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(54) Titre : MELANGE POLYMERIQUE A BASE DE POLY(TEREPHTALATE DE BUTYLENE) ET D'ELASTOMERE THERMOPLASTIQUE; ARTICLES A BASE DE CE MELANGE  
(54) Title: POLYMER MIXTURE COMPRISING POLYBUTYLENE TEREPHTHALATE AND THERMOPLASTIC ELASTOMER; ARTICLES FORMED THEREFROM

(57) **Abrégé/Abstract:**

The invention relates to polymer mixtures which comprise a polybutylene terephthalate, a thermoplastic elastomer, and a brominated polystyrene. The polymer mixtures according to the invention have a combination of the following favourable properties: a good flexibility, a good flow of the melt, good flame-retarding properties and a high tracking index.



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1 ABSTRACT:

The invention relates to polymer mixtures which comprise a polybutylene terephthalate, a thermoplastic elastomer, and a brominated polystyrene. The polymer mixtures according to the invention have a combination of the following favourable properties: a good flexibility, a good flow of the melt, good flame-retarding properties and a high tracking index.

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Polymer mixture comprising polybutylene terephthalate and thermoplastic elastomer; articles formed therefrom.

10           The invention relates to a polymer mixture which comprises the following constituents:

- A. a polybutylene terephthalate;
- B. a thermoplastic elastomer.

15           The invention also relates to articles formed from the polymer mixture according to the invention. Polymer mixtures which comprise a polybutylene terephthalate and a thermoplastic elastomer, for example, a polyether-ester block copolymer, are known, 20 for example, from DE-C-2338615 and DE-B-2460257. The addition of flame-retardants is not mentioned in these publications.

25           The addition of flame-retarding, halogen-containing compounds to polybutylene terephthalates is described in EP-A-0065777. A great number of compounds are mentioned as suitable flame-retardants including, for example, polycarbonate oligomers prepared starting from brominated bisphenol A and brominated polystyrene.

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Polymer mixtures which comprise a polybutylene terephthalate, a brominated polystyrene, an aromatic polycarbonate and an agent to improve the impact strength are described in EP 322,934. The polymer mixtures just mentioned hereinbefore have a good tracking index and good flame-retarding properties.

The invention provides polymer mixtures which comprise a polybutylene terephthalate and a thermoplastic elastomer. The polymer mixtures according to the invention have the following combination of properties: a good flexibility, a good flow of their melt, good flame-retarding properties and a high tracking index.

The polymer mixture according to the invention is characterised in that the polymer mixture comprises in addition C. a brominated polystyrene as a flame-retardant.

The polymer mixture according to the invention preferably comprises a polyether-ester block copolymer, or a polyester-ester urethane or a polyether imide ester as constituent B.

The composition of the polymer mixture according to the invention is preferably such that the polymer mixture comprises per 100 parts by weight of the sum of the constituents A, B and C

50-80 parts by weight of a constituent A,

5-20 parts by weight of a constituent B, and

15-30 parts by weight of a constituent C.

The polymer mixture according to the invention comprises as constituent A, a polymer or a copolymer

1 which is built up for more than 70 mol% from units  
derived from terephthalic acid and butane-1,4-diol.

It is possible to use in the polymer mixtures  
5 according to the invention a metal compound (for  
example, antimony oxide) which cooperates synergisti-  
cally with the brominated polystyrene.

The polymer mixture according to the invention  
10 comprises at any rate the following constituents:

- A. a polybutylene terephthalate
- B. a thermoplastic elastomer, and
- C. a brominated polystyrene.

15 A. Polybutylene terephthalate

Polybutylene terephthalate is a polymer known  
per se. It is derived from butane-1,4-diol and  
terephthalic acid. It is possible to use in the polymer  
mixtures copolyesters in which a part of the diol com-  
20 pound and/or a part of the terephthalic acid have/has  
been replaced by another diol and/or carboxylic acid.  
For the polymer mixture according to the invention it is  
to be preferred to use a polybutylene terephthalate which  
is built up for more than 70 mol% from units derived  
25 from butane-1,4-diol and terephthalic acid.

It is possible to use polybutylene terephtha-  
late in which a branching agent, for example, a glycol  
having three or more hydroxyl groups or a trifunctional  
30 or multifunctional carboxylic acid, has been incor-  
porated.

B. Thermoplastic elastomer

Various types of thermoplastic elastomers may  
35 be used in the polymer mixture according to the inven-  
tion. Examples are the known polyether-ester block copo-

1 lymers, polyester-ester urethanes and polyether imide  
esters.

Polyether-ester block copolymers are generally  
5 obtained by condensation polymerisation of one or more  
dicarboxylic acids, one or more long-chain glycols and  
one or more short-chain glycols. Examples of suitable  
polyether-ester block copolymers are described in  
DE-C-2338615 and DE-B-2460258 as well as in the litera-  
10 ture references mentioned therein.

As a polyester-ester urethane is preferably  
used a block copolymer which is built up from  
polyester-ester units which are bonded by means of  
15 groups of the formula  $R_1 [NHC(O)]_p$ , wherein  $R_1$  is a  
polyfunctional group having at most 30 carbon atoms and  
 $p$  has a value of 2 or 3, and in which the polyester-  
-ester units are built up from blocks having a number of  
ester units of the formula  $-OGOC(O)R_2C(O)-$  and blocks  
20 having a number of ester units which are capable of  
forming a bifunctional polyester or polyester amide  
having a melting-point lower than  $100^\circ\text{C}$ , which blocks are  
bonded together by ester bonds, in which at least 80  
mol% of the  $R_2$  groups are 1,4-phenylene radicals and in  
25 which at least 80 mol% of the G-groups are a tetramethy-  
lene group.

Such polymers and their mode of preparation  
are described for example in EP-A-0102115,  
30 US-A-4,186,257 and US-A-4,228,249.

As a polyether imide ester is preferably used  
a block copolymer obtained by reaction of a diol having  
a low molecular weight, a dicarboxylic acid, a  
35 poly(oxyalkylene) diamine and a tricarboxylic acid or a  
derivative thereof.

1 Such polymers are described, for example, in  
US-A-4,544,734, US-A-4,556,705 and US-A-4,556,688.

C. Brominated polystyrene

5 Brominated polystyrenes are known flame-  
-retardants for synthetic resins. They are commercially  
available. They may comprise, for example, approximately  
40-70% by weight of bromine.

10 The use of other flame-retardants seems not  
possible: in that case the above-mentioned combination  
of properties is not obtained.

In addition to the constituents mentioned  
15 hereinbefore the polymer mixture according to the inven-  
tion may moreover comprise conventionally used  
additives. Suitable additives are pigments and dyes,  
reinforcing fibres, stabilisers, transesterification  
inhibitors, flame-retardants, mould-release agents, etc.

20 The polymer mixture according to the invention  
can be obtained according to the methods conventionally  
used for the preparation of polymer mixtures, for exam-  
ple, by melt extrusion.

25 Examples I to IV, comparative examples A and B

The following constituents were used in the  
examples:

- 30 PBT: a polyester derived from butanediol and  
terephthalic acid having an intrinsic visco-  
sity of 1.20 dl/g, measured in a 60/40 mixture  
of phenol/tetrachloroethane at 25°C.
- TPE-1: a polyether imide ester commercially available  
under the indication LOMOD® J10.
- 35 TPE-2: a polyether ester block copolymer commercially  
available under the indication HYTREL® 4059.

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- TPE-3: a polyether ester having polypropylene glycol blocks commercially available under the indication ARNITEL\* PM 380.
- FR-1: a brominated polystyrene having a bromine content of approximately 65% by weight.
- FR-2: a brominated carbonate polymer.
- AOC: a mixture of 85% by weight of  $Sb_2O_3$  and 15% of a carrier polymer.
- T: a polytetrafluorethylene compound.
- O: a mixture of other conventionally used additives (stabiliser + mould-release agent).

The constituents mentioned hereinbefore were compounded in an extruder and pelletised in the quantities as indicated in the table hereinafter. Standardised test pieces were injection-moulded from the resulting pellets to determine the flammability class according to UL-94, to determine the comparative tracking index according to IEC, 1979 (3<sup>rd</sup> edition), to test the glow wire test according to IEC: 695-21 (1<sup>st</sup> edition), and to determine a few mechanical properties.

The results obtained are also recorded in the table hereinafter.

\* A trade-mark.

1 <u>TABLE</u>						
Example No.	A	B	I	II	III	IV
Composition (parts by weight)						
5	.PBT	74.3	57.2	64.3	59.3	64.3
	.TPE-1	-	10	10	15	-
	.TPE-2	-	-	-	-	10
	.TPE-3	-	-	-	-	10
	.FR-1	18	-	18	18	18
10	.FR-2	-	26	-	-	-
	.AOC	7	6.1	7	7	7
	.T	0.25	0.25	0.25	0.25	0.25
	.O	0.45	0.45	0.45	0.45	0.45
15						
<u>Properties</u>						
	.Tensile strength at flow (MPa)	51	47	42	36	40
20	.Elongation at fracture (%)	6	9	13	16	9
	.Bending strength (MPa)	84	80	71	61	65
25	.Bending modulus (MPa)	2520	1940	2020	1670	1980
	.Comparative trac- king Index (V)	250	250	over 600	over 600	over 600
	.Glow wire test	pass	pass	pass	pass	pass
30	.UL-94 (1.6 mm rod)	V-0	V-0	V-0	V-0	V-0
	.FLAME OUT TIME (sec.)	1	1	2	2	1

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CLAIMS:

1. A polymer mixture which comprises the following constituents:

A. 50 to 80 parts by weight of a polybutylene terephthalate;

5 B. 5 to 20 parts by weight of a thermoplastic elastomer selected from the group consisting of: a polyetherester block copolymer, a polyester ester urethane and a polyether imide ester; and

C. 15 to 30 parts by weight of a brominated polystyrene as a flame-retardant.

10 2. A polymer mixture as claimed in Claim 1, characterised in that the polymer mixture comprises as constituent A. a polymer or a copolymer which is built up for more than 70 mol% from units derived from terephthalic acid and butane-1, 4-diol.

15 3. A polymer mixture as claimed in Claim 1, characterised in that the polymer mixture comprises a metal compound which cooperates synergistically with the brominated polystyrene.

20 4. A polymer mixture as claimed in Claim 1, characterised in that in addition to the constituents mentioned hereinbefore sub A, B, and C, the polymer mixture comprises 1-100 parts by weight of conventionally used additives per 100 parts by weight of A plus B plus C.

25 5. Articles formed from the polymer mixture as claimed in Claims 1 to 3.

6. Articles formed from the polymer mixture as claimed in claim 4.