METHOD OF PRODUCING DROP-IN DIESEL

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Methods are disclosed for producing renewable diesel from hydrocarbon-containing feedstock using microwave energy and hydrotreatment.
Input carbon-containing feedstock into microwave transparent reaction chamber

Direct microwaves through the walls of the microwave transparent reaction chamber

Heat/irradiate the feedstock via the microwaves until depletion of the feedstock

Collect the products yielded by the feedstock

Figure 6
METHOD OF PRODUCING DROP-IN DIESEL

CROSS-REFERENCE TO RELATED APPLICATIONS

This application claims priority to U.S. Provisional Application No. 61/645,896, filed May 11, 2012 and titled “Method of Producing Drop-In Diesel,” the entirety of which is incorporated herein by reference.

FIELD OF THE INVENTION

The invention relates generally to the production of fuel from a hydrocarbon-containing biomass feedstock.

BACKGROUND OF THE INVENTION

The predicted shortage of fossil fuels and related attendant environmental concerns have attracted significant attention to the conversion of biomass into fuels, including renewable diesel.

First-generation renewable diesels were obtained from vegetable oils and animal fats. The direct use of straight vegetable oils as fuels in diesel engines was investigated. However, the high viscosity of straight vegetable oils caused a range of problems and lead to poor fuel characteristics. These problems were addressed through engine modifications, blending straight vegetable oils with fossil diesel, micro-emulsification, transesterification, and/or thermal cracking. The transesterification of oils to the corresponding fatty acid methyl esters (FAME) has been more widely applied because it is easy, cost-effective technology and leads to favorable physicochemical fuel parameters. Hydrotreating/cracking technology is another promising approach to convert vegetable oils and fats into quality fuel. This technology produces a diesel fuel with a chemical composition similar to that of conventional diesel (hydrocarbons). Both types of renewable diesel can replace petroleum diesel without the need for modification or adjustment of engines. In addition, renewable diesel with reduced CO₂ emissions offers significant environmental advantages, such as the lack of SOX emissions as a result of the absence of sulfur in the feedstock. In the case of FAME biodiesel, the higher oxygen content with respect to petroleum diesel ensures more complete combustion and a lower content of CO₂ hydrocarbons, and particulate emissions.

The continuing growth in demand of renewable diesel following several legislations in a number of countries that have mandated renewable diesel blends in transportation fuels has been a challenge to develop innovative and more efficient processes for producing renewable diesel from vegetable oils, including those that may be derived from biomass. Accordingly, it would be desirable to provide a renewable diesel derived from biomass in more efficient processes. The methods of the present invention are directed toward these, as well as other, important ends.

SUMMARY OF THE INVENTION

The invention in general is directed to methods for producing fuel, comprising:

forming pellets comprising at least one hydrocarbon-containing feedstock and at least one catalyst;

depolymerizing said hydrocarbon-containing feedstock in the presence of said catalyst using high-frequency microwave energy at a temperature of about 275°C to about 350°C to form a composition comprising:

- crude oil;
- biochar; and
- optionally, syngas comprising:
  - H₂;
  - CO;
  - CO₂;

reacting said crude oil with H₂ to remove impurities and to form an upgraded crude oil;

distilling said upgrading crude oil to form a phenol fraction and a diesel fraction.

The above summary is not intended to describe each embodiment or every implementation of the present invention. Advantages and attainments, together with a more complete understanding of the invention, will become apparent and appreciated by referring to the following detailed description and claims taken in conjunction with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

The accompanying drawings, which are included to provide a further understanding of the invention and are incorporated in and constitute a part of this specification, illustrate embodiments of the invention and together with the description serve to explain the principles of the invention. In the drawings:

FIG. 1 is a flow diagram of a process for generating fuel from hydrocarbon-containing feedstock in accordance with embodiments of the invention.

FIGS. 2A and 2B illustrate side and cross sectional views, respectively, of a system configured to convert carbon-containing materials to fuel in accordance with embodiments of the invention.

FIG. 2C is a diagram of a tilted reaction chamber system in accordance with embodiments of the invention.

FIG. 2D is a diagram of a side view of a reaction chamber system in accordance with embodiments of the invention.

FIG. 3A is a block diagram of a system that uses the reaction chamber systems illustrated in FIGS. 2A and 2B for water/air extraction and a reaction process in accordance with embodiments of the invention.

FIG. 3B illustrates a reaction system that includes feedback control in accordance with embodiments of the invention.

FIG. 4A shows a system which includes multiple stationary magnetrons arranged on a drum that is disposed outside a cylindrical reaction chamber having one or more microwave-transparent walls.

FIG. 4B illustrates a system having a drum supporting magnetrons which may be rotated around the longitudinal axis of the reaction chamber while the reaction chamber is concurrently rotated around its longitudinal axis.

FIG. 4C shows a reaction chamber with a feedstock transport mechanism comprising baffles.

FIG. 5 illustrates a system having a rotating magnetron in addition to a secondary heat source.

FIG. 6 is a flow diagram of a process for generating fuel from carbon-containing feedstock in accordance with embodiments of the invention.

While the invention is amenable to various modifications and alternative forms, specifics thereof have been shown by way of example in the drawings and will be described in detail below. It is to be understood, however, that the intention is not to limit the invention to the particular
embodiments described. On the contrary, the invention is intended to cover all modifications, equivalents, and alternatives falling within the scope of the invention as defined by the appended claims.

DETAILED DESCRIPTION OF THE INVENTION

As employed above and throughout the disclosure, the following terms, unless otherwise indicated, shall be understood to have the following meanings.

As used herein, the singular forms “a,” “an,” and “the” include the plural reference unless the context clearly indicates otherwise.

As used herein, “biomass” is biological material from living, or recently living organisms useful either directly or indirectly (through conversion) into an energy source. Biomass includes lignocellulosic biomass or a component part thereof, manure, and the like.

As used herein, “lignocellulosic biomass or a component part thereof” refers to plant biomass containing cellulose, hemicellulose, and lignin from a variety of sources, including, without limitation (1) agricultural residues (including corn stover and sugarcane bagasse), (2) dedicated energy crops, (3) wood and wood residues (including sawmill and paper mill discards), and (4) municipal waste, and their constituent parts including without limitation, lignocellulose biomass itself, lignin, C_6 saccharides (including cellulose, celllobiose, C_6 oligosaccharides, C_6 monosaccharides, and C_6 saccharides (including hemicellulose, C_5 oligosaccharides, and C_5 monosaccharides).

As used herein, “plant-based biomass” includes, but is not limited to, wood, energy grasses, algae, rapeseed, soybean, waste vegetable oil, agricultural waste (such as corn cobs, corn stover, sugarcane bagasse, orange rinds, and pea shells), timber industry waste (such as sawmill offcuts and sawdust), and the like.

As used herein, “animal-based biomass” includes, but is not limited to, animal husbandry waste, chicken litter, cow dung, slaughterhouse renderings, and the like.

As used herein, “municipal waste” includes, but is not limited to, landfill components such as sludge, construction and demolition waste, household waste, industrial waste, and the like.

As used herein, “industrial waste” includes, but is not limited to, carpet trimmings, used carpeting, and any other waste containing hydrocarbons or plastics, and the like.

As used herein, “agricultural waste” includes, but is not limited to, residual stalks, straw, leaves, roots, husk, nut or seed shells, waste wood, animal husbandry waste, chicken litter, cow dung, slaughterhouse renderings and the like.

As used herein with respect to fuel, “hydrotreatment” is a reaction process employing hydrogen in the presence of inorganic catalysts that converts the intermediate or product stream to a higher quality, oxygen-free fuel and removes unwanted compounds, including oxygen, sulfur, nitrogen, and unsaturated hydrocarbons, from the fuel.

The use of numerical values in the various quantitative values specified in this application, unless expressly indicated otherwise, are stated as approximations as though the minimum and maximum values within the stated ranges were both preceded by the word “about.” In this manner, slight variations from a stated value can be used to achieve substantially the same results as the stated value. Also, the disclosure of ranges is intended as a continuous range including every value between the minimum and maximum values recited as well as any ranges that can be formed by such values. Also disclosed herein are any and all ratios (and ranges of any such ratios) that can be formed by dividing a recited numeric value into any other recited numeric value. Accordingly, the skilled person will appreciate that many such ratios, ranges, and ranges of ratios can be unambiguously derived from the numerical values presented herein and in all instances such ratios, ranges, and ranges of ratios represent various embodiments of the present invention.

Accordingly, the invention is directed to methods for producing fuel, comprising:

1. forming pellets comprising at least one hydrocarbon-containing feedstock and at least one catalyst;
2. depolymerizing said hydrocarbon-containing feedstock in the presence of said catalyst using high-frequency microwave energy at a temperature of about 275°C. to about 350°C. to form a composition comprising:
3. crude oil;
4. biochar; and
5. optionally, syngas comprising:
6. H₂;
7. CO;
8. CO₂;
9. reacting said crude oil with H₂ to remove impurities and to form an upgraded crude oil; and
10. distilling said upgrading crude oil to form a phenol fraction and a diesel fraction.

One embodiment of the method of the invention is shown schematically in FIG. 1, where wood is the hydrocarbon-containing feedstock. A wide variety of other hydrocarbon-containing feedstocks may be substituted for the wood, as described herein.

The components of the process shown in FIG. 1 are described below:

<table>
<thead>
<tr>
<th>Number</th>
<th>Component</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>600</td>
<td>Feedstock Receiving and Preparation</td>
<td>This phase of the process is where the material(s) are received from the suppliers and cleaned before entering the pelletizing process.</td>
</tr>
<tr>
<td>601</td>
<td>Receiving</td>
<td>The materials are weighed and analyzed before being unloaded and stored. This process may include a method for payment to the suppliers. The computer program and scale management system are commercially available, such as, for example, from Rayouer.</td>
</tr>
<tr>
<td>602</td>
<td>Unloading and storage</td>
<td>The wood is unloaded by crane and stored before processing.</td>
</tr>
<tr>
<td>603</td>
<td>Debarking</td>
<td>This process removes the bark and other contaminants (such as soil and sand) from the wood.</td>
</tr>
</tbody>
</table>
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<table>
<thead>
<tr>
<th>Number</th>
<th>Component</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>604</td>
<td>Clean logs</td>
<td>The debarked logs are conveyed to the chipper.</td>
</tr>
<tr>
<td>605</td>
<td>Chipper</td>
<td>The logs are reduced to wood chips.</td>
</tr>
<tr>
<td>606</td>
<td>Bark</td>
<td>The bark is reduced in to a uniform size and conveyed to storage.</td>
</tr>
<tr>
<td>300</td>
<td>Pelletizing</td>
<td>This phase of the process the feedstock is reduced in size, excess moisture is removed, catalyst is added and the mixture converted into compressed into pellets.</td>
</tr>
<tr>
<td>301</td>
<td>Chip Storage</td>
<td>The wood chips (or any other feedstock) are conveyed to a storage pile (may be open or closed, silos).</td>
</tr>
<tr>
<td>302</td>
<td>Bark Storage</td>
<td>The sized bark is conveyed to a storage pile.</td>
</tr>
<tr>
<td>303</td>
<td>Size reduction</td>
<td>The feedstock is reduced in size using, for example, hammermills or similar equipment, to a size that is consistent with the pelletizing process.</td>
</tr>
<tr>
<td>304</td>
<td>Drying</td>
<td>Excess moisture is removed from the feedstock. Heat for the drying process is obtained from: 1. Combustion of the bark (606); 2. Heat that has been captured from the cooling of the biochar or the oils after distillation (402); 3. Combustion of the syngases (700).</td>
</tr>
<tr>
<td>305</td>
<td>Storage</td>
<td>The dry feedstock is stored in a covered container (silos).</td>
</tr>
<tr>
<td>306</td>
<td>Pelletizing</td>
<td>The feedstock is combined with a measured amount of catalyst (307) as it is fed into the pellet mills. This process includes all the necessary conveyors and a system to cool the pellets prior to storage.</td>
</tr>
<tr>
<td>308</td>
<td>Pellet Storage</td>
<td>This is the system to capture and cool the Biochar before it is stored.</td>
</tr>
<tr>
<td>400</td>
<td>Biochar System</td>
<td>Biochar is the charcoal that remains after the other hydrocarbons have been converted into syngas or crude oil.</td>
</tr>
<tr>
<td>401</td>
<td>Biochar</td>
<td>Biochar is cooled using a series of jacketed screw conveyors and a combination of thermal oil, water and mechanical cooling systems. Some of the heat is captured and utilized in the drying process (304).</td>
</tr>
<tr>
<td>402</td>
<td>Biochar Cooling</td>
<td>Biochar is stored before being shipped to other end users for consumption as a fuel or as a fertilizer.</td>
</tr>
<tr>
<td>403</td>
<td>Biochar Storage</td>
<td>Biochar is stored before being shipped to other end users for consumption as a fuel or as a fertilizer.</td>
</tr>
<tr>
<td>500</td>
<td>Refining Process</td>
<td>In this module of the process, the crude oil from the reactors is treated in a hydrotreatment unit. This upgraded crude is then passed through a distillation column where the various fractions are separated. Finally additives may be added to the diesel fraction to create transportation ready fuel. This process also allows for the creation of other fuels, such as aviation fuel, in addition or instead of diesel.</td>
</tr>
<tr>
<td>501</td>
<td>Crude Oil</td>
<td>After cooling in a condensation unit (241) from the reactor(s) the condensed liquid is what is referred to as crude oil.</td>
</tr>
<tr>
<td>502</td>
<td>Hydrotreating</td>
<td>The oil is processed through a hydrotreating unit. By contacting the crude oil with hydrogen gas and a suitable catalyst to remove impurities, such as oxygen, sulfur, nitrogen, and unsaturated hydrocarbons to form an upgraded crude oil.</td>
</tr>
<tr>
<td>503</td>
<td>Distillation</td>
<td>Different oil fractions vaporize at different temperatures. The distillation process utilizes this fact to separate the various fractions in the upgraded crude oil. Depending on the feedstock, the catalyst used and the reector &quot;recipe&quot; the composition of the crude oil will vary. Generally, the fractions will consist of mostly diesel fuel (about 80%) and other aromatic oils, including phenols.</td>
</tr>
<tr>
<td>504</td>
<td>Diesel Storage</td>
<td>There may be fuel storage tanks.</td>
</tr>
<tr>
<td>505</td>
<td>Phenol Storage</td>
<td>There may be storage tanks.</td>
</tr>
<tr>
<td>506</td>
<td>Blending</td>
<td>Certain additives (508) may be mixed with the diesel fuel to enable it to meet the ASTM standards for transportation fuel. If aviation fuel is produced, additives specific to that fuel may be added.</td>
</tr>
<tr>
<td>507</td>
<td>Fuel Storage</td>
<td>The fuel is stored in tanks before being shipped to the consumers.</td>
</tr>
<tr>
<td>700</td>
<td>Syngas</td>
<td>This is the non-condensable portion of the vapors that are collected from the reactors. This gas is combustible and may be used to provide heat or to fuel an electrical generator.</td>
</tr>
</tbody>
</table>
| 800    | Control System   | The entire system is designed with temperature, pressure, proximity and other monitoring and measuring devices. Information from all these devices is fed into a central control system that automatically or semi-automatically
controls the entire process. Software may manage and control the process.

<table>
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<td></td>
<td>controls the entire process. Software may manage and control the process.</td>
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</tbody>
</table>

[0055] In one embodiment, where wood is the hydrocarbon-containing feedstock, logs are unloaded and stacked. The logs are then debarked. The bark may be stored before being consumed as a fuel for heating or drying. The cleaned logs are chipped and may be stored before being sent for further size reduction and then to a dryer for moisture removal. Reclaimed heat may be used to assist in the drying of the wood chips. After drying, the bark and wood chips may be further reduced in size. The feedstock material is then mixed with a suitable catalyst and pelleted. The pellets are then optionally being preheated before being sent to the reactor for microwave catalytic depolymerization, which is described in detail below. Reclaimed heat may be used to assist in the preheating step. Three fractions are produced in the microwave catalytic depolymerization: crude oil, biochar, and syngas. The biochar may be cooled, where the reclaimed heat may be used in the drying or preheating steps, before storage. The syngas may be collected. The crude oil is then hydrotreated by contacting the crude oil with hydrogen gas and a suitable catalyst to remove impurities, such as oxygen, sulfur, nitrogen, and unsaturated hydrocarbons to form an upgraded crude oil. The upgraded crude oil is then distilled to form a phenol fraction and a diesel fraction. Both fractions may be stored. Additives (or fossil fuel) may be blended with the diesel fraction and then stored. The final fuel may be used as a drop-in fuel or as a blend with fossil fuels.

[0056] Suitable hydrotreating catalysts include, but are not limited to, CoMo catalysts and NiMo (http://www.topsoe.com/business_areas/refining/~media/PDF_files/Refining/, Topsoe_hydroprocessing_catalysts.aspx).

[0057] Embodiments of the invention are directed to methods for producing fuel from a hydrocarbon-containing feedstock, particularly solid feedstock, using microwave energy and a catalyst. The methods may employ at least one reaction chamber comprising at least one microwave-transparent chamber wall and a reaction cavity configured to hold the carbon-containing feedstock. A microwave subsystem may include at least one device configured to emit microwaves when energized. The microwave device is positioned relative to the reaction chamber so that the microwaves are directed through the microwave-transparent chamber wall and into the reaction cavity. The system also includes a mechanism that provides relative motion between the microwave device and the reaction chamber.

[0058] Another embodiment of the invention involves a process for converting a hydrocarbon-containing compound to fuel. A hydrocarbon-containing feedstock is input into a substantially microwave-transparent reaction chamber. Microwaves are directed from a microwave source through walls of the reaction chamber to impinge on the feedstock. The feedstock is microwaved until it reacts to produce a fuel.

[0059] While the present invention is capable of being embodied in various forms, the description herein of several embodiments is made with the understanding that the present disclosure is to be considered as an exemplification of the invention, and is not intended to limit the invention to the specific embodiments illustrated. Headings are provided for convenience only and are not to be construed to limit the invention in any manner. Embodiments illustrated under any heading may be combined with embodiments illustrated under any other heading.

[0060] The following description relates to approaches for processing the solid hydrocarbon-containing feedstock into fuels, such as, for example, drop-in fuels, by thermally and/or microwave enhanced reaction processes, including depolymerization. For example, depolymerization may be caused by heating or by exposure to microwaves. Depolymerization, also referred to as “cracking,” is a refining process that uses heat to break down (or “crack”) hydrocarbon molecules into shorter polymer chains which are useful as fuels. Depolymerization and other reaction processes may be enhanced by adding a catalyst to the feedstock which increases the speed of the reaction and/or reduces the temperature and/or the radiation exposure required for the processes. Furthermore, the catalyst, such as zeolite, has a nanostructure which allows only molecules of a certain size to enter the crystalline grid or activate the surface areas of the catalyst and to interact with the catalyst. Thus, the catalyst advantageously is very effective at controlling the product produced by the reaction processes because only substances having a specified chain length may be produced using the catalytic process. Catalytic depolymerization is particularly useful for transforming biomass and other carbon-containing feedstock into fuels usable as transportation or heating fuels.

[0061] In certain embodiments, the method further comprises:

[0062] blending said diesel fraction with at least one additive.

Suitable additives include, but are not limited to pH adjusting agents, emission reduction additives, lubricants, cetane enhancers, and the like. Other suitable additives may be found at http://www.epa.gov/otaq/regs/fuels/additive/web-dies.htm, the entire disclosure of which is incorporated herein by reference.

[0063] In certain embodiments, the method further comprises:

[0064] blending said diesel fraction with at least one fossil fuel.

In certain embodiments, the fuel produced may be used as a drop-in fuel without blending said diesel fraction with a fossil fuel.

[0065] In certain embodiments, the method further comprises:

[0066] wherein said depolymerizing step is carried out at a temperature of about 275°C. to about 350°C., preferably, at a temperature of about 285°C.

[0067] In certain embodiments, the pellets have a density sufficient to permit said depolymerizing step and said hydrotreating step without substantially mechanically degrading said pellets.

[0068] In certain embodiments, the method further comprises:

[0069] drying said hydrocarbon-containing feedstock prior to forming said pellets.
In certain embodiments, the method further comprises: combusting at least a portion of said syngas in said drying step. In certain embodiments, syngas or non-condensable gasses are formed. Generally, these are available for combustion to generate electricity or to support the heating needs of the process. The amount and composition of the syngases will depend on the particular feedstock. In certain embodiments, the method further comprises: recovering heat from said biochar to form a cooled biochar. In certain embodiments, the method further comprises: storing said cooled biochar. The biochar forms a sterile matter containing mainly carbon and non-organic substances (such as, for example, minerals and salts) originally present in the feedstock. Some of these materials may be further processed. For example, the biochar may be processed and used as fertilizer. In certain embodiments, the method further comprises: storing said phenol fraction. In certain embodiments, said phenol fraction comprises methoxyphenol. In certain embodiments, the method further comprises: storing said diesel fraction. In certain embodiments, said catalyst is a zeolite. Suitable zeolites include, but are not limited to, hydrogen-exchanged zeolites and the like. In certain embodiments, said hydrocarbon-containing feedstock is selected from the group consisting of plant-based biomass, animal-based biomass, tires, municipal waste, industrial waste, and combinations thereof, as defined above. In certain embodiments, said plant-based biomass is selected from the group consisting of lignocellulosic biomass or a component thereof, algae, and combinations thereof, as defined above. One aspect of efficient depolymerization (and other reactions) is the ability to heat and/or irradiate the feedstock substantially uniformly to the temperature that is sufficient to cause depolymerization as well as activate the catalyst. Upon depolymerization, long hydrocarbon chains “crack” into shorter chains. Microwave heating has been shown to be particularly useful in heating systems for thermal depolymerization. Heating systems such as flame, steam, and/or electrical resistive heating, heat the feedstock by thermal conduction through the reaction chamber wall. These heating systems operate to heat the feedstock from the outside of the reaction chamber walls to the inside of the feedstock, whereas microwaves heat from the inside of the feedstock toward the reaction chamber walls. Using non-microwave heating sources, the heat is transferred from the heat source outside wall to the inside of the vessel wall that is in direct contact with the feedstock mixture. The heat is then transferred to the surfaces of the feedstock and then transferred, again, through the feedstock until the internal areas of the feedstock are at a temperature near the temperature of the reaction chamber wall. Embodiments of the invention are directed to a reaction chamber system that can be used to process any carbon-containing feedstock, whether gas, solid and/or liquid, to extract the volatile organic compounds in the feedstock at a temperature range that will produce transportation fuels. The reaction chamber can also act in a reactive mode that will convert gaseous carbon feedstock into different gaseous chemical species. The system involves a heating system that enhances uniform heating and/or microwave irradiation throughout the feedstock, thereby increasing the efficiency of the reaction.

Some reaction chamber systems described herein use microwave heating to achieve more uniform temperature within the feedstock, and, as a result, a more efficient reaction. Microwaves are absorbed by the water molecules in the material that is irradiated in the microwave. When the water molecules absorb the microwaves, the molecules vibrate, which creates heat by friction, and the heat is conducted to the surrounding material. The methods of the invention employ microwave radiation and a catalyst to produce oil by cracking or depolymerizing hydrocarbon-containing feedstock. In certain embodiments, a reaction chamber system described herein takes advantage of microwave irradiation and/or heating in processing feedstock that contains hydrocarbon and can be converted to transportation and heating fuels. Such reactors are described, for example, in US 2011/0036706, the entire disclosure of which is incorporated herein by reference. The reactor may be made from a substantially microwave-transparent substance such as quartz, a glass-like material that is substantially transparent to microwave radiation. Because quartz can be manipulated into many shapes, it provides design discretion for shaping the reaction chamber, but in one example the reaction chamber is configured in the shape of a tube or cylinder. The cylindrical shape allows for the feedstock to feed in one end and exit at the opposite end. An example of a suitable reaction chamber would be a quartz tube that is about four feet long with a wall thickness of $\frac{3}{8}$". Microwave radiation may be generated by a magnetron or other suitable device. One or more microwave producing devices, such as, for example, magnetrons can be mounted external to the quartz tube wall. Magnetrons come in different power ranges and can be controlled by computers to irradiate the processing feedstock with the proper energy to convert the feedstock to most desirable fuel products efficiently. In one application, the magnetron can be mounted on a cage that would rotate around the outside of the reactor tube as well as travel the length of the reactor tube. Feedstock traveling through the length of the inside of the tube will be traveling in a plug flow configuration and can be irradiated by fixed and/or rotating magnetrons. A computer may be used to control the power and/or other parameters of the microwave radiation so that different feedstock, with different sizes and densities can be irradiated at different parameter settings specific to the feedstock and thus convert the feedstock more efficiently. These configurations of a reactor will allow efficient processing of feedstocks, from relatively pure feedstock streams to mixed feedstock streams that include feedstocks of different densities, moisture contents, and chemical composition. Efficiencies can occur because the fuel products are extracted from the reactor chamber as they are vaporized from the feedstock, but further processing of the remaining feedstock occurs until different fuel products are vaporized and extracted. For example, dense feedstock, such as plastics, take longer to process into a useable fuel than less dense
feedstock, such as foam or wood chips. The system described herein continues to process dense feedstock without over-processing the earlier converted products from the less dense feedstock. This is accomplished by using both stationary and rotating microwave generators. [0092] One example of a mixed feedstock would be unsorted municipal solid waste. In some implementations, catalyst may be added to the feedstock which helps in the conversion of the feedstock as well as the speed at which the conversion can progress. A catalyst can be designed to react at the preset processing temperature inside the reactor or to react with the impinging microwave radiation. In some embodiments, no catalyst is required. In other embodiments, the catalyst may be a rationally designed catalyst for a specific feedstock.

[0093] The plug flow configuration with the reactors described herein will allow adjustments to the residence time that the feedstock resides within the reactor core for more efficient exposure to the heat and the radiation of the microwaves to produce the desired end products.

[0094] Inlets and/or outlets, such as, for example, quartz inlets and/or outlets can be placed along the walls of the reaction chamber to allow for pressure and/or vacuum control. The inlets and outlets may allow the introduction of inert gases, reactive gases and/or the extraction of product gases.

[0095] Thus, the design of the microwave-transparent reaction chamber, the use of microwaves as a heating and/or radiation source with fixed and/or rotating magnetrons, plug flow processing control, with or without the use of catalysts, will allow the processing of any carbon-containing feedstock in any physical phase.

[0096] A system useful in the various embodiments of the methods of the invention includes a reaction chamber having one or more substantially microwave-transparent walls and a microwave heating/radiation system. The microwave heating/radiation system is arranged so that microwaves generated by the heating/radiation system are directed through the substantially microwave-transparent walls of the reaction chamber and into the reaction cavity where the feedstock material is reacted without substantially heating the walls of the reaction chamber. To enhance the temperature uniformity of the feedstock, the reaction chamber and the heating/radiation system may be in relative motion, such as, for example, relative rotational and/or translational motion. In some implementations, the heating system may rotate around a stationary reaction chamber. In some implementations, the feedstock within the reaction chamber may rotate by the use of flights with the heating/radiation system remaining stationary. In some implementations, the reaction chamber may rotate with the heating system remaining stationary. In yet other implementations, both the reaction chamber and the heating/radiation system may rotate, such as, for example, in countercurrent, opposing directions. To further increase temperature uniformity, the system may include a mechanism for stirring and/or mixing the feedstock material within the reaction chamber. The reaction chamber may be tilted during reaction process, for example, to force the feedstock to go through the catalytic bed.

[0097] FIGS. 2A and 2B illustrate side and cross sectional views, respectively, of a system 100 for converting carbon-containing feedstock to fuel in accordance with embodiments of the invention. Although the reaction chamber 110 may be any suitable shape, the reaction chamber 110 is illustrated in FIGS. 2A and 2B as a cylinder having a cylindrical wall 111 that is substantially transparent to microwaves in the frequency range and energy used for the reaction process. The reaction chamber 110 includes a reaction cavity 112 enclosed by the cylindrical wall 111. The system 100 includes a transport mechanism 118 configured to move the feedstock through the reaction chamber. The operation of the system 100 with regard to the reactions taking place within the reaction chamber 110 may be modeled similarly to that of a plug flow reactor.

[0098] As illustrated in FIG. 2A, system includes a transport mechanism 118 for moving the feedstock material through the reaction chamber 110. The transport mechanism 118 is illustrated as a screw auger, although other suitable mechanisms, such as, for example, conveyer, may also be used. The transport mechanism 118 may further provide for mixing the feedstock within the reaction chamber. In some embodiments, the reaction chamber wall 111 may have a thickness of about ⅛ inches. The smoothness of the reaction chamber wall 111 facilitates the movement of the feedstock through the reaction chamber 110. A heating/radiation subsystem 115 may include any type of heating and/or radiation sources, but preferably includes a microwave generator 116 such as a magnetron that is configured to emit microwaves 113 having a frequency and energy sufficient to heat the carbon-containing feedstock to a temperature sufficient to facilitate the desired reaction of the feedstock, for example, for depolymerization of the feedstock, microwaves in a frequency range of about 0.3 GHz to about 300 GHz may be used. For example, the operating power of the magnetrons may be in the range of about 1 Watt to 500 kilowatts. The magnetron 116 is positioned in relation to the reaction chamber 110 so that the microwaves 113 are directed through the wall 111 of the reaction chamber 110 and into the reaction cavity 112 to heat and/or irradiate the material therein. A mechanism 117 provides relative motion between the magnetron 116 and the reaction chamber 110 along and/or around the longitudinal axis 120 of the reaction chamber 110. In some embodiments, the mechanism 117 may facilitate tilting the reaction chamber 110 and/or the magnetron 116 at an angle 0 (FIG. 2C) to facilitate the reaction of the feedstock and/or the extraction of gases, for example. In the embodiment illustrated in FIGS. 2A-C, the magnetron 116 is positioned on a rotational mechanism 117, such as a rotatable cage or drum, that rotates the magnetron 116 around the stationary reaction chamber 110. In some implementations, the rotation around the chamber may not be complete, but the rotation path may define an arc around the circumference of the reaction chamber. The rotation may occur back and forth along the path of the arc. As previously mentioned, in some embodiments, the reaction chamber 110 may be the rotating component, or both the heating/radiation subsystem 116 and the reaction chamber 110 may rotate, such as, for example, in opposing, countercurrent directions. The rotation between the reaction chamber and the magnetron provides more even heating and more even microwave exposure of the feedstock within the reaction cavity 112, thus enhancing the efficient reaction chemistry of the feedstock and/or other processes that are temperature/radiation dependent, such as removal of water from the feedstock. The rotation lessens the temperature gradient and/or maintains a more constant microwave flux across the plug inside the reaction chamber.

[0099] The reaction chamber 110 may include one or more entry ports 120, such as, for example, quartz entry ports, configured to allow the injection or extraction of substances
into the reaction cavity 112. In one implementation, the quartz ports may be used to extract air and/or oxygen from the reaction cavity. Extraction of air and/or oxygen may be used to suppress combustion which is desirable for some processes.

For example, in certain embodiments, the system 100 may be used to preprocess the feedstock through compression and/or removal of air and/or water. In this application, gases such as hydrogen and/or nitrogen may be injected through one or more ports 120 to hydrogenate and/or suppress combustion of the feedstock. The reaction chamber 110 may also include one or more exit ports 121, such as, for example, quartz exit ports, configured to allow passage of water, water vapor, air, oxygen and/or other substances and/or by-products from the reaction chamber 110.

FIG. 2D is a diagram illustrating a reaction chamber system 150 for producing fuel from carbon-containing feedstock in accordance with embodiments of the invention. The system 150 includes an input hopper (also referred to as a load hopper) 151 configured to allow introduction of the feedstock material into the system 150. A gearmotor auger drive 152 provides a drive system for the auger 153 that transports the feedstock through the system 150. As the feedstock is compressed in the load hopper 151, air is extracted through the atmosphere outlet 154. A seal 155 isolates the load hopper 151 from the reaction chamber 156 to maintain a level of vacuum. The reaction chamber 156 includes walls of a microwave-transparent material. One or more stationary microwave heads 157 are positioned at the walls of the reaction chamber 156. In addition, the system 150 includes one or more rotating microwave heads 158. In one implementation, each rotating microwave head is located at a fixed position with respect to the longitudinal axis 160 of the reaction chamber 156. The rotating microwave head is mounted on a slipping bearing 159 that allows the microwave head to rotate around the reaction chamber 156. In some implementations the rotating microwave head(s) 158 may rotate around the longitudinal axis 160 of the reaction chamber 156 as well as moving back and forth along the longitudinal axis 160. The system 150 includes a seal at the exit of the reaction chamber 156 to maintain the reaction chamber vacuum.

FIG. 3A is a block diagram of a system 200 that uses one or more of the reaction chamber systems 100 illustrated in FIGS. 2A and 2B. The reaction chamber systems 220, 230 may be arranged and/or operated in series or in a parallel configuration. The extraction process 220 and the reaction process 230 depicted in FIGS. 3A and 3B are illustrated as occurring in two separate reaction chambers, such as, for example, that operate at different temperatures. Alternatively, the extraction process and the reaction process may be implemented in a single reaction chamber with two separate zones, such as, for example, two separate temperature zones.

In the system 200 of FIG. 3, one or both of the water/air extraction subsystem 220 and the reaction subsystem 230 may be similar to the reaction chamber system 100 of FIGS. 2A and 2B. Hydrocarbon-containing feedstock, such as manure, wood chips, plant-based cellulose, tires, and/or municipal solid waste, enters the system through a hopper 211, and traverses an airlock 212 to enter a feedstock preparation module 213. If needed, a catalyst, such as zeolite, and/or other additives that enhance the reaction process, for example to adjust the pH, may be introduced into the system 200 through the input hopper 211 and/or the entry ports (shown in FIG. 2B). In the feedstock preparation module 213, the feedstock material is shredded to a predetermined particle size may be dependent on the properties of the feedstock, such as the purity, density, and/or chemical composition of the feedstock. If used, the catalyst may be added at the time that the feedstock is being prepared so that the catalyst is evenly dispersed within the feedstock material before entering the reaction chamber 231. In general, the less uniform the feedstock, the smaller the particle size needed to provide efficient reaction.

After the initial feedstock preparation stages 600 and 300, the shredded and mixed feedstock is transported by a transport mechanism 215 into the reaction chamber 221 of the next stage of the process. The air/water extraction subsystem 220, which performs the optional processes of water and/or extraction prior to the reaction process, has similarity to the system illustrated in FIGS. 2A and 2B, includes a heating/radiation module 222 comprising at least one magnetron 223 configured to generate microwaves 226 which may be mounted on a rotational or stationary mechanism 227. If mounted on a rotational mechanism, the mechanism rotates the magnetron 223 either partially or fully around the reaction chamber 221 as the microwaves 226 are directed through the wall 224 of the reaction chamber 221 and into the reaction cavity 225 impinging on and heating the feedstock therein. In some embodiments, the heating module 222 may utilize only one magnetron 223 or only two or more magnetrons without using other heat/radiation sources.

In some embodiments, the heating/radiation module 222 may utilize one magnetron 223 in addition to other heat sources, such as heat sources that rely on thermal conduction through the wall of the reaction chamber, such as, for example, flame, steam, electrical resistive heating, recycled heat from the process, and/or other heat sources. During the air and/or water extraction process, the feedstock may be heated to at least 100°C, the boiling point of water, to remove excess water from the feedstock. The excess water (such as, for example, in the form of steam) and/or other substances may exit the reaction chamber 221 via one or more exit ports. Additives to the feedstock, such as inert and/or reactive gases including hydrogen and/or nitrogen, may be introduced via one or more input ports into the reaction chamber 221 of the water/air extraction process. In addition to being heated and/or irradiated by microwaves, the feedstock may also be subjected to a pressurized atmosphere and/or a vacuum atmosphere and/or may be mechanically compressed to remove air from the reaction chamber 221.

After the optional air and/or water extraction process, the transport mechanism 215 moves the feedstock to the next processing stage 230 which involves the reaction process, such as, for example, thermal depolymerization, of the feedstock. After the feedstock/catalyst mixture enters the reaction chamber 231, it is heated to a temperature that is sufficient to facilitate the desired reaction. For example, to produce drop-in fuel through depolymerization, a temperature of in a range of about 265°C to about 295°C is used to crack the hydrocarbons in the feedstock into shorter chains. In addition to being heated, the feedstock may also be subjected to a pressurized atmosphere, a vacuum atmosphere and/or may be mechanically compressed in the reaction chamber 231.

In some embodiments, heating/radiation in the reaction chamber 231 is accomplished using a magnetron 233 emitting microwaves 236. The magnetron 233 may rotate relative to the reaction chamber 231. As previously described
in connection with the water extraction stage 220, the rotating magnetron 233 may be supported by rotational mechanism 237, such as a cage or drum. The rotational mechanism 237 allows relative rotational motion between the magnetron 233 and the reaction chamber 231. For example, the magnetron 233 may rotate completely around the reaction chamber 231 or the rotation of the magnetron 233 may proceed back and forth along an arc that follows the circumference of the reaction chamber 231. The rotating magnetron heating system 233 may be supplemented using a stationary magnetron, and/or other conventional heat sources such as a flame or electrical resistive heating. Rotating the magnetron 233 provides more even heating/radiation of the feedstock material and catalyst within the reaction cavity 235 and enhances the heating properties over that of stationary heat sources.

0108] In certain embodiments, the cracked hydrocarbons vaporize and are collected in a condenser 241 and liquefy and then are sent to the hydrotreater 502 and then to the hydrotreater (not shown) and then to the distiller 240 to the drop-in fuel. In other embodiments, the vapors may hydrotreated directly without first liquefying. Heavier, longer chain hydrocarbon molecules may be recycled back to the reaction chamber. In some implementations, distillation may not be necessary, and the fuel product only needs to be filtered.

0109] In some configurations, it is desirable to control the processes of the reaction to allow a higher efficiency of fuel extraction from the feedstock. FIG. 3B is a block diagram of a system 205 that includes the system components described in connection with FIG. 3A along with a feedback control system 250. The illustrated feedback control system 250 includes a controller 251 and one or more sensors 252, 253, 254 which may be configured to sense parameters at various stages during the process. The feedback control system 250 may include sensors 252 at the feedstock preparation stage which are configured to sense parameters of the feedstock and/or feedstock preparation process. For example, the sensors 252, may sense the chemical composition of the feedstock, density, moisture content, particle size, energy content or other feedstock parameters. The sensors 252 may additionally or alternatively sense the conditions within the feedstock preparation chamber, such as, for example, flow, pressure, temperature, humidity, composition of the gases present in the chamber, etc. The sensors 252 develop signals 255a that are input to the controller electronics 251 where they are analyzed to determine the condition of the feedstock and/or the feedstock preparation process. In response to the sensed signals 255a, the controller 251 develops feedback signals 255b which control the operation of the feedstock preparation module 213. For example, in some implementations, the controller 251 may control the feedstock preparation module 213 to continue to shred and/or grind the feedstock material until a predetermined particle size and/or a predetermined particle size variation is detected. In another example, based on the sensed chemical composition of the feedstock, the controller 251 may cause a greater or lesser amount of catalyst to be mixed with the feedstock or may cause different types of catalyst to be mixed with the feedstock.

0110] The control system 250 may also develop feedback signals 256b, 257b to control the operation of the water extraction module 220 and/or the reaction module 230, respectively, based on sensed signals 256a, 257a. For example, the sensors 253, 254 may sense the temperature of the water extraction and/or reaction processes and the controller 251 may develop feedback signals 256b, 257b to control the operation of the heating/radiation systems 222, 232, such as, for example, power, frequency, pulse width, rotational or translational velocity, etc. of one or both of the magnetrons 223, 233. The controller 251 may develop feedback signals to the magnetrons to control the amount of radiation impinging on the feedstock so that the feedstock will not be over- or undercooked and development of hot spots will be avoided. The controller 250 may control the operation of various sensors into one or both of the extraction chamber and/or the reaction chamber 221, 231 through the entry ports to control the processes taking place within the chambers 221, 231. The residue of the depleted feedstock (char) is sent to a storage unit. After the distillation stage, the heavy hydrocarbons may be recycled back into the reaction chamber and the lighter hydrocarbons may be sent on to a polymerization stage.

0111] As previously discussed, the reaction chambers may be made of quartz, glass, ceramic, plastic, and/or any other suitable material that is substantially transparent to microwaves in the frequency and energy range of the reaction processes. In some configurations, the heating/radiation systems described herein may include one or more magnetrons that rotate relative to the reaction chamber. In some embodiments, the magnetrons may be multiple and/or may be stationary. FIG. 4A illustrates a system 300 which includes multiple stationary magnetrons 311 arranged on a drum 312 that is disposed outside a cylindrical reaction chamber 313 having one or more microwave-transparent walls. The operation of the magnetrons may be continuous, or may be pulsed, such as, for example, in a multiplexed pattern. In some embodiments (FIG. 4B), the drum 312 supporting the magnetrons 311 may be rotated 320 around the longitudinal axis 350 of the reaction chamber 313 and/or the reaction chamber 313 may be rotated 330 around its longitudinal axis 350.

0112] A feedstock transport mechanism may be disposed within the reaction chamber. For example, as illustrated in FIG. 4C, the feedstock transport mechanism may comprise one or more baffles 361 that are configured to move the feedstock through the reaction chamber 360 as the reaction chamber rotates. The baffles 361 may be mounted to the walls of the reaction chamber 360 and/or may be otherwise installed within the reaction chamber to provide movement of feedstock within and through the reaction chamber 360, such as, for example, longitudinally through the reaction chamber.

0113] In some embodiments, illustrated in FIG. 5, one or more secondary heat sources 450, such as a flame, steam, and/or electric resistive heating, or recycled heat, may be used in addition to magnetrons 416, which are stationary, or are supported on a mechanism 417 that rotates around the circumference of the reaction chamber 420. In some configurations, the magnetrons 416 may not make a complete revolution around the reaction chamber 420, but may rotate back and forth 419 along an arc that follows the circumference of the reaction chamber 420.

0114] FIG. 6 is a flow chart illustrating a process for producing fuel from a carbon-containing feedstock in accordance with embodiments of the invention. A carbon-containing feedstock, such as biomass, municipal solid waste, plant material, wood chips and the like is input 610 to a reaction chamber having walls that are substantially transparent to microwaves used to heat and/or irradiate the feedstock. The feedstock may be a gas, liquid and/or solid matter. The heating and/or radiation occur(s) by directing 620 the microwave
energy through the walls of the reaction chamber so that it impinges on the feedstock disposed within the reaction chamber. The feedstock is heated/irradiated by the microwaves, optionally in the presence of a catalyst, until reaction of the carbon-containing molecules occurs to produce the desirable end fuel product. The fuel product created by the reaction processes are collected.

While the preferred forms of the invention have been disclosed, it will be apparent to those skilled in the art that various changes and modifications may be made that will achieve some of the advantages of the invention without departing from the spirit and scope of the invention. Therefore, the scope of the invention is to be determined solely by the claims to be appended.

When ranges are used herein for physical properties, such as molecular weight, or chemical properties, such as chemical formulae, all combinations, and subcombinations of ranges specific embodiments therein are intended to be included.

The disclosures of each patent, patent application, and publication cited or described in this document are hereby incorporated herein by reference, in their entirety.

Those skilled in the art will appreciate that numerous changes and modifications can be made to the preferred embodiments of the invention and that such changes and modifications can be made without departing from the spirit of the invention. It is, therefore, intended that the appended claims cover all such equivalent variations as fall within the true spirit and scope of the invention.

What is claimed is:

1. A method for producing a fuel, comprising:
   - forming pellets comprising at least one hydrocarbon-containing feedstock and at least one catalyst;
   - depolymerizing said hydrocarbon-containing feedstock in the presence of said catalyst using high-frequency microwave energy at a temperature of about 275°C and 350°C to form a composition comprising: crude oil; biochar; and optionally, syngas comprising:
     - H₂;
     - CO; and optionally, CO₂;
   - reacting said crude oil with H₂ to remove impurities and to form an upgraded crude oil; and distilling said upgrading crude oil to form a phenol fraction and a diesel fraction.

2. A method of claim 1, further comprising: blending said diesel fraction with at least one additive.

3. A method of claim 1, further comprising: blending said diesel fraction with at least one fossil fuel.

4. A method of claim 1, wherein said depolymerizing step is carried out at a temperature of about 280°C.

5. A method of claim 1, wherein said pellets have a density sufficient to permit said depolymerizing step and said hydrotreating step without substantially mechanically degrading said pellets.

6. A method of claim 1, further comprising: drying said hydrocarbon-containing feedstock prior to forming said pellets.

7. A method of claim 6, further comprising: combusting at least a portion of said syngas in said drying step.

8. A method of claim 1, further comprising: recovering heat from said biochar to form a cooled biochar.


10. A method of claim 1, further comprising: storing said phenol fraction.

11. A method of claim 1, wherein said phenol fraction comprises methoxyphenol.

12. A method of claim 1, further comprising: storing said diesel fraction.

13. A method of claim 1, wherein said catalyst is a zeolite.

14. A method of claim 1, wherein said hydrocarbon-containing feedstock is selected from the group consisting of plant-based biomass, animal-based biomass, tires, municipal waste, industrial waste, and combinations thereof.

15. A method of claim 14, wherein said plant-based biomass is selected from the group consisting of lignocellulosic biomass or a component part thereof, algae, and combinations thereof.

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