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Field of the invention

The invention relates to multi-layered moulded plastics bodies consisting largely of aromatic polycarbonate and having a finish of a polymethacrylate plastics layer containing UV-absorbers as a protection against the effect of UV-radiation.

Prior art

Polycarbonate plastics only have a limited weather resistance. They are damaged in particular by UV-radiation which is demonstrated by the original shiny surfaces becoming dull. This dullness is the visible sign of the molecular disintegration due to UV-radiation by forming a plurality of very fine capillary fissures beginning on the plastics surface. UV-absorbers incorporated into the plastics are able to protect the inside of a plastics body such as a plastics board made of polycarbonate but not the surface thereof. As a protection, a layer of varnish made of polymethyl methacrylate which also contains an UV-absorber is prepared from a solution of the components of this protective layer, according to German patent application No. 1 694 273 (= US 3 582 398).

US 4 666 779 also discloses UV-stabilised articles consisting of thermoplastics substrates, in particular those consisting of polycarbonates which have a protective layer of a copolymer which largely consists of methylmethacrylate units and of 0.5 to 10% of the number of monomer units of other methacrylate units, i.e. ethyl methacrylate, n-butyl methacrylate, i-butylmethacrylate or mixtures thereof and of 0 to 5% of a number of monomer units of other α , β -unsaturated monomer units. The protective layer of polymethacrylate

is applied as a solution on to the substrate, the low-molecular UV-stabilisers required for the UV-stabilisation being incorporated together with the polymer-solvent system.

In addition, plastics bodies are known from US 4 576 870 and EP-A 0 203 487 in which the UV-absorbent protective layer is a copolymer consisting largely of (meth)acrylate monomers and monomers containing UV-protecting groups. These protective layers, are, in general, more brittle than the low molecular protective layers containing UV-absorbers as described above.

According to DE-A 2 832 676, a polymethylmethacrylate layer containing sufficient UV-absorber is applied when a polycarbonate plastics board is produced by coextrusion. According to DE-A 3 244 953 (=EP-A 110 238) an improved laminated body is obtained, in the multi component extrusion process, by applying to the plastics layer containing UV-absorbers, which may be polymethylmethacrylate or also polycarbonate, another covering containing substantially few UV-absorbers, which consist of polymethylmethacrylate or (meth)acrylate copolymers.

In all the cases of the prior art, the polymethacrylate plastics, in particular polymethylmethacrylate or also copolymers of methylmethacrylate and lesser amounts of C_2 - to C_{10} -alkyl- or aryl(meth)acrylates, are preferably used as a top layer containing UV-protectants, since they are highly weather-resistant and their surfaces are less damaged by UV-radiation than those of polycarbonates. EP-B 110 221 discloses polycarbonate top layers containing UV-absorbers.

The known polymethacrylate top layers containing UV-absorbers, used for polycarbonate plastics, have the disadvantage that their adherence to the polycarbonate to be covered is insufficient and that their viscosities are lower. This deficiency in the properties present is

a disadvantage to the entire plastics composite, in particular in the mechanical processing of the multi-layered moulded plastics bodies as for example in the bending thereof.

Object and solution

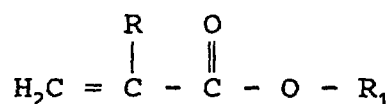
The invention aims to provide multi-layered plastics bodies which largely consist of polycarbonate, the layer of polymethacrylate containing UV-absorbers directly applied to the polycarbonate plastics, which permanently adheres to the polycarbonate-containing plastics body to be protected, thus cancelling out the deterioration of its mechanical properties such as in particular deteriorations of bending strength or breaking resistance, hitherto caused by the polymethylmethacrylate layer.

Surprisingly, the above disadvantages of the top layers known from the prior art are avoided while fully maintaining the protective effect, if the multi-layered plastics bodies on a polycarbonate basis are synthesised such that a polymethacrylate layer containing UV-absorber is applied to the polycarbonate core to be protected, said polymethacrylate layer being an impact strength-modified polymethacrylate plastics, one phase of which consisting of methacrylate copolymers which, per se, just as the impact strength-modified polymethacrylate plastics, together with polycarbonate, in particular polycarbonate of bisphenol A, form compatible mixtures.

The invention is a multi-layered plastics body having a core layer consisting of more than 50 wt.-% of aromatic polycarbonate and, applied thereto, a thermoplastic, impact strength-modified polymethacrylate plastics layer containing UV-absorbers, which may also be coated with additional layers, characterised in that the polymethacrylate plastics layer is a two-phase mixed

polymer which contains UV-absorbers or UV-absorbing groups in amounts of from 0.01 to 50 wt.-%, and the two-phase mixed polymer consists of

- (A) 10 to 90 wt.-% of optionally cross-linked polymers P having a glass transition temperature $T_g \leq 10^\circ\text{C}$, preferably $T_g \leq -10^\circ\text{C}$, in which 0 to 30 wt.-%, based on P, of a polymerisable UV-absorber may be incorporated, and
- (B) 90 to 10 wt.-% of a methylmethacrylate copolymer having a glass temperature $T_g \geq 30^\circ\text{C}$ which is at least partially covalently linked, synthesised from
 - (b1) 99.9 to 5 wt.-%, more particularly 95 to 20 wt.-% of methylmethacrylate units and optionally other α, β -unsaturated monomer units in amounts from 0 to 40 wt.-%, and
 - (b2) 0 to 50 wt.-% of a polymerisable UV-absorber, and
 - (b3) 0.1 to 95 wt.-%, more particularly 5 to 80 wt.-% of acrylate and/or methacrylate units having carbocyclic groups in the ester radical of formula I



I, wherein

R represents H or CH_3 and

R_1 represents Y or A-Y, whilst

Y is a cycloalkyl or a monoalkyl- or polyalkyl-

substituted C_{5-12} -cycloalkyl group, or an optionally

alkyl- or oxyalkyl-substituted C_{6-12} -aryl group and A

is a C_{1-6} -alkylene group, which may also be

branched, or a C_{2-4} -oxyalkylene group, whilst the copolymer (B) per se has a molecular weight $M_w \geq 30,000$ and any desired mixture of aromatic polycarbonate (PC) and the copolymer (B) is compatible and this mixture has a lower critical solution temperature (LCST) of $\geq 120^\circ C$.

The invention also relates to a process for preparing a multi-layered plastics body having a core layer consisting of more than 50 wt.-% of aromatic polycarbonate and, applied thereto, a thermoplastic polymethacrylate plastics layer containing UV-absorbers, which may also be coated with additional layers, characterised in that the thermoplastic polymethacrylate plastics layer containing UV-absorbers is an impact strength-modified polymethacrylate copolymer which is compatible with the aromatic polycarbonate and is applied to the core layer by coextrusion or painting on layer thicknesses of from 1 to 500 μm .

The additives providing UV-protection may be added in the form of known, low molecular, soluble UV-absorbing compounds, to the two-phase copolymer (A)-(B) before said copolymer is applied to the polycarbonate plastics, or else polymerisable compounds containing UV-absorbing structures in the molecule i.e. the known, so-called polymerisable UV-absorbers are homopolymerised when preparing (A) and/or (B). The polymethacrylate plastics layer may also contain the two UV-absorber species as UV-protection (c.f. R. Gächter & H. Müller, Taschenbuch der Kunststoff-Additive, p. 96 - 111, Carl Hanser Verlag 1979).

The polymethacrylate layer of the invention which is UV-absorbing, impact-resistant and adheres well on the polycarbonate plastics is applied, according to the invention, to the plastics material to be protected consisting predominantly of aromatic polycarbonate, in particular the polycarbonate of bisphenol A, using known

methods such as coextrusion and coating. If necessary, the polymethacrylate layer may be coated with another top layer such as a weather-resistant polymethylmethacrylate layer whereby this layer may be applied simultaneously with the other layers during the coextrusion or it may be applied by subsequent coating such as e.g. a scratch-proof coating on the basis of silicone resins or cross-linked polyacrylates.

The methacrylate copolymers (B) may be processed per se thermoplastically to form glass-clear, colourless substances. The unpublished German patent application P 37 19 239.6 largely discloses those methacrylate copolymers (B) of (b1) and (b3) which are compatible with aromatic polycarbonate.

Examples of such acrylate and methacrylate comonomers of Formula 1, i.e. having carbocyclic groups in the ester radical, include cyclopentyl methacrylate, cyclohexyl acrylate, cyclohexyl methacrylate, 3, 3, 5-trimethyl cyclohexyl methacrylate, 4-t-butyl cyclohexyl acrylate, 4-t-butyl cyclohexyl methacrylate, 3-cyclohexyl propyl methacrylate, phenyl methacrylate, 4-t-butyl phenyl methacrylate, 4-methoxy phenyl methacrylate, benzylmethacrylate, 1-phenyl ethyl methacrylate, 2-phenyl ethylacrylate, 2-phenyl ethyl methacrylate, 3-phenyl propyl acrylate, 3-phenyl propyl methacrylate, 2-phenoxy ethyl methacrylate, 2-naphthylmethacrylate.

Acryl- or methacrylamides which are comonomers rendering polymethacrylate resin compatible, according to DE-A 36 32 946 (=US 4 749 749), may also be homopolymerised in (B). Such monomers include for example N-cyclohexyl acrylamide, N-cyclohexyl methacrylamide, N-2-methyl cyclohexyl methacrylamide, N-phenyl methacrylamide, N-benzyl methacrylamide.

The methacrylate copolymers disclosed in the above patent specifications produce, together with aromatic polycarbonate, in particular polycarbonate of bisphenol

A, the completely compatible polymer mixtures disclosed in that document which are noted for high transparency.

N-carbocyclicly substituted maleinimide comonomers, such as N-cyclohexyl maleinimide, N-2- and 4-methyl cyclohexyl maleinimide, N-3,3,5-trimethyl cyclohexyl maleinimide, N-4-ethyl cyclohexylmaleinimide, N-2-phenyl cyclohexyl maleinimide, render methylmethacrylate polymers compatible with aromatic polycarbonates, according to German patent application DE 37 09 562 (published on 6.10.1988)

Implementation of the invention

1. Polycarbonate (PC) Plastics.

The plastics body according to the invention consists largely of aromatic carbonate to be protected from ultraviolet light. Polycarbonates of bisphenol A, in particular, are used as aromatic polycarbonates in technical applications. Polycarbonates of bisphenol A are glass-clear, viscous plastics and commercially available e.g. under the trademark of ^RMakrolon. The molecular weights M_w , in g per mol, which may be determined e.g. by means of gel permeation chromatography or light scattering, of the carbonate polymers from which these plastics are derived are within the range of approximately 20,000 to 60,000, in particular 20,000 to 40,000, and the Vicat softening temperatures, VST, of these polymers measured according to DIN 53460 are 140°C to 160°C.

The PC-plastics may be used for producing pre-fabricated plastics bodies according to the invention, e.g. in the form of solid boards, cavity boards, in the form of hoods or virtually any imaginable shape, the layer providing protection then being applied, preferably, by coating. When preparing moulded parts, according to the invention, by coextrusion, the

polycarbonates used and the methacrylate layer of (A) and (B) applied for the protection thereof may be present in the form and quality usually suitable for extrusion, e.g. as granules or powder.

The PC plastics used for preparing the plastics bodies according to the invention may preferably also be compatible mixtures which consist predominantly, i.e. in amounts of 99.9 to 80 wt.%, of aromatic polycarbonate, in particular polycarbonate of bisphenol A, and of the alloying components thereof i.e. in amounts of 0.1 to 20 wt.%, said components consisting of one or more methacrylate copolymers (B) and/or the two-phase copolymer of (A) and (B) which are completely compatible with the aromatic polycarbonate as already mentioned.

2. Two-phase copolymer of (A) and (B)

Because of its structure and the UV-absorbers contained in the copolymer layer, the two-phase copolymer of (A) and (B) ensures a permanent UV-protection of the polycarbonate plastics, even during greater mechanical stress or thereafter. It is an impact-resistant, transparent material which is compatible with polycarbonate plastics (PC), as largely disclosed by the unpublished German patent application P 37 43 199.4 and P 38 03 405.0.

In order to prepare the impact-resistant UV-protective layer, according to the invention, polymers P having a glass transition temperature $T_g \leq 10^\circ\text{C}$, preferably $\leq -10^\circ\text{C}$ are used as viscous phase (A). These are polymers which are normally referred to as Caoutchouc. The polymers of component (A) are, optionally, cross-linked. Their glass transition temperature T_g may be determined according to "Thermal Characterization of Polymeric Materials", p. 169 ff., Academic Press, New York 1981.

Polymers P for the viscous phase (A) are,

preferably, those from the group consisting of polyolefins, polydienes, ethylene-vinylacetate-copolymers, polyacrylates, polysiloxane, (cf. Ullmann's Encyclopädie der technischen Chemie, 4th ed, vol. 13, p. 595 to 635, Verlag Chemie 1977). The literature reference mentioned lists the methods of preparing these Caoutchoucs, their properties such as solubilities and viscosities, as well as their molecular weights M_w .

Preferred viscous phases (A) are terpolymers consisting of ethylene and propylene and a diene, the so-called EPDM-types, and acrylate caoutchoucs, in particular on the basis of butyl acrylate, are particularly preferred. These polyacrylates, are, preferably, prepared by polymerisation in aqueous emulsion, particularly also in suspension using conventionally known methods (Ullmann's Encyclopädie der technischen Chemie, 4th ed. vol. 19. p. 125 to 137, Verlag Chemie 1980).

Monomers of formula I to be incorporated into polymer components are known in the preparation of methyl methacrylate copolymer (B) combined with (A). In addition to other essential comonomers for methyl methacrylate, they are disclosed, for this purpose, in German patent application P 37 19 239.6 mentioned above. R represents CH_3 in a preferred embodiment, i.e. the monomers I are preferably synthesised from methacrylic acid. Examples of I include, in particular phenyl methacrylate, benzyl methacrylate, phenyl ethyl methacrylate, phenyl ethyl acrylate, cyclohexyl methacrylate.

The two-phase copolymer (A)-(B) consists, in amounts of 10 to 90 wt.%, preferably in amounts of 10 to 60 wt.%, and more particularly in amounts of 20 to 50 wt.%, of polymers P and, in amounts of 90 to 10 wt.%, preferably in amounts of 90 to 40 wt.% and more particularly in amounts of 80 to 50 wt.%, of the copolymer (B) which is, at least partially, covalently

bonded to the polymers (P), forming the polymer phase (A).

The copolymer (B) is synthesised from 99.9 to 5 wt.%, particularly 95 to 20 wt.%, more particularly 80 to 40 wt.% of methyl methacrylate units, and 0.1 to 95 wt.%, particularly 5 to 80 wt.%, more particularly 20 to 50 wt.% of acrylic and/or methacrylic monomers of formula I. It may contain, in amounts of 0 to 40 wt.%, in particular in amounts of 0.5 to 20 wt.%, further α , β - unsaturated units such as styrene, α -methylstyrene, acrylic acid, methacrylic acid, C_1 - C_6 esters of acrylic acid such as methylacrylate and further esters (C_2 - C_6 esters) of methacrylic acid such as ethyl methacrylate and the above-mentioned carbocyclicly substituted acrylamide and methacrylamide which were polymerised in. It is also possible and optionally advantageous to incorporate compounds with several polymerisable double-bonds, i.e. of cross-linking agents.

The methacrylate copolymers (B) per se are polymers having a glass transition temperature $T_g \geq 30^\circ\text{C}$ which may be processed thermoplastically to form colourless moulded bodies having Vicat softening temperatures VST of approximately 100 to 150°C , measured according to DIN 53460, and which form compatible mixtures with aromatic polycarbonate.

The preparation of the two-phase copolymers (A)-(B) for the impact strength-modified top layer may be carried out according to known processes.

In general, polymer P of phase (A) is introduced, for example in the form of a solution, to a solvent suitable for further processing e.g. a solvent which is suitable for the radical polymerisation of monomers according to (B) (H. Rauch-Puntigam, Th. Völker, "Acryl- und Methacrylverbindungen", p. 230 to 234, Springer-Verlag, Heidelberg 1967; J. Brandrup, E.H. Immergut, Polymer Handbook, III-144 bis III-148, John Wiley 1975). In so doing, the tendency of the solvent to transfer

reactions has also to be taken account of. Examples of solvents include esters such as butyl acetate, ethyl acetate, hydrocarbons such as toluene, ketones such as acetone. In general, solutions having a content of approximately 15 to 50 wt.% of polymer P are used to which the monomer and polymerisation adjuvants are added, for example by pouring. As a rule, the polymerisation is carried out at an elevated temperature e.g. within the range of 80 to 140°C. The initiators used are conventional ones such as peroxide or azo compounds (cf. H. Rauch-Puntigam, Th. Völker, Acryl- und Methacrylverbindungen loc.cit.). In solvents which have a lower boiling point, dibenzoyl peroxide may for example be used, whereas in solvents having a higher boiling point, tert. butyl compounds such as tert. butyl peroctoate may suitably be used.

When grafting EPDM and using for example butyl acetate as a solvent, the polymerisation of monomers to form component (B) may be started at approximately 120°C and then finished at approximately 80°C, while adding further methyl methacrylate, cross-linking agents and initiators. The polymerisation may suitably be carried out in the presence of regulators, e.g. mercaptans.

Preferably, the two-phase copolymer (A)-(B) is prepared as an emulsion polymer in an aqueous medium whereby in particular polybutyl acrylate or a phase consisting substantially of polybutyl acrylate is prepared as phase (A) forming the core onto which one or more shells of (B) are applied by polymerisation, using known methods of emulsion polymerisation (loc.cit. Ullmann, vol. 13 and Ullmann, vol. 19). The viscous phase (A) may also initially be polymerised onto a hard primary particle e.g. a particle of polymethyl methacrylate. In this way, it is possible to prepare usable copolymers (A)-(B) which have more than two shells. The emulsion polymerisation of the individual shells is carried out successively at temperatures of

approximately 30 to 100°C, in the presence of water-soluble polymerisation initiators, such as potassium persulphate or Redox systems with e.g. metal ions of varying valency such as $\text{Fe}^{\text{II}}/\text{Fe}^{\text{III}}$. Dispersions containing approximately 30 to 60 wt.% of solids with particle sizes (radii) of the solids of 10 to 300 nm may be prepared. Dispersions whose particles have radii of 20 to 100 nm are preferred.

The two-or multi-phase copolymer is preferably obtained from the solution by precipitation, e.g. using methanol as precipitant. When preparing the polymer by means of emulsion polymerisation, it is obtained by freeze-coagulation or spray drying or preferably by squeezing the emulsion polymer by means of an extruder.

3. UV-absorbers

The UV-absorbers are present in the polymethacrylate layer, in amounts of 0.01 to 50 wt.%, in particular in amounts of 0.1 to 30 wt.% and more particularly in amounts of 1 to 20 wt.%. The UV-absorbers may, as low-molecular absorbers i.e. having molecular weights $M_w \leq 5,000$, be distributed, i.e. dissolved, homogenously in the polymethacrylate layer (A)-(B), and/or the polymer phases (A) and/or (B) contain UV-absorbing partial structures by incorporating copolymerisable compounds, the known polymerisable UV-absorbers as for example disclosed in US 45 76 870. Examples of polymerisable UV-absorbers include 2-(2'-hydroxyphenyl)-5-methacrylamido-benzotriazole or 2-hydroxy-4-methacryloxybenzophenone. (cf. also Houben-Weyl, 4th ed. vol. 15, p. 256 - 260, Verlag Chemie).

The polymerisable compounds may, however, also be advantageously used in the polymethacrylate layer (A)-(B), in their monomeric form, as low-molecular, soluble UV-absorbers. Other UV-absorbers with a molecular weight $M_w \leq 5,000$ are

2-hydroxy-4-n-octoxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-(2'-hydroxy-5'-methylphenyl)benzotriazole and other derivatives of 2-hydroxy-benzophenone or benzotriazole, besides 2,4-dihydroxy-benzoylfuran, phenyl salicate, resorcinol disalicate, resorcinol mono-and dibenzoate, benzyl benzoate, stilbene, β -methyl umbelliferon and the benzoate thereof. Numerous other UV-absorbers are known and commercially available. UV-absorbers of low volatility at the processing temperature, i.e. in particular those having as high a molecular weight as possible are particularly preferred. If possible, the UV-absorber should, in the chosen concentration be homogenously distributed in the polymethacrylate layer. A PC-containing core layer may be completely free of UV-absorbers or contain such absorbers in a clearly lower concentration than the top layer. A concentration of 0.5 wt.% is not exceeded.

Even those UV-absorbers which contain, in their molecular structure, several UV-absorbing partial structures of different chemical constitutions, are suitable as homogenously mixable UV-absorbers having molecular weights of $\leq 5,000$ and of a lower volatility. Such oligomers may be obtained according to known polycondensation reactions of multi-functional starting compounds or as a result of polymerisation reactions of known polymerisable UV-absorbers (see for example US 4 576 870), and also as a result of copolymerisation of other conventional comonomers. By providing suitable condensation and polymerisation conditions (e.g. starting quantity, amount of transmission regulator), molecular weights $\leq 5,000$ may be obtained.

The UV-absorber containing top layer (A)-(B) is applied, by means of coextrusion or painting, in layer thicknesses of from 1 to 500 μm , usually 1 to 100 μm , more particularly in layer thicknesses up to approximately 50 μm using conventionally known methods.

In the extrusion method, known multi-component slot nozzles are used. The extrusion is carried out at temperatures of between 240 and 300°C. When using the method of painting, solvents must be used in which the system (A)-(B) is soluble or highly swellable. For this purpose, 1-methoxypropanol-2 may, for example, be used.

The methacrylate copolymers (A)-(B) noted for their excellent adherence and viscosity which are used according to the invention as protective layers for polycarbonate plastics form, together with the polycarbonate of bisphenol A as disclosed in unpublished German application P 38 03 405.0, compatible mixtures according to the criteria of the "Transition Temperature Method" and the "Optical Method", as required for compatible polymer mixtures and described in the chapter "Compatible Polymers" in Polymer Handbook, Second Edition, Copyright © 1975, by John Wiley & Sons; III, 211. These compatible mixtures may also be characterised by their LCST behaviour (D.R. Paul, Polymer Blends and Mixtures, 1985, p. 1 - 3; Martinus Nijhoff Publishers, Dordrecht, Boston Lancaster; Kirk-Othmer, 3rd. Ed. Vol 18, pg. 443 - 478, J. Wiley 1982). They have a lower critical solution temperature (LCST) of $\geq 120^{\circ}\text{C}$, particularly $\geq 150^{\circ}\text{C}$. For this purpose the cloud point T_{lr} (turbidity point) is determined experimentally, for example on the Kofler heating bench (Chem. Ing.-Technik 1950, p. 289), which characterises the phase transition from the homogenous to the heterogenous state of the mixture, depending on the qualitative and quantitative composition of mixing.

Advantages of the invention

The multi-layered plastics bodies on polycarbonate basis, according to the invention, are used particularly outdoors e.g. as glazing and construction elements where they are subjected to sunlight and the prevailing

weather conditions. Weathering tests such as the Xenotest or the QUV-Test (DIN 53 387) and the hailstorm test demonstrate the obvious superiority of the new material over the comparison materials of the known prior art. The hailstorm test serves to examine the materials as to their hail-resistance, since great damage was suffered through hailstorms in the past because of the less hail-resistant materials used e.g. in the construction of glass houses. As a result of the invention, damages to the plastics material caused by UV-radiation are avoided thus also obviating any further damages due to destructive effects such as hailstorms.

The hailstorm test is the non-standardised test carried out by the company itself, in which e.g. boards are hit by polyamide balls of different sizes and different kinetic energies. The hailstorm energy values (HE) ascertained in the test indicate the energy in Joules [J], at which the protective layer/substrate compound breaks.

Further quantities to be measured for characterising the surface and thus serving as proof of the advantages of the invention include the determination of the surface roughness by means of the perthometer according to DIN 4768, the determination of the surface lustre, the reflectometer value according to DIN 67530, the value of yellowing according to DIN 6167 and the determination of the layer thickness of the UV-absorber-containing layer, e.g. according to US 4 477 521.

Examples 1 - 4

Preparation of the materials for the (meth)acrylate protective layers.

Example 1

Impact strength-modified (meth)acrylate-copolymer according to the invention, by emulsion polymerisation.

770 g of water (dist.), 0.5 g of acetic acid (100%), 0.04 g of iron (II) sulfate, 18 g of C15-paraffin sulfonate and 125 g of a monomer emulsion I are introduced to a reaction vessel.

The polymerisation is started at ambient temperature, together with 0.15 g of diisopropylbenzenemonohydroperoxide and 1.3 g sodium hydroxy methyl sulfinat.

When the peak temperature is reached, the remaining monomer emulsion I is added which consists, as a whole, of 730 g of water (dist.), 3,4 g of C15-paraffin sulfonate, 772 g of butyl acrylate and 23.5 g of allyl methacrylate and is initiated with 1.26 g of diisopropylbenzenemonohydroperoxide and 0.32 g sodium hydroxy methyl sulfinat.

At the end of the reaction, the dispersion obtained is heated to 90°C and the monomer emulsion II is added over a period of two hours, said monomer emulsion consisting of 2705 g of water (dist.), 4.2 g C15-paraffin sulfonate, 2.2 g of potassium peroxodisulphate, 709.8 g of methyl methacrylate, 668.4 g of phenyl methacrylate, 73.9 g of 3-(2-benzotriazolyl)-2-hydroxy-5-tert.-octylbenzyl methacrylamide, 29.6 g methylacrylate and 8.9 g of 2-ethylhexyl thioglycolate.

Characterisation of the dispersion

Particle diameter: = 98 nm

Solids content : = 35 wt.%

The solid matter is obtained by freeze coagulation, filtration and subsequent drying in the drying cupboard at 60°C.

Example 2 (Comparison example according to US 4 576 870)

10 parts by weight of 3-(2-benzotriazolyl)-2-hydroxy-5-tert.-octylbenzylmethacrylamide are dissolved in a mixture consisting of 80 parts by weight of methyl methacrylate (MMA) and 10 parts by weight of phenyl methacrylate while heating to 50°C and continuously stirring. After adding 0.05 parts by weight of tert.butyl perneodecanoate and 0.03 parts by weight of 2,2-bis-(t-butylperoxy)butane as initiators and 0.3 parts by weight of dodecylmercaptan as regulator, the mixture is polymerised in a polymerisation chamber for 7 hours at 50°C and for 17 hours at 40°C. For the final polymerisation, the polymer is tempered for 12 hours at 110°C. The average molecular weight is determined at $M_w = 160,000$ and the glass transition temperature is determined at $T_g = 110^\circ\text{C}$.

Example 3 (Comparison example)

A mixture consisting of 80 parts by weight of MMA and 20 parts by weight of ethyl acrylate is mixed with 0.05 parts by weight of 2,2'-azobisisobutyronitrile and 0.35 parts by weight of dodecylmercaptan and polymerised in a tubular sheeting for 12 hours at 50°C. For the final polymerisation, the polymer is tempered for 12 hours at 110°C. The average molecular weight is determined at $M_w = 150,000$ and the glass transition temperature at $T_g = 77^\circ\text{C}$.

Example 4 (Comparison example)

The polymer matrix used is a polymethyl methacrylate

prepared as follows:

0.04 parts by weight of dilauroylperoxide and 0.62 parts by weight of 2-ethylhexylthioglycolate are added to a mixture of 94 parts by weight of MMA, 5.7 parts by weight of ethyl acrylate and 0.3 parts by weight of methacrylic acid. The mixture is polymerised in a tubular sheeting for 5 hours at 65°C and 27 hours at 55°C. For the final polymerisation, the polymer is tempered for 8 hours at 120°C. The molecular weight is determined at $M_w = 90,000$ and the glass transition temperature at $T_g = 95^\circ\text{C}$.

Examples 5 - 8

Preparation of the top layers

Example 5 (according to the invention)

The emulsion polymer prepared according to Example 1, is, firstly, extruded and granulated. In the subsequent coextrusion step, the granulate is applied, in an extrusion tool, onto a web double plate made of polycarbonate, by means of a multi-channel nozzle. The thickness of the coextrusion layer is 10 - 30 μm .

Example 6 (Comparison)

The polymer prepared according to Example 2 is ground and extruded and granulated in a second compounding step. The subsequent coextrusion is carried out analogous to Example 5.

Example 7 (Comparison)

The polymer prepared according to Example 3 is ground and extruded and granulated in a second compounding step. In a drum mixer, 10 parts by weight (based on the total amount) of 2-(2-hydroxy-3,5-

diisopropylphenyl)-benzotriazole are compounded into the granulate. The subsequent coextrusion steps and the thickness of the coextruded layer is implemented according to Example 5.

Example 8 (Comparison)

Coat of paint: The exact formulation is:

80 parts by weight of 1-methoxypropanol-2
14 parts by weight of polymethyl methacrylate of
Example 4
6 parts by weight of UV-absorber ®Cyasorb 531
(2-hydroxy-4-n-octoxybenzophenone)

The formulation of paint is applied, by means of a 15 μm doctor blade, onto the PC-web double plate (WDP), corresponding to a dry film thickness of approximately 3 μm .

Examples 9 - 12

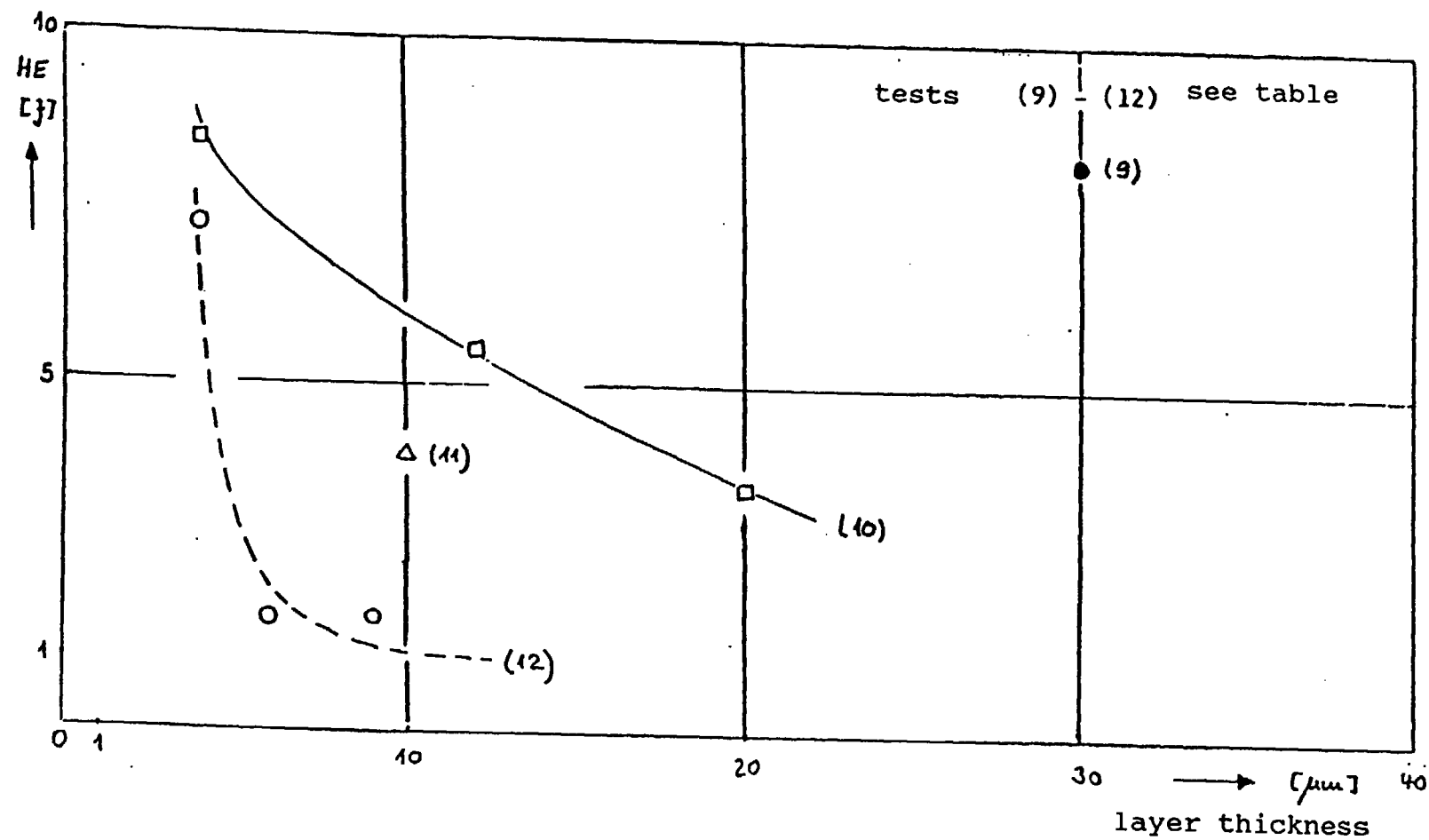
Weathering and fracture mechanics of the PC-WDP coated according to the Examples 5 - 8.

The following table lists the weathering data (weathering according to the Xenotest) and the hailstorm test data as a function thickness of the top layer (see also diagram 1) of (meth)acrylate-UV-protective layers.

Table
Properties of the PC-WDP described

Example	Coextrusion Layer/layer of paint according to	Weather-resistance in the Xenotest (+ unchanged) (h)	Storm energy (HE) in (J) at breakage as a function of the layer thickness d (μm) / HE (J)	
9	Example 5	1000 +	30 / 8.3	
10	Example 6 (Comparison)	1000 +	4 / 8.5 12 / 5.5 20 / 3.5	
11	Example 7 (Comparison)	5000 +	10 / 4.0	After 5 years of weathering outdoors
12	Example 8 (Comparison)	5000 +	4 / 7.3 6 / 1.6 9 / 1.6	

Diagram 1: Hailstorm resistance as a function of the layer thickness of coextruded or painted methacrylate - UV - protective layer on PC - WDP



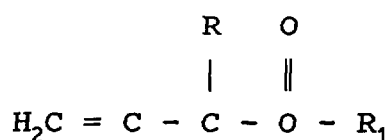
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Claims

1. A multi-layered plastics body having a core layer consisting of more than 50 wt.-% of aromatic polycarbonate and, applied thereto, a thermoplastic, impact strength-modified polymethacrylate plastics layer containing UV-absorbers, which may also be coated with additional layers, characterised in that the polymethacrylate plastics layer is a two-phase mixed polymer which contains UV-absorbers or UV-absorbing groups in amounts of from 0.01 to 50 wt.-%, and the two-phase mixed polymer consists of

- (A) 10 to 90 wt.-% of optionally cross-linked polymers P having a glass transition temperature $T_g \leq 10^\circ\text{C}$, preferably $T_g \leq -10^\circ\text{C}$, in which 0 to 30 wt.-%, based on P, of a polymerisable UV-absorber may be incorporated, and
- (B) 90 to 10 wt.-% of a methylmethacrylate copolymer having a glass temperature $T_g \geq 30^\circ\text{C}$ which is at least partially covalently linked, synthesised from
 - (b1) 99.9 to 5 wt.-%, more particularly 95 to 20 wt.-% of methylmethacrylate units and optionally other α, β -unsaturated monomer units in amounts from 0 to 40 wt.-%, and
 - (b2) 0 to 50 wt.-% of a polymerisable UV-absorber, and
 - (b3) 0.1 to 95 wt.-%, more particularly 5 to 80 wt.-% of acrylate and/or methacrylate units having carbocyclic groups in the ester radical of formula I



I, wherein

R represents H or CH₃ and
R₁ represents Y or A-Y, whilst
Y is a cycloalkyl or a monoalkyl- or polyalkyl-
substituted C₅₋₁₂-cycloalkyl group, or an optionally
alkyl- or oxyalkyl-substituted C₆₋₁₂-aryl group and A
is a C₁₋₆-alkylene group, which may also be
branched, or a C₂₋₄-oxyalkylene group,
whilst the copolymer (B) per se has a molecular
weight M_w ≥ 30,000 and any desired mixture of
aromatic polycarbonate (PC) and the copolymer (B)
is compatible and this mixture has a lower critical
solution temperature (LCST) of ≥ 120°C.

2. Multi-layered plastics body according to claim 1,
characterised in that the polycarbonate plastics core
layer is a compatible polymer mixture of 99.9 to
80 wt.-% of aromatic polycarbonate (PC) and 0.1 to
20 wt.-% of one or more methacrylate copolymers (B)
and/or (A)-(B).

3. Process for preparing a multi-layered plastics body
having a core layer consisting of more than 50 wt.-% of
aromatic polycarbonate and, applied thereto, a
thermoplastic polymethacrylate plastics layer containing
UV-absorbers, which may also be coated with additional
layers, characterised in that the thermoplastic
polymethacrylate plastics layer containing UV-absorbers
is an impact strength-modified polymethacrylate
copolymer which is compatible with the aromatic
polycarbonate and is applied to the core layer by
coextrusion in layer thicknesses of from 1 to 500 μm.



4. Process for preparing a multi-layered plastics body having a core layer consisting of more than 50 wt.-% of aromatic polycarbonate and, applied thereto, a thermoplastic polymethacrylate plastics layer containing UV-absorbers, which may also be coated with additional layers, characterised in that the thermoplastic polymethacrylate plastics layer containing UV-absorbers is an impact strength-modified polymethacrylate copolymer which is compatible with the aromatic polycarbonate and is applied to the core layer by painting on in layer thicknesses of from 1 to 500 μm .

5. Process for preparing a multi-layered plastics body according to claim 3 or 4, characterised in that a multi-layered plastics body according to claim 1 or 2 is thus obtained.

6. Process for preparing a multi-layered plastics body according to claims 3 to 5, characterised in that the polymethacrylate plastics layer is applied to the polycarbonate plastics in layer thicknesses of from 1 to 50 μm .