

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
29 March 2001 (29.03.2001)

PCT

(10) International Publication Number  
**WO 01/21711 A1**

- (51) International Patent Classification<sup>7</sup>: C08L 77/00, 67/02, C08K 3/22, C08L 69/00
- (74) Agent: DERKS, Wilhelmus, Hubertus, Petrus; DSM Patents & Trademarks, P.O. Box 9, NL-6160 MA Geleen (NL).
- (21) International Application Number: PCT/NL00/00647
- (81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (22) International Filing Date:  
12 September 2000 (12.09.2000)
- (25) Filing Language: English
- (26) Publication Language: English
- (84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).
- (30) Priority Data:  
1013134 24 September 1999 (24.09.1999) NL
- (71) Applicant (*for all designated States except US*): DSM N.V. [NL/NL]; Het Overloon 1, NL-6411 TE Heerlen (NL).
- (72) Inventor; and
- (75) Inventor/Applicant (*for US only*): OTTENHEIJM, Johannes, Hendrikus, Godefried [NL/NL]; Heer Ottostraat 28, NL-6121 NA Born (NL).
- Published:**  
— *With international search report.*
- For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*



WO 01/21711 A1

(54) Title: FLAME-RETARDANT THERMOPLASTIC COMPOSITION WITH IMPROVED PROPERTIES

(57) Abstract: The invention relates to a flame-retardant thermoplastic composition which comprises at least (A) a thermoplastic polymer and (B) a flame-retardant system and which also contains (C) an n-alkanelactam-substituted polymer. Such a composition shows both a good flame retardancy, in particular a short afterflame time and little dripping, and also excellent mechanical properties. In particular, the n-alkanelactam-substituted polymer is a polyolefine, substituted with n-alkanelactam groups, with n being chosen from between 2 and 12. More in particular, the n-alkanelactam is polyvinyl pyrrolidone. Preferably, the thermoplastic polymer is a polycondensation polymer, in particular a polyamide, a polyester or a polycarbonate. Particularly suitable for use as the polyamide are PA-6, PA-6,6, PA-4,6 and semi-aromatic polyamides. The flame-retardant system may be both a halogen-containing and a halogen-free system. Preferably, the flame-retardant system comprises a halogen-containing compound and an antimony-containing compound.

FLAME-RETARDANT THERMOPLASTIC COMPOSITION  
5                   WITH IMPROVED PROPERTIES

The invention relates to a flame-retardant thermoplastic composition that comprises (A) a thermoplastic polymer and (B) a flame-retardant system.

10                   Such a composition is inter alia known from WO-95/18178 (DuPont), describing a polyamide composition which contains a combination of a brominated polystyrene and sodium antimonate as a flame-retardant system. Thermoplastic polymers are  
15 usually insufficiently flame-retardant per se, except for, for example, completely aromatic polyamides, for example aramide, and in practice a thermoplastic polymer is hence used in combination with a flame-retardant system consisting of one or more flame  
20 retardants and one or more synergists, i.e. compounds that promote the flame retardancy.

The disadvantage of such a composition is that the flame-retardant system has such an adverse effect on the composition's properties, in particular  
25 the mechanical, thermal, electrical and processing properties, that the amount of the flame-retardant system used is a compromise between on the one hand the required flame retardancy, for example for obtaining a V0 classification according to the UL-94 test of  
30 Underwriters Laboratories, and on the other the composition's required mechanical, thermal, processing and electrical properties. For example, the presence of antimony trioxide leads to thermal degradation of the composition at high processing temperatures, in

particular at temperatures above 280°C, which among other things leads to discolouration, fouling of the mould and corrosion of the processing equipment. In practice, the smallest possible amount of flame-  
5 retardant system is used in a thermoplastic composition and a certain degree of dripping is accepted in the UL-94 test.

The aim of the invention is hence to provide a thermoplastic composition with good flame-  
10 retardant, thermal, processing and mechanical properties, in particular at a high temperature, more in particular at a temperature above 280°C.

The inventors have now surprisingly found that a flame-retardant thermoplastic composition that  
15 comprises at least (A) a thermoplastic polymer and (B) a flame-retardant system and that also contains (C) an n-alkanelactam-substituted polymer, shows both a good flame retardancy, particular a short after-flame time, more particular a short total after-flame time and  
20 excellent mechanical properties. After-flame time is defined according to the UL-94 test as the time a test specimen burns till extinction after a flame application has stopped. Total after-flame time is the combined time ( $t_1+t_2$ ) for 5 specimen in which  $t_1$  is the  
25 after-flame time after a first flame application and  $t_2$  is the after-flame time after a second flame application.

As an added advantage of the composition according to the invention it was found that the  
30 dripping according to UL-94 could be greatly reduced, in particular in compositions according to the invention that also contain glass fibres. On account of

the increasingly stringent requirements that are being imposed with respect to flame-retardant compositions, minimal dripping, preferably the absence of dripping, is most desirable.

5                   Another advantage of the composition according to the invention is that the amount of synergist, in particular the amount of antimony compound in halogen-containing flame-retardant systems can be reduced without the flame retardancy of the  
10 composition being adversely affected. As a result, compositions can be obtained which contain less synergist, in particular less antimony compounds, while they have particularly good flame-retardant properties.

                  Another advantage of the composition  
15 according to the invention is that discolouration of the composition, which may for example occur during the processing of the composition at high temperatures, can be reduced, for example by reducing the amount of synergist.

20                   From EP-A-401,740 (DuPont, USA) it is known to use polyvinyl pyrrolidone (PVP) as an additive in para-aramide fibres to obtain improved properties, in particular fibre strength, paintability, UV stability, strength after ageing under the influence of heat, and  
25 adhesion in composite materials. A fabric made from fibres prepared from about 11 wt.% PVP (relative to the total composition) and para-aramide also showed a greater resistance to heat of radiation (singeing behaviour) to which the fabric was exposed (Test Method  
30 1971, Section 5-1, of the National Fire Protection Association (NFPA)). Said publication does not mention the composition's dripping behaviour, in particular at

PVP concentrations equal to or less than 10 wt.%, relative to the total weight of the composition.

From US-5,236,982 (Cossement et al.) it is known that PVP is used as an additive in a surface  
5 layer on glass fibres which can subsequently be mixed into a thermoplastic polymer to obtain a glass-fibre reinforced thermoplastic composition. The concentration of PVP in a composition is typically approximately 0.008 wt.% (relative to the total composition).

10 According to the aforementioned publication, PVP is used as a lubricant to retain critical properties of the employed glass fibres, such as length and amount of fine particles.

In the context of this application an n-  
15 alkanelactam-substituted polymer is understood to be a polymer that contains a number of n-alkanelactam groups that are bound to the polymer via the N atom of the lactam. Suitable for use as the n-alkanelactam group is a group derived from an n-alkanelactam in which n is an  
20 integer and  $n > 1$ , in particular an n-alkanelactam in which n is an integer and is chosen from 2 up to and including 12. More in particular the n-alkanelactam is 2-ethanelactam (azacyclopropan-2-one), 3-propanelactam ( $\beta$ -propiolactam), 4-butanelactam ( $\gamma$ -butyrolactam or 2-  
25 pyrrolidone),  $\gamma$ -valerolactam, 5-pentanelactam ( $\delta$ -valerolactam), 3-methylvalerolactam, 6-  
methylvalerolactam, 6-hexanelactam ( $\epsilon$ -caprolactam), 7-  
heptanelactam ( $\phi$ -enantholactam), 8-octanelactam ( $\gamma$ -caprylolactam), 9-nonanelactam ( $\theta$ -pelargolactam), 10-  
30 decanelactam ( $\omega$ -caprinolactam), 11-undecanelactam or 12-dodecanelactam ( $\omega$ -laurolactam).

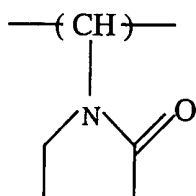
Any aliphatic or aromatic polymer can be chosen as the polymer for the n-alkanelactam substituted polymer. In particular, the polymer is a polyolefine, for example a polyethylene, polypropene or a copolymer thereof. The n-alkanelactam groups may, independently of one another, be the same or differ per polymer molecule.

In particular, the n-alkanelactam-substituted polymer is a compound that consists of units according to Formula 1



where  $R_1$  and  $R_2$ , independently of one another, are H or an n-alkanelactam group, providing that each molecule contains at least 1 n-alkanelactam group. Preferably, the n-alkanelactam-substituted polymer is polyvinyl pyrrolidone (PVP). Polyvinyl pyrrolidone is commercially available in several molecular weights as Luviskol<sup>®</sup> (BASF, Germany) and can be obtained through linear polymerization of the monomer N-vinyl-2-pyrrolidone. Polyvinyl pyrrolidone consists of units according to formula 2

25



(2)

An n-alkanelactam-substituted polymer with virtually any molecular weight can be used in the composition according to the invention. The molecular weight may vary in a very wide range, in particular  
5 from approximately 7,000 to approximately 2,000,000. Preferably, a molecular weight of between approximately 45,000 and approximately 2,000,000 is chosen. The amount of n-alkanelactam-substituted polymer can be freely chosen. Preferably the amount is between 0.01  
10 and 10 wt.%; preferably, the amount is between 0.1 and 5 wt.%, relative to the weight of the thermoplastic polymer. N-alkanelactam-substituted polymer that contains a small amount of impurities or unreacted monomer, for example PVP with a small amount of 2-  
15 pyrrolidone, for example less than 1 wt.%, relative to the n-alkanelactam-substituted polymer, is also covered by the definition of n-alkanelactam-substituted polymer according the invention.

Any thermoplastic polymer known to a person  
20 skilled in the art can be used as thermoplastic polymer (A) in the composition according to the invention. Preferably, a polycondensation polymer is chosen, in particular a polyester, a polycarbonate or a polyamide.

Examples of suitable polyesters are  
25 poly(cyclo)alkylene terephthalates or copolyesters thereof with isophthalic acid, for example polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polycyclohexylene dimethylene terephthalate (PCT), polyalkylene naphthalates, for example polyethylene  
30 naphthalate (PEN), polypropylene naphthalate (PPN) and polybutylene naphthalate (PBN), polyalkylene dibenzoates, for example polyethylene dibenzoate, and

copolyesters of said polyesters. Preferably, PET, PBT, PEN or PBN is chosen. Also suitable are block copolyesters which, in addition to hard polyester segments chosen from the aforementioned group, also  
5 contain soft polyester segments derived from at least a polyether or an aliphatic polyester. Examples of such block copolyesters with elastomeric properties are for example described in "Encyclopedia of Polymer Science and Engineering", Vol. 12, p. 75 ff. (1988), John Wiley  
10 & Sons, and in "Thermoplastic Elastomers", 2nd Ed., Chapter 8 (1996), Hanser Verlag, the relevant contents of which are hereby understood to have been mentioned.

Examples of a suitable polyamide are aliphatic polyamides, for example PA-6, PA-6,6, PA-9,  
15 PA-11, PA-4,6, polyamides based on 2-methylpentamethylene diamine and adipic acid and copolyamides thereof, semi-aromatic polyamides based on aromatic dicarboxylic acids, for example isophthalic acid and terephthalic acid, and aliphatic diamines, for  
20 example hexane diamine, for example PA-6/T, PA-6.6/T, PA-6/6.T, PA-6,6/6,T, PA-6,6/6/6,T, PA-6T/6I, PA-6,6/6,I/6,T and PA-6,6/2-MPMD,6, and completely aromatic polyamides.

In particular, the thermoplastic polymer  
25 has a melting point equal to or above 280° C.

Surprisingly, it has also been found that the composition according to the invention can be processed at higher temperatures, in particular above 280°C, without noticeable chemical degradation. This  
30 makes the use of n-alkanelactam-substituted polymer in thermoplastic polymers with high melting points, for example polyamide-4,6 and semi-aromatic polyamides,



particularly suitable. Preferably, the polyamide is chosen from the group comprising PA-6, PA-6,6, PA-4,6 and semi-aromatic polyamides.

The composition according to the invention comprises a flame-retardant system (B), in particular a halogen-containing system, comprising at least a halogen-containing compound. Suitable for use as the halogen-containing system are for example systems that contain halogen-containing flame retardants as the flame retardant, for example brominated polystyrene, for example Pyrochek<sup>®</sup> 68PB from Ferro Corporation (USA) and Saytex<sup>®</sup> HP7010 from Albemarle (USA), brominated polyphenylene ether, for example PO64P<sup>®</sup> from Great Lakes (USA), polydibromostyrene, for example PDBS80<sup>®</sup> from Great Lakes, polytribromostyrene, polypentabromostyrene, polydichlorostyrene, polytrichlorostyrene, polypentachlorostyrene, polytribromo-alpha-methylstyrene, polydibromo-p-phenylene oxide, polytribromo-p-phenylene oxide, polydichloro-p-phenylene oxide, polybromo-p-phenylene oxide, polybromo-o-phenylene oxide, pentabromobenzyl acrylate, for example FR1025<sup>®</sup> from AmeriBrom (USA), ethylene bis-tetrabromophthalimide, for example Saytex<sup>®</sup> BT-93W from Albemarle (USA), polybromobiphenyl, brominated phenoxy- and chlorine-containing flame retardants such as DeChlorane<sup>®</sup> (Occidental Chemical Corporation, USA) and other brominated compounds such as Saytex<sup>®</sup> 8010 from Albemarle (USA).

The flame-retardant systems may also contain a synergist. Suitable synergists are:

- antimony-containing compounds, for example antimony trioxide, for example Bluestar<sup>®</sup> RG (Campine,

- Belgium), antimony tetraoxide, antimony pentoxide, sodium antimonate, for example Pyrobloc<sup>®</sup> SAP-2 (Cookson Specialty Additives), antimony tartrate;
- 5 - zinc borate, for example Firebrake<sup>®</sup> ZB (Borax Inc., USA)
  - magnesium hydroxide, aluminium hydroxide, iron oxide, zinc oxide, calcium oxide and analogous substances.

In particular, the halogen-containing system contains an antimony-containing compound.

10 A halogen-free flame-retardant system may also be chosen as the flame-retardant system. Suitable halogen-free flame retardants are:

- metal-containing compounds such as magnesium hydroxide and aluminium hydroxide;
- 15 - nitrogen-containing compounds such as melamine, melamine cyanurate, melam, melem and melon;
- phosphorus-containing compounds such as red phosphorus, melamine phosphate, melamine polyphosphate, for example Melapur<sup>®</sup> 200 (DSM, the
- 20 Netherlands) and PMP-100<sup>®</sup> (Nissan Chemical Industries, Japan), melam polyphosphate, for example PMP-200<sup>®</sup> (Nissan Chemical Industries), melem polyphosphate, for example PMP-300<sup>®</sup> (Nissan Chemical Industries), phosphazene-based compounds and ammonium
- 25 polyphosphate.

The composition according to the invention may also contain other additives known to a person skilled in the art that are customarily used in polymer compositions, providing they do not essentially detract

30 from the invention, in particular pigments, processing aids, for example mould release agents, agents accelerating crystallization, nucleating agents,

softeners, UV and heat stabilizers and the like. Other substances that promote the flame retardancy may optionally also be added, for example carbon-forming substances such as polyphenylene ether and  
5 polycarbonate and substances modifying the dripping behaviour, for example fluoropolymers such as polytetrafluoroethylene. In particular, the composition according to the invention contains an inorganic filler or reinforcing agent. Suitable for use as an inorganic  
10 filler or reinforcing agent are all the fillers known to a person skilled in the art, for example glass fibres, metal fibres, graphite fibres, aramide fibres, glass beads, aluminium silicates, asbestos, mica, clay, calcined clay and talcum. Preferably, glass fibres are  
15 chosen.

Preferably, the composition according to the invention contains

- A) 40-90 wt.% of a thermoplastic polymer, to be chosen from the group comprising polyamides, polycarbonates  
20 and polyesters
  - B) 10-40 wt.% of a halogen-containing flame-retardant system
  - C) 0.1-5 wt.% polyvinyl pyrrolidone with a molecular weight between 45,000 and 2,000,000
  - 25 D) 0-50 wt.% glass fibres
  - E) 0-20 wt.% other additives,
- where (A+B+C+D+E) equals 100 wt.%.

The thermoplastic composition according to the invention can be obtained in a simple way by means  
30 of melt-mixing. Preferably, use is made of an extruder fitted with means for dosing all the desired components to the extruder, either to the extruder's throat or to

the melt. According to one embodiment the n-alkanelactam-substituted polymer may form part of the flame-retardant system B. In another embodiment the n-alkanelactam-substituted polymer can be fed directly to the melt separately or it can first be mixed with the polymer granules.

The composition accordingly to the invention is suitable for making any object known to the skilled person, in particular a film, a fiber, a sheet and a moulded part.

The invention will be further elucidated with reference to the following examples and comparative examples.

#### 15 Examples

##### Examples I - XII and Comparative Examples A - E

A number of compositions were prepared using the following components:

#### 20 (A) thermoplastic polymer:

- polyamide 4,6: Stanyl<sup>®</sup> KS200, DSM N.V., the Netherlands
- polyamide 6,6/6T: Grivory<sup>®</sup> FE5011, EMS Chemie. Switzerland
- 25 • polyester PBT: Arnite<sup>®</sup> T04200, DSM N.V., the Netherlands
- polyamide 6: Akulon<sup>®</sup> K122, DSM N.V., the Netherlands

#### 30 (B) flame-retardant system:

- Pyrochek<sup>®</sup> 68PBi (brominated polystyrene - Ferro Corp., USA)

- PDBS80<sup>®</sup> (polymerized dibromostyrene - Great Lakes, USA)
- Saytex<sup>®</sup> BT-93W (ethylene bis-tetrabromo-phthalimide - Albemarle, USA)
- 5 • GR2617<sup>®</sup> (Sb<sub>2</sub>O<sub>3</sub>, as a PA-6 masterbatch, containing 80% Sb<sub>2</sub>O<sub>3</sub> - Campine, Belgium)
- GR2616<sup>®</sup> (Sb<sub>2</sub>O<sub>3</sub>, as a PBT masterbatch, containing 80% Sb<sub>2</sub>O<sub>3</sub> - Campine, Belgium)
- Saytex<sup>®</sup> HP-7010 (brominated polystyrene -
- 10 Albemarle, USA)
- Saytex<sup>®</sup> 8010 (Albemarle, USA)

(C) polyvinyl pyrrolidone:

- Luviskol<sup>®</sup> K90 (BASF, Germany), M<sub>w</sub> = 1,200,000-2,000,000
- 15 • Luviskol<sup>®</sup> K30 (BASF, Germany), M<sub>w</sub> = 45,000-55,000
- Luviskol<sup>®</sup> K17 (BASF, Germany), M<sub>w</sub> = 7,000-11,000

Glass fibres:

- 20 • OC 173X-10c<sup>®</sup> (Owens-Corning, USA)
- OC 183-11c<sup>®</sup> (Owens-Corning, USA)

Lubricant : AC 540A<sup>®</sup> (Allied, USA)

Stabilizer : Irganox<sup>®</sup> 1098 (Ciba Geigy, Switzerland)

25 Filler: Stealene (talc, Luzenac, France)

The compositions are given in Tables 1, 3, 5, 7 and 9. All the amounts are wt.%, relative to the total composition. All the compositions were prepared

30 in the melt.

The flame retardancy according to UL-94 (classification, total after-flame time), the dripping behaviour (expressed as number of dripping specimen) and mechanical properties of all the compositions were determined using 5 specimen rods. The results are summarized in Tables 2, 4, 6, 8 and 10. The spiral flow was determined as the length of a polymer flow in a flat spiral with a width of 15 mm and a cross-section of 1 mm mounted on an Arburg CMD injection-moulding machine at an effective pressure of 900 bar.

#### Discussion

Comparison of Comparative Example A with Examples I, II and III shows that a small amount of PVP in the composition according to the invention leads to a substantial reduction in the total after-flame time (of up to 50%) and total absence of dripping. The mechanical properties, in particular the modulus of elasticity, tensile strength, elongation-at-break, notched impact resistance and spiral flow, remain unchanged or improve, in particular the tensile strength.

Comparison of Comparative Example B with Examples IV and V, of Comparative Example C with Examples VI and VII, of Comparative Example D with Example VIII and of Comparative Example E with Example IX shows that when the synergist concentration is lowered in the presence of an amount of PVP, the mechanical properties, in particular the tensile strength and the elongation-at-break, increase, whereas the total after-flame time remains more or less unchanged ; dripping is completely absent with these

compositions according to invention, too.

Examples X, XI and XII (Tables 7 and 8) illustrate that the flame retardancy effect according to the invention does not depend on the molecular weight of the PVP. For low molecular weights however, in particular lower than 10,000, the concentration can be increased, a combination of molecular weight and concentration which can be selected by a skilled person by simple optimisation. When Experiment XII was repeated with a concentration of 6 weight%, the flame retardancy properties were comparable with those of Example X and XI.

Table 1: PA-4,6 compositions

Component	A	I	II	III	B	IV	V	C	VI	VII
<b>Polymer</b>										
Stanyl® KS200	42.74	42.49	42.24	41.74	41.00	43.00	44.00	32.84	34.64	35.59
<b>Flame-retardant</b>										
Pyrochek® 68PBi	18.75	18.75	18.75	18.75	-	-	-	-	-	-
PDBS80®	-	-	-	-	21.00	21.00	21.00	19.50	19.50	19.50
GR2617®	7.81	7.81	7.81	7.81	7.50	5.00	3.75	7.00	4.70	3.50
<b>PVP</b>										
Luviskol® K90	-	0.25	0.50	1.00	-	0.50	0.75	-	0.50	0.75
<b>Glass fibre</b>										
OC 173X-10c®	30	30	30	30	30	30	30	40	40	40
<b>Lubricant and stabilizer</b>										
	0.70	0.70	0.70	0.70	0.50	0.50	0.50	0.66	0.66	0.66



Table 2 : Properties of the compositions of Table 1

Property	Unit	ISO	A	I	II	III	B	IV	V	C	VI	VII
<b>Mechanical properties</b>												
Modulus of elasticity	GPa	527/1A	11.6	11.5	11.4	11.6	11.3	11.3	11.3	15.0	15.4	15.4
Tensile strength	MPa	527/1A	172	183	179	181	150	165	176	161	180	186
Elongation-at-break	%	527/1A	2.3	2.4	2.3	2.2	1.8	1.9	2.1	1.5	1.6	1.7
Notched impact resistance (IZOD)	kJ/m <sup>2</sup>	180/1A	10.6	10.4	10.3	9.3	12.2	11.5	11.6	15.7	13.9	16.1
Spiral flow (900 bar)	mm		102	95	103	100	127	128	127	115	113	117
UL94V 0.8 mm thick												
Specimen 125x13x0.8 mm												
48 hours/23°C/50% R.H.												
Classification			V0	V0	V0	V0	V0	V0	V0	V0	V0	V0
Total after-flame time	sec.		21	13	10	10	24	23	28	10	10	9
Dripping specimens			1	0	0	0	4	0	0	0	0	0
168 hours/70°C												
Classification			V0	V0	V0	V0	V0	V0	V0	V0	V0	V0
Total after-flame time	sec.		23	24	12	12	31	16	39	10	10	10
Dripping specimen			1	0	0	0	5	0	0	0	0	0

Table 3 : PA-6,6/6T compositions

Component	D	VIII
<b>Polymer</b>		
Grivory® FE5011	42.87	42.27
<b>Flame-retardant</b>		
Pyrochek® 68PBi	18.75	18.75
GR2617®	7.81	3.91
<b>FVP</b>		
Luviskol® K90	-	0.50
<b>Glass fibre</b>		
OC 173X-10c®	30	30
<b>Lubricant and stabilizer</b>	0.57	0.57

5 Table 4 : Properties of the compositions of Table 3

Property	Unit	ISO	D	VIII
<b>Mechanical properties</b>				
Modulus of elasticity	GPa	527/1A	12.5	12.4
Tensile strength	MPa	527/1A	178	188
Elongation-at-break	%	527/1A	2.0	2.1
Notched impact resistance (IZOD)	kJ/m <sup>2</sup>	180/1A	10.1	10.1
Spiral flow (900 bar)	mm		120	120
UL94V 0.8 mm thick				
Specimen 125x13x0.8 mm				
<u>48 hours/23°C/50% R.H.</u>				
Classification			V0	V0
Total after-flame time	sec.		10	14
Dripping specimens			1	0
<u>168 hours/70°C</u>				
Classification			V0	V0
Total after-flame time	sec.		10	10
Dripping specimen			0	0

Table 5: PBT-compositions

Component	E	IX
<b>Polymer</b>		
Arnite® T04200	56	57.5
<b>Flame-retardant</b>		
Saytex® BT-93W	9	9
GR2616®	5	2.5
<b>PVP</b>		
Luviskol® K90	-	1.0
<b>Glass fibre</b>		
OC 183-11c®	30	30

5 Table 6 : Properties of the compositions of Table 5

Property	Unit	ISO	E	IX
UL94V 2 mm thick				
Specimen 125x13x2 mm				
48 hours/23°C/50% R.H.				
Classification			V0	V0
Total after-flame time	sec.		10	10
Dripping specimens			0	0
<u>168 hours/70°C</u>				
Classification			V0	V0
Total after-flame time	sec.		10	10
Dripping specimen			0	0

Table 7: PA-4,6 compositions with different amounts of PVP

Component	X	XI	XII
<b>Polymer</b>			
Stanyl® KS200	44	44	44
<b>Flame-retardant</b>			
PDBS80®	21	21	21
GR2617®	3.75	3.75	3.75
<b>PVP</b>			
Luviskol® K90	0.75	-	-
Luviskol® K30	-	0.75	-
Luviskol® K17	-	-	0.75
<b>Glass fibre</b>			
OC 173X-10c®	30	30	30
<b>Lubricant and stabilizer</b>	0.50	0.50	0.50

5

Table 8 : Properties of the compositions of Table 7

Property	Unit	ISO	X	XI	XII
<b>Mechanical properties</b>					
Modulus of elasticity	GPa	527/1A	11.2	11.1	11.2
Tensile strength	MPa	527/1A	181	181	174
Elongation-at-break	%	527/1A	2.2	2.3	2.1
Notched impact resistance (IZOD)	kJ/m <sup>2</sup>	180/1A	11.1	10.9	11.8
<b>UL94V 0.8 mm thick</b>					
Specimen 125x13x0.8 mm					
48 hours/23°C/50% R.H.					
Classification			V0	V0	V0
Total after-flame time	sec.		22	28	55
Dripping specimens			0	0	5
<b>168 hours/70°C</b>					
Classification			V0	V0	V2
Total after-flame time	sec.		22	24	51
Dripping specimen			1	0	5

Table 9: PA-6 compositions

Component	F	XIII	G	XIV
<b>Polymer</b>				
Akulon® K122	44.45	46.02	53.1	54.65
<b>Flame-ratardant</b>				
Saytex® HP7010	20.6	20.6	-	-
Saytex® 8010	-	-	12	12
GR 2617®	4.15	2.08	4.1	2.05
<b>PVP</b>				
Luviskol® K90	-	0.5	-	0.5
<b>Glassfibre</b>				
OC 173X-10c®	30	30	-	-
<b>Filler</b>				
Stealene	-	-	30	30
<b>Other additives</b>	0.8	0.8	0.8	0.8

Table 10: Properties of the compositions of Table 9

Component	Unit	Iso	F	XIII	G	XIV
<b>Mechanical properties</b>						
Modulus of elasticity	GPa	527/1A	10.7	11.6	7.4	7.5
Tensile strength	MPa	527/1A	148	152	69	66
Elongation at break	%	527/1A	2.5	2.2	1.6	1.3
Notched Impact resistance (Izod)	Kj/m <sup>2</sup>	180/1A	-	-	-	-
UL 94V 0.8 mm thick						
Specimen 125 x 13 x 0.8 mm						
48 hours/23°C/50%R.H.						
Classification						
Total after-flame time	sec.		V0	V0	V2	V2
(5 specimen rods)			45	10	13	20
Number of specimens that show dripping			100%	100%	100%	100%
168 hours/70°C						
Classification						
Total after-flame time	Sec.		V0	V0	V2	V2
(5 specimen rods)			38	10	15	18
Number of specimens that show dripping			100%	100%	100%	100%

CLAIMS

1. Flame-retardant thermoplastic composition that  
comprises at least (A) a thermoplastic polymer and  
5 (B) a flame-retardant system, characterized in  
that the composition also contains (C) an n-  
alkanelactam-substituted polymer.
2. Flame-retardant thermoplastic composition  
according to Claim 1, characterized in that the  
10 amount of n-alkanelactam-substituted polymer is  
0.01 - 10 wt.%, relative to the total composition.
3. Flame-retardant thermoplastic composition  
according to any of Claims 1-2, characterized in  
that the molecular weight of the n-alkanelactam-  
15 substituted polymer is between 7,000 and  
2,000,000.
4. Flame-retardant thermoplastic composition  
according to any of Claims 1-3, characterized in  
that the n-alkanelactam-substituted polymer is a  
20 polyolefine substituted with n-alkanelactam  
groups, with n being chosen from between 2 and 12.
5. Flame-retardant thermoplastic composition  
according to Claim 4, characterized in that the n-  
alkanelactam-substituted polymer is polyvinyl  
25 pyrrolidone.
6. Flame-retardant thermoplastic composition  
according to any one of Claims 1-5, characterized  
in that the thermoplastic polymer is a  
polycondensation polymer.
- 30 7. Flame-retardant thermoplastic composition  
according to Claim 6, characterized in that the

- polycondensation polymer is chosen from the group comprising polyesters, polycarbonates and polyamides.
- 5 8. Flame-retardant thermoplastic composition according to Claim 7, characterized in that the polyamide is chosen from the group comprising PA-6, PA-6,6, PA-4,6 and semi-aromatic polyamides.
- 10 9. Flame-retardant thermoplastic composition according to any one of Claims 1-8, characterized in that the flame-retardant system comprises at least a halogen-containing compound and an antimony-containing compound.
- 15 10. Flame-retardant thermoplastic composition according to any one of Claims 1-9, characterized in that a filler is also present.
11. Flame-retardant thermoplastic composition according to Claim 10, characterized in that the filler is glass fibre.
- 20 12. Use of an n-alkanelactam-substituted polymer for improving the flame-retardant behaviour of a thermoplastic composition comprising at least a thermoplastic polymer (A) and a flame-retardant system (B).
- 25 13. Halogen containing flame-retardant thermoplastic composition containing
- A) 40-90 wt.% of a thermoplastic polymer, to be chosen from the group comprising polyamides, polycarbonates and polyesters
- B) 10-40 wt.% of a halogen-containing flame
- 30 retardant system
- C) 0.1-5 wt.% polyvinyl pyrrolidone with a molecular weight between 45,000 and 2,000,000.
- D) 0-50 wt.% glass fibres



E) 0-20 wt.% other additives which are not the same as A, B, C or D, where (A+B+C+D+E) amounts to 100 wt.%.

14. Film, fibre or moulded part obtainable using the composition of any of Claims 1-11 and Claim 13.
15. Composition as described and elucidated with reference to the examples.

# INTERNATIONAL SEARCH REPORT

Intern: al Application No  
PCT/NL 00/00647

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 C08L77/00 C08L67/02 C08K3/22 C08L69/00		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b>		
Minimum documentation searched (classification system followed by classification symbols) IPC 7 C08L C09K 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 practical, search terms used)  EPO-Internal		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 381 172 A (HOECHST AG) 8 August 1990 (1990-08-08) page 6, line 14-45 ---	
A	EP 0 401 740 A (DU PONT) 12 December 1990 (1990-12-12) cited in the application page 5, line 21-29 claims ---	
A	WO 95 18178 A (DU PONT ;HAYASHI RYUICHI (JP)) 6 July 1995 (1995-07-06) cited in the application page 2, line 6 -page 3, line 30 ---	
A	EP 0 802 268 A (TORAY INDUSTRIES) 22 October 1997 (1997-10-22) page 3, line 6-22; claims ---	
-/--		
<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C.		
<input checked="" type="checkbox"/> 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 "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed		
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. "&" document member of the same patent family		
Date of the actual completion of the international search	Date of mailing of the international search report	
13 November 2000	21/11/2000	
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer  Leroy, A	

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/NL 00/00647

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 144 175 A (GAF CORP) 12 June 1985 (1985-06-12) page 9, line 19-31 ----	
A	EP 0 866 097 A (KANEKAFUCHI CHEMICAL IND) 23 September 1998 (1998-09-23) claims page 13, line 12-25 -----	

## INTERNATIONAL SEARCH REPORT

Information on patent family members

Internat'l Application No

PCT/NL 00/00647

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 0381172	A	08-08-1990	DE 3903099 A	16-08-1990
			AU 618167 B	12-12-1991
			AU 4899590 A	09-08-1990
			CA 2009139 A	02-08-1990
			DE 59004370 D	10-03-1994
			JP 2233761 A	17-09-1990
EP 0401740	A	12-12-1990	US 5073440 A	17-12-1991
			AT 158030 T	15-09-1997
			AU 628177 B	10-09-1992
			AU 5626990 A	06-12-1990
			BR 9002639 A	20-08-1991
			CA 2017827 A	05-12-1990
			CN 1050748 A,B	17-04-1991
			DE 69031405 D	16-10-1997
			DE 69031405 T	09-04-1998
			HK 1001191 A	29-05-1998
			IL 94505 A	15-11-1992
			JP 2903173 B	07-06-1999
			JP 3069610 A	26-03-1991
			MX 171493 B	29-10-1993
			NZ 233918 A	26-07-1991
			PT 94271 A	20-03-1991
			TR 24636 A	01-01-1992
			US 5135687 A	04-08-1992
WO 9518178	A	06-07-1995	JP 7196875 A	01-08-1995
			CA 2178716 A	06-07-1995
			EP 0737226 A	16-10-1996
EP 0802268	A	22-10-1997	JP 9268433 A	14-10-1997
			CN 1167844 A	17-12-1997
			US 5922830 A	13-07-1999
EP 0144175	A	12-06-1985	US 4778839 A	18-10-1988
			US 4766164 A	23-08-1988
			US 4797437 A	10-01-1989
			AT 37556 T	15-10-1988
			BR 8405978 A	27-08-1985
			CA 1263778 A	05-12-1989
			DE 3474315 D	03-11-1988
			JP 1049425 B	24-10-1989
			JP 1561400 C	31-05-1990
			JP 60135450 A	18-07-1985
			ZA 8408745 A	31-07-1985
			IL 73480 A	31-03-1988
EP 0866097	A	23-09-1998	JP 10110090 A	28-04-1998
			US 6096818 A	01-08-2000
			WO 9815596 A	16-04-1998