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(54) Title: PROCESS FOR PRODUCING ETHYLENE-ALPHA-OLEFIN INTERPOLYMER FILMS

(57) Abstract: There is disclosed a process for producing films from ethylene- α -olefin interpolymers, which films have, in combination, the properties of improved surface haze, improved stiffness, and improved color. The process involves incorporating from about 0.1 to less than 1 % by weight, of a functionalized ethylene homopolymer or functionalized ethylene interpolymer into an ethylene- α -olefin interpolymer-containing composition, and forming a film from the composition.

PROCESS FOR PRODUCING ETHYLENE-ALPHA-OLEFIN
INTERPOLYMER FILMS

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FIELD OF THE INVENTION

This invention relates to a process for producing films of ethylene- α -olefin interpolymers having, in combination, the properties of improved surface haze, improved stiffness, and improved color. The process involves forming a film from a composition comprising an ethylene- α -olefin interpolymer and an amount of from about 0.1% to less than 1% by weight, based on the composition, of a functionalized ethylene homopolymer or a functionalized ethylene- α -olefin interpolymer.

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BACKGROUND OF THE INVENTION

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Ethylene- α -olefin interpolymer films are often employed in packaging and other applications where appearance is important. Usually in the case of packaging, it is desired that the film be optically transparent and glossy so that the product packaged within will be more attractive. Accordingly, it is desirable to provide a film that has not only improved surface haze properties, but also improved stiffness and color.

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SUMMARY OF THE INVENTION

The novel process of the present invention for the production of ethylene- α -olefin interpolymer films having improved surface haze, improved stiffness and improved color is comprised as follows. A composition comprising an ethylene- α -olefin interpolymer and an amount of from about 0.1 to less than 1% by weight, based on the composition, of a functionalized ethylene homopolymer or a functionalized ethylene- α -olefin interpolymer, as defined herein, is prepared. The composition is then formed into a film in any manner known in the art. In a preferred

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embodiment of the process, the film is formed utilizing the well-known blown film technique.

DETAILED DESCRIPTION OF THE INVENTION

5 The novel process of the present invention for the production of ethylene- α -olefin interpolymer films having improved surface haze, improved stiffness and improved color is comprised as follows. A composition comprising an ethylene- α -olefin interpolymer and an amount of from about 0.1 to less than 1% by weight, based on the composition, of a
10 functionalized ethylene homopolymer or a functionalized ethylene- α -olefin interpolymer, as defined herein, is prepared. The composition is then formed into a film in any manner known in the art. In a preferred embodiment of the process, the film is formed utilizing the well-known blown film technique.

15 More particularly, the ethylene- α -olefin interpolymer used in preparing the compositions, that are formed into the films of the present invention, is any ethylene- α -olefin interpolymer comprising ethylene and at least one, or more, other α -olefin wherein the ethylene content is at least about 50% by weight of the total monomers comprising the interpolymer.
20 Preferably, the ethylene content of the interpolymer ranges from greater than about 50% to about 99% by weight of the total monomers comprising the interpolymer. The α -olefin monomer(s) preferably contain from 3 to about 16 carbon atoms. Exemplary α -olefins that may be utilized herein are propylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 4-methyl-
25 1-pentene, 1-decene, 1-dodecene, 1-hexadecene and the like. Also utilizable herein as the α -olefin monomer are non-conjugated dienes and olefins formed in situ in the polymerization medium. The ethylene- α -olefin interpolymers may be linear or may contain rheologically significant long chain branches, such as the "substantially linear" polymers, and may be
30 produced by any process, such as solution, gas phase, or slurry loop, and

with any olefin polymerization catalyst, such as the constrained geometry, metallocene type, Ziegler Natta types, or chromium oxide types. The ethylene- α -olefin interpolymers are present in the compositions utilized to form the films of the present invention, in an amount of from about greater than 5 99% to about 99.9% by weight of the composition.

The functionalized ethylene homopolymer or functionalized ethylene- α -olefin interpolymers suitable for use herein are defined as follows. The polyethylene that is functionalized may be any homopolymer of ethylene. The interpolymers of ethylene that are functionalized and are suitable for use in 10 preparing the compositions from which the films of the present invention are formed are any ethylene- α -olefin interpolymers comprising ethylene and at least one, or more, other α -olefin wherein the ethylene content is at least about 50% by weight of the total monomers comprising the interpolymers. Preferably, the ethylene content of the interpolymers ranges from greater 15 than about 50% to about 99% by weight of the total monomers comprising the interpolymers. The α -olefin monomer(s) preferably contain from 3 to about 16 carbon atoms. Exemplary α -olefins that may be utilized herein are propylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 4-methyl-1-pentene, 1-decene, 1-dodecene, 1-hexadecene and the like. Also utilizable 20 herein as the α -olefin monomers are non-conjugated dienes and olefins formed in situ in the polymerization medium. The ethylene- α -olefin interpolymers may be linear or may contain rheologically significant long chain branches, such as the "substantially linear" polymers, and may be produced by any process, such as solution, gas phase, or slurry loop, and 25 with any olefin polymerization catalyst, such as the constrained geometry, metallocene types, Ziegler Natta types, or chromium oxide types.

In preparing the functionalized ethylene homopolymers or ethylene- α -olefin interpolymers suitable for use herein, there may be utilized as the functionalizing agent any unsaturated monomer containing one or more 30 carboxylic acid, carboxylic acid ester, or acid anhydride group. Examples of

suitable functionalizing agents herein are carboxylic acids such as acrylic and methacrylic acid, carboxylic acid esters such as methyl acrylate and methyl methacrylate, and acid anhydrides such as maleic anhydride.

Further exemplary functionalizing agents suitable for use herein are
5 unsaturated monocarboxylic acids and polycarboxylic acids and cyclic acid anhydrides. Specifically included herein are acids such as maleic acid, fumaric acid, himic acid, itaconic acid, citraconic acid, mesaconic acid, acrylic acid, methacrylic acid, crotonic acid, isocrotonic acid, and acid anhydrides such as maleic anhydride and himic anhydride. Preferred for
10 use herein as the functionalizing agent is maleic anhydride. Mixtures of functionalizing agents may be utilized in the present invention. The functionalizing agent is utilized in any amount that will functionalize the ethylene homopolymer and/or interpolymer such that the functionalized ethylene homopolymer and/or interpolymer will contain from about 0.01 to
15 about 3% by weight of the at least one functionalizing agent, based on the functionalized ethylene homopolymer or copolymer. Preferably, the functionalizing agent will be utilized in an amount of from about 0.20 to about 10 weight percent, based on the weight of the ethylene component in order to produce the desired functionalized polymer.

20 The functionalized ethylene homopolymers and ethylene- α -olefin interpolymers utilized herein may be prepared by any process known in the art. Exemplary of such processes for preparing functionalized ethylene homopolymers and copolymers are U.S. Patents Numbers 4,612,155; 4,762,890; 5,180,788; 5,705,565; and 5,728,776; and International
25 Publication No. WO 01/36495 A1.

The functionalized ethylene homopolymer or ethylene- α -olefin interpolymer is present in the compositions utilized to form the films of the present invention, in an amount of from about 0.1 to less than 1% by weight of the composition.

The compositions, from which the novel films are formed, comprising from greater than 99 to about 99.9% by weight of the composition, of the ethylene- α -olefin interpolymer described herein, and from about 0.1 to less than 1% by weight of the composition, of the functionalized ethylene
5 homopolymer or functionalized ethylene- α -olefin interpolymer described herein are prepared by any method known in the art. For example, the components of the composition may be admixed together on a conventional mixing machine such as a mill roll, an extruder or a continuous mixer. In preparing the compositions of the examples herein, pellets of the
10 components were placed into a fiber drum and mixed for five minutes on an INS Plastic Tumbling unit. In more detail, in each of the following examples wherein a composition is prepared, the following procedure was utilized. A predetermined amount of ethylene- α -olefin interpolymer and functionalized ethylene homopolymer or functionalized ethylene- α -olefin interpolymer,
15 both described herein, were weighed using a Toledo scale and placed into a fiber drum. The fiber drum was then placed on an INS Plastic Tumbling unit and mixed for a period of five minutes yielding the desired composition.

For many purposes, it may be desirable to incorporate other conventional additives with the compositions utilized in the present
20 invention. For example, there may be added antioxidants, heat and light stabilizers, dyes, antistatic agents, lubricants, preservatives, processing aids, slip agents, antiblocking agents, pigments, flame retardants, blowing agents, and the like.

The compositions may be fabricated into films by any technique
25 known in the art, and are particularly suitable for use with blown-film techniques. In further detail, the films of the examples were blown under the following conditions.

The tumbled pellet blends were fed to the hopper of a 2.5-inch (64-
30 mm) Egan extruder, having a length to diameter ratio of 24/1 and a barrier screw. The extruder was equipped with a 6-inch (152-mm) diameter

annular die having a die gap of 88 mil (2.2 mm). A dual-lip air ring was used for cooling the external surface of the tubular film as it exited the annular die. A "hump-back" extruder temperature profile was employed with setpoints of 193, 232, 221, 216, and 204°C for zones 1 through 5, respectively, and a die setpoint of 218°C was employed. A throughput rate of 69 kg/h was maintained while producing films of 1-mil (0.025-mm) thickness with a blow-up ratio of 2.4. A frost line height of 38 cm was maintained during the film formation. Films were wound into rolls of about 60 linear meters and aged for 40 hours at standard lab conditions (23°C and 50% relative humidity) before testing.

The invention will be more readily understood by reference to the following examples. There are, of course, many other forms of this invention which will become obvious to one skilled in the art, once the invention has been fully disclosed, and it will accordingly be recognized that these examples are given for the purpose of illustration only, and are not to be construed as limiting the scope of this invention in any way. Moreover, all U.S. patents referred to herein are incorporated by reference in their entirety.

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EXAMPLES

In the following examples, the test procedures listed below were used in evaluating the properties of the films herein.

Total film haze was measured according to ASTM D1003 with a BYK Gardner Haze-gard Plus. In order to measure the internal haze component, a non-interacting liquid of similar refractive index (1.48) to the polymer (1.51) was spread over the rough surface to "fill-in" the rough surface texture. The haze measured on such a coated sample represented the component originating from the bulk, or internal semicrystalline structure only (internal haze). The surface haze component was calculated by subtracting the internal haze from the total haze.

30

Acid number (AN) was measured according to ASTM D1386-83 with the following modifications: A 0.05N sodium hydroxide in methanol solution was substituted for the 0.1N potassium hydroxide in ethanol and the sample size was increased from between 1 and 2 g to 5 g, and the weighing
5 accuracy was changed from 0.001 to 0.0001g. The percent by weight of grafted functionalizing agent in the final functionalized polyethylene is equal to 0.178•AN.

Melt index (MI) and Melt Flow Ratio (MFR) were measured in accordance with ASTM D1238, conditions 190/2.16 and 190/21.6.

10 Secant modulus (at 1% strain) was measured in accordance with ASTM D882. Due to the anisotropic nature of polyethylene blown films, modulus measurements were made by testing the blown film in both the extrusion, or machine direction (MD), and the hoop, or transverse direction (TD).

15 Yellowness index of pellets was measured according to ASTM E313-96 with a BYK-Gardner Color-View Spectrophotometer. Pellets composed of blends of polyethylene and functionalized polyethylene were prepared by melt extrusion of a tumbled pellet/pellet blend. Melt extrusion was carried out with a 1.25-in (31.8-mm) single screw Killion extruder at a screw speed
20 of 50 rpm with the following temperature profile (barrel zones 1-4/adaptor/clamp ring): 177/221/216/216/216/216 °C. Pellets were fashioned with a standard water bath and pelletizer and conditioned at 23°C and 50% humidity for at least 24 hours before measurement of yellowness index.

25 Density was measured in accordance with ASTM D4883-89 with a Haake V Tecrad density measurement system.

EXAMPLE 1

30 In this example, blown films were prepared as described herein. The major component of the composition from which the films were formed is

ethylene/1-hexene interpolymers containing 8.3 weight % 1-hexene and having MI of 0.85 dg/min, and density of 0.917 g/cm³ (referred to herein as LLDPE). The compositions also comprised varying amounts of a maleic anhydride grafted ethylene/1-hexene interpolymers (referred to herein as mal-LLDPE) having an acid number of 7, a maleic anhydride content of 1.2% by weight, and an MI of 8 dg/min.

The maleic anhydride grafted ethylene/1-hexene interpolymers (referred to as mal-LLDPE herein) used in this example was prepared as follows. Ethylene/1-hexene interpolymers having a density of 0.917 g/cm³, a melt index of 0.8 dg/min, and a Melt Flow Ratio of 27 was prepared utilizing a continuous polymerization process in a fluidized-bed reactor for gas-phase polymerization, consisting of a vertical cylinder of diameter 0.74 meters and height seven meters and surmounted by a velocity reduction chamber. The reactor was provided in its lower part with a fluidization grid and with an external line for recycling gas, which connects the top of the velocity reduction chamber to the lower part of the reactor, at a point below the fluidization grid. The recycling line was equipped with a compressor for circulating gas and a heat transfer means such as a heat exchanger. In particular the lines for supplying ethylene, 1-hexene, hydrogen and nitrogen, which represent the main constituents of the gaseous reaction mixture passing through the fluidized bed, feed into the recycling line.

Above the fluidization grid, the reactor contained a fluidized bed weighing 800 lb (360 kg) consisting of a linear low-density polyethylene powder made up of particles with a weight-average diameter of about 1.3 mm. The gaseous reaction mixture, which contained ethylene, 1-hexene, hydrogen, nitrogen and minor amounts of other components, passed through the fluidized bed under a pressure of about 296 psig (2.04 MPa) with an ascending fluidization speed of about 1.7 ft/s (52 cm/s).

The transition metal component of the Ziegler-Natta catalyst used was prepared in accordance with Example 1-a of European Patent

Application EP 0 703 246 A1. The Ziegler-Natta catalyst was used in prepolymer form, and was prepared in accordance with Example 1-b of European Patent Application EP 0 703 246 A1. A prepolymer containing about 35.7 g of polyethylene per millimole of titanium, with a tri-n-octylaluminum (TnOA) to titanium molar ratio of about 1.0, was thus
5 obtained.

The catalyst was introduced intermittently into the reactor, the said catalyst comprising magnesium, chlorine and titanium. The rate of introduction of prepolymer into the reactor was adjusted to achieve the
10 desired production rate. During the polymerization, a solution of trimethylaluminum (TMA) in n-hexane, at a concentration of about 2 weight percent, was introduced continuously into the line for recycling the gaseous reaction mixture, at a point situated downstream of the heat transfer means. The feed rate of TMA is expressed as a molar ratio of TMA to titanium
15 (TMA/Ti), and is defined as the ratio of the TMA feed rate (in moles of TMA per hour) to the prepolymer feed rate (in moles of titanium per hour). Simultaneously, a solution of tetrahydrofuran (THF) in n-hexane, at a concentration of about 1 weight percent, was introduced continuously into the line for recycling the gaseous reaction mixture. The feed rate of THF is
20 expressed as a molar ratio of THF to titanium (THF/Ti), and is defined as the ratio of the THF feed rate (in moles of THF per hour) to the prepolymer feed rate (in moles of titanium per hour). Dinitrogen monoxide (N₂O) was added as a gas to the line for recycling the gaseous reaction mixture. The concentration of N₂O in the gas phase polymerization medium is expressed
25 in units of parts per million (ppm) by volume.

A solution of chloroform (CHCl₃) in n-hexane, at a concentration of about 0.5 weight percent, was introduced continuously into the line for recycling the gaseous reaction mixture. The feed rate of CHCl₃ is
30 expressed as a molar ratio of CHCl₃ to titanium (CHCl₃/Ti), and is defined as the ratio of the CHCl₃ feed rate (in moles of CHCl₃ per hour) to the

prepolymer feed rate (in moles of titanium per hour). The CHCl_3 was added as a solution in n-hexane to the line for recycling the gaseous reaction mixture.

The continuous gas phase process conditions are given in Table 1.

- 5 The molar ratio TMA/Ti was 4. The molar ratio CHCl_3 /Ti was 2.0. The molar ratio THF/Ti was 0.3. The concentration of dinitrogen monoxide (N_2O) in the polymerization medium was 304 ppm by volume. 1-Hexene was used as comonomer. Under these conditions there was produced the ethylene/1-hexene interpolymer having a density of 0.917 g/cm^3 , and an MI
- 10 of 0.8 dg/min. This is the polymer that was functionalized in Example 1.

Table 1

	Reactor Pressure, psig (MPa)	296(2.04)
	Reactor Temperature, °C	86
5	Fluidized Bulk Density, lb/ft ³ (g/cm ³)	16.3(0.261)
	Reactor Bed Height, ft (meter)	10.7(3.26)
	Ethylene, mole%	50.4
	H ₂ /C ₂ ¹	0.124
10	C ₆ /C ₂ ²	0.117
	TMA/Ti ³	4
	CHCl ₃ /Ti ⁴	2.0
	THF/Ti ⁵	0.3
	N ₂ O, ppm by volume	304
15	Prepolymer Rate, lb/h (kg/h)	0.73(0.33)
	Production Rate, lb/h (kg/h)	205(93)
	Space Time Yield, kg/h-m ³	66.9
	Productivity, mass ratio	281
	Activity ⁶	244
20	Residual Titanium, ppm	4.8

¹hydrogen to ethylene molar ratio; ²1-hexene to ethylene molar ratio;
³trimethylaluminum to titanium molar ratio; ⁴chloroform to titanium molar
ratio; ⁵tetrahydrofuran to titanium molar ratio; ⁶units of gram PE•(mmole Ti)
¹•h⁻¹•(0.1 MPa)⁻¹]

25 The maleic anhydride grafted ethylene/1-hexene interpolymer (mal-
LLDPE herein) was then prepared utilizing a 40-mm x 55-L/D (length/
diameter) Berstorff ZE 40A twin screw extruder, produced and sold by
Berstorff Corporation, of Florence, Kentucky. The extruder screw speed
30 was set at 270 rpm. Pellets of the ethylene/1-hexene interpolymer having a

density of 0.917 g/cc, a melt index of 0.8 dg/min, and a Melt Flow Ratio of 27, were fed to the feed throat of the extruder by a volumetric pellet feeder at a rate of 45 kg/h, and the pellets were melted in barrels 1 and 2. Maleic anhydride was injected at a rate of 1.1 kg/h through an injection device
 5 located between barrels 2 and 3 of the extruder. The maleic anhydride was then mixed with the interpolymer in three sequential reaction/mixing zones, each separated from the other by reverse conveying elements. The three sequential mixing/reaction zones were maintained at a temperature of 358°C. The resulting maleated interpolymer was passed under two
 10 sequential vacuum zones with 6840 Pa of vacuum, and 6960 Pa of vacuum, respectively, to remove volatiles. The resulting maleic anhydride grafted ethylene-1/hexene interpolymer (herein mal-LLDPE) was recovered from the end of the extruder by a conventional strand bath and pelletizer system. Analysis revealed that the mal-LLDPE had a melt index of 8
 15 dg/min, an acid number of 7, and a maleic anhydride content of 1.2% by weight of the interpolymer.

As mentioned herein, films were produced from the following compositions comprising the LLDPE and varying proportions of the mal-LLDPE. The data are reported in Table 2 below.

20

TABLE 2

mal-LLDPE content (wt.%)	total haze (%)	internal haze (%)	surface haze (%)	MD secant modulus (Pa)	TD secant modulus (Pa)
0 (100% LLDPE)	12.3	3.3	9.0	1.99(10 ⁸)	2.21(10 ⁸)
0.1	6.4	2.6	3.8	2.08(10 ⁸)	2.59(10 ⁸)
0.2	6.3	2.6	3.7	2.25(10 ⁸)	2.63(10 ⁸)
0.5	5.5	2.5	3.0	2.23(10 ⁸)	2.85(10 ⁸)
1	5.7	2.2	3.5	2.37(10 ⁸)	2.88(10 ⁸)
2	5.6	2.2	3.4	2.29(10 ⁸)	3.06(10 ⁸)

From the data in Table 2, it is observed that films formed from the compositions described herein comprising the required amounts of

ethylene- α -olefin interpolymer and functionalized component are characterized by having decreased surface haze, thereby having improved transparency. This is in comparison with a film formed from the LLDPE. Further, from the data in Table 2, it is observed that the films of the present invention formed from the compositions comprising the required amounts of the components have increased moduli, as compared to films formed from the LLDPE. The increased moduli values indicate that the films have increased stiffness.

The compositions of Table 2 that were formed into films, were measured for yellowness index prior to being formed into films. The yellowness index values are reported below in table 3.

TABLE 3

mal-LLDPE content (wt.%)	yellowness index
0 (100% LLDPE)	2.42±0.36
0.1	2.33±0.21
0.2	2.28±0.12
0.5	2.87±0.32
1	3.42±0.08
2	4.85±0.21

From the data in Table 3, it is observed that as the amount of mal-LLDPE blended with the LLDPE reaches a level of 1% by weight, the yellowness index of the blend composition is undesirably increased, as compared to the yellowness index of the composition containing no mal-LLDPE. Accordingly, films prepared from the compositions containing the required content of mal-LLDPE, as described herein, will have improved color.

It should be clearly understood that the forms of the invention herein described are illustrative only and are not intended to limit the scope of the invention. The present invention includes all modifications falling within the scope of the following claims.

CLAIMS

We Claim:

- 5 1. A process for producing films characterized by having decreased
 surface haze, increased secant modulus and improved color,
 comprising forming a film from a composition comprising from
 greater than 99% to about 99.9% by weight of an ethylene- α -olefin
10 interpolymer, based on the composition, and from about 0.1% to less
 than 1% by weight of a functionalized component selected from the
 group consisting of functionalized ethylene homopolymer and
 functionalized ethylene- α -olefin interpolymer, wherein the
 functionalized component comprises from about 0.01% to about 3%
15 by weight of functionalizing agent based on the functionalized
 component.
2. The process according to Claim 1 wherein the ethylene- α -olefin
 interpolymer comprises ethylene and at least one, or more, other α -
 olefin, and wherein the ethylene content is at least about 50% by
20 weight of the interpolymer.
3. The process according to Claim 2 wherein the other α -olefin has
 from 3 to about 16 carbon atoms.
- 25 4. The process according to Claim 1 wherein the functionalized
 ethylene- α -olefin interpolymer comprises a functionalized
 interpolymer of ethylene- α -olefin wherein the ethylene content is at
 least about 50% by weight of the interpolymer, and the α -olefin has
 from 3 to about 16 carbon atoms.

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5. The process according to Claim 1 wherein the functionalizing agent is selected from the group consisting of unsaturated monomers containing one, or more, carboxylic acid groups and unsaturated monomers containing one, or more, acid anhydride groups.
- 5
6. The process according to Claim 5 wherein the functionalizing agent is maleic anhydride.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 02/40772

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 7 C08F255/02 C08F8/46 C08L23/16 C08L23/04 C08J5/18

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)
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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)

WPI Data, PAJ, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 171 777 A (DOW CHEMICAL CO) 19 February 1986 (1986-02-19)	1-3,5,6
Y	abstract; claims 1-14 page 15, line 1-13 page 5, line 6-27	4
Y	----- WO 90 01504 A (DOW CHEMICAL CO) 22 February 1990 (1990-02-22)	4
A	abstract; claims 1-19 -----	1-3,5,6
A	WO 01 36495 A (EASTMAN CHEM CO) 25 May 2001 (2001-05-25) cited in the application abstract; claims 1-28 -----	1-6
E	US 6 514 583 B1 (SHAH GAUTAM P ET AL) 4 February 2003 (2003-02-04) abstract; claims 1-51 -----	1-6

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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Patent document cited in search report		Publication date	Patent family member(s)	Publication date
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