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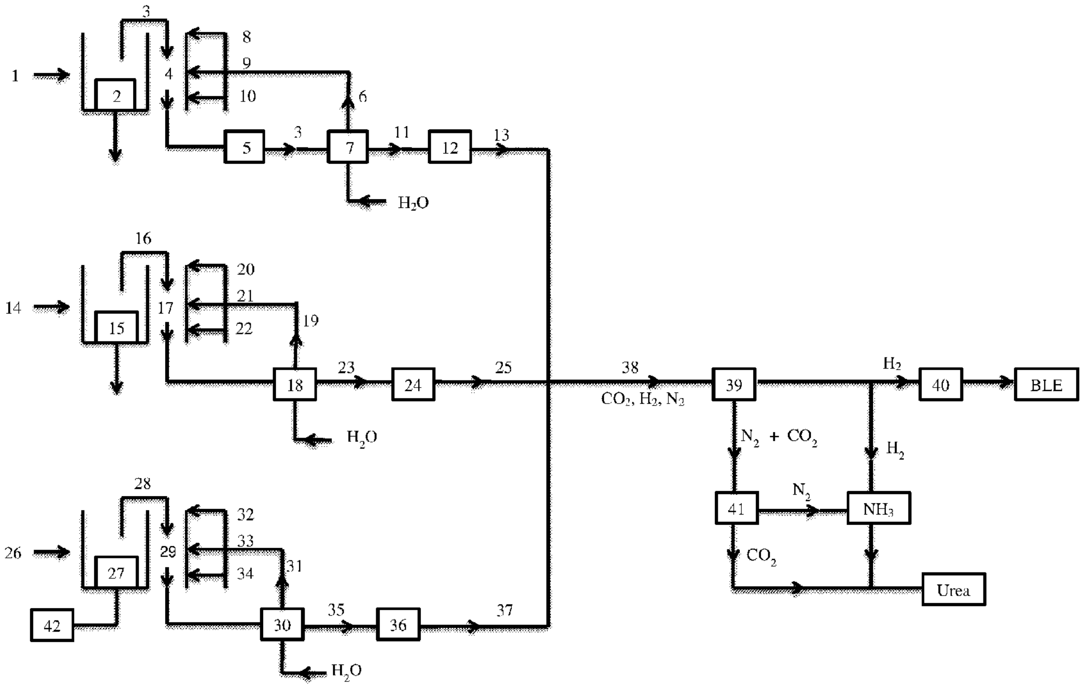

Figure 1

(57) Abstract: The invention is broadly directed to a continuous recycling process comprising: providing a homogenised waste material; forming the homogenised waste material into briquettes having a consistent size and density; feeding the briquettes into a first reactor and pyrolysing the briquettes so as to provide carbon and syngas, wherein heat from the syngas is used to drive further pyrolysis. The syngas may be subjected to a high-temperature water-gas shift reaction to enrich the hydrogen continent of the syngas, and scrubbed to remove contaminants, wherein heat is then exchanged from the syngas with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction and a cooled syngas.

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A process for converting waste into clean energy and value-added products

Field of the Invention

The present invention broadly relates to a process for converting waste, in particular carbonaceous waste, into clean energy and value-added products.

Background of the Invention

Across the globe the amount of waste being generated continues to grow at an alarming rate. In 2012, the worlds' cities generated 1.3 billion tonnes of solid waste. With rapid population growth and urbanization, municipal waste generation is expected to rise to 2.2 billion tonnes by 2025.

Residents in developing countries, especially the urban poor, are more severely impacted by unsustainably managed waste. In some countries waste is often disposed in unregulated dumps or openly burned. These practices create serious health, safety, and environmental consequences. Poorly managed waste serves as a breeding ground for disease vectors and contributes to global climate change through methane generation.

For many years waste has been converted into energy in so-called "waste-to-energy plants". A waste-to-energy plant is a waste management facility that combusts waste in order to produce electricity. However, following the combustion process, further waste is generated. This waste is typically in the form of slag which is disposed in landfill and air pollution control residues. Furthermore, waste-to-energy plants require a large input of energy to sustain the temperatures required to combust the waste.

Against this background there is a need for improved processes for converting waste into energy.

Summary of the Invention

In a first aspect the present invention provides a continuous recycling process comprising:

(i) providing a homogenised waste material;

(ii) forming the homogenised waste material into briquettes having a consistent size and density;

(iii) feeding the briquettes into a first reactor and pyrolysing the briquettes so as to provide carbon and syngas, wherein heat from the syngas is used to drive further pyrolysis;

(iv) subjecting the syngas to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the syngas;

(v) scrubbing the syngas to remove contaminants;

(vi) exchanging heat from the syngas with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (iv) and a cooled syngas;

(vii) subjecting the cooled syngas to a low-temperature water-gas shift reaction so as to provide a first gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(viii) transferring the carbon obtained following step (iii) into a second reactor and further heating the carbon in the presence of oxygen-enriched air so as to provide a second gas mixture comprising carbon monoxide and nitrogen, wherein heat from the second gas mixture is used to heat further carbon in step (viii);

(ix) subjecting the second gas mixture to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the second gas mixture;

(x) exchanging heat from the second gas mixture with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (ix) and a cooled second gas mixture;

(xi) subjecting the cooled second gas mixture to a low-temperature water-gas shift reaction so as to provide a third gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(xii) transferring the carbon obtained following step (viii) into a third reactor and further heating the carbon in the presence of oxygen-enriched air so as to provide a fourth gas mixture comprising carbon monoxide and nitrogen, wherein heat from the fourth gas mixture is used to heat further carbon in step (xii);

(xiii) subjecting the fourth gas mixture to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the fourth gas mixture;

(xiv) exchanging heat from the fourth gas mixture with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (xiii) and a cooled fourth gas mixture;

(xv) subjecting the cooled fourth gas mixture to a low-temperature water-gas shift reaction so as to provide a fifth gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(xvi) separating the hydrogen, carbon dioxide and nitrogen in the first, third and fifth gas mixtures, wherein at least a portion of the hydrogen separated is used to produce base load electricity, at least a portion of the nitrogen separated is used to produce ammonia and at least a portion of the carbon dioxide separated is used to produce urea; and,

(xvii) recovering the carbon obtained following step (xii).

The waste material may comprise a carbonaceous material.

The carbonaceous material may comprise plastic or a mixture of different plastics.

The carbonaceous material may comprise tyres.

The carbonaceous material may comprise coal fines.

The carbonaceous material may comprise biomass, such as for example a fibrous biomass.

The carbonaceous material may comprise municipal waste.

In some embodiments, the waste material comprises palm oil waste or coconut fibre.

The briquettes may be produced by extrusion.

The briquettes may have a density between about 500 kg/m^3 and about 1000 kg/m^3, or between about 600 kg/m^3 and about 900 kg/m^3, or between about 650 kg/m^3 and about 750 kg/m^3. In one embodiment the briquettes have a density of about 750 kg/m^3.

The briquettes may further comprise one or more of: sewage, wood pulp or fines.

The briquettes may comprise at least 20% moisture (w/w).

The briquettes may be pyrolysed at a temperature between about 500 °C and about 1200 °C, or between about 600 °C and about 1100 °C, or between about 700 °C and about 1100 °C, or between about 800 °C and about 1000 °C, or about 900 °C.

The high-temperature water-gas shift reaction in step (iv) may be performed by mixing the syngas with superheated steam.

The high-temperature water-gas shift reactions in steps (ix) and (xiii) may be performed by mixing the second gas mixture and the fourth gas mixture respectively with superheated steam.

The high-temperature water-gas shift reaction in steps (iv), (ix) and (xiii) may be performed at a temperature between about 500 °C and about 1200 °C, or at a temperature between about 500 °C and about 1100 °C, or at a temperature between about 500 °C and about 1000 °C, or at a temperature between about 600 °C and about 900 °C, or at a temperature between about 600 °C and about 800 °C, or at a temperature between about 650 °C and about 750 °C.

Exchanging heat from the syngas with water so as to provide superheated steam may be performed after step (v).

In step (v) the syngas may be scrubbed to remove one or more of the following contaminants: dioxins, heavy metals, furans and acid gases.

The cooled syngas, cooled second gas mixture and cooled fourth gas mixture may have a temperature below about 300 °C, or below about 275 °C, or below about 250 °C, or between about 200 °C and about 300 °C, or about 250 °C.

The low-temperature water-gas shift reaction in step (vii) may be performed by mixing the cooled syngas with superheated steam.

The low-temperature water-gas shift reactions in steps (xi) and (xv) may be performed by mixing the cooled second gas mixture and the cooled fourth gas mixture respectively with superheated steam.

The low-temperature water-gas shift reaction in steps (vii), (xi) and (xv) may be performed at a temperature above about 250 °C, or above about 300 °C, or above about 350 °C, or between about 250 °C and about 500 °C, or between about 250 °C and about 400 °C, or between about 250 °C and about 350 °C.

In steps (viii) and (xii) the carbon may be heated at a temperature between about 500 °C and about 1200 °C, or between about 700 °C and about 1200 °C, or between about 600 °C and about 1100 °C, or between about 700 °C and about 1100 °C, or between about 800 °C and about 1000 °C, or about 900 °C.

The oxygen-enriched air may have an oxygen content of at least about 23% (w/w), at least about 25% (w/w), at least about 27% (w/w), at least about 30% (w/w), at least about 35% (w/w), or at least about 50% (w/w).

The carbon dioxide, hydrogen and nitrogen may be separated using a positive swing adsorption (PSA) system.

The first, third and fifth gas mixtures may be combined prior to step (xvi).

The first, second and third reactors may be vertically stacked such that step (iii) is carried out above step (viii) which is carried out above step (xii).

In step (iii), the briquettes may be rotated during heating.

In steps (viii) and (xii) the carbon may be rotated during heating.

The first reactor may comprise a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein syngas produced in the reactor is fed into the conduit so as to transfer heat to the reaction chamber to drive further pyrolysis.

The second reactor may comprise a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein the gas mixture produced in the second reactor is fed into the conduit so as to transfer heat to the reaction chamber to heat further carbon.

The third reactor may comprise a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein the gas mixture produced in the third reactor is fed into the conduit so as to transfer heat to the reaction chamber to heat further carbon.

In the first, second and third reactors the inner wall may have a thermal conductivity that is about 15 times to 25 times, or about 20 times larger than the thermal conductivity of the outer wall.

The thermal conductivity of the inner wall may be between about 250 W/(m.K) and about 350 W/(m.K), or between about 300 W/(m.K) and about 350 W/(m.K). In one embodiment the thermal conductivity of the inner wall may be about 327 W/(m.K).

The inner wall may comprise, or be made of, a copper/ceramic material.

The superheated steam generated in steps (vi), (x) and (xiv) may be injected into the conduit of each of the first, second and third reactors at a plurality of spaced apart points thereby enabling the high-temperature water-gas shift reaction.

Carbon generated in steps (iii), (viii) and (xii) may be one or more of: biochar, carbon black or activated carbon.

Time elapsed from commencement of pyrolsis of a briquette to removal of carbon produced from the briquette from the third reactor may be between about 60 minutes and about 120 minutes, or between about 70 minutes and about 110 minutes, or between about 80 minutes and about 100 minutes, or about 90 minutes.

The first, third and fifth gas mixtures may be free, or substantially free, of carbon monoxide.

Definitions

The following are some definitions that may be helpful in understanding the description of the present invention. These are intended as general definitions and should in no way limit the scope of the present invention to those terms alone, but are put forth for a better understanding of the following description.

Throughout this specification, unless the context requires otherwise, the word "comprise", or variations such as "comprises" or "comprising", will be understood to imply the inclusion of a stated element, integer or step, or group of elements, integers or steps, but not the exclusion of any other element, integer or step, or group of elements, integers or steps.

The terms "a" and "an" are used herein to refer to one or to more than one (i.e. to at least one) of the grammatical object of the article. By way of example, "an element" means one element or more than one element.

In the context of this specification the term "about" is understood to refer to a range of numbers that a person of skill in the art would consider equivalent to the recited value in the context of achieving the same function or result.

In the context of the present specification, the term "substantially free" is understood to mean less than about 1%, or less than about 0.5%, or less than about 0.1%, or less than about 0.01 %, or less than about 0.005%, or less than about 0.001 %, or less than about 0.0001 % of the recited entity.

Brief Description of the Drawings

Figure 1: Schematic diagram of a process in accordance with one embodiment of the invention.

Detailed Description of the Invention

In one aspect the present invention provides a continuous recycling process comprising:

(i) providing a homogenised waste material;

(ii) forming the homogenised waste material into briquettes having a consistent size and density;

(iii) feeding the briquettes into a first reactor and pyrolysing the briquettes so as to provide carbon and syngas, wherein heat from the syngas is used to drive further pyrolysis;

(iv) subjecting the syngas to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the syngas;

(v) scrubbing the syngas to remove contaminants;

(vi) exchanging heat from the syngas with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (iv) and a cooled syngas;

(vii) subjecting the cooled syngas to a low-temperature water-gas shift reaction so as to provide a first gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(viii) transferring the carbon obtained following step (iii) into a second reactor and further heating the carbon in the presence of oxygen-enriched air so as to provide a second gas mixture comprising carbon monoxide and nitrogen, wherein heat from the second gas mixture is used to heat further carbon in step (viii);

(ix) subjecting the second gas mixture to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the second gas mixture;

(x) exchanging heat from the second gas mixture with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (ix) and a cooled second gas mixture;

(xi) subjecting the cooled second gas mixture to a low-temperature water-gas shift reaction so as to provide a third gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(xii) transferring the carbon obtained following step (viii) into a third reactor and further heating the carbon in the presence of oxygen-enriched air so as to provide a fourth gas mixture comprising carbon monoxide and nitrogen, wherein heat from the fourth gas mixture is used to heat further carbon in step (xii);

(xiii) subjecting the fourth gas mixture to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the fourth gas mixture;

(xiv) exchanging heat from the fourth gas mixture with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (xiii) and a cooled fourth gas mixture;

(xv) subjecting the cooled fourth gas mixture to a low-temperature water-gas shift reaction so as to provide a fifth gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(xvi) separating the hydrogen, carbon dioxide and nitrogen in the first, third and fifth gas mixtures, wherein at least a portion of the hydrogen separated is used to produce base load electricity, at least a portion of the nitrogen separated is used to produce ammonia and at least a portion of the carbon dioxide separated is used to produce urea; and,

(xvii) recovering the carbon obtained following step (xii).

The process of the present invention allows a wide range of waste materials to be converted to energy and value-added products. The process differs from traditional waste-to-energy processes in that emissions are used as an energy source to drive the process and carbon is used as a catalyst to produce hydrogen. Importantly, the process results in zero emissions and can therefore be descried as a "closed-loop" process.

The process generates substantial quantities of hydrogen, nitrogen and carbon dioxide which are used to produce base load electricity and value-added products, i.e., ammonia and urea. Ammonia has been suggested as a possible fuel for electric cars and urea is used extensively as a fertiliser.

A portion of the hydrogen produced is used to produce base-load electricty. This may be achieved, for example, by using a solid oxide fuel cell.

The process also generates carbon, which is typically in the form of biochar, activated carbon or carbon black. Each of these forms of carbon are widely used in industry. Biochar is used as a soil amendment and may increase soil fertility of acidic soils, increase agricultural productivity and provide protection against some foliar and soil-borne diseases. Carbon black is used as a filler in tyres and other rubber products and as a colour pigment in plastics, paints and inks. Activated carbon is used in a wide range of applications, including methane and hydrogen storage, air purification, decaffeination, gold purification, metal extraction, water purification, sewage treatment and teeth whitening.

The type of carbon produced depends on the nature of the waste material. As such, the waste material may be selected based on the type of carbon that is sought to be produced. For example, where the waste material is tyres, a substantial quantity of carbon black is produced. Alternatively, where the waste material is a fibrous biomass, such as cocunut fibre, a substantial quantity of activated carbon is produced. Municipal solid waste can be used where it is desired to produce biochar.

Waste materials suitable for use in the process include any and all carbonaceous materials, including but not limited to, plastic, biomass, municipal waste, coal fines and tyres. Plastic pollution remains a major global challenge. The present invention provides a convenient and efficient means for converting large amounts of plastic waste into energy and value-added products.

Step (i) involves providing a homogenised waste material. The waste material may comprise a single type of waste or different types of waste and be homogenised using, for example, a blender.

In step (ii) the homogenised waste material is formed into briquettes. The briquettes may be about the same size and the same density, thereby having a consistent carbon content. The consistent carbon content allows for consistent carbon monoxide production which translates into consistent hydrogen production following the initial high-temperature water-gas shift reaction. This provides the process with the necessary energy balance required for continuous operation and the production of base-load electricity. The briquettes may have a density between about 400 kg/m^3 and about 1000 kg/m^3, or between about 500 kg/m^3 and about 1000 kg/m^3, or between about 550 kg/m^3 and about 1000 kg/m^{3,} or between about 550 kg/m^3 and about 950 kg/m^3, or between about 550 kg/m^3 and about 900 kg/m^3, or between about 600 kg/m^3 and about 900 kg/m^3, or between about 600 kg/m^3 and about 850 kg/m^3, or between about 650 kg/m^3 and about 750 kg/m^3.

The briquettes are then fed into a first reactor and pyrolysed so as to provide carbon and a syngas which typically comprises carbon dioxide, carbon monoxide, hydrogen, hydrocarbons and contaminants, including dioxins, heavy metals, furans and acid gases.

The briquettes may be pyrolysed at a temperature between about 400 °C and about 1200 °C, or at a temperature between about 500 °C and about 1200 °C, or at a temperature between about 500 °C and about 1100 °C, or at a temperature between about 550 °C and about 1100 °C, or at a temperature between about 550 °C and about 1000 °C, or at a temperature between about 600 °C and about 1100 °C, or at a temperature between about 600 °C and about 1000 °C, or at a temperature between about 700 °C and about 1100 °C, or at a temperature between about 700 °C and about 1000 °C, or at a temperature between about 800 °C and about 1000 °C, or at a temperature between about 850 °C and about 1000 °C, or at a temperature between about 850 °C and about 950 °C. In some embodiments, the briquettes are pyrolysed at a temperature of about 900 °C.

Heat from the syngas produced is used to drive further pyrolysis. This may be achieved using a jacketed reactor whereby syngas produced in the reactor is extracted and fed into an outer gas jacket. Heat from the syngas is transferred from the outer jacket to the reaction chamber where pyrolysis takes place.

In one embodiment the first reactor comprises a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein syngas produced in the reactor is fed into the conduit so as to transfer heat to the reaction chamber to drive further pyrolysis.

The inner wall may have a thermal conductivity that is about 15 times to 25 times, or about 20 times larger than the thermal conductivity of the outer wall. This arrangement optimises heat transfer from the syngas to the reaction chamber. The inner wall may comprise, or be made of, a copper/ceramic material.

In step (iv) the syngas is subjected to a high-temperature water-gas shift reaction. The reaction may be carried out by mixing the syngas with superheated steam. The high-temperature water-gas shift reaction serves to enrich the amounts of hydrogen and carbon dioxide in the syngas stream at the expense of carbon monoxide.

The high-temperature water-gas shift reaction may be performed at a temperature between about 500 °C and about 1200 °C, or at a temperature between about 500 °C and about 1100 °C, or at a temperature between about 500 °C and about 1000 °C, or at a temperature between about 600 °C and about 900 °C, or at a temperature between about 600 °C and about 800 °C, or at a temperature between about 650 °C and about 750 °C.

In step (v) the syngas is scrubbed to remove contaminants including dioxins, heavy metals, furans, acid gases and particulates. Those skilled in the art will be familiar with methods for removing contaminants from a syngas stream.

Heat from the syngas is utilised by exchanging with water so as to provide superheated steam that is used in the high-temperature water-gas shift reaction in step (iv). Typically, the heat exchange is performed after scrubbing of the syngas. The heat exchange also provides a cooled syngas. The cooled syngas may have a temperature below about 300 °C, or below about 275 °C, or below about 250 °C, or between about 200 °C and about 300 °C, or about 250 °C.

When pyrolysis is carried out in a jacketed reactor, the superheated steam generated in step (vi) may be injected at a plurality of spaced apart points along the conduit thereby enabling the high-temperature water-gas shift reaction. This arrangement allows additional heat produced in the high-temperature water-gas shift reaction to be transferred to the reaction chamber to drive further pyrolysis.

In step (vii) the cooled syngas is subjected to a low-temperature water-gas shift reaction in order to remove any residual carbon monoxide. The carbon monoxide may be present as a result of methane being reduced to carbon monoxide and hydrogen in the high-temperature water-gas shift reaction according to the following reaction:

$$CH_4 + \text{superheated steam} \rightarrow CO + 3H_2$$

The low-temperature water-gas shift reaction may be performed at a temperature above about 250 °C, or above about 300 °C, or above about 350 °C, or between about 250 °C and about 500 °C, or between about 250 °C and about 400 °C, or between about 250 °C and about 350 °C.

The combination of high-temperature water-gas shift and low-temperature water-gas shift reactions optimises the forward direction of the following reaction:

$$CO + H_2O \leftrightarrow CO_2 + H_2$$

In step (viii) the carbon generated from pyrolysis is transferred to a second reactor and heated in the presence of oxygen-enriched air so as to provide a second gas mixture comprising carbon monoxide and nitrogen. The oxygen-enriched air may be obtained from the positive swing adsorption system. In some embodiments the oxygen-enriched air may have an oxygen content of at least about 23% (w/w), at least about 25% (w/w), at least about 27% (w/w), at least about 30% (w/w), at least about 35% (w/w), or at least about 50% (w/w).

Heat from the second gas mixture produced is used to heat further carbon in step (viii). This may be achieved using a jacketed reactor as described above, whereby gas produced in the reactor is extracted and fed into an outer gas jacket. Heat from the second gas mixture is transferred from the outer jacket to the reaction chamber where heating of the carbon takes place.

In one embodiment the second reactor comprises a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein gas produced in the second reactor is fed into the conduit so as to transfer heat to the reaction chamber to heat further carbon.

As per the first reactor, the inner wall of the second may have a thermal conductivity that is about 15 times to 25 times, or about 20 times larger than the thermal conductivity of the outer wall. This arrangement optimises heat transfer from the syngas to the reaction chamber. The inner wall may comprise, or be made of, a copper/ceramic material.

In step (ix) the second gas mixture is subjected to a high-temperature water-gas shift reaction. The reaction may be carried out by mixing the syngas with superheated steam. Again, the high-temperature water-gas shift reaction serves to enrich the amounts of hydrogen and carbon dioxide in the second gas mixture at the expense of carbon monoxide.

The high-temperature water-gas shift reaction in step (ix) may be performed at the temperatures described above in connection with step (iv).

Heat from the second gas mixture is utilised by exchanging with water so as to provide superheated steam that is used in the high-temperature water-gas shift reaction in step (ix). The heat exchange also provides a cooled second gas mixture. The cooled second gas mixture may have a temperature below about 300 °C, or below about 275 °C, or between about 250 °C, or between about 200 °C and about 300 °C, or about 250 °C.

When step (viii) is carried out in a jacketed reactor, the superheated steam generated in step (x) may be injected at a plurality of spaced apart points along the outer gas jacket thereby enabling the high-temperature water-gas shift reaction. This arrangement allows additional heat produced in the high-temperature water-gas shift reaction to be transferred to the reaction chamber to heat further carbon.

In step (xi) the cooled second gas mixture is subjected to a low-temperature water-gas shift reaction. This reaction serves to further enrich the amounts of carbon dioxide and hydrogen in the second gas mixture at the expense of carbon monoxide so as to provide the third gas mixture. The third gas mixture may be free, or substantially free, of carbon monoxide. The low-temperature water-gas shift reaction may be performed at a temperature above about 250 °C, or above about 300 °C, or about 350 °C, or between about 250 °C and about 500 °C, or between about 250 °C and about 400 °C.

In step (xii) the carbon generated following step (viii) is transferred to a third reactor and heated in the presence of oxygen-enriched air so as to provide a fourth gas mixture comprising carbon monoxide and nitrogen. The oxygen-enriched air may be obtained from the positive swing adsorption system. Steps (ix) to (xi) are then repeated as described above as steps (xiii) to (xv).

Following steps (vii), (xi) and (xv) the carbon dioxide, hydrogen and nitrogen are separated. Separation may be achieved using a positive swing adsorption system. In some embodiments, the gas mixtures obtained in steps (vii), (xi) and (xv) may be combined prior to step (xvi).

The first, second and third reactors may be vertically stacked such that step (iii) is carried out above step (viii) which is carried out above step (xii). In this arrangement, following step (iii) the carbon is dropped into the second reactor and following step (viii) the carbon is dropped into the third reactor.

By varying the dwell time of the briquettes and carbon in each reactor the amounts of carbon and hydrogen produced may be controlled. The dwell time may be varied by varying the rate of rotation of the briquettes and carbon. It has been found that increasing the rate of rotation maximises the amount of carbon produced and minimises the amount of hydrogen produced. Conversely, decreasing the rate of rotation minimises the amount of carbon produced and maximises the amount of hydrogen produced.

In some embodiments, the dwell time calculated from commencement of pyrolsis of a briquette to removal of carbon from the third reactor may be between about 60 minutes and about 120 minutes, or between about 70 minutes and about 110 minutes, or between about 80 minutes and about 100 minutes, or about 90 minutes.

Figure 1 shows a schematic diagram of a process in accordance with one embodiment of the invention. Jacketed pyrolysis reactor **1** is provided comprising briquette **2**. On heating, briquette **2** is pyrolysed liberating syngas **3** which is extracted and passes through the conduit **4** of the jacketed reactor **1**. Superheated steam **6** is injected into the conduit **4** at a plurality of spaced apart points **8**, **9** and **10** along the conduit **4** where mixing with syngas **3** occurs thereby enabling the high-temperature water-gas shift reaction. Following the high-temperature water-gas shift reaction syngas **3** is scrubbed in scrubber **5** to remove contaminants. The syngas **3** is then passed through heat exchanger **7** so as to provide superheated steam **6** which is piped to points **8**, **9** and **10** for injection into the conduit **4**, and cooled syngas **11**. The cooled syngas **11** is then subjected to a low temperature water-gas shift reaction **12** to provide the first gas mixture **13**, which comprises carbon dioxide, nitrogen and hydrogen.

Following pyrolysis, the resulting carbon **15** is dropped into jacketed reactor **14** where heating in oxygen-rich air is performed. On heating, carbon **15** liberates the second gas mixture **16** which is extracted and passes through the conduit **17** of the jacketed reactor **14**. Superheated steam **19** is injected into the conduit **17** at a plurality of spaced part points **20**, **21** and **22** along the conduit **17** where mixing with the second gas mixture **16** occurs thereby enabling the high-temperature water-gas shift reaction. The second gas mixture **16** is then passed through heat exchanger **18** so as to provide superheated steam **19** which is piped to points **20**, **21** and **22**, for injection into the conduit **17**, and the cooled second gas mixture **23**. The cooled second gas mixture **23** is then subjected to a low temperature water-gas shift reaction **24** to provide the third gas mixture **25**, which comprises carbon dioxide, nitrogen and hydrogen.

The resulting carbon **27** is dropped into jacketed reactor **26** where heating in oxygen-rich air is performed. On heating, carbon **27** liberates the fourth gas mixture **28** which is extracted and passes through the conduit **29** of the jacketed reactor **26**. Superheated steam **31** is injected into the conduit **29** at a plurality of spaced part points **32**, **33** and **34** along the conduit **29** where mixing with the fourth gas mixture **28** occurs thereby enabling the high-temperature water-gas shift reaction. The fourth gas mixture **28** is then passed through heat exchanger **30** so as to provide superheated steam **31** which is piped to points **32**, **33** and **34**, for injection into the conduit **29**, and the cooled fourth gas mixture **35**. The cooled second gas mixture **35** is then subjected to a low temperature water-gas shift reaction **36** to provide the fifth gas mixture **37**, which comprises carbon dioxide, nitrogen and hydrogen.

The first gas mixture **13**, third gas mixture **25** and fifth gas mixture **37** are then combined into a single gas mixture **38**. The single gas mixture **38** is passed through PSA **39** which separates the single gas mixture **38** into a gas stream comprising hydrogen gas and a gas stream comprising nitrogen gas and carbon dioxide gas. A portion of the hydrogen gas is directed to a single oxide fuel cell **40** for generation of base load electricity (BLE). The gas stream comprising nitrogen gas and carbon dioxide gas is passed through PSA **41** which separates the mixture into nitrogen and gas and carbon dioxide gas. A portion of the hydrogen gas is reacted with the nitrogen gas to give ammonia (NH_3). A portion of the ammonia is reacted with the carbon dioxide to provide urea. Carbon **42** (in the form of one or more of biochar, activated carbon and carbon black) is recovered from the jacketed reactor **26**.

The citation of any reference herein should not be construed as an admission that such reference is available as prior art to the present application. Further, the reference in this specification to any prior publication (or information derived from it), or to any matter which is known, is not, and should not be taken as an acknowledgement or admission or any form of suggestion that that prior publication (or information derived from it) or known matter forms part of the common general knowledge in the field of endevour to which this specification relates.

Those skilled in the art will appreciate that the invention described herein is susceptible to variations and modifications other than those specifically described. It is to be understood that the invention includes all such variations and modifications.

CLAIMS:

1. A continuous recycling process comprising:

(i) providing a homogenised waste material;

(ii) forming the homogenised waste material into briquettes having a consistent size and density;

(iii) feeding the briquettes into a first reactor and pyrolysing the briquettes so as to provide carbon and syngas, wherein heat from the syngas is used to drive further pyrolysis;

(iv) subjecting the syngas to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the syngas;

(v) scrubbing the syngas to remove contaminants;

(vi) exchanging heat from the syngas with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (iv) and a cooled syngas;

(vii) subjecting the cooled syngas to a low-temperature water-gas shift reaction so as to provide a first gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(viii) transferring the carbon obtained following step (iii) into a second reactor and further heating the carbon in the presence of oxygen-enriched air so as to provide a second gas mixture comprising carbon monoxide and nitrogen, wherein heat from the second gas mixture is used to heat further carbon in step (viii);

(ix) subjecting the second gas mixture to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the second gas mixture;

(x) exchanging heat from the second gas mixture with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (ix) and a cooled second gas mixture;

(xi) subjecting the cooled second gas mixture to a low-temperature water-gas shift reaction so as to provide a third gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(xii) transferring the carbon obtained following step (viii) into a third reactor and further heating the carbon in the presence of oxygen-enriched air so as to provide a fourth gas mixture comprising carbon monoxide and nitrogen, wherein heat from the fourth gas mixture is used to heat further carbon in step (xii);

(xiii) subjecting the fourth gas mixture to a high-temperature water-gas shift reaction so as to enrich the hydrogen content of the fourth gas mixture;

(xiv) exchanging heat from the fourth gas mixture with water so as to provide superheated steam which is used in the high-temperature water-gas shift reaction in step (xiii) and a cooled fourth gas mixture;

(xv) subjecting the cooled fourth gas mixture to a low-temperature water-gas shift reaction so as to provide a fifth gas mixture comprising hydrogen, nitrogen and carbon dioxide;

(xvi) separating the hydrogen, carbon dioxide and nitrogen in the first, third and fifth gas mixtures, wherein at least a portion of the hydrogen separated is used to produce base load electricity, at least a portion of the nitrogen separated is used to produce ammonia and at least a portion of the carbon dioxide separated is used to produce urea; and,

(xvii) recovering the carbon obtained following step (xii).

2. The process of claim 1, wherein the waste material comprises a carbonaceous material.

3. The process of claim 2, wherein the carbonaceous material comprises plastic or a mixture of different plastics.

4. The process of claim 2, wherein the carbonaceous material comprises tyres or coal fines.

5. The process of claim 2, wherein the carbonaceous material comprises biomass.

6. The process of claim 5, wherein the biomass is palm oil waste or coconut fibre.

7. The process of claim 2, wherein the carbonaceous material comprises municipal waste.

8. The process of any one of claims 1 to 7, wherein the briquettes are produced by extrusion.

9. The process of any one of claims 1 to 8, wherein the briquettes have a density between about 500 kg/m^3 and about 1000 kg/m^3, or between about 600 kg/m^3 and about 900 kg/m^3, or between about 650 kg/m^3 and about 750 kg/m^3.

10. The process of any one of claims 1 to 9, wherein the briquettes further comprise one or more of: sewage, wood pulp or fines.

11. The process of any one of claims 1 to 10, wherein the briquettes are pyrolysed at a temperature between about 500 °C and about 1200 °C, or between about 600 °C and

about 1100 °C, or between about 700 °C and about 1100 °C, or between about 800 °C and about 1000 °C, or about 900 °C.

12. The process of any one of claims 1 to 11, wherein the high-temperature water-gas shift reactions in steps (iv), (ix) and (xiii) are performed at a temperature between about 500 °C and about 1200 °C, or at a temperature between about 500 °C and about 1100 °C, or at a temperature between about 500 °C and about 1000 °C, or at a temperature between about 600 °C and about 900 °C, or at a temperature between about 600 °C and about 800 °C, or at a temperature between about 650 °C and about 750 °C.

13. The process of any one of claims 1 to 12, wherein exchanging heat from the syngas with water so as to provide superheated steam may be performed after step (v).

14. The process of any one of claims 1 to 13, wherein in step (v) the syngas is scrubbed to remove one or more of the following contaminants: dioxins, heavy metals, furans and acid gases.

15. The process of any one of claims 1 to 14, wherein the cooled syngas, cooled second gas mixture and cooled fourth gas mixture have a temperature below about 300 °C, or below about 275 °C, or below about 250 °C, or between about 200 °C and about 300 °C, or about 250 °C.

16. The process of any one of claims 1 to 15, wherein the low-temperature water-gas shift reaction in step (vii) is performed by mixing the cooled syngas with superheated steam.

17. The process of any one of claims 1 to 16, wherein the low-temperature water-gas shift reactions in steps (xi) and (xv) are performed by mixing the cooled second gas mixture and the cooled fourth gas mixture respectively with superheated steam.

18. The process of any one of claims 1 to 17, wherein the low-temperature water-gas shift reaction in steps (vii), (xi) and (xv) is performed at a temperature above about 250 °C, or above about 300 °C, or above about 350 °C, or between about 250 °C and about 500 °C, or between about 250 °C and about 400 °C, or between about 250 °C and about 350 °C.

19. The process of any one of claims 1 to 18, wherein in steps (viii) and (xii) the carbon is heated at a temperature between about 500 °C and about 1200 °C, or between about 700 °C and about 1200 °C, or between about 600 °C and about 1100 °C, or between about 700 °C and about 1100 °C, or between about 800 °C and about 1000 °C, or about 900 °C.

20. The process of any one of claims 1 to 19, wherein the oxygen-enriched air may have an oxygen content of at least about 23% (w/w), at least about 25% (w/w), at least about 27% (w/w), at least about 30% (w/w), at least about 35% (w/w), or at least about 50% (w/w).

21. The process of any one of claims 1 to 20, wherein the carbon dioxide, hydrogen and nitrogen are separated using a positive swing adsorption system.

22. The process of any one of claims 1 to 21, wherein the first, third and fifth gas mixtures are combined prior to step (xvi).

23. The process of any one of claims 1 to 22, wherein the first, second and third reactors are vertically stacked such that step (iii) is carried out above step (viii) which is carried out above step (xii).

24. The process of any one of claims 1 to 23, wherein in step (iii), the briquettes are rotated during heating.

25. The process of any one of claims 1 to 24, wherein in steps (viii) and (xii) the carbon is rotated during heating.

26. The process of any one of claims 1 to 25, wherein the first reactor comprises a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein syngas produced in the reactor is fed into the conduit so as to transfer heat to the reaction chamber to drive further pyrolysis.

27. The process of any one of claims 1 to 26, wherein the second reactor comprises a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein the gas mixture produced in the second reactor is fed into the conduit so as to transfer heat to the reaction chamber to heat further carbon.

28. The process of any one of claims 1 to 27, wherein the third reactor comprises a reaction chamber and an outer gas jacket attached to at least a portion of the reaction chamber, the outer gas jacket comprising an inner wall and an outer wall defining a conduit, wherein the gas mixture produced in the third reactor is fed into the conduit so as to transfer heat to the reaction chamber to heat further carbon.

29. The process of any one of claims 26 to 28, wherein in the first, second and third reactors the inner wall has a thermal conductivity that is about 15 times to about 25 times, or about 20 times, larger than the thermal conductivity of the outer wall.

30. The process of claim 29, wherein the thermal conductivity of the inner wall is between about 250 W/(m.K) and about 350 W/(m.K), or between about 300 W/(m.K) and about 350 W/(m.K).

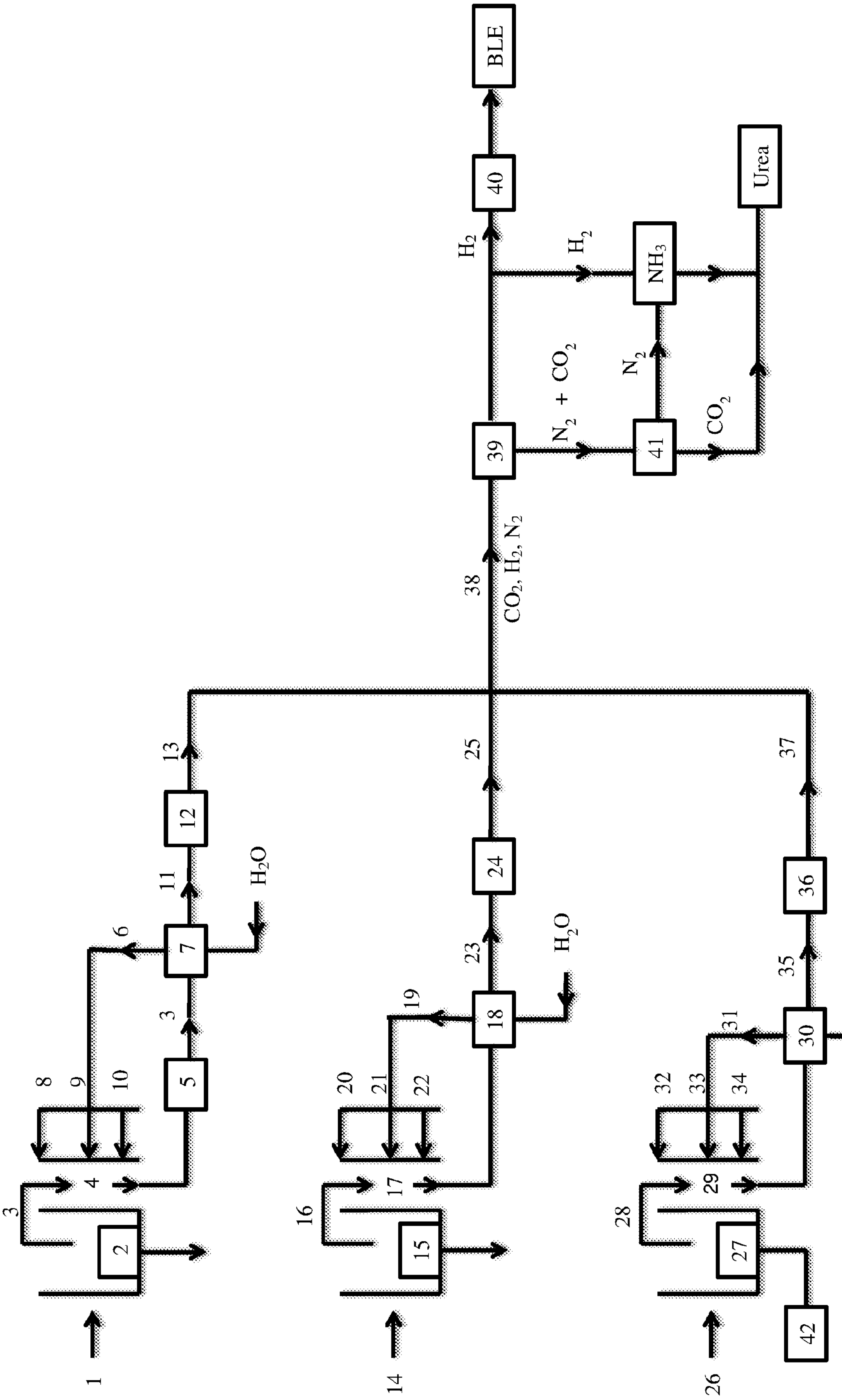
31. The process of any one of claims 26 to 30, wherein the inner wall comprises, or is made of, a copper/ceramic material.

32. The process of any one of claims 26 to 31, wherein the superheated steam generated in steps (vi), (x) and (xiv) is injected into the conduit of each of the first, second and third reactors at a plurality of spaced apart points thereby enabling the high-termperature water-gas shift reaction.

33. The process of any one of claims 1 to 32, wherein carbon generated in steps (iii), (viii) and (xii) is one or more of: biochar, carbon black or activated carbon.

34. The process of any one of claims 1 to 33, wherein time elapsed from commencement of pyrolsis of a briquette to removal of carbon produced from the briquette from the third reactor may be between about 60 minutes and about 120 minutes, or between about 70 minutes and about 110 minutes, or between about 80 minutes and about 100 minutes, or about 90 minutes.

35. The process of any one of claims 1 to 34, wherein the first, third and fifth gas mixtures may be free, or substantially free, of carbon monoxide.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H_2O
42
38
CO_2, H_2, N_2
39
N_2 + CO_2
41
N_2
CO_2
H_2
40
BLE
H_2
NH_3
Urea

Figure 1

A. CLASSIFICATION OF SUBJECT MATTER

C10K 3/04 (2006.01) ***C01B 3/02 (2006.01)*** ***C10K 1/00 (2006.01)*** ***C01B 3/02 (2006.01)*** ***C01B 3/32 (2006.01)***
C01C 1/04 (2006.01)

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)

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)

Database: PATENW: IPCs & CPCs: C01B3/34/LOW, C01B3/32/LOW, C01B3/36/LOW, C10J2300/0956/LOW, C10J3/721/LOW, C10J2300/0926/LOW, C10J2300/0926/LOW, C10J2300/0943/low, C10J2300/094/low, C10J2300/1637/LOW, C10K3/04/LOW, C07C273/10/LOW, C01C1/04/LOW, C10J2300/0913/LOW, C10J2300/1643/LOW, C10J2300/1618/LOW, C10J2300/1603/LOW, C10J2300/1612/LOW, Y02E20/18/LOW, C10J2300/1643/low, C10J2300/1884/low, C10J2300/1892/low, C10J2300/1656/low, C10J2300/1668/low, C10J2300/1671/low, C10J2300/1861/low; and key words: high-temp, low-temp, water-gas, carbon, wood, briquette, biomass, pyrolysis, scrub, clean, waste and similar terms. Google Patents & ESPACENET with key words: pyrolysis, water-gas, waste, biomass, hydrogen, ammonia, briquettes, carbon, oxygen, nitrogen, urea, electricity, syngas, scrub, air, low-temp, high temp and similar terms. Applicant and Inventor names searched in ESPACENET and AUSPAT internal databases provided by IP Australia.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	Documents are listed in the continuation of Box C	

☒ 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	"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
"D"	document cited by the applicant in the international application		
"E"	earlier application or patent but published on or after the international filing date	"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
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"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
"O"	document referring to an oral disclosure, use, exhibition or other means	"&"	document member of the same patent family
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search 25 February 2022	Date of mailing of the international search report 25 February 2022
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaustralia.gov.au	**Authorised officer** Santosh Gupta AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. +61262837947

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	US 2009/0324460 A1 (ROBINSON ET AL.) 31 December 2009	
X	Abstract; paras [0005], [0013], [0029]-[0032], [0044], [0088], [0167]-[0168], [0184], [0226]-[0227], [0244], [0270]-[0271], [0275]; claim 1	1-20, 22, 26-32, 34-35
Y	Abstract; paras [0005], [0013], [0029]-[0032], [0044], [0088], [0167]-[0168], [0184], [0226]-[0227], [0244], [0270]-[0271], [0275]; claim 1	21, 23-25, 33
	US 2010/0297001 A1 (GUYOMARC'H) 25 November 2010	
Y	Abstract; paras [0042]-[0046]	23-25
	WO 2007/134075 A2 (ALCHEMIX CORPORATION) 22 November 2007	
Y	Abstract; page 2, line 14-page 3, line 2; page 3, lines 24-28; page 7, lines 10-11; page 12, lines 13-15; page 17, lines 1-3; page 18, lines 15-24; page 27, lines 7-14; page 38, lines 8-11; page 41, line 3-page 42, line 6; page 44, lines 13-14; page 49, lines 23-28; claims 8, 29, 37-38	21, 33
	US 2017/0152452 A1 (TESNA ENERGY CO., LTD.) 01 June 2017	
A	Abstract	1-8, 34-35

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/AU2021/051532

This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document/s Cited in Search Report		**Patent Family Member/s**	
Publication Number	**Publication Date**	**Publication Number**	**Publication Date**
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		AU 2009262073 A1	30 Dec 2009
		AU 2009262073 B2	06 Sep 2012
		CA 2729003 A1	30 Dec 2009
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		CN 102076829 A	25 May 2011
		CN 102076829 B	28 Aug 2013
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		WO 2016185635 A1	24 Nov 2016

Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.

INTERNATIONAL SEARCH REPORT Information on patent family members	International application No. **PCT/AU2021/051532**

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Patent Document/s Cited in Search Report		**Patent Family Member/s**	
Publication Number	**Publication Date**	**Publication Number**	**Publication Date**

End of Annex

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