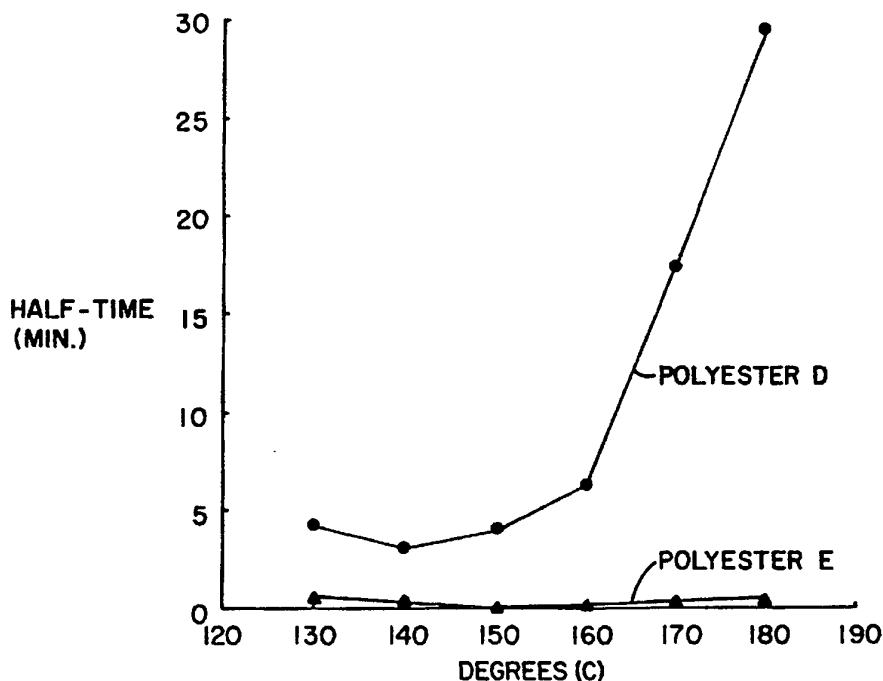




## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification<sup>6</sup> : C08J 5/18, B32B 27/36, C08G 63/199, B65D 65/40</p>	A1	<p>(11) International Publication Number: <b>WO 95/09885</b> (43) International Publication Date: 13 April 1995 (13.04.95)</p>
<p>(21) International Application Number: PCT/US94/11348 (22) International Filing Date: 6 October 1994 (06.10.94) (30) Priority Data: 133,550 7 October 1993 (07.10.93) US (71) Applicant: EASTMAN CHEMICAL COMPANY [US/US]; 100 North Eastman Road, Kingsport, TN 37660 (US). (72) Inventors: MERCER, James, Wilson, Jr.; 1033 Hanover Court, Kingsport, TN 37660 (US). BELL, Emily, Tedrow; 182 Harmony Acres Drive East, Jonesborough, TN 37659 (US). SHIH, Wayne, Keng; 1032 Norfolk Place, Kingsport, TN 37660 (US). (74) Agent: BOSHEARS, Betty, Joy; P.O. Box 511, Kingsport, TN 37662-5075 (US).</p>		<p>(81) Designated States: AU, CA, CN, JP, KR, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>

(54) Title: POLYESTER FILM



(57) Abstract

Monolayer and multilayer polyester film which comprises at least one layer of a heat-sealing polyester which is crystallizable to the extent of allowing mixtures thereof to be made with other crystallizable polyesters for crystallization without sticking is disclosed. The heat-sealing polyester is polyethylene terephthalate modified with about 7-15 ml. % cyclohexanedimethanol. In multilayered films, another layer of readily crystallizable polyethylene terephthalate is used.

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	GB	United Kingdom	MR	Mauritania
AU	Australia	GE	Georgia	MW	Malawi
BB	Barbados	GN	Guinea	NE	Niger
BE	Belgium	GR	Greece	NL	Netherlands
BF	Burkina Faso	HU	Hungary	NO	Norway
BG	Bulgaria	IE	Ireland	NZ	New Zealand
BJ	Benin	IT	Italy	PL	Poland
BR	Brazil	JP	Japan	PT	Portugal
BY	Belarus	KE	Kenya	RO	Romania
CA	Canada	KG	Kyrgystan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SI	Slovenia
CI	Côte d'Ivoire	LI	Liechtenstein	SK	Slovakia
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CN	China	LU	Luxembourg	TD	Chad
CS	Czechoslovakia	LV	Latvia	TG	Togo
CZ	Czech Republic	MC	Monaco	TJ	Tajikistan
DE	Germany	MD	Republic of Moldova	TT	Trinidad and Tobago
DK	Denmark	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	US	United States of America
FI	Finland	MN	Mongolia	UZ	Uzbekistan
FR	France			VN	Viet Nam
GA	Gabon				

- 1 -

POLYESTER FILMTechnical Field

The present invention relates to polyester film, both monolayer and multilayer. The film comprises at least one layer of a heat-sealing polyester which is crystallizable to the extent of allowing mixtures thereof to be made with other crystallizable polyesters for crystallization without sticking. The heat-sealing polyester is polyethylene terephthalate modified with about 7-15 mol % cyclohexanedimethanol. In multilayered films, another layer of readily crystallizable polyethylene terephthalate (PET) is used.

Background of the Invention

It is known in the art that amorphous polyesters can be extruded with crystallizable polyesters to improve performance of the film. Performance improvements over monolayer crystallizable polyester films include high temperature sealing, solvent sealing, and lower cutting force during trimming. Crystallizable polyesters used as a heat-sealing layer often results in poor performance. Amorphous polyester resins cause problems when being handled as waste material (hereinafter sometimes called "regrind") which is generated during processing. When mixed with crystalline polyester pellets and dried at a temperature above the glass transition temperature of the amorphous polyester, the regrind softens and sticks together, forming large clumps. These clumps make drying of the regrind very difficult and cause significant problems with air flow in a dryer and also when feeding to an extruder.

Some techniques for overcoming this sticking problem include melting the regrind with crystallizable

- 2 -

polyesters using devices specially equipped for this process and then reforming the extrudate into pellets that can later be thermally crystallized. Although this technique may alleviate the sticking problems, the cost can be relatively expensive.

This invention provides a polyester composition which offers advantages due to its heat-sealability, while at the same time crystallize to sufficient levels to allow mixing and drying with crystallizable polyesters.

Polyesters containing repeat units from terephthalic acid, ethylene glycol and cyclohexanedimethanol are known in the art. For example, see U. S. Patent Nos. 4,373,002; 4,091,150; 4,405,400; 4,011,358; 4,765,999; 4,399,179; 4,946,743; 4,375,494 and 2,901,466. Some of the films disclosed in these patents are oriented. In others, the heat-sealing layer is not as described and claimed herein. For example, U. S. Patent No. 4,765,999 discloses a dual layered film wherein the heat-sealing layer is a polyester having repeat units from terephthalic acid, ethylene glycol and 1,4-cyclohexanedimethanol. This heat-sealing polyester contains a greater number of repeat units from 1,4-cyclohexanedimethanol than claimed herein, making the heat-sealing layer too difficult to crystallize and thus, would result in sticking when mixed with other polyesters during crystallization.

Japanese Patent 62,222,845 discloses a laminated heat-sealable polyester film comprised of (1) a crystallizable PET containing up to 10 mol % of comonomer that has a heat of fusion of at least 7 cal/g and (2) a PET copolymer containing 10-20 mol % isophthalic acid that has a heat of fusion of up to 5 cal/g. The film is oriented and used for pouches for boiling food for sterilization. An important relation

- 3 -

between the layers is the refraction index. A preferred range is disclosed for optimum properties. This patent does not address regrind nor the crystallizability of the second layer. Furthermore, it does not mention the use of PET copolymers containing 1,4-cyclohexane-  
5 dimethanol.

Japanese Patent 60,253,545 discloses a laminated polyester film for shrink-packaging comprised of (1) a copolyester modified with 5 to 50 mol % 1,4-cyclohexane-  
10 dimethanol and (2) PET. The total thickness of the copolyester layer is 20-70% of the entire structure. An important property of the film is shrinkage. Again, this patent does not address regrind nor the crystallizability of the copolyester layer.

15

#### Detailed Description of the Drawings

Fig. 1 - A graphical representation of a plot of half-times of crystallization versus temperature.

Fig. 2 - A graphical representation of a plot of half-times of crystallization versus level of 1,4-cyclohexanedimethanol.  
20

Fig. 3 - A graphical representation of a plot of half-times of crystallization versus temperature.

#### Description of the Invention

According to one embodiment of this invention, there is provided a non-oriented film of an amorphous, heat-sealing, slowly crystallizable polyester consisting essentially of repeat units from terephthalic acid,  
30 about 85-93 mol % ethylene glycol and about 15-7 mol % cyclohexanedimethanol.

According to another embodiment of the invention, there is provided a non-oriented film comprising

a) an amorphous, readily crystallizable layer of  
35 polyester which is essentially polyethylene

- 4 -

terephthalate having a melting point greater than about 238°C, a melt heat of fusion of greater than about 9 cal/g as measured by a differential scanning calorimeter using a scan rate of about 20°C/min, an I.V. of about 0.50 dL/g to about 1.00 dL/g, and

5

b) an amorphous, heat-sealing layer of slowly crystallizable polyester having an I.V. of about 0.50 dL/g to about 0.90 dL/g and

10

consisting essentially of repeat units from terephthalic acid, about 85-93 mol % ethylene glycol and about 15-7 mol % cyclohexane-dimethanol, the total thickness of slowly crystallizable polyester being less than about

15

50% of the total thickness of said film.

The monolayer film described above is useful in that it is readily heat-sealable to various substrates such as paper, paperboard, plastic, metal and wood, but at the same time is slowly crystallizable such that it

20

can be reground or chopped into particles and crystallized with virgin material to an extent that sticking of particles is not a problem. It is useful as packaging material, such as in fabricated boxes, food trays and blister packaging. The film may be used as

25

such or may be molded, thermoformed, or the like to make articles such as trays. Also, it may be extruded or coextruded directly into mold cavities. It has been discovered that the properties of heat-sealability and crystallizing ability can be carefully balanced by

30

providing a polyester consisting essentially of repeat units from terephthalic acid, about 85-93 mol % ethylene glycol and about 15-7 mol % cyclohexanedimethanol. Below about 7 mol % cyclohexanedimethanol, the polyester becomes too readily crystallizable, and heat-sealability

35

is adversely affected. Above about 15 mol %

- 5 -

cyclohexanedimethanol, the polyester becomes too difficult to crystallize, and has a tendency to stick when crystallization with other polyester particles is attempted. For example, polyethylene terephthalate modified with 3.5 mol % 1,4-cyclohexanedimethanol is too readily crystallizable and heat-sealability is poor. On the other hand, polyethylene terephthalate modified with about 30 mol % 1,4-cyclohexanedimethanol is so difficult to crystallize, sticking problems are encountered when crystallization with other polyesters is attempted.

For the purposes of this invention, the term "readily crystallizable" is preferably defined as when the crystallization half-time from the glassy state is in the range of 2 minutes, more preferably 1 minute, or less.

The term "slowly crystallizable" is preferably defined as when the crystallization half-time from the glassy state is in the range of 2 minutes or greater, preferably 3 minutes or greater.

The technique for following the rate of crystallization consists primarily in following the increase in depolarization of plane-polarized light by the polyester. The method used in this invention is primarily that shown in "A New Method for Following Rapid Rates of Crystallization", I. Poly (hexamethylene adipamide), J. H. Magill, Polymer, Vol. 2, page 221-233 (1961) with the exception that Magill uses a polarizing microscope as the source of light and light-collection lenses. In measuring the crystallization half-times of the present invention, a helium-neon laser [with a small angle light scattering technique (SALS)] was used as was shown by Adams and Stein in J. Polymer Sci. A2, Vol. 6 (1962).

Crystallization from the "glass or glassy state" is a term well-known in the art. For instance, it is

- 6 -

discussed in J. Polymer Science, Vol. 118, p. 334 (1980)  
by R. S. Stern and A. Misra.

Crystallization half-times are measured at the time  
in which the transmitted intensity is half of the  
5 maximum intensity achieved.

The method used is generally as follows:

- (1) Melt the sample to remove existing  
crystallinity;
- (2) Bring the temperature of the sample polyester  
10 to a temperature below the glass transition  
temperature (crystallization from the glass or  
glassy state);
- (3) Crystallize the sample polyester at a pre-  
determined temperature;
- 15 (4) Record the transmitted light intensity plotted  
versus time;
- (5) Find the time at which the transmitted  
intensity is half of the maximum intensity  
achieved.

20 In multilayered structures, the layer which is  
essentially polyethylene terephthalate provides  
strength, support, and potentially lower cost to the  
film. By "essentially polyethylene terephthalate", it  
is meant polyethylene terephthalate homopolymer or a  
25 copolymer having up to a total of about 10 mol % repeat  
units from one or more other conventional dicarboxylic  
acids, glycols, or combinations thereof. Included as  
examples of the more conventional acids are terephthalic  
acid, isophthalic acid, naphthalene dicarboxylic acid,  
30 cyclohexanedicarboxylic acid and the like or their alkyl  
esters. Included as examples of the more conventional  
glycols are diethylene glycol, butanediol, hexanediol,  
neopentyl glycol, etc.

The cyclohexanedimethanol may be 1,4- or  
35 1,3-isomers and may be cis, trans, or a mixture thereof.

- 7 -

The polyesters may be mixtures of polyesters achieving the aforementioned level of cyclohexanedimethanol.

The readily crystallizable polyester and the heat-sealing polyester are produced by esterification and polycondensation techniques well known in the art. By the term "polyester", we intend to include copolyester.

The "crystallizable polyester", as defined by this invention, is further characterized by having a melting point greater than about 238°C and a melt heat of fusion of greater than about 9 cal/g as measured by a differential scanning calorimeter, using a scan rate of about 20°C/min. The monolayer film may be produced by conventional extrusion or casting techniques. The multilayered films may be produced by conventional coextrusion, lamination, or the like. The readily crystallizable layer may have a layer of the heat-sealable layer applied to one or both sides. The film may be of any convenient thickness, but total thickness will normally be between about 5 and about 50 mil. Normally, the heat-sealing polyester in multilayered films will account for about 5-50% of the total thickness of the film.

#### Examples

The following examples are submitted for a better understanding of the invention.

In the examples, "Polyester A" is polyethylene terephthalate modified with about 1.5 mol % 1,4-cyclohexanedimethanol, a readily crystallizable copolyester having an I.V. of about 0.76 dL/g. "Polyester B" is a copolyester having repeat units from terephthalic acid, about 90 mol % ethylene glycol and about 10 mol % 1,4-cyclohexanedimethanol, a slowly crystallizable copolyester having good heat sealability and an I.V. of about 0.68 dL/g. "Polyester C" is a crystallizable

- 8 -

polyester having an I.V. of about 0.76 dL/g, and having repeat units from terephthalic acid, 96.5 mol % ethylene glycol and 3.5 mol % 1,4-cyclohexanedimethanol.

"Polyester D" is essentially non-crystallizable, has an I.V. of 0.75 dL/g and has repeat units from terephthalic acid, about 70 mol % ethylene glycol and about 30 mol % 1,4-cyclohexanedimethanol. "Polyester D" is a crystallizable polyester having repeat units from terephthalic acid, 88 mol % ethylene glycol and 12 mol % 1,4-cyclohexanedimethanol, "Polyester E" is a crystallizable polyester having repeat units from terephthalic acid, 98.5 mol % ethylene glycol and 1.5 mol % 1,4-cyclohexanedimethanol. "Polyester F" is a crystallizable polyester having repeat units from terephthalic acid, 84.9 mol % ethylene glycol and 15.1 mol % 1,4-cyclohexanedimethanol. "Polyester G" is a crystallizable polyester having repeat units from terephthalic acid, 87.6 mol % ethylene glycol and 12.4 mol % 1,4-cyclohexanedimethanol. "Polyester H" is crystallizable polyester having repeat units from terephthalic acid, 89.3 mol % ethylene glycol and 10.7 mol % 1,4-cyclohexanedimethanol.

The first 11 examples were prepared with a 3.5" Welex (main) extruder and a 1.5" Davis Standard (satellite) extruder in conjunction with a 3 layer Dow feed block.

Example 1 - A 10 mil film was extruded from Polyester A. One mil (0.001") of Polyester B was coextruded on each side of the 10 mil structure.

A control sample was also prepared by coextruding 1 mil of Polyester C on each side of the 10 mil film.

The film structures were then evaluated for high temperature heat sealing properties. The test consisted of sealing films to themselves using a heated bar at

375°F for 2 seconds with 60 psi bar pressure. Samples were cut to produce a one inch by 4 inch test specimen. Bond strengths were determined using an Instron tensile testing machine to pull the bonded specimens at a 180°  
5 angle. The results, measured in grams/mm, unexpectedly indicated about a 30 % improvement in peel strength with samples made from the coextruded modified polyester as compared to the control (125.3 g/mm versus 96.1 g/mm).

Example 2 - An 8-mil film was extruded from  
10 Polyester A. Two mils (0.002") of Polyester B, as described in Example 1, were coextruded on each side of the 8-mil structure.

A control sample was also prepared by coextruding 2 mils of Polyester C on each side of the 8-mil film.

15 The films were heat sealed and tested as described in Example 1. The coextruded modified copolyester, unexpectedly, had about a 50% higher peel strength as compared to the control sample (207.6 g/mm versus 137.3 g/mm).

20 Example 3 - A 6-mil film was extruded from Polyester A. Three mils (0.003") of Polyester B, as described in Example 1, were coextruded on each side of the 6-mil structure.

A control sample was also prepared by coextruding  
25 3 mils of Polyester C on each side of the 6-mil film.

The films were heat sealed and tested as described in Example 1. The coextruded modified copolyester, surprisingly, had about a 290% higher peel strength as compared to the control sample (193.4 g/mm versus  
30 49.5 g/mm). This significant increase in peel strength was totally unexpected.

Example 4 - Films were coextruded as described in Example 1. A control sample was also prepared as described in Example 1.

- 10 -

The film structures were then evaluated for high temperature heat sealing properties. The test consisted of sealing the films to 10 mil substrates made from Polyester D using a heated bar at 375°F for 2 seconds with 60 psi bar pressure. Samples were cut to produce a one-inch by 4-inch test specimen. Bond strengths were determined using an Instron tensile testing machine to pull the bonded specimens at a 180° angle. The results, measured in grams/mm, unexpectedly indicated about a 32% improvement in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (141.4 g/mm versus 107.3 g/mm).

Example 5 - Films were coextruded as described in Example 2. A control sample was also prepared as described in Example 2. The films were heat sealed and tested as described in Example 4. The results unexpectedly showed about a 36% increase in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (136.5 g/mm versus 100.4 g/mm).

Example 6 - Films were coextruded as described in Example 3. A control sample was also prepared as described in Example 3. The films were heat sealed and tested as described in Example 4. The results surprisingly showed about a 327% increase in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (168.8 g/mm versus 39.5 g/mm). This significant increase in peel strength was totally unexpected.

Example 7 - A 10-mil film was extruded from Polyester A, a crystallizable polyester. One mil (.001") of Polyester B was coextruded on each side of

- 11 -

the 10-mil film. A control sample was also prepared as described in Example 1.

The films were heat sealed and tested as described in Example 1. The results unexpectedly showed about a  
5 55% increase in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (148.8 g/mm versus 96.1 g/mm).

Example 8 - A 6-mil film was extruded from  
10 Polyester A, a crystallizable polyester. Three mils (0.003") of Polyester B was coextruded on each side of the 6-mil film. A control sample was also prepared as described in Example 3.

The films were heat sealed and tested as described  
15 in Example 1. The results unexpectedly showed about a 353% increase in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (223.9 g/mm versus 49.5 g/mm).

Example 9 - A 10-mil film was extruded from  
20 Polyester A. One mil (0.001") of a modified copolyester, containing about 100 mol % terephthalic acid, about 85 mol % ethylene glycol and about 15 mol % 1,4-cyclohexanedimethanol, was coextruded on each side  
25 of the 10-mil film. A control sample was also prepared as described in Example 1.

The films were heat sealed and tested as described  
in Example 4. The results unexpectedly showed about a  
30 99% increase in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (213.5 g/mm versus 107.3 g/mm).

Example 10 - A 6-mil film was extruded from  
Polyester A. Three mils (0.003") of a modified  
35 copolyester, as described in Example 9, was coextruded

- 12 -

on each side of the 6-mil film. A control sample was also prepared as described in Example 3.

The films were heat sealed and tested as described in Example 4. The results unexpectedly showed about a  
5 252% increase in peel strength with samples made from the coextruded modified polyester, as described in this invention, as compared to the control (139.0 g/mm versus 39.5 g/mm).

Example 11 - Film samples as prepared in Example 1,  
10 with a control sample consisting of Polyester A, coextruded with an amorphous copolyester (Polyester D), were put through a regrind operation for the purpose of conducting drying evaluations. The regrind had a bulk density of 15-22 lb/ft<sup>3</sup>, compared to 55 lb/ft<sup>3</sup> for  
15 virgin crystallizable polyester pellets, which have already been crystallized. Regrind from each film sample was tumble blended with the virgin crystallizable polyester pellets, using a 50/50 weight ratio of regrind to pellets in the mixture. A ConAir crystallizer/dryer  
20 hopper utilizing desiccant air at 300°F was used to evaluate the drying and recrystallizing of the amorphous regrind mixed with virgin crystallizable polyester pellets. The evaluation unexpectedly showed that modified copolyesters that show a melting heat of fusion  
25 greater than 0.5 cal/g (as measured in a DSC at 20°C/min) will not stick in a dynamic dryer set at crystalline polyester drying temperatures. The highly modified control regrind sample, which showed no melting heat of fusion, stuck together and prevented plug flow  
30 of the material through the hopper, which is highly undesirable.

Example 12 - Single layer films were prepared using a 3.5" Welex extruder and a 3 roll stack nip polish take-up unit. The films were 50 mils (0.050") in  
35 thickness and consisted of the modified copolyester,

- 13 -

containing about 100 mol % terephthalic acid, about 88 mol % ethylene glycol and about 12 mol % 1,4-cyclohexanedimethanol, and Polyester C as the control.

5           An internal test has been developed to measure the force and energy required to cut through a plastic film sample. An Instron tensile/compression testing machine has been outfitted with a compression load cell and a jig to hold a typical steel rule blade. The blade is  
10 pushed into the plastic while the force is monitored. Results of this test unexpectedly showed that it took about 9% less force and about 16% less energy for the modified polyester to be cut, as compared to the control (753 lb versus 826 lb and 1.51 ft-lb versus 1.79 ft-lb).

15           Example 13 - The samples for Polyesters D-H were prepared as in Example 1.

(1) The samples were melted to remove the existing crystallinity. The polymer samples were placed between two microscope cover glasses (thin slips of glass  
20 approximately 1 inch square) on a temperature-controlled hot plate for a pre-determined time, usually a minute or two;

(2) The samples were quenched to the glassy state (below the glass transition temperature rapidly enough  
25 to prevent crystallization). Using forceps, the sample-cover glass sandwich, was rapidly transferred to a metal block which is at room temperature;

(3) The samples are crystallized at a pre-determined temperature. The equipment or "sandwich" of  
30 (2) is transferred to the heated sample holder;

(4) The data is collected by recording the transmitted light intensity versus time;

(5) The data is analyzed. The time is recorded at which the transmitted intensity is half the maximum

- 14 -

intensity achieved. This is a measure on crystallization rate;

(6) Repeat for different temperatures as shown in Figures 1-3.

5           Inherent viscosity (I.V.) is determined herein using 0.50 grams of polymer per 100 mL of a solvent consisting of 60% by weight phenol and 40% by weight tetrachloroethane.

10           The invention has been described in detail with particular reference to preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention.

- 15 -

CLAIMS

We claim:

- 5 1. A non-oriented film of an amorphous, heat-sealing, slowly crystallizable polyester consisting essentially of repeat units from terephthalic acid, about 85-93 mol % ethylene glycol and about 15-7 mol % cyclohexanedimethanol.
- 10 2. A non-oriented layered film comprising
- 15 a) an amorphous, readily crystallizable layer of polyester which is essentially polyethylene terephthalate having a melting point greater than about 238°C and a melt heat of fusion of greater than about 9 cal/g as measured by a differential scanning calorimeter using a scan rate of about 20°C/min, and
- 20 b) an amorphous, heat-sealing layer of slowly crystallizable polyester consisting essentially of repeat units from terephthalic acid, about 85-93 mol % ethylene glycol and about 15-7 mol % cyclohexanedimethanol, the total thickness of slowly crystallizable
- 25 polyester being less than about 50% of the total thickness of said film.
- 30 3. The film according to Claim 2 wherein said heat-sealing layer is contained on both sides of said readily crystallizable layer.
- 35 4. The film according to Claim 2 wherein said slowly crystallizable polyester accounts for about 5-50% of the total thickness of said film.

- 16 -

5. The film according to Claim 3 wherein said slowly crystallizable polyester accounts for about 5-50% of the total thickness of said film.
- 5      6. The film according to Claim 1 wherein said polyester consists essentially of repeat units from terephthalic acid, about 87-91 mol % ethylene glycol and about 13-9 mol % cyclohexanedimethanol.
- 10     7. The film according to Claim 2 wherein said polyester consists essentially of repeat units from terephthalic acid, about 87-91 mol % ethylene glycol and about 13-9 mol % cyclohexanedimethanol.
- 15     8. A package comprising the film according to Claim 1.
9. A package comprising the film according to Claim 2.
10. A molded article comprising the film of Claim 1.
- 20     11. A molded article comprising the film of Claim 2.
12. The film according to Claim 2 wherein said layers are coextruded.
- 25     13. The film according to Claim 2 wherein at least one of said layers is preformed and the other is applied to the preformed layer as a coating.

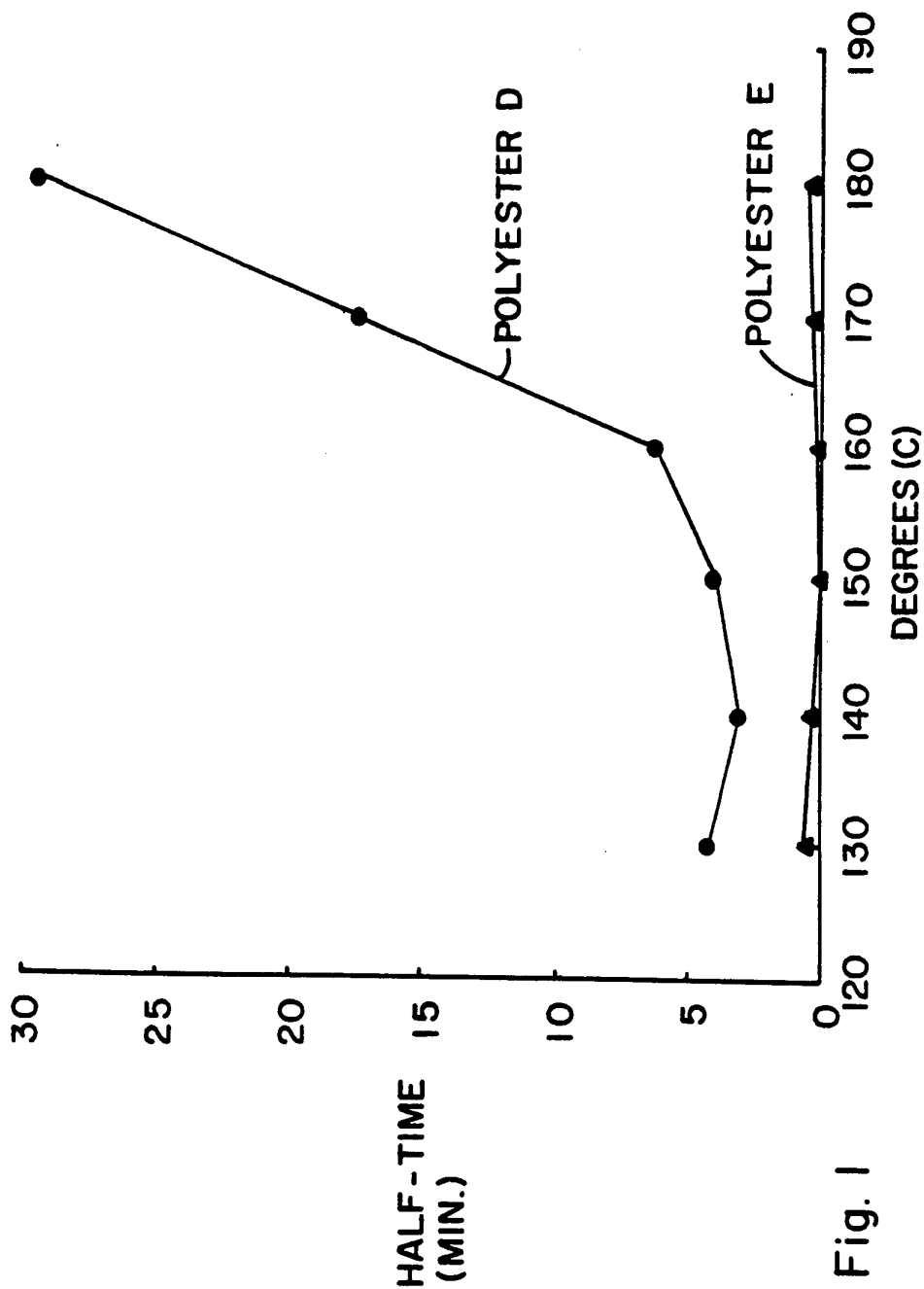


Fig. 1

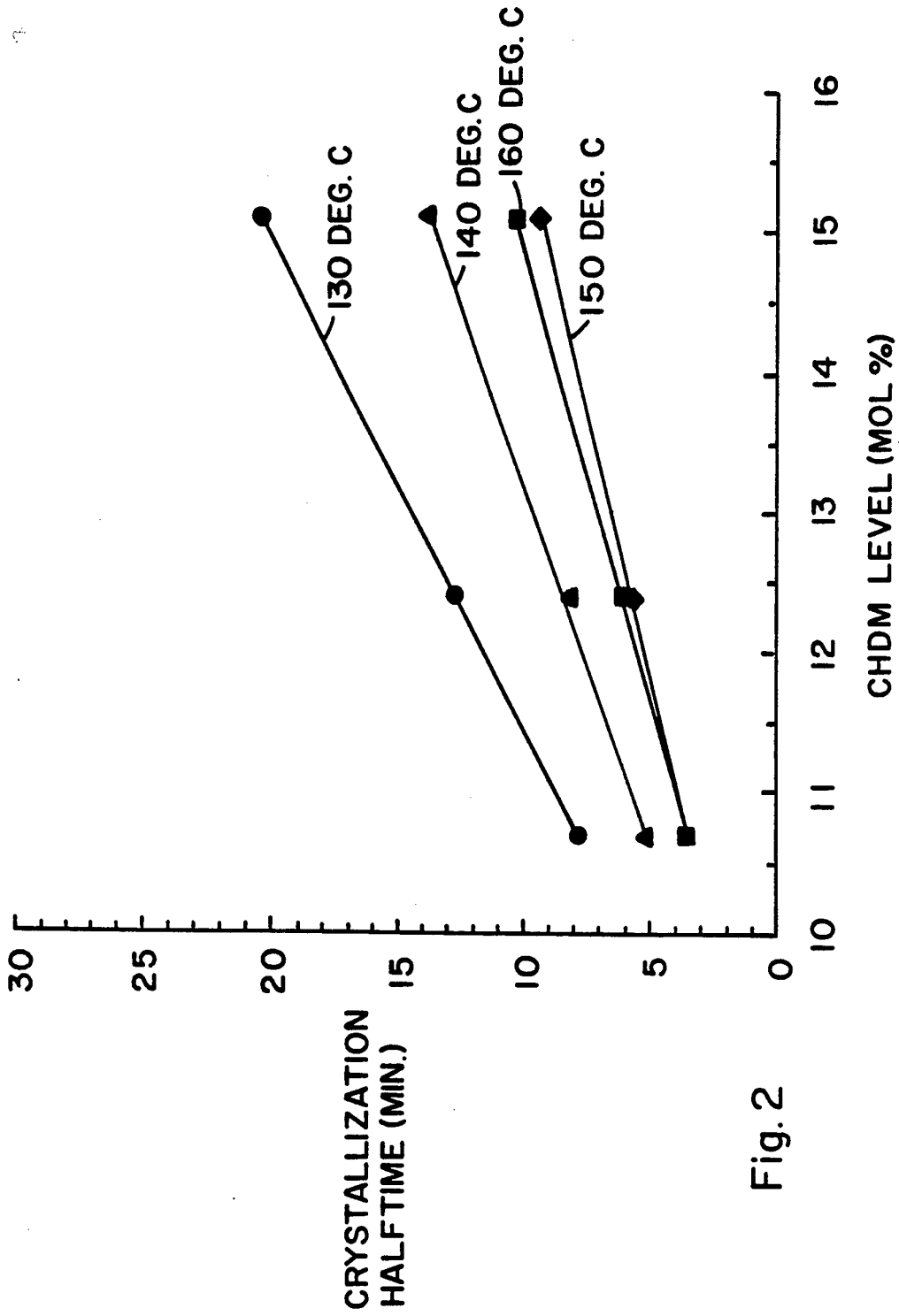


Fig. 2

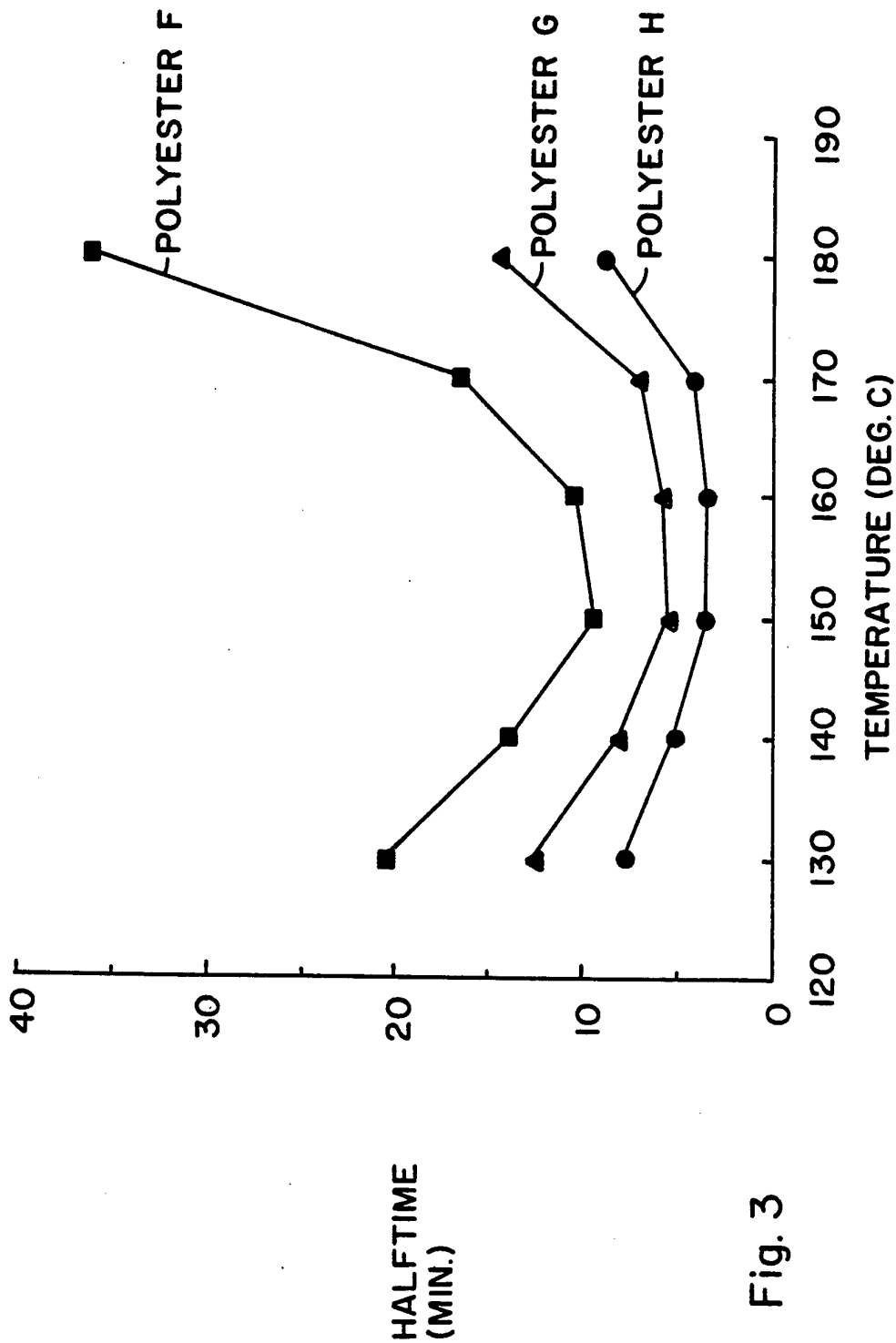


Fig. 3

INTERNATIONAL SEARCH REPORT

Int. l. Application No  
PCT/US 94/11348

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C08J5/18 B32B27/36 C08G63/199 B65D65/40

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C08J C08G B32B B65D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A,4 765 999 (WINTER) 23 August 1988 cited in the application see example 1 see column 2, line 33 - line 36 ---	1-13
X	EP,A,0 517 171 (DU PONT-MITSUI POLCHEMICALS CO., LTD.) 9 December 1992 see page 2, line 1 - page 3, line 11 see page 5, line 14 - line 31 see example 3 --- -/--	1

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

\* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

17 January 1995

Date of mailing of the international search report

31. 01. 95

Name and mailing address of the ISA  
European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+ 31-70) 340-3016

Authorized officer

De Jonge, S

## INTERNATIONAL SEARCH REPORT

Int. Application No

PCT/US 94/11348

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Week 8605, Derwent Publications Ltd., London, GB; AN 86-032457 & JP,A,60 253 545 (OKURA INDUSTRIAL KK) 14 December 1985 cited in the application see abstract ---	1,2
A	WO,A,91 08107 (EASTMAN KODAK COMPANY) 13 June 1991 see claims 1-5; example 2 see page 5, line 20 - line 23 ---	1,2
P,X	DATABASE WPI Week 9433, Derwent Publications Ltd., London, GB; AN 94-271890 & US,A,5 340 907 (EASTMAN CHEM CO) 23 August 1994 see abstract -----	1

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International Application No

PCT/US 94/11348

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A-4765999	23-08-88	NONE	
EP-A-0517171	09-12-92	JP-A- 5209116 US-A- 5310787	20-08-93 10-05-94
WO-A-9108107	13-06-91	US-A- 5034263 EP-A- 0502960 JP-T- 5501682	23-07-91 16-09-92 02-04-93