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(54) Title: UV PROTECTION OF POLYMERS



(57) Abstract: The present invention includes a method of producing dispersed of high quality graphene/graphite oxides in a powder matrix to then be reacted to form a UV absorbing/reflecting layer on a polymeric substrate and sealed with a compatible encapsulation coating.

UV PROTECTION OF POLYMERS

TECHNICAL FIELD OF THE INVENTION

The present invention relates in general to the field of graphite, and more particularly, to compositions and methods of graphite oxide entrainment in cement and asphalt composites.

BACKGROUND OF THE INVENTION

Without limiting the scope of the invention, its background is described in connection with composite materials.

Above 155 °C, polycarbonate flows similar to a liquid allowing it to be formed into complex mechanical structures. In general, the polycarbonate material is held to form or mold tools at temperatures above 80 °C to reduce and eliminate strain and stress in the final structure. Low molecular weight grades of polycarbonate are easier to mold but, because of the lower molecular density, results in reduced strength. The strongest grades of polycarbonate have the highest molecular mass and are more difficult to process and form.

Polycarbonate can undergo large plastic deformations without cracking or breaking. Polycarbonate can be processed and formed at room temperature using sheet metal manipulation techniques that require sharp angle bends and tight radii, with minimal heating. Polycarbonate material is useful in making transparent or electrically insulating devices. Polycarbonate resins can be extruded into tubes, rods and other profiles including multiwall structure. Polycarbonate films thinner than 1 mm can be shaped using thermoforming or more traditional machining techniques, such as bending, drilling, routing, laser cutting, etc.

SUMMARY OF THE INVENTION

A UV-protected structure comprising: a polymeric substrate, and a UV protective layer on an outside surface of said polymeric substrate with a thickness of greater than 1 nm and less than 10 μ m of the UV protective layer, that comprises a protective layer of graphene, graphene oxide, or thin flakes graphite, whereby the layer can reduce transmitted UV radiation by at least 90%, and allows transmitting at least 70% of visible spectrum of light, whereby polymer yellowing is significantly reduced. In one aspect, the UV protective layer has a thickness of greater than 1 nm and less than 1 μ m. In another aspect, the UV protective layer further comprises an adhesive layer that is compatible with the polymeric substrate. In another aspect, the UV protective layer further the UV protective layer transmits at least 70% of the visible light and transmits less than 10% of the incident UV light. In another aspect, the UV protective layer transmits at least 70% of the visible light and transmits at least 78% of the visible light and transmits less than 5%

of the incident UV light. In another aspect, the UV protective layer transmits at least 75% of the visible light and transmits less than 1% of the incident UV light. In another aspect, the structure further comprises an encapsulation layer that is chemically compatible with the polymeric substrate, wherein the encapsulation layer is less than 100 μ m thick layer and greater than 100 nm thick and encapsulates the UV protective layer and the polymeric substrate. In another aspect, the UV protective layer reduces embrittlement of the polymer. In another aspect, the polymer is selected from at least one of polycarbonate, polystyrene, poly(methyl methacrylate), poly(lactic acid), polyethylene terephthalate, polybutylene terephthalate, nylon, nylon 6, polypropylene, polyethylene, nylon 6,6, or rubber. In another aspect, the UV protective layer is applied by spraying, aerosol, spin coating, electroplating, Langmuir Blodgett immersion, roll-to-roll coating, dip coating, knife coating, roller coating, or deposition.

Another embodiment of the present invention includes a method of making an UV-protected structure comprising: providing a polymeric substrate; and applying a UV protective layer on the outside surface of said polymeric substrate with a layer thickness of greater than 1 nm and less than 10 µm, wherein the UV protective layer comprises a protective layer of graphene, graphene oxide, or thin flakes graphite, whereby the layer can reduce transmitted UV radiation by at least 90% and allows transmitting at least 70% of visible spectrum of light, whereby the polymer vellowing is significantly reduced. In one aspect, the thin layer has a thickness of greater than 1 nm and less than 1 µm of graphene/graphene oxide. In another aspect, the UV protective layer is encapsulated by adhesive layer that is compatible with polymeric substrate. In another aspect, the UV protective structure transmits at least 70% of the visible light and transmits less than 10% of the incident UV light. In another aspect, the UV protective layer transmits at least 78% of the visible light and transmits less than 5% of the incident UV light. In another aspect, the UV protective layer transmits at least 75% of the visible light and transmits less than 1% of the incident UV light. In another aspect, the UV protective structure reduces embrittlement of the polymeric sustrate. In another aspect, the polymeric substrate is selected from at least one of polycarbonate, polystyrene, poly(methyl methacrylate), poly(lactic acid), polyethylene terephthalate, polybutylene terephthalate, nylon, nylon 6, polypropylene, polyethylene, nylon 6,6, or rubber. In another aspect, the method further comprises the step of adding an adhesive layer to the UV protective structure that is compatible with the polymeric substrate. In another aspect, the method further comprises the step of adding an encapsulation layer that is chemically compatible with the polymeric substrate, wherein the encapsulation layer is less than $100 \,\mu\text{m}$ thick layer and greater than $100 \,\text{nm}$ thick and encapsulates the UV protective layer and the polymeric substrate. In another aspect, the UV

protective layer is applied by spraying, aerosol, spin coating, electroplating, Langmuir Blodgett immersion, roll-to-roll coating, dip coating, knife coating, roller coating, or deposition.

BRIEF DESCRIPTION OF THE DRAWINGS

For a more complete understanding of the features and advantages of the present invention, reference is now made to the detailed description of the invention along with the accompanying figures and in which:

FIG. 1 shows the transmission/absorption data for a single layer of GO, represented with solid lines, in the UV through the near IR in addition the plot shows calculated transmission/absorption of multiple layers GO; and

FIG 2 shows the transmission of 1 to 7 layers of GO on a transparent substrate from the visible to near IR wavelengths.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

While the making and using of various embodiments of the present invention are discussed in detail below, it should be appreciated that the present invention provides many applicable inventive concepts that can be embodied in a wide variety of specific contexts. The specific embodiments discussed herein are merely illustrative of specific ways to make and use the invention and do not delimit the scope of the invention.

To facilitate the understanding of this invention, a number of terms are defined below. Terms defined herein have meanings as commonly understood by a person of ordinary skill in the areas relevant to the present invention. Terms such as "a", "an" and "the" are not intended to refer to only a singular entity, but include the general class of which a specific example may be used for illustration. The terminology herein is used to describe specific embodiments of the invention, but their usage does not delimit the invention, except as outlined in the claims.

In one example, polycarbonate is a durable, high impact-resistance, low scratch-resistance hard polymer, and its transparency has made it the material of choice for lenses and exterior automotive components. Polycarbonate has a glass transition temperature near 147 °C. Polycarbonate is highly transparent to visible light, with better light transmission than many kinds of glass but forms color centers that produce a yellow hue with prolonged exposure to UV. The resulting damage to the polycarbonate limits the functional lifetime particularly in outdoor environments where there is exposure to high levels of UV light. Other non-limiting examples of polymers for use with the present invention include polystyrene, poly(methyl methacrylate), poly(lactic acid), polyethylene

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terephthalate, polybutylene terephthalate, nylon, nylon 6, polypropylene, polyethylene, nylon 6,6, or rubber.

Graphene is one of the strongest materials ever tested. Various research institutes have loaded hosts with carbon allotropes such as carbon nanotubes (CNT), graphene flakes (GF), graphene oxide (GO), and graphite oxide and have seen up to a 200 % increase in tensile strength in the loaded host, but with inconsistent results. Measurements have shown that graphene has a breaking strength 200 times greater than steel, with a tensile modulus (stiffness) of 1 TPa (150,000,000 psi). All forms of carbon absorb light through soft x-rays, Ultraviolet (UV) to the Infrared. The strongest absorption occurs in the short wavelengths soft x-rays and Ultraviolet (UV).

Graphene is an allotrope of carbon. Graphene's purest form is a one-atom-thick planar sheet of sp²bonded carbon atoms that are densely packed in a honeycomb or hexagonal crystal lattice. In the last few years scientists have identified multi-layer graphene oxide or a few layers of graphite oxide to be of sufficient strength and electrical conductivity to be useful as an additive in a host for providing superior mechanical, chemical, thermal, gas barrier, electrical, flame retardant and other properties compared to the native host. Improvement in the physicochemical properties of the host depends on: 1) a uniform distribution and entrainment of the graphene flake, 2) optimizing the interfacial bonding between the graphene flake and host matrix, 3) removal of gasses entrained in the host during processing, 4) optimizing the additive's innate properties, e.g. flatness, and 5) optimizing the thickness to surface-area ratio of the graphene flake and/or the chemical functionalization or decoration on the flake/particle.

A thin layer (e.g. a thickness of greater than 1 nm and less than 1 μ m) of graphene/graphene oxide can be applied to the outside surface of a polymer absorbing a large UV radiation while transmitting a large amount of visible spectrum of light that protecting the polymer from yellowing or other damage as Polymers such as Polycarbonate develop color centers or yellowing when it is exposed to short wavelength radiation where the wavelength is shorter than the visible spectrum. Thus the thin layer of graphene can generally block the yellowing radiation while transmitting visible light.

The transmission and absorption of graphene/graphene oxide can be seen in Figures 1 and 2. The graphene/graphene oxide can be applied by an aerosol or spin coating techniques, electroplating, Langmuir Blodgett, roll-to-roll coating methods such as dip coating, knife coating, roller coating or other similar deposition technique. A aerosol process paints the graphene/graphene oxide on the surface of the polymeric structure. The aerosolized carrier fluid needs to be compatible with the graphene/graphene oxide suspension to obtain a uniform dispersion. The interaction between the graphene/graphite oxide flake and a liquid host, as well as the effects gravity has on a suspension of

a carrier fluid, represents a significant technical challenge in creating and maintaining a uniform dispersion of the graphene/graphite oxide flakes in the liquid host. Powder mixing of two or more diverse materials has been shown to be effective in uniformly mixing and dispersing disparate powders independent of the polar (hydrophilic) or non-polar (hydrophobic) nature of those powders.

The carrier fluid should not evaporate too much during the time of flight of the deposition. If the carrier fluid dries in flight the surface tension of the carrier fluid will distort the flat graphene/graphene oxide structure to a crumpled structure reducing the incident surface area to the incident UV light preventing the coating from being an effective UV protective coating. Once the aerosolized suspension is deposited on the surface of the polymer, the wicking forces created from the surface tension of carrier fluid as it dries can force the graphene/graphene oxide flakes to lay flat and parallel to the surface of the polymer host, and this may be increased by some water in the carrier fluid. The graphene/graphene oxide flakes are loosely bound to the surface of the polymeric substrate and can be removed by abrasion; as such, the deposited graphene/graphene oxide flakes require a top encapsulation layer to retain the deposited layer. The encapsulation layer can be a visibly transparent epoxy, polymer or other layer that is compatible with the polymeric substrate. The polymeric substrate may be heated to facilitate uniform deposition of the aerosolized graphene/graphene oxide flakes and carrier fluid.

Graphene Oxide (GO) and/or reduced Graphene Oxide (rGO) can be suspended in either a polar or a non-polar solvent. GO is suspendable in a polar solvent such as water. rGO can be suspended in a non-polar solvent such as Dimethylformamide (DMF), ethanol, or acetone. We suspend rGO in ethanol at 1% by weight of rGO to ethanol. The suspension is sonicated for at least 30 minutes and is then atomized and deposited on the surface of the polymer substrate. For depositing on a non-planer surface, the substrate should preferably be held at a temperature ~30% lower than the evaporation temperature of ethanol. As the ethanol evaporates, surface tension draws the rGO/GO planar to the surface of the polymer substrate. The transmission and absorption of graphene/graphene oxide can be seen in Figures 1 and 2. Each layer is on the order of 10 nm thick, making 7 layers approximately 70 nm thick. The deposition can be monitored in real time by measuring the increase in mass by a precision scale or change in optical transmission by a simple imaging system. The rGO/GO is preferably deposited to a thickness less than 1 µm and greater than 1 nm. In general we deposit a layer of rGO/GO to 200nm thickness. After depositing the rGO/GO it is encapsulated with a tin coating of sprayable transparent epoxy or resin. The structure can then cured in open air or a drying chamber for no more than 30 min.

Although the present invention and its advantages have been described in detail, it should be understood that various changes, substitutions and alterations can be made herein without departing from the spirit and scope of the invention as defined by the appended claims. Moreover, the scope of the present application is not intended to be limited to the particular embodiments of the process, machine, manufacture, composition of matter, means, methods and steps described in the specification. As one of ordinary skill in the art will readily appreciate from the disclosure of the present invention, processes, machines, manufacture, compositions of matter, means, methods, or steps, presently existing or later to be developed, that perform substantially the same function or achieve substantially the same result as the corresponding embodiments described herein may be utilized according to the present invention. Accordingly, the appended claims are intended to include within their scope such processes, machines, manufacture, compositions of matter, means, methods, or steps.

Although the present invention and its advantages have been described in detail, it should be understood that various changes, substitutions and alterations can be made herein without departing from the spirit and scope of the invention as defined by the appended claims. Moreover, the scope of the present application is not intended to be limited to the particular embodiments of the process, machine, manufacture, composition of matter, means, methods and steps described in the specification. As one of ordinary skill in the art will readily appreciate from the disclosure of the present invention, processes, machines, manufacture, compositions of matter, means, methods, or steps, presently existing or later to be developed, that perform substantially the same function or achieve substantially the same result as the corresponding embodiments described herein may be utilized according to the present invention. Accordingly, the appended claims are intended to include within their scope such processes, machines, manufacture, compositions of matter, means, methods, or steps.

It is contemplated that any embodiment discussed in this specification can be implemented with respect to any method, kit, reagent, or composition of the invention, and vice versa. Furthermore, compositions of the invention can be used to achieve methods of the invention.

It will be understood that particular embodiments described herein are shown by way of illustration and not as limitations of the invention. The principal features of this invention can be employed in various embodiments without departing from the scope of the invention. Those skilled in the art will recognize, or be able to ascertain using no more than routine experimentation, numerous equivalents to the specific procedures described herein. Such equivalents are considered to be within the scope of this invention and are covered by the claims.

All publications and patent applications mentioned in the specification are indicative of the level of skill of those skilled in the art to which this invention pertains. All publications and patent applications are herein incorporated by reference to the same extent as if each individual publication or patent application was specifically and individually indicated to be incorporated by reference.

The use of the word "a" or "an" when used in conjunction with the term "comprising" in the claims and/or the specification may mean "one," but it is also consistent with the meaning of "one or more," "at least one," and "one or more than one." The use of the term "or" in the claims is used to mean "and/or" unless explicitly indicated to refer to alternatives only or the alternatives are mutually exclusive, although the disclosure supports a definition that refers to only alternatives and "and/or." Throughout this application, the term "about" is used to indicate that a value includes the inherent variation of error for the device, the method being employed to determine the value, or the variation that exists among the study subjects.

As used in this specification and claim(s), the words "comprising" (and any form of comprising, such as "comprise" and "comprises"), "having" (and any form of having, such as "have" and "has"), "including" (and any form of including, such as "includes" and "include") or "containing" (and any form of containing, such as "contains" and "contain") are inclusive or open-ended and do not exclude additional, unrecited elements or method steps. In embodiments of any of the compositions and methods provided herein, "comprising" may be replaced with "consisting essentially of" or "consisting of". As used herein, the phrase "consisting essentially of" requires the specified integer(s) or steps as well as those that do not materially affect the character or function of the claimed invention. As used herein, the term "consisting" is used to indicate the presence of the recited integer (e.g., a feature, an element, a characteristic, a property, a method/process step or a limitation) or group of integers (e.g., feature(s), element(s), characteristic(s), propertie(s), method/process steps or limitation(s)) only.

The term "or combinations thereof" as used herein refers to all permutations and combinations of the listed items preceding the term. For example, "A, B, C, or combinations thereof" is intended to include at least one of: A, B, C, AB, AC, BC, or ABC, and if order is important in a particular context, also BA, CA, CB, CBA, BCA, ACB, BAC, or CAB. Continuing with this example, expressly included are combinations that contain repeats of one or more item or term, such as BB, AAA, AB, BBC, AAABCCCC, CBBAAA, CABABB, and so forth. The skilled artisan will understand that typically there is no limit on the number of items or terms in any combination, unless otherwise apparent from the context.

As used herein, words of approximation such as, without limitation, "about", "substantial" or "substantially" refers to a condition that when so modified is understood to not necessarily be absolute or perfect but would be considered close enough to those of ordinary skill in the art to warrant designating the condition as being present. The extent to which the description may vary will depend on how great a change can be instituted and still have one of ordinary skilled in the art recognize the modified feature as still having the required characteristics and capabilities of the unmodified feature. In general, but subject to the preceding discussion, a numerical value herein that is modified by a word of approximation such as "about" may vary from the stated value by at least ± 1 , 2, 3, 4, 5, 6, 7, 10, 12 or 15%.

Additionally, the section headings herein are provided for consistency with the suggestions under 37 CFR 1.77 or otherwise to provide organizational cues. These headings shall not limit or characterize the invention(s) set out in any claims that may issue from this disclosure. Specifically and by way of example, although the headings refer to a "Field of Invention," such claims should not be limited by the language under this heading to describe the so-called technical field. Further, a description of technology in the "Background of the Invention" section is not to be construed as an admission that technology is prior art to any invention(s) in this disclosure. Neither is the "Summary" to be considered a characterization of the invention(s) set forth in issued claims. Furthermore, any reference in this disclosure to "invention" in the singular should not be used to argue that there is only a single point of novelty in this disclosure. Multiple inventions may be set forth according to the limitations of the multiple claims issuing from this disclosure, and such claims accordingly define the invention(s), and their equivalents, that are protected thereby. In all instances, the scope of such claims shall be considered on their own merits in light of this disclosure, but should not be constrained by the headings set forth herein.

All of the compositions and/or methods disclosed and claimed herein can be made and executed without undue experimentation in light of the present disclosure. While the compositions and methods of this invention have been described in terms of preferred embodiments, it will be apparent to those of skill in the art that variations may be applied to the compositions and/or methods and in the steps or in the sequence of steps of the method described herein without departing from the concept, spirit and scope of the invention. All such similar substitutes and modifications apparent to those skilled in the art are deemed to be within the spirit, scope and concept of the invention as defined by the appended claims.

What is claimed is:

 A UV-protected structure comprising: a polymeric substrate; and

a UV protective layer on an outside surface of said polymeric substrate with a thickness of greater than 1 nm and less than 10 μ m of the UV protective layer that comprises a protective layer of graphene, graphene oxide, or thin flakes graphite, whereby the layer can reduce transmitted UV radiation by at least 90% and allows transmitting at least 70% of visible spectrum of light, whereby polymer yellowing is significantly reduced.

2. The structure of claim 1, wherein the UV protective layer has a thickness of greater than 1 nm and less than 1 μ m.

3. The structure of claim 1, wherein the UV protective layer further comprises an adhesive layer that is compatible with the polymeric substrate.

4. The structure of claim 1, wherein the UV protective layer transmits at least 70% of the visible light and transmits less than 10% of the incident UV light.

5. The structure of claim 1, wherein the UV protective layer transmits at least 78% of the visible light and transmits less than 5% of the incident UV light.

6. The structure of claim 1, wherein the UV protective layer transmits at least 75% of the visible light and transmits less than 1% of the incident UV light.

7. The structure of claim 1, further comprising an encapsulation layer that is chemically compatible with the polymeric substrate, wherein the encapsulation layer is less than 100 μ m thick layer and greater than 100 nm thick and encapsulates the UV protective layer and the polymeric substrate.

8. The structure of claim 1, wherein the UV protective layer reduces embrittlement of the polymeric substrate.

9. The structure of claim 1, wherein the polymer is selected from at least one of polycarbonate, polystyrene, poly(methyl methacrylate), poly(lactic acid), polyethylene terephthalate, polybutylene terephthalate, nylon, nylon 6, polypropylene, polyethylene, nylon 6,6, or rubber.

10. A method of making an UV-protected structure comprising: providing a polymeric substrate; and

applying a UV protective layer on the outside surface of said polymeric substrate with a layer thickness of greater than 1 nm and less than 10 μ m, wherein the UV protective layer

comprises a protective layer of graphene, graphene oxide, or thin flakes graphite, whereby the layer can reduce transmitted UV radiation by at least 90% and allows transmitting at least 70% of visible spectrum of light, whereby the polymer yellowing is significantly reduced.

11. The method of claim 10, wherein the thin layer has a thickness of greater than 1 nm and less than 1 μ m of graphene/graphene oxide.

12. The method of claim 10, wherein the UV protective layer is encapsulated by adhesive layer that is compatible with polymeric substrate.

13. The method of claim 10, wherein the UV protective layer transmits at least 70% of the visible light and transmits less than 10% of the incident UV light.

14. The method of claim 10, wherein the UV protective layer transmits at least 78% of the visible light and transmits less than 5% of the incident UV light.

15. The method of claim 10, wherein the UV protective layer transmits at least 75% of the visible light and transmits less than 1% of the incident UV light.

16. The method of claim 10, wherein the UV protective layer reduces embrittlement of the polymeric substrate.

17. The method of claim 10, wherein the polymeric substrate is selected from at least one of polycarbonate, polystyrene, poly(methyl methacrylate), poly(lactic acid), polyethylene terephthalate, polybutylene terephthalate, nylon, nylon 6, polypropylene, polyethylene, nylon 6,6, or rubber.

18. The method of claim 10, further comprising the step of adding an adhesive layer to the UV protective layer that is compatible with the polymeric substrate.

19. The method of claim 10, further comprising the step of adding an encapsulation layer that is chemically compatible with the polymeric substrate, wherein the encapsulation layer is less than $100 \mu m$ thick layer and greater than 100 nm thick and encapsulates the UV protective layer and the polymeric substrate.

20. The method of claim 10, wherein the UV protective layer is applied by spraying, aerosol, spin coating, electroplating, Langmuir Blodgett immersion, roll-to-roll coating, dip coating, knife coating, roller coating, or deposition.

21. A UV-protected structure comprising:

a polymeric substrate; and

a thin layer on the outside surface of said polymeric substrate with a thickness of greater than 1 nm and less than 10 μ m of UV protective structure on that comprises a protective layer of graphene, graphene oxide, or thin flakes graphite, whereby the layer can reduce transmitted UV radiation by at least 90% and allows transmitting at least 70% of visible spectrum of light, whereby the polymer yellowing is significantly reduced.

22. A method of making an UV-protected structure comprising: providing a polymeric substrate; and

applying a thin layer on the outside surface of said polymeric substrate with a layer thickness of greater than 1 nm and less than 10 μ m of UV protective structure on that comprises a protective layer of graphene, graphene oxide, or thin flakes graphite, whereby the layer can reduce transmitted UV radiation by at least 90% and allows transmitting at least 70% of visible spectrum of light, whereby the polymer yellowing is significantly reduced.



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WAVELENGTH (nm)

A. CLASSIFICATION OF SUBJECT MATTER C08J 7/06(2006.01)i, C08L 101/00(2006.01)i

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 7/06; C09D 183/08; B32B 3/00; H05K 1/09; H01B 1/24; B32B 27/14; B32B 27/06; H01B 1/04; B32B 9/04; B32B 27/20; C08L 101/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Korean utility models and applications for utility models Japanese utility models and applications for utility models

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) eKOMPASS(KIPO internal) & Keywords: polymeric substrate, UV protective layer, graphene, praphene oxide, praphite

DOCUMENTS CONSIDERED TO BE RELEVANT C. Category Relevant to claim No. Citation of document, with indication, where appropriate, of the relevant passages US 2011-0088931 A1 (LETTOW, JOHN S. et al.) 21 April 2011 1 - 22Y See paragraphs [0015], [0019], [0020], [0024], [0027], [0034], [0037], [0097] ; claims 1, 8, 20; and figure 1. US 2010-0239870 A1 (BOWEN, DAVID A. et al.) 23 September 2010 Y 1 - 22See paragraphs [0032]-[0034], [0055]; claim 6; and figures 1, 2. US 2011-0049437 A1 (CRAIN, JOHN M. et al.) 03 March 2011 1 - 22А See paragraphs [0049], [0051]; and figure 1. US 2007-0219336 A1 (ITO, KAZUHIKO) 20 September 2007 1 - 22A See claim 1. А US 2005-0191471 A1 (HAGGQUIST, GREGORY W.) 01 September 2005 1 - 22See claims 1. 2. X See patent family annex. Further documents are listed in the continuation of Box C. Special categories of cited documents: later document published after the international filing date or priority "T" "A" document defining the general state of the art which is not considered date and not in conflict with the application but cited to understand to be of particular relevance the principle or theory underlying the invention "E" earlier application or patent but published on or after the international "X" document of particular relevance; the claimed invention cannot be filing date considered novel or cannot be considered to involve an inventive "1." document which may throw doubts on priority claim(s) or which is step when the document is taken alone cited to establish the publication date of another citation or other $^{\parallel}V^{\parallel}$ document of particular relevance; the claimed invention cannot be special reason (as specified) considered to involve an inventive step when the document is "O' document referring to an oral disclosure, use, exhibition or other combined with one or more other such documents, such combination means being obvious to a person skilled in the art "P' document published prior to the international filing date but later "&" document member of the same patent family than the priority date claimed Date of mailing of the international search report Date of the actual completion of the international search 13 May 2016 (13.05.2016) 12 May 2016 (12.05.2016) Name and mailing address of the ISA/KR Authorized officer International Application Division Korean Intellectual Property Office CHO, KI YUN 189 Cheongsa-ro, Seo-gu, Daejeon, 35208, Republic of Korea Telephone No. +82-42-481-5655 Facsimile No. +82-42-481-8578

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/US2016/014873

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