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(54) Title: INJECTION FLOW MOLDABLE POLYESTER RESIN, PROCESS FOR MAKING THE SAME AND INJECTION FLOW MOLDED ARTICLE.

(57) Abstract: P.E.T. resin composition used for injection flow molding of molded articles and a process for manufacturing the same.

INJECTION FLOW MOLDABLE POLYESTER RESIN, PROCESS FOR MAKING THE
SAME AND INJECTION FLOW MOLDED ARTICLE.

FIELD OF THE INVENTION

8 The present invention relates to poly (ethylene terephthalate)
polyester resin composition and to a process for manufacturing
the same.

12 In particular it relates to resin compositions which are
suitable for injection flow molding and to the manufacture of
molded articles therefrom.

16 Under Injection Flow Molding in this context is understood a
further development of the current Selective Injection
Compression Molding process (I.C.M.), as taught by Jack Avery,
in the book "Injection Moulding Alternatives, a guide ", 1998,
page 133-136, ISBN 1-56990-8.

20 The standard I.C.M. molding process uses mechanically
controlled moveable mold cores, located inside of a fully
closed and clamped mold, to achieve an in mold compression and
24 still does not overcome the fundamental inherent problem of
process instability and lack of process control.

28 This typically manifests itself in quality issues such as:
part weight variance, wall thickness deviation, low output
efficiency, cavity to cavity part variation, overfilled
overweight parts and poor surface finish at flow end.

32 In Injection Flow Molding however as understood in this
context the process is entirely determined and driven by the

resin melt flow rheology alone, meaning in turn, that the process is inherently self-controlled and self-adjusting.

4 This is in complete contrast to Selective and less
sophisticated I.C.M. methods which are mechanically
controlled, relying on specific external physical parameter
settings, such as pressure, time, distance and velocity, to
8 control the whole process of first injecting a disk like pre-
form and then subsequently compressing it into the final part.

In Injection Flow Molding the thermoplastic resin is injected
12 into a fully closed and clamped mold using injection
conditions of temperature and velocity that are appropriate to
the particular resin type and product type.

16 The injection of the resin melt pressurizes the mold cavity,
which contains a moveable mold part on the opposite side to
the injection side of the cavity. As soon as the cavity
pressure starts to increase, as a direct function of the resin
20 melt viscosity, the mold cavity expands at a rate and to an
extent that is dictated by the resin melt viscosity, as the
material is then more free to follow its natural flow profile
without externally imposed controls.

24
Once the cavity volume expansion starts to offset the increase
in the internal cavity pressure and the cavity pressure then
starts to fall, the pre-pressurized flexible mold part is able
28 to return to close the mold back to its original dimensions
with a resulting compression of the melt.

This compression phase is then, just as with the injection
32 phase, dictated and controlled naturally by the resin melt
behavior. This factor explains why the process is inherently
self-regulated and controlled. As a result variations in melt

viscosity between different cavities or between different shots are automatically corrected for by the material which adapts the in mold process according to any changes in melt viscosity and pressure without any externally applied process adaptations.

Typically the injection flow molded articles consist of thin walled containers featuring good color, optical, mechanical, chemical and thermal properties and have low susceptibility to degradation, for both food and non-food applications, and which require the use of high melt strength and high melt flow polyester resin.

The injection flow molded articles offer significant economic benefits as well as quality improvements with a wide design window.

BACKGROUND OF THE INVENTION

Injection molding in general is a well-established and widespread conversion process for the manufacture of various articles, particularly liquid containers and packaging items, having applications involving both food and non-food contact. It is being used very successfully since many decades.

Both the process and machinery is very well developed and today many companies offer injection molding machines in various sizes and degrees of sophistication, for a wide range of applications, together with appropriate molding compositions. The latter typically consist of polypropylene (P.P.) and polystyrene (P.S.) resins.

Poly (ethylene terephthalate) (P.E.T.) on the other hand is a polymer having outstanding properties.

P.E.T. resins can be semi-rigid to rigid, and are very lightweight. They make good gas and fair moisture barriers, as well as good barriers to alcohol and other solvents, particularly after additional barrier treatment, like for instance metallization, sandwiching with polyvinyl alcohol etc.

They are moreover strong and impact resistant and naturally colorless with a high level of transparency. P.E.T is an excellent material for food contact as it has extremely low levels of absorption and migration. Moreover they can be easily recycled and the base materials re-used.

P.E.T. resins are therefore used extensively in synthetic fibers; beverage, food and other liquid containers; thermoforming applications; and engineering resins often in combination with glass fiber reinforcement.

Depending on their processing and thermal history, they may exist both as an amorphous and as a semi-crystalline polymer.

The amorphous polymer is transparent. Whereas the semi-crystalline material might appear transparent when it has crystal particles in sizes under about 500 nm or opaque and white when it has crystal particle with sizes up to a few microns.

If used for thermoforming applications melt extrusion of sheets subsequently followed by a thermoforming process is necessary, generating nevertheless up to 35 % of wastage which needs to be recycled. This unavoidably results in a considerable increase in overall investment and operating costs and has a negative impact on the environment.

If used in molding applications it exhibits a high melt viscosity and a high tendency to degrade thermally and hydrolytically at elevated temperatures. The latter invariably tend to cause deterioration of its mechanical, chemical and esthetic properties. For that reason it is not being used in common injection molding applications, particularly not for the manufacture of molded articles having wall thicknesses of less than 0,5 mm.

In order to produce molded articles suitable for high temperature applications from P.E.T. resins, a certain degree of crystallinity is required in order to maintain dimensional stability of the article above the glass transition temperature (T_g) of the resin.

16

Typically the melting temperature of P.E.T. resins lies in the range of about 250°C to about 260°C as taught by A.K. van der Vegt, L.E.Govaert in "Polymeren, van keten tot kunststof", ISBN 90-407-2388-5, and the glass transition temperature at about 75°C.

In order to overcome the typical very slow crystallization rate of the resin inorganic as well as organic nucleating agents can be added.

The disadvantage is however that often the resin produced this way has inferior mechanical strength and other desirable properties. Furthermore resins that are converted by common injection processes into molded articles, and that are not oriented such as in the manufacture of bottles or films, tend to crystallize on cooling with the formation of large spherulites. This invariably results in a higher level of opacity of the molded article, which for the application it is

intended to , may be undesirable.

The P.E.T. polymer itself is usually produced in a reactor by
4 forming a pre-polymer first by an esterification reaction
between terephthalic acid and ethylene glycol with water as a
byproduct, and in which monomers and pre-polymers are formed
in the presence of a catalyst. This esterification reaction is
8 then immediately followed by polymerization reaction of the
monomers and pre-polymers through polycondensation, with again
water as the by product.

12 This polymerization reaction through polycondensation is
typically carried out at a temperature range of about 270°C to
about 305°C, and under reduced pressure of less than 10 mbar.

16 In case additives, stabilizers, rheology modifiers,
lubricating agents, nucleating agents etc. are incorporated,
they are usually compounded in only after solid state
polymerization of the mass.

20

This known compounding process however has severe shortcomings
in that it consists of an additional process step which is not
only expensive, time and energy consuming and needs additional
24 equipment, but also exposes the polymer to further degradation
by oxygen, temperature and moisture. It also often results in
the formation of gel particles in the final polymer which, in
turn, tend to affect adversely the color, clarity and
28 mechanical properties of the final product.

Typically, as taught by Ulrich K. Thiele e.a. in "Polyester
bottle resin, production, processing and recycling, pp. 85 ff,
32 Heidelberg, Germany, 2007 ISBN 978-3-9807497-4-9, and by
Stoyko Fakirov in "Handbook of thermoplastic polyesters",
chapter 7 by V.B.Gupta, Z.Bashir, p.320, ISBN 978352730113,

the polyester resin formed has an intrinsic viscosity (I.V.) range of :

- 4 0,40 to 0,70 dl/g for textile fiber grade;
0,72 to 0,98 dl/g for technical applications such as tire cord;
0,60 to 0,70 dl/g for biaxially oriented film;
- 8 0,70 to 1.00 for sheet grade for thermoforming;
0,70 to 0,78 dl/g for flat water bottles grade;
0,78 to 0,85 dl/g for carbonated soft drink bottles grade;
1,00 to 2,00 dl/g for monofilament spinning or engineering
12 plastics.

The standard grades used for making preforms, bottles and sheets typically have a melt flow index (M.F.I.) of 25 g/min
16 at 270°C with a 3,2 kg load.

OBJECTIVES OF THE PRESENT INVENTION

20 It is therefore an objective of the present invention to offer
a solution to the above mentioned and other disadvantages and
shortcomings by providing a process for making polyester resin
which is suitable for injection flow molding of molded
24 articles without degradation of their desirable properties.

It is still a further objective of the present invention to
provide a process for the preparation of injection flow
28 moldable polyester resin that can be directly used without
recurring to an additional process step of blending or
compounding in of additives.

32 It is another objective of the present invention to provide an
injection flow moldable polyester resin that is gel free.

It is yet another objective of the present invention to provide an injection flow moldable polyester resin that has good clarity i.e. low haze, high gloss, good color, and is free from any yellow tinge.

It is still a further objective of the present invention to provide injection flow molded articles that at least locally have a very thin wall thickness.

It is yet another objective of the present invention to provide P.E.T. injection flow molded articles that can be particularly well, though in no way exclusively, used in the high volume packaging market and compete on quality and cost with articles produced by thermoforming of extruded sheet in the thin wall container market.

It is yet another objective of the present invention to provide an injection flow moldable polyester resin which is suitable for making clear high crystalline transparent ovenable trays, such as particularly used for chilled and frozen ready meals, and thin wall hot fill containers, such as particularly used for pasteurizing or sterilizing shelf stable foods.

SUMMARY OF THE INVENTION

In accordance with one aspect of the present invention, there is provided a process for the preparation of injection flow moldable polyester resin compositions, essentially consisting of a poly (ethylene terephthalate-isophthalate) co-polymer, wherein said process comprises the following steps:

a. charging a reactor with a diol-dicarboxylic acid pair, along with co-monomer to obtain a reaction mixture;

b. subjecting said reaction mixture to an esterification reaction yielding a pre-polymer;

4

c. adding to the pre-polymer at least one resin modifying additive and forming a pre-polymer - additive mixture;

8 d. subjecting the pre-polymer-additive mixture to a

polycondensation polymerization reaction at a temperature in the range of about 255 to about 265°C, under vacuum and increased rate of agitation, to obtain a final polyester resin

12 having a melt flow index (MFI) of about 130 g/10 min. to about 140 g/10 min. at 270°C with a load of 3.2 kg, and an intrinsic viscosity (I.V.) of about 0,50 dl/g to about 0,80 dl/g.

16 Under 'diol-dicarboxylic acid pair' is understood a reactive pair of chemical compounds, consisting of a diol and a dicarboxylic acid, typically mono ethylene glycol and therephtalic acid, which react together in an esterifaction
20 reaction to form the structural unit with molecular formula $C_{10}H_8O_4$ from which the main polymer chain is essentially built in a consecutive polymerization reaction based on a polycondensation process.

24

Under 'co-monomer' is understood a reactive chemical compound, either diol or dicarboxylic acid, capable of forming a monomer that is incorporated into the polymer chain by an
28 esterification reaction followed by polymerization through polycondensation, but forms repeat units that are different from those the main chain is essentially constituted of.

32 Under 'pre-polymer' is understood a monomer, oligomer, polyester or co-polyester, or a blend of any of those, having low molecular weight and hence low intrinsic viscosity.

In accordance with another aspect of the present invention there is provided a P.E.T. polyester resin suitable for injection flow molding applications, particularly for the manufacture of molded articles having a local wall thickness not exceeding 300 μ .

In accordance with still another aspect of the present invention there is provided an injection flow molded article in the form of a thin walled container capable of being filled at up to 95°C.

12

In accordance with a further aspect of the present invention there is provided an injection flow molded article in the form of a transparent ovenable tray capable of withstanding up to 210°C for an extended period of time.

DETAILED DESCRIPTION OF THE PRESENT INVENTION

The known methods for making P.E.T. polyester resin employ the compounding approach for incorporation of various additives along with the polyester wherein they are only introduced after solid state polymerization of the latter through polycondensation.

In the present invention however there is provided a process for making injection flow moldable polyester resin wherein the various additives are introduced in the reaction mixture during the monomer, oligomer or pre-polymer formation, when the viscosity of the reaction mixture is still very low.

In accordance with a first aspect of the present invention, there is provided a process for the preparation of injection flow moldable polyester resin, essentially consisting of a

poly (ethylene terephthalate-isophthalate) co-polymer, wherein said process comprises the following steps:

4 a. charging a reactor with a diol-dicarboxylic acid pair, along with co-monomer to obtain a reaction mixture;

b. subjecting the reaction mixture to esterification yielding
8 a pre-polymer;

c. adding to the pre-polymer at least one resin modifying additive forming a pre-polymer - additive mixture;
12

d. subjecting the pre-polymer additive mixture to a polycondensation polymerization reaction at a temperature in the range of about 255 to about 265°C, under vacuum and increased rate of agitation, to obtain a final polyester resin
16 having a Melt flow Index (MFI) of about 130 g/10 min. to about 140 g/10 min. at 270°C with a load of 3.2 kg, and an intrinsic viscosity (I.V.) of about 0,50 dl/g to about 0,80 dl/g.
20

As is known to those skilled in the art, P.E.T. resins are commonly prepared by using either a 'diol-dicarboxylic acid' pair, a 'diol-dicarboxylic ester pair' or a 'diol-P.E.T.' pair
24 as polyester forming materials.

The latter pair is used when considerable amounts of recycled P.E.T. resin are used which need to be glycolyzed first before
28 further processing and building of new resin from it can take place.

In case, as in the present invention, a 'diol-dicarboxylic
32 acid' pair is used, the esterification step b) is typically carried out at a temperature in the range of about 200 to about 300°C.

It is during this esterification reaction or immediately after that according to one important aspect of the present invention useful additives are added to the reaction mixture.

With the consecutive polymerization reaction through polycondensation, catalysts are normally used which are not different from those known by people skilled in the art, for instance at least one catalyst selected from the group consisting of antimony, germanium, titanium, tin, or aluminum compounds.

12

This polymerization reaction, step d), of the reaction mixture through polycondensation is typically carried out at a temperature range of about 290 to about 300°C.

16

It is an essential characteristic of the present invention that the temperature at which the polycondensation reaction is carried out is significantly reduced and adjusted in the range of about 255°C to about 265°C, under vacuum, and with increased rate of agitation.

This has the advantage that less thermal degradation and less side reactions such as branching occur resulting in a final polyester resin that has improved melt flow characteristics and higher mechanical, optical and chemical properties.

It also has improved thermal stability compared with conventionally manufactured P.E.T. resin, which is clearly corroborated by a lesser drop in intrinsic viscosity (I.V.) on molding and by a lower color change, thermal oxidation, hydrolytic degradation, and lower leaching out rate of catalyst and acetaldehyde residues in injection flow molded articles.

The injection flow molded articles made there from also have better color and esthetic appearance.

4

This also has the advantage that lower cycle times and better productivity can be obtained in consecutive injection flow molding processes.

8

They also offer the advantage of better mold release and superior surface properties.

12 Typically for standard P.E.T. grades , for instance used for making pre-forms for bottles or for making sheet, the melt flow index (M.F.I) lies in the range of 25 g/10 min. at 270°C with a 3,2 kg load, whereas the M.F.I. of the polyester
16 resin made according to the invention lies at about 135 g/10 min. under the same conditions. This indicates that there is over a 500 % increase of flow compared with that of a conventional P.E.T. resin of corresponding intrinsic viscosity
20 (I.V.).

One of the important benefits of the present invention is that useful materials in the form of additives are added at the
24 melt polymerization stage c) which largely prevents the formation of gels in the final polymer or the need for addition of those additives in a subsequent compounding operation. At the same time avoiding additional costs,
28 investments and adverse environmental effects.

It also makes it possible to introduce crystallization growth controlling segments into the polymer chain which , together
32 with suitable nucleating agents added as a solution or a slurry in a co-monomer prior to the polyester forming process step c).

It also offers the possibility to control the crystallization speed and the size of the crystals formed, so that molded articles can be produced by injection flow molding techniques that have thin side walls while still being capable of withstanding hot fill or retort applications or remain transparent and withstand hot oven conditions for an extended period of time.

It also has the advantage that the P.E.T. resin according to the present invention can be supplied in crystallized form, so that it can be dried at about 160°C to about 180°C without hydrolytic degradation taking place.

The investment and operating costs of a sheet extrusion line, and expensive thermoforming line, can be saved because they are no longer needed.

DESCRIPTION OF PREFERRED EMBODIMENTS

In accordance with one embodiment of the present invention the dicarboxylic acid of the diol-dicarboxylic acid pair of step a) is at least one selected from the group consisting of terephthalic acid and isophthalic acid, and the diol is monoethylene glycol.

In accordance with another embodiment of the present invention about 4 wt% to about 20 wt% of terephthalic acid is replaced by isophthalic acid.

This had the advantage of forming repeat units in the polymer chain that work as a flow enhancers lowering the melting point of the resin from about 250°C to about 210°C and improving the flow characteristics at the same time.

A shorter cycle time during injection flow molding invariable results in economic benefits and higher optical and mechanical properties of the molded article due to less exposure to thermal and hydrolytic degradation.

In a further embodiment of the present invention the co-monomer in step a) is at least one dicarboxylic acid selected from the group consisting of malonic-, succinic-, glutaric- and adipic acid, and is present in a concentration in the range of about 0,5 wt% to about 2,0 wt% in respect of the mass of the reaction mixture.

The structural units that are incorporated in the main chain this way by the consecutive esterification and polymerization reaction are capable of cooperating with specific nucleating agents to control the crystallization rate and the size of the crystals formed in the final product.

According to another embodiment of the present invention the co-monomer in step a) is at least one diol selected from the group consisting of di-ethylene glycol (DEG), poly-ethylene glycol having a molecular weight up to 4000, neo-pentylglycol (N.P.G.) 1,3- and 1,4-cyclohexane dimethanol (1,3 C.H.D.M. and 1,4 C.H.D.M.), 1,4-butanediol, and is present in a concentration up to 20 wt% in respect to the mass of the reaction mixture.

In yet another embodiment of the present invention the resin modifying additive in step c) is at least one stabilizer selected from the group consisting of phosphoric acid, aliphatic phosphoric acid compounds and phosphites, and is incorporated in a concentration of up to 6000 ppm.

In yet another preferred embodiment of the present invention the resin modifying additive in step c) is at least one internal lubricating agent selected from the group consisting of fatty acid esters, synthetic waxes and micronized waxes, particularly alkanes with 50 to 100 carbon atoms, silicone fluids, and is incorporated in a concentration of up to 1 wt%.

In still another preferred embodiment of the present invention the resin modifying additive in step c) is at least one nucleating agent selected from the group consisting of:

A. solids, particularly silicon dioxide, talc, clay particles;

B. alkali and alkaline metal earth salts of aromatic carboxylic acids, particularly sodium benzoate, sodium salicylate;

C. salts of aliphatic acids, particularly sodium acetate,

and that has a range size of about 20 nm to about 10 μ . It is added in a concentration ranging from about 0,1 wt% to about 1,0 wt%.

It was surprisingly found, as a main aspect of the present invention, that these nano-sized nucleating agents can be very well dispersed the P.E.T. matrix and then act synergistically with the co-monomers mentioned above, in particular gluconic, succinic, malonic and adipic acid, which can interfere locally with the large scale crystallization of P.E.T.

They obviously allow the production of very large numbers of small sized spherulites that do not interfere with the light transmittance and also do not interfere with the mechanical

and other desirable properties of the resin or the injection flow molded article.

4 In practice the co-monomers mentioned above are melt mixed with the P.E.T raw materials and the nano-sized nucleating agents mentioned above in the esterification phase of the manufacturing process, i.e during formation of pre-polymer,
8 and before polymerization through polycondensation.

The requirements of high crystalline resin combined with high transparency, high heat resistance, good mechanical and
12 optical properties, as well as good flow are particularly common for the P.E.T. grades of the present invention successfully used for instance for injection flow molding of hot fill containers and ovenable trays.

16

In still another preferred embodiment of the present invention the resin modifying additive in step c) is at least one flow promoter selected from the group consisting of finely
20 dispersed silica, micronized graphite particles, metallic and non-metallic stearates, particularly calcium stearate, zinc stearate, butyl stearate, and is suspended or dissolved in at least one diol before being added to the mixture in a
24 concentration of up to 5 wt%.

In accordance with a further embodiment of the present invention an injection flow molded article is provided
28 consisting of a parison, a container, a film, a tube, or a tray.

In accordance with yet another preferred embodiment of the present invention an injection flow molded article is provided that has at least a local wall thickness of less than
32 300 μ .

In a further preferred embodiment of the present invention an injection flow molded article formed is provided having a thin wall and capable of being hot filled at about 95°C without dimensional deterioration.

In still a further preferred embodiment of the present invention an injection flow molded article is provided consisting in a transparent ovenable tray that can withstand up to 210°C.

In a further preferred embodiment of the present invention thin wall containers are provided produced in multi-cavity molds with typical wall thicknesses of 300 μ by injection flow molding of high flow P.E.T. grades having a Melt Flow Index (I.M.F.) of 90 g/10 min., (at 270 °C, under 2.16 kg load).

16

In a further preferred embodiment of the present invention Injection flow molded containers with a wall thicknesses of 200 μ are provided using a P.E.T. grade with a Melt Flow Index (I.M.F.) of 120 g/10 min., (at 270 °C, under 2.16 kg load).

20

In a further embodiment, according of the present invention, injection flow molded containers are provided that are heat sealed with lids made from a high flow P.E.T grade of different formulation in such a way as to allow the lid to be subsequently removed by peeling the seal area manually.

24

In still another preferred embodiment of the present invention said sealable lid is designed with clips that allow the lid to be clipped back onto the container, after opening, to act as a re-closable lid.

32

In a further embodiment, according to the present invention, injection flow molded high heat resistant crystallized

containers, resistant up to 220 °C, are provided that are suitable for chilled and frozen ready meals.

4 In a further embodiment, according to the present invention, injection flow molded high heat resistant crystallized transparent thin wall ovenable containers are provided, resistant up to 220 °C, and in which transparency is achieved
8 by designing the crystal wall structure so that the crystal size is kept smaller than the wavelength of visible light so as not to scatter the light.

12 In a further embodiment, according to the present invention, transparent high oxygen barrier injection flow molded containers are provided that suitable for packaging shelf stable processed foods, in which the foods are aseptically
16 processed either by pasteurizing or sterilizing using Ohmic heating and filled under clean, ultra clean or aseptic conditions, and that have an oxygen barrier with a maximum oxygen ingress of 1 ppm oxygen per year.

20 In a further embodiment, according to the present invention, transparent high oxygen barrier injection flow molded containers are provided that are coated with an aqueous 0.1 μ
24 SiO₂ coating in the mold, and in which the coating prevents bacterial growth on the container surface, not requiring chemical sterilization of the containers anymore immediately prior to the entrance of the filling machine.

28 In still a further embodiment, according to the present invention, both transparent and opaque high oxygen barrier injection flow molded containers are provided that are
32 suitable for hot filling and pasteurizing high acid shelf stable foods at 100 °C.

In still a further embodiment, according to the present invention, both transparent and opaque high oxygen barrier injection flow molded P.E.T. containers are provided that are
4 suitable for hot filling and sterilizing low acid shelf stable foods at 121 °C.

In yet a further embodiment, according to the present
8 invention, reusable washable injection flow molded beer glasses are provided that can subsequently pass the washing cycle of 90 °C for 2 minutes as typically used for cleaning beer glasses.

12

In yet another preferred embodiment of the present invention said glasses are coated in the with a 0.1 μ SiO₂ coating and rendered scratch resistant for the lifetime of the glasses.

16

In a further embodiment, according to the present invention, single use injection flow molded beer containers are provided which contain the CO₂ level in the beer over a 4 month shelf
20 life and which have a fully peel off lid allowing the consumer to drink directly from the wide mouth container.

In a further embodiment, according to the present invention,
24 high stretch ratio thin wall P.E.T. injection flow molded preforms are provided which are suitable for manufacturing of ultra-light weight 0.5 liter carbonated soft drink bottles which include a step design for manufacturing a champagne base
28 containing a thin walled gate.

In yet another embodiment, according to the present invention, high barrier, creep resistant injection flow molded aerosol
32 containers are provided which exhibit increased heat resistant to pass the 50 °C standard pressure test.

In still a further embodiment, according to the present invention, transparent and opaque injection flow molded squeezable tubes are provided by using a high flow soft P.E.T.grade.

The present invention is in no way limited to the form of embodiment described by way of an example, however, such an invention of a process for making injection flow moldable polyester resin, the resin composition produced herewith, and the molded articles made from it by injection flow molding techniques can be realized in various ways or forms without leaving the scope of the invention.

All the compositions and methods disclosed and claimed herein can be made and executed without undue experimentation in the light of the present disclosure.

While the compositions and methods of his invention have been described in terms of preferred embodiments it will be apparent to those skilled in the art that variations may be applied to the compositions and the steps or in the sequence of the steps of the method described herein without departing from the concept, spirit and scope of the invention.

More specifically, it will be apparent that certain agents which are both chemically and physiologically related may be substituted for the agents described herein while the same or similar results would be achieved. All such similar substitutes and modifications apparent to those skilled in the art are deemed to be within the spirit, scope and concept of the invention as defined by the appended claims.

32

CLAIMS

What is claimed is:

4

1.- A process for the preparation of injection flow moldable polyester resin, essentially consisting of a poly (ethylene terephthalate-isophthalate) co-polymer, characterized in that
8 said process comprises the following steps:

12

a. charging a reactor with a diol-dicarboxylic acid pair, along with co-monomer to obtain a reaction mixture;

b. subjecting the reaction mixture to esterification yielding a pre-polymer;

16

c. adding to the pre-polymer at least one resin modifying additive forming a pre-polymer - additive mixture;

20

d. subjecting the pre-polymer additive mixture to a polycondensation polymerization reaction at a temperature in the range of about 255 to about 265°C, under vacuum and increased rate of agitation, to obtain a final polyester resin having a melt flow index (MFI) of about 130 g/10 min. to about
24 140 g/10 min. at 270°C with a load of 3.2 kg, and an intrinsic viscosity (I.V.) of about 0,50 dl/g to about 0,80 dl/g.

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2.- The process of claim 1, characterized in that the dicarboxylic acid is at least one selected from the group consisting of terephthalic acid and isophthalic acid, and the diol is monoethyleneglycol.

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3.- The process of claim 2, characterized in that about 4 wt% to about 20 wt% of terephthalic acid is replaced by isophthalic acid.

4.- The process of claim 1, characterized in that said co-
monomer is at least one dicarboxylic acid selected from the
4 group consisting of malonic-,succinic-, glutaric-, adipic acid
and is present in a concentration in the range of about 0,5
wt% to about 2,0 wt% in respect of the mass of the reaction
mixture.

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5.- The process of claim 1, characterized in that said co-
monomer is at least one diol selected from the group
consisting of di-ethylene glycol (DEG), poly-ethylene glycol
12 having a molecular weight up to 4000, neo-pentylglycol
(N.P.G.) 1,3- and 1,4-cyclohexane dimethanol (1,3 C.H.D.M. and
1,4 C.H.D.M.), 1,4-butanediol, and is present in a
concentration up to 20 wt% in respect to the mass of the
16 reaction mixture.

6.-The process of claim 1, characterized in that said resin
modifying additive is at least one stabilizer selected from
20 the group consisting of phosphoric acid , aliphatic phosphoric
acid compounds and phosphites, and is present in a
concentration of up to 6000 ppm.

24 7.- The process of claim 1, characterized in that said resin
modifying additive is at least one internal lubricating agent
selected from the group consisting of fatty acid esters,
synthetic waxes and micronized waxes, particularly alkanes
28 with 50 to 100 carbon atoms, silicone fluids, and is present
in a concentration of up to 1 wt%.

8.- The process of claim 1, characterized in that said resin
32 modifying additive is at least one nucleating agent selected
from the group consisting of:

A. solids, particularly silicon dioxide, talc and clay particles;

4 B. alkali and alkaline metal earth salts of aromatic carboxylic acids, particularly sodium benzoate, sodium salicylate;

8 C. salts of aliphatic acids, particularly sodium acetate,

and has a range size of about 20 nm to about 10 μ , and is added in a concentration ranging from about 0,1 wt% to about
12 1,0 wt%.

9.- The process of claim 1, characterized in that said resin modifying additive is at least one flow promoter selected from
16 the group consisting of finely dispersed silica, micronized graphite particles, metallic and non-metallic stearates, particularly calcium stearate, zinc stearate, butyl stearate, and is suspended or dissolved in at least one co-monomer diol
20 in a concentration of up to 5 wt%.

10.- An injection flow moldable polyester resin essentially consisting of poly (ethylene terephthalate- isophthalate) co-
24 polymer, characterized in that it has a melt flow index (MFI) of about 130 g/10 min. to about 140 g/10 min. at 270°C with a load of 3.2 kg, and an intrinsic Viscosity (I.V.) of about 0,50 dl/g to about 0,80 dl/g.

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11.- An injection flow molded article formed from the co-polymer resin composition as claimed in claim 10, characterized in that the molded article is at least one
32 selected from the group consisting of a parison, a container, a film, a tube, a tray.

12.- An injection flow molded article formed from the co-
polymer resin as claimed in claim 10, characterized in that
4 the molded article has at least a local wall thickness of less
than 300 μ .

13.- An injection flow molded article formed from the co-
8 polymer resin as claimed in claim 10, characterized in that
the molded article is a thin wall hot fill container capable
of being filled at about 95°C.

12 14.- An injection flow molded article formed from the co-
polymer resin as claimed in claim 10, characterized in that
the molded article is a transparent ovenable tray that can
withstand up to 210°C.

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