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(54) Title: METHOD AND COMPOSITION FOR THE IN SITU FORMATION OF A WATER BARRIER

(57) Abstract: A water barrier can be formed *in situ* in or on at least one substrate located at e.g. a building or construction site by a method comprising: a) providing a first composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group; b) providing a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 –93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature; c) forming a mixture of said first composition and said second composition, and, before all the reactive groups of the crosslinking agent have reacted with carboxyl groups on the polymer, so forming a water-impermeable composition comprising water and a network of crosslinked organic polymer; d) conveying andapplying said mixture to the substrate(s) where said water barrier is required; wherein the amount of said first composition and said second composition in the mixture is such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8. The water barrier may be used as a damp course in building structures, geomembrane, as a barrier to inhibit or prevent leaching of chemicals into the water table, streams, rivers and oceans or to control spilt or waste fluids, or as a sealant or capping compound for tanks and pipes.

Method and Composition for the *in situ* Formation of a Water Barrier

The present invention concerns a method for the *in situ* formation of a water barrier in or on at least one substrate or between at least two substrates, and a multi-pack composition suitable for use in such a method.

Background

Crosslinked superabsorbent polymers (SAPs) are well known and are polymers that can absorb and retain large amounts of water relative to their own mass. The largest use of SAPs, typically derived from crosslinking of sodium and potassium salts of hydrophilic organic polymers, is in a variety of hygiene products, such as diapers, sanitary napkins and adult protective underwear.

Such crosslinked superabsorbent polymers have also been used for blocking water penetration in underground power or communications cables, as horticultural water retention agents, in geomembranes and in the control of spill and waste aqueous fluids.

Many applications utilise the SAPs in the form of dried powders. However, as powders can be difficult to handle, SAPs have often been provided as a coating on a nonwoven or woven sheet, with the coated product being used as a water blocking layer for a variety of applications. For example, JP-A-11319553 discloses the manufacture of a sheet comprising a water absorbing gel binder sandwiched between two sheet-like fibre materials. The gel binder is prepared by coating the sheet-like fibre material with a solution comprising a polyacrylic acid and/or polyacrylate, a polyhydric alcohol and a crosslinker, before desiccation to a moisture content of 4 to 20 weight percent.

WO2010/142568 discloses the manufacture of a swellable non-woven by a process which involves contacting a fibrous web with a polymer composition and drying the impregnated web.

JP51-143799 discloses a liquor for textile treatment to improve oil and water repellency, wherein the liquor contains polyethyleneglycol diglycidyl ether and a copolymer of perfluoroalkyl acrylate and meth(acrylic) acid.

WO-A-2011/121319 discloses a method of forming a crosslinked superabsorbent polymer on a non-woven or woven substrate, said method comprising (i) coating a non-woven or woven substrate with an aqueous composition comprising a dispersion

of a hydrophilic organic polymer comprising carboxyl functionality, a base having a boiling point no greater than the boiling point of water, and a water soluble crosslinking agent capable of crosslinking the polymer in the absence of base; and heating the coated substrate to a temperature above the boiling point of water to volatilize the base, initiate crosslinking and remove water, so as to form the crosslinked superabsorbent polymer on a non-woven substrate.

Although the prior art has provided a solution to the problems associated with the use of dried SAP powders, the provision of conventional SAP coated non-woven or woven sheets or tapes still require complex manufacturing processes to be undertaken in order to prepare, dry and apply a SAP composition to a said sheet or tape prior to the transportation of the resultant coated sheet or tape to the site for application in, or or to at least one substrate for end use as a water barrier.

Further, it is difficult to install such coated sheets and tapes as a water barrier in inaccessible areas such as, for example, within cavities between pre-formed metal, timber, concrete, masonry or composite structures, and/or in subterranean and/or subaquatic environments, as may be encountered at civil engineering project sites, such as building and construction sites. Accordingly, there remains a need for water barriers for inaccessible areas at project sites such as those identified above.

The present invention in its various aspects is as set out in the accompanying claims.

According to a first aspect, the present invention provides a method for the *in situ* formation in or on at least one substrate or between at least two substrates, of a water barrier consisting of a water-impermeable composition comprising water and a network of crosslinked organic polymer, wherein water comprises at least 50 wt% of said composition, said method comprising:

a) providing a first composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group,

b) providing a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 – 93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature,

c) forming a mixture of said first composition and said second composition, and, before all the reactive groups of the crosslinking agent have reacted with carboxyl groups on the polymer, so forming a water-impermeable composition comprising water and a network of crosslinked organic polymer,

d) conveying and applying said mixture to the substrate(s) where said water barrier is required,

wherein the amount of said first composition and said second composition in the mixture is such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8.

Preferably, the substrate or at least one of the substrates is earth e.g. soil.

Preferably, the substrate or at least one of the substrates is a component of a pre-formed metal, timber, concrete, masonry or composite structure or building, such as a house, office block, store, factory, oil rig or chemical plant.

Preferably, the substrate or at least one of the substrates is at least partially located, more preferably entirely located, in a subterranean, i.e. below the surface of the Earth, and/or subaquatic environment, i.e. below the surface of an aquatic environment such as a waterway, lake, reservoir or ocean. More preferably, the substrate or at least one of the substrates is located in a subterranean environment.

Preferably the substrate or at least one of the substrates is earth e.g. soil, or a component of a pre-formed metal, timber, concrete, masonry or composite structure or building, such as a house, office block, store, factory, oil rig or chemical plant.

In one embodiment, one substrate is earth and another substrate is a component of a pre-formed metal, timber, concrete, masonry or composite structure or building, such as a house, office block, store, factory, oil rig or chemical plant. In another embodiment, the water barrier is formed *in situ* between two substrates, each of which is a component of a pre-formed metal, timber, concrete, masonry or composite structure or building.

The water impermeable composition used in the present invention comprises at least 50 weight percent water, based upon the total weight of the composition. Preferably, the composition comprises at least 60 weight percent, more preferably at least 70 weight percent water. At the same time, it is also preferred that the composition comprises no more than 95 weight percent, more preferably no more than 80 weight

percent water. The water content of the resultant water impermeable composition can be controlled by any conventional means known to the skilled person. For example, the overall level of crosslinking and/or number of acid groups contained within the organic polymer may be modified in order to provide an end composition with the desired water absorption capacity.

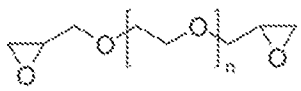
Although it is preferred that the water impermeable composition consists of a network of crosslinked organic polymer and water, it is envisaged that additional components, for example extender particles such as aggregate materials e.g. sand, pulverised rubber, etc, may be included within the composition. An extender is any particulate material that may be added to the composition to increase its volume that does not react with other components in the first or second compositions or the water-impermeable composition. A person skilled in the art will readily appreciate the amounts of extender that may be used will depend upon the intended application. Preferably, the amount of extender will not be greater than 98 wt%, more preferably not greater than 90 wt%, based on the dry weight of all components in the water-impermeable composition.

The first composition of the present invention comprises a water soluble crosslinking agent having at least two reactive groups, wherein each reactive group is capable of undergoing a condensation reaction upon contact with a carboxyl group at room temperature, i.e. at 25°C. The crosslinking agent can be a solid or a liquid. The first composition may be a solid, e.g. it may comprise a solid crosslinking agent, or the first composition may be a liquid, such as ethylene glycol diglycidyl ether. The first composition may be an aqueous solution comprising the crosslinking agent. For applications where it is desirable to provide a permanent or long term water barrier, it is preferable that the water soluble crosslinking agent does not comprise an ester functional group as it has been observed that the use of such crosslinking agents may lead to hydrolysis of the resultant crosslinked polymer over time. However, for applications where a temporary or short term water barrier is desirable, e.g. where the barrier may act as a temporary sealant or capping compound for tanks or pipes whilst maintenance or repairs are carried out, it is preferable that the water soluble crosslinking agent comprises such an ester functional group. A particularly preferred crosslinking agent comprising an ester functional group is pentaerythritol tris (3-(1-aziridinyl) propionate, which , when used in the method of the present invention, leads to hydrolysis of the crosslinked polymer and consequently a breakdown of the water barrier over a timescale of approximately 1 to 4 weeks.

Preferably, the water soluble crosslinking agent is a zirconium compound. Preferably, the zirconium compound is selected from ammonium zirconium carbonate, zirconium acetate, potassium or sodium zirconyl carbonate, or a mixture thereof. Most preferably, the zirconium compound is ammonium zirconium carbonate. Ammonium zirconium carbonate is a stabilised alkaline solution containing anionic zirconium species with bridging hydroxyl groups with carbonate groups bonded to the zirconium. It is known that zirconium reacts strongly with carboxyl groups forming strong bonds whilst hydrogen bonding occurs weakly with hydroxyl groups. Crosslinking of linear hydrophilic organic polymers comprising carboxyl functionality causes the polymer chain to uncoil which allows water to associate with the polymer chain.

Another preferred crosslinking agent is a poly(oxirane).The term poly(oxirane) refers to any compound comprising 2 or more oxirane moieties. Such compounds undergo surface cross-linking reactions when combined with a waterborne crosslinkable, hydrophilic organic polymer. The reactivity of the epoxy rings allows cross-linking to take place.

Preferably, the poly(oxirane) is a (poly)ethylene glycol diglycidyl ether, i.e. a compound of Formula (I):



Formula (I)

wherein n represents an integer from 1 to 10, preferably from 1 to 5. Most preferably, the poly(oxirane) is ethylene glycol diglycidyl ether, i.e. a compound of Formula (1), wherein n equals 1.

The second composition of the present invention comprises a waterborne crosslinkable hydrophilic organic polymer having a weight average molecular weight, measured according to ASTM D4001 – 93 (2006), of at least 200,000, wherein the polymer comprises at least 80 wt%, preferably at least 90 wt%, based on the total dry weight of the polymer, of polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group.

Preferably, the waterborne crosslinkable, hydrophilic organic polymer is a homopolymer or copolymer comprising polymerized units of acrylamide, methacrylic acid or acrylic acid, and is more preferably poly(acrylic acid). In preferred embodiments the crosslinkable, hydrophilic organic polymer has a minimum molecular weight of 300,000, more preferably 500,000 prior to the addition of a crosslinking agent. At the same time, the crosslinkable, hydrophilic organic polymer preferably has a maximum

molecular weight of 6 million, more preferably 2 million prior to the addition of a crosslinking agent.

It will be appreciated by a person skilled in the art that the water-impermeable composition and the second composition may contain small amounts of monomer and/or oligomer that are residual from the formation of the crosslinkable, hydrophilic organic polymer. However, if present, such low amounts of monomer are considered to have a negligible effect upon the performance of the water-impermeable composition.

The first and second compositions are manufactured as two separate composition packs which, prior to their combination, are transported to the project site within which the substrate(s) is(are) located. The first and second compositions are then mixed (thereby initiating a condensation reaction between the crosslinking agent of the first composition and the polymer of the second composition to crosslink the polymer), conveyed and applied to said substrate(s) at the location within the site where the water barrier is required. The water barrier is then formed *in situ* by completion of the crosslinking reaction and subsequent water absorption.

Without wishing to be bound by particular theory, it is believed that as the degree of crosslinking of the mixture increases, it becomes more difficult to convey the resultant composition to the application site as the composition increases in viscosity as the polymer swells as water is absorbed, thereby forming the water-impermeable composition. Therefore, to aid the conveying and application processes, it is preferred that the mixture is applied to the substrate(s) where said water barrier is required before no more than 10% of the reactive groups of the crosslinker have reacted with carboxyl groups on the polymer. More preferably, the mixture is applied to the substrate(s) before no more than 5% of the reactive groups of the crosslinker have reacted with carboxyl groups on the polymer. It is preferred that the mixture is prepared and applied to the substrate(s) within a period of 4 hours, more preferably within 1 hour, following the initiation of crosslinking upon combination of the first and second compositions.

Depending on the environment in which the water barrier is to be formed, the conveying and application process may be carried out during the early stages of crosslinking. By conducting the application process at this stage, the mixture is easily conveyable as subsequent water absorption and concomitant viscosity increase is minimised prior to application. However, following application at such an early stage, the composition will require further time before forming the water barrier, absorbing

water from the surrounding environment. Therefore, such application is preferred in dry environments such as within dry subterranean applications or within cavities between pre-formed metal, timber, concrete, masonry or composite structures.

Alternatively, the conveying and application process may be carried out during the later stages of crosslinking. By conducting the application process at this stage, the composition possesses water-resistant properties. However, due to the concomitant viscosity increase, conveying and applying the resultant mixture is more difficult.

The water-barrier is considered to have been formed once crosslinking has completed and the crosslinked polymer so formed has swollen with water.

The method of the present invention is advantageous over the prior art methods discussed above in that it dispenses with the necessity to employ a non-woven or woven sheet or tape as a carrier for the water barrier, and the consequence that it negates the drying step required in the manufacture of such SAP coated sheets or tapes. The method of the present invention is further advantageous because it may address some of the difficulties associated with producing a water barrier in areas where the application of sheets or tapes may be problematic dueto their inaccessibility.

The water barrier formed by the present invention may act e.g. as a damp course, inhibiting or preventing absorption of environmental water into building structures; as a geomembrane, to inhibit or prevent leakage of water from a reservoir or dam; as a barrier to inhibit or prevent leaching of chemicals into the water table, streams, rivers and oceans; as a barrier to control spilt or waste fluids; or as a sealant or capping compound for tanks and pipes. Other applications will be apparent to those skilled in the art.

As well forming a barrier to water, the water-impermeable composition may also provide a barrier to liquids, including aqueous liquids and non-aqueous liquids such as hydrocarbons.

The amount of mixture conveyed and applied to the substrates will depend upon the intended application, but must be sufficient for purpose, as would be apparent to a person skilled in the art.

In a second aspect, the present invention provides a multi-pack composition suitable for use in the method of the first aspect of the present invention, said multi-pack composition comprising

a) a first pack comprising a composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group, and

b) a second pack comprising a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 – 93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature,

wherein the amount of said first composition and said second composition are such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8. An extender, if required, may be provided as an additional pack in the multi-pack composition, or the extender may be included as a component in one or both of the first and second packs.

Preferably, the multi-pack composition is a two-pack composition consisting of

a) a first pack comprising a composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group, and

b) a second pack comprising a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 – 93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature,

wherein the amount of said first composition and said second composition are such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8. An extender, if required, may be provided as a component in one or both of the first and second packs.

The invention in its various embodiments shall now be further described by way of exemplification only:

Abbreviations used in the Example are listed below alongside their corresponding descriptions:

Polyacrylate:	Aqueous dispersion comprising 25 percent by weight potassium polyacrylate (weight average molecular weight = 500,000) based on the weight of the dispersion, available from BASF as Alcosorb™ CA10
EGDE:	ethylene glycol diglycidyl ether provided in a commercially available liquid form

Example 1

A first pack consisting of EGDE and a second pack consisting of Polyacrylate were individually prepared and delivered to the point of use. A polypropylene non-woven fabric substrate was installed inside a containment bund of a subterraneous storage tank. The first pack was combined with the second pack in a weight ratio of 1:49, and mixed for a 10 minute period by a handheld battery-powered stirrer, thereby providing a mixture wherein the ratio of oxirane groups to carboxyl groups was approximately 1:15. The mixture was then conveyed and applied over the polypropylene substrate. The coated fabric substrate was subsequently covered to prevent precipitation falling on the liquid system, and left for 24 hours to allow cross-linking to complete, thereby forming the water-impermeable composition.

The fully crosslinked system was then uncovered and left exposed to all weather conditions. Water egression measurements were recorded at several test control points over a 3 month period during which heavy rainfall was observed. The crosslinked composition provided an effective water barrier, keeping all rainwater inside the containment area surrounding the storage tank.

Claims

1. A method of *in situ* formation in or on at least one substrate or between at least two substrates, of a water barrier consisting of a water-impermeable composition comprising water and a network of crosslinked organic polymer, wherein water comprises at least 50 wt% of said composition, said method comprising:

a) providing a first composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group,

b) providing a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 – 93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature,

c) forming a mixture of said first composition and said second composition, and, before all the reactive groups of the crosslinking agent have reacted with carboxyl groups on the polymer, so forming a water-impermeable composition comprising water and a network of crosslinked organic polymer,

d) conveying and applying said mixture to the substrate(s) where said water barrier is required,

wherein the amount of said first composition and said second composition in the mixture is such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8.

2. A method as claimed in claim 1, wherein the hydrophilic organic polymer is a homopolymer or copolymer comprising polymerized units of acrylamide and/or (meth)acrylic acid.

3. A method as claimed in claim 2, wherein the hydrophilic organic polymer is poly(acrylic acid).

4. A method as claimed in any one of the claims 1-3, wherein the crosslinking agent does not comprise an ester functional group.

5. A method as claimed in any one of the preceding claims, wherein the crosslinking agent is a zirconium compound.

6. A method as claimed in claim 5, wherein the zirconium compound is ammonium zirconium carbonate, zirconium acetate, potassium or sodium zirconyl carbonate, or a mixture thereof.

7. A method as claimed in any one of claim 1-4, wherein the crosslinking agent is a poly(oxirane).

8. A method as claimed in claim 7, wherein the poly(oxirane) is ethylene glycol diglycidyl ether.

9. A method as claimed in any one of the preceding claims, wherein the water barrier is formed in situ in or on a substrate that is earth or a component of a pre-formed metal, timber, concrete, masonry or composite structure or building; or the water barrier is formed *in situ* between two substrates that are components of a pre-formed metal, timber, concrete, masonry or composite structure or building.

10. A method as claimed in any one of the preceding claims, wherein the water barrier is formed *in situ* in a subterranean and/or subaquatic environment.

11. A method as claimed in any one of the preceding claims, wherein the substrate(s) is(are) located at a building or construction site

12. A multi-pack composition suitable for use in the method as claimed in any one of the preceding claims comprising:

a) a first pack comprising a composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group, and

b) a second pack comprising a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 – 93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature,

wherein the amount of said first composition and said second composition are such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8.

13. A multi-pack composition as claimed in claim 12, wherein an extender is present as a component in at least one of the first and second pack.

14. A multi-component composition as claimed in either claim 12 or claim 13, comprising a third-pack comprising an extender.

15. A multi-pack composition as claimed in claim 12 or claim 13 that is a two-pack composition consisting of:

a) a first pack comprising a composition comprising a water soluble crosslinking agent having at least two reactive groups wherein each reactive group is capable at room temperature of undergoing a condensation reaction upon contact with a carboxyl group, and

b) a second pack comprising a second composition comprising a waterborne crosslinkable, hydrophilic organic polymer having a weight average molecular weight of at least 200,000 (according to ASTM D4001 – 93(2006)) wherein said polymer comprises at least 80 wt% polymerized units of an ethylenically unsaturated monomer having at least one carboxyl group, and wherein said carboxyl groups in said second composition will undergo condensation reactions with said crosslinking agent upon admixture of the first composition with the second composition at room temperature,

wherein the amount of said first composition and said second composition are such that the number of groups capable of undergoing a condensation reaction with a carboxyl group in the crosslinking agent to the number of carboxyl groups in said organic polymer is in the ratio of from 1:85 to 1:8. An extender, if required, may be provided as a component in one or both of the first and second packs.

INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2013/050307

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08J3/24
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08J

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

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP H02 11690 A (ASAHI CHEMICAL IND; NIPPON TELEGRAPH & TELEPHONE) 16 January 1990 (1990-01-16) abstract	1-15
A	EP 0 475 778 A2 (NIPPON CATALYTIC CHEM IND [JP]) 18 March 1992 (1992-03-18) the whole document	1-15
A	WO 2011/121319 A2 (H & R CHEMPHARM UK LTD [GB]; LYONS CHRISTOPHER MARK [GB]; MORLAND GAVI) 6 October 2011 (2011-10-06) cited in the application the whole document	1-15

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search	Date of mailing of the international search report
14 May 2013	23/05/2013

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Puttins, Udo

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/GB2013/050307

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