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(54) Title: ELECTRICALLY INSULATED HIGH THERMAL CONDUCTIVE POLYMER COMPOSITION

(57) Abstract: An electrically insulating highly thermally conductive resin composition includes (A) 100 parts by weight of a polyamide-based resin, and (B) 10 to 80 parts by weight of a long metal fiber including a metal selected from the group consisting of copper, nickel, aluminum, iron, chromium, molybdenum, and alloys thereof. The electrically insulating highly thermally conductive resin composition shows high hardness and high strength as well as an excellent electrically insulating property and thermal conductivity, so as to be applicable to various molded products requiring high thermal conductivity and excellent mechanical characteristics.



【SPECIFICATION】**ELECTRICALLY INSULATED HIGH THERMAL CONDUCTIVE
POLYMER COMPOSITION****【Technical Field】**

5 The present disclosure relates to an electrically insulating highly thermally conductive resin composition. More particularly, the present invention relates to an electrically insulating highly thermally conductive resin composition having excellent thermal conductivity, high hardness, and good mechanical characteristics.

10 **【Background Art】**

 Thermal conductivity materials tend to be widely used due to an increase of power consumption of electronic parts/devices. The conventional thermally conductive material is mainly composed of a metal that has low formability and productivity, and that has a limit in designing parts. Therefore,
15 endeavors have been undertaken to substitute the metal with a material having excellent formability and productivity. In addition, there have been many efforts to overcome the demerits of metal for developing thermally conductive materials by using polymers, and some metals have been successfully substituted.

20 The biggest merit of using thermally conductive polymer materials is to enable achievement of high productivity and fine design by using the injection molding method. Since a thermally conductive polymer material that can be substituted for metal has limits in that the thermal conductivity reaches about 10

[W/mK] at most, metal should still be used for parts requiring high thermal conductivity.

The thermally conductive polymer material is developed mainly through combining a polymer/thermally conductive filler, but other methods of remarkably increasing the thermal conductivity of polymer materials have rarely been found and developed. A general polymer material has thermal conductivity of 0.1 to 0.4 [W/mK], which is a thermal insulator value. When thermally conductive fillers are combined therewith, it is possible to provide thermal conductivity of up to 10 [W/mK], but the viscosity is remarkably increased and the mechanical properties are highly decreased, so that it is substantially hard to obtain merits of a thermally conductive polymer material. Recently, in order to secure an appropriate level of physical properties and sufficient fluidity for injection molding, the development of a thermal conductive polymer material has focused on providing optimal thermal conductivity by adding a minimum amount of thermally conductive fillers, but it still has drawbacks of a low mechanical strength property compared to that of a general enforced resin composite material.

Properties of such thermally conductive resin composite materials may be roughly classified into electrically conductive and electrically insulative properties. The electrical conductivity should be suitably determined depending upon the application field to be used. For example, semi-conductor or electric/electronic parts that heat up in which the electrical interference should be prohibited should include the thermally conductive resin having an electrically insulating characteristic. Generally, as metal or graphite fillers that

are materials used for the conductive fillers in the thermally conductive resin composite materials are electrical conductors, the composite material also has electrical conductivity. The electrically insulating thermally conductive resin composite materials include ceramic fillers, and the ceramic fillers, such as BN, 5 AlN, SiC, and the like, have drawbacks of inferior thermal conductivity to those of metals or graphite, considerably high costs, and poor physical properties when combined in a resin. However, since there are no alternatives for the ceramic filler so far, the polymer/ceramic filler composite is being used to develop the electrically insulating thermally conductive resin in spite of such 10 drawbacks.

Japanese Patent Laid-Open Publication No. 2006-22130, which is one study on the conventional thermally conductive polymer composite, discloses a composite material including a low-melting point metal and metal or alloy powders for increasing the dispersing property of the metal in the crystalline 15 polymer, and including an inorganic powder having incompatibility with the metal powder and an enforcing material of a glass fiber. However, in this case, since the main thermal conductor is composed of a low-melting point metal and an inorganic powder having incompatibility with the metal powder, contact efficiency between thermally conductive fillers is decreased, and a matrix of the 20 crystalline polymer includes a high amount of materials having incompatibility with each other so as to deteriorate the physical properties.

Japanese Patent Laid-Open Publication No. 2005-074116 discloses a thermal conductive polymer composite prepared by using the expansion graphite and the general graphite in a sequential ratio of 1/9 to 5/5. However,

this technique relates to a composite material that increases thermal conductivity by adjusting the ratio of the expansion graphite to the graphite and increases the contact possibility, but it has drawbacks of high viscosity of the material and fragility, and drawbacks of slurping in which the graphite is smeared on the material surface.

In addition, U.S. Patent No. 6,048,919 discloses a technique of combining a thermally conductive filler having an aspect ratio of at least 10:1 and a thermally conductive filler having an aspect ratio of 5:1 or less in a volume ratio ranging from 30 to 60% and from 25 to 60%, respectively. However, this invention also has problems of a low possibility of contact between thermally conductive fillers.

Further, U.S. Patent No. 5,011,872 discloses a technology of combining a 130 to 260 μ m-sized ceramic filler material such as BN, SiC, and AlN with a polymer, and U.S. Patent No. 5,232,970 discloses a technology of adding at least 35 volume% of a ceramic filler that is capable of including silica in a polyamide and polybenzocyclobutene.

As another invention of an electrically insulating thermal conductive resin composite material, U.S. Patent No. 6,822,018 discloses an electrically insulating material-coated metal filler and a ceramic filler that are combined with epoxy silicon or a polyurethane binder. All the electrically insulating thermally conductive resin composite materials are developed by using ceramic fillers.

【DETAILED DESCRIPTION】

【Technical Problem】

An exemplary embodiment of the present invention provides an electrically insulating highly thermally conductive resin composition. Another embodiment of the present invention provides an electrically insulating highly thermally conductive resin composition having an excellent electrically
5 insulating property and high thermal conductivity, and simultaneously excellent mechanical characteristics such as high hardness and high strength, by using the electric insulating characteristic of a metal composite and a polyamide-based resin and further by being enforced with a long metal fiber. Yet another embodiment of the present invention provides a molded product obtained from
10 the electrically insulating highly thermally conductive resin composition.

The embodiments of the present invention are not limited to the above technical purposes, and a person of ordinary skill in the art can understand other technical purposes.

【Technical Solution】

15 According to one embodiment of the present invention, an electrically insulating highly thermally conductive resin composition is provided that includes (A) 100 parts by weight of a polyamide-based resin, and (B) 10 to 80 parts by weight of a long metal fiber including a metal selected from the group consisting of copper, nickel, aluminum, iron, chromium, molybdenum, and alloys
20 thereof.

The electrically insulating highly thermally conductive resin composition may include a low-melting point metal (C) in an amount of 0.5 to 10 parts by weight based on 100 parts by weight of the polyamide-based resin (A).

According to another embodiment of the present invention, a molded product fabricated using the electrically insulating highly thermally conductive resin composition is provided.

Hereinafter, further embodiments of the present invention will be
5 described in detail.

【Advantageous Effects】

The electrically insulating highly thermally conductive resin composition according to the present exhibits high hardness and high strength as well as an excellent electrically insulating property and thermal conductivity, so it is
10 applicable to various molded products requiring excellent mechanical characteristics as well as high thermal conductivity.

【Best Mode】

Exemplary embodiments of the present invention will hereinafter be described in detail. However, these embodiments are only exemplary, and the
15 present invention is not limited thereto.

The electrically insulating highly thermally conductive resin composition according to one embodiment of the present invention includes (A) 100 parts by weight of a polyamide-based resin, and (B) 10 to 80 parts by weight of a long metal fiber including a metal selected from the group consisting of copper,
20 nickel, aluminum, iron, chromium, molybdenum, and alloys thereof.

Exemplary components included in the electrically insulating highly thermally conductive resin composition according to embodiments of the present invention will hereinafter be described in detail.

(A) Polyamide-based resin

The polyamide-based resin means that an amide (-NHCO-) group is bound to the polymer main chain. Specific examples may be selected from the group consisting of polycaproamide (nylon 6), polytetramethylene adipamide (nylon 46), polyhexamethylene adipamide (nylon 66), poly(hexamethylene nonanediamide) (nylon 69), poly(hexamethylene sebacamide) (nylon 610), poly(hexamethylenedodecanediamide), polyhexamethylene dodecanamide (nylon 612), nylon 611, nylon 1212, nylon 1012, polyundecanoamide (nylon 11), polydodecanamide (nylon 12), polyhexamethylene terephthalamide (nylon 6T), polyhexamethylene isophthalamide (nylon 6I), nylon 9T, nylon 10T, polyundecamethylene terephthalamide (nylon 11T), nylon 12T, nylon 12I, polyphthalamide (PPA), a polycaproamide/polyhexamethylene adipamide copolymer (nylon 6/66), a polyhexamethylene terephthalamide/polyhexamethylene isophthalamide copolymer (nylon 6T/6I), a polyhexamethylene adipamide/polyhexamethylene terephthalamide copolymer (nylon 66/6T), poly-bis-(4-aminocyclohexyl)methanedodecamide (nylon PACM12), poly-bis-(3-methyl-4-aminocyclohexyl)methanedodecamide (nylon dimethyl PACM12), polymetaxylene adipamide (MXD 6), polyundeca methylene hexahydroterephthalamide (nylon 11T(H)), and combinations thereof.

Alternatively, the polyamide-based resin may be a blend including a polyamide-based polymer resin. Examples of the blend include polyphthalamide (PPA)/polyphenylene ether (PPE), polyamide (PA)/polyphenylene sulfide (PPS), polyamide (PA)/acrylonitrile-butadiene-styrene (ABS), and the like.

The conventional polymer/metal composite material has electrical conductivity, but the polyamide-based resin exhibits an electrically insulating property when it is used with a certain level or less of a metal component by including a polar amide (-NHCO-) group, due to the phenomenon of trapping
5 electrons. Accordingly, the polyamide-based resin is very suitable for a resin composition requiring both an electrical insulating property and high thermal conductivity together with excellent formability.

According to one embodiment, it includes nylon 66 or PPA. The PPA is advantageously applicable to a material requiring high thermal resistance used
10 for electrical insulation/thermal conductivity since it has high thermal resistance. The material uses may include a connector, a lamp socket, a lamp reflector, and the like, which are applied in a lead-free soldering process.

(B) Long metal fiber

The long metal fiber plays a role of improving the hardness and the
15 strength characteristics of the composition, and it is formed as roving that is bundled with a plurality of metal fibers having a diameter of 1 to 65 μ m and that is capable of being continuously immersed into the resin in a glass roving device.

The long metal fiber according to one embodiment of the present
20 invention may be prepared from one selected from the group consisting of copper, nickel, aluminum, iron, chromium, molybdenum, and alloys thereof. According to one embodiment, it includes one prepared from aluminum, or a

metal alloy referred to as stainless steel such as copper/nickel/molybdenum, nickel/chromium/iron and the like.

The long metal fiber is included at 10 to 80 parts by weight based on 100 parts by weight of a polyamide-based resin, and in one
5 embodiment, it is included at 50 to 75 parts by weight. When the long metal fiber is included within the range, it is possible to secure an excellent property balance of mechanical strength and impact strength.

(C) Low-melting point metal

The electrically insulating highly thermally conductive resin composition
10 according to the present invention may further include a low melting point metal.

The low-melting point metal plays a role of maximizing contact between thermal conductive fillers, and may be one kind of metal or a solid solution consisting of two or more kinds of metal elements. In one embodiment, it is a solid solution.

15 According to one embodiment, the low-melting point metal of the solid solution is a metal solid solution having a lower solidus temperature than the melting point of the crystal polymer of the (A) polyamide-based resin. Particularly, when the solidus temperature is lower than the melting point of the polyamide-based resin at 20° C or more, it improves the stability during
20 preparation of the electrically insulating highly thermal conductive resin composition. The solidus temperature is preferably at least 100 ° C higher than the environment of using the composition.

The solid solution of low-melting point metal includes a first metal element selected from the group consisting of tin, bismuth, lead, and combinations thereof, and a second metal element selected from the group consisting of copper, aluminum, nickel, silver, germanium, indium, zinc, and combinations thereof. For example, when the aluminum is a thermally conductive filler, the aluminum is included to the component of solid solution; in addition, when copper is a thermally conductive filler, the copper is included to a component of solid solution. Furthermore, it is advantageous to use tin for the first metal element considering the environmental view.

In addition, it is possible to control a physical property such as solidus temperature, liquidus temperature, mechanical strength, and the like of the low-melting point metal by adjusting the amount ratio of the first metal element and the second metal element. According to one embodiment, the first metal element and the second metal element are included in a weight ratio of 99.5:0.5 to 89:11. When they are included in the weight ratio range, it is possible to provide a low-melting point metal with the optimal solidus temperature.

The low-melting point metal according to one embodiment of the present invention is added at 0.5 to 10 parts by weight based on 100 parts by weight of polyamide-based resin, but in another embodiment, it ranges from 0.5 to 5 parts by weight. When the low-melting point metal is added within the range, it is mixed well during the preparation of the resin composition, so as to improve the formation of the network and thermal conductivity. Therefore, it is advantageous for the balance of thermal conductivity improvement.

(D) Other additives

Other electrically insulating highly thermally conductive resin compositions according to one embodiment of the present invention may further include other additives such as glass fiber, glass beads, calcium carbonate (CaCO_3), and the like in order to maximize contact between thermally
5 conductive fillers or improve the mechanical property of the resin composition.

In addition, it may further include an antioxidant, a weather resistance agent, a flame retardant, a release agent, a lubricant, a colorant, and the like.

The antioxidant may include phenol, phosphite, thioether, or amine antioxidants, and the weather-resistance agent may include benzophenone or
10 amine weather-resistance agents.

The flame retardant is not specifically limited, but it may include a halogen-based, phosphorous, metal salt-based, or silicone-based flame retardant.

The release agent may include a fluorine-included polymer, silicone oil,
15 a metal salt of stearic acid, a metal salt of montanic acid, montanic acid ester wax, or a polyethylene wax, and the colorant may include a dye or pigment.

The electrically insulating highly thermally conductive resin composition may be prepared in accordance with the general method of preparing a resin
20 composition. For example, it may be prepared by simultaneously mixing the constituting components and other additives, and fusion-extruding in an extruder to provide a pellet shape.

When the constituting components are mixed, the long metal fiber may be charged by inputting it into the same inlet of the mixture of other components

or in an inlet other than that of the mixture. According to one embodiment, it uses a glass roving device in which the roving long metal fiber is charged by continuously adding it to the fused mixture, and a plurality of metal fiber strands are used.

5 The electrically insulating highly thermally conductive resin composition according to one embodiment of the present invention may be prepared by the method including the step of charging a long metal fiber in a mixture including the polyamide-based resin and selectively the low-melting point metal to provide a pellet. According one embodiment, the charging step of the long-metal fiber
10 is performed in a glass roving device by melting the mixture including the polyamide-based resin and selectively the low-melting point metal and continuously adding a long-metal fiber having a roving shape to the melted mixture.

 The pellet obtained from the method has a shape in which the fiber is
15 oriented in a certain direction. In one embodiment, it had a length ranging from 5 to 30mm, and in another embodiment, the length ranges from 10 to 15 mm. Since it is formed to have such a length range, it enforces the hardness and impact resistance strength of the resin composition, and it causes fewer problems in input.

20 The electrically insulating highly thermally conductive resin composition may be used for forming various articles, and particularly, a main body, a chassis, or a heat dissipating plate and the like of electro-electronic products such as TVs, a computers, a mobile phones, and office automating devices requiring an excellent electrical insulating property and high thermal conductivity.

In addition, it is applicable to lamp sockets, lamp reflectors, electron chip sockets, connector, and the like requiring electrical insulation/thermal conduction characteristics. According to another embodiment of the present invention, provided is a molded product fabricated by the electrically insulating
5 highly thermally conductive resin composition.

【Mode for Invention】

The following examples illustrate the present invention in more detail. However, they are exemplary embodiments of the present invention and are not limiting.

10 [Examples]

The (A) polyamide-based resin, (B) long metal fiber, and (C) low-melting point metal used in examples and comparative examples are as follows.

(A) Polyamide-based resin

PPA (polyphthalic amide) having a melting point (T_m) of 300° C and a
15 glass transition temperature (T_g) of 125° C was used as polyamide-based resin.

(A') Polyphenylenesulfide resin

A polyphenylenesulfide (PPS) resin having a melting point of 280° C was used.

(B) Long metal fiber

20 A SUS316L stainless steel roving strand composed of 1000 bundles having a diameter of 8 μ m was used as a long metal fiber. The long metal fiber had an elastic coefficient of 197 GPa and a tensile strength of 485 MPa.

(C) Low-melting point metal

A tin/aluminum low-melting point metal having a main component of tin was used. The tin/aluminum solid solution that was used included tin mixed at a weight ratio of 99.7 wt% and aluminum mixed at a weight ratio of 0.3 wt%, and the solidus temperature thereof was 228° C.

5 (D-1) Ceramic inorganic filler

Boron nitride (BN) having an average particle diameter of 45 μ m was used as a ceramic inorganic filler.

(D-2) Ceramic inorganic filler

Aluminum nitride (AlN) having an average particle diameter of 2.5 μ m
10 was used as a ceramic inorganic filler.

Examples 1 to 3 and Comparative Example 1

With the above-mentioned components, pellets of each of highly thermally conductive resin compositions according to Examples 1 to 3 and Comparative Example 1 were prepared in accordance with the compositions
15 shown in Table 1. They were fabricated by using a glass roving device with a plurality of bundled fiber strands, and particularly by immersing long metal fibers in the resin melted in a resin bath of the glass roving device, extruding, and cold-cutting the same.

Comparative Examples 2 and 3

20 Each component was mixed in a composition shown in the following Table 1 in a mixer and extruded by using a twin screw extruder of L/D=35, Φ =45mm under the condition of a process temperature suitable for each resin (320° C in the case of PPA, and 300° C in the case of PPS), a screw rotation

speed of 150 rpm, the first vent pressure of about -600mmHg, and a self-supplying speed of 60 kg/h. The extruded strand was cooled in water and cut into pellets by a rotary cutter.

Table 1

5 (units: parts by weight)

	Examples			Comparative Examples		
	1	2	3	1	2	3
(A) polyamide-based resin	100	100	100	-	100	100
(A') polyphenylenesulfide resin	-	-	-	100	-	-
(B) long metal fiber (SUS316L)	29	41	52	52	-	-
(C) low melting point metal (Sn/Al)	2	2	2	2	-	-
(D-1) inorganic filler (BN)	-	-	-	-	40	-
(D-2) inorganic filler (AlN)	-	-	-	-	-	40

Pellets obtained from Examples 1 to 3 and Comparative Examples 1 to 3 were dried with hot air at 80° C for about 5 hours and extruded in a 10 oz extruder to provide a specimen for determining physical properties.

10 The obtained specimen for determining physical properties was measured in accordance with the following method, and the results are shown in the following Table 2.

(1) Thermal Conductivity: measured in accordance with guarded heat flow method.

(2) Electrical characteristic: determined by measuring surface resistance in accordance with ASTM D257.

(3) Mechanical properties: determined by measuring flexural modulus and flexural strength in accordance with ASTM D790.

5 Table 2

	Example			Comparative Example		
	1	2	3	1	2	3
Thermal conductivity [W/mK]	0.9	1.8	2.8	2.9	1.7	1.0
Surface resistance [$\Omega/\square$]	$\geq 10^{12}$	$\geq 10^{12}$	$\geq 10^{12}$	$\leq 10^3$	$\geq 10^{12}$	$\geq 10^{12}$
Flexural modulus [kgf/cm ²]	142,000	168,000	189,000	185,000	115,000	200,000
Flexural strength [kgf/cm ²]	1,600	2,650	2,700	2,600	770	650

As shown in Table 2, each specimen obtained by the compositions according to Examples 1 to 3 showed excellent thermal conductivity, electrically insulating property, and mechanical characteristic balance, but Comparative Example 1 in which polyphenylenesulfide was used instead of a polyamide-based resin showed low surface resistance, which means that the electrically insulating property thereof was deteriorated. Comparative Examples 2 and 3 in which the low-melting point metal and long metal fiber were not included and

ceramic fillers were combined in the polymer showed significantly lower flexural strength than those of Examples 1 to 3, so it is understood that the mechanical characteristics of Comparative Examples 2 and 3 were deteriorated.

From comparing the results of the examples to those of the comparative
5 examples, it is confirmed that the polyamide-based resin/low-melting point metal/long metal fiber resin composite material had excellent mechanical strength and hardness, excellent thermal conductivity, and simultaneously good electrically insulating characteristics.

The present invention is not limited to the embodiments illustrated with
10 the drawings and tables, but can be fabricated with various modifications and equivalent arrangements included within the spirit and scope of the appended claims by a person who is ordinarily skilled in this field. Therefore, the aforementioned embodiments should be understood to be exemplary but not limiting the present invention in any way.

【CLAIMS】**【Claim 1】**

An electrically insulating highly thermally conductive resin composition, comprising:

- 5 (A) 100 parts by weight of a polyamide-based resin; and
- (B) 10 to 80 parts by weight of a long metal fiber selected from the group consisting of copper, nickel, aluminum, iron, chromium, molybdenum, and an alloy thereof.

10 **【Claim 2】**

The electrically insulating highly thermally conductive resin composition of claim 1, wherein the electrically insulating highly thermally conductive resin composition further comprises (C) 0.5 to 10 parts by weight of a low-melting point metal based on 100 parts by weight of the polyamide-based resin (A).

15

【Claim 3】

The electrically insulating highly thermally conductive resin composition of claim 1, wherein the long metal fiber (B) is included in an amount of 50 to 75 parts by weight based on 100 parts by weight of the polyamide-based resin (A).

20

【Claim 4】

The electrically insulating highly thermally conductive resin composition of claim 1, wherein the polyamide-based resin (A) comprises one selected from

the group consisting of nylon 6, nylon 46, nylon 66, nylon 69, nylon 610, nylon 612, nylon 611, nylon 1212, nylon 1012, nylon 11, nylon 12, nylon 6T, nylon 6I, nylon 9T, nylon 10T, nylon 11T, nylon 12T, nylon 12I, polyphthalamide (PPA), nylon 6/66, nylon 6T/6I, nylon 66/6T, poly-bis-(4-aminocyclohexyl)methanedodecamide (nylon PACM12), nylon dimethyl PACM12, polymetaxylene adipamide (MXD 6), nylon 11T(H), and combinations thereof.

【Claim 5】

10 The electrically insulating highly thermally conductive resin composition of claim 2, wherein the low-melting point metal (C) has a lower solidus temperature than a melting point of the polyamide-based resin.

【Claim 6】

15 The electrically insulating highly thermally conductive resin composition of claim 2, wherein the low-melting point metal (C) comprises a first metal element selected from the group consisting of tin, bismuth, lead, and combinations thereof, and a second metal element selected from the group consisting of copper, aluminum, nickel, silver, germanium, indium, zinc, and
20 combinations thereof.

【Claim 7】

The electrically insulating highly thermally conductive resin composition of claim 1, wherein the long metal fiber (B) is a roving strand bundled with a plurality of metal fibers having a diameter of 1 to 65 μ m.

5

【Claim 8】

A method of fabricating an electrically insulating highly thermally conductive resin composition comprising

charging 10 to 80 parts by weight of long metal fiber in a mixture
10 comprising 100 parts by weight of a polyamide-based resin to provide a pellet.

【Claim 9】

The method of fabricating an electrically insulating highly thermally conductive resin composition of claim 8, wherein the mixture further comprises
15 a low-melting point metal at 0.5 to 10 parts by weight based on 100 parts by weight of the polyamide-based resin.

【Claim 10】

The method of fabricating an electrically insulating highly thermally
20 conductive resin composition of claim 8, wherein the long metal fiber charging step is performed by melting the mixture comprising the polyamide-based resin, and continuously immersing roved long metal fiber in the melted mixture.

【Claim 11】

A pellet manufactured from the electrically insulating highly thermally conductive resin composition according to any one of claims 1 to 7.

5 **【Claim 12】**

The pellet of claim 11, wherein the pellet has a length of 5 to 30 mm.

【Claim 13】

10 A molded product fabricated using the electrically insulating highly thermally conductive resin composition according to one of claims 1 to 7.

A. CLASSIFICATION OF SUBJECT MATTER***C08L 77/00(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)

IPC 8: C08L 77/00

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)

eKOMPASS(KIPO internal), PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	JP03-079663 A (TEIJIN LTD) 04 Apr. 1991 Whole document	1-13
A	EP00997496 B1 (TORAY INDUSTRIES, INC.) 01 Mar. 2006 Whole document	1-13



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

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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