

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
9 April 2009 (09.04.2009)

PCT

(10) International Publication Number
WO 2009/043081 A1

(51) International Patent Classification:

C07C 1/12 (2006.01) B01J 19/08 (2006.01)
C07C 13/18 (2006.01) C07C 15/04 (2006.01)

(21) International Application Number:

PCT/AU2008/001040

(22) International Filing Date: 17 July 2008 (17.07.2008)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:

2007905394 3 October 2007 (03.10.2007) AU

(71) Applicant (for all designated States except US):

NOKUTA PTY LTD [AU/AU]; 24/10 Yalgar Road,
Kirrawee, NSW 2232 (AU).

(72) Inventor; and

(75) Inventor/Applicant (for US only): KLIEBER, Hans
[AU/AU]; 24/10 Yalgar Road, Kirrawee, NSW 2232 (AU).

(74) Agent: SHELSTON IP; 60 Margaret Street, Sydney,
MSW 2000 (AU).

(81) Designated States (unless otherwise indicated, for every

kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every

kind of regional protection available): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, NO, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Declaration under Rule 4.17:

— of inventorship (Rule 4.17(iv))

Published:

— with international search report

(54) Title: METHOD AND APPARATUS FOR PERFORMING A CHEMICAL REACTION

(57) Abstract: The present invention combines the benefits of an atomic reactor, providing energy through particle rays to convert, for example, carbon dioxide and hydrogen to aromatic and/or aliphatic compounds. In the inventive process, carbon dioxide (as gas, liquid (e.g. supercritical), or dry ice) and hydrogen gas are combined with the help of the secondary rays of an atomic reactor to produce useful aliphatic and/or aromatic compounds. Preferably, the atomic reactor is an accelerator driven system (ADS). More preferably, the accelerator driven system is a thorium accelerator driven system. The invention provides for a double benefit in replacing fossil fuels as source of power, energy and valuable limited raw materials, whilst at once reducing the greenhouse gas carbon dioxide. Preferably the produced organic compounds/raw materials are aromatic and/or aliphatic compounds. The present invention is envisaged as being applicable to a carbon-trading system.



WO 2009/043081 A1

METHOD AND APPARATUS FOR PERFORMING A CHEMICAL REACTION

Related Application

5 The present application claims convention priority from Australian Provisional Patent Application No. AU 2007905394 (dated 3 October 2007), the content of which is incorporated herein by reference in its entirety.

Field of the Invention

10 The present invention relates to chemical reactions and in particular to a method and apparatus for the synthesis of organic compounds from carbon dioxide and hydrogen.

 The invention has been developed primarily to synthesise organic compounds using atomic energy, on an industrial scale, and in turn reduce
15 greenhouse gases. However, it will be appreciated that the invention is not limited to this particular field of use.

Background of the Invention

 Any discussion of the prior art throughout the specification should in no
20 way be considered as an admission that such prior art is widely known or forms part of common general knowledge in the field.

 The most fundamental means of obtaining carbon monoxide from carbon dioxide is to heat the dioxide molecules to around 2400 °C, in turn producing carbon monoxide and oxygen. Unfortunately, the vast amounts of energy
25 required to effect such a transformation render it unfeasible when applied on an industrial scale.

 One source of energy is sunlight. Los Alamos Renewable Energy (LARE) have produced a prototype reactor that can funnel sunlight through a chamber window and channel it onto a ceramic rod set which collects heat and raises the
30 temperature within the chamber to around 2400 °C. When carbon dioxide comes into contact with the ceramic rods at this temperature, it decomposes, as above, to

- 2 -

release carbon monoxide. Notwithstanding the desirability of harnessing the sun's energy, one disadvantage of such a system is the high operating temperature, which may lead to thermal losses, and can reduce efficiency. Another disadvantage is that the equipment required construct and maintain a system capable of withstanding these temperatures is somewhat prohibitive.

An alternate method to convert carbon dioxide to carbon monoxide is through a system known as "CR5" (Counter-Rotating Ring Receiver Reactor Recuperator). Whilst this system also utilises the sun's energy, it operates at less extreme temperatures (*ca.* 1500 °C). However, one disadvantage of such a system is that successive heating and cooling cycles of cobalt ferrite ceramic rings is crucial to the process, which imparts undesirable labour intensity upon such a system.

Another method is to use sunlight to convert carbon dioxide into a carbon-based fuel in an electrochemical cell. However, the so-formed hydrocarbons liberate relatively little energy when used as fuels.

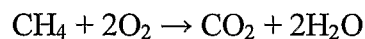
Carbon dioxide is seen as the most predominant and damaging greenhouse gas in the earth's atmosphere. Carbon dioxide is an odourless, colourless non-flammable gas that is recycled through the atmosphere by the process of photosynthesis, which occurs when green plants and other organisms transform light energy into chemical energy. Light energy is used to convert carbon dioxide, water, and other compounds into oxygen and energy rich organic compounds.

Deforestation and burning of fossil fuel results in a large proportion of human-made carbon dioxide emissions. Carbon-containing fossil fuels such as coal, oil and natural gas are used for example to generate electricity, heat houses, power factories and run cars. When they are burned, carbon from these fuels combines with oxygen to form carbon dioxide. Accordingly, over time, the concentration of greenhouse gases in the earth's atmosphere has steadily increased, insulating the earth and causing it to heat up. This, in turn, results in global warming and related problems such as the melting of the polar ice-caps.

Carbon dioxide is currently at a globally averaged concentration of approximately 387 ppm by volume in the earth's atmosphere, although this is steadily increasing due to human activity. Carbon dioxide is an important greenhouse gas because it transmits visible light but absorbs strongly in the infrared and near-infrared.

Carbon dioxide is produced by all animals, plants, fungi and microorganisms during respiration and is used by plants during photosynthesis, to make sugars which may either be consumed again in respiration or used as the raw material for plant growth. It is, therefore, a major component of the carbon cycle. Carbon dioxide is generated as a by-product of the combustion of fossil fuels or vegetable matter, among other chemical processes. Some carbon dioxide is output by volcanoes and other geothermal processes such as hot springs.

The combustion of all carbon containing fuels, such as methane (natural gas), petroleum distillates (gasoline, diesel, kerosene, propane), but also of coal and wood, will yield carbon dioxide and, in most cases, water. As an example, the chemical reaction between methane and oxygen is:



Carbon dioxide is produced from six principal sources and processes: As a by-product in ammonia and hydrogen plants, where methane is converted to CO₂; From combustion of wood and fossil fuels; As a by-product of fermentation of sugar in the brewing of beer, whisky and other alcoholic beverages; From thermal decomposition of limestone, CaCO₃, in the manufacture of lime; As a by-product of sodium phosphate manufacture; Directly from natural carbon dioxide springs, where it is produced by the action of acidified water on limestone or dolomite. However, the greatest production of CO₂ is not man made but produced by the tectonic movement of the earth's plates.

Greenhouse gases reduce the loss of heat into space and therefore contribute to global temperatures through the greenhouse effect. Greenhouse gases are essential to maintaining the temperature of the earth; without them the

- 4 -

planet would be so cold as to be uninhabitable. However, an excess of greenhouse gases can raise the temperature of a planet to lethal levels, as on Venus where the 90 bar partial pressure of carbon dioxide contributes to a surface temperature of about 467 °C.

5 Based on ice-core samples and records, current levels of carbon dioxide are approximately 100 ppmv higher than during immediately pre-industrial times, when direct human influence was negligible.

Most greenhouse gases have both natural and anthropogenic sources. During the pre-industrial holocene, concentrations of these gases were roughly
10 constant. Since the industrial revolution, concentrations of all the long-lived greenhouse gases have increased due to human actions.

Gas	Preindustrial Level	Current Level	Increase since 1750
Carbon dioxide	280 ppm	384ppm	104 ppm
Methane	700 ppb	1,745 ppb	1,045 ppb
Nitric oxide	270 ppb	314 ppb	44 ppb
CFC-12	0	533 ppt	533 ppt

Aside from water vapor, which has a residence time of days, most
15 greenhouse gases take many years to leave the atmosphere. Although it is not easy to know with precision how long it takes specific greenhouse gases to leave the atmosphere, there are estimates for the principal greenhouse gases.

Greenhouse gases can be removed from the atmosphere by various processes, including but not limited to:

- 20 1. As a consequence of a physical change (condensation and precipitation remove water vapor from the atmosphere).
2. As a consequence of chemical reactions within the atmosphere. This is the case for methane. It is oxidised by reaction with naturally occurring hydroxyl radicals, OH· and degraded to CO₂ and water vapor at the end of
25 a chain of reactions (the contribution of the CO₂ from the oxidation of

methane is not included in the methane Global Warming Potential). This also includes solution and solid phase chemistry occurring in atmospheric aerosols.

3. As a consequence of a physical interchange at the interface between the atmosphere and the other compartments of the planet. An example is the mixing of atmospheric gases into the oceans at the boundary layer.
4. As a consequence of a chemical change at the interface between the atmosphere and the other compartments of the planet. This is the case for CO₂, which is reduced by photosynthesis of plants, and which, after dissolving in the oceans, reacts to form carbonic acid and bicarbonate and carbonate ions.
5. As a consequence of a photochemical change. Halocarbons are dissociated by UV light releasing Cl· and F· as free radicals in the stratosphere with harmful effects on ozone (halocarbons are generally too stable to disappear by chemical reaction in the atmosphere).
6. As a consequence of dissociative ionisation caused by high energy cosmic rays or lightning discharges, which break molecular bonds. For example, lightning forms N anions from N₂ which then react with O₂ to form NO₂.

The lifetime τ of an atmospheric species X in a one-box model is the average time that a molecule of X remains in the box. Mathematically τ can be defined as the ratio of the mass m (kg) of X in the box to its removal rate, which is the sum of the flow of X out of the box (F_{out}), chemical loss of X (L), and deposition of X (D) (all in kg/sec).

The atmospheric lifetime of a species therefore measures the time required to restore equilibrium following an increase in its concentration in the atmosphere. Individual atoms or molecules may be lost or deposited to sinks such as the soil, the oceans and other waters, or vegetation and other biological systems, reducing the excess to background concentrations. The average time taken to achieve this is the mean lifetime.

- 6 -

The atmospheric lifetime of CO₂ is often incorrectly stated to be only a few years because that is the average time for any CO₂ molecule to stay in the atmosphere before being removed by mixing into the ocean, photosynthesis, or other processes. However, this ignores the balancing fluxes of CO₂ into the atmosphere from the other reservoirs. It is the net concentration changes of the various greenhouse gases by all sources and sinks that determines atmospheric lifetime, not just the removal processes.

Examples of the atmospheric lifetime and Global Warming Potential (GWP) for several greenhouse gases include:

- 10 • CO₂ has a variable atmospheric lifetime, and cannot be specified precisely. Recent work indicates that recovery from a large input of atmospheric CO₂ from burning fossil fuels will result in an effective lifetime of tens of thousands of years. Carbon dioxide is defined to have a GWP of 1 over all time periods.
- 15 • Methane has an atmospheric lifetime of 12 ± 3 years and a GWP of 62 over 20 years, 23 over 100 years and 7 over 500 years. The decrease in GWP associated with longer times is associated with the fact that the methane is degraded to water and CO₂ by chemical reactions in the atmosphere.
- 20 • Nitrous oxide has an atmospheric lifetime of 120 years and a GWP of 296 over 100 years.
- CFC-12 has an atmospheric lifetime of 100 years and a GWP of 10600 over 100 years.
- HCFC-22 has an atmospheric lifetime of 12.1 years and a GWP of 1700 over 100 years.
- 25 • Tetrafluoromethane has an atmospheric lifetime of 50,000 years and a GWP of 5700 over 100 years.
- Sulfur hexafluoride has an atmospheric lifetime of 3,200 years and a GWP of 22000 over 100 years.

30

- 7 -

Accordingly, one will readily appreciate that carbon dioxide is not only the most abundant of greenhouse gases, but also one of the longest-lived. It would thus be desirable to conceive of, establish and implement new technologies that may operate in unison with photosynthesis as means of reducing atmospheric carbon dioxide levels.

It would thus be advantageous to have means to utilise carbon dioxide in the atmosphere to produce relatively harmless organic compounds or hydrocarbons. It would be even more advantageous to achieve same on an industrial scale with minimal waste by-products contaminating the environment.

It is an object of the present invention to overcome or ameliorate at least one of the disadvantages of the prior art, or to provide a useful alternative.

It is further conceivable that even where carbon dioxide could be converted to undesirable organics, these too may be *in situ*, or independently converted to relatively harmless products.

To this end, hexaketocyclohexane is a chemical contaminant, found principally in soil. Destruction of this and related contaminants is possible via the use of industrially important enzyme laccase from *Coriolus versicolor*, as described in US 7,169,965.

Attempts to decompose hazardous chemical substances directly in plant cells using transformant plants into which an enzyme gene for decomposing hazardous chemical substances derived from microorganisms are introduced, have been made with respect to 2,4,6-trichlorophenol (Japan Society for Bioscience, Biotechnology, and Agrochemistry, Abstracts for the Annual Meeting, p.164, 1998) or gamma-hexacyclohexane (Japan Society for Bioscience, Biotechnology, and Agrochemistry, Abstracts for the Annual Meeting, p.89, 1997). It has been clarified that laccase can decompose various chemical substances which are not readily degradable. Laccase can oxidatively decompose endocrine disrupting chemicals including chlorophenols, agricultural chemicals, polycyclic aroma hydrocarbons, alkyl phenol, aroma hydrocarbons, and nitro compounds. Accordingly, when genes for phenoxidase, *e.g.* laccase, are incorporated and plants which can express a function of the genes are prepared, a method of

producing phenoloxidase at high yields and desirable cost levels can be established. Also, it is further possible to accomplish phyto-remediation which is useful for decomposing and removing hazardous chemical substances in the environment.

5 Thorium, as well as uranium and plutonium, can be used as fuel in a nuclear reactor. Although not fissile itself, ^{232}Th will absorb slow neutrons to produce (^{233}U), which is fissile. Hence, like ^{238}U , it is fertile. In one significant respect ^{233}U is better than the other two fissile isotopes used for nuclear fuel, ^{235}U and plutonium-239 (^{239}Pu), because of its higher neutron yield per neutron
10 absorbed. Given a start with other fissile material (^{235}U or ^{239}Pu), a breeding cycle similar to, but more efficient than that currently possible with the ^{238}U -to- ^{239}Pu cycle (in slow-neutron reactors), can be set up. The ^{232}Th absorbs a neutron to become ^{233}Th which normally emits an electron and an anti-neutrino (ν_{e^-}) by β^- decay to become protactinium-233 (^{233}Pa) and then emits another electron and
15 anti-neutrino by a second β^- decay to become ^{233}U .

The irradiated fuel can then be unloaded from the reactor, the ^{233}U separated from the thorium (a relatively simple process since it involves chemical instead of isotopic separation), and fed back into another reactor as part of a closed nuclear fuel cycle.

20 Problems include the high cost of fuel fabrication due partly to the high radioactivity of ^{233}U which is a result of its contamination with traces of the short-lived ^{232}U ; the similar problems in recycling thorium due to highly radioactive ^{228}Th ; some weapons proliferation risk of ^{233}U ; and the technical problems (not yet satisfactorily solved) in reprocessing.

25 When ^{233}U absorbs a neutron, it either fissions or becomes the next heavier isotope, ^{234}U . The chance of not fissioning on absorption of a thermal neutron is about 10% or less, which is less than the corresponding capture/fission ratios for ^{235}U , ^{239}Pu or ^{241}Pu . U-234, like most actinide nuclides with an even number of neutrons, is not easily fissionable with slow neutrons, but further
30 neutron capture produces fissile ^{235}U ; if this in turn fails to fission on neutron capture, it will produce ^{236}U , ^{237}Np , ^{238}Pu , and eventually fissile ^{239}Pu . Thus

production of heavy transuranic nuclides (the minor actinides other than neptunium) is far less than in the $^{238}\text{U}/^{239}\text{Pu}$ cycle, because 98-99% of thorium cycle fuel nuclei would fission before reaching even ^{236}U . On the other hand, the thorium cycle produces some ^{231}Pa (half-life 33,000 years) via the (n,2n) reaction on ^{232}Th . Because the thorium/uranium-233 cycle produces a smaller amount of long-lived actinide isotopes, the long-term radioactivity of the spent nuclear fuel is less. Common fission products have half-lives up to 30 years (^{90}Sr , ^{137}Cs) or more than 200,000 years (^{99}Tc), and radioactivity in the period intermediate between these two scales is chiefly from actinide wastes. Another positive, if a solid-fuel reactor is used, is that thorium dioxide melts around 3,300 °C compared to 2,800 °C for uranium dioxide cycle.

Nevertheless, the thorium fuel cycle, with its potential for breeding fuel without fast neutron reactors, holds considerable potential long-term benefits. Thorium is significantly more abundant than uranium, and is a key factor in sustainable nuclear energy.

One of the earliest efforts to use a thorium fuel cycle took place at Oak Ridge National Laboratory in the 1960s. An experimental reactor was built based on Molten Salt Reactor technology to study the feasibility of such an approach, using thorium-fluoride salt kept hot enough to be liquid, thus eliminating the need for fabricating fuel elements. This effort culminated in the Molten-Salt Reactor Experiment that used ^{232}Th as the fertile material and ^{233}U as the fissile fuel. Due to a lack of funding, the MSR program was discontinued in 1976.

Although one skilled in the art will readily appreciate that any energy source is applicable to the present invention, they will also appreciate that atomic energy is preferable due to its relative cost effectiveness, *cf. e.g.* electricity. Of the potential atomic sources, thorium is preferred due to the relatively minimal waste it produces in comparison with uranium and plutonium.

Unless the context clearly requires otherwise, throughout the description and the claims, the words “comprise”, “comprising”, and the like are to be construed in an inclusive sense as opposed to an exclusive or exhaustive sense; that is to say, in the sense of “including, but not limited to”.

Although the invention will be described with reference to specific examples it will be appreciated by those skilled in the art that the invention may be embodied in many other forms.

5 **Summary of the Invention**

According to a first aspect of the invention there is provided a method for performing a chemical reaction, said method comprising the steps of:

introducing one or more predetermined reactants into a reaction chamber;

10 exciting said one or more reactants with energy derived from a fuel source in an atomic reactor, thereby to initiate said chemical reaction and provide one or more products; and

isolating said one or more products;

15 wherein said reaction chamber is in substantially non-dissipating energy communication with said fuel source, thereby to relatively optimise energy efficiency.

In an embodiment, said one or more predetermined reactants comprise greenhouse gases. In another embodiment, said one or more predetermined reactants comprise carbon dioxide and hydrogen.

20 In an embodiment, said one or more predetermined reactants are in one or more state selected from the group consisting of: solid, liquid (*e.g.* supercritical) and gas.

In an embodiment, said atomic reactor is an accelerator driven system (ADS). In another embodiment, said accelerator driven system is a thorium
25 accelerator driven system.

In an embodiment, said products comprise one or more aromatic or aliphatic compounds.

In an embodiment, said energy is in the form of particle rays. Preferably, said particle rays are secondary particle rays.

30 In an embodiment, the inventive method further comprises the step of providing one or more auxiliary elements selected from the group consisting of:

enzymes, catalysts, heat and pressure, thereby to relatively enhance reaction kinetics and/or effect a predetermined transformation of said one or more reactant species.

In an embodiment, said one or more enzymes and said one or more
5 catalysts are selected from the group consisting of: platinum, palladium, niobium, rhodium, ruthenium, earth elements, tin or biological enzymes such as cytochrome P-450, proteasomes, decarboxylase, carbonate dehydratase, biotin as a carboxylase, carbonic anhydrase and carboxylases.

In an embodiment, the inventive method further comprises the step of
10 isolating one or more intermediate products for transfer and subsequent reaction to a final product in a secondary reactor.

In an embodiment, said intermediate product is hexaketocyclohexane octahydrate (triquinoyl hydrate), and said final product is benzene or cyclohexane.

In an embodiment, said method is performed on an industrial scale. In an
15 embodiment, said introducing one or more predetermined reactants into said reaction chamber is performed actively or passively. Preferably, said active introduction of said one or more reactants into said reaction chamber is by means of sequestering.

In an embodiment, the inventive method is applicable within a system of
20 "carbon-trading" credits or debits.

According to a second aspect of the present invention there is provided one or more products of a chemical reaction, when so-formed by a method according to the first aspect of the present invention.

According to a third aspect of the present invention there is provided an
25 apparatus for performing a chemical reaction, said apparatus comprising:

means for introducing one or more predetermined reactants into a reaction chamber;

means for exciting said one or more reactants with energy derived from a fuel source in an atomic reactor, thereby to initiate said chemical
30 reaction and provide one or more products; and

means for isolating said one or more products;

wherein said reaction chamber is in substantially non-dissipating energy communication with said fuel source, thereby to relatively optimise energy efficiency.

In an embodiment, said one or more predetermined reactants comprise greenhouse gases. In an embodiment, said one or more predetermined reactants
5 comprise carbon dioxide and hydrogen.

In an embodiment, said one or more predetermined reactants are in one or more state selected from the group consisting of: solid, liquid (*e.g.* supercritical) and gas.

In an embodiment, said atomic reactor is an accelerator driven system (ADS). Preferably, said accelerator driven system is a thorium accelerator driven system.
10

In an embodiment, said products comprise one or more aromatic or aliphatic compounds.

In an embodiment, said energy is in the form of particle rays. Preferably, said particle rays are secondary particle rays.
15

In an embodiment, the inventive apparatus further comprises means for providing one or more auxiliary elements selected from the group consisting of: enzymes, catalysts, heat and pressure.

In an embodiment, said one or more enzymes and said one or more catalysts are selected from the group consisting of: platinum, palladium, niobium, rhodium, ruthenium, earth elements, tin or biological enzymes such as cytochrome P-450, proteasomes, decarboxylase, carbonate dehydratase, biotin as a carboxylase, carbonic anhydrase and carboxylases.
20

In an embodiment, the inventive apparatus further comprises means for isolating one or more intermediate products for transfer and subsequent reaction to a final product in a secondary reactor.
25

In an embodiment, said intermediate product is hexaketocyclohexane octahydrate (triquinoyl hydrate), and said final product is benzene or cyclohexane.

In an embodiment, the inventive apparatus is scalable to an industrial scale. In an embodiment, said means for introducing one or more predetermined
30

- 13 -

reactants into said reaction chamber is active or passive. Preferably, said active means for introduction of said one or more reactants into said reaction chamber is by sequestering.

In an embodiment, the inventive apparatus is applicable within a system of
5 “carbon-trading” credits or debits.

Accordingly, it will be appreciated that the present invention combines the benefits of an atomic reactor, providing energy through particle rays to convert, for example, carbon dioxide and hydrogen to aromatic and/or aliphatic compounds. The inventive process further provides a partial solution to depletion
10 of oil reserves as energy source and raw material for many chemical compounds. The use of a thorium reactor especially provides synergistic benefits for the environment through the reduction of carbon dioxide and relatively little radioactive waste. Indeed, under the inventive process, the waste generated would need to be contained only for around 500 years, itself only around five
15 percent of the “locked-up” time necessary for most nuclear waste. Any consideration of “carbon trading”, *i.e.* the giving and receiving of “credits” for reduction in carbon dioxide emissions, will enhance the economic potential of the present invention.

In the inventive process, carbon dioxide (as gas, liquid (*e.g.* supercritical)
20 or dry ice) and hydrogen gas are combined with the help of the secondary rays of an atomic reactor to produce useful aliphatic and/or aromatic compounds.

Thus, the present invention broadly provides a method for producing/synthesising organic compounds/raw materials from carbon dioxide and hydrogen using high energy radiation from an atomic reactor.

25 Preferably, the atomic reactor is an accelerator driven system (ADS). More preferably, the accelerator driven system is a thorium accelerator driven system.

Preferably the organic compounds/raw materials are aromatic and/or aliphatic compounds.

30 Preferably, the method further includes use of enzymatic processes and/or catalyst/s and/or heat and/or pressure to catalyse the initiation and/or enhance the

speed of the reaction and to produce a predetermined one or more organic molecules.

The catalysts are preferably elements or mixtures of elements such as platinum, palladium, niobium, rhodium, ruthenium, earth elements, tin or
5 biological enzymes such as cytochrome P-450, proteasomes, decarboxylase, carbonate dehydratase, biotin as a carboxylase, carbonic anhydrase and carboxylases

Catalytic and/or enzymatic processes and/or pressure and/or heat can be used to convert carbon dioxide (as gas, liquid (*e.g.* supercritical), or dry ice) and
10 hydrogen gas into hexaketocyclohexane octahydrate (triquinoyl hydrate), which may, in turn, be reduced to benzene or other useful chemicals.

Preferably, the method is performed on an industrial scale.

The present invention also broadly provides a method of reducing green house gases comprising sequestering carbon dioxide for use in a method
15 according to the broad form of the invention, as described above.

With regard to climate change, the term "sequestration" denotes a technique for the permanent storage of carbon dioxide or other IR active compounds so they will not be released to the atmosphere where they would contribute to the greenhouse gas effect.

20 According to another broad form of the present invention there is provided an atomic reactor system for producing/synthesising organic compounds/raw materials from carbon dioxide and hydrogen, the reactor adapted such that the vessel/chamber for synthesis of the compound is adjacent/in close contact with the fuel of the reactor to receive the necessary density of incident rays.

25 Preferably the atomic reactor is an accelerator driven system (ADS). Even more preferably, the accelerator driven system is a thorium accelerator driven system.

The organic compounds/raw materials are preferably aromatic and/or aliphatic compounds.

Preferred Embodiment of the Invention

In an especially preferred embodiment, the present invention provides a method and apparatus for synthesising, on an industrial scale, organic compounds from carbon dioxide and hydrogen by using high-energy radiation from a thorium
5 atomic reactor.

The use of carbon dioxide and hydrogen in such a method may provide product intermediates similar to the biological process of photosynthesis, which provides material to be used as an energy source or raw material for other organic compounds such as plastics.

10 Those skilled in the art will readily appreciate that depending upon the precise reaction conditions employed (stoichiometric ratio of carbon dioxide to hydrogen, temperature, pressure, catalysis, residence time, *etc.*), the inventive method and apparatus may give rise to any conceivable oxygen-hydrocarbon product mixture. Most preferably, triquinolyl hexahydrate ($C_6H_6 \cdot 6H_2O$) is an
15 intermediate product of the reaction of carbon dioxide with hydrogen, which is itself, conceivably reducible to benzene. The so-formed benzene can be subjected to industrial use, such as in the plastics industry.

In particular, the invention concerns the production of aromatic and/or aliphatic compounds from carbon dioxide and hydrogen by using the high energy
20 delta radiation of an atomic reactor, preferably a thorium accelerator driven system (ADS). The use of an atomic reactor provides high-energy rays to enable the reaction to take place.

Accelerator-driven systems are safer than normal fission reactors as they are subcritical and stop when the input current is switched off. Such a system
25 may be thorium fueled, although the accelerator-driven system is applicable to other isotopes of uranium, plutonium or any other radioisotope.

Thorium cannot maintain criticality on its own, therefore the thorium reactor is "sub-critical". This means it cannot produce a chain reaction that could lead to a meltdown and is thereby a further advantage of the present invention.
30 The starting materials for a thorium reactor are generally a mixture of thorium with plutonium and uranium to provide the necessary neutrons. Alternatively, a

particle accelerator, which fires protons into a target such as lead, which in turn releases neutrons that collide with thorium to initiate fission is applicable to the present invention.

Additionally, the abundance of minable thorium ore is about 500 times
5 that of ^{235}U . Australia and India have especially high resources of thorium ore. The present invention prefers a thorium reactor as a source of high energy. The thorium reactor is also less likely to produce radioactivity in the synthesis vessel.

A delta ray (sometimes called "secondary radiation") is characterised by very fast electrons produced in quantity by alpha particles or other fast energetic
10 charged particles knocking orbiting electrons out of atoms. Collectively, these electrons are defined as delta radiation when they have sufficient energy to ionise further atoms through subsequent interactions on their own. Delta rays appear as branches in the main track of a cloud chamber. These branches will appear nearer the start of the track of a heavy charged particle, where more energy is imparted
15 to the ionised electrons.

Otherwise called a knock-on electron, the term "delta ray" is also used in high-energy physics to describe single electrons in particle accelerators that are exhibiting characteristic deceleration. In a bubble chamber, electrons will lose their energy more quickly than other particles through Bremsstrahlung and will
20 create a spiral track due to their small mass and the magnetic field. The Bremsstrahlung rate is proportional to the square of the acceleration of the electron. The secondary rays have an enormous speed but only a short penetration.

The secondary rays activate carbon dioxide and hydrogen to the extent that
25 they react even at room temperature. The reactor provides the energy to produce aromatic and/or aliphatic compounds from carbon dioxide and hydrogen on a large/industrial scale. To enhance the kinetics of the reaction or to achieve a certain outcome of product molecule/s, one can additionally use a catalytic process and/or pressure and/or heat.

30 The catalysts are preferably elements or mixtures of elements such as platinum, palladium, niobium, rhodium, ruthenium, earth elements, tin or

- 17 -

biological enzymes such as cytochrome P-450, proteasomes, decarboxylase, carbonate dehydratase, biotin as a carboxylase, carbonic anhydrase and carboxylases. The hydrogen and carbon dioxide can either be produced on site using the reactor's energy or provided from an outside source.

5 To achieve the desired effect, the vessel for the synthesis needs to be in close contact with the fuel of the reactor to achieve the necessary density of rays. This can be achieved by using a design similar to the one shown by Paul Harteck (1960) to produce nitrous oxide from nitrogen and oxygen. The process can produce a mixture of organic compounds obeying the stoichiometric ratios of the
10 reactant molecules, which can be separated by conventional chemical means such as vaporization, distillation or chromatography.

The waste from a conventional reactor can reduce the ecological benefit of sequestering or using carbon dioxide to produce raw material for organic compounds. Therefore the preferred reactor type would be a thorium reactor,
15 which produces much less waste and can be better controlled.

Examples

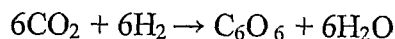
The following Examples illustrate preferred embodiments in accordance with the present invention.

20

Example 1: Cyclic Molecules from CO₂

The reaction proceeds via the following stoichiometry, in the presence of the thorium reactor's radiation energy, and optionally, one or more selected from the group consisting of: enzyme/catalyst/heat/pressure:

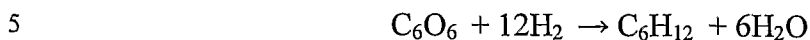
25



The so-formed hexaketocyclohexane hexahydrate can be removed via conventional processes and subjected to remediation via the process of, for
30 example, US 7,169,965, as discussed above – or else reacted further, be it in

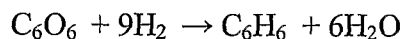
- 18 -

isolation or *in situ*, in the presence of the thorium reactor's radiation energy, and optionally, one or more selected from the group consisting of: enzyme/catalyst/heat/pressure, to give cyclohexane as per:



Alternatively, the hexaketocyclohexane can be reacted in similar manner, under different reactor conditions to give benzene for use in the chemical industry to make products such as styrene, cumene, nylon-6 and cyclohexane.

10 The worldwide demand for styrene is approximately 18 million tonnes per annum; that of cumene is approximately 12 million tonnes per annum; and that of cyclohexane is estimated to reach 500 million gallons for 2008.

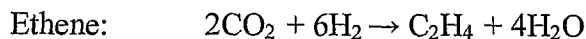
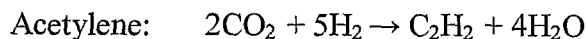


15

Example 2: Aliphatic Molecules from CO₂

The reaction proceeds via the following stoichiometry, in the presence of the thorium reactor's radiation energy, and optionally, one or more selected from the group consisting of: enzyme/catalyst/heat/pressure, to give acetylene, ethane

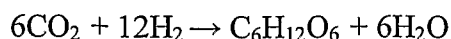
20 or ethane:



Example 3: Sugars from CO₂

The reaction proceeds via the following stoichiometry, in the presence of the thorium reactor's radiation energy, and optionally, one or more selected from

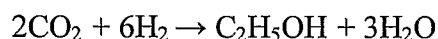
30 the group consisting of: enzyme/catalyst/heat/pressure:



The so-formed glucose is applicable to the food industry, for example, in
5 the manufacture of chocolate or other confectionary products.

Example 4: Alcohols from CO₂

The reaction proceeds via the following stoichiometry, in the presence of
the thorium reactor's radiation energy, and optionally, one or more selected from
10 the group consisting of: enzyme/catalyst/heat/pressure:



The so-formed ethanol is applicable to the beverage industry, for example,
15 in the distillation of spirits and other alcohol-containing products or as biofuels.

Example 5: Carbonyls from CO₂

The reaction proceeds via the following stoichiometry, in the presence of
the thorium reactor's radiation energy, and optionally, one or more selected from
20 the group consisting of: enzyme/catalyst/heat/pressure:



The so-formed acetone is applicable to the solvents industry. Other
25 ketones and esters may be applicable to the perfume or paint industries.

It will be appreciated by persons skilled in the art that numerous variations
and/or modifications may be made to the invention as shown in the specific
embodiments without departing from the spirit or scope of the invention as
30 broadly described. To achieve a certain mix of resulting organic molecules, a
change in the radiation energy, catalyst and/or heat and/or pressure can be used.

The present embodiments are, therefore, to be considered in all respects as illustrative and not restrictive.

Reference throughout this specification to “one embodiment” or “an embodiment” means that a particular feature, structure or characteristic described in connection with the embodiment is included in at least one embodiment of the present invention. Thus, appearances of the phrases “in one embodiment” or “in an embodiment” in various places throughout this specification are not necessarily all referring to the same embodiment, but may. Furthermore, the particular features, structures or characteristics may be combined in any suitable manner, as would be apparent to one of ordinary skill in the art from this disclosure, in one or more embodiments.

Similarly it should be appreciated that in the above description of exemplary embodiments of the invention, various features of the invention are sometimes grouped together in a single embodiment, or description thereof for the purpose of streamlining the disclosure and aiding in the understanding of one or more of the various inventive aspects. This method of disclosure, however, is not to be interpreted as reflecting an intention that the claimed invention requires more features than are expressly recited in each claim. Rather, as the following claims reflect, inventive aspects lie in less than all features of a single foregoing disclosed embodiment. Thus, the claims following the Preferred Embodiment of the Invention are hereby expressly incorporated into this Preferred Embodiment of the Invention, with each claim standing on its own as a separate embodiment of this invention.

Furthermore, while some embodiments described herein include some but not other features included in other embodiments, combinations of features of different embodiments are meant to be within the scope of the invention, and form different embodiments, as would be understood by those in the art. For example, in the following claims, any of the claimed embodiments can be used in any combination.

Furthermore, some of the embodiments are described herein as a method or combination of elements of a method that can be implemented by a processor

of a computer system or by other means of carrying out the function. Thus, a processor with the necessary instructions for carrying out such a method or element of a method forms a means for carrying out the method or element of a method. Furthermore, an element described herein of an apparatus embodiment is an example of a means for carrying out the function performed by the element for the purpose of carrying out the invention.

In the description provided herein, numerous specific details are set forth. However, it is understood that embodiments of the invention may be practiced without these specific details. In other instances, well-known methods, structures and techniques have not been shown in detail in order not to obscure an understanding of this description.

As used herein, unless otherwise specified the use of the ordinal adjectives “first”, “second”, “third”, etc., to describe a common object, merely indicate that different instances of like objects are being referred to, and are not intended to imply that the objects so described must be in a given sequence, either temporally, spatially, in ranking, or in any other manner.

In the claims below and the description herein, any one of the terms comprising, comprised of or which comprises is an open term that means including at least the elements/features that follow, but not excluding others. Thus, the term comprising, when used in the claims, should not be interpreted as being limitative to the means or elements or steps listed thereafter. For example, the scope of the expression a device comprising A and B should not be limited to devices consisting only of elements A and B. Any one of the terms including or which includes or that includes as used herein is also an open term that also means including at least the elements/features that follow the term, but not excluding others. Thus, including is synonymous with and means comprising.

Thus, while there has been described what are believed to be the preferred embodiments of the invention, those skilled in the art will recognise that other and further modifications may be made thereto without departing from the spirit of the invention, and it is intended to claim all such changes and modifications as fall within the scope of the invention. For example, any formulas given above are

- 22 -

merely representative of procedures that may be used. Steps may be added or deleted to methods described within the scope of the present invention.

Unless the context clearly requires otherwise, throughout the description and the claims, the words 'comprise', 'comprising', and the like are to be
5 construed in an inclusive sense as opposed to an exclusive or exhaustive sense; that is to say, in the sense of "including, but not limited to".

Although the invention has been described with reference to specific examples it will be appreciated by those skilled in the art that the invention may be embodied in many other forms.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:-

1. A method for performing a chemical reaction, said method comprising the steps of:
 - 5 introducing one or more predetermined reactants into a reaction chamber;
 - exciting said one or more reactants with energy derived from a fuel source in an atomic reactor, thereby to initiate said chemical reaction and provide one or more products; and
 - 10 isolating said one or more products;
 - wherein said reaction chamber is in substantially non-dissipating energy communication with said fuel source, thereby to relatively optimise energy efficiency.
- 15 2. A method according to claim 1, wherein said one or more predetermined reactants comprise greenhouse gases.
3. A method according to claim 1 or claim 2, wherein said one or more predetermined reactants comprise carbon dioxide and hydrogen.
- 20 4. A method according to any one of the preceding claims, wherein said one or more predetermined reactants are in one or more state selected from the group consisting of: solid, liquid (*e.g.* supercritical) and gas.
- 25 5. A method according to any one of the preceding claims, wherein said atomic reactor is an accelerator driven system (ADS).
6. A method according to any one of the preceding claims, wherein said accelerator driven system is a thorium accelerator driven system.

- 24 -

7. A method according to any one of the preceding claims, wherein said products comprise one or more aromatic or aliphatic compounds.
8. A method according to any one of the preceding claims, wherein said energy is in the form of particle rays.
9. A method according to claim 8, wherein said particle rays are secondary particle rays.
10. A method according to any one of the preceding claims, further comprising the step of providing one or more auxiliary elements selected from the group consisting of: enzymes, catalysts, heat and pressure.
11. A method according to claim 10, wherein said one or more enzymes and said one or more catalysts are selected from the group consisting of: platinum, palladium, niobium, rhodium, ruthenium, earth elements, tin or biological enzymes such as cytochrome P-450, proteasomes, decarboxylase, carbonate dehydratase, biotin as a carboxylase, carbonic anhydrase and carboxylases.
12. A method according to any one of the preceding claims, further comprising the step of isolating one or more intermediate products for transfer and subsequent reaction to a final product in a secondary reactor.
13. A method according to claim 12, wherein said intermediate product is hexaketocyclohexane octahydrate (triquinoyl hydrate), and said final product is benzene or cyclohexane.
14. A method according to any one of the preceding claims, wherein said method is performed on an industrial scale.

- 25 -

15. A method according to any one of the preceding claims, wherein said introducing one or more predetermined reactants into said reaction chamber is performed actively or passively.
- 5 16. A method according to claim 15, wherein said active introduction of said one or more reactants into said reaction chamber is by means of sequestering.
17. A method according to any one of the preceding claims, when applied
10 within a "carbon-trading" credits system.
18. One or more products of a chemical reaction, when so-formed by a method according to any one of the preceding claims.
- 15 19. An apparatus for performing a chemical reaction, said apparatus comprising:
means for introducing one or more predetermined reactants into a reaction chamber;
means for exciting said one or more reactants with energy derived
20 from a fuel source in an atomic reactor, thereby to initiate said chemical reaction and provide one or more products; and
means for isolating said one or more products;
wherein said reaction chamber is in substantially non-dissipating energy communication with said fuel source, thereby to relatively optimise
25 energy efficiency.
20. An apparatus according to claim 19, wherein said one or more predetermined reactants comprise greenhouse gases.
- 30 21. An apparatus according to claim 19 or claim 20, wherein said one or more predetermined reactants comprise carbon dioxide and hydrogen.

- 26 -

22. An apparatus according to any one of claims 19 to 21, wherein said one or more predetermined reactants are in one or more state selected from the group consisting of: solid, liquid (*e.g.* supercritical) and gas.
- 5
23. An apparatus according to any one of claims 19 to 22, wherein said atomic reactor is an accelerator driven system (ADS).
24. An apparatus according to any one of claims 19 to 23, wherein said
10 accelerator driven system is a thorium accelerator driven system.
25. An apparatus according to any one of claims 19 to 24, wherein said products comprise one or more aromatic or aliphatic compounds.
- 15
26. An apparatus according to any one of claims 19 to 25, wherein said energy is in the form of particle rays.
27. An apparatus according to claim 26, wherein said particle rays are secondary particle rays.
- 20
28. An apparatus according to any one of claims 19 to 27, further comprising means for providing one or more auxiliary elements selected from the group consisting of: enzymes, catalysts, heat and pressure.
- 25
29. An apparatus according to claim 28, wherein said one or more enzymes and said one or more catalysts are selected from the group consisting of: platinum, palladium, niobium, rhodium, ruthenium, earth elements, tin or biological enzymes such as cytochrome P-450, proteasomes, decarboxylase, carbonate dehydratase, biotin as a carboxylase, carbonic
30 anhydrase and carboxylases.

- 27 -

30. An apparatus according to any one of claims 19 to 29, further comprising means for isolating one or more intermediate products for transfer and subsequent reaction to a final product in a secondary reactor.
- 5 31. An apparatus according to claim 30, wherein said intermediate product is hexaketocyclohexane octahydrate (triquinoyl hydrate), and said final product is benzene or cyclohexane.
- 10 32. An apparatus according to any one of claims 19 to 31, scalable to an industrial scale.
33. An apparatus according to any one of claims 19 to 32, wherein said means for introducing one or more predetermined reactants into said reaction chamber is active or passive.
- 15 34. An apparatus according to claim 33, wherein said active means for introduction of said one or more reactants into said reaction chamber is by sequestering.
- 20 35. An apparatus according to any one of claims 19 to 34, applicable within a "carbon-trading" credits system.
- 25 36. A method for performing a chemical reaction, said method substantially as herein described with reference to any one of the embodiments of the invention illustrated in the accompanying examples.
- 30 37. One or more products of a chemical reaction, when so-formed by a method substantially as herein described with reference to any one of the embodiments of the invention illustrated in the accompanying examples.

- 28 -

38. An apparatus for performing a chemical reaction, said apparatus substantially as herein described with reference to any one of the embodiments of the invention illustrated in the accompanying examples.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU2008/001040

A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl.

C07C 1/12 (2006.01) C07C 13/18 (2006.01)
 B01J 19/08 (2006.01) C07C 15/04 (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)

DWPI, USPTO, ESPACE and GOOGLE Advanced Patent Search with IPC marks; C07C 1/12, 13/18 & 15/04, B01J 19/00 & 19/08 and keywords (atomic or nuclear and reactor, greenhouse, carbon dioxide, hydrogen and accelerator driven system)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	AU 2007100529 A4 (BIOVITAL Pty. LTD) 02 August 2007 Abstract and pages 1 & 2	1 - 4, 7 - 13 5, 6, 14 - 35
X Y	US 3228848 A (FELLOWS) 11 January 1966 Column 5 at lines 20 - 32, column 26 at lines 35 - 52 and Fig. 1	1, 8, 9 5, 6, 14 - 35
X Y	US 4362689 A (TEICHMANN et al.) 7 December 1982 Abstract, column 2 at lines 3 - 12, column 3 at lines 43 - 47 and Fig. 1	1, 8, 9 5, 6, 14 - 35

 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
"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
10 September 2008

Date of mailing of the international search report
18 SEP 2008

Name and mailing address of the ISA/AU
 AUSTRALIAN PATENT OFFICE
 PO BOX 200, WODEN ACT 2606, AUSTRALIA
 E-mail address: pct@ipaustalia.gov.au
 Facsimile No. +61 2 6283 7999

Authorized officer
KOSALA GUNATILLAKA
 AUSTRALIAN PATENT OFFICE
 (ISO 9001 Quality Certified Service)
 Telephone No : +61 2 6222 3652

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU2008/001040

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	US 4140601 A (GOMBERG) 20 February 1979 Abstract, column 2 at lines 23 – 59 and Figs 1 & 2	1, 8, 9 5, 6, 14 – 35
X Y	US 2958637 A (VOORHEES) 1 November 1960 Column 1 at lines 15 – 28, column 6 at lines 38 – 54 and Fig. 1	1, 8, 9 5, 6, 14 – 35
Y	US 2005/0013397 A1 (CSOM et al.) 20 January 2005 Abstract and claim 1	5, 6, 14 – 35
A	WO 1994/029872 A (KUNO) 22 December 1994 Whole document	
A	US 3334020 A (DEUTSCH et al.) 1 August 1967 Whole document	
A	Derwent Abstract Accession No. 90-098175/13, Class E36 J04 SU 1490074 A (KALINICHENKO) 30 June 1989 Whole document	
	<p>Note for the Y indications: (1) for claims 19 – 22 and 25 -31: AU 2007100529 can be combined with any one of US 3228848, US 4362689, US 4140601 and US 2958637. (2) for claims 5, 6, 23, 24: Any one of AU 2007100529, US 3228848, US 4362689, US 4140601 and US 2958637 can be combined with US 2005/0013397.</p>	

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/AU2008/001040

This Annex lists the known "A" publication level 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 Cited in Search Report		Patent Family Member					
AU	2007100529						
US	3228848	BE	596566	DE	1417722	FR	1280619
		GB	970864	US	3228849	US	3228850
US	4362689	CA	1025800	DE	2443626	FR	2250818
		GB	1492785	JP	50079501	NL	7414488
US	4140601	AU	74311/74	BE	821815	BR	7409591
		CA	1026705	CH	592472	DE	2454564
		DK	599474	ES	432035	FR	2251890
		GB	1494089	IL	45832	JP	50083692
		NL	7414337	NO	744127	SE	7414440
		ZA	7406528				
US	2958637	NONE					
US	2005013397	CA	2461102	EP	1428226	HU	0103762
		RU	2004111795	WO	03025951		
WO	9429872	JP	7005283				
US	3334020	NONE					
SU	1490074	NONE					
Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.							
END OF ANNEX							