

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter I of the Patent Cooperation Treaty)

(PCT Rule 44bis)

Applicant's or agent's file reference 77177-WO-PCT	FOR FURTHER ACTION	See item 4 below
International application No. PCT/US2015/059972	International filing date (<i>day/month/year</i>) 10 November 2015 (10.11.2015)	Priority date (<i>day/month/year</i>) 31 December 2014 (31.12.2014)
International Patent Classification (8th edition unless older edition indicated) See relevant information in Form PCT/ISA/237		
Applicant DOW GLOBAL TECHNOLOGIES LLC		

1.	This international preliminary report on patentability (Chapter I) is issued by the International Bureau on behalf of the International Searching Authority under Rule 44 bis.1(a).																								
2.	This REPORT consists of a total of 6 sheets, including this cover sheet.																								
In the attached sheets, any reference to the written opinion of the International Searching Authority should be read as a reference to the international preliminary report on patentability (Chapter I) instead.																									
3.	This report contains indications relating to the following items: <table style="width: 100%;"> <tr> <td style="width: 5%; text-align: center;">☒</td> <td style="width: 25%;">Box No. I</td> <td style="width: 70%;">Basis of the report</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. II</td> <td>Priority</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. III</td> <td>Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. IV</td> <td>Lack of unity of invention</td> </tr> <tr> <td style="text-align: center;">☒</td> <td>Box No. V</td> <td>Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. VI</td> <td>Certain documents cited</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. VII</td> <td>Certain defects in the international application</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. VIII</td> <td>Certain observations on the international application</td> </tr> </table>	☒	Box No. I	Basis of the report	☐	Box No. II	Priority	☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability	☐	Box No. IV	Lack of unity of invention	☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement	☐	Box No. VI	Certain documents cited	☐	Box No. VII	Certain defects in the international application	☐	Box No. VIII	Certain observations on the international application
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☐	Box No. VIII	Certain observations on the international application																							
4.	The International Bureau will communicate this report to designated Offices in accordance with Rules 44bis.3(c) and 93bis.1 but not, except where the applicant makes an express request under Article 23(2), before the expiration of 30 months from the priority date (Rule 44bis .2).																								

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Date of issuance of this report 04 July 2017 (04.07.2017)
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PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To: see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference see form PCT/ISA/220		FOR FURTHER ACTION See paragraph 2 below
International application No. PCT/US2015/059972	International filing date (day/month/year) 10.11.2015	Priority date (day/month/year) 31.12.2014
International Patent Classification (IPC) or both national classification and IPC INV. B32B27/08 B32B27/30 B32B27/32		
Applicant DOW GLOBAL TECHNOLOGIES LLC		

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:  European Patent Office P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Fax: +31 70 340 - 3016	Date of completion of this opinion see form PCT/ISA/210	Authorized Officer Hillebrand, Gerhard Telephone No. +31 70 340-0
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Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:
 - ☒ the international application in the language in which it was filed.
 - ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).
2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))
3. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:
 - a. ☐ forming part of the international application as filed:
 - ☐ in the form of an Annex C/ST.25 text file.
 - ☐ on paper or in the form of an image file.
 - b. ☐ furnished together with the international application under PCT Rule 13ter.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.
 - c. ☐ furnished subsequent to the international filing date for the purposes of international search only:
 - ☐ in the form of an Annex C/ST.25 text file (Rule 13ter.1(a)).
 - ☐ on paper or in the form of an image file (Rule 13ter.1(b) and Administrative Instructions, Section 713).
4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
5. Additional comments:

Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	<u>1-12</u>
	No: Claims	
Inventive step (IS)	Yes: Claims	
	No: Claims	<u>1-12</u>
Industrial applicability (IA)	Yes: Claims	<u>1-12</u>
	No: Claims	

2. Citations and explanations

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Reference is made to the following documents; the numbering will be adhered to in the rest of the procedure.

- D1 WO 2010/034456 A1 (BOREALIS AG [AT]; OEYSAED HARRY [NO]; GAHLEITNER MARKUS [AT]; SKAU KAR) 1 April 2010 (2010-04-01)
- D2 WO 2012/164585 A2 (ESSEL PROPACK LTD [IN]; BHATTACHARYA SATYA PRASAD [IN]; BANERJEE MRINA) 6 December 2012 (2012-12-06)

1. Inventive step(Art. 33(3) PCT)

a)

Document D1 discloses a multi-layer structure comprising a polyolefin layer comprising a compatibiliser and a barrier layer of EVOH. In addition D1 discloses the compatibiliser being an acid or anhydride modified polyolefin(claims 1 and 6).

The difference of claim 1 of the present application versus D1 is that D1 does not disclose an additional tie layer comprising a maleic-grafted anhydride grafted polymer. In addition the present application claims a specific compatibilizer component, which is also not disclosed in D1.

Tie layer made of maleic-anhydride grafted polymers are well known in the prior art.

Document D2 discloses a tie layer of maleic anhydride grafted polyethylene(claims 1 and 15) applied in a multilayer structure between polyolefin and polar layers.

Thus the subject-matter of claims 1,3 and 9 is not inventive in view of the prior art.

b)

The remaining claims, which are not mentioned under point 1.a), do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step, the reasons being as follows:

The features of these dependent claims are merely selected from several straight forwarded possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed.

2. Others

a)

To meet the requirements of the PCT, the document D1 should be identified in the description and the relevant background art disclosed therein should be briefly summarised in an objective way.

b)

The title of the application is misleading and not supported by the claims.

The claim comprised in the title "Self-recyclable barrier packaging" is not supported by the present set of claims, because the present set of claims refer to a multi-layer structure and not to a packaging.

In addition these claims comprise no disclosure how such a multi-layer structure will be re-obtained by following the claimed method of claim 10 in which the structure is cut into flakes and pellets. Obviously the same well defined multi-layered structure comprising a specific order of the layers within said structure will not be obtained by using such pellets as a starting material for making a new structure, because the pellets will be randomly fused together and thus also the order of the layers will appear randomly.



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- (74) **Agent:** **WELVAERT, Bronwyn**; The Dow Chemical Company, Intellectual Property, P.O. Box 1967, Midland, Michigan 48641-1967 (US).

- (81) **Designated States** (*unless otherwise indicated, for every kind of national protection available*): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.
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(54) **Title:** SELF-RECYCLABLE BARRIER PACKAGING

(57) **Abstract:** A multi-layer structure comprising: a) at least one layer comprising a polyolefin component comprising i) 60 to 94 weight percent of a first component selected from the group consisting of ethylene homopolymer, ethylene copolymer, polypropylene homopolymer, polypropylene copolymer, and combinations thereof ii) 0 - 35 weight percent of a functional polymer component, and iii) 1 -35 weight percent of a compatibilizer component comprising an anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolymer having a melt viscosity (177°C) less than, or equal to, 200,000 cP and a density from 0.855 to 0.94 g/cc; b) at least one tie layer comprising maleic-anhydride grafted polymer with a melt index of less than 50 dg/min, wherein the tie layer does not contain the compatibilizer component; and c) at least one polar layer comprising a polar polymer, is disclosed.



WO 2016/109023 A1

SELF-RECYCLABLE BARRIER PACKAGING

REFERENCE TO RELATED APPLICATIONS

The present application claims the benefit of EP Application No. 14382589.1, filed
5 December 31, 2014, which is incorporated herein by reference in its entirety.

FIELD OF THE INVENTION

The instant invention relates to multi-layer structures and the recycling thereof.

10 BACKGROUND

Every year, a considerable amount of flexible packaging barrier film is being disposed by landfill or incineration, due to the difficulty of dispersing typical functional barrier polymers such as Ethylene vinyl alcohol (EVOH) or Polyamide (PA) within a more conventional Polyolefin (PO) waste stream for further recycling.

15 Typically, flexible film converters and recyclers get value out of post industrial waste by submitting the material to a compatibility process, in which a recycling compatibilizer is added to the waste stream for further conversion of the material into pellets, allowing its reuse. However, when a post consumer barrier film structure is collected and mixed with a conventional PO waste stream it is very difficult to know when to use such compatibilizers
20 and the amounts required.

An all-PO film structure can facilitate the recycling of waste by using only this family of raw material in its structure, therefore avoiding the compatibilizer step, however, when gas barrier is required the package will be compromised. Therefore, a solution is desired to allow the incorporation of a gas barrier material in a film structure while still allowing for recycling
25 without the need of a secondary process.

SUMMARY OF THE INVENTION

The present invention enables improved compatibilization, when compared to adding a compatibilizer component as a separate stream in a recycling process.

30 In one broad embodiment of the present invention, there is disclosed a multi-layer structure comprising, consisting of, or consisting essentially of: a) at least one layer comprising a polyolefin component comprising i) 60 to 94 weight percent of a first component selected from the group consisting of ethylene homopolymer, ethylene

copolymer, polypropylene homopolymer, polypropylene copolymer, and combinations thereof ii) 0 – 35 weight percent of a functional polymer component, and iii) 1 -35 weight percent of a compatibilizer component comprising an anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolymers having a melt viscosity (177°C) less than, or equal to, 200,000 cP and a density from 0.855 to 0.94 g/cc; b) at least one tie layer comprising maleic-anhydride grafted polymer with a melt index of less than 50 dg/min, wherein the tie layer does not contain the compatibilizer component; and c) at least one polar layer comprising a polar polymer, is disclosed.

In another embodiment of the present invention, there is disclosed a method for making a recycled structure comprising a) converting the above-mentioned multi-layer structure into flakes, b) optionally converting the flakes into pellets, and c) forming the recycled structure from the flakes or the pellets wherein the method does not involve the use of the compatibilizer component.

BRIEF DESCRIPTION OF THE FIGURES

Fig. 1 shows a 3-layer structure embodiment

Figs. 2a and 2b show embodiments of 5-layer structures

Figs. 3a and 3b show embodiments of 7-layer structures

Figs. 4a, 4b, and 4c show embodiments of 9-layer structures

DETAILED DESCRIPTION

In one broad embodiment of the present invention, there is disclosed a multi-layer structure comprising: a) at least one layer comprising a polyolefin component comprising i) 60 to 94 weight percent of a first component selected from the group consisting of ethylene homopolymer, ethylene copolymer, polypropylene homopolymer, polypropylene copolymer, and combinations thereof, ii) 0 – 35 weight percent of a functional polymer component, and iii) 1 -35 weight percent of a compatibilizer component comprising an anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolymers having a melt viscosity (177°C) less than, or equal to, 200,000 cP and a density from 0.855 to 0.94 g/cc; b) at least one tie layer comprising maleic-anhydride grafted polymer with a melt index of less than 50 dg/min, wherein the tie layer does not contain the compatibilizer component ; and c) at least one polar layer comprising a polar polymer.

POLYOLEFIN COMPONENT LAYER

The polyolefin layer comprises a first component selected from the group consisting of ethylene homopolymer (eg. DMDA-8007 NT 7 sold by The Dow Chemical Company), ethylene copolymer (eg. DOWLEX 2045 G sold by the Dow Chemical Company),
5 polypropylene homopolymer (eg. H110-02N sold by Braskem), polypropylene copolymer (eg. DS6D81 sold by Braskem), and combinations thereof.

The first component is present in the polyolefin layer in the range of from 60 weight percent to 94 weight percent. Any and all ranges between 60 and 94 weight percent are included herein and disclosed herein, for example, the first component can be present in the
10 range of from 65 to 90 weight percent, 70 to 87 weight percent, or 75 to 82 weight percent.

The polyolefin component can also contain a functional polymer component. Examples of polymers that can be used include, but are not limited to a maleic anhydride functionalized polyolefin (eg. AMPLIFY™ TY 1353 sold by the Dow Chemical Company), acrylate ethylene copolymer (eg. AMPLIFY™ EA 101 sold by the Dow Chemical
15 Company), ethylene vinyl acetate (eg. ELVAX 450 sold by DuPont), and mixtures thereof.

In an embodiment, the polyolefin component comprises a blend of 5 to 30 weight percent of a sealant polymer and 70 to 95 weight percent of an ethylene homopolymer or ethylene alpha-olefin copolymer.

The functional polymer component can be present in the polyolefin layer in the range of from 0 to 35 weight percent. Any and all ranges between 0 and 35 weight percent are included herein and disclosed herein, for example, the functional polymer component can be present in the range of from 2 to 16 weight percent, 4 to 12 weight percent, or 6 to 11 weight percent.

25 COMPATIBILIZER - ANHYDRIDE AND/OR CARBOXYLIC ACID FUNCTIONALIZED ETHYLENE/ALPHA-OLEFIN INTERPOLYMER

The polyolefin component further comprises a compatibilizer component which is an anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolymer.

The term "interpolymer," as used herein, refers to polymers prepared by the
30 polymerization of at least two different types of monomers. The generic term interpolymer thus includes copolymers (employed to refer to polymers prepared from two different types of monomers), and polymers prepared from more than two different types of monomers.

The term, "ethylene/ α -olefin interpolymer," as used herein, refers to an interpolymer

that comprises, in polymerized form, a majority amount of ethylene monomer (based on the weight of the interpolmer), and at least one α -olefin.

The term "anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolmer," as used herein, refers to an ethylene/ α -olefin interpolmer that comprises
5 at least one anhydride group and/or at least one acid group (for example, -COOH formed by the hydrolysis of an anhydride) linked by a covalent bond.

The compatibilizer is present in the polyolefin layer in the range of from 1 weight percent to 35 weight percent. Any and all ranges from 1 to 35 weight percent are included herein and disclosed herein, for example, the compatibilizer can be present in the polyolefin
10 layer in the range of from 5 to 30 weight percent, 10 to 25 weight percent, or 15 to 22 weight percent.

The base polymer used to form the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolmer is an ethylene/ α -olefin interpolmer. In various embodiments, α olefins are C3-C20 α (α)-olefins. Any and all ranges between C3 and C20 are
15 included herein and disclosed herein, for example, the α -olefins are C3-C10 α -olefins. Examples of α -olefins that can be used in the present invention include, but are not limited to propylene, 1-butene, 1-pentene, 1-hexene, 1-heptene and 1-octene, and more preferably include propylene, 1-butene, 1-hexene and 1-octene.

In one embodiment, the ethylene/ α -olefin interpolmer is a homogeneously branched
20 linear interpolmer, and further a copolymer, or a homogeneous branched substantially linear interpolmer, and further a copolymer.

In one embodiment, the ethylene/ α -olefin interpolmer is a homogeneously branched linear interpolmer, and further a copolymer.

In one embodiment, the ethylene/ α -olefin interpolmer is a homogeneously branched
25 substantially linear interpolmer.

The terms "homogeneous" and "homogeneously-branched" are used in reference to an ethylene/ α -olefin interpolmer, in which the α -olefin comonomer is randomly distributed within a given polymer molecule, and all of the polymer molecules have the same or substantially the same comonomer-to-ethylene ratio.

30 The homogeneously branched linear ethylene interpolymers are ethylene polymers, which lack long chain branching, but do have short chain branches, derived from the comonomer polymerized into the interpolmer, and which are homogeneously distributed, both within the same polymer chain, and between different polymer chains. These

ethylene/ α -olefin interpolymers have a linear polymer backbone, no measurable long chain branching, and a narrow molecular weight distribution. This class of polymers is disclosed, for example, by Elston in US Patent No. 3,645,992, and subsequent processes to produce such polymers, using bis-metallocene catalysts, have been developed, as shown, for example, in EP 0 129 368; EP 0 260 999; US Patent No. 4,701,432; US Patent No. 4,937,301; US Patent No. 4,935,397; US Patent No. 5,055,438; and WO 90/07526; each incorporated herein by reference. As discussed, the homogeneously branched linear ethylene interpolymers lack long chain branching, just as is the case for the linear low density polyethylene polymers or linear high density polyethylene polymers. Commercial examples of homogeneously branched linear ethylene/ α -olefin interpolymers include TAFMER polymers from the Mitsui Chemical Company, and EXACT and EXCEED polymers from ExxonMobil Chemical Company.

The homogeneously branched substantially linear ethylene/ α -olefin interpolymers are described in U.S. Patent Nos. 5,272,236; 5,278,272; 6,054,544; 6,335,410 and 6,723,810; each incorporated herein by reference. The substantially linear ethylene/ α -olefin interpolymers have long chain branching. The long chain branches have the same comonomer distribution as the polymer backbone, and can have about the same length as the length of the polymer backbone. "Substantially linear," typically, is in reference to a polymer that is substituted, on average, with "0.01 long chain branches per 1000 carbons" to "3 long chain branches per 1000 carbons." The length of a long chain branch is longer than the carbon length of a short chain branch, formed from the incorporation of one comonomer into the polymer backbone.

Some polymers may be substituted with 0.01 long chain branches per 1000 total carbons to 3 long chain branch per 1000 total carbons, further from 0.01 long chain branches per 1000 total carbons to 2 long chain branch per 1000 total carbons, and further from 0.01 long chain branches per 1000 total carbons to 1 long chain branch per 1000 total carbons.

The substantially linear ethylene/ α -olefin interpolymers form a unique class of homogeneously branched ethylene polymers. They differ substantially from the well-known class of conventional, homogeneously branched linear ethylene/ α -olefin interpolymers, as discussed above, and, moreover, they are not in the same class as conventional heterogeneous "Ziegler-Natta catalyst polymerized" linear ethylene polymers (for example, ultra low density polyethylene (ULDPE), linear low density polyethylene (LLDPE) or high density polyethylene (HDPE), made, for example, using the technique disclosed by Anderson et al.,

in U.S. Patent 4,076,698); nor are they in the same class as high pressure, free-radical initiated, highly branched polyethylenes, such as, for example, low density polyethylene (LDPE), ethylene-acrylic acid (EAA) copolymers and ethylene vinyl acetate (EVA) copolymers.

5 The homogeneously branched, substantially linear ethylene/ α -olefin interpolymers useful in the invention have excellent processability, even though they have a relatively narrow molecular weight distribution. Surprisingly, the melt flow ratio (I10/I2), according to ASTM D 1238, of the substantially linear ethylene interpolymers can be varied widely, and essentially independently of the molecular weight distribution (Mw/Mn or MWD). This
10 surprising behavior is contrary to conventional homogeneously branched linear ethylene interpolymers, such as those described, for example, by Elston in U.S. 3,645,992, and heterogeneously branched, conventional "Ziegler-Natta polymerized," linear polyethylene interpolymers, such as those described, for example, by Anderson et al., in U.S. 4,076,698. Unlike substantially linear ethylene interpolymers, linear ethylene interpolymers (whether
15 homogeneously or heterogeneously branched) have rheological properties, such that, as the molecular weight distribution increases, the I10/I2 value also increases.

Long chain branching can be determined by using ^{13}C Nuclear Magnetic Resonance (NMR) spectroscopy, and can be quantified using the method of Randall (Rev. Macromol. Chem. Phys., C29 (2 &3), 1989, p. 285-297), the disclosure of which is incorporated herein
20 by reference. Two other methods are Gel Permeation Chromatography, couple with a Low Angle Laser Light Scattering detector (GPC/LALLS), and Gel Permeation Chromatography, coupled with a Differential Viscometer detector (GPC-DV). The use of these techniques for long chain branch detection, and the underlying theories, have been well documented in the literature. See, for example, Zimm, B.H. and Stockmayer, W.H., J. Chem. Phys., 17, 1301
25 (1949), and Rudin, A., Modern Methods of Polymer Characterization, John Wiley & Sons, New York (1991) pp. 103-112.

In contrast to "substantially linear ethylene polymer," "linear ethylene polymer" means that the polymer lacks measurable or demonstrable long chain branches, that is, the polymer is substituted with an average of less than 0.01 long chain branch per 1000 carbons.

30 An example of an ethylene/ α -olefin copolymer is AFFINITY GA Polyolefin Plastomer available from The Dow Chemical Company, and LICOCENE Performance Polymers from Clariant. Other examples of ethylene/ α -olefin polymers suitable for the

invention include the ultra low molecular weight ethylene polymers described in U.S. Patent Nos. 6,335,410, 6,054,544 and 6,723,810, each fully incorporated herein by reference.

An example of a functionalized ethylene/ α -olefin copolymer is RETAIN 3000 available from The Dow Chemical Company.

5 In various embodiments, the anhydride and/or carboxylic acid functionality is greater than or equal to 0.5 weight percent, based on the total weight of the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer. Any and all ranges greater than or equal to 0.5 weight percent are included herein and disclosed herein, for example, the anhydride and/or carboxylic acid functionality can be greater than or equal to 0.8 weight
10 percent, greater than or equal to 0.9 weight percent, greater than or equal to 1.0 weight percent, from 0.6 to 1.9 weight percent, from 0.8 to 1.7 weight percent, or from 0.9 to 1.5 weight percent based on the total weight of the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer.

In one embodiment, the anhydride and/or carboxylic acid functionalized
15 ethylene/ α -olefin interpolymer is grafted to the polyolefin.

In various embodiments, the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer has a melt viscosity in the range of from 2000 cP to 50,000 cP at 350°F (177°C). Any and all ranges between 2000 cP and 50,000 cP are included herein and disclosed herein, for example, the melt viscosity can be in the range of
20 from 3000 cP to 40,000 cP, from 4000 cP to 30,000 cP, or from 5000 cP to, 20,000 cP, at 350°F (177°C).

In one embodiment, the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer has a density from 0.855 g/cm³ to 0.940 g/cm³. Any and all ranges from 0.855 g/cm³ to 0.940 g/cm³ are included herein and disclosed herein, for
25 example, the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer can have a density from 0.860 g/cm³ to 0.900 g/cm³, from 0.860 g/cm³ to 0.895 g/cm³, from 0.865 g/cm³ to 0.890 g/cm³, or from 0.865 g/cm³ to 0.880 g/cm³.

In one embodiment, the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer has a molecular weight distribution (MWD) (Mw/Mn)
30 from 1.1 to 5.0. Any and all ranges from 1.1 to 5.0 are included herein and are disclosed herein, for example, the anhydride and/or carboxylic acid functionalized ethylene/ α -olefin interpolymer can have a MWD of from 1.3 to 4.0, 1.5 to 2.8, or 2.0 to 2.5, or from 2.0 to 3.0.

In one embodiment, the anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolpolymer has a weight average molecular weight (Mw) in the range of from 2000 g/mole to 50,000 g/mole. Any and all ranges from 2000 g/mole to 50,000 g/mole are included herein and disclosed herein, for example, the anhydride and/or
5 carboxylic acid functionalized ethylene/alpha-olefin interpolpolymer can have a Mw in the range of from 3000 g/mole to 40,000 g/mole, or from 4000 g/mole to 30,000 g/mole.

In one embodiment, the anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolpolymer has a melt index (I2), or calculated melt index (I2) (2.16 kg, 190 C), in the range of from 300 g/10 min to 1500 g/10min. Any and all ranges between
10 300 g/10 min and 1500 g/10 min are included herein and disclosed herein, for example, the anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolpolymer can have a melt index (I2), or calculated melt index (I2) in the range of from 400 g/10 min to 1200 g/10min, or from 500 g/10 min to 1000 g/10 min.

In one embodiment, the anhydride and/or carboxylic acid functionalized
15 ethylene/alpha-olefin interpolpolymer has a percent crystallinity, as determined by DSC, in the range of from 2 percent to 40 percent. Any and all ranges from 2 % to 40% are included herein and disclosed herein, for example, the anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolpolymer can have a percent crystallinity in the range of from 5% to 30 %, from 10% to 25%, or from 15% to 20%.

TIE LAYER

MALEIC-ANHYDRIDE GRAFTED POLYMER

The tie layer comprises a polymer with a maleic anhydride (MAH)-grafted
25 functionality. In a further embodiment, the MAH-graft level is from 0.05 to 1.20 weight percent, based on the weight of the MAH-grafted polymer. In a further embodiment, the MAH-graft level is from 0.07 to 1.00 weight percent, based on the weight of the MAH-grafted polymer. In a further embodiment, the MAH-graft level is from 0.10 to 0.60 weight percent, based on the weight of the MAH-grafted polymer.

30 In one embodiment, the tie layer is a MAH-grafted ethylene-based polymer. In a further embodiment, the MAH-grafted ethylene-based polymer has a melt index (I2) from 0.5 to 10 g/10 min, or from 1 to 6 g/10 min.

In one embodiment, the functionalized MAH-grafted ethylene-based polymer comprises at least one functional group selected from the following:



, anhydride, and combinations thereof; and wherein R is hydrogen or alkyl, R' is hydrogen or alkyl. In a further embodiment, each alkyl group is, independently, methyl, ethyl, propyl or butyl. In one embodiment, the functionalized ethylene-based polymer is selected from a functionalized ethylene homopolymer or a functionalized ethylene/alpha-olefin interpolymers. In a further embodiment, the functionalized ethylene-based polymer is a functionalized ethylene homopolymer. In another embodiment, the functionalized ethylene-based polymer is a functionalized ethylene/alpha-olefin interpolymers, and further a functionalized ethylene/alpha-olefin copolymer. Preferred alpha-olefins include C3-C8 alpha-olefins, and further propylene, 1-butene, 1-hexene and 1-octene.

The addition of low levels of maleic anhydride grafted polymer into the tie layer is known in the art. The addition of a sufficient level of maleic anhydride grafted polymer to the tie layer to enable compatibilization can diminish the adhesion properties of the tie layer.

POLAR LAYER

The multi-layer structure also comprises a polar, or barrier layer comprising a polar polymer.

The term "polar polymer," as used herein, refers to polymer formed from at least one monomer that comprises at least one heteroatom. Some examples of heteroatoms include O, N, P and S.

In various embodiments, the polar polymer has a melt index (I2) (2.16 kg, 190°C) from 0.1 to 40 g/10 min, further from 0.2 to 20 g/10 min, and further from 0.5 to 10 g/10 min.

In various embodiments, the polar polymer has a density from 1.00 to 1.30 g/cc, further from 1.10 to 1.20 g/cc (1 cc = 1 cm³).

In various embodiments, the polar polymer is selected from an ethylene vinyl alcohol polymer (EVOH) (such as Eval H171B sold by Kuraray) or polyamide (PA) (such as Nylon 6, Nylon 66, and Nylon 6/66 sold by DuPont) and combinations thereof.

In various embodiments, the at least one polar layer comprises at least one layer of EVOH.

In various embodiments, the at least one polar layer comprises at least one layer comprising a nylon selected from the group consisting of nylon 6, nylon 66, nylon 6/66 and combinations thereof.

In various embodiments, the polar layer comprises at least one layer of at least one of
5 the above-mentioned nylon compounds and at least one layer of EVOH.

In various embodiments, the polar layer can further comprise up to 15 weight percent of the compatibilizer described above. Any and all ranges up to 15 weight percent are included herein and disclosed herein, for example, the polar layer can comprise 0.25 weight
10 percent, 0.5 weight percent, 1 weight percent, 5 weight percent, 6 weight percent, 8 weight percent, or 10 weight percent of the compatibilizer component.

To one skilled in the art it is known that the incorporation of a recycle compatibilizer component could be added to any layers of the multi-layer structure. Surprisingly, addition to specific layers is required to achieve the targeted film performance. The addition of low
15 levels of maleic anhydride grafted polymer into the polar polymer layer is known in the art. The addition of a sufficient level of maleic anhydride grafted polymer to the polar polymer layer to enable compatibilization can diminish the properties of the polar polymer.

The figures show several embodiments of the present invention. Fig. 1 shows a three-layer structure, with a polyolefin layer comprising a polyethylene and compatibilizer
20 component, a tie layer, and a polar layer comprising EVOH or PA.

Figs. 2a and 2b each show 5-layer structures. In Fig. 2a, there is a polyolefin layer comprising a polyethylene and a compatibilizer component, a tie layer, a polar layer comprising EVOH or PA, a second tie layer, and a second polyolefin layer comprising
25 EVOH or PA. Fig. 2b shows a 5-layer structure with a polyolefin layer comprising a polyethylene, a tie layer, a polar layer comprising EVOH or PA, a second tie layer, and a polyolefin layer comprising a polyethylene and a compatibilizer component.

Figs. 3a and 3b show 7-layer structures. In Fig. 3a, there is a polyolefin layer comprising a polyethylene, a polyolefin layer comprising a polyethylene and a compatibilizer component, a tie layer, a polar layer comprising EVOH or PA, another tie layer, a polyolefin
30 layer comprising a polyethylene and a compatibilizer component, and a polyolefin layer comprising a polyethylene. Fig. 3b shows a 7-layer structure with a polyolefin layer comprising a polyethylene and a compatibilizer, a tie layer, a polar layer comprising PA, a

polar layer comprising EVOH, another polar layer comprising PA, another tie layer, and another polyolefin layer comprising a polyethylene and a compatibilizer component.

Figs. 4a, 4b, and 4c show 9-layer structure embodiments. In Fig. 4a, there is a polyolefin layer comprising a polyethylene, a polyolefin layer comprising a polyethylene and a compatibilizer component, a tie layer, a polar layer comprising PA, a polar layer comprising EVOH, a second polar layer comprising PA, a second tie layer, a second polyolefin layer comprising a polyethylene and a compatibilizer component, and a second polyolefin layer comprising a polyethylene. Fig. 4b shows a 9-layer structure with a polyolefin layer comprising a polyethylene, a second polyolefin layer comprising a polyethylene, a polyolefin layer comprising a polyethylene and a compatibilizer component, a tie layer, a polar layer comprising EVOH or PA, a second tie layer, a second polyolefin layer comprising polyethylene and a compatibilizer component, a third polyolefin layer comprising a polyethylene, and a fourth polyolefin layer comprising a polyethylene. Fig. 4c shows a nine-layer structure with a polyolefin layer comprising a polyethylene, a polyolefin layer comprising a polyethylene and a compatibilizer component, a second polyolefin layer comprising a polyethylene, a tie layer, a polar layer comprising EVOH or PA, a second tie layer, a third polyolefin layer comprising a polyethylene, a second polyolefin layer comprising a polyethylene and a compatibilizer component, and a fourth polyolefin layer comprising a polyethylene.

The multi-layer structures can be used for a variety of applications including, but not limited to film applications such as flexible films, semi-flexible films, rigid films, and semi-rigid films. Films can be prepared by any method known to those skilled in the art. The multi-layer structures or self-recyclable films disclosed herein may be converted into films, sheet, or rigid structures.

RECYCLING METHOD

In various embodiments, this invention discloses a method for making a recycled structure comprising a) converting the multi-layer structure of any previous embodiment into flakes, b) optionally converting the flakes into pellets, and c) forming the recycled structure from the flakes or the pellets wherein the method does not involve the addition of a compatibilizer component during this unit operation.

In another embodiment, this invention discloses a method for making a recycled structure comprising a) continuously transferring the edge trim from a blown or cast film

process directly back into the feed stream, b) optionally combining the trim with other polymers to form a layer in a multi-layer structure.

The recycled structure can be formed from the flakes and/or pellets by undergoing any useful process known to those skilled in the art. These processes include, but are not limited to an extrusion process, a blown-film process, a cast-film process, and combinations thereof.

EXAMPLES

Materials

Materials are shown in Table 1 below.

Table 1. Materials used in the Examples

Material	Density (g/cm ³)	Melt Index (g 10/min)	Comments
RETAIN 3000	0.87	660	Recycle Compatibilizer (RC). Maleic anhydride (MAH) grafted ethylene-octene plastomer
Dowlex 5056G	0.919	1.1	LLDPE
AMPLIFY™ TY 1353	0.921	2.0	MAH grafted polymer
EVAL F171B	1.19	1.6	Ethylene Vinyl-Alcohol Copolymer. 32 mol % Ethylene content

EXAMPLES 1-3 AND COMPARATIVE EXAMPLES A-C

SAMPLE PREPARATION AND RECYCLING PROCESS

A control multi-layer film sample which contained EVOH, tie layers, and outer polyolefin layers is compared to a self-recyclable multi-layer structure which contained EVOH, tie layers, and an outer polyolefin layer that was a blend of polyolefin and the recycle compatibilizer, Table 2. Both films are 5 layer A/B/C/B/A structures with an overall thickness of 100 microns, distributed as 42.5/5/5/5/42.5 micron layers. These structures were produced on a Collin co-extrusion blown film line. These samples were designated as the “original film structures”, since they would be later submitted to a recycling process and correspond to the structure depicted in Fig. 2a. Table 2 shows the designed structures.

Table 2. Designed original film structures

Structures	Layer	Comparative Example A	Inventive Example 1
A/B/C/B/A 100 μ 42.5 / 5 / 5 / 5 / 42.5 μ (42.5 / 5 / 5 / 5 / 42.5 %)	A	Dowlex 5056G	94% Dowlex 5056G + 6% RETAIN 3000
	B	Amplify TY 1353	Amplify TY 1353
	C	EVAL F171B	EVAL F171B

The ratio EVAL F171B : RETAIN 3000 for this study was 1:1, meaning that the amount of EVOH within these structures was 5%, while the amount of built-in compatibilizer within the self recyclable structure was 5% which was evenly split between the two outer layers.

5

The original film structures were ground in a Mateu & Sole grinder machine, a machine composed of rotary blades designed to reduce fed film into flakes of approximately 0.5 x 0.5 cm, by cutting and compressing the film against the drum.

Once the films were converted into flakes, they were then processed on a Buss kneader-compounder in order to convert the flakes to pellets.

10

Monolayer and 3 layer film structures were produced on a Collin co-extrusion blown film line using a 2.5 BUR and a 1.8mm die gap. Prior to the extrusion process, the pellets were dried overnight in a lab oven at 100°C to remove any remaining moisture.

The films produced are shown in Table 3.

15

Table 3. Recycled Monolayer and 3 Layer Films

Monolayer Films 50 μ		3 Layer films (A/B/A) 50 μ (15/20/15 μ)	
Comparative Example B	Inventive Example 2	Comparative Example C	Inventive Example 3
100% recycled Comparative Example A	100% recycled Inventive Example 1	Layer A = Dowlex 5056 Layer B = Recycled Comparative Example A	Layer A = Dowlex 5056 Layer B = Recycled Inventive Example 1

The extrusion conditions for these samples are shown in Table 4.

20

Table 4. Extruding Conditions Collin co-extrusion blown film line

	Units	Comp Ex A	Inv Ex 1	Comp Ex B	Inv Ex 2	Comp Ex C	Inv Ex 3
Amps - Ext. A	A	4	2.7	8	8.3	3.3	3.4
Amps - Ext. B	A	2.2	2.4	-	-	5.2	6.3
Amps - Ext. C	A	1	0.9	-	-	3.3	3.1
Amps - Ext. D	A	3.7	3.1	-	-	-	-
Layer Percentage - Ext. A	%	42	42	100	100	30	30
Layer Percentage - Ext. B	%	10	10	-	-	40	40
Layer Percentage - Ext. C	%	5	5	-	-	30	30
Layer Percentage - Ext. D	%	42	42	-	-	-	-
Melt Pressure - Ext. A	bar	213	179	278	273	203	208
Melt Pressure - Ext. B	bar	48	44	-	-	184	214
Melt Pressure - Ext. C	bar	52	46	-	-	162	165
Melt Pressure - Ext. D	bar	173	155	-	-	-	-
Melt Temperature - Ext. A	°C	248	248	221	223	235	234
Melt Temperature - Ext. B	°C	235	235	-	-	221	203
Melt Temperature - Ext. C	°C	225	226	-	-	228	228
Melt Temperature - Ext. D	°C	242	241	-	-	-	-
RPM - Ext. A	rpm	90	90	85	85	68	65
RPM - Ext. B	rpm	12	12	-	-	45	45
RPM - Ext. C	rpm	6	5	-	-	68	65
RPM - Ext. D	rpm	92	92	-	-	-	-
Take-off speed	m/min	3.4	3.3	5.2	5.2	6.8	6.8
Total Output	kg/h	10	10	8	8	10	10

Table 5 shows the operating conditions for the Buss compounder.

5

Table 5. Operating conditions for Buss kneader compounder

	Units	Pellets derived from Comp Ex A	Pellets derived from Inv Ex 1
Kneader Motor Current	A	13	13
Kneader Speed	Rpm	120	120
Motor Current	A	3	3
Output Rate	Kg/h	4	4
Screw Speed	Rpm	45	30
Thermocouple K1	°C	75	75
Thermocouple K2	°C	150	150
Thermocouple K3	°C	170	170
Thermocouple K5	°C	130	130
Thermocouple K4	°C	135	135

The mechanical and optical properties of the recycled monolayer structures are presented in Table 6.

Table 6. Properties of monolayer recycled films

Test	Comparative Example B	Inventive Example 2
Dart Drop Impact (g)	342	305
Elmendorf Tear CD (g)	1.060	1.320
Elmendorf Tear MD (g)	893	1.050
Gloss 45° (Gloss)	30,4	64,5
Haze (%)	30,9	13,1

5

The physical properties for the Example B and the Inventive Example 2 are compared. The CD and MD tear properties are improved. The optical properties measured, Gloss and Haze, are significantly improved in Inventive Example 2 without the inclusion of a distinct unit operation step where the recycle compatibilizer is added.

10 Table 7 shows the mechanical and optical properties of the 3 layer film structures with recycled materials in the core layer.

Table 7. Properties of 3 layer films with recycled material in the core layer

Test	Comparative Example C A/B/A A = Dowlex 5056 B = Recycled Comparative Example A	Inventive Example 3 A/B/A A = Dowlex 5056 B = Recycled Inventive Example 1
Dart Drop Impact (g)	377	319
Elmendorf Tear CD (g)	1.160	1.210
Elmendorf Tear MD (g)	984	1.020
Gloss 45° (Gloss)	78.4	76.9
Haze (%)	7.9	7.2

15

When comparing the 3 layer structure with the recycled control sample (Comparative Example A) and the 3 layer structure with the Inventive Example 1 in the core layer, an improvement in mechanical properties can be observed.

Other test methods used in these examples are listed in Table 8.

20

Table 8. Mechanical Properties Tests

Test	Method
Dart Drop Impact	ASTM D 1709
Elmendorf Tear	ASTM D 1922
Gloss 45°	ASTM D2457
Haze	ASTM D1003
Density	ASTM 792-08
Melt Index	ISO 1133

EXAMPLES 4-7 AND COMPARATIVE EXAMPLES D-E**SAMPLE PREPARATION AND RECYCLING PROCESS**

A control multi-layer film simple which contained EVOH, tie layers and outer polyolefin layers is compared to two self recyclable multilayer structures which contained EVOH, tie layers and outer layers comprised of polyolefin and a blend of polyolefin and the recycle compatibilizer, as seen in Table 9.

All films are 7 layer A/B/C/D/C/B/A structures with and overall thickness of 100 microns distributed as 20/22,5/5/5/5/22,5/20 micron layers. These structures were produced on a Alpine co-extrusion blown film line. These structures are designated as the “original films structures”, since they would be later submitted to a recycling process.

Table 9. Original film structures

Structures	Layer	Comparative Example D	Inventive Example 4	Inventive Example 5
A/B/C/D/C/B/A 100 μ 20 /22,5 / 5 / 5 / 5 / 22.5 /20 μ (20 /22,5 / 5 / 5 / 5 / 22.5 /20 %)	A	Dowlex 2056G	94% Dowlex 2056 + 6% RETAIN 3000	Dowlex 2056G
	B	Dowlex 2056G	94% Dowlex 2056 + 6% RETAIN 3000	89% Dowlex 2056 + 11% RETAIN 3000
	C	Amplify TY 1353	Amplify TY 1353	Amplify TY 1353
	D	EVAL F171B	EVAL F171B	EVAL F171B

The ratio EVAL F171B : RETAIN 3000 was 1:1 in both of the inventive examples, meaning that the amount of EVOH within these structures was 5% while the amount of built-in compatibilizer within the self recyclable structures was 5%. In the first example, the compatibilizer was evenly split between the two B layers and in the second example it was evenly split between the four outer layers (A and B).

The original film structures were ground up to produce film flakes and then processed on a Buss kneader-compounder in order to convert the film flakes into pellets.

The produced pellets were dried overnight in an oven at 100°C to remove any remaining moisture and then Monolayer films were produced on a Collin blown film line. The films produced are shown in Table 10.

Table 10. Recycled monolayer films

Recycled monolayer films 50 μ		
Comparative Example E	Inventive Example 6	Inventive Example 7
100% recycled Comparative Example D	100% recycled Inventive Example 4	100% recycled Inventive Example 5

The gel count data (a gel is considered as an imperfection in the films) of the recycled monolayer structures was measured using the Dow method DOWM 101898-I12H and is presented in Table 11.

5

Table 11. Gel count data

gel size (μ)	Comparative Example E (Gel/m ²)	Inventive Example 6 (Gel/m ²)	Inventive Example 7 (Gel/m ²)
64	763,67	34,98	237,90
100	422,32	34,98	119,95
200	515,78	50,48	156,43
300	139,44	20,49	50,98
400	41,48	15,49	19,99
500	13,99	5,00	10,00
800	8,5	4,50	6,00
1000	1,00	0	1,50
1200	0	0	0
1200	0	0	0
Total	1906,18	165,92	602,75

Gel data for Comparative Example E and Inventive Examples 6 and 7 were compared. Gel level in the recycled films was significantly reduced, and thus, increasing the quality of the recycled films without the inclusion of a distinct unit operation step where the recycle compatibilizer was added.

10

CLAIMS

1. A multi-layer structure comprising:

a) at least one layer comprising a polyolefin component comprising

5 i) 60 to 94 weight percent of a first component selected from the group consisting of ethylene homopolymer, ethylene copolymer, polypropylene homopolymer, polypropylene copolymer, and combinations thereof

ii) 0 – 35 weight percent of a functional polymer component, and

10 iii) 1 -35 weight percent of a compatibilizer component comprising an anhydride and/or carboxylic acid functionalized ethylene/alpha-olefin interpolymer having a melt viscosity (177°C) less than, or equal to, 200,000 cP and a density from 0.855 to 0.94 g/cc;

b) at least one tie layer comprising maleic-anhydride grafted polymer with a melt index of less than 50 dg/min, wherein the tie layer does not contain the compatibilizer component ; and

15 c) at least one polar layer comprising a polar polymer .

2. The multi-layer structure of claim 1, wherein the ratio of the compatibilizer component to the polar polymer is in the range of 0.2 – 4.0 : 1.0 by weight.

3. The multi-layer structure of claims 1 or 2, wherein the polar layer comprises a layer of EVOH.

20 4. The multi-layer structure of any of the preceding claims, wherein the polar layer comprises a layer comprising a nylon selected from the group consisting of nylon 6, nylon 66, nylon 6/66 and combinations thereof.

5. The multi-layer structure of claim 1, wherein the polar layer comprises at least one layer of EVOH and at least one layer of a nylon selected from the group consisting of
25 nylon 6, nylon 66, nylon 6/66 and combinations thereof.

6. The multi-layer structure of any of the preceding claims, wherein the ethylene homopolymer or ethylene alpha-olefin copolymer is selected from the group consisting of homopolymer polyethylene, an ethylene alpha-olefin copolymer, polypropylene, random polypropylene, and mixtures thereof.

30 7. The multi-layer structure of claim 6, wherein the polyolefin component comprises a blend of 5 to 30 weight percent of a sealant polymer and 70 to 95 weight percent of the ethylene homopolymer or ethylene alpha-olefin copolymer.

8. The multi-layer structure of any of the preceding claims wherein the functional polymer component is selected from the group consisting of a maleic anhydride functionalized polyolefin, acrylate ethylene copolymer, ethylene vinyl acetate, and mixtures thereof.
- 5 9. An article prepared from the multi-layer structure of claim 1.
10. A method for making a recycled structure comprising
- a) converting the multi-layer structure of claim 1 into flakes
 - b) optionally converting the flakes into pellets
 - c) forming the recycled structure from the flakes or the pellets
- 10 wherein the method does not involve the use of the compatibilizer component.
11. The method of claim 10, wherein the flakes and/or pellets form the recycled structure in step c) by i) continuously transferring edge trim from a blown or cast film process directly back into the feed stream, and ii) optionally combining the trim with other polymers to form a layer in a multi-layer structure.
- 15 12. A film having at least one layer which comprises the recycled structure of claim 10.

1/2

FIG. 1

PE + Compatibilizer
Tie
EVOH or PA

FIG. 2A

PE + Compatibilizer
Tie
EVOH or PA
Tie
PE + Compatibilizer

FIG. 2B

PE
Tie
EVOH or PA
Tie
PE + Compatibilizer

FIG. 3A

PE
PE + Compatibilizer
Tie
EVOH or PA
Tie
PE + Compatibilizer
PE

FIG. 3B

PE + Compatibilizer
Tie
PA
EVOH
PA
Tie
PE + Compatibilizer

2/2

FIG. 4A

PE
PE + Compatibilizer
Tie
PA
EVOH
PA
Tie
PE + Compatibilizer
PE

FIG. 4B

PE
PE
PE + Compatibilizer
Tie
EVOH or PA
Tie
PE + Compatibilizer
PE
PE

FIG. 4C

PE
PE + Compatibilizer
PE
Tie
EVOH or PA
Tie
PE
PE + Compatibilizer
PE

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2015/059972

A. CLASSIFICATION OF SUBJECT MATTER INV. B32B27/08 B32B27/30 B32B27/32 ADD.		
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) B32B		
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 practicable, search terms used) EPO-Internal, COMPENDEX, WPI Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2010/034456 A1 (BOREALIS AG [AT]; OEYSAED HARRY [NO]; GAHLEITNER MARKUS [AT]; SKAU KAR) 1 April 2010 (2010-04-01) claims 1,6 -----	1-12
X	WO 2012/164585 A2 (ESSEL PROPACK LTD [IN]; BHATTACHARYA SATYA PRASAD [IN]; BANERJEE MRINA) 6 December 2012 (2012-12-06) claims 1,15 -----	1-12
☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
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Date of the actual completion of the international search 20 January 2016	Date of mailing of the international search report 27/01/2016	
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Hillebrand, Gerhard	

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2015/059972

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 2010034456 A1	01-04-2010	CN 102159396 A EP 2340165 A1 US 2011236708 A1 WO 2010034456 A1	17-08-2011 06-07-2011 29-09-2011 01-04-2010
WO 2012164585 A2	06-12-2012	CN 104093568 A EP 2714395 A2 US 2014170399 A1 WO 2012164585 A2	08-10-2014 09-04-2014 19-06-2014 06-12-2012

PATENT COOPERATION TREATY

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Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference see form PCT/ISA/220		FOR FURTHER ACTION See paragraph 2 below
International application No. PCT/US2015/059972	International filing date (day/month/year) 10.11.2015	Priority date (day/month/year) 31.12.2014
International Patent Classification (IPC) or both national classification and IPC INV. B32B27/08 B32B27/30 B32B27/32		
Applicant DOW GLOBAL TECHNOLOGIES LLC		

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:  European Patent Office P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Fax: +31 70 340 - 3016	Date of completion of this opinion see form PCT/ISA/210	Authorized Officer Hillebrand, Gerhard Telephone No. +31 70 340-0
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Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:
 - ☒ the international application in the language in which it was filed.
 - ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).
2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))
3. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:
 - a. ☐ forming part of the international application as filed:
 - ☐ in the form of an Annex C/ST.25 text file.
 - ☐ on paper or in the form of an image file.
 - b. ☐ furnished together with the international application under PCT Rule 13ter.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.
 - c. ☐ furnished subsequent to the international filing date for the purposes of international search only:
 - ☐ in the form of an Annex C/ST.25 text file (Rule 13ter.1(a)).
 - ☐ on paper or in the form of an image file (Rule 13ter.1(b) and Administrative Instructions, Section 713).
4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
5. Additional comments:

Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	<u>1-12</u>
	No: Claims	
Inventive step (IS)	Yes: Claims	
	No: Claims	<u>1-12</u>
Industrial applicability (IA)	Yes: Claims	<u>1-12</u>
	No: Claims	

2. Citations and explanations

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Reference is made to the following documents; the numbering will be adhered to in the rest of the procedure.

- D1 WO 2010/034456 A1 (BOREALIS AG [AT]; OEYSAED HARRY [NO]; GAHLEITNER MARKUS [AT]; SKAU KAR) 1 April 2010 (2010-04-01)
- D2 WO 2012/164585 A2 (ESSEL PROPACK LTD [IN]; BHATTACHARYA SATYA PRASAD [IN]; BANERJEE MRINA) 6 December 2012 (2012-12-06)

1. Inventive step(Art. 33(3) PCT)

a)

Document D1 discloses a multi-layer structure comprising a polyolefin layer comprising a compatibiliser and a barrier layer of EVOH. In addition D1 discloses the compatibiliser being an acid or anhydride modified polyolefin(claims 1 and 6).

The difference of claim 1 of the present application versus D1 is that D1 does not disclose an additional tie layer comprising a maleic-grafted anhydride grafted polymer. In addition the present application claims a specific compatibilizer component, which is also not disclosed in D1.

Tie layer made of maleic-anhydride grafted polymers are well known in the prior art.

Document D2 discloses a tie layer of maleic anhydride grafted polyethylene(claims 1 and 15) applied in a multilayer structure between polyolefin and polar layers.

Thus the subject-matter of claims 1,3 and 9 is not inventive in view of the prior art.

b)

The remaining claims, which are not mentioned under point 1.a), do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step, the reasons being as follows:

The features of these dependent claims are merely selected from several straight forwarded possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed.

2. Others

a)

To meet the requirements of the PCT, the document D1 should be identified in the description and the relevant background art disclosed therein should be briefly summarised in an objective way.

b)

The title of the application is misleading and not supported by the claims.

The claim comprised in the title "Self-recyclable barrier packaging" is not supported by the present set of claims, because the present set of claims refer to a multi-layer structure and not to a packaging.

In addition these claims comprise no disclosure how such a multi-layer structure will be re-obtained by following the claimed method of claim 10 in which the structure is cut into flakes and pellets. Obviously the same well defined multi-layered structure comprising a specific order of the layers within said structure will not be obtained by using such pellets as a starting material for making a new structure, because the pellets will be randomly fused together and thus also the order of the layers will appear randomly.