Process for preparation of chlorinated poly(vinyl chloride) thermoplastic alloys.

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BE-A- 663 921
DE-A- 2 304 894
FR-A- 2 258 420
US-A- 4 105 711

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Description

TECHNICAL FIELD

The present invention relates to a process for preparing thermoplastic alloys containing chlorinated poly(vinyl chloride) having a chlorine content of at least 57% by weight. More specifically, the present invention relates to a process for preparing chlorinated PVC containing alloys which have low temperature processing properties.

BACKGROUND OF THE INVENTION

It is well known that a chlorinated vinyl polymer referred to as chlorinated polyvinyl chloride, hereinafter CPVC, has excellent high temperature performance characteristics, among other desirable physical properties. Typically, commercial CPVC has in excess of about 57 percent by weight (% by weight) bound chlorine, and is most conveniently prepared by the chlorination of polyvinyl chloride as described in U.S. Patent Nos. 2,996,489; 3,100,762; 3,334,077; 3,334,078; 3,506,837; 3,534,013; 3,591,571; 4,049,517; 4,350,798; 4,377,459; 4,412,898; and 4,459,387 inter alia.

The term CPVC is used herein to define a chlorinated vinyl chloride polymer having in excess of about 57% by weight bound chlorine.

Chlorinated PVC (CPVC) has become an important specialty polymer due to its relatively low cost, high glass transition temperature, high heat distortion temperature, outstanding flame and smoke properties, chemical inertness and low sensitivity to hydrocarbon feed stock costs. The glass transition temperature of CPVC generally increases as the percentage of chlorine increases. However, a well known undesirable characteristic of CPVC resin is that it inherently has low impact properties, a characteristic which is also common to vinyl chloride homopolymers. Also, as the chlorine content increases, the CPVC resin becomes more difficult to melt process, and also becomes more brittle.

The poor melt processability of CPVC resins is exemplified by milling CPVC on a roll mill which results in high torque and high temperatures as well as decomposition of the CPVC. Softening additives or plasticizers have been added to CPVC in order to improve its processability. Although its processability is somewhat improved, these additives produce undesirable effects. Some of the more significant detrimental effects produced by inclusion of these softening or plasticizer additives are lower heat distortion temperatures, softness and weakness in terms of lower tensile strength, and less desirable chemical properties than that exhibited by CPVC alone. These negative attributes of the additives on CPVC limit usefulness of the modified CPVC in the manufacture of rigid plastic articles.

The burgeoning demand for CPVC pipes, vessels, valve bodies and fittings, and the fact that an impact-deficient CPVC matrix can be improved by compounding and alloying it with other polymers, has instigated concerted efforts to develop better impact modified CPVC compositions having increased heat distortion temperatures, and increased ease of melt-processing. Most of these efforts have been channeled toward rigid CPVC applications where acceptable impact strength and dimensional stability under heat are critical.

Such applications include the manufacture of exterior structural products, rigid panels, pipe and conduit, injection-molded and thermoformed industrial parts, appliance housing, and various types of containers both large and small.

U.S. Patent No. 3,882,192 to Elghani, et al relates to molding compositions consisting of from 5 to 95 parts by weight of a polycarbonate, from 5 to 95 parts by weight of a vinyl chloride polymer, and from 5 to 95 parts by weight of an ABS graft polymer, a styrene/maleic anhydride copolymer or an ethylene/ethylene acetate copolymer. Molding compositions are made by solution casting, that is by separately dissolving each component in a suitable inert organic solvent. The three components can then be mixed together and the molding composition isolated from the solutions by precipitation with non-solvents or by removal of the solvents by distillation. Processing of the molding compositions is carried out at a temperature of about 250 °C. In addition to high processing temperatures, which is too high for processing of high chlorine CPVC, no mention is made of the utilization of an ethylene-based functional polymer.

U.S. Patent Nos. 4,105,711; 4,239,361; and 4,515,925 while relating to polymer mixtures containing polycarbonate, do not utilize any ethylene-based functional polymer.

At present, no entirely satisfactory means is available for improving properties such as impact strength, heat distortion temperatures, improved ease of melt processing of chlorinated PVC resins, or any combination thereof.

Since PVC processes easily and CPVC does not, since CPVC has heat resistance but PVC does not, and furthermore, since CPVC has a high melt viscosity but PVC does not, it should be apparent that CPVC
and PVC are different materials and that PVC prior art is not analogous to patentability issues relating to CPVC.

SUMMARY OF THE INVENTION

In accordance with the present invention there is provided a process for making a low temperature melt processable CPVC polymer alloy composition comprising the steps of:

- forming a polycarbonate premixture, said polycarbonate premixture containing from 10% to 80% by weight of said polymer alloy of an aromatic polycarbonate, wherein the weight average molecular weight of said polycarbonate is from 10,000 to 200,000 and
- 1% to 15% by weight of said polymer alloy of an ethylene-based functional polymer being a copolymer of ethylene and at least one other monomer containing functional groups thereon to provide said polycarbonate premixture with a melt processing temperature or 230 °C or less;

- forming a CPVC premixture, said CPVC premixture comprising from 20% to 70% by weight of said polymer alloy of a chlorinated poly(vinyl chloride) containing from 57% to 75% by weight of chlorine therein and optionally
- 1% to 25% by weight of said polymer alloy of an impact modifier having a Tg of 0 °C or less, and optionally
- 0.1% to 20% by weight of said polymer alloy of a viscosity improver polymer,

- forming the low temperature melt processable CPVC polymer alloy by mixing said CPVC premixture with said polycarbonate premixture at a temperature of 230 °C or less.

DETAILED DESCRIPTION OF THE INVENTION

The thermoplastic polymer alloy generally contains a chlorinated poly(vinyl chloride), a polycarbonate, an ethylene-based functional polymer, an optional impact modifier and an optional viscosity improver.

The chlorinated poly(vinyl chloride), i.e. CPVC, resins include any post-chlorinated poly(vinyl chloride), hereinafter CPVC, containing at least 57% by weight of chlorine. The CPVC resins contain from 57% to 75% of chlorine by weight and preferably from 63% to 72% of chlorine by weight. Generally, any poly-(vinyl chloride) (PVC) can be utilized for chlorination having an intrinsic viscosity, obtained utilizing a 99% by weight tetrahydrofuran solution with the PVC being the remaining 1% by weight, of from about 0.46 to about 1.4 and desirably from about 0.54 to about 1.0. The PVC can be prepared in any conventional manner such as by suspension, emulsion, or mass polymerization. Such methods of preparation are well known to the art as well as to the literature. The chlorination process can be carried out according to any method such as by a solution process, a fluidized bed process, a water slurry process, a thermal process, or a liquid chlorine process. Inasmuch as CPVC resins are known to the art as well as to the literature, they will not be discussed in great detail herein. Rather, reference is hereby made to the several CPVC patents set forth in the background, e.g. U.S. Patent Nos. 2,996,489; 3,100,762.

The CPVC resins utilized in the present invention generally have a density of from about 1.45 to about 1.67 gm/cc at 25 °C and a glass transition temperature (Tg) of from about 95 °C to about 200 °C. Such Tgs are at least 20 °C higher than the glass transition temperature of unchlorinated PVC resins. The glass transition temperature was measured by a differential scanning calorimeter.

The preferred CPVC resins have densities in the range of from about 1.55 to about 1.60 gm/cc at 25 °C and a glass transition temperature of at least 100 °C.

Based upon the total amount of the above polymers forming the thermoplastic alloy blend, the amount of the CPVC resins therein is from 20% to 70% and desirably from 30% to 65% by weight. Inasmuch as the thermoplastic alloy can be either polycarbonate rich or CPVC rich, the preferred amount of CPVC will vary accordingly. In the polycarbonate rich embodiment, the preferred amount of the CPVC will be from about 20% to about 50% by weight. In the CPVC rich embodiment, the preferred amount of CPVC will be from about 50% to about 70% by weight. It is to be understood that such weight percentages is based upon only the polymer alloy and not upon any additives, stabilizers, etc.

The polycarbonate utilized in the polymer alloy composition is an aromatic polycarbonate. The thermoplastic polymer alloy of CPVC and polycarbonate is generally a two-phase system. That is, a single overall glass transition temperature Tg is not obtained. Depending upon whether the thermoplastic polymer alloy is CPVC rich or polycarbonate rich, the rich component will often exist in a continuous phase. The non-rich phase will often exist as a discontinuous phase within the continuous phase. Should generally equal amounts of these two polymers be utilized, portions of a continuous phase as well as a discontinuous phase of each material within the overall polymer alloy will often exist. Inasmuch as the CPVC and the
polycarbonate polymers are partially compatible, small homogeneous phases will also exist. It is thus to be understood that various continuous, discontinuous, and even small homogeneous phases can exist within the polymer alloy but that the overall polymer alloy system will generally contain two separate and distinct phases.

The aromatic polycarbonates have a low molecular weight, that is a weight average molecular weight of from 10,000 to 200,000 and preferably from 10,000 to 30,000. Aromatic polycarbonates include various homopolycarbonates, various copolycarbonates and/or mixtures of various homo- and copolycarbonates. The amount of such aromatic polycarbonates is from 10% to 80% by weight and desirably from 30% to 70% by weight based upon the total weight of the various polymers forming the thermoplastic polymer alloy. When a CPVC rich composition is desired, the amount of the polycarbonate is from about 20% to about 50% by weight based upon the total weight of the alloy forming polymers. When a polycarbonate rich alloy is desired the amount of the aromatic polycarbonate is from about 60% to about 80% by weight.

The specific types of aromatic polycarbonate utilized are generally not critical to the present invention provided that they are utilized in an amount and have a molecular weight as set forth herein above. Accordingly, various conventional types of aromatic polycarbonates can be utilized as well as other polycarbonates known to the art and to the literature. As part of the literature, numerous polycarbonate patents exist and the specific types of aromatic polycarbonate set forth therein can be utilized.

Suitable aromatic polycarbonates which can be utilized are set forth in U.S. Patent No. 4,515,925. Generally, such aromatic polycarbonates are made from diphenols corresponding to the formula

\[
\text{HO-Z-OH}
\]

wherein Z represents a divalent mono- or polynuclear aromatic radical having from 6 to 30 carbon atoms, and is constructed such that each of the two OH groups is directly bound to a carbon atom of an aromatic system.

Examples of other types of polycarbonates are set forth in U.S. Patent Nos. 3,544,514; 4,005,037; 4,105,711; 4,239,861; and 4,513,119.


In polycarbonates, groups of dihydric or polyhydric phenols are linked through carbonate groups. Typically, polycarbonates are derived from bisphenol A and diphenyl carbonate through an ester exchange. Polycarbonates can also be made utilizing small amounts of other polyhydric phenols. The polycarbonates of the present invention are usually melt processable at temperatures of from about 260 °C to about 300 °C. Such temperatures are generally too high for melt mixing or blending with CPVC resins.

Aromatic polycarbonates are commercially available with specific examples including General Electric Lexan polycarbonates, desirably Lexan® High Flow; Dow Chemical’s Calibre® polycarbonates, and preferably such high melt flow polycarbonates; and Mobay Chemical’s Merlon® polycarbonate resins, preferably Merlon FCR series.

Ethylene-based functional polymers are utilized to reduce the processing temperature, to reduce the melt viscosity of the polycarbonate and also to impart high impact properties to the polymer alloy. An effective amount is utilized such that the Izod notch impact strength of the polymer alloy is at least 53.38 J/m (1.0 ft-lbs/inch) and desirably at least 106.76 J/m (2.0 ft-lbs/inch) or greater and the processing temperature is 230 °C or less, desirably 220 °C or less and preferably 215 °C or less. Generally, the ethylene-based functional polymer is premixed with the polycarbonate at a temperature of about or below 220 °C. Usually, an effective amount is from 1% to 15%, and preferably from 3% to 10% by weight of the ethylene-based functional polymers based upon the total weight of compounds forming the polymer alloy, that is the CPVC, the aromatic polycarbonate, the optional impact modifier, the optional viscosity improver, and the ethylene-based functional polymer.

Large improvements in impact strength and/or decreases in processing temperatures are unexpectedly obtained when the ethylene-based functional polymer is premixed with the aromatic polycarbonate. Although the viscosity improver can also be mixed therewith, it is generally mixed with the CPVC. While not being intended to be bound to theory, it is thought that the ethylene type functional polymer reduces the viscosity of the polycarbonate and thus can lower the processing temperature. Mixing of the polycarbonate premixure can occur by utilizing any conventional melt method. For example, the various premixture components can be mixed on a two-roll mill, in a compounding screw extruder, in a twin screw extruder.

Regardless of mixing method, the temperature is such to generally produce a melt mixture of from about 210 °C to about 230 °C, desirably from about 210 °C to about 220 °C, and preferably from about 215 °C to about 220 °C.
A stabilized CPVC premixture can also be made and contains the CPVC, a heat stabilizer, the optional impact modifiers, and the optional viscosity improvers. The CPVC premixture can also contain small amounts of the ethylene-based functional polymers although such is desirably utilized in the polycarbonate premixture. Mixing of the CPVC premixture can occur in any conventional melt mixing method such as by extrusion mixing, roll mill mixing. The temperatures are maintained generally below 230 °C and preferably below 220 °C. Otherwise, the CPVC tends to degrade or to decompose. If this occurs, the overall polymer alloy will have poor physical properties. The CPVC premixture is then mixed with the polycarbonate premixture in any conventional melt mix manner, for example extrusion mixing and roll mix mixing, at comparatively low temperatures, that is generally at temperatures of 230 °C or less and preferably at 220 °C or less.

The polymer alloy is made utilizing the polycarbonate premixture, since as noted above, surprisingly good high impact properties and relatively low processing temperatures are achieved. In contrast thereto, should all the various components be simultaneously mixed together, high mixing temperatures, that is generally in excess of 220 °C and usually in excess of 230 °C are required with resulting deleterious effects upon the CPVC compound. Simultaneous mixing, furthermore, does not result in producing a polymer alloy having good impact strength. In addition to having good impact resistant properties, the properties of the polymer alloy of the present invention include good ductility, good flame retardancy, good high heat distortion temperatures, and also good thermal and color stability. The polymer alloys can thus be utilized wherever such properties are desired, for example in structural plastics, calendaring sheet and injection molding for equipment housings.

The ethylene-based functional polymer is generally a copolymer of ethylene and at least one other monomer containing functional groups thereon. Generally, the remaining comonomer can be a vinyl ester having a total of from 1 to 10 carbon atoms with acetate being preferred. The amount of the vinyl ester component of the ethylene-based functional copolymer is from 5 to 50% by weight and desirably from 10 to 35% by weight. The ethylene-vinyl ester copolymers are often utilized as a blend with a small amount of another ethylene copolymer. The additional monomers of the second copolymer which contains functional groups are usually vinyl organic acids or organic anhydrides. For example, the additional comonomer can be acrylic acid, methacrylic acid, and various hydrocarbon derivatives thereof as well as maleic anhydride, or various derivatives thereof, wherein said acids and said anhydrides have a total of from 3 to 12 carbon atoms. Methacrylic acid is a preferred comonomer. The additional monomer of the second copolymer generally exists in a small amount as from 1% to 25% by weight based upon the total weight of the second or additional copolymer. When utilized, the amount of the second copolymer is generally small, as from 0 or 1% to 50% by weight and desirably from 5% to 40% by weight. Thus, the amount of the ethylene-vinyl ester copolymer exists in amount of from 50% to 100% by weight and preferably from 60% to 95% by weight. Other types of ethylene-based functional polymers or copolymers are set forth in U.S. Patent No. 4,230,830. This patent relates to a polymer blend consisting essentially of
a. about from 80-99% by weight of a first olefinic polymer selected from
   (i) non-polar ethylene polymers and copolymers having a density of about from 0.930 to 0.965 g/cc and
   (ii) copolymers of ethylene having up to about 30 weight percent of at least one ethylenically unsaturated ester having from 4 to 12 carbon atoms; and
b. about from 1-19% of a second olefinic polymer selected from the group consisting of
   (i) non polar ethylene polymers and copolymers having a density of about from 0.945 to 0.965 g/cc and
   (ii) terpolymers of ethylene, at least one a-olefin having from 3-6 carbon atoms, and at least one nonconjugated diene; the second olefinic polymer being thermally grafted with an unsaturated acid or anhydride to give a copolymer having about from 0.002 to 4.0 weight percent of grafted succinic groups, provided, however, that when the second olefinic polymer is (ii), then the first olefinic polymer is (ii).

Regardless of whether or not a second or a terpolymer ethylene-based functional copolymer is utilized, as described in the preceding paragraph the melt index of the overall ethylene-based functional polymer, that is an ethylene-vinyl ester copolymer or blends thereof, is from about 2 to 40g/10 min and desirably from about 6 to about 25g/10 min. The density is from about 0.92 to about 1.1 g/cc and desirably from about 0.92 to about 0.96 g/cc. The ethylene-based functional polymer or blends thereof useful in the present invention are available from DuPont under the name Bynel® CXA resins such as CXA resin 1123 and/or resin 1124. A specific example of a suitable ethylene-based functional polymer is a blend of 92.5% by weight of an ethylene-vinyl acetate copolymer containing 28% by weight of vinyl acetate therein and 7.5% by weight of an ethylene-methacrylic acid copolymer containing 15% by weight of methacrylic acid.
The impact modifiers which can be optionally utilized in the CPVC premixture are generally added in an effective amount which helps improve the impact strength, for example, the Izod notch strength of the polymer alloy. Such amounts when utilized generally range from 1 to 25%, desirably from about 2 to 15% and preferably 3 to 5% by weight based upon the total weight of the polymer alloy forming compounds. Suitable impact modifiers which can be utilized in the present invention have a Tg of generally 0°C or less and very often a much lower Tg is desirable. As known to the art as well as to the literature, many polymeric compounds which could impart good impact strength for rigid PVC can be utilized. Accordingly, various impact modifiers which impart improved impact resistance to the polymer alloy can be utilized including those known to the literature, such as for example those set forth in The Encyclopedia of PVC, Volume 2, Chapter 12, Marcel Dekker, Inc., New York, 1977. Examples of specific impact modifiers include the various acrylonitrile-butadiene-sytrene (ABS) polymers, the various chlorinated polyethylene, the various acrylic rubbers, the various poly(ethylene-co-vinyl acetates, styrene-butadiene-styrene block copolymers, poly(methacrylate-co-butadiene-co-sytrene) (MBS), and the like. Impact modifiers of these types are commercially available. Preferred impact modifiers include ABS, MBS, and chlorinated polyethylene.

Various viscosity improvers can optionally be utilized in the present invention. Generally, effective amounts are utilized to reduce the viscosity of the polymer alloy and more specifically to reduce the viscosity of the CPVC compound as in the CPVC premixture. Such compounds also tend to prevent the CPVC from degrading and hence also stabilize the same. Effective amounts, when utilized, are from 0.1 to 20% by weight, desirably from 0.5 to 10% by weight and preferably from 3 to 10% by weight based upon the total weight of the polymer alloy forming compounds as defined hereinabove. Such viscosity improvers are known to the art as well as to the literature. Examples of such various types include conventional compounds such as the various styrene-acrylonitrile copolymers, for example those described in the Encyclopedia of Polymer Science and Engineering, Volume I, pages 452-470, 1985, John Wiley & Sons, Inc., New York; a polymer of alpha methylstyrene, styrene, methyl methacrylate, or an acrylonitrile as described in U.S. Patent No. 4,304,884 to Okamoto.

Other viscosity improvers include various lubricants such as low molecular weight oxidized polyethylene, stearic acid, low molecular weight polyethylene. Although these types of viscosity improvers can be utilized, it is to be understood that various other types can be utilized and that such are known to the literature as well as to the art. The important aspect is that they generally lower the viscosity and hence also improve the stability of the CPVC compounds.

In addition to the above polymer alloy forming compounds, the CPVC premixture according to the present invention preferably comprises a CPVC resin which has been premixed with heat stabilizers. The stabilization of CPVC resins and compounds finds their roots in PVC stabilization technology. Consequently, the stabilizers being used for CPVC resins are based on, for example, lead, barium and/or cadmium, calcium and/or zinc and organotin stabilizers. Among these, organotin stabilizers are widely used. Other secondary stabilizers (or called a co-stabilizer) can also be utilized. For example, epoxy compounds; e.g., epoxidized soybean oil, epoxy esters, are strongly synergistic co-stabilizers for metal-based stabilizer system, as well as for organic stabilizers. A detailed description of stabilizers is available in PVC Technology, Fourth Edition, Chapters 9 and 10, W.V. Titow (Editor), Elsevier Applied Science Publishers, London and New York, 1984.

In addition to the above stabilizers, various conventional antioxidants can be utilized in typical amounts with regard to the various polycarbonates and the various ethylene-based functional polymers. These compounds are well known to the art as well as to the literature.

The invention will be better understood by reference to the following examples.

The following recipes with ingredients as specified, are mechanically melt-mixed in a 6 inch electric mill at 215°C for about 2 minutes and compression molded at 215°C for one and a half minutes in a laboratory press. In Examples 1-2, TempRite® T-1310 CPVC, manufactured by The BFGoodrich Company, is masterbatch with stabilizer, antioxidants, with and without Bynel® CXA 1123, an ethylene-based functional polymer available from DuPont. The ethylene-based functional compound has a melt index of 6.6 and comprises a blend of 92.5% of ethylene/28% vinyl acetate copolymer and 7.5% of ethylene/15% methacrylic acid copolymer. TempRite® T-1310 CPVC contains 69.6% bound chlorine, and has an intrinsic viscosity of about 0.68 in tetrahydrofuran (THF). Each batch of 100 parts of CPVC contains 4 parts stabilizer, such as T-31 THERMOLITE® stabilizer, available from M&T which is dibutyl tin bisisoctylthioglycolate; also 1 part of Irganox® 1010 which is 2,2-bis[[3,5-bis(1,1-Dimethylallyl)-4-hydroxyphenyl]-1-oxopropyloxy][methyl]-1,3-propanediyl.
3,5-bis(1,1-dimethyl-ethyl)-4-hydroxybenzeneepropanoate available from Ciba Geigy, and 0.5 parts of BHT which is (2,6-di-tert-butyl-4-methylphenol) available from Alrich. Example 1 does not contain any ethylene-based polymers. Example 2 contains 10 parts by weight of the ethylene-based copolymer blend.

In Examples 3-4, Lexan® HP 2110-111, which is a high flow polycarbonate resin available from General Electric Company, having a weight average molecular weight of about 25,000, is melt mixed with 1 part by weight of Irganox® 1010 and 0.5 parts BHT, with and without Bynel® CXA 1123. Example 3 does not contain Bynel® CXA resin. Example 4 contains 10 parts Bynel® CXA 1123. The thermomechanical properties of these examples are listed in Table I.

**TABLE I**

<table>
<thead>
<tr>
<th>EXAMPLE</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
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<tbody>
<tr>
<td>CPVC TempRite T-1310</td>
<td>100</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Bynel CXA 1123</td>
<td>0</td>
<td>10</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>Lexan HP 2110-111</td>
<td>0</td>
<td>0</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>

**Izod Impact (ft-lb/in) **

(ASTM D256)

Notched

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>0.6</td>
<td>5</td>
<td>8</td>
<td></td>
</tr>
</tbody>
</table>

Unnotched

|   | 1 | 2 | 18 | 24 |

Heat Distortion Temperature

(ASTM 684, 264 psi) (annealed)

|   | 99 | 94 | 130 | 130 |

**Apparent Shear Viscosity (Pa-S) @ 220°C**

Shear Rate = 85 sec\(^{-1}\)

| 4780 | 3110 | 10900 | 3460 |

Shear Rate = 850 sec\(^{-1}\)

| 850  | 700  | 2580  | 1050  |

* The shear viscosity was measured using an Instron capillary rheometer, die diameter = 0.06 inch.

**A ft-lb/inch = 53.38 J/m**

The above data demonstrates that the Bynel® CXA resin reduces the shear viscosity of unmodified CPVC and also very drastically lowers the shear viscosity of polycarbonate. At 220°C, the magnitude of shear viscosity of polycarbonate premixture containing 10 parts Bynel® CXA resin (Example 4) is similar to that of unmodified CPVC resin (Example 1). This improves the processability of blending polycarbonate with CPVC resin as will be discussed below.

The compositions tested in Table II hereinbelow are identified as follows:

In Example 5-7, each batch is formulated with 50 parts CPVC TempRite® T-1310, with 2 parts T-31 THERMOLITE® stabilizer, and also 50 parts Lexan HP 2110-111 with 1 part Irganox® 1010 and 0.5 parts BHT antioxidant. Example 5 does not include Bynel® CXA resin, while Examples 6 and 7 both contain 10
parts Bynel® CXA 1123. Examples 5 and 6 are prepared by mixing all the ingredients simultaneously at 215°C for 2 mins. Example 7 was prepared by premixing the Bynel® CXA resin and polycarbonate and antioxidants at 215°C to form a polycarbonate premixture, and then adding this premixture to the CPVC (plus stabilizer) at 215°C. The Izod impact strength and heat distortion temperature are listed in Table II.

**TABLE II**

<table>
<thead>
<tr>
<th>EXAMPLE</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>CPVC TempRite T-1310</td>
<td>50</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>Bynel CXA 1123</td>
<td>0</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Lexan HP 2110-111</td>
<td>50</td>
<td>50</td>
<td>50</td>
</tr>
</tbody>
</table>

**Izod Impact (ft-lb/in.)**

(ASTM D256)

Notched 0.7  1.4  6
Unnotched 4   9   29

**Heat Distortion Temperature (°C)**

(1 ft-lb/in. = 53.38 J/m)

<table>
<thead>
<tr>
<th></th>
<th>112</th>
<th>101</th>
<th>110</th>
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</thead>
<tbody>
<tr>
<td>(unannealed)</td>
<td>53.38 J/m</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The foregoing data demonstrate, that the blend, Example 7, prepared according to the present invention by mixing Bynel CXA resin with the polycarbonate at 215°C to form a premixture and then mixing the same with the premix stabilized CPVC, exhibits better impact strength and also better heat distortion temperature, when compared with Example 6, an identical composition but wherein all of the ingredients were mixed simultaneously.

Examples 8 - 11 relate to blend compounds comprising CPVC TempRite® T-1310 resin, polycarbonate, Bynel® CXA, impact modifiers, and processing improvers. The detailed compositions and processes are described hereinbelow. In Examples 12 and 13, however, a commercially available stabilized CPVC compound, TempRite® 3503, is used, a CPVC compound having a 67% Chlorine CPVC resin, and having an intrinsic viscosity of about 0.68 in THF.

**EXAMPLE 8**

A CPVC mixture was prepared by mixing at 215°C in an electric two-roll mill, 70 parts by weight of CPVC TempRite® T-1310 (69.8% chlorine) with 10 parts by weight of styrene-acrylonitrile, 15 parts by weight of ABS modifier, 5 parts by weight of chlorinated polyethylene impact modifier, 2.8 parts of T-31 THERMOLITE® stabilizer and 0.5 parts of oxidized polyethylene lubricant. Various properties of this mixture are set forth in Table III.

**EXAMPLE 9**

A polycarbonate mixture was prepared by mixing at 215°C in an electric mill, 30 parts by weight of polycarbonate with 5 parts by weight Bynel® CXA 1123 resin. The polycarbonate was Lexan® HP 2110-111 available from General Electric. The antioxidants used for polycarbonate premixture were 1 part by weight.
of Irganox® 1010 available from Ciba-Geiigi and 1 part by weight of BHT available from Airich. The melted polycarbonate premixture was then mixed at 215°C with 100 parts by weight of the stabilized CPVC-premixture as described in Example 8. Various properties are set forth in Table III.

EXAMPLE 10

The composition was prepared as described in Example 9, using the same polymers with appropriate additives. The polycarbonate premixture, however, consisted of 100 parts by weight of polycarbonate and 5 parts by weight of Bynel® CXA 1123. Various properties are set forth in Table III.

EXAMPLE 11

The composition was prepared as described in Example 9, using the same polymers with appropriate additives. The polycarbonate premixture consisted of 200 parts by weight of polycarbonate, and 10 parts by weight of Bynel® CXA 1123. Various properties are set forth in Table III.

EXAMPLE 12

A 100 parts by weight of TempRite 3503 was melt-mixed at 215°C. The TempRite® 3503 was a CPVC compound based on 67% chlorine CPVC available from the BFGoodrich Company. Various properties are set forth in Table III.

EXAMPLE 13

The polycarbonate premixture was prepared as described in Example 9, using 60 parts of Lexan® HP 2110-111, and 5 parts of Bynel® CXA 1123. A 100 parts of TempRite® 3503 as described in Example 12 was then mixed with the polycarbonate premixture at 215°C. Various properties are set forth in Table III.

| TABLE III |

| EXAMPLE |

| Izod Impact (ft-lb/in.) | X |
| (ASTM D256) | |
| Notched | 2 6 7 8 1 4 |
| Unnotched | 17 25 40 35 11 33 |

Heat Distortion Temperature °C
(ASTM D-684: 264 psi)
| 86 96 105 118 98 99 |

* 1 ft-lb/in. = 53.38 J/m

It is obvious from Table III that the polymer mixtures, Examples 9 - 11, according to the present invention, containing a high chlorine CPVC resin TempRite T-1310, polycarbonate, and also fully compounded with impact modifier and processing improvers, have much better impact strength and heat
distortion temperatures than that without a premixture of polycarbonate-Bynel® CXA resin, i.e. Example 8. Example 13 containing a 67% chlorine CPVC compound TempRite® 3503 with polycarbonate and Bynel® CXA resin also had a much better impact strength and higher heat distortion temperature than that of TempRite 3503, Example 12.

Claims

1. A process for making a low temperature melt processable CPVC polymer alloy composition comprising the steps of:
   - forming a polycarbonate premixture, said polycarbonate premixture containing
     from 10 % to 80 % by weight of said polymer alloy of an aromatic polycarbonate, wherein the
     weight average molecular weight of said polycarbonate is from 10,000 to 200,000 and
     1 % to 15 % by weight of said polymer alloy of an ethylene-based functional polymer being a
     copolymer of ethylene and at least one other monomer containing functional groups thereon to provide
     said polycarbonate premixture with a melt processing temperature of 230 °C or less;
   - forming a CPVC premixture, said CPVC premixture comprising
     from 20 % to 70 % by weight of said polymer alloy of a chlorinated poly(vinyl chloride) containing
     from 57 % to 75 % by weight of chlorine therein and optionally
     1 % to 25 % by weight of said polymer alloy of an impact modifier having a Tg of 0 °C or less, and
     optionally
     0.1 % to 20 % by weight of said polymer alloy of a viscosity improver polymer,
   - forming the low temperature melt processable CPVC polymer alloy by mixing said CPVC premix-
     ture with said polycarbonate premixture at a temperature of 230 °C or less.

2. A process according to claim 1, wherein the amount of said polycarbonate is from 10 % to 80 % by
   weight of said polymer alloy, wherein the weight average molecular weight of said polycarbonate is
   from 10,000 to 200,000 wherein the amount of said ethylene-based functional polymer is from 1 % to
   15 % by weight of said polymer alloy, including melt processing said polycarbonate premixture at a
   temperature of 230 °C or less, wherein the amount of said CPVC is from 20 % to 70 % by weight of said
   polymer alloy, wherein said CPVC has a chlorine content of from 57 % to 75 % by weight, and
   including melt processing said CPVC premixture at a temperature of 230 °C or less.

3. A process according to claim 2, wherein said ethylene-based functional polymer is a blend of an
   ethylene-vinyl ester and an ethylene-vinyl and functional containing copolymer, wherein said vinyl ester
   portion of said ethylene-vinyl ester copolymer has a total of from 1 to 10 carbon atoms, wherein the
   amount of said ethylene-vinyl ester copolymer is from 50 % to 100 % by weight based upon the total
   amount of said ethylene-vinyl ester copolymer and said ethylene-vinyl and functional containing
   copolymer, and wherein the amount of said ethylene-vinyl and functional containing copolymer is from
   0 % or 1 % to 50 % by weight, and wherein said vinyl and functional portion of said ethylene-vinyl and
   functional copolymer is a vinyl organic acid or an organic anhydride having from 3 to 12 carbon atoms.

4. A process according to claim 3, wherein the amount of said CPVC is from 30 % to 65 % by weight of
   said polymer alloy, including an impact modifier, wherein the amount of said impact modifier is from 2
   % to 15 % by weight of said polymer alloy, wherein the amount of said polycarbonate is from 30 % to
   70 % by weight of said polymer alloy, wherein said polycarbonate has a weight average molecular
   weight of from 10,000 to 30,000, wherein the amount of said chlorine in said CPVC is from 63 % to 72
   % by weight, and wherein the total amount of said ethylene copolymers is from 3 % to 10 % by weight
   of said polymer alloy, and wherein the Izod notch impact strength of said polymer alloy is at least 53.38
   J/m (1.0 ft-lb/in).

5. A process according to claim 1, wherein said ethylene-based functional copolymer is a blend of an
   ethylene-vinyl ester copolymer and at least one ethylene-vinyl and functional containing copolymer,
   wherein the amount of said ethylene-vinyl ester copolymer is from 50 % to 100 % by weight based
   upon the total weight of said ethylene-vinyl ester copolymer and said ethylene-vinyl and functional
   containing copolymer, wherein the amount of said ethylene-vinyl and functional containing copolymer is
   from 0 % or 1 % to 50 % by weight, wherein said ester portion of said ethylene-vinyl ester copolymer
   contains a total of from 1 to 10 carbon atoms, wherein said vinyl and functional portion of said ethylene-
   vinyl and functional copolymer contains a vinyl organic acid or an organic anhydride having from 3 to
12 carbon atoms and wherein the weight of said vinyl ester portion of said ethylene-vinyl ester copolymer is from 5 % to 50 % by weight.

6. A process according to claim 5, wherein the amount of said CPVC is from 30 % to 65 % by weight of said polymer alloy, wherein the amount of said polycarbonate is from 30 % to 70 % by weight of said polymer alloy, wherein said polycarbonate has a weight average molecular weight of from 10,000 to 30,000.

7. A process according to claim 6, wherein the amount of said chlorine in said CPVC is from 63 % to 72 % by weight, wherein the total amount of said ethylene-based functional copolymers is from 3 % to 10 % by weight of said polymer alloy, and wherein said polymer alloy has a melt processing temperature of 220 °C or less.

8. A process according to claim 7 wherein the amount of said ethylene-vinyl ester copolymer is from 60 % to 95 % by weight, wherein said vinyl ester is vinyl acetate, wherein the amount of said vinyl acetate in said ethylene-vinyl acetate copolymer is from 10 % to 35 % by weight, wherein the amount of said ethylene-vinyl and functional containing copolymer is from 5 % to 40 % by weight, wherein said vinyl and functional containing portion of said ethylene-vinyl and functional containing copolymer is a methacrylic acid, and wherein the amount of said methacrylic acid in said ethylene-methacrylic acid copolymer is from 1 % to 25 % by weight.

9. A process according to claim 8, including from 2 % to 15 % by weight of an impact modifier, and including from 0.5 % to 10 % by weight of a viscosity improver.

Patentansprüche

1. Verfahren zur Herstellung einer bei niedrigen Temperaturen schmelzverarbeitbaren CPVC Polymer-Legierung, das die Schritte umfaßt:
   Herstellen einer Polycarbonat-Vormischung, wobei die Polycarbonat-Vormischung
   10 bis 80 Gew.-%, bezogen auf die Polymer-Legierung eines aromatischen Polycarbonats, worin das
ein Molekulargewicht des Polycarbonats von 10 000 bis 200 000 reicht und
   1 bis 15 Gew.-%, bezogen auf die Polymer-Legierung eines funktionellen Polymers auf Ethylenbasis,
das ein Copolymer aus Ethylen und wenigstens einem weiteren Monomer ist, das funktionelle Gruppen
   auf diesem enthält, um die Polycarbonat-Vormischung mit einer Schmelz-Verarbeitungstemperatur von
   230 °C oder weniger zu versehen, enthält;
   Herstellen einer CPVC-Vormischung, wobei die CPVC-Vormischung
   20 bis 70 Gew.-%, bezogen auf die Polymer-Legierung eines chlorierten Poly(vinylchlorid)s, das 57 bis
   75 Gew.-% Chlor enthält und wahlweise
   1 bis 25 Gew.-%, bezogen auf die Polymer-Legierung eines Schlagfestmachers mit einer Einfriertempe-
   ratur von 0 °C oder weniger und wahlweise
   0,1 bis 20 Gew.-%, bezogen auf die Polymer-Legierung eines Viscositäts-Verbesserungs-Polymers
   umfaßt;
   Herstellen der bei niedriger Temperatur schmelzverarbeitbaren CPVC-Polymer-Legierung durch Mi-
   schen der CPVC-Vormischung mit der Polycarbonat-Vormischung bei einer Temperatur von 230 °C
   oder weniger.

2. Verfahren nach Anspruch 1, worin die Menge des Polycarbonats 10 bis 80 Gew.-% der Polymer-
   Legierung beträgt, worin das mittlere Molekulargewicht des Polycarbonats von 10 000 bis 200 000
   reicht, worin die Menge des funktionellen Polymers auf Ethylenbasis 1 bis 15 Gew.-% der Polymer-
   Legierung beträgt, wobei die Schmelzverarbeitung der Polycarbonat-Vormischung bei einer Temperatur
   von 230 °C oder weniger erfolgt, worin die Menge des CPVC 20 bis 70 Gew.-% der Polymer-
   Legierung beträgt, worin CPVC einen Chorgehalt von 57 bis 75 Gew.-% hat und Schmelzverarbeitung
   der CPVC-Vormischung bei einer Temperatur von 230 °C oder weniger erfolgt.

3. Verfahren nach Anspruch 2, worin das funktionelle Polymer auf Ethylenbasis eine Mischung aus einem
   Ethylenvinylester und einem Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymer ist, worin
   der Vinylsternanteil des Ethylenvinylester-Copolymers insgesamt 1 bis 10 Kohlenstoffatome hat, worin
   die Menge des Ethylenvinylester-Copolymers 50 bis 100 Gew.-%, bezogen auf die Gesamtmenge des

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Ethylenvinylester-Copolymere und Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymere, beträgt, und worin die Menge des Ethylenvinyls und funktionelle Gruppen enthaltenden Copolymers 0 oder 1 bis 50 Gew.-% beträgt, und worin der Vinyl- und funktionelle Gruppenanteil des Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymers eine vinylogische Säure oder ein organisches Anhydrid mit 3 bis 12 Kohlenstoffatomen ist.

4. Verfahren nach Anspruch 3, worin die Menge des CPVC 30 bis 65 Gew.-% der Polymer-Legierung beträgt, einschließlich einem Schlagfestmacher, worin die Menge des Schlagfestmachers 2 bis 15 Gew.-% der Polymer-Legierung beträgt, worin die Menge des Polycarbonats 30 bis 70 Gew.-% der Polymer-Legierung beträgt, worin das Polycarbonat ein mittleres Molekulargewicht von 10 000 bis 30 000 hat, worin die Menge an Chlor im CPVC 63 bis 72 Gew.-% beträgt und worin die Gesamtmenge des Ethylen-Copolymer 3 bis 10 Gew.-% der Polymer-Legierung beträgt und worin die Izod-Kerbschlagzähigkeit der Polymer-Legierung wenigstens 53,38 J/m (1,0 ft-lb/inch) ist.

5. Verfahren nach Anspruch 1, worin das funktionelle Copolymer auf Ethylenbasis eine Mischung aus einem Ethylenvinylester-Copolymer und wenigstens einem Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymer ist, worin die Menge des Ethylenvinylether-Copolymer 50 bis 100 Gew.-%, bezogen auf das Gesamtgewicht des Ethylenvinylester-Copolymer und des Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymers, beträgt, worin die Menge des Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymers 0 oder 1 bis 50 Gew.-% beträgt, worin der Esteranteil des Ethylenvinylester-Copolymer insgesamt 1 bis 10 Kohlenstoffatome enthält, worin der Vinyl- und funktionelle Anteil des Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymers eine vinylogische Säure oder ein organisches Anhydrid mit 3 bis 12 Kohlenstoffatomen enthält und worin das Gewicht des Vinylsteranteils des Ethylenvinylester-Copolymer 5 bis 50 Gew.-% beträgt.

6. Verfahren nach Anspruch 5, worin die Menge des CPVC 30 bis 65 Gew.-% der Polymer-Legierung beträgt, worin die Menge des Polycarbonats 30 bis 70 Gew.-% der Polymer-Legierung beträgt, worin das Polycarbonat ein mittleres Molekulargewicht von 10 000 bis 30 000 hat.

7. Verfahren nach Anspruch 6, worin die Menge an Chlor im CPVC 63 bis 72 Gew.-% beträgt, worin die Gesamtmenge des funktionellen Copolymers auf Ethylenbasis 3 bis 10 Gew.-% der Polymer-Legierung beträgt und worin die Polymer-Legierung eine Schmelz-Verarbeitungstemperatur von 220 °C oder weniger hat.

8. Verfahren nach Anspruch 7, worin die Menge des Ethylenvinylether-Copolymer 60 bis 95 Gew.-% beträgt, worin der Vinylster Vinylacetat ist, worin die Menge des Vinylacetats in dem Ethylenvinylether-Copolymer 10 bis 35 Gew.-% beträgt, worin die Menge des Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymers 5 bis 40 Gew.-% beträgt, worin der Vinyl- und funktionelle Gruppen enthaltende Anteil des Ethylenvinyl und funktionelle Gruppen enthaltenden Copolymers eine Methacrylsäure ist und worin die Menge der Methacrylsäure in dem Ethylenmethacrylsäure-Copolymer 1 bis 25 Gew.-% beträgt.

9. Verfahren nach Anspruch 8, das 2 bis 15 Gew.-% eines Schlagfestmachers und 0,5 bis 10 Gew.-% eines Viscositäts-Verbesserers einschließt.

Reverdications

1. Procédé pour produire une composition d’alliage polymérique de CPVC pouvant être mis en œuvre à l’état fondu à basse température, qui comprend les étapes consistant :
   - à former un prémélange à base de polycarbonate, ledit prémélange à base de polycarbonate contenant de 10 à 80 % en poids, par rapport audit alliage polymérique, d’un polycarbonate aromatique, la masse moléculaire moyenne en masse dudit polycarbonate étant de 10 000 à 200 000, et de 1 à 15 % en poids, par rapport audit alliage polymérique, d’un polymère fonctionnel à base d’éthylène, qui est un copolymère de l’éthylène et d’au moins un autre monomère contenant des groupes fonctionnels de façon que le prémélange à base de polycarbonate ainsi obtenu ait une température de mise en œuvre à l’état fondu de 230 °C ou moins ;
   - à former un prémélange de CPVC, ledit prémélange de CPVC contenant de 20 à 70 % en poids, par rapport audit alliage polymérique, d’un poly(chlorure de vinyle) chloré contenant de 57 à 75
% en poids de chlore, et éventuellement de 1 à 25 % en poids, par rapport audit alliage polyémérique, d'un modificateur de choc ayant une Tg de 0°C ou moins, et éventuellement de 0,1 à 20 % en poids, par rapport audit alliage polyémérique, d'un polyémère améliorant la viscosité ;
- à former l'alliage polyémérique de CPVC pouvant être mis en oeuvre à l'état fondu à basse température, par mélange dudit prémélange de CPVC audit prémélange à base de polycarbonate, à une température de 230°C ou moins.

2. Procédé selon la revendication 1, dans lequel la quantité dudit polycarbonate est de 10 à 80 % en poids par rapport audit alliage polyémérique, la masse moléculaire moyenne en masse dudit polycarbonate étant de 10 000 à 200 000, la quantité dudit polyémère fonctionnel à base d'éthylène étant de 1 à 15 % en poids par rapport audit alliage métallique, qui comprend les étapes consistant à mettre en oeuvre à l'état fondu ledit prémélange à base de polycarbonate à une température de 230°C ou moins, la quantité dudit CPVC étant de 20 à 70 % en poids par rapport audit alliage polyémérique, ledit CPVC ayant une teneur en chlore de 57 à 75 % en poids, et comprenant en outre l'étape consistant à mettre en oeuvre à l'état fondu ledit prémélange de CPVC à une température de 230°C ou moins.

3. Procédé selon la revendication 2, dans lequel ledit polyémère fonctionnel à base d'éthylène est un mélange d'un copolymère d'éthylène et d'un ester vinylque et d'un copolymère éthylène-vinylque contenant des groupes fonctionnels, ladite portion ester vinylque dudit copolymère éthylène-ester vinylque ayant en tout de 1 à 10 atomes de carbone, la quantité dudit copolymère éthylène-ester vinylque étant de 50 à 100 % en poids par rapport au poids total dudit copolymère éthylène-ester vinylque et dudit copolymère éthylène-vinylque et contenant des groupes fonctionnels, la quantité dudit copolymère éthylène-vinyle contenant des groupes fonctionnels étant de 0 ou 1 à 50 % en poids, ladite portion vinylque et fonctionnelle dudit copolymère éthylène-vinylque comportant des groupes fonctionnels étant acide organique vinylque ou un anhydride organique ayant de 3 à 12 atomes de carbone.

4. Procédé selon la revendication 3, dans lequel la quantité dudit CPVC est de 30 à 85 % en poids par rapport audit alliage polyémérique, procédé dans lequel on utilise un modificateur de choc, la quantité dudit modificateur de choc étant de 2 à 15 % en poids par rapport audit alliage polyémérique, la quantité dudit polycarbonate étant de 30 à 70 % en poids par rapport audit alliage polyémérique, ledit polycarbonate ayant une masse moléculaire moyenne en masse de 10 000 à 30 000, la quantité dudit chlore dans ledit CPVC étant de 63 à 72 % en poids, la quantité totale desdits copolymères de l'éthylène étant de 3 à 10 % en poids par rapport audit alliage polyémérique, la résistance au choc Izod sur barreau entaillé dudit alliage polyémère étant d'au moins 53.36 J/m (1.0 ft-lb/in).

5. Procédé selon la revendication 1, dans lequel ledit copolymère fonctionnel à base d'éthylène est un mélange d'un copolymère éthylène-ester vinylque et d'au moins un copolymère éthylène-vinylque et contenant des groupes fonctionnels, la quantité dudit copolymère éthylène-ester vinylque étant de 50 à 100 % en poids par rapport au poids total dudit copolymère éthylène-ester vinylque et dudit copolymère éthylène-vinylque et contenant des groupes fonctionnels, la quantité dudit copolymère éthylène-vinylque et contenant des groupes fonctionnels étant de 0 ou 1 à 50 % en poids, ladite portion ester dudit copolymère éthylène-ester vinylque contenant en tout de 1 à 10 atomes de carbone, ladite portion vinylque et fonctionnelle dudit copolymère éthylène-vinylque et contenant des groupes fonctionnels contenant un acide organique vinylque ou un anhydride organique ayant de 3 à 12 atomes de carbone, la masse de ladite portion ester vinylque dudit copolymère éthylène-ester vinylque étant de 5 à 50 % en poids.

6. Procédé selon la revendication 5, dans lequel la quantité dudit CPVC est de 30 à 85 % en poids par rapport audit alliage polyémérique, la quantité dudit polycarbonate étant de 30 à 70 % en poids par rapport audit alliage polyémérique, ledit polycarbonate ayant une masse moléculaire moyenne en masse de 10 000 à 30 000.

7. Procédé selon la revendication 6, dans lequel la quantité dudit chlore dans ledit CPVC est de 63 à 72 % en poids, la quantité totale desdits copolymères fonctionnels à base d'éthylène étant de 3 à 10 % en poids par rapport audit alliage polyémère, ledit alliage polyémérique ayant une température de mise en oeuvre à l'état fondu de 220°C ou moins.
8. Procédé selon la revendication 7, dans lequel la quantité dudit copolymère éthylène-ester vinylique est de 60 à 95 % en poids, ledit ester vinylique étant l'acétate de vinylique, la quantité dudit acétate de vinylique dans ledit copolymère éthylène-acétate de vinylique étant de 10 à 35 % en poids, la quantité dudit copolymère éthylène-vinylique et contenant des groupes fonctionnels étant de 5 à 40 % en poids, ladite portion vinylique et contenant des groupes fonctionnels dudit copolymère éthylène-vinylique et contenant des groupes fonctionnels étant un acide méthacrylique, la quantité dudit acide méthacrylique dans ledit copolymère éthylène-acide méthacrylique étant de 1 à 25 % en poids.

9. Procédé selon la revendication 8, dans lequel on a de 2 à 15 % en poids d'un modificateur de choc, et de 0,5 à 10 % en poids d'un agent améliorant la viscosité.