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(54) Title: LOW WARP POLYBUTYLENE TEREPHTHALATE MOLDING COMPOSITIONS

(57) Abstract: A low warp, reinforced polybutylene terephthalate molding composition includes: from about 30 weight percent to about 70 weight percent of polybutylene terephthalate resin; from about 10 weight percent to about 30 weight percent of a styrene-acrylonitrile copolymer; from about 15 to about 60 weight percent of a reinforcing agent; and optionally includes one or more impact modifiers, lubricants and stabilizers.



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LOW WARP POLYBUTYLENE TEREPHTHALATE MOLDING
COMPOSITIONS

5 Technical Field

The present invention relates generally to reinforced thermoplastic molding compositions. More specifically the invention relates to glass fiber reinforced polybutylene terephthalate molding compositions provided with a warp-reducing amount of styrene-acrylonitrile copolymer.

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Background

General background with respect to blended thermoplastic molding compositions is seen in United States Patent No. 4,665,122 entitled "Polycarbonate Blends" of *Robeson et al.*. The '122 patent discloses a blend of polycarbonate and a styrenic polymer. The styrenic polymer used is a styrene acrylonitrile copolymer (SAN). The ratio of styrene copolymer to the polycarbonate varies according to end properties sought and in the usual case is from about 25 to 75 weight percent of the polycarbonate to about 75 to 25 weight percent of the styrenic polymer. Typical additives include UV stabilizers, antioxidants, flame retardants, inorganic fillers, fiberglass, and carbon fibers.

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Another thermoplastic blend is found in United States Patent No. 4,493,921 entitled "Impact Resistant Blend" of *Wefer*. Here, the blends are based on thermoplastic polyester resin such as polybutylene terephthalate, aromatic polycarbonate resin and graft copolymers of EPDM or EPM. The grafting monomer may be styrene/acrylonitrile. Preferred compositions will include a major proportion of the polyester resin component along with sufficient polycarbonate resin and graft copolymer to provide high notched impact strength.

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United States Patent No. 5,106,907 entitled "Engineering Thermoplastic with Improved Processability and Performance" of *Boutni* discloses a four component blend including an aromatic carbonate polymer, a styrene-acrylonitrile

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copolymer, a methacrylate-diene-styrene shell-core copolymer and polybutylene terephthalate "PBT". The styrene-acrylonitrile copolymer is present in about 2% to about 15% (preferably from about 4% to 10%), from about 2% to about 20% (preferably from about 3% to about 12%) of the core-shell polymer, and from
5 about 2% to about 50% (preferably for about 4% to about 15%) of the polybutylene terephthalate, with the remainder of the four-component blend being essentially the aromatic carbonate. The blend may be modified by the addition of additives such as clay or talc fillers, reinforcing agents such as glass fibers, impact
10 modifiers, other resins, plasticizers, flow promoters and other processing aids, stabilizers, colorants, mold release agents, flame retardants, ultraviolet screening agents, and the like.

Turning more specifically to PBT reinforced resin compositions, there is taught in United States Patent No. 4,900,610 to *Hochberg et al.* PBT molding
15 compositions wherein the matrix resin is augmented with cyclohexane dimethanol/terephthalic acid copolymer. As noted in Patent No. 4,900,610, reinforced PBT compositions have a tendency to warp. See Col. 1, lines 29 and following.

20 One known way to reduce warpage in PBT molding compositions is to add polycarbonate to the blend; however such compositions tend to exhibit very high melt viscosities resulting in poor mold cavity filling. Moreover, added polycarbonate tends to reduce heat resistance significantly as is seen in the example compositions described hereafter.

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Summary of Invention

It has been found in accordance with the present invention that styrene-acrylonitrile copolymer significantly reduces warp exhibited by reinforced PBT compositions. The inventive compositions do not exhibit the high melt viscosities
30 seen in PBT/polycarbonate blends and have surprisingly high deflection temperatures under load as compared with other PBT/polymer blends.

There is thus provided in accordance with the present invention a low warp, reinforced polybutylene terephthalate molding composition comprising: (a) from about 30 weight percent to about 70 weight percent of polybutylene terephthalate resin; (b) from about 10 weight percent to about 30 weight percent of a styrene-acrylonitrile copolymer; (c) from about 15 to about 60 weight percent of a reinforcing agent; and (d) optionally including one or more impact modifiers, lubricants and stabilizers. Preferably, the polybutylene terephthalate resin is present in an amount of from about 40 weight percent to about 60 weight percent and has an intrinsic viscosity of from about 0.4 to about 4.0. An intrinsic viscosity of from about 0.6 to about 1.5 is still more preferred.

Typically the styrene-acrylonitrile copolymer is present in an amount of from about 15 to about 25 weight percent and has a melt index of from about 2g/10min to about 10g/10min at 230 °C/3.8kg. A melt index of from about 2g/10min to about 9g/10min at 230 °C/3.8kg is more preferred, while a melt index of from about 3g/10min to about 3.5g/10min at 230 °C/3.8kg is still more preferred.

In most cases the amount of polybutylene terephthalate resin present is at least 1.5 times the amount of styrene-acrylonitrile copolymer present on a weight basis and preferably the amount of polybutylene terephthalate resin present is at least 2 times the amount of styrene-acrylonitrile copolymer present on a weight basis. The reinforcing agent preferably includes fiberglass and may consist of fiberglass in some cases while in others there is further provided a mineral filler.

Preferably, when molded the compositions exhibit a relative warp of less than about 150 percent and still more preferably exhibit a relative warp of less than about 125 percent. The composition generally has a melt viscosity at 250 °C and 1000 l/sec of less than about 3000 poise and preferably has a melt viscosity at 250 °C and 1000 l/sec of less than about 2500 poise. So also, the composition generally has a temperature of deflection under load of at least about 170 °C at 1.8

Mpa; typically even higher. Optionally, there is further provided to the composition an impact modifier selected from the group consisting of ionomeric impact modifiers, multiphase polymers and ethylene/acrylic copolymer impact modifiers. The impact modifier may be present in an amount of from about 2
5 weight percent to about 15 weight percent; preferably the impact modifier is present in an amount of from about 4 weight percent to about 10 weight percent.

In another aspect of the invention, there is provided a low warp, reinforced composition consisting essentially of a melt blend of polybutylene terephthalate,
10 styrene-acrylonitrile copolymer and a reinforcing agent, optionally including one or more impact modifiers, stabilizers and lubricants wherein: (i) the polybutylene terephthalate resin has an intrinsic viscosity of from about 0.4 to about 4; (ii) the styrene-acrylonitrile copolymer has a melt index of from about 2g/10 minutes to about 10g/10 minutes at 230 °C/3.8kg; and (iii) the polybutylene terephthalate
15 resin is present in an amount of from about 1.5 to about 3 times the amount of styrene-acrylonitrile copolymer present on a weight basis.

Still yet another aspect of the invention is directed to a method of making an injection molded part comprising: (a) melt blending a composition comprising:
20 (i) from about 30 weight percent to about 70 weight percent of polybutylene terephthalate resin; (ii) from about 10 weight percent to about 30 weight percent of a styrene-acrylonitrile copolymer; (iii) from about 15 weight percent to about 60 weight percent of a reinforcing agent, (iv) optionally including one or more impact modifiers, lubricants and stabilizers; and (b) injection molding the melt
25 blend of step (a) into a shaped article. Preferably, the process further includes the step of pelletizing the melt blend. Generally, the components are melt blended at a temperature of from about 215 °C to about 245 °C.

Further aspects and advantages of the present invention will become
30 apparent from the following detailed description.

Detailed Description

The invention is described in detail below with reference to numerous embodiments for purposes of exemplification and illustration only. Modifications to particular embodiments within the spirit and scope of the present invention, set
5 forth in the appended claims, will be readily apparent to those of skill in the art.

Unless more specifically defined, terminology is given its ordinary meaning. Percent, for example, refers to weight percent.

10 “Styrene-acrylonitrile copolymer” or “SAN” is a copolymer which is typically prepared by copolymerization of from about 68% to about 80% (preferably from about 70% to about 78%) styrene and from about 20% to about 32% (preferably from about 22% to about 30%) of acrylonitrile. The molecular weight of SAN can be varied within a wide range, generally from about 30,000 to
15 about 600,000. SAN is readily produced by known processes such as mass, solution, suspension, or emulsion polymerization. “SAN copolymer” as used in this specification and appended claims means SAN copolymer at least 80 mol percent of which consists of styrene and acrylonitrile derived repeating units. While the use of minor amounts of other co monomers is not excluded, the present
20 invention is not intended to encompass the use of acrylonitrile-butadiene-styrene copolymer (ABS) or other impact modifier as the SAN copolymer component recited in the claims. Such impact modifiers are not excluded as optional additional components and may be present if so desired. Two preferred SAN resins and Tyril ® 880B and 990 available from Dow Plastics. Representative
25 properties of these resins appear in Table 1 below.

Table 1- Selected Styrene/Acrylonitrile Resin Properties

Property	Test Method	Tyrl ® 880B	Tyrl ® 990
Melt Index, 230 °C/3.8kg	ASTM D 1238	3.2g/10min	8.5g/10min
DTUL at 1.8 Mpa	ASTM D 648	86 °C	97 °C
Vicat Softening Point	ASTM D 1525	108 °C	107 °C

“Polybutylene terephthalate”, “PBT” is a polyester obtained by polymerizing a glycol component, at least 70 mole preferably at least 80 mole %, of which consists of tetramethylene glycol (1,4-butanediol or 1,4-butylene glycol) and an acid component at least 70 mole %, preferably at least 80 mole %, of which consists of terephthalic acid, or polyester-forming derivatives thereof. Also contemplated are mixtures of the ester with minor amounts, e.g., from 0.5 to 2% by weight, of units derived from aliphatic or aromatic dicarboxylic acids and/or aliphatic polyols, e.g., glycols, i.e., copolyesters. These can be made by following the teachings outlined in *Whinfield et al.*, U.S. Pat. No. 2,465,316 and *Pengilly*, U.S. Pat. No. 3,047,539, for example. Among the units which can be present in the copolyesters are those derived from aliphatic dicarboxylic acids, e.g., of up to about 50 carbon atoms, including straight and branched chain acids, such as adipic acid, dimerized C₁₆-C₁₈ unsaturated acids (which have 32 to 36 carbon atoms), trimerized such acids, and the like. Among the units in the copolyesters can also be minor amounts derived from aromatic dicarboxylic acids, e.g., of up to about 36 carbon atoms, such as isophthalic acids and the like. In addition to the 1,4-butylene glycol units, there can also be minor amounts of units derived from other aliphatic glycols and polyols, e.g., of up to about 50 carbon atoms, including ethylene glycol, propylene glycol, glycerol and the like. Such copolyesters can be made by techniques well known to those skilled in the art. Poly(1,4-butylene terephthalate) copolymer is the preferred polyester and is commercially available.

These polymeric 1,4-butylene glycol terephthalates have an intrinsic viscosity of at least 0.4 and preferably at least about 0.7 deciliters/gram as measured in o-chlorophenol, a 60/40 phenol tetrachloroethane mixture or a similar solvent at 25 °-30 °C. The upper limit is not critical, but it will generally be about 4.0 dl./g. Especially preferred PBT resins will have an intrinsic viscosity in the range of about 0.7 to 2.0.

The reinforcing agents used are typically reinforcing fibers. Suitable reinforcing agents include, for example, glass fiber, carbon fiber, ceramic fiber, fibrous potassium titanate, iron whiskers, and the like. Glass is the most preferred. While fiber is the most preferred form for the reinforcing agent, other suitable forms may also be employed in the practice of the invention. Where reinforcing fibers are used, such fibers should normally have diameters between about 5 and about 30 microns, typically from 10-21 microns and preferably from 11-16 microns. Aspect ratios (ratio of length of fiber to diameter of fiber) are desirably at least about 5. The reinforcing fiber typically has a length prior to compounding of generally from less than 1-10 mm, preferably from 2-6 mm and more preferably from 3-5 mm. After compounding and/or molding, the fibers are considerably shorter, generally in the range of 0.2-0.5 mm in length with the average length typically toward the lower value of 0.2 mm. Glass fibers, where used, preferably have diameters between about 10 and about 15 microns and an initial aspect ratio of at least about 20. Fillers may also be included. Suitable fillers include, but are not limited to, mica, talcum, clay, titanium dioxide, calcium carbonate and the like. There may be variants within the same filler type such as, for example, the muscovite type mica (supplied by KMG, Inc.), the phlogopite type mica (Suzorite, Inc.) and the like. Nanofillers, that is, exfoliated minerals, are considered both reinforcing agents and mineral fillers for purposes of the present invention. Suitable nanofillers are exfoliated layered minerals including exfoliated clays such as montmorillonite, other exfoliated silicates and so forth as are known in the art.

“Impact modifier” and such terminology means and includes polymers used to toughen engineering resin compositions, including core-shell elastomers, ethylene/methacrylate copolymers, ionomers and so forth as are known in the art. Lotader ® impact modifiers, available from Atofina are used in some
5 compositions. These resins are copolymers of ethylene and acrylic esters with a reactive functionality. This reactive group can either be maleic anhydride (MAH), very polar and giving chemical reaction on -NH₂, -OH, epoxy group, or glycidyl methacrylate (GMA) of which epoxy group can react on COOH, -OH, -NH₂. The acrylic ester can be methyl, ethyl or butyl acrylate.

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Also as part of the polyester resin of the polyester composition are conventional additives known to the art. Some of them include, for example, antioxidants, stabilizers, lubricants, nucleating agents, colorants, mold release agents, ultraviolet light stabilizers, and the like. Examples of suitable antioxidants
15 include phosphites. Examples of suitable stabilizers include bis-phenol A based epoxy. Examples of suitable lubricants include olefinic waxes, EBS waxes and the like.

In preparing molded compositions of the invention, the components may
20 be intimately blended by any suitable means. Melt-compounding by extrusion at about 230 °C is preferred. The extrusion may be carried out in a suitable extruder such as for example a twin screw extruder with down-stream feeding capability. Many such extruders are commercially available such as, for example, the 40 mm Werner Pleiderer twin screw extruder. The extruder is fed with the resin
25 composition and temperatures are kept at a suitable level, for example, the temperature may range 210 °C to 250 °C. Likewise, in molding parts, barrel temperatures between about 230 °C and 275° C. are preferred. In a preferred embodiment, the molding composition of the invention is formed by extrusion and pelletized. Products of the invention are then produced by injection molding the
30 pelletized extrudate into a mold having a surface temperature of 40 °C or so.

The fiberglass reinforced polybutylene and styrene-acrylonitrile composition of the invention and optionally containing the impact modifier, when obtained by the end user, may be dried by any convenient method, re-melted and molded.

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Unless otherwise indicated, the following test procedures are used to characterize the compositions and products of the invention.

10	Relative Warp	ISO 9001/ASTM D 792
	Melt Index or Viscosity	ISO 1133/ASTM D 1238
	DTUL	ISO 75/ASTM D 648
	Tensile Properties	ISO 527/ASTM D 638
	Izod Impact Notched	ISO 180/ASTM D 256
	Shrinkage	ISO 294/ASTM D 955
15	Injection Molded Specimen Preparation	ISO 7792-2

Unless otherwise indicated, the test method version in effect on July 1, 2004 is used.

20

The following examples illustrate preferred compositions and methods of the present invention. These examples are illustrative only and do not limit the scope of the invention. All percentages are by weight unless otherwise indicated.

25 Comparative Examples A, B and invention Examples 1-4.

A variety of PBT/SAN blends reinforced with fiberglass and optional impact modifier were studied. The indicated components were incorporated into compositions at the amounts indicated in Table 2 by melt-compounding, followed by injection molding test specimens in accordance with ISO test method 7792-2.

30

In connection with Table 2 and 3 the following ingredients were used:

	Fiberglass	PPG 3540 (PPG)
5	PBT	Virgin PBT injection molding grade resin (Ticona)
	SAN	Tyrl ® 880B and 990 (Dow)
10	Polycarbonate	Recycled Polycarbonate
	Impact Modifier	Lotader ® 8930 (Atofina)
	Additives	Lubricants, antioxidants
15	“Commercial Product: indicates a commercial reinforced PBT-based, 30 percent glass fiber reinforced product marketed by Ticona, based on the same Virgin PBT resin used in the other examples.	

Property data appears for the various compositions in Table 3.

Table 2 -- Test Compositions

Component/Example	A	1	2	3	4	B
Commercial Product	100%	---	---	---	---	---
PBT	---	50%	45%	50%	45%	45%
Fiberglass (PPG 3540)	---	30%	30%	30%	30%	30%
Polycarbonate	---	---	---	---	---	20%
Tyrl 880B SAN	---	20%	20%	---	---	---
Tyrl 990 SAN	---	---	---	20%	20%	---
Impact Modifier	---	---	5%	---	5%	5%
Additives	---	---	---	---	---	---

Table 3—Properties of Compositions and Molded Products.

Example	A	1	2	3	4	B
Tensile Strength at Break (MPa)	115.7	118.9	111.5	119.8	112.3	111.4
Tensile Elongation (%)	2.3	2.0	2.2	2.0	2.1	3.2
Tensile Modulus (Mpa)	9255	9832	9664	9719	9227	8547
Notched Izod (kJ/m ²)	7.8	6.9	7.1	7.1	7.7	14.0
DTUL @ 1.80 Mpa (C°)	200	176	172	177	170	154
Melt Viscosity (poise)	2444	2470	2749	2088	2183	6207
Rate of Change (% min)	-0.68	-0.28	0.22	-0.17	0.46	-1.09
Relative Warp (%)	257	117	119	125	128	100
Shrinkage @ 4mm (mils/in)	1.1	0.9	1.3	1.3	0.9	1.0

As will be appreciated from the foregoing, compositions of the invention exhibit reduced warp, as well as melt viscosities and DTUL temperatures comparable to compositions with PBT only as the matrix resin. On the other
5 hand, the PBT/polycarbonate composition exhibited high melt viscosity and reduced deflection temperatures under load.

Articles prepared by way of the invention include many automotive applications including connectors, sensors, housings, and other products
10 benefiting from the combination of warp resistance, easy mold filling and high DTUL. Competitive products do not have this unique combination of properties.

While the invention has been described in connection with several examples, modifications to those examples within the spirit and scope of the
15 invention will be readily apparent to those of skill in the art. In view of the foregoing discussion, relevant knowledge in the art and references including co-pending applications discussed above in connection with the Background and Detailed Description, the disclosures of which are all incorporated herein by reference, further description is deemed unnecessary.

WHAT IS CLAIMED IS:

1. A low warp, reinforced polybutylene terephthalate molding composition comprising:

- 5 (a) from about 30 weight percent to about 70 weight percent of
polybutylene terephthalate resin;
- (b) from about 10 weight percent to about 30 weight percent of a styrene-
acrylonitrile copolymer;
- 10 (c) from about 15 to about 60 weight percent of a reinforcing agent; and
- (d) optionally including one or more impact modifiers, lubricants and
stabilizers.
- 15 2. The composition according to Claim 1, wherein the polybutylene terephthalate
resin is present in an amount of from about 40 weight percent to about 60
weight percent.
- 20 3. The composition according to Claim 1, wherein the polybutylene terephthalate
resin has an intrinsic viscosity of from about 0.4 to about 4.0.
4. The composition according to Claim 1, wherein the polybutylene terephthalate
resin has an intrinsic viscosity of from about 0.6 to about 1.5.
- 25 5. The composition according to Claim 1, wherein the styrene-acrylonitrile
copolymer is present in an amount of from about 15 to about 25 weight
percent.

6. The composition according to Claim 1, wherein the styrene-acrylonitrile copolymer has a melt index of from about 2g/10min to about 10g/10min at 230°C/3.8kg.
- 5 7. The composition according to Claim 1, wherein the styrene-acrylonitrile copolymer has a melt index of from about 2g/10min to about 9g/10min at 230°C/3.8kg.
8. The composition according to Claim 1, wherein the styrene-acrylonitrile
10 copolymer has a melt index of from about 3g/10min to about 3.5g/10min at 230°C/3.8kg.
9. The composition according to Claim 1, wherein the amount of polybutylene terephthalate resin present is at least 1.5 times the amount of styrene-
15 acrylonitrile copolymer present on a weight basis.
10. The composition according to Claim 1, wherein the amount of polybutylene terephthalate resin present is at least 2 times the amount of styrene-
acrylonitrile copolymer present on a weight basis.
20
11. The composition according to Claim 1, wherein the reinforcing agent comprises fiberglass.
12. The composition according to Claim 1, wherein the reinforcing agent consists
25 of fiberglass.
13. The composition according to Claim 1, further comprising a mineral filler.
14. The composition according to Claim 1, having a relative warp of less than
30 about 150 percent.

15. The composition according to Claim 1, having a relative warp of less than about 125 percent.
16. The composition according to Claim 1, having a melt viscosity at 250 °C and
5 1000 l/sec of less than about 3000 poise.
17. The composition according to Claim 1, having a melt viscosity at 250 °C and 1000 l/sec of less than about 2500 poise.
- 10 18. The composition according to Claim 1, wherein the composition has a temperature of deflection under load of at least about 170 °C at 1.8 Mpa.
19. The composition according to Claim 1, including an impact modifier selected from the group consisting of ionomeric impact modifiers, multiphase polymers
15 and ethylene/acrylic copolymer impact modifiers.
20. The composition according to Claim 19, wherein the impact modifier is present in an amount of from about 2 weight percent to about 15 weight percent.
20
21. The composition according to Claim 19, wherein the impact modifier is present in an amount of from about 4 weight percent to about 10 weight percent.
- 25 22. A low warp, reinforced composition consisting essentially of a melt blend of polybutylene terephthalate, styrene-acrylonitrile copolymer and a reinforcing agent, optionally including one or more impact modifiers, stabilizers and lubricants wherein:
- 30 (i) the polybutylene terephthalate resin has an intrinsic viscosity of from about 0.4 to about 4;

(ii) the styrene-acrylonitrile copolymer has a melt index of from about 2g/10 minutes to about 10g/10 minutes at 230 °C/3.8kg; and

5 (iii) the polybutylene terephthalate resin is present in an amount of from about 1.5 to about 3 times the amount of styrene-acrylonitrile copolymer present on a weight basis.

23. A method of making an injection molded part comprising:
10

(a) melt blending a composition comprising:

(i) from about 30 weight percent to about 70 weight percent of polybutylene terephthalate copolymer;
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(ii) from about 10 weight percent to about 30 weight percent of a styrene-acrylonitrile resin;

(iii) from about 15 weight product to about 60 weight percent of a reinforcing agent;
20

(iv) optionally including one or more impact modifiers, lubricants and stabilizers; and

25 (b) injection molding the melt blend of step (a) into a shaped article.

24. The method according to Claim 23, further comprising the step of pelletizing the melt blend.

30 25. The method according to Claim 23, wherein the components are melt blended at a temperature of from about 215 °C to about 245 °C.

INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US2005/024466

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08L67/02 C08L25/12

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 7 C08L

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 practical, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 03/085046 A (WIN TECH POLYMER LTD; MATSUSHIMA, MITSUNORI; SAKATA, KOUICHI; TAKAYAMA) 16 October 2003 (2003-10-16)	1,2,5,9, 11,12, 14,15, 23-25
X	examples 4,14 & US 2005/165176 A1 (WIN TECH POLYMER LTD; MATSUSHIMA, MITSUNORI; SAKATA, KOUICHI; TAKAYAMA) 28 July 2005 (2005-07-28)	1,2,5,9, 11,12, 14,15, 23-25
X	----- DE 198 45 317 A1 (BASF AG) 6 April 2000 (2000-04-06) page 2, line 27 - line 60 page 4, line 56 - line 67 examples 1-3 ----- -/-	1,2,5, 9-15, 18-21, 23-25

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
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INTERNATIONAL SEARCH REPORT

International Application No
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 290 864 A (MOCHIZUKI ET AL) 1 March 1994 (1994-03-01) column 2, line 3 - line 67 column 3, line 54 - line 60 column 4, line 33 - line 40 column 5, line 67 - column 7, line 2 example 15 -----	1,2,5, 9-15, 19-21, 23-25
X	US 4 968 731 A (LAUSBERG ET AL) 6 November 1990 (1990-11-06) column 1, line 1 - line 54 column 2, line 12 - line 19 column 2, line 46 - line 53 column 5, line 34 - line 52 claims; examples -----	1-25
X	US 6 605 665 B1 (FISCHER MICHAEL ET AL) 12 August 2003 (2003-08-12) column 2, line 18 - line 57 column 3, line 12 column 6, line 31 - line 54 column 7, line 15 - line 28 claims; examples -----	1-25
X	US 6 479 617 B1 (FISCHER MICHAEL ET AL) 12 November 2002 (2002-11-12) example comparative -----	1-25
X	DE 100 03 270 A1 (BASF AG) 2 August 2001 (2001-08-02) claims; examples -----	1-25
A	EP 0 614 945 A (GENERAL ELECTRIC COMPANY; GEN ELECTRIC) 14 September 1994 (1994-09-14) claims -----	1-25
A	US 4 342 846 A (SILBERBERG ET AL) 3 August 1982 (1982-08-03) column 1, line 51 - line 67; claims -----	1-25

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No.

PCT/US2005/024466

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 03085046	A	16-10-2003	CN 1646629 A	27-07-2005
			US 2005165176 A1	28-07-2005
US 2005165176	A1	28-07-2005	CN 1646629 A	27-07-2005
			WO 03085046 A1	16-10-2003
DE 19845317	A1	06-04-2000	AT 274028 T	15-09-2004
			AU 5982499 A	26-04-2000
			BR 9914113 A	12-06-2001
			CN 1323330 A	21-11-2001
			CZ 20011174 A3	14-11-2001
			WO 0020500 A1	13-04-2000
			EP 1117741 A1	25-07-2001
			ES 2226499 T3	16-03-2005
			JP 2002526625 T	20-08-2002
US 5290864	A	01-03-1994	NONE	
US 4968731	A	06-11-1990	DE 3733838 A1	20-04-1989
			EP 0310976 A2	12-04-1989
			JP 1144449 A	06-06-1989
US 6605665	B1	12-08-2003	AT 272682 T	15-08-2004
			AU 2541600 A	18-08-2000
			BR 0007753 A	30-10-2001
			CN 1345354 A	17-04-2002
			CZ 20012737 A3	14-11-2001
			DE 19903073 A1	10-08-2000
			WO 0044831 A1	03-08-2000
			EP 1153083 A1	14-11-2001
			ES 2223457 T3	01-03-2005
			JP 2002535471 T	22-10-2002
US 6479617	B1	12-11-2002	AU 4547000 A	10-11-2000
			BR 0010035 A	15-01-2002
			CN 1349550 A	15-05-2002
			CZ 20013762 A3	13-03-2002
			DE 19918912 A1	02-11-2000
			WO 0064974 A2	02-11-2000
			EP 1183309 A2	06-03-2002
			MX PA01010387 A	27-03-2002
DE 10003270	A1	02-08-2001	AU 2515701 A	07-08-2001
			WO 0155255 A1	02-08-2001
EP 0614945	A	14-09-1994	JP 7011083 A	13-01-1995
US 4342846	A	03-08-1982	AU 7954682 A	16-09-1982
			CA 1183630 A1	05-03-1985
			DD 204936 A5	14-12-1983
			EP 0060601 A1	22-09-1982
			JP 57159845 A	02-10-1982