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(54) Title: A PACKAGING COMPOSITE

(57) Abstract: The present invention is in the field of packaging composites. More particularly the present invention relates to a packaging composite providing improved mechanical properties. It has been found that a combination of graphene or derivative thereof in the range 0.01 to 1.0 % by weight and select slip additive in the range 1.0 to 10 % by weight and a polymer provides a packaging composite, which shows synergistic improvement in mechanical properties of the composite without compromising its barrier properties. The present invention further describes a packaging film and a container comprising the composite. It also describes a process for providing the packaging composite comprising steps of providing a polymer in liquid form, mixing graphene or derivative thereof and a slip additive with the polymer and extruding or casting the polymer to form the composite.



A PACKAGING COMPOSITE

Field of the Invention

The present invention is in the field of packaging composites. More particularly, the present invention relates to a packaging composite providing improved mechanical properties.

Background of the Invention

5 Consumers often use various products such as, laundry detergent, surface cleaner, food products, soap, skin creams etc. in their day-to-day activities. Most of such products are marketed in packaged condition, where the products are wrapped or packed using suitable packaging materials. There are various types of packaging depending on products and/or their
10 applications, for example flexible packaging, rigid packaging, packaging film etc., they may be used as primary or secondary packaging depending on the purpose and functionality. Typically, a packaging embodiment contains multiple layers made of different composites providing various functional benefits.

Those functional benefits include, protecting the products from environmental stress, working as
15 a moisture barrier, reducing accidental damage during transportation, reducing microbial contamination etc. Therefore, it is desired that the packaging composites to have improved mechanical and barrier properties. The packaging composite with improved barrier properties, reduces moisture ingress into the product from surroundings, thus maintains the product performance and enhances shelf life of the product. Further improved mechanical properties
20 provide better protection from external stress and reduces damages during handling and transportation.

One way to improve the barrier properties of a packaging composite is to incorporate a hydrophobic compound into it. However, such compounds may affect the mechanical properties, such as, tensile strength, elongation etc. of the composite, which is not preferred.

25 In this context CN107353507 (Anhui Songtai Packaging Mat Co Ltd, 2017) describes an anti-static easily degradable environment-friendly packaging material. The anti-static easily-degradable environment-friendly packaging material is prepared from the following raw materials in parts by weight: 60-70 parts of polypropylene, 4-8 parts of silicon dioxide, 2-4 parts of magnesium oxide, 3-7 parts of oleamide, 1-2 parts of a dispersing agent, 2-6 parts of
30 chitosan, 0.5-1.5 parts of a slipping agent, 2-5 parts of nanocellulose, 3-10 parts of oxidized graphene, 2-6 parts of active manganese oxide, 3-8 parts of calcium carbonate, 1-2 parts of a silane coupling agent, 1-2 parts of an anti-oxidant, 3-6 parts of nano titanium dioxide, 1-5 parts

of conductive fibres, 1-3 parts of an anti-static agent, 3-5 parts of zinc oxide crystal whiskers and 0.3-1 part of a degradation promoter.

CN111976244 (Wuxi Chun Yu Environmental Product Co Ltd, 2020) describes a full biodegrade air bubble film composite material and a preparation method thereof. It describes an air bubble
5 film comprising 70 parts of aliphatic-aromatic polyester copolymer PBAT, 20 parts of aliphatic polyester polylactic acid, 15 parts of polypropylene carbonate, and graphene oxide 1 part, 5 parts of inorganic filler superfine talc, 1 part of ADR4468 containing active epoxy group, 1 part of slip agent erucamide.

CN112644123A (Xiamen Changsu Ind Co Ltd., 2021) describes a toughened biaxially oriented
10 polylactic acid film and a preparation method thereof. The film is sequentially provided with a first surface layer, a core layer and a second surface layer; the first surface layer and the second surface layer are composed of, in percentage by mass, 10 to 20% of toughening resin, 0.5 to 3% of nano inorganic filler, 0.1 to 1% of an anti-blocking agent, 0.1 to 1% of a slipping agent and 75 to 89.3% of PLLA resin; the core layer is composed of, in percentage by mass,
15 10-20% of toughening resin, 0.5-3% of nano inorganic filler and 77-89.5% of PLLA resin; and the toughening resin is at least one of a polylactic acid stereo complex or a copolymer.

CN111171433A (Shantou Jinping Dongxing Packaging Co Ltd., 2020) describes a packaging film comprising following components in parts by weight: 8 to 15 parts of graphene oxide, 5 parts of lignin, 5 parts of corn starch, 2 to 3 parts of polybutylene succinate, 2 to 4 parts of
20 polycyclic lactone, 2 to 4 parts of an antioxidant, 6 to 8 parts of ethylene glycol, and 20 to 40 parts of polyethylene.

CN111976244 (Wuxi Chun Yu Environmental Product Co Ltd, 2020) describes fully biodegradable bubble membrane comprising the following raw materials in parts by weight: 50 to 85 parts of an aliphatic-aromatic polyester copolymer, 5 to 30 parts of aliphatic polyester, 3 to
25 10 parts of an inorganic filler, 0.1 to 2 parts of a compound containing an active epoxy group, and 0.1 to 2 parts of a slipping agent.

Although various packaging embodiments are disclosed, there is still a need for an improved packaging composite for providing enhanced mechanical properties, yet without compromising other characteristics, like, barrier properties.

30 **Objects of the invention**

It is an object of the present invention to provide an improved packaging composite.

It is another object of the present invention is to provide a packaging composite with enhanced mechanical properties.

It is yet another object of the present invention is to provide a packaging composite without compromising barrier properties.

- 5 The present inventors surprisingly have found that a packaging composite comprising a combination of graphene or derivative thereof and select slip additive in specific amount provides synergistic improvement in mechanical properties of the composite without compromising barrier properties.

Thus, satisfying one or more of the above-mentioned objects.

10 **Summary of the Invention**

In a first aspect, the present invention provides a packaging composite comprising:

- a) 0.01 to 1.0 % by weight of graphene or derivative thereof;
- b) 1.0 to 10 % by weight of a slip additive; and
- c) a polymer,

- 15 wherein the slip additive is a fatty acid amide.

In a preferred aspect, the present invention provides a packaging film comprising a packaging composite according to the first aspect.

In another preferred aspect, the present invention provides a container comprising a packaging composite according to the first aspect.

- 20 In another aspect, the present invention relates to a process for providing a packaging composite according to the first aspect comprising steps of:

- a) providing a polymer in liquid form;
- b) mixing graphene or graphene derivative and a slip additive with the polymer; and
- c) extruding the polymer to form the composite.

- 25 These and other aspects, features and advantages will become apparent to those of ordinary skill in the art from reading of the following detailed description. For the avoidance of doubt, any feature of one aspect of the present invention may be utilized in any other aspect of the invention. The word “comprising” is intended to mean “including” but not necessarily “consisting of” or “composed of.” In other words, the listed steps or options need not be exhaustive. It is
30 noted that the examples given in the description below are intended to clarify the invention and

are not intended to limit the invention to those examples per se. Similarly, all percentages are weight/weight percentages unless otherwise indicated. Except in the operating and comparative examples, or where otherwise explicitly indicated, all numbers in this description indicating amounts of material or conditions of reaction, physical properties of materials and/or use are to be understood as modified by the word "about". Numerical ranges expressed in the format "from x to y" are understood to include x and y. When for a specific feature multiple preferred ranges are described in the format "from x to y", it is understood that all ranges combining the different endpoints are also contemplated.

Detailed Description of the Invention

According to the present invention there is provided a packaging composite comprising 0.01 to 1.0 % by weight graphene or derivative thereof and 1.0 to 10 % by weight a slip additive and a polymer, wherein the slip additive is a fatty acid amide.

Graphene or derivative thereof

The composite comprises 0.01 to 1.0% by weight graphene or derivative thereof.

Graphene is an allotrope, formed by a single layer carbon atoms arranged in hexagonal lattice structure. It is reported that graphene or derivative thereof has good thermal and electrical conductivity, commonly used in various sector of electrical and electronics, e.g., semiconductor, communication, sensors, etc.

Graphene is hydrophobic and can be obtained in two manners. The first is by peeling layers from graphite until you achieve a graphene monolayer. The second is known as Chemical Vapor Deposition (CVD) and where large-scale uniformity can be obtained and controlled.

Preferably the graphene derivative suitable for the invention is graphene oxide. Graphene oxide (GO) is hydrophilic and can be manufactured through Hummer's method. Graphene oxide (GO) is also commercially available and may be procured from suppliers such as Platonic Nanotech.

Preferably the composite comprises 0.01 to 0.8 %. More preferably the composite comprises 0.02 to 0.8% by weight, furthermore preferably 0.05 to 0.7 % by weight, yet more preferably 0.08 to 0.6 % by weight and even more preferably 0.09 to 0.5 % by weight and most preferably 0.09 to 0.4 % by weight of the graphene or derivative thereof.

Slip additive

The composite comprises 1.0 to 10 % by weight of a slip additive. Slip additive or slip agent are compounds known in packaging context. Such compounds are commonly used in packaging to reduce friction in polymer films to avoid sticking. It also helps in moulding processes by reducing friction between a polymer resin and an apparatus. Examples of slip additive or slip agent include inorganic materials, e.g., talc; organic materials e.g., select fatty acid amides.

Surprisingly, in the present invention the slip additive works in synergy with the graphene or derivative and improves mechanical properties of the composite without compromising its barrier properties.

Mechanical properties may be assessed in terms of tensile strength, elongation etc. Further, barrier properties of the composite are assessed terms of Water Vapour Transmission Rate (WVTR).

Preferably the composite comprises 1.2 to 8 % by weight, more preferably 1.2 to 7 % by weight, even more preferably 1.2 to 6 % by weight, furthermore preferably 1.2 to 5% by weight, yet more preferably 1.20 to 4% by weight and most preferably 1.2 to 3% by weight of the composite.

The composite may comprise at least 1.2 % by weight, more preferably at least 1.3 % by weight, even more preferably at least 1.4 % by weight and most preferably 1.5 % by weight of the slip additive. Preferably the composite comprises at most 6 % by weight, more preferably at most 5.5% by weight, even more preferably at most 5% by weight and most preferably at most 4.5 % by weight of the slip additive.

The slip additive is a fatty acid amide. Preferably the slip additive comprises fatty acid amide with 10 to 30 carbon atoms, more preferably 11 to 28 carbon atoms, even more preferably 12 to 26 and yet more preferably 12 to 24 and most preferably 12 to 22 carbon atoms. Preferably, the fatty acid amide is saturated or unsaturated. Where it is unsaturated it is preferred that it is mono- or di-, more preferably mono-unsaturated.

Most preferred slip additives include stearamide, erucamide and oleamide and mixture thereof.

Most preferred slip additives are C18 to C22 mono-unsaturated fatty acid amides and which includes erucamide and oleamide.

The composite may further contain other organic and/or inorganic slip additives known in the arts, example talc. Such slip additives may present in addition to the fatty acid amides.

Polymer

The composite comprises a polymer. The polymers may be selected from polyesters, polyolefins, polyamides, polystyrene (PS), polyanhydrides, polyacrylates, poly hydroxy alkanoates, poly vinyl chloride, thermoplastic polyurethanes, polycarbonate (PC), polylactic acid (PLA), acrylonitrile/butadiene/styrene copolymer (ABS), styrene/acrylonitrile copolymer (SAN), polyoxymethylene (POM), biodegradable thermoplastics, starch-based thermoplastics, their derivatives, and combinations thereof. It is also possible to have the composite comprising blend of two or more polymers.

Preferably the composite comprises at 50 to 99% by weight, more preferably 60 to 97% by weight, even more preferably, 70 to 95% by weight and most preferably 80 to 90% by weight of the composite.

Preferably the polymer is selected from polyesters, polyolefins, polyamides, polystyrene, polyanhydrides, polyacrylates, polyhydroxy alkanoates, poly vinyl chloride, thermoplastic polyurethanes, polycarbonate, polylactic acid, and combinations thereof.

More preferably the polymer is selected from polyesters, polyolefins, polyamides, polystyrene, polyanhydrides, polyacrylates, polyhydroxy alkanoates, poly vinyl chloride, thermoplastic polyurethanes and combinations thereof.

The composite preferably comprises a polymer is selected from polyolefins, polypropylenes and their combination.

Preferably the polymer comprises a polyolefin containing olefin monomer in polymerized form. Examples of monomer include ethylene, propylene and optionally may comprise one or more co-monomers.

Polyolefins are typically produced from a simple olefin (also called an alkene with the general formula C_nH_{2n}) as a monomer. For example, polyethylene (PE) is the polyolefin produced by polymerizing the olefin ethylene (C_2H_4). Polypropylene (PP) is another common polyolefin which is made from the olefin propylene (C_3H_6). Copolymers of ethylene and propylene are also useful polymers in accordance with the present disclosure.

A preferred polyolefin according to the present invention is a polyethylene. This includes polyethylene homopolymers or copolymers. Common forms of polyethylene known in the art include Ultra Low-Density Polyethylene (ULDPE), Very Low-Density Polyethylene (VLDPE),

Linear Low-Density Polyethylene (LLDPE), Low Density Polyethylene (LDPE), Medium Density Polyethylene (MDPE), and High-Density Polyethylene (HDPE).

LLDPE is a linear ethylene/ α -olefin copolymer containing heterogeneous short-chain branching distribution comprising units derived from ethylene and units derived from at least one C3 to C10 α -olefin comonomer. LLDPE has a density from 0.910 g/cc to less than 0.940 g/cc. Non-limiting examples of LLDPE include TUFLIN™ linear low density polyethylene resins (available from The Dow Chemical Company), DOWLEX™ polyethylene resins (available from the Dow Chemical Company), FINGERPRINT™ polyethylene resins (available from Dow Chemical Company), DOWLEX™ 2038.68G, ELITE™, ELITE™ AT, and AFFINITY™, enhanced polyethylene, such as ELITE™ 5400G, available from the Dow Chemical Company, and MARLEX™ polyethylene (available from Chevron Phillips). The copolymers may comprise just one comonomer or terpolymers, i.e., copolymers of ethylene with two further comonomers, is particularly preferred. Said terpolymer is one specifically preferred in linear low-density polyethylene (LLDPE). The linear low-density polyethylene (LLDPE) contains preferably just one or two type(s) of C3 to C10 α -olefin comonomer(s). Still more preferably, the comonomer(s) is/are selected from the group consisting of 1-butene, 1-hexene, 1-octene and mixtures thereof. In one preferred embodiment the comonomer employed is 1-octene. In another preferred embodiment the linear low-density polyethylene (LLDPE) is a terpolymer consisting of ethylene, 1-butene and 1-hexene. Preferably the linear low-density polyethylene (LLDPE) has a Melt Flow Rate (MFR) (at 2.16 Kg load and a temperature of 190°C,) in the range of 0.15 to 8.0 g/10 min, more preferably in the range of 0.15 to 4.0 g/10 min, more preferably in the range from 0.15 to 2 g/10 min, more preferably in the range of 0.3 to 1 g/10 min and still preferably in the range from 0.5 to 1 g/10 min.

Low density polyethylene (LDPE) comprises ethylene homopolymer, or ethylene/ α -olefin copolymer comprising at least one C3 to C10 α -olefin and has a density preferably from 0.915 g/cc to less than 0.940 g/cc. It contains long chain branching with broad molecular weight distribution. LDPE is typically produced by way of high pressure free radical polymerization (tubular reactor or autoclave with free radical initiator). Non-limiting examples of LDPE include MarFlex™ (Chevron Phillips), LUPOLEN™ (LyondellBasell), as well as LDPE products from Borealis, INEOS, ExxonMobil, and others. Preferably the low-density polyethylene (LDPE) has a MFR (at 2.16 Kg load and a temperature of 190°C,) in the range of 0.05 to 2.0 g/10 min, more preferably in the range of 0.10 to 1.8 g/10 min, and more preferably in the range of 0.15 to 1.5 g/10 min. Accordingly, one example of such low-density polyethylene (LDPE) is the commercial product FT5230 of Borealis AG. Examples of other commercially available polyethylene that

can be used in accordance with the present invention include those available from the Dow Chemical Company under the names DOW LDPE™ and AGILITY™.

HDPE is an ethylene homopolymer or an ethylene/ α -olefin copolymer with at least one C4 to C10 α -olefin comonomer, or C4 to C8 α -olefin comonomer and a density from 0.940 g/cc to 0.980 g/cc. HDPE can be a monomodal copolymer or a multimodal copolymer. A monomodal ethylene copolymer is an ethylene C4 to C10 α -olefin copolymer that has one distinct peak in a gel permeation chromatography (GPC) showing the molecular weight distribution. A multimodal ethylene copolymer is an ethylene C4 to C10 α -olefin copolymer that has at least two distinct peaks in a GPC showing the molecular weight distribution. Multimodal includes copolymer having two peaks (bimodal) as well as copolymer having more than two peaks. Nonlimiting examples of HDPE include DOW™ High Density Polyethylene (HDPE) Resins (available from The Dow Chemical Company), CONTINUUM™ Bimodal Polyethylene Resins (available from The Dow Chemical Company), LUPOLEN™ (available from LyondellBasell), as well as HDPE products from Borealis, INEOS, and ExxonMobil. Preferably HDPE has a Melt Flow Rate (MFR) less than 0.5 gm/10 min, preferably less than 0.4 grams/10 minutes, and will have polymerized therein at least about 98 mol % ethylene with any comonomer polymerized therein being an alpha-mono olefin containing about 3 to 12 carbon atoms.

HDPE is commercially available for example, RELENE F46003 is a high-density polyethylene (HDPE) having a density of 0.946 g/cc and a MFR of 0.38 g/10 minutes (at 2.16 Kg load and a temperature of 190°C), from the Dow Chemical Company under the names DOW™ HDPE resins and DOWLEX™.

Polyamide may be used as suitable polymer material. Typically, polyamide is made by condensation of a diacid and a diamine. Useful polyamide homopolymers include nylon 6 (polycaprolactam), nylon 11 (polyundecanolactam), and nylon 12 (polylauryllactam), nylon 4,2 (polytetramethylene ethylenediamide), nylon 4,6 (polytetramethylene adipamide), nylon 6,6 (polyhexamethylene adipamide), nylon 6,9 (polyhexamethylene azelamide), nylon 6,10 (polyhexamethylene sebacamide), nylon 6,12 (polyhexamethylene dodecanediamide), nylon 7,7 (polyheptamethylene pimelamide), nylon 8,8 (polyoctamethylene suberamide), nylon 9,9 (polynonamethylene azelamide), nylon 10,9 (polydecamethylene azelamide), and nylon 12,12 (polydodecamethylene dodecanediamide).

Other useful polyamide copolymer also includes nylon 6,6/6 copolymer (polyhexamethylene adipamide/caprolactam copolymer), nylon 6,2/6,2 copolymer (polyhexamethylene

ethylenediamide/hexamethylene ethylenediamide copolymer), nylon 6,6/6,9/6 copolymer (polyhexamethylene adipamide/hexamethylene azelaamide/caprolactam copolymer), as well as other nylons which are not particularly delineated here. One of the examples of commercially available nylon is grade of Nylon is ULTRAMID™ B33L ex. BASF.

- 5 Polyethylene terephthalate (PET) is one of the polyesters, which can be used in the present invention. PET exists both as amorphous (transparent) and as semi-crystalline (opaque and white) polymer material. Semicrystalline PET has good strength, ductility, stiffness, and hardness. Amorphous PET has better ductility but less stiffness and hardness. Other examples of polyester material are polyethylene naphthalate (PEN), poly butylene terephthalate,
10 polycarbonate etc.

In another aspect, the polymer materials may be recycled polymers, which includes post-consumer recycled (PCR) or post-industrial recycled (PIR). The recycled polymers may be a mechanically recycled or a chemically recycled polymer.

- 15 Post-consumer recycled (PCR) refers to polymers that has reached the intended end user or consumer, is no longer being used for its intended purpose, and which has been collected or reclaimed after it is discarded by the end user or consumer. In other words, the term refers to material that would have otherwise been disposed of as waste but has instead been collected and recovered (reclaimed) as a material input, in lieu of new virgin material, for a recycling or manufacturing process. The term is inclusive of such collected or reclaimed materials which
20 have been further treated or processed to facilitate re-use of the material. Preferably the recycled polymer is a mechanically recycled material. Example of commercially available recycled polyolefin are, from Corpela (Italian Consortium for the collection, recovery, recycling of packaging plastic wastes), Resource Plastics Corp. (Brampton, ON), Kruschitz GmbH, Plastics and Recycling (AT), Vogt Plastik GmbH (DE), recycled LDPE from Ecoplast etc.

- 25 In another preferred aspect, the composite comprises a water-soluble polymer, such as polyvinyl alcohol or derivative thereof. Such composite is suitable for a unit dose product, where the product as such is dissolved in water providing a cleaning composition.

The composite may comprise polyvinyl alcohol or derivatives thereof as the polymer.

- 30 Preferably the water-soluble polymer is polyvinyl alcohol (PVOH), including homopolymers thereof (e.g., including substantially only vinyl alcohol and vinyl acetate monomer units) and

copolymers thereof (e.g., including one or more other monomer units in addition to vinyl alcohol and vinyl acetate units). Preferably the polymer comprises partially hydrolysed PVOH.

Other water soluble polymers for use in addition to the PVOH polymers and PVOH copolymers in the blend can include, but are not limited to modified polyvinyl alcohols, polyacrylates, water-
5 soluble acrylate copolymers, polyvinyl pyrrolidone, polyethyleneimine, pullulan, water-soluble natural polymers including, but not limited to, guar gum, gum Acacia, xanthan gum, carrageenan, and starch, water-soluble polymer derivatives including, but not limited to, modified starches, ethoxylated starch, and hydroxypropylated starch, copolymers of the forgoing and combinations of any of the foregoing. Yet other water-soluble polymers can
10 include polyalkylene oxides, polyacrylamides, polyacrylic acids and salts thereof, celluloses, cellulose ethers, cellulose esters, cellulose amides, polyvinyl acetates, polycarboxylic acids and salts thereof, polyaminoacids, polyamides, gelatines, methylcelluloses, carboxymethylcelluloses and salts thereof, dextrans, ethylcelluloses, hydroxyethyl celluloses, hydroxypropyl methylcelluloses, maltodextrins, and polymethacrylates. Such water-soluble polymers, whether
15 PVOH or otherwise are commercially available from a variety of sources. Any of the foregoing water-soluble polymers are generally suitable for the invention. In general, the water-soluble polymer can include copolymers and/or blends of the foregoing resins.

Plasticizer

Preferably the composite comprises a plasticiser. Preferably the plasticiser is present at from
20 0.1 to 25% wt. of the film. More preferably the plasticiser is present at from 0.5 to 20% wt., even more preferably at from 1.0 to 15% wt. of the film. Suitable plasticisers include glycerol and sorbitol.

Optional ingredients

The composite may comprise one or more optional ingredients. Examples of such ingredients
25 include, but are not limited to, an antioxidant, a foaming agent, a dye, fillers, colourants, a cross-linking agent, an inhibitor, pigment, stabiliser, biocidal and/or an accelerator. The composite may comprise conventional additives used in packaging industry, such as enhancers, mold releasing agents, coating materials, humectants, sealing materials, thickening agents, diluting agents, binders etc. Typically, antioxidants help in preventing degradation of
30 polymer during processing. An example of antioxidant suitable for the present invention is Chemtura Corporation's Naugard B25 (a mixture of tris (2,4-di-tert-butyl phenyl) phosphine and tetrakis methylene (3,5-di-tert-butyl-4-hydroxyhydrocinnamate) methane).

Foaming agent is added to decrease density of resin composition by foaming. Examples of foaming agents include Chemtura Corporation's Celogen TSH (toluene sulfonyl hydrazide), Celogen AZ (azodicarbonamide), Celogen OT (p-p'-oxybis(benzenesulfonylhydrazide)), Celogen RA (p-toluene sulfonyl semicarbazide), Opex 80 (dinitrosopentamethylenetetramine),
5 and Expandex 5-PT (5-phenyltetrazole).

Colorants are added in packaging for improving aesthetic. Colorants are typically either pigments or dyes. Dyes are generally organic compounds and soluble in the polymer resin. On the other hands, pigments are generally dispersed in the polymer resin, and they can be organic or inorganic compounds and are viable in a variety of forms including dry powders,
10 colour concentrates, liquids, and precolor resin pellets. Examples of inorganic pigments include oxides, sulfides, chromates, and other complexes based on a heavy metal such as cadmium, zinc, titanium, lead, molybdenum, iron, combinations thereof, sulfide-silicate complexes containing sodium and aluminum. Often pigments comprise mixtures of two, three or more oxides of iron, barium, titanium, antimony, nickel, chromium, lead, in specific ratios. Titanium
15 dioxide is a widely used and known bright white thermally stable inorganic pigment. Other known organic pigments include azo or diazo pigments, pyrazolone pigments, permanent red 2B, nickel azo yellow, litho red, and pigment scarlet. Cross-linking agents can optionally be added to strengthen the bond between recycled polyolefin, as described above, into a final homogenous product. Cross-linking agent bonds across the pendent hydroxyl groups on the
20 polyolefin chain. Cross-linking agents must have the characteristics of forming a strong bond at relatively low temperatures. Examples of cross-linking agents that can be used include polyurethanes such as isocyanate, phenolic resin, unsaturated polyester and epoxy resin and combinations thereof. Phenolic resin may be any single stage or two-stage resin, preferably with a low hexane content. Inhibitors can be added to retard the speed of the cross-linking
25 reaction. Examples of known inhibitors include organic acids, such as citric acid. Accelerators can be added to increase the speed of the cross-linking reaction. Examples of accelerators include amine catalysts such as Dabco BDO (Air Products), and DEH40 (Dow Chemical). The amounts of the various additional ingredients in the composition can be adjusted by those skilled in the art depending on the specific materials being used and the intended use of the
30 material. Another kind of components optionally present in the compositions of the present invention are reinforcing agents, in particular mineral fillers, like talc or calcium carbonate, or fibres, like glass fibres. The composite may further comprise various polymer components and with polar additives, such as, fillers and fibres.

Application

In a preferred aspect, the present invention provides a container comprising the packaging composite according to the present invention.

Preferably, the container is a substantially rigid container such as a bottle or jar and provides structural support to maintain its own shape. Preferably the container is in the form of bottle,
5 jar, jug etc. Preferably the container is provided with a cap. Preferably the container comprises polymer materials such as HDPE, polypropylene, polystyrene etc. Preferably the polymers constitute the body of the container.

In another aspect, the container may preferably be flexible containers such as a bottle, which may be squeezed to eject the container's contents when desired. Preferably the container
10 comprises flexible polymeric material.

In another preferred aspect, the container is a flexible container such that sachet, pouch, bag, stand-up pouch, tubes, gusseted pouch and other flexible pouch known in the art. Preferably the flexible container comprises polymer materials such as LLDPE, LDPE etc.

In another preferred aspect, the present invention provides a packaging film comprising the
15 packaging composite. Preferably the packaging film comprises more than one layers. Preferably the packaging film is laminated with one or more polymeric layer, such as polyethene, polypropylene, biaxially oriented polypropylene etc. Such packaging film may be adhered to a container, preferably from outside. It may also be possible to use the packaging film to wrap a product such as detergent tablets, soap bar to reduce water transfer where
20 possible. This helps in maintaining the integrity of the product in the wrapping and without the need for expensive secondary packaging. Preferably the packaging film is printed on one or both surfaces providing information about the product to consumers. Preferably the print comprises a UV curable ink.

In another preferred aspect, the packaging film comprises water-soluble polymer such as
25 polyvinyl alcohol and preferably used for unit dose applications. Often a unit does product contain an amount of product suitable for single use, and dissolve, as such, in water forming a liquid composition for subsequent use.

Preferably the composite is suitable for packaging various consumer products such as detergent powder, detergent bar, laundry liquid detergent, fabric conditioner, dish washing
30 liquid, hard surface cleaner, machine dish washing tablets in home care domain. In another scenario the composite may be used for packaging wide range of personal care products

including skin cream, sunscreen, bathing bar, body lotions, shampoo, conditioner, tooth paste etc.

The present invention also provides a process for providing a packaging composite according to the invention comprising steps of:

- 5 a) providing a polymer in liquid form;
- b) mixing graphene or graphene derivative and a slip additive with the polymer; and
- c) extruding the polymer to form the composite.

10 In the process, graphene or graphene derivative and slip additive is added directly to the polymer, unlike conventional process of mixing additives in a masterbatch and subsequently adding to the polymer. Preferably the polymers, such as, polyesters, polyolefins, polyamides, polystyrene, polyanhydrides, polyacrylates, polyhydroxy-alkanoates, poly vinyl chloride, thermoplastic polyurethanes are melted to liquid state by heating, subsequently and graphene or graphene derivative and slip additive are mixed in thoroughly in the polymer melt.

15 Preferably the composite is used in blow moulding extruder to form packaging films. Preferably such films are laminated with one or more packaging layers, for further improving the performance.

Alternatively, the polymer may be solubilized in a suitable solvent to facilitate the mixing process. In such scenario casting the composite to form a packaging embodiment may also be possible.

20 The invention is now be illustrated by means of following non-limiting examples.

Examples

Following materials were used for preparing various examples:

Table 1

Material	Supplier
Graphene Oxide	Platonic Nanotech Pvt. Ltd.
Erucamide (slip additive)	Croda Chemicals India.
Polyethylene polymer resin (PE)	Haldia Petrochemicals Ltd.
Biaxially Oriented Polypropylene (BOPP)	Reliance Industries Ltd.

Different Examples were prepared according to below recipe:

Table 2

Material (%w/w)	Ex-A	Ex-B	Ex-C	Ex-1	Ex-2
Graphene oxide	0	0.1	0	0.25	0.1
Erucamide	0	0	2	1.5	1
Polyethylene (PE)	100	Up to 100	Up to 100	Up to 100	Up to 100

Graphene oxide and erucamide according to above recipe were added to the polyethylene resin and mixed thoroughly in the temperature range of 120 to 130 °C forming a homogenised mixture. A preliminary film was made using Three Layer Blown Film Extrusion Line (Rajoo
 5 Engineers Ltd.) and subsequently, the preliminary film was laminated with a biaxially oriented poly propylene (BOPP) using Adhesive Lamination Instrument (Nordmecannica, Italy) producing the films of examples Ex- A to C and Ex- 1 to 2.

Ex- 1 and 2 are according to the present invention and Ex- A to C are comparative.

Various mechanical parameters and moisture barrier parameters were measured on the film
 10 following ASTM method as mentioned below:

Table 3

Parameters	Method	Instrument
Thickness	ASTM*-F2251	Mitutoyo thickness tester
Tensile strength & elongation	ASTM*-D882	Presto - Zeus Ultimo (Cap: 50 Kgf.)
WVTR (Water Vapour Transmission Rate)	ASTM*-F 1249	Mocon WVTR tester.

*ASTM - American Society for Testing and Materials

Results are summarized below:

Table 4

Parameters	Ex-A	Ex-B	Ex-C	Ex-1	Ex-2
Thickness (micron)	67.0	64.0	67.5	68.0	63.5
GSM (Gram per Square Meter)	55.4	54.1	59.1	56.2	55.4
Tensile strength (N/mm ²)	70.0	71.8	77.6	91.8	80.1
Elongation (%)	12.1	8.5	9.1	15.6	13
WVTR (Water Vapour Transmission Rate) (gm/m ² /24hr)	4.0	2.6	2.4	2.5	2.5

Higher values of tensile strength and elongation of films with similar physical characteristics (i.e., thickness, GSM) is better.

5 Form table 4, it is evident that Ex- 1 and 2 provide higher values for tensile strength, elongation indicating improved mechanical properties compared to Ex-A to C. Further Ex 1 and 2 also shows lower water vapour transmission rate and higher elongation compared to Ex-A, and it is comparable with Ex- B and C.

Claims

1. A packaging composite comprising:
 - a) 0.01 to 1.0 % by weight of graphene or derivative thereof;
 - b) 1.0 to 10 % by weight of a slip additive; and
 - c) a polymer,wherein the slip additive is a fatty acid amide.
2. A composite as claimed in claim 1 wherein the polymer is selected from polyesters, polyolefins, polyamides, polystyrene, polyanhydrides, polyacrylates, polyhydroxyalkanoates, poly vinyl chloride, thermoplastic polyurethanes, polycarbonate, polylactic acid, and combinations thereof.
3. A composite as claimed in claim 2 wherein the polymer is selected from polyolefins, polypropylenes and their combination.
4. A composite as claimed in claim 1 wherein the polymer comprises polyvinyl alcohol or derivatives thereof.
5. A composite as claimed in any one of claims 1 to 4 wherein the amount of said graphene or derivative thereof is in the range from 0.01 to 0.8 % by weight of the composite.
6. A composite as claimed in any one of claims 1 to 5 wherein the graphene derivative is graphene oxide.
7. A composite as claimed in any one of claims 1 to 6 wherein the amount of said slip additive is in the range from 1.2 to 8 % by weight of the composite.
8. A composite as claimed in any one of claims 1 to 7 wherein the fatty acid amide comprises 10 to 30 carbon atoms.
9. A composite as claimed in claim 8 wherein the fatty acid amide is selected from erucamide, oleamide, stearamide and combinations thereof.
10. A packaging film comprising a packaging composite as claimed in any one of claims 1 to 9.
11. A container comprising a packaging composite as claimed in any one of claims 1 to 9.

12. A process for providing a packaging composite as claimed in any one of claims 1 to 9 comprising steps of:
- a) providing a polymer in liquid form;
 - b) mixing graphene or derivative thereof and a slip additive with the polymer; and
 - c) extruding or casting the polymer to form the composite.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2023/082654

A. CLASSIFICATION OF SUBJECT MATTER		
INV.	C08K3/04	C08L23/06
ADD.	C08J5/18	B32B27/18
		C08K5/20
		B32B27/32
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) C08K C08L 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		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CN 112 644 123 A (XIAMEN CHANGSU IND CO LTD) 13 April 2021 (2021-04-13) paragraphs [0001] - [0002]; claims; examples 1-4 -----	1-12
A	CN 111 171 433 A (SHANTOU JINPING DONGXING PACKAGING CO LTD) 19 May 2020 (2020-05-19) claims -----	1-12
A	CN 111 976 245 A (WUXI CHUN YU ENVIRONMENTAL PROT PRODUCT CO LTD) 24 November 2020 (2020-11-24) claims; example 3 ----- --/--	1-12
☒ 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 "E" earlier application or patent but published on or after the international filing date "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) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "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 "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 "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 "&" document member of the same patent family		
Date of the actual completion of the international search 15 January 2024		Date of mailing of the international search report 25/01/2024
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Masson, Patrick

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2023/082654

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CN 111 976 244 A (WUXI CHUN YU ENVIRONMENTAL PROT PRODUCT CO LTD) 24 November 2020 (2020-11-24) cited in the application claims; examples -----	1-12
A	CN 109 627 574 A (GUANGZHOU YUYI PACKAGING MAT CO LTD) 16 April 2019 (2019-04-16) claims; examples -----	1-12

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2023/082654

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
CN 112644123	A	13-04-2021	NONE	

CN 111171433	A	19-05-2020	NONE	

CN 111976245	A	24-11-2020	NONE	

CN 111976244	A	24-11-2020	NONE	

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