



(51) International Patent Classification:

C08G 69/26 (2006.01) *C08J 3/20* (2006.01)
C08G 69/28 (2006.01) *C08K 7/14* (2006.01)

(21) International Application Number:

PCT/EP2017/076579

(22) International Filing Date:

18 October 2017 (18.10.2017)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

16194580.3 19 October 2016 (19.10.2016) EP

(71) Applicant: **DSM IP ASSETS B.V.** [NL/NL]; Het Over-
loon 1, 6411 TE HEERLEN (NL).

(72) Inventors: **WOLFFS, Martin**; P.O. Box 4, 6100 AA Echt
(NL). **RULKENS, Rudy**; P.O. Box 4, 6100 AA ECHT
(NL). **KIERKELS, Renier Henricus Maria**; P.O. Box 4,
6100 AA ECHT (NL).

(74) Agent: **VAN DER PLOEG, Antonius Franciscus M.J.**;
P.O. Box 4, 6100 AA ECHT (NL).

(81) Designated States (*unless otherwise indicated, for every
kind of national protection available*): AE, AG, AL, AM,
AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ,
CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO,
DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN,
HR, HU, ID, IL, IN, IR, IS, JO, JP, KE, KG, KH, KN, KP,
KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME,
MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ,
OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA,
SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN,
TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (*unless otherwise indicated, for every
kind of regional protection available*): ARIPO (BW, GH,
GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ,
UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ,
TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK,
EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV,
MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM,
TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW,
KM, ML, MR, NE, SN, TD, TG).

Published:

- with international search report (Art. 21(3))
- before the expiration of the time limit for amending the
claims and to be republished in the event of receipt of
amendments (Rule 48.2(h))

(54) Title: SEMI-CRYSTALLINE SEMI-AROMATIC POLYAMIDE AND COMPOSITIONS COMPRISING THE SAME

(57) Abstract: The present invention relates to a semi-crystalline semi-aromatic polyamide consisting of 95 – 100 mole% of monomeric units derived from diamine comprising aliphatic diamine comprising 5 – 40 mole% of a cycloaliphatic diamine, and dicarboxylic acid comprising at least 50 mole % of naphthalene dicarboxylic acid (N), or biphenyl dicarboxylic acid (B), or a combination thereof.



SEMI-CRYSTALLINE SEMI-AROMATIC POLYAMIDE AND COMPOSITIONS
COMPRISING THE SAME

5 The present invention relates to a semi-crystalline semi-aromatic polyamide, and to compositions comprising the same.

 Semi-crystalline semi-aromatic polyamides are widely known and applied in various compositions for use in electrical, electronic and automotive applications. In many of these applications semi-crystalline semi-aromatic polyamides are preferred over their aliphatic counterparts for their better performance under more
10 demanding conditions involving high temperature processing, such as in surface mount technologies (SMT), high continuous use temperatures, such as in automotive under the hood applications, and better performance under humid conditions. In particular, retention of mechanical properties and good dimensional stability are important properties. Even better performance under humid conditions are obtained with special
15 amorphous semi-aromatic polyamides. However, these polyamides lack the required high temperature mechanical properties. Moreover, with these amorphous semi-aromatic polyamides it appears more difficult to obtain a good balance between flow properties and mechanical performance, such as high temperature stiffness and creep, impact and fatigue properties. For achieving good mechanical properties generally a
20 high load of reinforcing agents is needed, which in turn has a draw back on the flow properties. Furthermore, there is a systematic trend towards miniaturization of parts, requiring improved flow while increasing demands are put on mechanical performance at high temperature and under humid conditions.

 Therefore, there is need for polyamides, and for compositions
25 comprising the same, which have a good flow, good mechanical properties and good dimensional stability and exhibit an improved balance in performance under different conditions.

 The aim of the present invention is to provide such a polyamide having an improved balance in performance, and for compositions comprising the
30 same.

 This aim has been achieved with the semi-crystalline semi-aromatic polyamide according to the invention, and to compositions comprising the same.

 According to the first embodiment of the invention, the polyamide is a semi-crystalline semi-aromatic polyamide consisting of 95 – 100 mole%, preferably 98
35 – 100 mole%, of monomeric units derived from diamine and dicarboxylic acid, and 0 –

5 mole%, preferably 0 – 2 mole%, of one or more other monomeric units, the mole% being relative to the total mole % of monomeric units, wherein

- 5 a) diamine comprises at least 90 mole% of aliphatic diamine comprising 5 – 40 mole%, relative to the total molar amount of diamine, of a cycloaliphatic diamine chosen from the group consisting of 1,4-diaminocyclohexane, 4,4'-methylene-bis(cyclohexylamine) (PAC), 4,4'-methylene-bis(2-methylcyclohexylamine) (MAC); 3,3',5,5'-tetramethyl-4,4'-diaminocyclohexylmethane; 2,2',3,3'-tetramethyl-4,4'-diaminocyclohexylmethane; norbornanediamine; and isophoronediamine (IPD), and any mixtures thereof; and
- 10 b) the dicarboxylic acid comprises at least 95 mole% of an aromatic dicarboxylic acid comprising 50 – 100 mole%, relative to the total molar amount of dicarboxylic acid, of naphthalene dicarboxylic acid (N), or biphenyl dicarboxylic acid (B), or a combination thereof.

15 According to the second embodiment of the invention, the composition is a thermoplastic polymer composition, comprising

(A) a semi-crystalline semi-aromatic polyamide as mentioned here above; and

(B) a reinforcing agent; and optionally

(C) one or more further ingredient.

20 The effect of the semi-crystalline semi-aromatic polyamide according to the invention is that the composition comprising the polyamide has a good processability and good mechanical properties, as well as a good dimensional stability and improved retention of mechanical properties under humid conditions at elevated temperature.

25 The polyamide according to the invention comprises 5 – 40 mole%, relative to the total molar amount of diamine, of a cycloaliphatic diamine as specified above. With the amount in the range of 5 – 40 mole% the balance of properties of the polyamide and compositions thereof is better than for those with the amount below 5 mole% or above 40 mole%.

30 Herein, the amount of the cycloaliphatic diamine is preferably in the range of 10 – 35 mole%, more preferably 15 - 32 mole%.

The cycloaliphatic diamine is chosen from the group consisting of 1,4-diaminocyclohexane, 4,4'-methylene-bis(cyclohexylamine) (abbreviated herein as PAC), 4,4'-methylene-bis(2-methylcyclohexylamine) (which is equivalent to 3,3'-dimethyl-4,4'-diaminocyclohexylmethane; abbreviated herein as MAC); 3,3',5,5'-

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tetramethyl-4,4'-diaminocyclohexylmethane; 2,2',3,3'-tetramethyl-4,4'-diaminocyclohexylmethane; norbornanedi-amine; and isophoronediamine (which is equivalent to 3-aminomethyl-3,5,5-trimethylcyclohexylamine; abbreviated herein as IPD), and any mixtures thereof.

5 Preferably, the cycloaliphatic diamine comprises PAC, MAC or IPD, or any mixture thereof. More preferably, the cycloaliphatic diamine comprises, or even better fully consists of PAC or MAC, or any mixture thereof

 The diamine comprises at least 90 mole% of aliphatic diamine. Preferably, the diamine comprises, next to the said cycloaliphatic diamine, a linear
10 aliphatic α - ω diamine in an amount in the range of 60 – 95 mole%, relative to the total molar amount of diamine. More preferably, the linear aliphatic α - ω diamine comprises a C2 – C20 diamine, or any mixtures thereof, still more preferably a C4 – C18 diamine, or any mixtures thereof.

 With a linear aliphatic α - ω diamine is herein understood a diamine
15 comprising two amine groups (-NH₂), wherein both NH₂ groups sit at the ends of a linear aliphatic chain, i.e. wherein one -NH₂ group sits at the α -position and the other at the ω -position. Examples of such diamines are: 1,2-diaminoethane (synonym: 1,2-ethylene diamine; 2 carbons); 1,3-diaminopropane (synonym: propane-1,3-diamine or 1,3-propylene diamine; 3 carbons); butane-1,4-diamine (synonym for 1,4-butane
20 diamine; 4 carbons); pentane-1,5-diamine (synonym for 1,5-pentane diamine; 5 carbons); hexamethylenediamine (synonym: hexane-1,6-diamine or 1,6-hexane diamine; 6 carbons), heptamethylene diamine (synonym: heptane-1,7-diamine or 1,7-heptane diamine) and octamethylene diamine (synonym: octane-1,8-diamine or 1,8-octane diamine), 1,9-nonane diamine, 1,10-decane diamine, 1,11-undecane diamine,
25 and 1,12-dodecane diamine and 1,18-octadecane diamine.

 Furthermore, the diamine may optionally comprise one or more other diamine. The other diamine may be, for example, a branched aliphatic diamine, a cyclic aliphatic diamine not being a cycloaliphatic diamine as mentioned above, or an aralkyl
30 diamine, or any combination thereof. An example of a branched diamine is 2-methyl, 1,5-pentane diamine. Examples of a such a cyclic diamine are 1,3-cyclohexanedi-amine and 1,4-cyclohexanedi-amine. Example of aralkyl diamines are metaxylylene diamine (MXD) and paraxylylene diamine (PXD).

 The polyamide according to the invention may comprise, if at all, one or more other diamine, not being a cycloaliphatic diamine chosen from the group

mentioned above or a linear aliphatic α - ω diamine, preferably in an amount of 0 – 10 mole%, more preferably 0 – 5 mole%, relative to the total molar amount of diamine.

The dicarboxylic acid in the semi-crystalline semi-aromatic polyamide according to the first embodiment comprises at least 95 mole% of an aromatic dicarboxylic acid comprising naphthalene dicarboxylic acid (abbreviated herein also as N), or biphenyl dicarboxylic acid (abbreviated herein also as B), or a combination thereof, in an amount in the range of 50 – 100 mole%, relative to the total molar amount of dicarboxylic acid. Preferably, the amount is in the range of 60 – 100 mole%, more preferably, of 75 – 100 mole%, relative to the total molar amount of dicarboxylic acid.

Suitably, the naphthalene dicarboxylic acid (N) comprises one of 2,6-naphthalene dicarboxylic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalene dicarboxylic acid, 2,7-naphthalene dicarboxylic acid, or a combination thereof, and preferably comprises, or even consists of 2,6-naphthalene dicarboxylic acid.

Suitably, the biphenyl dicarboxylic acid (B) comprises one of 3,3'-biphenyl dicarboxylic acid, 3,4'-biphenyl dicarboxylic acid and 4,4'-biphenyl dicarboxylic acid, and preferably comprises, or even consists of 4,4'-biphenyl dicarboxylic acid.

The dicarboxylic acid suitably comprises another aromatic dicarboxylic acid, if any, in an amount in the range of 0 – 50 mole%, preferably 0 – 40 mole%, and more preferably 0 – 25 mole%, relative to the total molar amount of dicarboxylic acid, of another dicarboxylic acid, different from naphthalene dicarboxylic acid (N) and biphenyl dicarboxylic acid (B). Suitably, the other aromatic dicarboxylic acid comprises terephthalic acid (T) or isophthalic acid (I), or a mixture thereof, and preferably comprises or even better consists of terephthalic acid.

The polyamide may optionally comprise an aliphatic dicarboxylic acid. The amount thereof is limited to a range of 0 – 5 mole%, preferably 0 – 2 mole%, relative to the total molar amount of dicarboxylic acid.

The polyamide comprises monomeric units derived from diamines and dicarboxylic acid. The polyamide may further comprise other monomeric units, for example monomeric units derived from one or more monomers comprising one functional group, or three- functional groups, or more than three functional groups, or a mixture thereof. Herein the functional group suitably is an amine group or a carboxylic acid. Mono-functional monomers can be used as chain stoppers. Tri-functional monomers and higher can be used as branching agent. Other monomeric units may

also be derived from difunctional monomers such as aminoacids, or lactams, or a mixture thereof.

In a preferred embodiment of the invention, the polyamide is a semi-crystalline semi-aromatic polyamide, wherein

- 5 a. the diamine consists of
- i. 5 – 40 mole% of a cycloaliphatic diamine chosen from the group consisting of 1,4-diaminocyclohexane, 4,4'-methylene-bis(cyclohexylamine) (PAC), 3,3'-dimethyl-4,4'-diaminocyclohexylmethane (MAC); 3,3',5,5'-tetramethyl-4,4'-diaminocyclohexylmethane; 2,2',3,3'-tetramethyl-4,4'-diaminocyclohexylmethane; norbornanediamine; and isophoronediamine (IPD), and any mixtures thereof; wherein the cycloaliphatic diamine comprises 10 – 40 mole% of PAC, MAC and IPD, or a combination thereof, and
- 10 ii. 60 – 90 mole% of a linear aliphatic α - ω diamine, and optionally
- 15 iii. 0 – 10 mole% of another diamine;
- wherein the mole % is relative to the total molar amount of diamine
- b. the dicarboxylic acid consists of
- i. 50 – 100 mole% of naphthalene dicarboxylic acid and /or biphenyl dicarboxylic acid; and optionally
- 20 ii. 0 – 50 mole% of another aromatic dicarboxylic acid, and/or
- iii. 0 – 5 mole% of an aliphatic dicarboxylic acid,
- wherein the mole % is relative to the total molar amount of dicarboxylic acid.

In a more preferred embodiment, the polyamide is a semi-crystalline semi-aromatic polyamide, wherein

- 25 a. the diamine consists of
- i. 10 – 40 mole% of a cycloaliphatic diamine chosen from PAC, MAC and IPD, or a combination thereof, and
- ii. 60 – 90 mole% of a linear aliphatic α - ω diamine, and optionally
- iii. 0 – 10 mole% of another diamine, not being a linear aliphatic α - ω diamine
- 30 or a cycloaliphatic diamine chosen from PAC, MAC and IPD;
- wherein the mole % is relative to the total molar amount of diamine; and
- b. the dicarboxylic acid consists of

- i. 75 – 100 mole%, relative to the total molar amount of dicarboxylic acid, of naphthalene dicarboxylic acid and /or biphenyl dicarboxylic acid, and optionally
- ii. 0 – 25 mole%, relative to the total molar amount of dicarboxylic acid, of another aromatic dicarboxylic acid, and/or
- iii. 0 – 5 mole%, relative to the total molar amount of dicarboxylic acid, of an aliphatic dicarboxylic acid,

wherein the mole % is relative to the total molar amount of dicarboxylic acid.

Semi-crystalline polymers typically have a morphology comprising crystalline domains, characterized by a melting temperature and a melting enthalpy, and amorphous domains characterized by a glass transition temperature.

With the term semi-crystalline in semi-crystalline polyamide is herein understood that the polyamide has a melting temperature (T_m) and a melting enthalpy (ΔH_m), as well as a glass transition temperature (T_g).

Suitably, the melting temperature (T_m) of the semi-crystalline semi-aromatic polyamides according to the invention, and the various preferred and particular embodiments thereof, suitably is at least 250 °C, or better at least 270 °C. Preferably, T_m is in the range of 280 – 350 °C, more preferably 290 – 330 °C.

With the term melting temperature is herein understood the temperature, measured by the differential scanning calorimetry (DSC) method according to ISO-11357-1/3, 2011, on pre-dried samples, in a N₂ atmosphere with heating and cooling rate of 20°C/min. Herein T_m has been calculated from the peak value of the highest melting peak in the second heating cycle.

Suitably the semi-crystalline polyamides according to the invention, and the various preferred and particular embodiments thereof, have a melting enthalpy of at least 10 J/g, preferably at least 20 J/g, and even more preferably at least 25 J/g.

With the term melting enthalpy (ΔH_m) is herein understood the melting enthalpy, measured by the DSC method according to ISO-11357-1/3, 2011, on pre-dried samples in a N₂ atmosphere with heating and cooling rate of 20°C/min. Herein (ΔH_m) has been calculated from the surface under the melting peak in the second heating cycle.

Suitably the semi-crystalline polyamides according to the invention, and the various preferred and particular embodiments thereof, have a glass transition temperature of at least 135 °C, preferably at least 145 °C, more preferably at least 150 °C.

With the term glass transition temperature (T_g) is herein understood the temperature, measured by the DSC method according to ISO-11357-1/2, 2011, on pre-dried samples in a N_2 atmosphere with heating and cooling rate of $20\text{ }^\circ\text{C/min}$. Herein T_g has been calculated from the value at the peak of the first derivative (with
5 respect of time) of the parent thermal curve corresponding with the inflection point in the second heating cycle of the parent thermal curve.

In a preferred embodiment of the invention, the semi-crystalline semi-aromatic polyamide has a T_m of at least $250\text{ }^\circ\text{C}$, a ΔH_m of at least 20 J/g , and a T_g of at least $135\text{ }^\circ\text{C}$. More preferably, the semi-crystalline semi-aromatic polyamide
10 has a T_m of at least $270\text{ }^\circ\text{C}$, a ΔH_m of at least 25 J/g , and a T_g of at least $145\text{ }^\circ\text{C}$.

The present invention also relates to a thermoplastic polymer composition. Herein the composition comprises:

- (A) a semi-crystalline semi-aromatic polyamide according to invention as described above;
- 15 (B) a reinforcing agent; and optionally
- (C) one or more further ingredient.

Suitably, the composition comprises

- (A) $30 - 70\text{ wt.}\%$ of the semi-crystalline semi-aromatic polyamide;
- (B) $5 - 60\text{ wt.}\%$ of an inorganic filler or a fibrous reinforcing agent, or a
20 combination thereof;
- (C) $0 - 40\text{ wt.}\%$ of one or more further ingredient.

The effect of the composition according to the invention is the combined effect between the semi-crystalline semi-aromatic polyamide and the reinforcing agent. The reinforcing agent contributes to the mechanical properties of the
25 composition, in particular contributing to the tensile strength and tensile modulus, while the semi-crystalline semi-aromatic polyamide according to the invention contributes to the retention of the mechanical properties under humid conditions at elevated temperatures.

Suitably, the semi-crystalline semi-aromatic polyamide therein is one
30 according to the first embodiment of the present invention, or any preferred or particular embodiment thereof.

The reinforcing agent in the composition according to the invention suitable comprises, at least one component selected from reinforcing fibers and inorganic fillers, or a combination thereof. Herein a wide range of fibers and fillers may
35 be used. Examples of such fillers include, but are not limited to, silica, metasilicates,

alumina, talc, diatomaceous earth, clay, kaolin, quartz, glass, mica, titanium dioxide, molybdenum disulphide, gypsum, iron oxide, zinc oxide, powdered polytetrafluoroethylene, montmorillonite, calcium carbonate, glass powder and glass beads.

5 As reinforcing fibers, for example aromatic fibers as well as inorganic fibers may be used. Preferably, inorganic fibers are used. Examples thereof are glass fibers, carbon fibers, boron fibers, ceramic fibers and whiskers of wollastonite and potassium titanate.

10 In a preferred embodiment of the invention, the reinforcing agent in the polymer composition comprises, or even consists of inorganic fibers or inorganic fillers, or a combination thereof.

 Preferably, the reinforcing agent (component (B)) comprises glass fibers or carbon fibers. More preferably glass fibers are used. These glass fibers may be of various composition and shape, for example, S-glass, E-glass and basalt glass,
15 as well as round glass and flat glass fibers.

 The polymer composition may comprise one or more other components (component (C)), different from the semi-crystalline semi-aromatic polyamide (component (A)), and the reinforcing agent (component (B)). As component (C), any auxiliary additive used in polyamide molding compositions may be used.

20 It is noted that the terms "one or more" and "at least one" are herein meant to have the same meaning and can be used interchangeably. When the amount of a component can be 0 wt.%, for example, when the component is present in an amount in a range starting from 0 wt.%, the amount of 0 wt.% it is to be understood that no such component is present at all.

25 Suitable additives include stabilizers, flame retardants, plasticizers, conductive and/or anti-static agents, lubricants and mold release agents, nucleating agents, dyes and pigments, and any other auxiliary additives that may be used in polyamide compositions. Examples of heat stabilizers include copper (I) halides e.g. copper bromide and copper iodide, and alkali halides e.g. lithium, sodium and
30 potassium bromides and iodides.

 The invention is further illustrated with the following Examples and Comparative Experiments.

Polymer preparation

35 Examples 1-5 and Comparative Experiments A-D.

Diacids and diamines were weighed in the appropriate amounts and molar ratio's according to their desired composition, and put into a glass vial. It was ensured that an equimolar balance between diamine and diacid was added to the glass vial. Subsequently the glass vial was placed in a metal container in an autoclave and
5 the atmosphere was made inert by passing over nitrogen. The metal container was closed and placed in a heater that was preheated to 290 °C and kept there for 2 hours. Subsequently the container was cooled to room temperature and the resulting polymer was removed from the glass vial.

10 Compounding

Glass fiber reinforced compositions, 70 wt.% polyamide, 30 wt.% glass fiber, were prepared on a twin-screw extruder employing standard compounding conditions for glass fiber reinforced thermoplastic polyamide compositions

15 The polymer compositions and physical properties of the polymers and mechanical properties of the polymers and compositions are listed in Table 1. Herein the dynamic modulus at 115 °C of test bars made of the polymers and compositions before and after conditioning at 100% RH are tested and compared. The trends in the mechanical performance of the polymers and the compositions are
20 comparable. The polymers and compositions according to the invention (E-1-5) retain their properties better than the other materials (CE-A-D).

Table 1. Polymer compositions, physical properties of the polymers and mechanical properties of the compositions for Examples 1-5 and Comparative Experiments A-D.

Examples /Comparative Experiments	Polyamide (mole %)	T _m (°C)	ΔH (J/g)	T _g (°C)	E' (115 °C), dry	E' (115 °C), 100% RH conditioned
E-1	6N/MACN (70/30)	348	26	191	+	+
E-2	6N/MACN (80/20)	346	52	175	+	+
E-3	10N/MACN (70/30)	293	26	152	+	+
E-4	12N/MACN (80/20)	285	24	146	+	+
E-5	18BB/MACBB (70/30)	283	25	150	+	+
CE-A	10N	325	82	139	+	-
CE-B	12N	295	88	120	+	-
CE-C	6T/MACT (70/30)	317	51	175	+	+/-
CE-D	6T/MACT (75/25)	320	43	168	+	+/-

CLAIMS

1. Semi-crystalline semi-aromatic polyamide consisting of 95 – 100 mole% of monomeric units derived from diamine and dicarboxylic acid, and 0 – 5 mole%
5 of one or more other monomeric units, the mole% being relative to the total mole % of monomeric units, wherein
 - a) the diamine comprises at least 90 mole% of aliphatic diamine comprising 5 – 40 mole%, relative to the total molar amount of diamine, of a cycloaliphatic diamine chosen from the group consisting of 1,4-diaminocyclohexane, 4,4'-methylene-bis(cyclohexylamine) (PAC), 3,3'-dimethyl-4,4'-
10 diaminocyclohexylmethane (MAC); 3,3',5,5'-tetramethyl-4,4'-diaminocyclohexylmethane; 2,2',3,3'-tetramethyl-4,4'-diaminocyclohexylmethane; norbornanediamine; and isophoronediamine (IPD), and any mixtures thereof;
 - 15 b) the dicarboxylic acid comprises at least 95 mole% of an aromatic dicarboxylic acid comprising 50 – 100 mole%, relative to the total molar amount of dicarboxylic acid, of naphthalene dicarboxylic acid (N), or biphenyl dicarboxylic acid (B), or a combination thereof.
2. Polyamide according to claim 1, wherein the cycloaliphatic diamine comprises
20 PAC, MAC or IPD, or any combination thereof.
3. Polyamide according to 1 or 2, wherein the diamine comprises 60 – 95 mole%, relative to the total molar amount of diamine, of a linear aliphatic α - ω diamine.
4. Polyamide according to claim 3, wherein the linear aliphatic α - ω diamine
25 comprises a C2 – C20 diamine.
5. Polyamide according to any of claims 1 - 4, wherein the diamine comprises 0 – 10 mole%, relative to the total molar amount of diamine, of another diamine, not being a linear aliphatic α - ω diamine or a cycloaliphatic diamine chosen from the group of claim 1.
- 30 6. Polyamide according to any of claims 1 - 5, wherein the dicarboxylic acid comprises 0 – 40 mole%, relative to the total molar amount of dicarboxylic acid, of another aromatic dicarboxylic acid.

7. Polyamide according to claim 6, wherein the other aromatic dicarboxylic acid comprises terephthalic acid or isophthalic acid, or a mixture thereof, and preferably comprises terephthalic acid.
8. Polyamide according to any of claims 1 - 7, wherein the dicarboxylic acid
5 comprises 0 – 5 mole%, preferably 0 – 2 mole%, relative to the total molar amount of dicarboxylic acid, of an aliphatic dicarboxylic acid.
9. Polyamide according to any of claims 1 - 8, wherein the polyamide has a T_m of at least 250 °C, a ΔH_m of at least 20 J/g, and a T_g of at least 135 °C
10. Thermoplastic polymer composition, comprising
10 (A) a semi-crystalline semi-aromatic polyamide according to any of claims 1 - 9; and
(B) a reinforcing agent; and optionally
(C) one or more further ingredient.
11. Composition according to claim 10, wherein the composition comprises
15 (A) 30 – 70 wt.% of the semi-crystalline semi-aromatic polyamide;
(B) 5 – 60 wt.% of an inorganic filler or a fibrous reinforcing agent, or a combination thereof;
(C) 0 – 40 wt.% of one or more further ingredient.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/076579

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08G69/26 C08G69/28 C08J3/20 C08K7/14
ADD.

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)

C08G C08J C08K

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, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 2 749 603 A1 (CHEIL IND INC [KR]) 2 July 2014 (2014-07-02) claims 1, 2, 4, 5 paragraph [0001] - paragraph [0002] paragraph [0049] - paragraph [0051]; examples 1-7 paragraph [0016] - paragraphs [0017], [0019]	1-11
A	US 2016/137782 A1 (KORY GAD [DE] ET AL) 19 May 2016 (2016-05-19) paragraphs [0001], [0042]; claim 3 paragraph [0081] paragraph [0083]	1-11



Further documents are listed in the continuation of Box C.



See patent family annex.

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Date of the actual completion of the international search

12 January 2018

Date of mailing of the international search report

21/02/2018

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

Paulus, Florian

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2017/076579

Patent document cited in search report		Publication date		Patent family member(s)		Publication date
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