

Amorphous, transparent sheet of a crystallizable thermoplastic having high standard viscosity

- The invention relates to an amorphous, transparent sheet of a crystallizable thermoplastic having high standard viscosity whose thickness is in the range from 1 to 20 mm. The sheet is distinguished by very good optical and mechanical properties. The invention furthermore relates to a process for the production of this sheet, and to the use of the sheet.
- 10 Amorphous, transparent sheets having a thickness of from 1 to 20 mm are well known. These two-dimensional structures comprise amorphous, non-crystallizable thermoplastics. Typical examples of such thermoplastics which can be converted into sheets are, for example, 15 polyvinyl chloride (PVC), polycarbonate (PC) and polymethyl methacrylate (PMMA). These semifinished products are produced on so-called extrusion lines (cf. Polymer Werkstoffe, Volume II, Technologie 1, p. 136, Georg Thieme Verlag, Stuttgart, 1984). The pulverulent or 20 granular raw material is melted in an extruder. After extrusion, the amorphous thermoplastics can easily be shaped via polishing stacks or other shaping tools as a consequence of their viscosity, which continuously increases with decreasing temperature. After shaping, 25 amorphous thermoplastics then have adequate stability, i.e. a high viscosity, in order to be self-supporting in the calibration die. However, they are still sufficiently soft to be shaped by the die. The melt viscosity and inherent rigidity of amorphous thermoplastics is so high 30 in the calibration die that the semifinished product does not collapse in the calibration die before cooling. In the case of easily decomposed materials, for example PVC, special processing aids, for example processing stabilizers against decomposition and lubricants against 35 excessive internal friction and thus uncontrollable warming, are necessary during extrusion. External lubricants are necessary to prevent sticking to walls and

rolls.

The processing of PMMA is carried out using, for example, a vented extruder to enable removal of moisture.

5 The production of transparent sheets from amorphous thermoplastics sometimes requires expensive additives, which can migrate and can cause production problems owing to evaporations and surface coatings on the semifinished product. PVC sheets can be recycled only with difficulty or using special neutralization or electrolysis processes. 10 PC and PMMA sheets can likewise only be recycled with difficulty and only with loss or extreme impairment of the mechanical properties.

In addition to these disadvantages, PMMA sheets also have extremely poor impact strength and splinter on fracture 15 or mechanical loading. Furthermore, PMMA sheets are readily combustible, which means that they cannot be employed, for example, for internal applications and in exhibitions.

PMMA and PC sheets furthermore cannot be shaped when 20 cold; PMMA sheets disintegrate to form dangerous splinters, while PC sheets undergo hairline cracking and stress whitening.

EP-A-0 471 528 describes a process for shaping an article from a polyethylene terephthalate (PET) sheet. The 25 intrinsic viscosity of the PET employed is in the range from 0.5 to 1.2. The PET sheet is heat-treated on both sides in the thermofilming mold in a temperature range between the glass transition temperature and the melting point. The shaped PET sheet is removed from the mold when 30 the degree of crystallization of the shaped PET sheet is in the region of 25 to 50%. The PET sheets disclosed in EP-A-0 471 528 have a thickness of from 1 to 10 mm. Since the thermoformed molding produced from the PET sheet is partially crystalline and thus no longer transparent and

the surface properties of the molding are determined by the thermoforming process and by the thermoforming temperature and mold, it is unimportant what optical properties (for example gloss, haze and light
5 transmission) the PET sheets employed have. In general, the optical properties of these sheets are poor and in need of optimization.

US-A-3,496,143 describes the vacuum thermoforming of a 3 mm thick PET sheet whose degree of crystallization is
10 said to be in the range from 5 to 25%. However, the crystallinity of the thermoformed molding is greater than 25%. Again, no requirements regarding optical properties are made of these PET sheets. Since the crystallinity of the sheets employed is already between 5 and 25%, these
15 sheets are hazy and non-transparent.

The object of the present invention is to provide an amorphous, transparent sheet having a thickness of from 1 to 20 mm which has both good mechanical and optical properties.

20 The good optical properties include, for example, high light transmission, high surface gloss, extremely low haze and high clarity.

The good mechanical properties include, inter alia, high impact strength and high breaking strength.

25 In addition, the novel sheet should be recyclable, in particular without loss of the mechanical properties, and have low combustibility so that it can also be employed, for example, for indoor applications and in exhibitions.

This object is achieved by a transparent, amorphous sheet
30 having a thickness in the region of 1 to 20 mm which contains, as principal constituent, a crystallizable thermoplastic, wherein the crystallizable thermoplastic has a standard viscosity SV (DCA), measured in

dichloroacetic acid in accordance with DIN 53728, in the range from 1800 to 6000.

The standard viscosity SV (DCA) of the crystallizable thermoplastic, measured in dichloroacetic acid in accordance with DIN 53728, is preferably from 2000 to 5000, particularly preferably from 2500 to 4000.

The intrinsic viscosity IV (DCA) is calculated from the standard viscosity SV (DCA) as follows:

$$IV (DCA) = 6.67 \cdot 10^{-4} SV (DCA) + 0.118$$

The surface gloss, measured in accordance with DIN 67530 (measurement angle 20°), is greater than 120, preferably greater than 130, the light transmission, measured in accordance with ASTM D 1003, is greater than 84%, preferably greater than 86%, and the haze of the sheet, measured in accordance with ASTM D 1003, is less than 15%, preferably less than 11%.

The transparent, amorphous sheet contains, as principal constituent, a crystallizable thermoplastic. Suitable crystallizable or partially crystalline thermoplastics are, for example, polyethylene terephthalate, polybutylene terephthalate, cycloolefin polymers and cycloolefin copolymers, preference being given to polyethylene terephthalate.

For the purposes of the invention, the term crystallizable thermoplastics is taken to mean

- crystallizable homopolymers,
- crystallizable copolymers,
- crystallizable compounds,
- crystallizable recycle and
- other variations of crystallizable thermoplastics.

For the purposes of the present invention, the term amorphous sheet is taken to mean a sheet which is non-

crystalline, although the crystallizable thermoplastic employed preferably has a crystallinity of from 5 to 65%, particularly preferably from 25 to 65%. Non-crystalline or essentially amorphous means that the degree of crystallinity is generally less than 5%, preferably less than 2%, particularly preferably 0%.

An amorphous sheet of this type is essentially unoriented.

Processes for the preparation of the crystallizable thermoplastics are known to the person skilled in the art.

For example, polyethylene terephthalates are usually prepared by polycondensation in the melt or by a two-step polycondensation in which the first step is carried out to a mean molecular weight - corresponding to a mean intrinsic viscosity IV of from about 0.5 to 0.7 - in the melt and the further condensation is carried out by solid-state condensation. The polycondensation is generally carried out in the presence of known polycondensation catalysts or catalyst systems. In the solid-state condensation, PET chips are warmed at temperatures in the range of from 180 to 320°C under reduced pressure or under a protective gas until the desired molecular weight has been reached.

The preparation of polyethylene terephthalate is described in detail in a large number of patents, such as, for example in JP-A-60-139 717, DE-C-2 429 087, DE-A-27 07 491, DE-A-23 19 089, DE-A-16 94 461, JP-63-41 528, JP-62-39 621, DE-A-41 17 825, DE-A-42 26 737, JP-60-141 715, DE-A-27 21 501 and US-A-5,296,586.

Polyethylene terephthalates having particularly high molecular weights can be prepared by polycondensation of dicarboxylic acid/diol precondensates (oligomers) at

elevated temperature in a liquid heat-transfer medium in the presence of conventional polycondensation catalysts and, if desired, cocondensable modifiers if the liquid heat-transfer medium is inert and free from aromatic moieties and has a boiling point in the range from 200 to 320°C, the weight ratio between dicarboxylic acid/diol/precondensate (oligomer) employed and liquid heat-transfer medium is in the range from 20:80 to 80:20, and the polycondensation is carried out in the boiling reaction mixture in the presence of a dispersion stabilizer.

In the case of polyethylene terephthalate, measurement of the Charpy impact strength a_n (measured in accordance with ISO 179/1D) of the sheet is preferably not accompanied by fracture. In addition, the Izod notched impact strength a_k (measured in accordance with ISO 180/1A) of the sheet is preferably in the range from 2.0 to 8.0 kJ/m², particularly preferably in the range from 4.0 to 6.0 kJ/m².

The clarity of the sheet measured at an angle of less than 2.5° (ASTM D 1003) is preferably greater than 96%, particularly preferably greater than 97%.

Polyethylene terephthalate polymers having a crystalline melting point T_m , as measured by DSC (differential scanning calorimetry) at a heating rate of 10°C/min, of from 220° to 280°C, in particular from 220°C to 260°C, preferably from 230°C to 250°C, a crystallization temperature range T_c from 75°C to 280°C, preferably from 75°C to 260°C, a glass transition temperature T_g of from 65°C to 90°C and a density, measured in accordance with DIN 53479, of from 1.30 to 1.45 g/cm³ and a crystallinity of from 5 to 65%, are preferred polymers as starting materials for production of the novel sheet.

The bulk density, measured in accordance with DIN 53466, is preferably from 0.75 to 1.0 kg/dm³, particularly

preferably from 0.80 to 0.90 kg/dm³.

The polydispersity of the polyethylene terephthalate M_w/M_n , measured by GPC, is usually from 1.5 to 6.0, preferably from 2.5 to 6.0, particularly preferably from 3.0 to 5.0.

In a particularly preferred embodiment, the novel sheet is provided with a UV light stabilizer.

The concentration is preferably from 0.01 to 5% by weight based on the weight of the crystallizable thermoplastic.

Light, in particular the ultra-violet part of sunlight, i.e. having a wavelength in the range of 280 to 400 nm, initiates degradation processes in thermoplastics, resulting not only in a change in the visual appearance as a consequence of color change or yellowing, but also adversely affecting the mechanical-physical properties.

The inhibition of these photooxidative degradation processes is of considerable industrial and economic importance, since otherwise the potential uses of numerous thermoplastics are drastically limited.

Polyethylene terephthalates begin to absorb UV light, for example, even at below 360 nm, their absorption increases considerably below 320 nm and is very pronounced at below 300 nm. The maximum absorption is between 280 and 300 nm.

In the presence of oxygen, principally chain cleavage, but no crosslinking is observed. Carbon monoxide, carbon dioxide and carboxylic acids represent the predominant photooxidation products in terms of amount. Besides direct photolysis of the ester groups, consideration must also be given to oxidation reactions, which likewise result in the formation of carbon dioxide via peroxide free radicals.

The photooxidation of polyethylene terephthalates can also result, by elimination of hydrogen in the α -position of the ester groups, in hydroperoxides and decomposition products thereof, and associated chain cleavage (H. Day, 5 D. M. Wiles: J. Appl. Polym. Sci 16, 1972, page 203).

UV stabilizers and UV absorbers as light stabilizers are chemical compounds which can engage in the physical and chemical processes of photoinduced degradation. Carbon black and other pigments can provide partial protection 10 from light. However, the substances are unsuitable for transparent sheets since they result in discoloration or a color change. For transparent, amorphous sheets, the only suitable compounds are organic and organometallic compounds which impart only an extremely weak color or 15 color change, or none at all, in the thermoplastics to be stabilized.

Examples of suitable light stabilizers or UV stabilizers are 2-hydroxybenzophenones, 2-hydroxybenzotriazoles, organonickel compounds, salicylic esters, cinnamic ester 20 derivatives, resorcinol monobenzoates, oxanilides, hydroxybenzoic esters, sterically hindered amines and triazines, preference being given to 2-hydroxybenzotriazoles and triazines.

In a particularly preferred embodiment, the novel transparent, amorphous sheet contains, as principle constituents, a crystallizable polyethylene terephthalate and 25 from 0.01 to 5.0% by weight of 2-(4,6-diphenyl-1,3,5-triazin-2-yl)-5-(hexyl)oxyphenol (structure in Fig. 1a) or from 0.01 to 5.0% by weight of 2,2'-methylene-bis(6- 30 (2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)phenol (structure in Fig. 1b). In a preferred embodiment, it is also possible to employ mixtures of these two UV stabilizers or mixtures of at least one of these two UV stabilizers with other UV stabilizers, where the total 35 concentration of light stabilizers is preferably from 0.01 to 5.0% by weight, based on the weight of

crystallizable polyethylene terephthalate.

Weathering tests have shown that the novel UV-stabilized sheets exhibit no yellowing, no embrittlement, no loss of gloss on the surface, no cracking on the surface and no
5 impairment of the mechanical properties even after use outside for from 5 to 7 years.

In addition, entirely unexpectedly, good cold formability without fracture, hairline cracking and/or stress whitening was observed, meaning that the novel sheet can
10 be shaped and bent without the action of heat.

In addition, measurements showed that the novel sheet has low combustibility and low flammability, so that it is suitable, for example, for internal applications and in exhibitions.

15 Furthermore, the novel sheet can easily be recycled without environmental pollution and without loss of the mechanical properties, meaning that it is suitable, for example, for use as short-term advertising hoardings or other advertising articles.

20 In the UV-stabilized embodiment, the sheet has improved weathering resistance and increased UV stability. This means that the sheets are only damaged to an extremely small extent, or not at all, by weathering and sunlight or by other UV radiation, so that the sheets are suitable
25 for external applications and/or critical internal applications.

The novel transparent, amorphous sheet can be produced, for example, by extrusion in an extrusion line.

An extrusion line of this type is shown diagrammatically
30 in Fig. 2. It essentially comprises

- an extruder (1) as plastication unit,
- a sheet die (2) as shaping tool,

- a polishing stack/calender (3) as calibration die,
- a cooling bed (4) and/or a roller conveyor (5) for aftercooling,
- a take-off roll (6),
- 5 - a separating saw (7),
- an edge trimming device (9), and, if desired,
- a stacking unit (8).

The process comprises drying the crystallizable thermo-
plastic if necessary, then melting the dry polymer in the
10 extruder, if desired together with the UV stabilizer,
extruding the melt through a die, calibrating, smoothing
and cooling the sheet in the polishing stack and then
cutting the sheet to size.

The process for the production of the novel sheet is
15 described in detail below using the example of polyethy-
lene terephthalate.

The polyethylene terephthalate is preferably dried for
from 4 to 6 hours at from 160 to 180°C before extrusion.

The polyethylene terephthalate is then melted in the
20 extruder. The temperature of the PET melt is preferably
in the range from 250 to 320°C, it being possible for the
temperature of the melt to be adjusted essentially both
through the temperature of the extruder and through the
residence time of the melt in the extruder.

25 If a light stabilizer is used, this can already have been
added by the producer of the raw material or be metered
into the extruder during production of the sheet.

The light stabilizer is particularly preferably added via
masterbatch technology, where the light stabilizer is
30 fully dispersed in a solid carrier material. Suitable
carrier materials are certain resins, the crystallizable
thermoplastic itself, such as for example, polyethylene
terephthalate, or alternatively other polymers which are

sufficiently compatible with the crystallizable thermoplastic.

It is important that the particle size and bulk density of the masterbatch are similar to the particle size and bulk density of the crystallizable thermoplastic, so that a homogeneous distribution and thus homogeneous UV stabilization can take place.

The melt then leaves the extruder through a die. This die is preferably a sheet die.

10 The PET melted by the extruder and shaped by a sheet die is calibrated, i.e. intensively cooled and smoothed, by smoothing calender rolls. The calender rolls can be arranged, for example, in an I-, F-, L- or S-shape (see Fig. 3).

15 The PET material can then be cooled on a roller conveyor, cut to size in width, cut into appropriate lengths and finally stacked.

The thickness of the PET sheet is determined essentially by the take-off unit, which is arranged at the end of the cooling zone, the chill (smoothing) rolls, which are coupled to the take-off unit with respect to speed, and the conveying rate of the extruder on the one hand and the separation between the rolls on the other hand.

25 The extruders employed can be either single-screw or twin-screw extruders.

The sheet die preferably comprises a collapsible die body, the lips and the restrictor bar for flow regulation over the width. To this end, the restrictor bar can be bent by means of tension and pressure screws. The thickness is set by adjusting the lips. It is important to ensure a uniform temperature of the PET and of the lip, since otherwise the PET melt flows out through the

various flow channels in different thicknesses.

5 The calibration tool, ie. the smoothing calender, gives the PET melt shape and dimensions. This is achieved by freezing at below the glass transition temperature by cooling and smoothing. Shaping should not be carried out in this state, since otherwise surface defects would form in this cooled state. For this reason, the calender rolls are preferably driven jointly. The temperature of the calender rolls must be lower than the crystalline melting point in order to avoid sticking of the PET melt. The PET melt leaves the sheet die at a temperature of from 240 to 300°C.

15 The first smoothing/chill roll is at a temperature of from 50°C to 80°C, depending on the output rate and sheet thickness. The second, somewhat colder roll cools the second or other surface.

20 In order to obtain a uniform sheet having excellent surface properties, it is essential here that the temperature of the first smoothing/chill roll is in the range from 50 to 80°C.

25 While the calibration unit freezes the sheet surfaces as smoothly as possible and cools the profile until it is rigid in shape, the aftercooling device lowers the temperature of the sheet virtually to room temperature. The aftercooling can be carried out on a roller board. The take-off rate should be matched accurately to the calender roll speed in order to prevent defects and thickness variations.

30 As additional equipment, the extrusion line for the production of the sheets can also include a separating saw for cutting the sheet to length, a side trimmer, a stacking unit and a monitoring station. The side or edge trimmer is advantageous since the thickness in the edge region may under certain circumstances not be uniform. At

the monitoring station, the thickness and optical properties of the sheet are measured.

The surprising multiplicity of excellent properties makes the novel, transparent, amorphous sheet highly suitable for a multiplicity of different uses, for example for interior room panels, for exhibitions and exhibition articles, as displays, for signs, in the lighting sector, in shop fitting and shelf construction, as advertizing articles, as menu holders, as basketball backboards, as room dividers, as information panels and as brochure and newspaper stands.

In the UV-stabilized embodiment, the novel sheet is also suitable for external applications, such as, for example, for greenhouses, roofing systems, external cladding, covers, applications in the construction sector, illuminated advertising profiles, balcony cladding and roof exit doors.

The invention is described in greater detail below with reference to working examples without this representing a limitation.

The individual properties are measured in accordance with the following standards or by the following processes.

Measurement methods

Surface gloss:

The surface gloss is measured at a measurement angle of 20° in accordance with DIN 67530. The reflector value is measured as an optical parameter for the surface of a sheet. In accordance with the standards ASTM-D 523-78 and ISO 2813, an angle of incidence is set at 20°. A light beam hits the planar test surface at the set angle of incidence and is reflected or scattered thereby. The light beams hitting the photoelectronic receiver are indicated as a proportional electrical quantity. The

measured value is dimensionless and must be given together with the angle of incidence.

Light transmission:

The light transmission is taken to mean the ratio between
5 the total amount of transmitted light and the amount of incident light.

The light transmission is measured using a "Hazegard plus" instrument in accordance with ASTM D 1003.

Haze and clarity:

10 Haze is the percentage of transmitted light deflected from the incident ray bundle by an average of more than 2.5° . The clarity is measured at an angle of less than 2.5° .

The haze and clarity are measured using a "Hazegard plus"
15 instrument in accordance with ASTM D 1003.

Surface defects:

The surface defects are determined visually.

Charpy impact strength a_n :

This parameter is determined in accordance with
20 ISO 179/1D.

Izod notched impact strength a_k :

The Izod notched impact strength a_k is measured in accordance with ISO 180/1A.

Density:

25 The density is determined in accordance with DIN 53479.

SV (DCA) and IV (DCA):

The standard viscosity SV (DCA) is measured in accordance with DIN 53726 in dichloroacetic acid.

The intrinsic viscosity (IV) is calculated as follows

from the standard viscosity (SV):

$$IV \text{ (DCA)} = 6.67 \times 10^{-4} \text{ SV (DCA)} + 0.118.$$

Thermal properties:

- 5 The thermal properties, such as crystalline melting point T_m , crystallization temperature range T_c , after- or cold-crystallization temperature T_{CN} and glass transition temperature T_g are measured by differential scanning calorimetry (DSC) at a heating rate of 10°C/min.

Molecular weight and polydispersity:

- 10 The molecular weights M_w and M_n and the resultant polydispersity M_w/M_n are measured by gel permeation chromatography (GPC).

Weathering (on both sides), UV stability:

- 15 The UV stability is tested as follows in accordance with ISO 4892 test specification:

	Test instrument	: Atlas Ci 65 Weather-O-meter
	Test conditions	: ISO 4892, i.e. artificial weathering
	Exposure time	: 1000 hours, (per side)
20	Exposure	: 0.5 W/m ² , 340 nm
	Temperature	: 63°C
	Relative atmospheric humidity	: 50%
25	Xenon lamp	: inner and outer filters made of borosilicate
	Exposure cycles	: 102 minutes UV light, then 18 minutes UV light with water spraying of the samples, then a further 102 minutes UV light, etc.

- 30 Color change:

The color change of the samples after artificial weathering is measured in accordance with DIN 5033 using a spectrophotometer.

The following symbols are used:

ΔL : Difference in brightness

+ ΔL : The sample is brighter than the standard

- ΔL : The sample is darker than the standard

5 ΔA : Difference in the red-green region

+ ΔA : The sample is redder than the standard

- ΔA : The sample is greener than the standard

ΔB : Difference in the blue-yellow region

+ ΔB : The sample is yellower than the standard

10 - ΔB : The sample is bluer than the standard

ΔE : Overall color change

$$\Delta E = \sqrt{\Delta L^2 + \Delta A^2 + \Delta B^2}$$

The larger the numerical deviation from the standard, the greater the color difference.

15 Numerical values of ≤ 0.3 can be neglected and mean that there is no significant color change.

Yellow value:

The yellow value G is a deviation from colorlessness toward "yellow" and is measured in accordance with DIN
20 6167. Yellow values of ≤ 5 are invisible.

The examples and comparative examples below each relate to single-layer transparent sheets of various thicknesses produced on the extrusion line described.

25 All sheets were weathered in accordance with ISO 4892 test specification on both sides for 1000 hours per side using the Atlas Ci 65 Weather-O-meter and subsequently tested for their mechanical properties discoloration, surface defects, haze and gloss.

Example 1:

30 The polyethylene terephthalate from which the transparent sheet is produced has a standard viscosity SV (DCA) of

3490, which corresponds to an intrinsic viscosity IV (DCA) of 2.45 dl/g. The moisture content is < 0.2% and the density (DIN 53479) is 1.35 g/cm³. The crystallinity is 19%, and the crystalline melting point, according to
5 DSC measurements, is 243°C. The crystallization temperature range T_c is between 82°C and 243°C. The polydispersity M_w/M_n of the polyethylene terephthalate is 4.3, where M_w is 225070 g/mol and M_n is 52400 g/mol. The glass transition temperature is 82°C.

10 Before extrusion, the polyethylene terephthalate is dried in a drier for 5 hours at 170°C and then extruded in a single-screw extruder at an extrusion temperature of 292°C through a sheet die onto a smoothing calender whose rolls are arranged in an S-shape, and smoothed to give a
15 sheet with a thickness of 3 mm. The first calender roll has a temperature of 73°C and the subsequent rolls each have a temperature of 67°C. The take-off rate and the calender roll speed are 6.5 m/min.

Following aftercooling, the transparent PET sheet with a
20 thickness of 3 mm is trimmed at the edges using separating saws, cut to length and stacked.

The transparent PET sheet produced has the following property profile:

	- thickness	: 3 mm
25	- surface gloss, 1st side	: 215
	(measurement angle 20°) 2nd side	: 214
	- light transmission	: 94%
	- clarity	: 100%
	- haze	: 0.8%

- surface defects per m^2 : none
(fisheyes, orange peel, bubbles, etc)
- Charpy impact strength a_n : no fracture
- 5 - Izod notched impact strength a_k : 4.6 kJ/m^2
- cold formability : good, no defects
- crystallinity : 0%
- density : 1.33 g/cm^3

Example 2

10 A transparent sheet is produced analogously to Example 1 using a polyethylene terephthalate with the following properties:

- SV (DCA) : 2717
- IV (DCA) : 1.93 dl/g
- 15 - density : 1.38 g/cm^3
- crystallinity : 44%
- M_w : 175640 g/mol
- M_n : 245°C
- crystalline melting point T_m : 49580 g/mol
- 20 - crystallization temperature range T_c : $82^\circ\text{C to } 245^\circ\text{C}$
- polydispersity M_w/M_n : 3.54
- glass transition temperature : 82°C

25 The extrusion temperature is 280°C . The first calander roll has a temperature of 66°C and the subsequent rolls have a temperature of 60°C . The take-off rate and the calender roll speed are 2.9 m/min .

The transparent PET sheet produced has the following property profile:

- thickness : 6 mm
- surface gloss, 1st side : 192
- 5 (measurement angle 20°) 2nd side : 190
- light transmission : 92.1%
- clarity : 99.8%
- haze : 2.0%
- surface defects per m² : none
- 10 (fisheyes, orange peel, bubbles, etc)
- Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 4.8 kJ/m²
- cold formability : good, no defects
- 15 - crystallinity : 0%
- density : 1.33 g/cm³

Example 3:

A transparent sheet is produced analogously to Example 2. The extrusion temperature is 275°C. The first calender roll has a temperature of 57°C and the subsequent rolls have a temperature of 50°C. The take-off rate and the calender roll speed are 1.7 m/min.

The PET sheet produced has the following property profile:

- 25 - thickness : 10 mm
- surface gloss, 1st side : 173
- (measurement angle 20°) 2nd side : 171
- light transmission : 88.5%
- clarity : 99.4%

- haze : 3.2%
- surface defects per m² : none
(fisheyes, orange peel, bubbles,
etc)
- 5 - Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 5.0 kJ/m²
- cold formability : good, no defects
- crystallinity : 0%
- density : 1.33 g/cm³
- 10 Example 4:

A transparent sheet is produced analogously to Example 3
using a polyethylene terephthalate having the following
properties:
 - SV (DCA) : 3173
 - 15 - IV (DCA) : 2.23 dl/g
 - density : 1.34 g/cm³
 - crystallinity : 12%
 - crystalline melting point T_m : 240°C
 - crystallization temperature range
 - 20 T_c : 82°C to 240°C
 - polydispersity M_w/M_n : 3.66
 - glass transition temperature : 82°C
 - M_w : 204,660 g/mol
 - M_n : 55,952 g/mol
- 25 The extrusion temperature is 274°C. The first calender
roll has a temperature of 50°C and the subsequent rolls
have a temperature of 45°C. The take-off rate and the
calender roll speed are 1.2 m/min.

The PET sheet produced has the following property profile:

- thickness : 15 mm
- surface gloss, 1st side : 162
- 5 (measurement angle 20°) 2nd side : 159
- light transmission : 89.3%
- clarity : 98.9%
- haze : 5.8%
- surface defects per m² : none
- 10 (fisheyes, orange peel, bubbles, etc.)
- Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 5.1 kJ/m²
- cold formability : good, no defects
- 15 - crystallinity : 0%
- density : 1.33 g/cm³

Example 5:

A transparent sheet produced analogously to Example 2. 70% of polyethylene terephthalate from Example 2 are
20 blended with 30% of this polyethylene terephthalate after recycling.

The transparent PET sheet produced has the following property profile:

- thickness : 6 mm
- 25 - surface gloss, 1st side : 188
- (measurement angle 20°) 2nd side : 186
- light transmission : 92.2%
- clarity : 99.6%

- haze : 2.2%
- surface defects per m² : none
(fisheyes, orange peel, bubbles,
etc.)
- 5 - Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 4.7 kJ/m²
- cold formability : good, no defects
- crystallinity : 0%
- density : 1.33 g/cm³
- 10 Example 6:
A transparent, amorphous sheet with a thickness of 3 mm
which contains, as principal constituents, the
polyethylene terephthalate from Example 1 and 1.0% by
weight of the UV stabilizer 2-(4,6-diphenyl-1,3,5-
15 triazin-2-yl)-5-hexyl)oxyphenol (®Tinuvin 1577 from Ciba-
Geigy) is produced analogously to Example 1. Tinuvin 1577
has a melting point of 149°C and is thermally stable up
to about 330°C.
1.0% by weight of the UV stabilizer are incorporated into
20 the polyethylene terephthalate directly by the raw-
material producer.
The drying, extrusion and process parameters are selected
as in Example 1.

The transparent PET sheet produced has the following
25 property profile:

- thickness : 3 mm
- surface gloss, 1st side : 208
(measurement angle 20°) 2nd side : 205
- light transmission : 92%
30 - clarity : 100%

- haze : 1.0%
- surface defects per m² : none
(fisheyes, orange peel, bubbles, etc.)
- 5 - Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 4.6 kJ/m²
- cold formability : good, no defects
- crystallinity : 0%
- density : 1.33 g/cm³
- 10 After weathering for 1000 hours per side using the Atlas Ci 65 Weather-O-Meter, the PET sheet exhibits the following properties:
- thickness : 3 mm
- surface gloss, 1st side : 202
- 15 (measurement angle 20°) 2nd side : 200
- light transmission : 91.7%
- clarity : 100%
- haze : 1.2%
- overall discoloration ΔE : 0.22
- 20 - dark discoloration ΔL : -0.18
- red-green discoloration ΔA : -0.08
- blue-yellow discoloration ΔB : 0.10
- surface defects
(cracks, embrittlement) : none
- 25 - yellow value G : 4
- Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 4.1 kJ/m²

- cold formability : good

Example 7:

A transparent, amorphous sheet with a thickness of 3 mm
5 is produced analogously to Example 6. The UV stabilizer
2-(4,6-diphenyl-1,3,5-triazin-2-yl)-5-(hexyl)oxyphenol
([®]Tinuvin 1577) is metered-in in the form of a
masterbatch. The masterbatch consists of 5% by weight of
[®]Tinuvin 1577 as active component and 95% by weight of
10 the polyethylene terephthalate from Example 1.

Before the extrusion, 80% by weight of polyethylene
terephthalate from Example 1 are dried for 5 hours at
170°C together with 20% by weight of the masterbatch. The
extrusion and sheet production are carried out
15 analogously to Example 1.

The transparent, amorphous PET sheet produced has the
following property profile:

	- thickness	: 3 mm
	- surface gloss, 1st side	: 204
20	(measurement angle 20°) 2nd side	: 201
	- light transmission	: 91.8%
	- clarity	: 100%
	- haze	: 0.9%
	- surface defects	: none
25	(fisheyes, orange peel, bubbles, etc.)	
	- Charpy impact strength a_n	: no fracture
	- Izod notched impact strength a_k	: 4.0 kJ/m ²
	- cold formability	: good
30	- crystallinity	: 0%

- density : 1.33 g/cm³

After weathering for 1000 hours per side using the Atlas Ci 65 Weather-O-Meter, the PET sheet exhibits the following properties:

5	- thickness	: 3 mm
	- surface gloss, 1st side	: 200
	(measurement angle 20°) 2nd side	: 198
	- light transmission	: 91.7%
	- clarity	: 100%
10	- haze	: 1.0%
	- overall discoloration ΔE	: 0.24
	- dark discoloration ΔL	: -0.19
	- red-green discoloration ΔA	: -0.08
	- blue-yellow discoloration ΔB	: 0.12
15	- surface defects	
	(cracks, embrittlement)	: none
	- yellow value G	: 5
	- Charpy impact strength a_n	: no fracture
	- Izod notched impact strength a_k	: 4.0 kJ/m ²
20	- cold formability	: good

Example 8:

A transparent, amorphous sheet with a thickness of 6 mm containing, as principal constituent, the polyethylene terephthalate described in Example 2 and 0.6% by weight of the UV stabilizer 2,2'-methylenebis(6-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)phenol ([®]Tinuvin 360 from Ciba-Geigy), based on the weight of the polymer is produced analogously to Example 2. Tinuvin 360 has a melting point of 195°C and is thermally stable

up to 250°C.

As in Example 6, 0.6% by weight of the UV stabilizer are incorporated into the polyethylene terephthalate directly by the raw material producer.

- 5 The extrusion temperature is 280°C. The first calender roll has a temperature of 66°C and the subsequent rolls have a temperature of 60°C. The take-off rate and the calender roll speed are 2.9 m/min.

10 The transparent PET sheet produced has the following property profile:

- | | | | |
|----|--|---|------------------------|
| | - thickness | : | 6 mm |
| | - surface gloss, 1st side | : | 187 |
| | (measurement angle 20°) 2nd side | : | 185 |
| | - light transmission | : | 91.8% |
| 15 | - clarity | : | 99.6% |
| | - haze | : | 2.5% |
| | - surface defects per m ² | : | none |
| | (fisheyes, orange peel, bubbles, etc.) | | |
| 20 | - Charpy impact strength a _n | : | no fracture |
| | - Izod notched impact strength a _k | : | 4.8 kJ/m ² |
| | - cold formability | : | good, no defects |
| | - crystallinity | : | 0% |
| | - density | : | 1.33 g/cm ³ |
| 25 | After weathering for 1000 hours per side using the Atlas Ci 65 Weather-O-Meter, the PET sheet exhibits the following properties: | | |

	- thickness	: 6 mm
	- surface gloss, 1st side	: 182
	(measurement angle 20°) 2nd side	: 179
	- light transmission	: 90.9%
5	- clarity	: 99.5%
	- haze	: 2.7%
	- overall discoloration ΔE	: 0.56
	- dark discoloration ΔL	: -0.21
	- red-green discoloration ΔA	: -0.11
10	- blue-yellow discoloration ΔB	: +0.51
	- surface defects (cracks, embrittlement)	: none
	- yellow value G	: 6
	- Charpy impact strength a_n	: no fracture
15	- Izod notched impact strength a_k	: 4.6 kJ/m ²
	- cold formability	: good, no defects

Example 9:

A transparent, amorphous sheet is produced analogously to Example 8. The extrusion temperature is 275°C. The first
20 calender roll has a temperature of 57°C and the subsequent rolls have a temperature of 50°C. The take-off rate and the calender roll speed are 1.7 m/min. The sheet is stabilized as described in Example 3.

The transparent PET sheet produced has the following
25 property profile:

- thickness	: 10 mm
- surface gloss, 1st side	: 168

	(measurement angle 20°) 2nd side	: 167
	- light transmission	: 88.5%
	- clarity	: 99.2%
	- haze	: 3.95%
5	- surface defects per m ² (fisheyes, orange peel, bubbles, etc.)	: none
	- Charpy impact strength a _n	: no fracture
	- Izod notched impact strength a _k	: 5.1 kJ/m ²
10	- cold formability	: good, no defects
	- crystallinity	: 0%
	- density	: 1.33 g/cm ³
15	After weathering for 1000 hours per side using the Atlas Ci 65 Weather-O-Meter, the PET sheet exhibits the following properties:	
	- thickness	: 10 mm
	- surface gloss, 1st side	: 164
	(measurement angle 20°) 2nd side	: 162
	- light transmission	: 88.2%
20	- clarity	: 99.1%
	- haze	: 5.0%
	- overall discoloration ΔE	: 0.47
	- dark discoloration ΔL	: -0.18
	- red-green discoloration ΔA	: -0.09
25	- blue-yellow discoloration ΔB	: +0.42
	- surface defects (cracks, embrittlement)	: none

- yellow value G : 5
- Charpy impact strength a_n : no fracture
- Izod notched impact strength a_k : 4.5 kJ/m²
- cold formability : good, no defects

5 Comparative Example 1:

A transparent sheet is produced analogously to Example 1. The polyethylene terephthalate employed has a standard viscosity SV (DCA) of 760, which corresponds to an intrinsic viscosity IV (DCA) of 0.62 dl/g. The other
10 properties are identical with the properties of the polyethylene terephthalate from Example 1 within the limits of measurement accuracy. The process parameters and the temperature were selected as in Example 1. As a
15 consequence of the low viscosity, sheet production is impossible. The melt stability is inadequate, so that the melt collapses before cooling on the calender rolls.

Comparative Example 2:

A transparent sheet is produced analogously to Example 2, also using the polyethylene terephthalate from Example 2.
20 The first calender roll has a temperature of 98°C and the subsequent rolls each have a temperature of 92°C.

The sheet produced is extremely hazy. The light transmission, the clarity and the gloss are significantly reduced. The sheet exhibits surface defects and
25 structures. The optical properties are unacceptable for a transparent application.

The sheet produced has the following property profile:

- thickness : 6 mm
- surface gloss, 1st side : 95
- 30 (measurement angle 20°) 2nd side : 93

	- light transmission	:	74%
	- clarity	:	90%
	- haze	:	52%
	- surface defects per m ²	:	bubbles,
5	(fisheyes, orange peel, bubbles, etc.)		orange peel
	- Charpy impact strength a _n	:	no fracture
	- Izod notched impact strength a _k	:	5.0 kJ/m ²
	- cold formability	:	good
10	- crystallinity	:	approx. 8%
	- density	:	1.34 g/cm ³



Claims

1. A transparent, amorphous sheet having a thickness in the range from 1 to 20 mm which contains, as principal constituent, a crystallizable thermoplastic, wherein a crystallizable thermoplastic has a standard viscosity SV (DCA), measured in dichloroacetic acid in accordance with DIN 53728, in the range from 1800 to 6000, with the exception of a sheet containing a crystallizable thermoplastic having a standard viscosity SV (DCA) of 1800 as principal constituent and a UV stabilizer.
2. A sheet as claimed in claim 1, wherein the standard viscosity is in the range from 2000 to 5000.
3. A sheet as claimed in claim 1, wherein the standard viscosity is in the range from 2500 to 4000.
4. A sheet as claimed in claims 1 to 3, wherein the surface gloss, measured in accordance with DIN 67530 (measurement angle 20°), is greater than 120.
5. A sheet as claimed in any one of the preceding claims, wherein the light transmission, measured in accordance with ASTM D 1003 is greater than 84%.
6. A sheet as claimed in any one of the preceding claims, wherein the haze, measured in accordance with ASTM D 1003, is less than 15%.
7. A sheet as claimed in one of the preceding claims, wherein the crystallizable thermoplastic used is selected from polyethylene terephthalate, polybutylene terephthalate, a cycloolefin polymer and a cycloolefin copolymer.
8. A sheet as claimed in claim 7, wherein the crystallizable thermoplastic used is polyethylene tere-



phthalate.

9. A sheet as claimed in claim 8, wherein the polyethylene terephthalate contains recycled polyethylene terephthalate.
- 5 10. A sheet as claimed in claim 8 or 9, wherein the measurement of the Charpy impact strength a_n , measured in accordance with ISO 179/1D, is not accompanied by a fracture.
- 10 11. A sheet as claimed in any one of claims 8 to 10, wherein the Izod notched impact strength a_k , measured in accordance with ISO 180/1A, is in the range from 2.0 to 8.0 kJ/m².
- 15 12. A sheet as claimed in any one of claims 8 to 11, wherein the clarity, measured in accordance with ASTM D 1003 at an angle of less than 2.5°, is greater than 96%.
- 20 13. A sheet as claimed in any one of claims 8 to 12, wherein the polyethylene terephthalate has a crystalline melting point, measured by DSC at a heating rate of 10°C/min, in the range from 220° to 280°C.
- 25 14. A sheet as claimed in any one of claims 8 to 13, wherein the polyethylene terephthalate has a crystallization temperature, measured by DSC at a heating rate of 10°C/min, in the range from 75° to 280°C.
15. A sheet as claimed in at least one of claims 8 to 14, wherein the polyethylene terephthalate employed has a crystallinity in the range from 5 to 65%.
- 30 16. A sheet as claimed in any one of the preceding claims, which contains at least one UV light stabilizer.



17. A sheet as claimed in claim 16, wherein the concentration of the UV stabilizer is in the range from 0.01 to 5% by weight, based on the weight of the crystallizable thermoplastic.
- 5 18. A sheet as claimed in claim 16 or 17, wherein the UV stabilizer is selected from 2-hydroxybenzotriazoles and triazines.
- 10 19. A process for the production of a transparent, amorphous sheet as claimed in any one of claims 1 to 18, which comprises melting the crystallizable thermoplastic in the extruder, extruding the melt through a die, calibrating, smoothing and cooling the extrudate in a polishing stack with at least two rolls, and cutting of the sheet to size, where the first roll in the polishing stack has a temperature in the range from 50 to 80°C.
- 15 20. A process as claimed in claim 19, wherein the crystallizable thermoplastic is melted in the extruder together with the UV stabilizer.
- 20 21. The process as claimed in claim 19 or 20, wherein the crystallizable thermoplastic is dried before melting in the extruder.
- 25 22. The process as claimed in claim 19 to 21, wherein the crystallizable thermoplastic is polyethylene terephthalate (PET).
23. The process as claimed in claim 22, wherein the polyethylene terephthalate is dried at from 160 to 180°C for from 4 to 6 hours before extrusion.
- 30 24. The process as claimed in claim 22 or 23, where the temperature of the PET melt is in the range from 250 to 320°C.

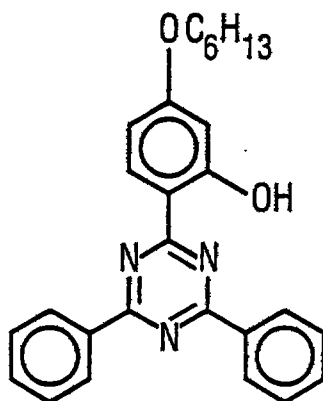


25. The process as claimed in any one of claims 20 to 24, wherein the UV stabilizer is added by master-batch technology.
26. The use of a transparent, amorphous sheet as claimed in any one of Claims 1 to 18 for internal and external applications.
- 5

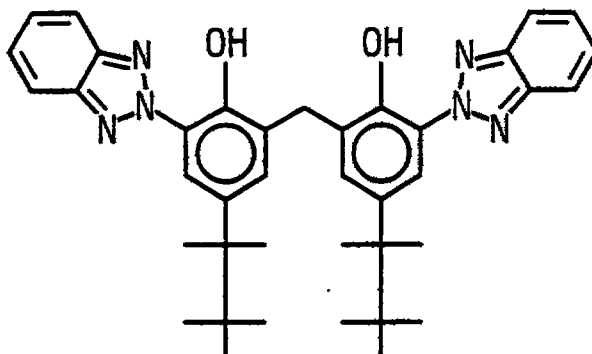
Abstract of the disclosure

Amorphous, transparent sheet of a crystallizable thermoplastic having high standard viscosity

The invention relates to a transparent, amorphous sheet having a thickness in the range from 1 to 20 mm which contains, as principal constituent, a crystallizable thermoplastic, wherein the crystallizable thermoplastic has a standard viscosity SV (DCA) in the range from 1800 to 6000, to a process for the production of the sheet, and to the use of the sheet.

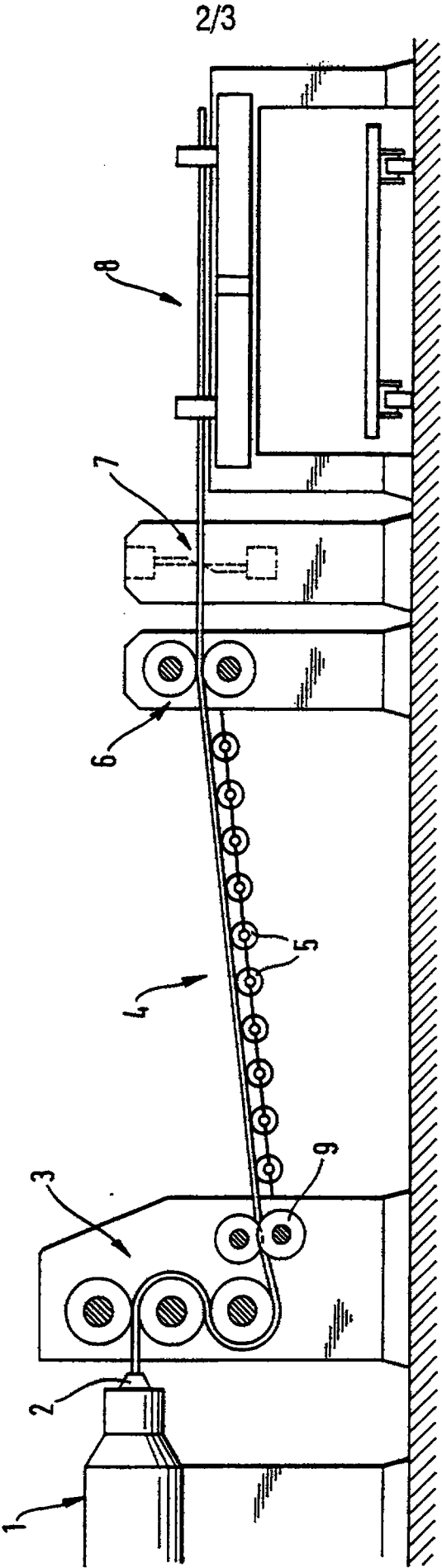
Fig. 1a

2-(4,6-Diphenyl-1,3,5-triazin-2-yl)-5-(hexyl)oxyphenol

Fig. 1b

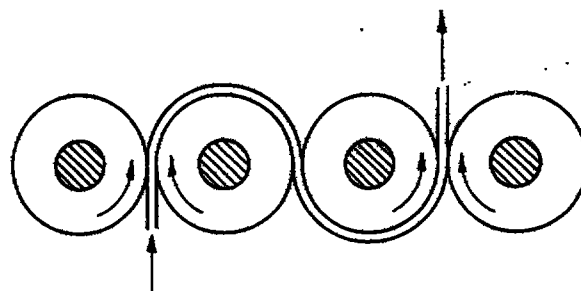
2,2'-Methylene-bis(6-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)-phenol

Fig. 2



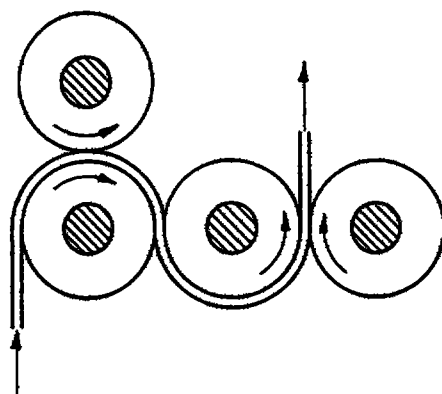
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Fig. 3a



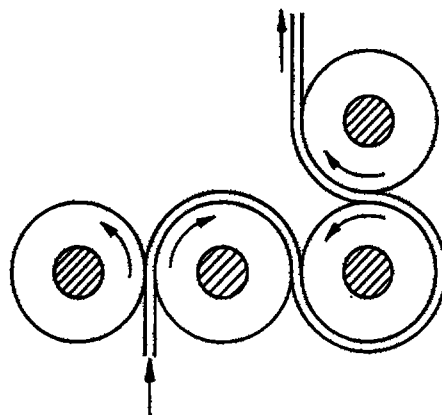
| - CALANDER

Fig. 3b



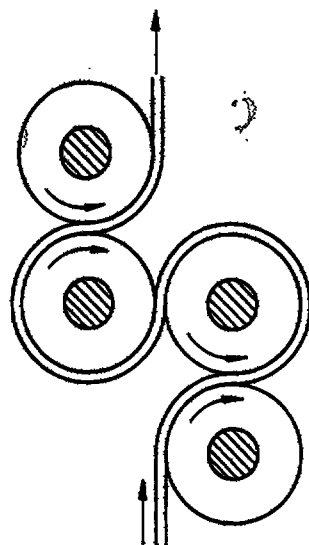
F - CALANDER

Fig. 3c



| - CALANDER

Fig. 3d



S - CALANDER

Fig. 3