

(19) United States

(12) Patent Application Publication (10) Pub. No.: US 2005/0167259 A1

Wagenaar et al.

Aug. 4, 2005 (43) Pub. Date:

(54) METHOD FOR THE PYROLYSIS OF A PYROLYSABLE MASS

(75) Inventors: Bertus Michiel Wagenaar, Hengelo (NL); Robert Hendrikus Venderbosch, Enschede (NL); Elwin Gansekoele, Enschede (NL); Jan Hendrik Florijn,

Haarlo (NL)

Correspondence Address: PEACOCK MYERS AND ADAMS P C

PO BOX 26927 ALBUQUERQUE, NM 871256927

(73) Assignee: BIOMASS TECHNOLOGY BEHEER B.V. (formerly known asTNO Biom-

assTechnology Group B.V.), Woudweg

(NL)

(21) Appl. No.: 11/008,318

(22) Filed: Dec. 9, 2004

Related U.S. Application Data

(63) Continuation of application No. PCT/NL03/00442, filed on Jun. 16, 2003.

(30)Foreign Application Priority Data

Jun. 14, 2002 (NL)...... 1020861

Publication Classification

- (51) **Int. Cl.**⁷ **C10B 49/00**; C10G 1/00 **U.S. Cl.** 201/25; 585/240
- **ABSTRACT** (57)

A method of pyrolysis wherein pyrolysable particles mixed with heated carrier particles are treated at an elevated temperature yielding a flammable product. According to the invention, use is made for mixing a rotating, vibrating or shaking element or by mixing at a volume density of less than 40%, and mixing occurs to a specified minimum degree after the mixture is deposited and is pyrolysed in the reactor after depositing.

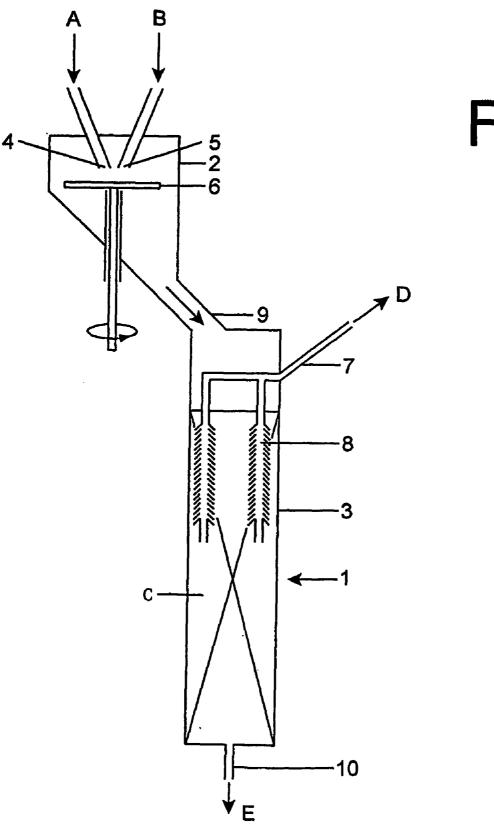


Fig.

METHOD FOR THE PYROLYSIS OF A PYROLYSABLE MASS

[0001] The present invention relates to a method for the pyrolysis of pyrolysable mass in the form of particles, wherein the pyrolysable particles are treated in the presence of carrier particles at elevated temperature, yielding at least one combustible product, wherein

[0002] the carrier particles are heated;

[0003] the heated carrier particles are contacted with the pyrolysable particles and are mixed therewith;

[0004] the mixture of pyrolysable particles and heated carrier particles pyrolyses in a reactor yielding a hot, gaseous, combustible product and a pyrolysable mass-depleted mixture; and

[0005] the pyrolysable mass-depleted mixture is removed for heating the carrier particles.

[0006] Such a method is described by the Lurgi Company where anorganic particles (ash residue particles) are fed into an extruder-like apparatus, which extruder possesses two horizontal screws for the transport of the anorganic particles. In the apparatus a hydrocarbon-comprising material, such as oil residue, pitch sand and heavy oil fractions, are added to the anorganic particles. The treatment at 500-600° C. causes the development of gasses and vapours, as well as cokes. The screws effectuate mixing between the anorganic particles and the mass to undergo pyrolysis, wherein little mixing occurs in the axial direction and satisfactory mixing in the radial direction. The screws also ensure that the mixture is transported through the apparatus. The advantage of the method described is that in contrast with methods based on a fluidised bed, there is no need to add large amounts of carrier gas, which greatly dilute the gaseous product.

[0007] The known method has the drawback that the screws encounter much resistance and driving them therefore requires much energy. This at the cost of the total energetic yield of the process carried out with the apparatus. The considerable resistance also means wear of the apparatus, in particular the screws, which has an adverse effect on the method's economy.

[0008] The object of the present invention is to provide a method of the kind mentioned in the preamble, wherein the energy losses caused by mixing the pyrolysable mass with carrier particles is reduced, and which is economically advantageous.

[0009] To this end the method according to the invention is characterised in that the carrier particles are supplied at a rate A and the pyrolysable particles at a rate B, and mixing is carried out such

[0010] that by using a method of mixing chosen from i) mixing by means of a rotating element, wherein the pyrolysable particles and the carrier particles at the centre of the rotating element come into contact with the rotating element and the rotating element rotates at a speed such that the pyrolysable particles and the carrier particles move over the surface of the rotating element to be hurled away, ii) mixing by means of a vibrating or shaking element, and the pyrolysable particles and the carrier particles leave

the vibrating or shaking element over an edge thereof or hole therein, and iii) mixing at a volume density of less than 40%; and

[0011] that before the mixture is deposited it is mixed at least well enough that in a sample of one cubic decimetre of the thus obtained mixture the A/B ratio deviates less than 20% from the ratio at which A and B are added, and the mixture obtained is deposited, and in a condition after pouring undergoes pyrolysis in the reactor.

[0012] In this way the pyrolysable particles and the carrier particles are mixed very quickly, without any extensive pyrolysis occurring before the mixture is deposited producing a mixed mass, whose further mixing would involve high energy costs. The fact that as carrier particles sand can be used is in itself known. In order to limit the loss of pyrolysis gasses via feed openings for carrier particles and pyrolysable particles the feed openings are connected to substantially vertical feed pipes whose inside diameter and length are chosen such that they form a resistance for the pyrolysis gasses. In case of the use of a rotating, vibrating or shaking element, the mixture of particles will in general fall over the edge of said element. Holes in said element can be provided to allow passage of said mixture. In the present invention, the term pyrolysis is understood to mean any thermal treatment producing decomposition. The pyrolysable mass preferable comprises at least one component chosen from a hydrocarbon and a carbohydrate. The present invention is in particular suitable for the formation of fuel that is liquid at room temperature and obtained by means of condensation. The risk that the thus formed compounds that are liquid at room temperature will be cracked further is very small. Also, in contrast to known methods, dilution by a carrier gas can be avoided. In the present application a carrier particle is understood to be a non-pyrolysable particle that is capable of absorbing and dissipating heat. The particle is, for example, an inert particle such as a sand grain, but may also be a catalytic particle. Such a catalytic particle preferably contributes to the formation of pyrolytic products that are liquid at room temperature. The term volume density denotes the ratio of the volume of the solid in relation to the total volume (i.e. inclusive of interstitial space) taken up by the solid. The volume density is preferably less than 25%, such as less than 15%. Lower volume density not only ensures low-friction mixing, but also limit pyrolysis during mixing.

[0013] The deviation is preferably less than 10%.

[0014] In practice this means that no further mixing is required. If further mixing is desirable, it usually suffices to carry out this mixing at the surface of the deposited mixture, where still fairly little force is needed for mixing.

[0015] Contacting and mixing is preferably realised by means of a rotating cone.

[0016] Such a cone is shown to be very useful for quickly and efficiently mixing the pyrolysable mass and the carrier particles.

[0017] Mixing and pyrolysis preferably occurs in two separate chambers, which are in communication by means of a partition provided with at least one opening.

[0018] The partition is designed such that carrier particles or pyrolysable particles are prevented from remaining

thereon, and is, for example, a grid or a funnel. This promotes the avoidance of pyrolytic gasses from the deposited mixture coming into the mixing chamber from where they might be able to escape via the feed pipes for the carrier particles and pyrolysable particles still to be mixed.

[0019] The combustible product is preferably cooled and separated into a liquidised oil component and a gaseous component.

[0020] In particular the liquidised oil component is usable as liquid fuel.

[0021] The gaseous component may be combusted and used for drying the pyrolysable mass.

[0022] In this way it is possible to influence the moisture content of the pyrolysable mass, and consequently the composition of the liquid and gaseous products.

[0023] Pyrolysable mass-depleted mixture is preferably discharged from the bottom of the deposited mixture.

[0024] The mixture is thus transported into the reactor at low energy cost, while at the top side newly deposited mixture comprising relatively much mass still to be pyrolysed is supplied.

[0025] If the pyrolysable mass contains components or products causing caking, this can be effectively remedied if during pouring of the mixture in the reactor carrier particles are poured against the inner wall of the reactor.

[0026] Preferably, the carrier particles are heated by passing an oxygen-comprising gas through the pyrolysable mass-depleted mixture.

[0027] Then, carbon-containing material still present is combusted and the carrier particles are brought again to a temperature allowing efficient pyrolysis in the reactor.

[0028] According to an important embodiment, the pyrolysable mass comprising a carbohydrate is biomass.

[0029] Thus, the method according to the present invention allows efficient pyrolysis of biomass, such as for the form ation of a bio-fuel, in particular bio-diesel.

[0030] The present invention will now be illustrated by an exemplary embodiment and with reference to the drawing, where the only FIGURE schematically depicts an apparatus suitable for performing the method according to the invention.

[0031] In FIG. 1 an apparatus 1 is depicted comprising a mixing chamber 2 and a reactor chamber 3. At the top side of the mixing chamber 2 a first feed opening 4 is provided for pyrolysable particles A, such as wood flour having a particle size of, for example 1-3 mm, and a second feed line 5 for hot carrier particles B, for example sand grains having a diameter of ca. 0.5 mm. The supplied particles A, B fall, in the embodiment disclosed here, on a rotating disc 6 present in the mixing chamber, near its centre. By the chosen number of revolutions per minute, and optionally further determined by the shape thereof (such as declining or ascending outwardly) the velocity with which the particles A, B move over the surface of disc 6 can be varied. A suitable rotational speed is for example 300 rpm. Thus, the particles A, B are mixed very quickly yielding a particle mixture.

[0032] After being hurled from the rotating disc 6 and possibly colliding with the inner wall of the mixing chamber 2, the mixture of particles formed falls downwardly and is deposited. It is in particular in the deposited mixture C that pyrolysis takes place, and the pyrolysis products D formed are discharged via a discharge line 7.

[0033] In the reactor zone of the reactor chamber 3 where the mixture C is deposited and pyrolysis takes place, collection tubes 8 may be present for reducing the (average) distance that pyrolysis products D have to travel through the deposited mixture C. This increases the yield of combustible pyrolysis products D liquid at room temperature. It is possible to collect the pyrolysis gases D above the deposited mixture C, but in the embodiment shown here use is made of collection tubes 8 present in the deposited mixture C, having oblique slits through which the particles, even they are smaller than the slit width, can not pass. It is preferred that the collection tubes 8 are not provided with such slits near the top of the deposited mixture C. Because of the pyrolysis, some fluidisation may occur which could transfer particles from the mixture C in the collection tube 8, which is less desirable. Optionally, the collection tubes 8 may be open at their lower ends. Thus, particles which enter the collection tube 8 can be discharged.

[0034] When the pyrolysable mass, such as biomass, still contains some moisture (water), this may contribute to the prevention that pyrolysis products enter the mixing chamber 2. This is in particular the case if a partition (not shown) or separation channel 9 is present between the zone where mixing occurs and the pyrolysis zone, which the mixture of A and B can pass quickly. To this end, a partition is for example in the form of a grating. Moisture in the biomass contributes, apart in particular mixing quickly, that little pyrolysis occurs in the mixing chamber. If the particles A and B are supplied via lines, the undesirable escape of pyrolysis products D is impeded even further. This is in particular the case of the lines are longer and have a smaller inner diameter. Of course, the lines are dimensioned in accordance with the desired capacity of the apparatus 1. It is advantageous if the lines are filled relatively high with particles. To prevent undesirable escape of pyrolysis products via the supply openings 4, 5, the lines may optionally be provided with non-return valves (not shown).

[0035] The mixture subjected to pyrolysis leaves via a discharge line 10 as a pyrolysable material-depleted mixture E the reactor chamber 3. This mixture E may contain, apart from particles B, possibly carbonised particles A'. The mixture E is in such a case contacted with oxygen, as a result of which the carbonised particles A' are combusting yielding re-heated particles B which are fed back to the mixing chamber 2. Optionally, a fuel, such as natural gas, is supplied to ensure that the particles B are sufficiently hot for return in the apparatus 1. Supply of oxygen may be performed by supplying air. Air may also be used for transporting the particles B up, in order to return these as heated particles back in the mixing chamber 2. Transporting particles in such a way is known in the art.

[0036] Instead of combusting the carbonised particles A', they may be gasified in the presence of water and or CO₂ yielding a flammable gas.

[0037] Generally, discharge of pyrolysis products via line 10 is circumvented also. This may be done as described

above for preventing the loss via the supply openings 4, 5. Also, the reactor chamber 3 may have such a length, that in the lower part no pyrolysis occurs and the lower part of the deposited material, converted from C into E, acts as a barrier.

What is claimed is:

1. A method for the pyrolysis of pyrolysable mass in the form of particles, wherein the pyrolysable particles are treated in the presence of carrier particles at elevated temperature, yielding at least one combustible product, wherein:

the carrier particles are heated;

the heated carrier particles are contacted with the pyrolysable particles and are mixed therewith;

the mixture of pyrolysable particles and heated carrier particles pyrolyses in a reactor yielding a hot, gaseous, combustible product and a pyrolysable mass-depleted mixture; and

the pyrolysable mass-depleted mixture is removed for heating the carrier particles; and

wherein the carrier particles are supplied at a rate of A and the pyrolysable particles at a rate B, and mixing is carried out such that by using a method of mixing chosen from i) mixing by means of a rotating element, wherein the pyrolysable particles and the carrier particles at the centre of the rotating element come into contact with the rotating element and the rotating element rotates at a speed such that the pyrolysable particles and the carrier particles move over the surface of the rotating element to be hurled away; ii) mixing by means of a vibrating or shaking element, and the pyrolysable particles and the carrier particles leave the

vibrating or shaking element over an edge thereof or hole therein, and iii) mixing at a volume density of less than 40%; and wherein before the mixture is deposited it is mixed at least well enough that in a sample of one cubic decimeter of the thus obtained mixture the A/B ratio deviates less than approximately 20% from the ratio at which A and B are added, and wherein the mixture obtained is deposited, and in a condition after pouring undergoes pyrolysis in the reactor, and the carrier particles are heated by passing an oxygencomprising gas through the pyrolysable mass-depleted mixture discharged from the reactor.

- 2. A method according to claim 1, wherein the deviation is less than approximately 10%.
- 3. A method according to claim 1 wherein contacting and mixing is realized by means of a rotating cone.
- 4. A method according to claim 1 wherein mixing and pyrolysis occurs in two separate chambers.
- 5. A method according to claim 1, wherein the combustible product is cooled and separated into a liquidized oil component and a gaseous component.
- **6.** A method according to claim 5, wherein the gaseous component is to be combusted and used for drying the pyrolysable mass.
- 7. A method according to claim 1, wherein the pyrolysable mass-depleted mixture is discharged from the bottom of the cast mixture.
- **8**. A method according to claim 1, wherein during pouring of the mixture in the reactor carrier particles are poured against the inner wall of the reactor.
- 9. A method according to claim 1, wherein the pyrolysable mass comprising a carbohydrate comprises biomass.

* * * * *