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(54) Titre : PROCEDE DE FABRICATION DE COMPOSITIONS POLYAMIDES THERMOPLASTIQUES PRESENTANT  
UN ECOULEMENT DE FUSION AMELIORE ET ARTICLES ELABORES A PARTIR DE CELLES-CI  
(54) Title: A PROCESS FOR THE PREPARATION OF THERMOPLASTIC POLYAMIDE COMPOSITIONS EXHIBITING  
INCREASED MELT FLOW AND ARTICLES FORMED THEREFROM

(57) **Abrégé/Abstract:**

A process for preparing thermoplastic polyamide compositions that exhibit decreased melt flow and similar physical properties in comparison with conventional systems, in which polyamide is melt-blended with at least one organic acid, and, optionally, one or more additional components, such that the organic acid has a melting point that is no lower than about 10° C less than the onset temperature of the melting point endotherm of the polyamide. The process results in decreased levels of corrosion and erosion of standard processing equipment in comparison with other processes for decreasing the melt flow while maintaining the physical properties of thermoplastic polyamide compositions. Articles of manufacture made according to the instant process and composition described therein are also disclosed.



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(54) Title: A PROCESS FOR THE PREPARATION OF THERMOPLASTIC POLYAMIDE COMPOSITIONS EXHIBITING INCREASED MELT FLOW AND ARTICLES FORMED THEREFROM

(57) Abstract: A process for preparing thermoplastic polyamide compositions that exhibit decreased melt flow and similar physical properties in comparison with conventional systems, in which polyamide is melt-blended with at least one organic acid, and, optionally, one or more additional components, such that the organic acid has a melting point that is no lower than about 10° C less than the onset temperature of the melting point endotherm of the polyamide. The process results in decreased levels of corrosion and erosion of standard processing equipment in comparison with other processes for decreasing the melt flow while maintaining the physical properties of thermoplastic polyamide compositions. Articles of manufacture made according to the instant process and composition described therein are also disclosed.



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efficiently when the polymer matrix has a high viscosity. When two or more polymers, such as a polymer matrix and a rubber impact modifier, are blended, optimal dispersion is often obtained when the two or more polymers have similar melt viscosities.

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When the matrix polymer is a condensation polymer such as a polyamide, it is often possible to obtain a composition that has both well-dispersed additives and a low melt viscosity by using high molecular weight matrix polymer in conjunction with a molecular-weight reducing additive in the melt blending process. In this method, the matrix polymer will have a sufficiently high melt viscosity to ensure an adequate dispersion of additives and the action of the molecular-weight reducing agent will result in a lower molecular weight matrix polymer. U.S. patent application publication 2003/0018135 discloses the use of aliphatic organic acids in the preparation of impact modified polyamide compositions that have both good melt flow and toughness. However, it has been discovered that the use of aliphatic acids disclosed in this publication can lead to rapid corrosion of the steel elements of the processing equipment used in the melt blending processes.

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Thus, a method of preparing a polyamide composition with improved melt flow that also leads to a reduction in rates of corrosion of processing equipment without having to replace the components of standard melt-processing equipment with those made from costly materials with improved corrosion resistance would be desirable. Such a process would desirably result in decreased levels of corrosion of standard processing equipment in comparison with other processes for increasing melt flow while maintaining the physical properties of thermoplastic polyamide compositions.

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### **Summary of the Invention**

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There is disclosed and claimed herein a process for the preparation of a resin composition exhibiting high melt flow, comprising melt-blending a thermoplastic polymer comprising at least one polyamide with 0.01 to 10 weight percent, based on the total weight of the polyamide, of at least one organic acid, and, optionally, one or more additional components, such that



the organic acid has a melting point that is no lower than about 10 °C less than the onset temperature of the melting point endotherm of the polyamide.

In addition, articles of manufacture made from compositions according to the claimed process are disclosed and claimed herein.

### **Detailed Description of the Invention**

A process is provided for the preparation of polyamide compositions that have improved melt flow. The process comprises melt blending polyamide with an organic carboxylic acid that has a melting point that is no lower than about 10 °C less than the onset temperature of the melting point endotherm of the polyamide. The process of the present invention leads to reduced rates of corrosion of processing equipment.

The polyamide used in the process of the present invention is at least one thermoplastic polyamide. Suitable polyamides can be condensation products of dicarboxylic acids and diamines, and/or aminocarboxylic acids, and/or ring-opening polymerization products of cyclic lactams. Suitable dicarboxylic acids include adipic acid, azelaic acid, sebacic acid, dodecanedioic acid, isophthalic acid, and terephthalic acid. Suitable diamines include tetramethylenediamine, hexamethylenediamine, octamethylenediamine, nonamethylenediamine, dodecamethylenediamine, decamethylenediamine, 2-methylpentamethylenediamine, 2-methyloctamethylenediamine, trimethylhexamethylenediamine, bis(*p*-aminocyclohexyl)methane, *m*-xylylenediamine, and *p*-xylylenediamine. A suitable aminocarboxylic acid is 11-aminododecanoic acid. Suitable cyclic lactams are caprolactam and laurolactam. Preferred polyamides include aliphatic polyamide such as polyamide 6; polyamide 6,6; polyamide 4,6; polyamide 6,10; polyamide 6,12; polyamide 11; polyamide 12; and semi-aromatic polyamides such as poly(*m*-xylylene adipamide) (polyamide MXD,6), poly(dodecamethylene terephthalamide) (polyamide 12,T), poly(decamethylene terephthalamide) (polyamide 10,T), poly(nonamethylene terephthalamide) (polyamide 9,T), hexamethylenedipamide-hexamethyleneterephthalamide copolyamide (polyamide 6,T/6,6),

hexamethyleneterephthalamide-2-methylpentamethyleneterephthalamide copolyamide (polyamide 6,T/D,T); and copolymers and mixtures of these polymers.

5           The organic carboxylic acid used in the process of the present invention may be one or more monocarboxylic acids, dicarboxylic acids, tricarboxylic acids, higher acids, or aminoacids. The organic acid is chosen such that its melting point is no lower than about 10 °C less than the onset temperature of the melting point endotherm of the polyamide. The melting  
10   point of the organic acid will preferably be no lower than about 5 °C less than the onset temperature of the melting point endotherm of the polyamide. The melting point of the organic acid will more preferably be no lower than the onset temperature of the melting point endotherm of the polyamide. As used  
15   herein in reference to the organic carboxylic acid, the term "melting point" refers to sublimation point or decomposition point if the organic acid does not have a melting point.

By "onset temperature of the melting point endotherm" of the polyamide is meant the extrapolated onset temperature of the melting curve of  
20   the polyamide ( $T_f$ ) as measured by differential scanning calorimetry (DSC) following ASTM method D3418 -- 82 (Reapproved 1988). If the polyamide has two or more melting point endotherms, the onset temperature of the lowest melting point endotherm is selected. If two or more polyamides are used, the onset temperature of the melting point endotherm of the polyamide  
25   with the lowest melting point endotherm onset temperature is chosen.

The organic acid is preferably selected such that its melting point is no higher than about 10 °C greater than the maximum temperature of the melt during melt-blending with the polyamide. For polyamides with melting point  
30   endotherm onset temperatures that are between about 200 °C and about 310 °C, isophthalic acid and terephthalic acid are preferred organic acids. For polyamides with melting point endotherm onset temperatures that are between about 100 °C and about 160 °C, adipic acid and dodecanedioic acids are preferred organic acids.



The organic acid is used at about 0.01 to about 10 weight percent, preferably at about 0.05 to about 2 weight percent, or more preferably at about 0.1 to about 1 weight percent, where the weight percentages are based  
5 on the total weight of polyamide

The polyamide compositions produced by the process of the present invention may contain additives. Examples of additives include impact modifiers, fibrous reinforcing agents (such as glass fibers, carbon fibers,  
10 wollastonite, aramids etc.), plasticizers, thermal stabilizers, oxidative stabilizers, UV light stabilizers, flame retardants, chemical stabilizers, lubricants, colorants (such as carbon black, other pigments, dyes), mold-release agents, nucleating agents, nanoclays, etc.

15 The process of the present invention is particularly useful for preparing polyamide compositions that contain impact modifiers and/or reinforcing agents.

Preferred impact modifiers for use with polyamides include carboxyl-substituted polyolefins, which are polyolefins that have carboxylic moieties  
20 attached thereto, either on the polyolefin backbone itself or on side chains. By "carboxylic moieties" is meant carboxylic groups such as one or more of dicarboxylic acids, diesters, dicarboxylic monoesters, acid anhydrides, and monocarboxylic acids and esters. Useful impact modifiers include dicarboxyl-  
25 substituted polyolefins, which are polyolefins that have dicarboxylic moieties attached thereto, either on the polyolefin backbone itself or on side chains. By 'dicarboxylic moiety' is meant dicarboxylic groups such as one or more of dicarboxylic acids, diesters, dicarboxylic monoesters, and acid anhydrides.

30 The impact modifier used with polyamides will preferably be based on an ethylene/ $\alpha$ -olefin polyolefin. Diene monomers such as 1,4-hexadiene or dicyclopentadiene may optionally be used in the preparation of the polyolefin. Preferred polyolefins are ethylene-propylene-diene (EPDM) polymers made

from 1,4-hexadiene and/or dicyclopentadiene. The carboxyl moiety may be introduced during the preparation of the polyolefin by copolymerizing with an unsaturated carboxyl-containing monomer. Preferred is a copolymer of ethylene and maleic anhydride monoethyl ester. The carboxyl moiety may also be introduced by grafting the polyolefin with an unsaturated compound containing a carboxyl moiety, such as an acid, ester, diacid, diester, acid ester, or anhydride. A preferred grafting agent is maleic anhydride. A preferred impact modifier is an EPDM polymer grafted with maleic anhydride, such as Fusabond® N MF521D, which is commercially available from E. I. DuPont de Nemours & Co., Inc., Wilmington, DE. Blends of polyolefins, such as polyethylene, polypropylene, and EPDM polymers with polyolefins that have been grafted with an unsaturated compound containing a carboxyl moiety may be used as an impact modifier.

Impact modifiers suitable for use with polyamides may also include ionomers. By an ionomer is meant a carboxyl group containing polymer that has been neutralized or partially neutralized with metal cations such as zinc, sodium, or lithium and the like. Examples of ionomers are described in US patents 3,264,272 and 4,187,358, both incorporated by reference herein. Examples of suitable carboxyl group containing polymers include, but are not limited to, ethylene/acrylic acid copolymers and ethylene/methacrylic acid copolymers. The carboxyl group containing polymers may also be derived from one or more additional monomers, such as, but not limited to, butyl acrylate. Zinc salts are preferred neutralizing agents. Ionomers are commercially available under the Surlyn® trademark from E.I. du Pont de Nemours and Co., Wilmington, DE.

It is preferred that a thermoplastic acrylic impact modifier have a core-shell structure. The core-shell structure is one in which the core portion preferably has a glass transition temperature of 0 °C or less, while the shell portion is preferably has a glass transition temperature higher than that of the core portion. The core portion may be grafted with silicone. The shell section may be grafted with a low surface energy substrate such as silicone, fluorine,



and the like. An acrylic polymer with a core-shell structure that has low surface energy substrates grafted to the surface will aggregate with itself during or after blending with the polyester and other components of the composition of the invention and can be easily uniformly dispersed in the composition.

In the process of the present invention, the polyamide and organic acid and optional additional ingredients are melt-blended. All of the components may be dry-blended prior to melt-blending; previously melt-blended mixtures of polyamide with additional additives may be melt-blended with the organic acid; previously melt-blended mixtures of polyamide with additional additives may be melt-blended with the organic acid and additional additives.

Melt-blending may be carried out using any appropriate method known to those skilled in the art. Suitable methods may include using a single or twin-screw extruder, blender, kneader, Banbury mixer, molding machine, etc. Twin-screw extrusion is preferred, particularly when the process is used to prepare compositions that contain additives such as impact modifiers, and reinforcing agents.

The compositions made from the process of the present invention have a high melt flow and may be conveniently formed into a variety of articles using injection molding, rotomolding and other melt-processing techniques.

### **Examples**

#### **Example 1 and Comparative Example 1**

The ingredients shown in Table 1 were dry-blended by tumbling in a drum. Zytel® 101 NC010 is a polyamide 6,6 supplied by E.I. du Pont de Nemours & Co., Wilmington, DE. Fusabond® N MF521D is an EPDM elastomer grafted with maleic anhydride also supplied by DuPont. The black color concentrate is carbon black dispersed in a polymeric carrier. Equal molar amounts of dodecanedioic acid and terephthalic acid were used.

The mixtures were melt-blended in a ten barrel 30 mm Werner & Pfleiderer co-rotating twin-screw extruder with a barrel temperature of about 270 °C and a die temperature of about 280 °C. All of the ingredients were fed into the first barrel section. Extrusion was carried out with a port under vacuum. The screw speed was 250 rpm and the extruder feed rate was 30 pounds per hour. The resulting strand was quenched in water, cut into pellets, and sparged with nitrogen until cool. The extruder was run for about 6.5 to about 7 hours a day for 10 days.

Between barrels 3 and 4; 6 and 7; and 9 and 10, the extruder contained 0.25 inch wide steel spacers having a profile matching that of the extruder barrels. (The barrels are numbered sequentially such that barrel 10 is closest to the die.) The diameters of the openings at different points inside the spacers was also measured using a laser micrometer before and after extrusion. Measurements were taken for both the left and right side (relative to the polymer flow direction in the extruder) openings in each barrel. The results of these measurements were converted to a rate of metal loss from the inner surface of the extruder barrels in mils/year (mpy). Measurements at 10 points were made for each spacer for Comparative Example 1 and measurements at eight points were made for each spacer for Example 1 and the range of results is reported in Table 2. Due to experimental error, low rates of metal loss were sometimes calculated to be negative. Negative rates are reported as zero in Table 2. The surfaces of the openings in the spacers were also visually inspected at a 40-50 fold magnification. Visual inspection showed that the spacers used in Example 1 showed significantly less corrosion than those used in Comparative Example 1.

Notched Izod impact strengths and melt viscosities at 280 °C and 1,000 s<sup>-1</sup> were measured for the compositions of Comparative Example 1 and Example 1. ISO method ISO 180/1A was used for the notched Izod testing. The results are given in Table 1.



Table 1

	Comparative Ex. 1	Example 1
Zytel® 101 NC010	77.57	77.75
Fusabond® N MF521D	17.05	17.05
Black color concentrate	4.45	4.45
Dodecanedioic acid	0.65	--
Terephthalic acid	--	0.47
Stabilizers and lubricants	0.28	0.28
Notched Izod impact strength (kJ/m <sup>2</sup> )	74	72
Melt viscosity (Pa•s)	102	102

Ingredient quantities are in weight percent relative to the total weight of the composition.

5

Table 2

	Corrosion rate (mpy)			
	Comparative Example 1		Example 1	
Spacer location	Left side	Right side	Left side	Right side
Between barrels 3 and 4	379-487	183-291	0-21	0-15
Between barrels 6 and 7	9-117	0-74	0	0
Between barrels 9 and 10	25-133	0-100	0-29	0-3

The polyamide used in Example 1 and Comparative Example 1, polyamide 6,6, has a melting point of about 264 °C. The organic acid used in Comparative Example 1, dodecanedioic acid, has a melting point of about 128-130 °C, more than 135 °C less than that of polyamide 6,6. The use of dodecanedioic acid led to significant corrosion in the extrusion equipment used. The organic acid used in Example 1, terephthalic acid, has a sublimation point of greater than 300 °C, which is more than 30 °C greater than the melting point of polyamide 6,6. The use of terephthalic acid led to minimal corrosion in the extrusion equipment used, while leading to a composition that had an impact strength and melt viscosity that were very similar to those of the composition made in Comparative Example 1.

20

### Example 2 and Comparative Example 2

All of the ingredients shown in Table 3 with the exception of the brominated flame retardant and glass fibers were dry-blended and fed to the first barrel of a 40 mm Werner & Pfleiderer co-rotating twin-screw extruder with a barrel temperature of about 340-350 °C. The brominated flame retardant and glass fibers were added via a side feeder downstream in the extruder. The resulting strand was quenched in water and cut into pellets.

In Table 3, "polyamide 6,T/6,6" refers to a copolyamide with a melting point of about 312 °C made from terephthalic acid, adipic acid, and hexamethylenediamine. "Lubricants" refers to fatty acid based organic lubricants. "Surlyn® 8920" refers to a neutralized ethylene-methacrylic acid copolymer supplied by E.I. du Pont de Nemours & Co. "Glass fibers" refers to PPG 3540, supplied by PPG Industries, Inc. "Brominated flame retardant" refers to PDBS-80, supplied by Great Lakes Chemical, Inc. The black color concentrate is a 30 weight percent loading of carbon black in polyamide 6T/66 supplied by Clariant Corp., Tarrytown, NY.

The melt viscosities of the resulting compositions were determined at 325 °C and 1000 s<sup>-1</sup> using a Kayeness rheometer and the results are shown in Table 3. Tensile strength and elongation at break were determined using ISO method 527-1/-2, flexural modulus was determined using ISO 178, and notched Charpy impact strength was determined using ISO method 179 1eA. The results are shown in Table 3.

25



Table 3

	Example 3	Comp. Ex. 3
Polyamide 6,T/6,6	39.68	39.88
Zinc borate	4	4
Lubricants	0.45	0.45
Surlyn® 8920	1	1
Terephthalic acid	0.2	--
Black color concentrate	0.67	0.67
Brominated flame retardant	24	24
Glass fibers	30	30
Melt viscosity (Pa•s)	65.8	78.2
Tensile strength (MPa)	173	173
Elongation at break (%)	2.0	2.0
Flexural modulus (MPa)	10637	10869
Notched Charpy impact strength (kJ/m <sup>2</sup> )	10.1	11.2

Ingredient quantities are in weight percent relative to the total weight of the  
5 composition.

#### Examples 3-5 and Comparative Example 3

All of the ingredients shown in Table 4 with the exception of the glass  
fibers were dry-blended and fed to the first barrel of a 58 mm Werner &  
10 Pfeiderer co-rotating twin-screw extruder with barrel temperatures of up about  
280 °C. The glass fibers were added via a side feeder downstream in the  
extruder. The resulting strand was quenched in water and cut into pellets.

In Table 4, "polyamide 6,6" refers to Zytel® 101 NC010 supplied by E.I.  
15 du Pont de Nemours & Co. "Glass fibers" refers to PPG 3540, supplied by  
PPG Industries, Inc. "Black color concentrate" refers to a black colorant in  
polyamide 6,6.

The melt viscosities of the resulting compositions were determined at  
20 280 °C and 1000 s<sup>-1</sup> using a Kayeness rheometer and the results are shown  
in Table 4. Tensile strength and elongation at break were determined using  
ISO method 527-1/-2, flexural modulus was determined using ISO 178, and

notched Charpy impact strength was determined using ISO method 179 1eA. The results are shown in Table 5.

Table 4

	Ex. 3	Ex. 4	Ex. 5	Comp. Ex. 2
Polyamide 6,6	65.38	65.18	64.98	65.68
Glass fibers	33	33	33	33
Heat stabilizer and lubricants	0.32	0.32	0.32	0.32
Black color concentrate	1	1	1	1
Terephthalic acid	0.3	0.5	0.7	--
Melt viscosity (Pa•s)	130	101	79	216
Tensile strength (MPa)	208	206	202	203
Elongation at break (%)	3.0	2.8	2.6	3.3
Flexural modulus (MPa)	9524	9693	9702	9048
Notched Charpy impact strength (kJ/m <sup>2</sup> )	12.9	13.0	13.0	12.3

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Ingredient quantities are in weight percent relative to the total weight of the composition.

#### Example 6

10 The ingredients shown in Table 5 were dry-blended by tumbling in a drum. Elvamide® 8061 is a copolyamide 6,6/6 supplied by DuPont. Its melting point is 156 °C and the onset temperature of its melting point endotherm is 129 °C

15 The mixtures were melt-blended in a ten barrel 30 mm Werner & Pfleiderer co-rotating twin-screw extruder with a barrel temperature of about 240 °C and a die temperature of about 250 °C. All of the ingredients were fed into the first barrel section. Extrusion was carried out with a port under vacuum. The screw speed was 250 rpm and the extruder feed rate was 30  
20 pounds per hour. The resulting strand was quenched in water, cut into pellets, and sparged with nitrogen until cool. The extruder was run for about 6.5 to about 7 hours a day for 10 days.



Between barrels 3 and 4; 6 and 7; and 9 and 10, the extruder contained 0.25 inch wide steel spacers having a profile matching that of the extruder barrels. (The barrels are numbered sequentially such that barrel 10 is closest to the die.) An additional 0.25 inch wide spacer of Inconel® 625 was placed between the steel spacer and barrel 4. The diameters of the openings at different points inside the spacers was also measured using a laser micrometer before and after extrusion. Measurements were taken for both the left and right side (relative to the polymer flow direction in the extruder) openings in each barrel. The results of these measurements were converted to a rate of metal loss from the inner surface of the extruder barrels in mils/year (mpy). Measurements at 10 points were made for each spacer for Example 6 and the range of results is reported in Table 6. Due to experimental error, low rates of metal loss were sometimes calculated to be negative. Negative rates are reported as zero in Table 6. The surfaces of the openings in the spacers were also visually inspected at a 40-50 fold magnification. Visual inspection showed that the spacers used in Example 6 showed minimal corrosion and significantly less corrosion than those used in Comparative Example 1.

Notched Izod impact strengths and melt viscosities at 280 °C and 1,000 s<sup>-1</sup> were measured for the compositions of Comparative Example 1 and Example 1. ISO method ISO 180/1A was used for the notched Izod testing. The results are given in Table 5.

Table 5

	Example 6
Elvamide®	77.57%
Fusabond® N MF521D	17.05%
Black color concentrate	4.45%
Dodecanedioic acid	0.65%
Stabilizers and lubricants	0.28%
Notched Izod impact strength (kJ/m <sup>2</sup> )	78.9
Melt viscosity (Pa•s)	84

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Ingredient quantities are in weight percent relative to the total weight of the composition.

Table 6

	Corrosion rate (mpy)	
	Example 6	
Spacer location	Left Side	Right side
Between barrels 3 and 4 (steel spacer)	0-109	0-174
Between barrels 3 and 4 (Inconel® spacer)	0-107	0-125
Between barrels 6 and 7	0-117	0-146
Between barrels 9 and 10	0-86	0-114



**What is Claimed is:**

1. A process for the preparation of a resin composition exhibiting high melt  
5 flow, comprising melt-blending a thermoplastic polymer comprising at least one polyamide with 0.01 to 10 weight percent, based on the total weight of the polyamide, of at least one organic acid, and, optionally, one or more additional components, such that the organic acid has a melting point that is no lower than about 10 °C less than the onset temperature of the  
10 melting point endotherm of the polyamide.
2. The process of claim 1 wherein the organic acid has a melting point that is no lower than about 5 °C less than the onset temperature of the melting point endotherm of the polyamide.
- 15 3. The process of claim 1 wherein the organic acid has a melting point that is no lower than the onset temperature of the melting point endotherm of the polyamide.
- 20 4. The process of claim 1 wherein about 0.05 to about 2 weight percent, based on the total weight of polyamide, of at least one organic acid is used.
5. The process of claim 1 wherein about 0.1 to about 1 weight percent, based  
25 on the total weight of polyamide, of at least one organic acid, is used.
6. The process of claim 1 wherein the polyamide has a melting point endotherm with an onset temperature of between about 200 °C and about 310 °C.
- 30 7. The process of claim 6 wherein the organic acid is terephthalic acid and/or isophthalic acid.

8. The process of claim 1 wherein the polyamide has a melting point endotherm with an onset temperature of between about 100 °C and about 160 °C.
- 5 9. The process of claim 8 wherein the organic acid is adipic acid and/dodecanedioic acid.
- 10 10. The process of claim 6 wherein the polyamide is selected from one or more of polyamide 6; polyamide 6,6; polyamide 6,10; polyamide 6,12; polyamide 11; polyamide 12; polyamide 4,6; polyamide 9,T; polyamide 10,T; polyamide 12,T; polyamide 6,T/6,6; and polyamide 6,T/D,T.
- 15 11. The process of claim 7 wherein polyamide is selected from one or more of polyamide 6; polyamide 6,6; polyamide 6,10; polyamide 6,12; polyamide 11; polyamide 12; polyamide 4,6; polyamide 9,T; polyamide 10,T; polyamide 12,T; polyamide 6,T/6,6; and polyamide 6,T/D,T.
- 20 12. The process of claim 1 wherein the thermoplastic polymer is at least one polyamide and the additional components comprise an impact modifier.
13. The process of claim 4 wherein the impact modifier comprises an EPDM polyolefin grafted with maleic anhydride.
- 25 14. The process of claim 1 wherein the thermoplastic polymer is at least one polyamide the additional ingredients comprise at least one reinforcing agent.
15. The process of claim 14 wherein the reinforcing agent is glass fibers.
- 30 16. An article of manufacture made from compositions according to the process of Claim 1.