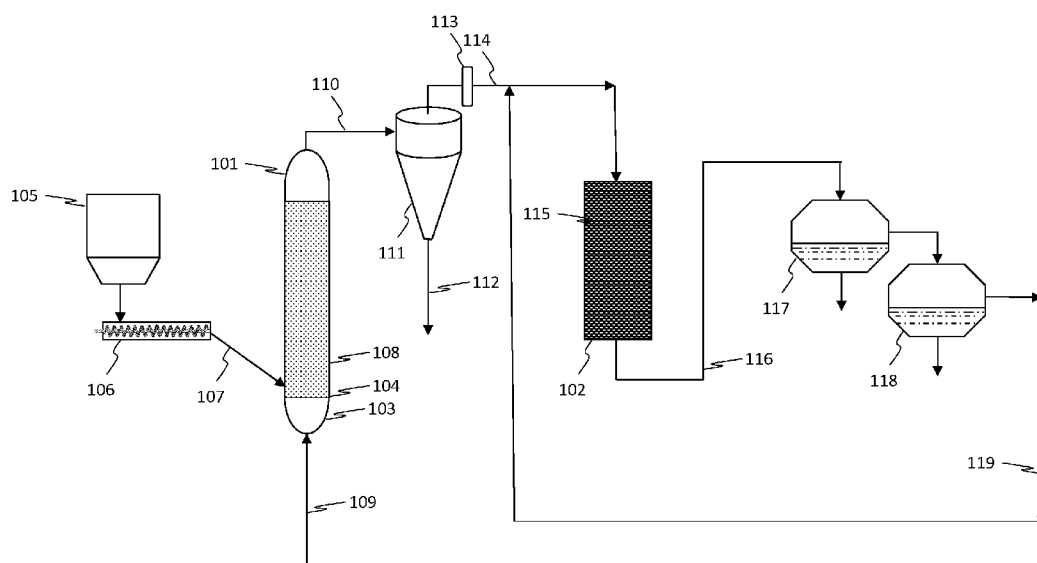


Figure 1

(57) Abstract: This invention relates to a process for the conversion of biomass-derived vapours, comprising: - a catalytic step, wherein at least part of the biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C, thereby producing a product mixture; and - a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction; wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours. The present invention also relates a process for the conversion of a solid biomass material, comprising: - a pyrolysis step, wherein at least part of the solid biomass material is pyrolysed, thereby producing biomass-derived vapours; - a catalytic step, wherein at least part of the

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biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C, thereby producing a product mixture; and - a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction; wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours.

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PROCESS FOR THE CONVERSION OF BIOMASS-DERIVED VAPOURS AND
PROCESS FOR THE CONVERSION OF A SOLID BIOMASS MATERIAL.

Field of the Invention

This invention relates to a process for the conversion of biomass-derived vapours and a process for the conversion of a solid biomass material.

Background of the invention

With the diminishing supply of fossil fuel resources, the use of sustainable and renewable biomass as an energy source is becoming increasingly important for the production of fuels and/or chemicals. The use of a biomass material as an energy source may also allow for a more sustainable production of fuels and/or chemicals and more sustainable CO₂ emissions that may help meet global CO₂ emissions standards under the Kyoto protocol. The fuels and/or chemicals derived from biomass materials are often referred to as biofuels and/or biochemicals.

Fuels and/or chemicals derived from non-edible biomass materials, such as cellulosic materials, are preferred as these do not compete with food production. Most of these non-edible biomass materials, however, are solid biomass materials that are cumbersome to convert.

In the past decades multiple processes have been developed to convert solid biomass materials into vapours and/or liquids. Examples of processes to convert solid biomass materials into vapours and/or liquids include pyrolysis processes, including so-called catalytic fast pyrolysis processes. With the help of such processes biomass-derived vapours and/or liquids can be obtained. These vapours and/or liquids can subsequently be converted further to produce hydrocarbons that can be

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used as a fuel and/or chemical component or as a precursor for such fuel and/or chemical component.

For example, WO2013/103872 describes a two-stage reactor/process for the conversion of solid particulate biomass material and includes a first stage in which solid particulate biomass material is pyrolyzed to primary reaction products, and a second stage in which the primary reaction products are catalytically converted. In the process as described in WO2013/103872 the second stage is operated at a temperature higher than that of the first stage.

In figure 1 of WO2013/103872, a two-stage reactor is shown, where biomass material, a lift gas and regenerated catalyst particles are entering the first stage of a two-stage reactor. After reaction in the two stages a gas/vapour/solids mixture leaves overhead from the second stage and is conveyed to a cyclone, where it is split into a gas/vapour stream and a solids stream. The vapour portion of the gas/vapour stream is condensed in a fractionator, forming a liquid bio-oil and a gas stream comprising non-condensables. It is indicated that this gas-stream can be removed and recycled to first stage of the reactor as at least a part of the lift gas.

During catalytic fast pyrolysis processes such as the one described in WO2013/103872 a considerable amount of carbon monoxide, carbon dioxide and other gasses are produced. Such gasses are produced at the expense of the production of hydrocarbons that could be useful as fuel and/or chemical components or as a precursor therefore.

It would be an advancement in the art to provide a process for the conversion of a solid biomass material and/or biomass-derived vapours, which would allow for a

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reduction in gas production and an improved hydrocarbon yield.

SUMMARY OF THE INVENTION

Such advancement has been achieved with the process according to the invention. Accordingly, in a first embodiment, the present invention provides a process for the conversion of biomass-derived vapours, comprising:

- a catalytic step, wherein at least part of the biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C, thereby producing a product mixture; and

- a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction;

wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours.

Preferably the biomass-derived vapours are derived from a solid biomass material and more preferably the biomass-derived vapours are pyrolysis vapours obtained or obtainable by pyrolysis of a solid biomass material.

Accordingly, in a second embodiment, the present invention also provides a process for the conversion of a solid biomass material, comprising:

- a pyrolysis step, wherein at least part of the solid biomass material is pyrolysed, thereby producing biomass-derived vapours;

- a catalytic step, wherein at least part of the biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C

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to equal to or less than 550°C, thereby producing a product mixture; and
- a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction;
wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours.

As illustrated in the examples, recycling at least part of the gaseous product fraction to the catalytic step instead or in addition to recycling such gaseous product to a pyrolysis step advantageously allows one to reduce gas-production and improve hydrocarbon yield.

Further, without wishing to be bound to any kind of theory it is believed that recycling at least part of the gaseous product fraction to the catalytic step can improve the catalyst lifetime of the catalyst used in this catalytic step.

BRIEF DESCRIPTION OF THE FIGURES

Figure 1 schematically illustrates an experimental set-up for a process according to the invention.

DETAILED DESCRIPTION OF THE INVENTION

By the word "comprising" respectively "comprise" is herein understood "including" respectively "include".

By biomass-derived vapours are herein understood vapours derived from a biomass material.

By a biomass material is herein understood a composition of matter of biological origin as opposed to a composition of matter obtained or derived from petroleum, natural gas or coal. Without wishing to be bound by any kind of theory it is believed that such biomass material may contain carbon-14 isotope in an

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abundance of about 0.0000000001 %, based on total moles of carbon.

The biomass material may suitably comprise animal fat, tallow and/or a solid biomass material. Preferably the biomass material is a solid biomass material.

Preferably such a solid biomass material contains cellulose and/or lignocellulose. Such a material containing cellulose respectively lignocellulose is herein also referred to as a "cellulosic", respectively "lignocellulosic" material. By a cellulosic material is herein understood a material containing cellulose and optionally also lignin and/or hemicellulose. By a lignocellulosic material is herein understood a material containing cellulose and lignin and optionally hemicellulose.

Preferably the solid biomass material is a lignocellulosic material comprising equal to or more than 20 weight percent (wt.%) lignin, more preferably equal to or more than 30 wt.% lignin, even more preferably equal to or more than 40 wt.% lignin, still more preferably equal to or more than 50wt.% lignin and possibly equal to or more than 75wt.% lignin, based on the total weight of lignocellulosic material. For practical purposes, the lignocellulosic material may contain equal to or less than 99 wt.% lignin, based on the total weight of lignocellulosic material. For example, willow wood may contain about 25 wt.% lignin, larch wood may contain about 35 wt.% lignin, straw may contain about 14 wt.% lignin, beech wood may contain about 12-23 wt.% lignin and coniferous wood may contain about 25-35 wt.% lignin, based on the total weight of lignocellulosic material.

It is also possible for the solid biomass material to be a lignin-containing material containing equal to or

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more than 90wt.% lignin and equal to or less than 100wt.% lignin, based on the total weight of lignin-containing material. More preferably such lignin-comprising material consists essentially of lignin (i.e. comprises essentially 100wt.% lignin based on the total weight of the material). Examples of a lignin-containing material with such high lignin contents include for example so-called organosolv lignin.

Preferably the solid biomass material is not a material used for food production.

Examples of suitable solid biomass materials include agricultural wastes such as corn stover, soybean stover, corn cobs, rice straw, rice hulls, oat hulls, corn fibre, cereal straws such as wheat, barley, rye and oat straw; grasses; forestry products and/or forestry residues such as wood and wood-related materials such as sawdust; waste paper; sugar processing residues such as bagasse and beet pulp; or mixtures thereof. More preferably the solid biomass material is selected from the group consisting of wood, sawdust, straw, grass, bagasse, corn stover and/or mixtures thereof. Preferred solid biomass materials also include non-wood lignocellulosic biomass materials such as grass-derived lignocellulosic biomass material. Typical grasses include wheat straw, but also miscanthus, sweet sorghum and bamboo.

A solid biomass material can suitably be first converted into a biomass-derived liquid and subsequently such biomass-derived liquid can be at least partly vapourized to produce biomass-derived vapours. It is also possible for a solid biomass material to be directly converted into biomass-derived vapours.

Preferably a solid biomass material is pyrolyzed into biomass-derived vapours. The biomass-derived vapours

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are preferably pyrolysis vapours obtained or obtainable by pyrolysis of a solid biomass material. Such pyrolysis vapours obtained or obtainable by pyrolysis of a solid biomass material are herein also referred to as biomass-derived pyrolysis vapours.

In a pyrolysis process a solid biomass material can conveniently be directly converted into biomass-derived vapours. In such a pyrolysis process solid biomass material may conveniently be partly or wholly converted, and is preferably wholly converted.

As indicated herein above, the present invention also provides a process for the conversion of a solid biomass material, comprising:

- a pyrolysis step, wherein at least part of the solid biomass material is pyrolysed, thereby producing biomass-derived vapours;
- a catalytic step, wherein at least part of the biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C, thereby producing a product mixture; and
- a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction;

wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours.

The solid biomass material can optionally be washed, steam exploded, dried, roasted, torrefied and/or reduced in particle size before being pyrolyzed or being converted via another process. In addition, if the solid biomass material is a cellulosic or lignocellulosic

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material it can be demineralized before being pyrolyzed or being converted via another process.

The solid biomass material preferably has a moisture content in the range from equal to or more than 1 weight percent (wt.%) to equal to or less than 20 wt.%. Any optional drying of the solid biomass material can be carried out by any method known by the person skilled in the art to be suitable for drying of a biomass material. If dried, the solid biomass material is preferably dried at a temperature in the range from equal to or more than 100°C to equal to or less than 200°C. The solid biomass material after drying may suitably have a water-content of equal to or less than 20 wt.%, more suitably of equal to or less than 15 wt.% and most suitably of equal to or less than 10 wt.%.

The solid biomass material can optionally be a torrefied solid biomass material. By torrefying or torrefaction is herein understood the treatment of the solid biomass material at a temperature in the range from equal to or more than 200°C to equal to or less than 350°C in the essential absence of a catalyst and in an oxygen-poor, preferably an oxygen-free, atmosphere.

If torrefied, the solid biomass material is preferably torrefied at a temperature of more than 200°C, more preferably at a temperature equal to or more than 210°C, still more preferably at a temperature equal to or more than 220°C, yet more preferably at a temperature equal to or more than 230°C. In addition torrefying of the solid biomass material is preferably carried out at a temperature less than 350°C, more preferably at a temperature equal to or less than 330°C, still more preferably at a temperature equal to or less than 310°C, yet more preferably at a temperature equal to or less

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than 300°C. Preferably torrefaction of the solid biomass material is carried out at atmospheric pressure (about 1 bar absolute, corresponding to about 0.1 MegaPascal).

Before being pyrolyzed or being converted via another process, the solid biomass material is preferably comminuted into small particles. Preferably, the solid biomass material consists of solid biomass material particles having a particle size distribution with a mean particle size in the range from equal to or more than 10 micrometer, more preferably equal to or more than 50 micrometer, more preferably equal to or more than 100 micrometer, even more preferably equal to or more than 200 micrometer; to equal to or less than 3000 micrometer, more preferably equal to or less than 1000 micrometer, even more preferably equal to or less than 500 micrometer and most preferably equal to or less than 300 micrometer.

By pyrolysis or pyrolyzing is herein understood the decomposition of the solid biomass material, in the presence or in the essential absence of a solid catalyst, effected by heating the solid biomass material at a temperature of equal to or more than 380°C. Preferably pyrolysis is carried out in an oxygen-poor, preferably an oxygen-free, atmosphere. By an oxygen-poor atmosphere is understood an atmosphere containing equal to or less than 10 volume percent (vol.%) oxygen, preferably equal to or less than 5 vol. % oxygen and more preferably equal to or less than 1 vol.% oxygen. By an oxygen-free atmosphere is understood an atmosphere where oxygen is essentially absent.

The solid biomass material is preferably pyrolyzed by heating the solid biomass material to a temperature of equal to or more than 400°C, more preferably equal to or more than 450°C, even more preferably equal to or more

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than 500°C and most preferably equal to or more than 525°C, and preferably equal to or less than 700°C, more preferably equal to or less than 600°C and most preferably equal to or less than 575°C.

More preferably the solid biomass material is pyrolyzed at a temperature in the range from equal to or more than 400°C to equal to or less than 600°C. Most preferably the temperature in any pyrolysis step is kept higher than the temperature in the catalytic step.

The pressure at which the solid biomass material is pyrolyzed can vary widely. Preferably this pressure lies in the range from equal to or more than 0.5 bar absolute (corresponding to 0.05 MegaPascal) to equal to or less than 10 bar absolute (corresponding to 1 MegaPascal), more preferably from equal to or more than 1.0 bar absolute (corresponding to 0.1 MegaPascal) to equal to or less than 3 bar absolute (corresponding to 0.3 MegaPascal), and most preferably atmospheric pressure (i.e. about 1.0 bar absolute corresponding to about 0.1 MegaPascal).

The pyrolysis step can be carried out in the essential absence of an externally added catalyst or the pyrolysis step can be carried out in the presence of one or more catalyst(s). Any catalyst(s) used in the pyrolysis step can be the same or different from any catalyst(s) used in the catalytic step. If the solid biomass material is contacted with a catalyst in the pyrolysis step, such catalyst is preferably a solid catalyst, sometimes also referred to as a heterogeneous catalyst. If present, the catalyst(s) in the pyrolysis step can comprise any catalyst(s) known to the skilled person to be suitable for use in a catalytic fast pyrolysis process. Preferably, any catalyst in the pyrolysis step contains a

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zeolite. Examples of suitable zeolites include mesoporous zeolites. By a mesoporous zeolite is herein preferably understood a zeolite containing pores with a pore diameter in the range from 2-50 nanometer, in line with IUPAC notation (see for example Rouquerol et al. (1994) "Recommendations for the characterization of porous solids (Technical Report)" Pure & Appl. Chem 66 (8) : 1739-1758).

If present, any catalyst in the pyrolysis step is most preferably a MFI or MWW type of zeolite. Especially preferred catalysts include ZSM-5 type zeolites, such as for example Zeolyst 5524G and 8014 and Albemarle UPV-2. The zeolites are preferably dispersed in an amorphous matrix component. For example the catalyst may contain amorphous silica alumina and one or more zeolites. In addition, the catalyst preferably contains a binder and/or a filler.

The pyrolysis step can further be carried out in the presence or absence of a solid heating medium, such as silica or sand.

The pyrolysis step can conveniently be carried out in a reactor. Such a reactor can be any kind of reactor known to the skilled person to be suitable for use in a pyrolysis process, such as for example a fixed bed reactor, fluidized bed reactor, entrained bed reactor or a cyclonic reactor. More preferably the solid biomass material is pyrolyzed in a fluidizing reactor.

By a fluidizing reactor is herein preferably understood a reactor wherein the solid biomass material is fluidized by introducing a carrier gas into such fluidizing reactor. The fluidizing reactor can for example be a riser reactor or a fluidized bed reactor, such as a bubbling fluidized bed reactor. In a riser

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reactor the carrier gas is preferably introduced at a location upstream of the location where the solid biomass material is introduced.

The solid biomass material can be introduced into a fluidizing reactor by any suitable type of feeding system known in the art, for example with the help of one or more hoppers and/or one or more screw feeders.

The solid biomass material and any potential solid catalyst and/or solid heating medium can suitably be fluidized within any fluidizing reactor by introducing a suitable carrier gas to such fluidizing reactor.

For example the carrier gas can include carbon monoxide, carbon dioxide, nitrogen, steam, any C_1 - C_4 hydrocarbon (such as a C_2 - C_4 olefin or C_1 - C_4 alkane) or a mixture of any of these.

By a C_x - C_y hydrocarbon (or an olefin or alkane, respectively) is herein understood a hydrocarbon (or an olefin or alkane, respectively) containing from equal to or more than x carbon atoms to equal to or less than y carbon atoms.

Preferably the C_1 - C_4 hydrocarbon is chosen from the group consisting of methane, ethane, propane, butane, ethene, propene and butene.

Preferably any C_1 - C_4 hydrocarbon carrier gas is introduced into any fluidizing reactor in an amount from equal to or more than 5wt.% to equal to or less than 50wt.%, more preferably in an amount from equal to or more than 10wt.% to equal to or less than 40wt.%, still more preferably in an amount from equal to or more than 15wt.% to equal to or less than 35wt.%, and most preferably in an amount from equal to or more than 15wt.% to equal to or less than 30wt.% or even equal to or less

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than 25wt.%, where all weight percentages are based on the total weight of the solid biomass material.

If a fluidizing reactor is used in the pyrolysis step, preferably part of the gaseous product fraction is recycled to the fluidizing reactor in such pyrolysis step as at least part of the carrier gas. Without wishing to be bound by any kind of theory it is believed that such a recycle can further improve the catalyst lifetime of any catalyst in the pyrolysis step and/or the subsequent catalytic step.

If a fluidizing reactor is used in the pyrolysis step, the fluidizing reactor is preferably a riser reactor. By a riser reactor is herein preferably understood an elongated, preferably essentially tube-shaped, reactor. The elongated reactor is preferably oriented in an essentially vertical manner. Examples of suitable riser reactors are described in the Handbook titled "Fluid Catalytic Cracking technology and operations", by Joseph W. Wilson, published by PennWell Publishing Company (1997), chapter 3, especially pages 101 to 112, herein incorporated by reference.

In the pyrolysis step the solid biomass material can be heated in any manner known to the skilled person in the art. The solid biomass material may for example be suitably preheated before entering any reactor and be subsequently heated further in the reactor where the pyrolysis is to take place.

In a preferred embodiment the solid biomass material is contacted with a solid heating medium, such as for example silica or sand.

For example, when a fluidizing reactor is used, the solid heating medium may be fluidized with the help of a carrier gas as described above. In such case, the solid

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biomass material may be fluidized within fluidized hot solid heating medium and subsequently the solid biomass material may be pyrolysed with the heat provided by such fluidized hot solid heating medium. Hereafter, the used solid heating medium may be separated from the biomass-derived vapours, for example in a cyclone, and any residual coke formed on the used solid heating medium may be burned off in a regenerator to regenerate hot solid heating medium. The coke that is burned off may conveniently supply the heat needed to heat the solid heating medium.

If present, the weight ratio of any solid heat carrier material to the solid biomass material (i.e. the solid heat carrier: solid biomass material weight ratio) at the location where the materials are supplied to the fluidizing reactor preferably lies in the range from equal to or more than 1:1, more preferably from equal to or more than 2:1 and most preferably from equal to or more than 3:1 to equal to or less than 50:1, more preferably to equal to or less than 10:1, most preferably to equal to or less than 5:1.

In addition to the solid heating medium or instead of the solid heating medium, a catalyst can be used as a source of heat. In this case, the solid biomass material can be contacted with a catalyst as described above.

For example, when a fluidizing reactor is used, the catalyst may be fluidized with the help of a carrier gas as described above in the fluidizing reactor. In such case, the solid biomass material may be fluidized within fluidized hot catalyst and subsequently the solid biomass material may be pyrolyzed with the heat provided by such fluidized catalyst. Hereafter, the used catalyst may be separated from the biomass-derived vapours, for example

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in a cyclone, and any residual coke formed on the used catalyst may be burned off in a regenerator to regenerate hot catalyst. The coke that is burned off may conveniently supply the heat needed to heat the catalyst.

If a catalyst is present, the weight ratio of catalyst to solid biomass material (catalyst:solid biomass material weight ratio) at the location where the solid biomass material is supplied to the reactor suitably lies in the range from equal to or more than 1:1, more preferably equal to or more than 2:1 to equal to or less than 100:1, more preferably equal to or less than 50:1.

The pyrolysis step can be carried out batch-wise, semi-batch wise or continuously.

Suitably the pyrolysis step can be carried out at a WHSV (weight hourly space velocity) that is equal to or more than 0.05 hr^{-1} , more preferably equal to or more than 0.10 hr^{-1} , and equal to or less than 40.0 hr^{-1} , more preferably equal to or less than 10.0 hr^{-1} and yet more preferably equal to or less than 1.0 hr^{-1} .

Preferably the pyrolysis step comprises pyrolyzing at least part of the solid biomass material by contacting the solid biomass material with a catalyst and/or a solid heat carrier medium at a temperature in the range from equal to or more than 400°C to equal to or less than 600°C in a fluidizing reactor, whilst introducing a carrier gas to the fluidizing reactor, thereby producing biomass-derived vapours.

In addition to the biomass-derived vapours the effluent of the pyrolysis step can potentially include char, ash and/or any entrained catalyst particles and/or solid heating medium particles. Preferably such potential char, ash and/or any entrained catalyst particles and/or

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solid heating medium particles is/are separated from the biomass-derived vapours. The effluent of the pyrolysis step is therefore preferably passed through a cyclone and/or a ceramic hot gas filter to separate any char, ash and/or any entrained catalyst particles and/or solid heating medium particles from the biomass-derived vapours. Such char, ash and/or any entrained catalyst particles and/or solid heating medium particles can conveniently be forwarded to a regenerator unit and/or be disposed of in an environmentally friendly manner.

In the catalytic step at least part of the biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C to produce a product mixture. Preferably such biomass-derived vapours are pyrolysis vapours obtained or obtainable by pyrolysis of a solid biomass material as described herein above.

The catalytic step can conveniently be carried out in a reactor. More preferably the catalytic step is carried out in a fixed bed reactor or a fluidizing reactor, most preferably a fixed bed reactor.

The fluidizing reactor may be a fluidizing reactor similar to the one described above for a pyrolysis step. In one embodiment any pyrolysis step and the catalytic step are carried out in the same reactor. The process according to the invention therefore comprises a process for the conversion of a solid biomass material, containing the following steps:

(a) pyrolyzing at least part of the solid biomass material by contacting the solid biomass material with a catalyst at a temperature in the range from equal to or more than 400°C to equal to or less than 600°C in a first zone of a fluidizing reactor, thereby producing biomass-

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derived vapours;

(b) contacting at least part of the biomass-derived vapours with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C in a second zone of the fluidizing reactor, thereby producing a product mixture;

(c) condensing part of the product mixture, thereby forming a condensed product fraction and a gaseous product fraction,

wherein at least part of the gaseous product fraction is recycled to step (b) and added to the biomass-derived vapours in the second zone of the fluidizing reactor.

For this embodiment the catalysts in step (a) and (b) are preferably the same. Further preferences for step (a) in this embodiment are as described for the pyrolysis step above. Preferences for step (b) are as described for the catalytic step below and preferences for step (c) are as described for the condensing step below.

Preferably, however, the catalytic step is carried out in a different reactor than any pyrolysis step. More preferably the catalytic step is carried out in one or more fixed bed reactors whilst any optional pyrolysis step is carried out in a fluidizing reactor. For example the catalytic step may be carried out in two fixed bed reactors operated in swing mode.

The preferences as described herein for the catalytic step apply to both the catalytic step in the process for the conversion of biomass-derived vapours and the catalytic step in the process for the conversion of a solid biomass material.

The catalyst in the catalytic step is preferably a MFI or MWW type zeolite catalyst. The catalysts preferably have a pore size ranging from 0.5 to 0.6nm and

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have a strong acidity. A preferred MWW type zeolite is MCM-22. By a MFI type zeolite is herein understood a zeolite having the characteristics of the MFI type as listed in the database approved by the Structure Commission of the International Zeolite Association (IZA-SC) that can be found at <http://www.iza-structure.org/databases>. More preferably, the catalyst in the catalytic step is a MFI type zeolite, even more preferably a zeolite belonging to the so-called pentasil family of zeolites. A highly preferred zeolite is ZSM-5, which has a pore structure that selectively creates mono-aromatics. A further preferred catalyst comprises ZSM-5 nanosheet material, which - similar to MFI nanosheets - has a multilamellar structure.

In a further embodiment of the invention, the MFI type zeolite catalyst is a mesoporous catalyst.

In the article titled "In situ assembly of zeolite nanocrystals into mesoporous aggregate with single-crystal like morphology without secondary template" by Yunming Fang et al. published in Chem. Mater. Vol. 20, pages 1670-1672 (2008) the synthesis of a so-called mesoporous ZSM-5 zeolite is described. A uniform mesoporous aggregate of zeolite nanocrystals with size smaller than 1 micrometer was formed. This is an example of a preferred mesoporous ZSM-5 catalyst.

The volume of mesopores (also called mesopore volume, cm^3/gram) in the MFI-type zeolite may vary widely. Preferably, however, the mesoporous volume lies in the range from equal to or more than $0.10 \text{ cm}^3/\text{gram}$, more preferably from equal to or more than $0.20 \text{ cm}^3/\text{gram}$, and most preferably from equal to or more than $0.30 \text{ cm}^3/\text{gram}$ to equal to or less than $1.50 \text{ cm}^3/\text{gram}$, more preferably

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from equal to or less than $1.25 \text{ cm}^3/\text{gram}$, most preferably equal to or less than $1.00 \text{ cm}^3/\text{gram}$.

The pore volumes of the zeolite catalyst can suitably be determined by nitrogen adsorption analysis according to ASTM standard ASTM D5604.

In addition to the MFI-type zeolite, the catalyst may contain one or more additional zeolites. Such additional zeolite is preferably chosen from the group consisting of Y zeolites; ultrastable Y zeolites (USY); X zeolites, zeolite beta, zeolite L, offretite, mordenite, faujasite (including synthetic faujasite), zeolite omega, Rare Earth zeolite Y (= REY) and Rare Earth USY (REUSY). Further the catalyst may contain additional zeolites for example chosen from the group consisting of MTW type zeolites (such as for example ZSM-12); MTT type zeolites (such as for example ZSM-23); the TON type zeolites (such as for example zeolite theta one or ZSM-22); and the FER structural type, for example, ferrierite.

The MFI-type zeolite and optionally additional further zeolites are preferably dispersed in an amorphous matrix component. For example the catalyst may contain amorphous silica alumina, the MFI-type zeolite and optionally one or more additional zeolites. In addition, the catalyst preferably contains a binder and/or a filler.

Examples of such an amorphous matrix include amorphous silica-alumina, amorphous silica, amorphous alumina, amorphous titania, amorphous zirconia and amorphous magnesium oxide, or combinations of two or more of these.

The MFI-type zeolite is preferably used in the form of extrudates, suitably with alumina, preferably with 35-45%, in particular 40%, of alumina. Preferably the MFI-

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type zeolite comprises a ratio of moles silica to moles alumina ($\text{SiO}_2:\text{Al}_2\text{O}_3$) in the range from equal to or more than 20 to 1 (20:1) to equal to or less than 100 to 1 (100:1), more preferably in the range from equal to or more than 30 to 1 (30:1) to equal to or less than 80 to 1 (80:1).

An example of a binder is silica sol. Examples of fillers include natural or synthetic clays, pillared or delaminated clays, or mixtures of one or more of these. Examples of clays which may be present in the catalyst include kaolin, hectorite, sepiolite and attapulgite.

The total amount of zeolite that is present in the catalyst is preferably in the range of 5 wt.% to 50 wt.%, more preferably in the range of 10 wt.% to 30 wt.%, and even more preferably in the range of 10 wt.% to 25 wt.% relative to the total mass of the catalyst, whilst the remainder is preferably amorphous matrix component, binder and/or filler.

In addition the catalysts may contain a metal and/or metal oxide. For example the catalyst may contain nickel, platinum, vanadium, palladium, manganese, molybdenum, iron, cobalt, zinc, copper, chromium, zinc, gallium and/or any of their oxides. Preferably the catalyst contains nickel, gallium or zinc or any combination or oxide thereof.

The catalytic step can be carried out batch-wise, semi-batch wise or continuously.

Suitably the catalytic step can be carried out at a WHSV (weight hourly space velocity) that is equal to or more than 0.05 hr^{-1} , more preferably equal to or more than 0.10 hr^{-1} , and equal to or less than 40.0 hr^{-1} , more preferably equal to or less than 10.0 hr^{-1} and yet more preferably equal to or less than 1.0 hr^{-1} .

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Preferably the catalytic step is carried out at a pressure in the range from equal to or more than 0.5 bar absolute (0.05 MegaPascal) to equal to or less than 10 bar absolute (1 MegaPascal), more preferably from equal to or more than 1.0 bar absolute (0.1 MegaPascal) to equal to or less than 3 bar absolute (0.3 MegaPascal). Most preferably the catalytic step is carried out at atmospheric pressure (i.e. about 1.0 bar absolute).

Preferably the catalytic step is carried out at a temperature from equal to or more than 350°C to equal to or less than 550°C, preferably from 400°C, more preferably from 450°C to equal to or less than 525°C. Preferably the catalytic step is carried out at a temperature equal to or lower than the temperature at which any pyrolysis step is carried out. The pyrolysis step is preferably operated at a temperature higher than the catalytic step.

As indicated above, it is possible for any pyrolysis step and the catalytic step to be carried out in the same reactor. Preferably, however, any pyrolysis step is a non-catalytic pyrolysis step preceding the catalytic step and this non-catalytic pyrolysis step and the catalytic step are carried out in separate reactors.

In the catalytic step a product mixture is produced. The product mixture of the catalytic step is suitably forwarded to one or more condensers.

The preferences as described herein for the condensing step apply to both the condensing step in the process for the conversion of biomass-derived vapours and the condensing step in the process for the conversion of a solid biomass material.

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In the condensing step, part of the product mixture is condensed, thereby forming a condensed product fraction and a gaseous product fraction.

The product mixture produced in the catalytic step can include hydrocarbon vapours, water vapour and other gasses, such as for example carbonmonoxide and carbon dioxide.

The product mixture can be partially condensed in any manner known by the person skilled in the art to be suitable for condensing a hydrocarbon vapour or water vapour. The condensation may for example be carried out by direct or indirect contact between the product mixture and a coolant. Preferably the product mixture is partially condensed by contacting it indirectly with a coolant in one or more condensers.

Preferably the product mixture is condensed in two or more stages. For example the product mixture can be cooled in a first stage condenser (at a pressure of about 1 bar absolute, corresponding to about 0.1 MegaPascal) to a temperature in the range from equal to or more than -10°C to equal to or less than 25°C and more preferably to a temperature in the range from equal to or more than 0°C to equal to or less than 15°C, to separate a first condensable liquid fraction, whereafter the remaining non-condensed vapours of the product mixture are cooled in a second stage condenser (at a pressure of about 1 bar absolute, corresponding to about 0.1 MegaPascal) to a temperature equal to or less than -25°C to separate a second condensable liquid fraction.

The condensed product fraction is herein also referred to as "bio-oil" and contains biomass-derived hydrocarbons and other biomass-derived compounds (such as for example oxygenates) that can be used for the

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production of a fuel component and/or chemical component or as a precursor for such a fuel and/or chemical component, as will be described in more detail below.

The product mixture is only partly condensed, some of the vapours are not condensed into a liquid and form the gaseous product fraction. The remaining gaseous product fraction can for example include carbon monoxide, carbon dioxide, and/or C1-C4 hydrocarbons such as ethene, propene, butene, methane, ethane, propane and butane.

In all embodiments of the invention at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours. The part of the gaseous product fraction being recycled to the catalytic step is herein also referred to as "recycled gaseous product fraction".

Preferably the recycled gaseous product fraction includes carbon monoxide, carbon dioxide and/or ethene, propene, butene, methane, ethane, propane and/or butane.

The recycled gaseous product fraction and the biomass-derived vapours can be combined in any manner known by the person skilled in the art to be suitable therefore. For example, the recycled gaseous product fraction can be added to the biomass-derived vapours or the biomass-derived vapours can be added to the recycled gaseous product fraction, or both the recycled gaseous product fraction and the biomass-derived vapours can be added simultaneously to a container or vessel and be mixed therein.

In one preferred embodiment the recycled gaseous product fraction is directly added to and mixed with the biomass-derived vapours, after production of the biomass-derived vapours and before the catalytic step, whereafter a mixture of the recycled gaseous product fraction and

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biomass-derived vapours is contacted with the catalyst in any fixed bed reactor or in any other reactor of the catalytic step.

In another preferred embodiment the recycled gaseous product fraction is added into a fixed bed reactor or other reactor in which the catalytic step is carried out, simultaneously with but separately from the biomass-derived vapours. In this case the recycled gaseous product fraction becomes added to and mixed with the biomass-derived vapours within such a fixed bed reactor or other reactor. Any addition of recycled gaseous product fraction to a fixed bed reactor or other reactor simultaneously with but separately from the addition of biomass-derived vapours to such same reactor, is preferably carried out via a set of multiple inlets. The multiple inlets are preferably arranged such to achieve a homogeneous mixture of recycled gaseous product fraction and biomass-derived vapours within the fixed bed reactor or other reactor.

In addition to the other advantages mentioned, the recycled gaseous product fraction can be used to assist in regulating the temperature and the reaction rate for the catalytic step.

In addition to recycling part of the gaseous product fraction to the catalytic step, another part of the gaseous product fraction can optionally be recycled to any optional pyrolysis step to be used as part of the carrier gas.

As illustrated in the examples, the process according to the invention advantageously allows one to improve hydrocarbon yield and reduce gas-production.

The hydrocarbons in the condensed product fraction can be fractionated into one or more hydrocarbon product

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fractions. Potentially one or more of these hydrocarbon product fractions may be subsequently hydrotreated to produce a hydrotreated hydrocarbon product fraction. The non-hydrotreated hydrocarbon product fraction(s) and/or hydrotreated hydrocarbon product fraction(s) may be used as fuel and/or chemical component(s) in respectively a fuel and/or chemical or as precursors thereof.

The invention is further illustrated by the following non-limiting examples.

Comparative examples A and B and example 1

The examples were carried out in an experimental set-up as illustrated in figure 1, comprising a bubbling fluidized bed (BFB) reactor (101) and a fixed bed reactor (102). The fluidized bed reactor had an inner diameter of 25 millimeter (mm), and a length of 600 mm, which included a gas preheating zone (103) below a 100 mm porous gas distribution plate (104). Both BFB reactor (101) and the fixed bed reactor (102) were externally heated with an electric furnace (not shown). For each of the examples, wood chips were dried, ground and sieved into a particle size of less than 40 mesh. The wood chips were subsequently fed via a feed hopper (105), a spring screw feeder system (106) and a high velocity nitrogen swept sloped feeder tube (107) into the BFB reactor (101) at a feed rate of approximately 200 gram per hour (g/h) on a dry basis. In the BFB reactor (101) the wood chips were back-mixed with sand (108).

The sand and the wood chips in the BFB reactor were fluidized with a carrier gas (109). The carrier gas that was used in each example is specified in table 1 below. In each example, temperature of the BFB reactor (101) was about 550°C. Temperatures within the reactor were

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measured and controlled with three thermocouples (not shown).

Upon exiting the BFB reactor (101), the biomass-derived vapours (110) produced in the BFB reactor (101) were passed through a cyclone (111) to remove residual sand and ashes (112) and through a ceramic hot gas filter (113). The cyclone (111) and ceramic hot gas filter (113) were both maintained at a temperature of 400°C. Then the resulting biomass-derived vapours (114) were passed to the fixed bed reactor (102).

As summarized in table 1, for comparative examples A and B only biomass-derived vapours were passed to the fixed bed reactor (102) and for example 1 a mixture of biomass-derived vapours and recycled gaseous product fraction was used.

The fixed bed reactor (102) was loaded with about 100 gram of a catalyst (115). The fixed catalyst (115) in the fixed bed reactor (102) was a mesoporous ZSM-5 catalyst with a mesopore volume of 0.34 cm³/gram. The fixed bed catalyst was the same in each example. In each example the temperature of the catalyst in the fixed bed was kept at about 500°C (i.e. below the temperature of the BFB reactor (101)). The product mixture (116) from the fixed bed reactor (102) was introduced into a liquid recovery system consisting of two condensers (117 and 118). The condensers were cooled using a 50/50 (vol.%/vol.%) mixture of ethylene glycol and water. The mixture had a temperature of about -5°C. The ethylene glycol/water mixture itself was cooled down using a 20 liter refrigerated circulating bath (not shown).

After condensation the still gaseous product fraction passed through a flow totalizer, which was followed by a gas chromatographer (GC) for gas composition analysis.

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The GC was configured to run three parallel channels (one Flame Ionization Detector and two Thermal Conductivity Detectors, not shown). All three detectors collected data at the same time. For each example the composition of the gaseous product fraction after condensation is specified in Table 2. In example 1 part of the gaseous product fraction (as indicated in Table 1) was recycled via line (119) to fixed bed reactor (102).

The yields of the condensed (liquid) product fraction and the char/coke were determined gravimetrically by weighing. Subsequently the condensed (liquid) product fraction was analyzed for its water content using volumetric Karl Fisher titration. The elemental composition (carbon, hydrogen, nitrogen, sulphur and oxygen) of the bio-oil was determined using a Vario EL organic elemental analyzer. For each example, the composition of the condensed product fraction is also specified in Table 2.

As illustrated by Comparative examples A and B and example 1, the recycle of gaseous product fraction to the catalytic step in the fixed bed reactor advantageously resulted in a lower total gas product fraction and a higher weight percentage of hydrocarbons in the condensed product fraction.

In addition it was surprisingly found that the recycle of the gaseous product fraction resulted in an improved catalyst lifetime of the catalyst in the fixed bed reactor. Table 3 illustrates that the catalyst lifetime in a situation where the gaseous product fraction was recycled was longer than that without a recycle of such gaseous product fraction. Table 4 illustrates the linear decrease of micropore volume of the catalyst over 9 hours as in example 1.

Table 1: Summary of comparative examples A* and B* and example 1.

Example	Carrier gas used in BFB reactor (101)	Gas feed used for fixed bed reactor (102)
A*	100 vol. % Nitrogen	100 vol. % biomass-derived vapours
B*	70 vol. % nitrogen and 30 vol. % recycled gaseous product fraction as listed for example B* in table 2.	100 vol. % biomass-derived vapours
1	100 vol. % Nitrogen	65 vol. % biomass-derived vapours and 35 vol. % recycled gaseous product fraction as listed for example 1 in table 2

* Comparative examples

Table 2: The product mixture, condensed product fraction and gaseous product fraction.

Example	Condensed product fraction		Water wt. %	Gaseous product fraction			Total gas product fraction wt. %	Char wt. %	Coke % wt	Total % wt
	wt. % HC	wt. % Oxyg.		wt. % CO	wt. % CO ₂	wt. % C1-C4 alkanes and/or alkenes				
A*	16.3	1.70	16.0	7.3	5.4	6.0	18.7	29.4	7.6	89.7
B*	17.2	1.90	17.0	5.7	9.2	2.1	17.0	29.8	7.8	90.7
1	18.15	1.95	17.2	3.1	9.7	1.4	14.2	30.4	8.1	90.0

* Comparative examples

wt. % = weight percentage based on the dry weight of the biomass feedstock.

HC = hydrocarbons, consisting solely of hydrogen and carbon.

Oxyg. = Oxygenates

CO = Carbon monoxide

CO₂ = Carbon dioxide

C1-C4 alkanes and/or alkenes = alkanes and/or alkenes comprising 1 to 4 carbon atoms.

Table 3: Catalyst lifetime** of the mesoporous ZSM-5 catalyst residing in the fixed bed reactor.

TOS (hours)	1	2	3	4	5	6	7	8	9	10	11
Oxygen content (wt.%), when recycling the gaseous product fraction into the fixed bed reactor (example 1)	0.2	0.3	0.4	0.8	2.3	2.4	2.5	4.0	8.5	12.5	20.1
Oxygen content (wt.%) when not recycling any gaseous product fraction into the fixed bed reactor (example A*)	0.2	0.3	1.0	1.7	6.2	9	18	-	-	-	-

** Catalyst lifetime was defined as the reaction time before the oxygen content in the produced liquid exceeded 1.5%
TOS = Time on stream

Table 4: The micropore volume of the mesoporous ZSM-5 catalyst residing in the fixed bed reactor as a function of time on stream

TOS (hours)	0	1	3	5	7	9
Micropore volume (millilitre/gram)	0.13	0.11	0.1	0.09	0.06	0.04

TOS = Time on stream

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C L A I M S

1. A process for the conversion of biomass-derived vapours, comprising:
 - a catalytic step, wherein at least part of the biomass-derived vapours are contacted with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C, thereby producing a product mixture; and
 - a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction;wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours.
2. A process according to claim 1, wherein the biomass-derived vapours are pyrolysis vapours obtained or obtainable by pyrolysis of a solid biomass material.
3. A process according to claim 2, where the solid biomass material contains cellulose and/or lignocellulose.
4. A process for the conversion of a solid biomass material, comprising:
 - a pyrolysis step, wherein at least part of the solid biomass material is pyrolyzed, thereby producing biomass-derived vapours;
 - a catalytic step, wherein at least part of the biomass-derived vapours are contacted with a catalyst

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at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C, thereby producing a product mixture; and
- a condensing step, wherein part of the product mixture is condensed in one or more condensers, thereby forming a condensed product fraction and a gaseous product fraction;
wherein at least part of the gaseous product fraction is recycled to the catalytic step and combined with the biomass-derived vapours.

5. A process according to claim 4, where the solid biomass material contains cellulose and/or lignocellulose.
6. A process according to claim 4 or 5, wherein the pyrolysis step comprises pyrolyzing at least part of the solid biomass material by contacting the solid biomass material with a catalyst and/or a solid heat carrier medium at a temperature in the range from equal to or more than 400°C to equal to or less than 600°C in a fluidizing reactor, whilst introducing a carrier gas to the fluidizing reactor, thereby producing biomass-derived vapours.
7. The process according to claim 6, wherein the carrier gas includes carbon monoxide, carbon dioxide, nitrogen, steam, any C₁-C₄ hydrocarbon or a mixture of any of these.
8. The process according to anyone of claims 4 to 7, wherein the catalytic step is carried out at a

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temperature equal to or lower than the temperature at which any pyrolysis step is carried out.

9. The process according to anyone of claims 1 to 8, wherein the catalytic step comprises contacting at least part of the biomass-derived vapours with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C in a fixed bed reactor, thereby producing a product mixture.
10. The process according to anyone of claims 1 to 9, wherein the catalyst in the catalytic step is a mesoporous MFI type catalyst.
11. The process according to anyone of claims 1 to 10, wherein the condensing step comprises condensing part of the product mixture in two or more condensers.
12. The process according to anyone of claims 1 to 11, wherein the part of the gaseous product fraction that is recycled to the catalytic step comprises carbon monoxide, carbon dioxide.
13. The process according to anyone of claims 1 to 12, wherein the part of the gaseous product fraction that is recycled to the catalytic step comprises methane, ethane, propane, butane, ethene, propene and/or butene.
14. A process for the conversion of a solid biomass material, comprising the following steps:

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(a) pyrolyzing at least part of the solid biomass material by contacting the solid biomass material with a catalyst at a temperature in the range from equal to or more than 400°C to equal to or less than 600°C in a first zone of a fluidizing reactor, thereby producing biomass-derived vapours;

(b) contacting at least part of the biomass-derived vapours with a catalyst at a temperature in the range from equal to or more than 350°C to equal to or less than 550°C in a second zone of a fluidizing reactor, thereby producing a product mixture;

(c) condensing part of the product mixture, thereby forming a condensed product fraction and a gaseous product fraction,

wherein at least part of the gaseous product fraction is recycled to step (b) and added to the biomass-derived vapours.

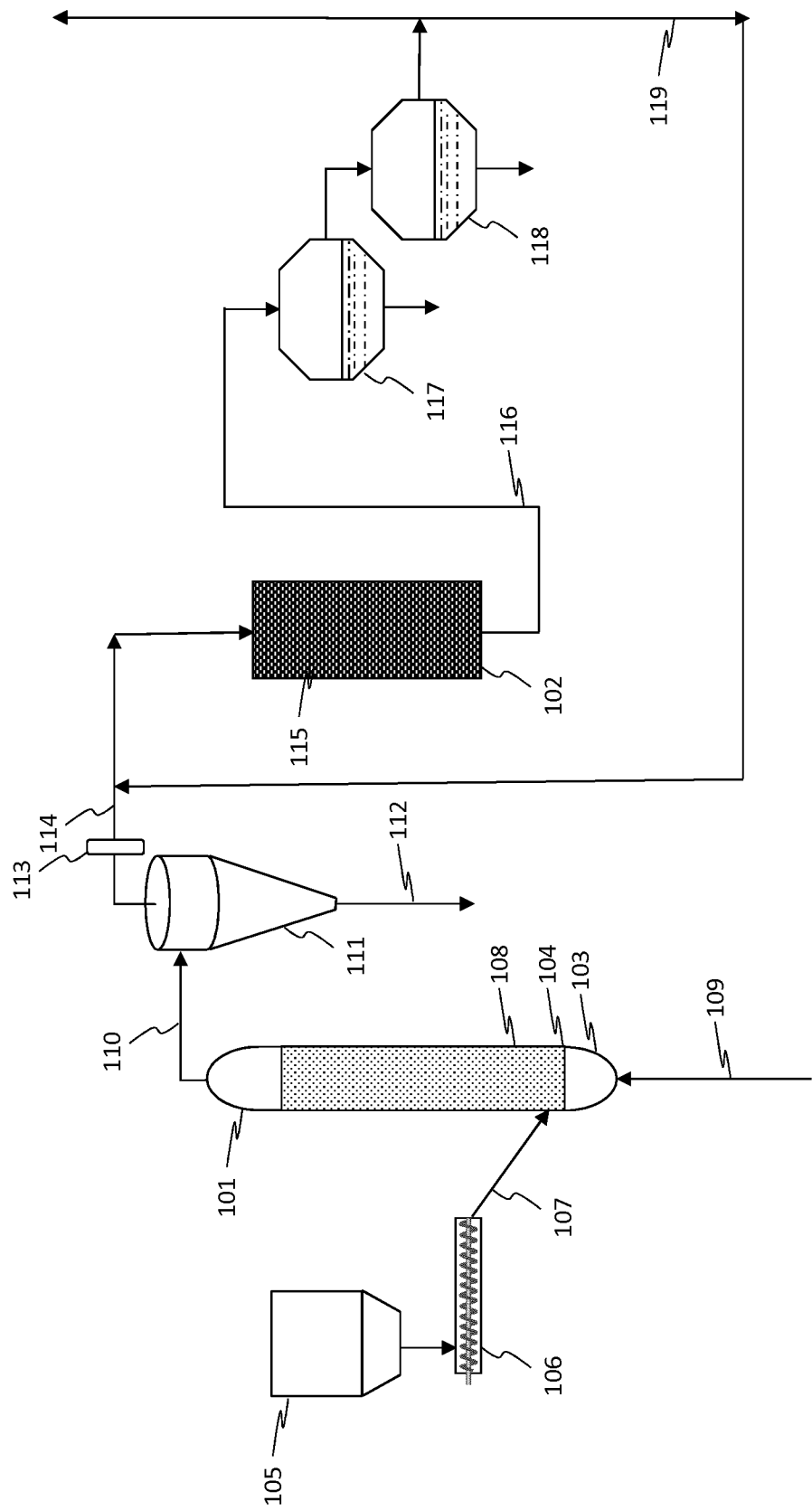


Figure 1

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/068304

A. CLASSIFICATION OF SUBJECT MATTER
INV. C10G1/00 C10G1/02 C10G1/08
ADD.

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)
C10G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	James Diebold ET AL: "Biomass to Gasoline" In: "Pyrolysis Oils from Biomass", 30 September 1988 (1988-09-30), American Chemical Society, Washington, DC, XP055114410, ISBN: 978-0-84-121228-2 vol. 376, pages 264-276, DOI: 10.1021/bk-1988-0376.ch023, pages 267-273	1-14
X	----- WO 2015/047085 A1 (BIOBTX B V [NL]; UNIV GRONINGEN [NL]; KNN ADVIES B V [NL]; SYNCOM B V) 2 April 2015 (2015-04-02) claims 1, 4, 5, 14, 18-22 figure 1 page 5, paragraph 5-6 page 9, paragraph 3 pages 10-12 ----- -/-	1-14



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

"E" earlier application or patent but published on or after the international filing date

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"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

"&" document member of the same patent family

Date of the actual completion of the international search

10 October 2017

Date of mailing of the international search report

18/10/2017

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INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/068304

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2014/072480 A1 (JONES SAMUEL T [US] ET AL) 13 March 2014 (2014-03-13) claims 1, 8 paragraphs [0022], [0027], [0030] - [0032], [0034], [0056], [0065], [0068] figure 1 -----	1-14
X	US 2015/073181 A1 (GORKE JOHNATHAN T [US] ET AL) 12 March 2015 (2015-03-12) claims 1, 7 paragraphs [0018] - [0020], [0024], [0026], [0027], [0041], [0045], [0057], [0059] -----	1-14

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2017/068304

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