CLAIMS:

1. A polymer blend, comprising:

a plurality of linear ethylene interpolymer blend components, each having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and a composition distribution breadth index of at least 50%;

wherein the blend is essentially free of blend components having both a higher average molecular weight and a lower average comonomer content than that of any other said polyethylene blend component; and

wherein the blend has either $M_w/M_n > 3$ or a composition distribution breadth index below 50%, or both.

2. The blend of claim 1, wherein said blend has a density in the range of 0.85 to 0.94 g/cm^3.

3. The blend of claim 1, wherein said blend components have an α-olefin comonomer content in the range of 0 to 30 mole percent and an ethylene content in the range of 100 to 70 mole percent.

4. The blend of claim 1, wherein said blend components are selected from the group consisting of:

poly(ethylene-co-α-olefin) elastomers; very low density linear polyethylenes; and linear, low density polyethylenes.

5. The blend of claim 1, wherein the blend components have essentially the same molecular weight and different comonomer contents.

6. The blend of claim 1, wherein the blend components have essentially the same comonomer content and different molecular weights.

7. The blend of claim 1, wherein the molecular weights and the comonomer contents of the blend components are both different.

8. The blend of claim 1, wherein the value of M_w/M_n for each blend component is less than or equal to 2.5.

9. The blend of claim 1, wherein the composition distribution breadth index for each blend component is greater than about 70%.

10. A polymer blend, comprising:

a plurality of linear ethylene polymer or interpolymer blend components, each having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and a composition distribution breadth index of at least 50%; and

wherein said blend components are selected from one of the groups consisting of: (1) blend components having essentially the same average molecular weight but different average comonomer contents; (2) blend components having essentially the same average comonomer content but different average molecular weights; and (3) blend components having different average molecular weights and comonomer contents wherein said components, taken serially in order of increasing average molecular weight, have an increasing comonomer content.

11. The blend of claim 10, wherein the blend has a density of 0.85 to 0.94 g/cm^3.

12. The blend of claim 10, wherein said blend has an average molar α-olefin comonomer content of 1-30 percent.

13. The blend of claim 10, wherein said blend components include poly(ethylene-co-α-olefin) elastomers.

14. The blend of claim 12, wherein said blend components include very low density, linear polyethylenes.

15. The blend of claim 12, wherein said blend components include linear, low density polyethylenes.

16. The blend of claim 10, wherein the number of said blend components is from 2 to 5.

17. The blend of claim 10, wherein said blend is essentially free of blend components having both a higher average molecular and a lower average comonomer content than another blend component.

18. The blend of claim 10, wherein the blend has plural modality with respect to molecular weight, comonomer content or both.

19. The blend of claim 12, wherein said α-olefins have from 3 to about 12 carbon atoms.

20. The blend of claim 10, wherein $M_w/M_n \leq 2.5$ for each blend component, and the composition distribution breadth index for each component is in the range from 50 to 70%.

21. The blend of claim 20, wherein $M_w/M_n \leq 2.0$ for each blend component.

22. The blend of claim 20, wherein each blend component has a composition distribution breadth index in the range from 50 to 90%.

23. A polymer blend exhibiting enhanced tear strength properties over that of the individual blend components comprising:

a plurality of linear ethylene interpolymer blend components, each having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and a composition distribution breadth index of at least 50%; and

wherein for said components taken serially, primarily in order of increasing molecular weight and secondarily in order of increasing average comonomer content, each succeeding component has substantially the same or a higher average comonomer content than each preceding component in said series.

24. A polymer blend of linear ethylene interpolymer having a molecular weight distribution such that $M_w/M_n > 2$ and a composition distribution breadth index greater than or equal to 50%.

25. A linear ethylene interpolymer comprising fractions having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ for each fraction, said fractions taken serially in order of increasing average molecular weight have an increasing average comonomer content.

26. A linear ethylene interpolymer comprising fractions having a composition distribution breadth index of 50% or more, said fractions taken serially in order of increasing comonomer content have an increasing average molecular weight.

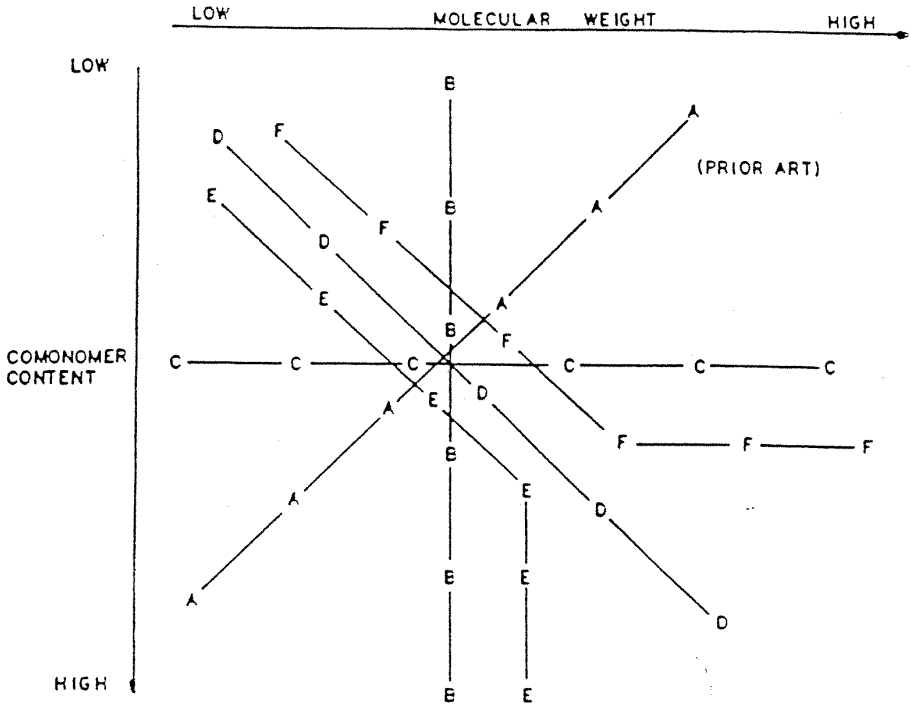

LOW
MOLECULAR WEIGHT
HIGH
LOW
COMONOMER CONTENT
HIGH
(PRIOR ART)
A
B
C
D
E
F

FIG. 1

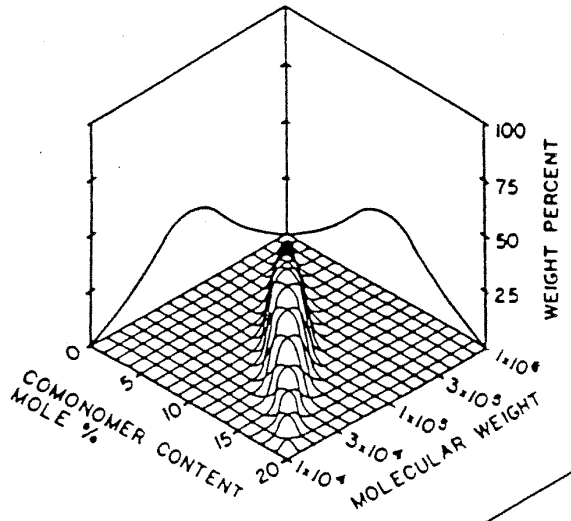
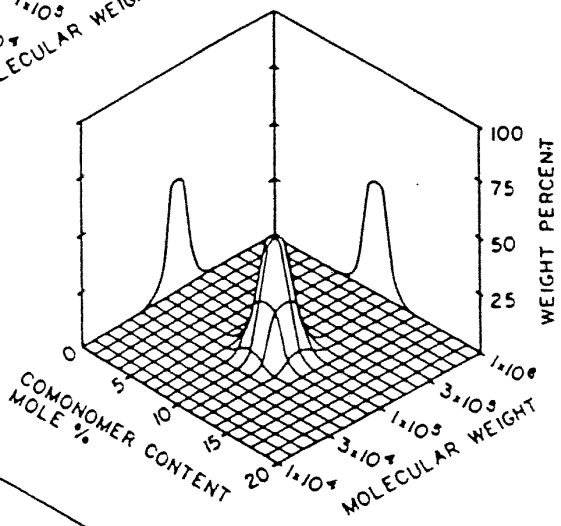


FIG. 2

FIG. 3



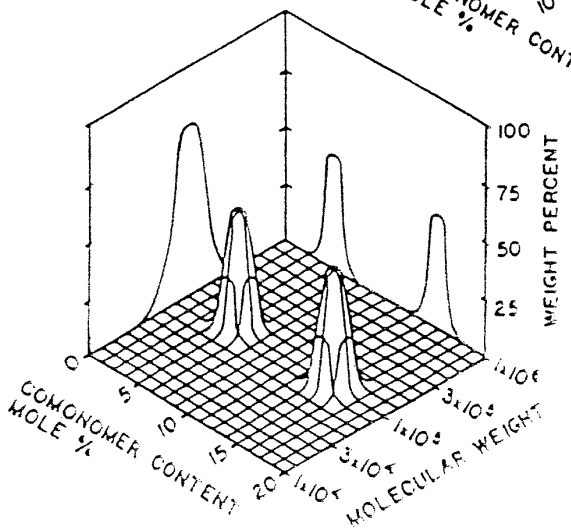


FIG. 4

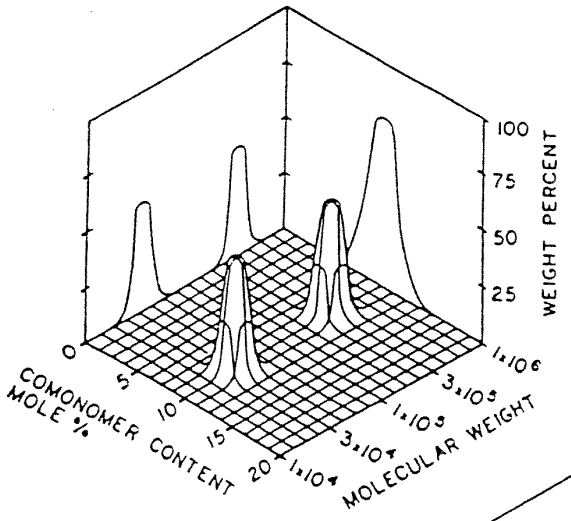
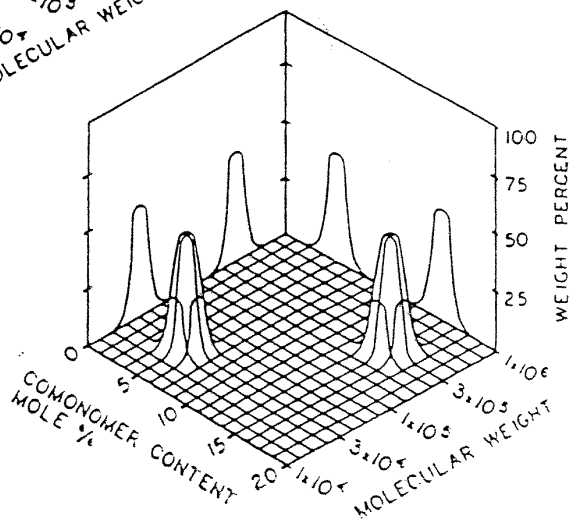

100
75
50
25
WEIGHT PERCENT
0
5
10
15
20
COMONOMER CONTENT
MOLE %
1x10⁶
3x10⁵
1x10⁵
3x10⁴
1x10⁴
MOLECULAR WEIGHT

FIG. 5

FIG. 6


100
75
50
25
WEIGHT PERCENT
0
5
10
15
20
COMONOMER CONTENT
MOLE %
1x10⁶
3x10⁵
1x10⁵
3x10⁴
1x10⁴
MOLECULAR WEIGHT

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 89/04259

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC5: C 08 L 23/08

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
IPC5	C 08 L

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
A	EP, A1, 0100843 (PHILIPS PETROLEUM COMPANY) 22 February 1984, see the whole document	1-26
A	EP, A2, 0168928 (BCL LIMITED) 22 January 1986, see the whole document	1-26
A	EP, A2, 0241234 (EXXON CHEMICAL PATENTS INC.) 14 October 1987, see the whole document	1-26
A	GB, A, 2077273 (NIPPON OIL COMPANY, LIMITED) 16 December 1981, see the whole document	1-26

* Special categories of cited documents: [10]

"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

"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

IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
21st December 1989	08. 01. 90
International Searching Authority	**Signature of Authorized Officer**
EUROPEAN PATENT OFFICE	T.K. WILLIS

Form PCT/ISA/210 (second sheet) (January 1985)

International Application No. PCT/US 89/04259

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
A	GB, A, 2148906 (OWENS-ILLINOIS INC (USA-DELAWARE)) 5 June 1985, see the whole document	1-26

--

Form PCT/ISA/210 (extra sheet) (January 1985)

ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO. PCT/US 89/04259

SA 31622

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
The members are as contained in the European Patent Office EDP file on 08/11/89
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A1- 0100843	22/02/84	JP-A- 59047242	16/03/84
		US-A- 4461873	24/07/84
		US-A- 4547551	15/10/85
		CA-A- 1216392	06/01/87
EP-A2- 0168928	22/01/86	GB-A-B- 2159091	27/11/85
EP-A2- 0241234	14/10/87	US-A- 4728325	01/03/88
		JP-A- 63249703	17/10/88
GB-A- 2077273	16/12/81	FR-A-B- 2481709	06/11/81
		JP-A- 56152853	26/11/81
		DE-A- 3117240	11/03/82
		US-A- 4413097	01/11/83
		CA-A- 1160792	17/01/84
GB-A- 2148906	05/06/85	AU-D- 34650/84	09/05/85
		FR-A-B- 2554454	10/05/85
		DE-A-C- 3437116	23/05/85
		JP-A- 60124643	03/07/85
		AU-A- 549622	06/02/86
		US-A- 4577768	25/03/86

EPO FORM P079

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

FEB 10 2000 JC60 PATENT & TRADEMARK OFFICE

GP 1713

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Assistant Commissioner for Patents
Washington, D.C. 20231

ASSOCIATE POWER OF ATTORNEY

First Named Inventor: Pak-Wing S. Chum	Attorney Docket No.: 40121BU
Application No.: 08/927,393	Examiner: Harlan, R.
Filing Date: August 27, 1997	Group Art Unit: 1713
Title: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS	

In the matter of the above-identified patent application, please recognize the practitioner(s) below as Associate Attorney(s)/Agent(s) with full power of substitution and revocation, to make alterations and amendments therein, and to transact all business in the U. S. Patent & Trademark Office in connection therewith, according to the box checked below:

#9

☒ **Practitioners at Customer No.:** 022515 PATENT TRADEMARK OFFICE

Please change the correspondence address for the above-identified application to that of the above-mentioned Customer Number.

Please direct all future telephonic communications to Osborne K. McKinney at (409) 238-7889.

☐ **Practitioner(s) named below:**

Name	Registration Number

Please change the correspondence address and direct all future telephonic communications for the above-identified application to:

NAME					
ADDRESS					
CITY		STATE		ZIP CODE	
TELEPHONE		FAX			

RECEIVED FEB 16 2000 TC 1700 MAIL ROOM

Signature: S. P. Krupp

Name: Stephen P. Krupp
Registration No.: 34,366
Date: February 3, 2000

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: ASSISTANT COMMISSIONER FOR PATENTS, WASHINGTON, DC 20231, ON: February 3, 2000

MaryAnn Navarrette

MaryAnn Navarrette

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant (s): Pak-Wing S. Chum, et al.

Serial No.: 08/927,393 Group Art Unit: 1713

Filed: August 27, 1997 Examiner: Harlan, R.

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS



I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: ASSISTANT COMMISSIONER FOR PATENTS, WASHINGTON, DC 20231, ON:

February 3, 2000
DATE OF DEPOSIT

MaryAnn Navarrette
PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

MaryAnn Navarrette
SIGNATURE OF PERSON SIGNING CERTIFICATE

2-3-00
DATE OF SIGNATURE

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

TRANSMITTAL OF TERMINAL DISCLAIMER

To supplement RESPONSE A, mailed Janaury 31, 2000, Applicants submit hereunder a Terminal Disclaimer to disclaim the terminal part of any patent granted in the present application which would extend beyond the patent term of U.S. Patent No. 5,677,383. Applicants respectfully request reconsideration of the claimed invention in view of RESPONSE A and the Terminal Disclaimer submitted hereunder.

RECEIVED
FEB 15 2000
TC 1700 MAIL ROOM

Respectfully submitted,

Osborne K. McKinney
Registration No. 40,084
Phone: (409) 238-7889

Date: 02-03-00
2301 Brazosport Blvd., B-1211
Freeport, Texas 77541

OKM/man

1713

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant (s): Pak-Wing S. Chum, et al.

Serial No.: 08/927,393 Group Art Unit: 1713

Filed: August 27, 1997 Examiner: Harlan, R.

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

OIPE JC42 FEB 10 2000 PATENT & TRADEMARK OFFICE

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: ASSISTANT COMMISSIONER FOR PATENTS, WASHINGTON, DC 20231, ON:

January 31, 2000
DATE OF DEPOSIT

Osborne K McKinney
PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

SIGNATURE OF PERSON SIGNING CERTIFICATE

01-31-00
DATE OF SIGNATURE

RECEIVED FEB 16 2000 TC 1700 MAIL ROOM

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

EXTENSION OF TIME

Applicant(s) hereby request(s) that the shortened statutory period for response now set to expire December 30, 1999 in this application be extended for a period of one month(s), the extended period then expiring on January 31, 2000. This is a first request for an extension of time.

Please charge $110.00 to our Deposit Account No. 04-1512. If this estimate is incorrect, please charge or credit our account accordingly. Two duplicate copies of this sheet are enclosed.

02/11/2000 SLUANG 00000129 041512 08927393
01 FC:115 110.00 CH

Respectfully submitted,

Osborne K. McKinney
Registration No. 40,084
Phone: (409) 238-7889

Date: 01-31-02
2301 Brazosport Blvd., B-1211
Freeport, Texas 77541

40121BU

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

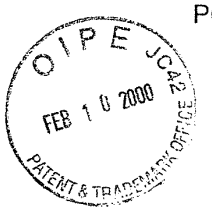
Applicants: Pak-Wing Steve Chum, et al.

Serial No.: 08/927,393
Filed: August 27, 1997
Attorney Docket No.: 40121BU

Art Unit: 1713
Examiner: Harlan, R.

#11
2/29/00

For: ETHYLENE POLYMER FILM MADE FROM ETHYLENE POLYMER BLENDS



I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: COMMISSIONER OF PATENTS AND TRADEMARKS, WASHINGTON, D.C. 20231, on

January 31, 2000
DATE OF DEPOSIT

Osborne K. McKinney
PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

SIGNATURE OF PERSON SIGNING CERTIFICATE

01-31-00
DATE OF SIGNATURE

RECEIVED
FEB 16 2000
TC 1700 MAIL ROOM

Hon. Commissioner of Patents & Trademarks
Washington, D.C. 20231

Sir:

RESPONSE A

The following is responsive to the Office Action mailed September 30, 1999 (paper number 8) wherein the PTO rejected all pending claims, Claims 36-51. Also submitted herewith is a petition for a one-month extension of time, extending the time for response to January 30, 2000. Applicants respectfully request reconsideration of the present invention in view of the remarks that follow.

The Rejection

1. Claims 36-51 stand rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as obvious over WO 90/03414 (WO '414).

The Examiner takes the position that although WO '414 fails to show the value of slope of strain hardening coefficient, in view of other substantially similar physical properties and processes for making the ethylene polymer, the Examiner has a reasonable basis to believe that the polymer inherently posses

the slope of strain hardening coefficient required by the presently claimed invention. And as such, the Examiner alleges the polymer blends disclosed in WO '414 are not necessarily different from the presently claimed polymer blends and the burden is on the Applicants to prove the claimed polymer blends are necessarily different and unobvious from those of WO '414.

The Examiner also takes the position that even if the properties of the claimed polymer composition and the prior examples in WO '414 are not the same, it would still have been obvious to one of ordinary skill in the art to make polymers having the claimed properties because it appears that the WO '414 generically embraces the claimed polymer blends and the person of ordinary skill in the art would have expected all embodiments of the reference to work; and that the Applicants have not demonstrated that the difference between the claimed product and prior art product give rise to unexpected results; and that the Markovich declaration is not persuasive as to unexpected results because according to the Examiner the comparative tests do not show unexpected of the claimed polymer as the toughness properties of the comparative runs are not necessarily inferior to those of inventive examples.

2. Claims 36-51 are also rejected under the judicially created doctrine of obvious-type double patenting as being unpatentable over Claims 1-18 of US Patent No. 5,677,383.

RECEIVED
FEB 16 2000
TC 1700 MAIL ROOM

APPLICANTS' RESPONSE

A. The claimed invention is <u>not</u> anticipated by WO '414 because the recited slope of strain hardening coefficient is <u>not</u> an inherent property

In formulating the rejection, the Examiner stated that there was a reasonable basis to believe the recited slope of strain hardening coefficient of the present invention is an inherent property of the '013 ethylene polymer described in WO '414.

Applicants do <u>not</u> believe the recited slope of strain hardening coefficient of the present invention is an inherent property of the homogeneously branched ethylene polymer (i.e. ethylene polymer '013) described in WO '414. More specifically, Applicants believe <u>not</u> all homogeneously branched ethylene polymers necessarily and consistently possess a slope of strain hardening coefficient value greater than or equal to 1.3. As evidence thereof, the Examiner's attention is directed to Table 1 at page 12 in the present specification. Table 1 clearly shows several homogeneously branched ethylene polymers which do <u>not</u> meet the recited requirement of the present invention. See Polymers A-D and S-V.

As other evidence that the recited slope of strain hardening coefficient value is not an inherent property, see Markovich's Third Declaration. The declaration shows three different homogeneously branched linear ethylene polymers that possess slope of strain hardening coefficient values that are outside of the recited requirement of the present invention. See Component Example B, Exact 3027 and Exact 3022 in Table 1 of the Markovich"s Third Declaration.

Because there is ample evidence of record that indicates not all homogeneously branched ethylene polymers have a slope of strain hardening coefficient equal to or greater than 1.3, the recited requirement of the present invention is clearly not an inherent property and, according, the Examiner's rejection under 35 U.S.C. § 102(b) should be withdrawn.

B. The present invention is nonobvious over WO '414 because the present invention provides unexpected results and comparative runs are necessarily and consistently inferior in regard to impact and tear performance.

The Examiner's attention is again directed to the Third Markovich Declaration. In Table 2, this declaration provides a comparison between Inventive Example AD1, comparative example BD1 and Inventive Example CD1 which clearly and convincingly demonstrates that the present invention provides unexpected results. Note from Table 1 of the declaration that the comparative example, BD1, has a SHC value of 1.0 (i.e., less the claimed value) and the two Inventive Examples, AD1 and CD1 have SHC values within the claimed range at 1.5 and 1.7, respectively. Note also from Table 2 that the Inventive Examples have substantially improved Dynatup impact performance and tear resistance relative to the comparative example. This result is unexpected because other than the difference in SHC values, the examples are the same. One of ordinary skill in the art would ordinarily measure the I_2, I_{10}/I_2, M_w/M_n and density properties and expect equivalent performance for such examples. The dramatic improvement provided by the present invention would be a completely unexpected surprise. Applicants believe that there is simply no way anyone could ever expect or predict that small slope of strain hardening differences between suitable and non-suitable Component (A) polymers would result in dramatic impact/toughness improvements when blended with a suitable Component (B) polymer.

Applicants believe the Examiner's position that the comparative runs are not necessarily inferior is untenable. While the Examiner does not provide much guidance, it seems the Examiner is concerned about the results reported for tensile break strength at 38 weight percent of the polymer representative of polymer '013 (and perhaps for tensile break energy at 72 weight percent of the polymer

representative of polymer '013). These are narrow, isolated instances where Inventive Examples and comparatives runs provide "similar" results. But the Applicants believe sameness of tensile properties in narrow, isolated instances does not and can not negate the superior to dramatically superior impact and tear resistances of the Inventive Examples as clearly shown in all of comparative tests. Note that as to impact and tear resistances, there is no instance where a comparative run is similar to any Inventive Example. Note also in regard to impact and tear resistances, the Inventive Examples are 18% to 716% better than their respective comparative runs; that is, with regard to impact and tear resistances, comparative runs have been shown to be consistently inferior.

Further note that for every narrow, isolated instance where a tensile property is reported as "similar" for a given weight percent combination, "dramatically superior" results are also reported for either impact resistance and/or tear resistance. For example, tear resistance as well as tensile break energy are dramatically superior (i.e. 56% to 118% higher and 256% to 793% higher, respectively) at 38 weight percent of the polymer representative of polymer '013 where tensile break strength is "similar". Applicants believe that these dramatic improvements 38 weight percent nullify any sameness as to tensile break strength.

As another example, tear resistance and impact resistance are "dramatically superior" (i.e. 121% to 237% higher and 48 to 127% higher, respectively) and tensile break strength is "superior" (i.e. 17% to 45% higher) at 72 weight percent of the polymer representative of polymer '013 where the tensile break energy is "similar". Again, the isolated instance of sameness for tensile break energy can not negate these dramatic improvements.

Applicants believe their showing of unexpected tear resistance and impact resistance results is thoroughly sufficient. If fact, the Applicants believe that it is even more surprising that impact and tear resistances are superior to dramatically superior where tensile properties are shown to be similar. Ordinarily one might expect product with similar tensiles properties to exhibit similar toughness properties.

In short, the Examiner obviousness rejection has been traversed because there is no requirement that the Inventive Examples be shown to be superior in all respects. The requirement is a showing of an unexpected result. But even if the Examiner's requirement is proper, Applicants have shown the comparative runs are consistently inferior in regard to tear and impact performance.

C. **Applicants respectfully request that the rejection under the judicial created doctrine of obviousness-type double patenting be held in abeyance.**

Applicants respectfully request that the obviousness-type double patent rejection be held in abeyance until claims of the present invention are found allowable by the Examiner. At that time, Applicants will promptly provide a terminal disclaimer to obviate the rejection.

Except the obviousness-type double patenting rejection, Applicants believe all outstanding rejections have been traversed and the claims are in a condition for allowance. Accordingly, Applicants respectfully request a notice of allowability as to Claims 36-51.

Respectfully submitted,

By _____
Osborne K. McKinney
Registration No. 40,084
Phone: (409) 238-7889

Dated: January 31, 2000
The Dow Chemical Company
Patent Department
Building B-1211
Freeport, Texas 77541
OKM/okm

ANNEX 1-10 attached
copy of Third Markovich Declaration
Flowchart of continuity chain for 08/834,050

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

#12

Applicant (s): **Pak-Wing S. Chum, et al.**

Serial No.:**08/927,393** Group Art Unit: **1713**

Filed:**August 27, 1997** Examiner: **Harlan, R.**

For: **FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS**



I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: ASSISTANT COMMISSIONER FOR PATENTS, WASHINGTON, DC 20231, ON:

February 3, 2000

DATE OF DEPOSIT

MaryAnn Navarrette

PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

MaryAnn Navarrette

SIGNATURE OF PERSON SIGNING CERTIFICATE

2-3-00

DATE OF SIGNATURE

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

TERMINAL DISCLAIMER

The undersigned registered patent attorney or agent states that:

(1) The undersigned is an attorney or agent of record in Application Serial No.08/927,393, filed on August 27, 1997, for "FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS" (the Present Application).

(2) The undersigned has reviewed the evidentiary assignment documents and, to his/her best knowledge and belief, The Dow Chemical Company, a Delaware corporation, is the sole and common assignee of the following patent properties:

(a) The Present Application:

[☒] The original documents are recorded at reel 8459, frame 0434.

[☐] The original documents have been submitted to Assignment Branch for recording. A copy of the assignment document(s) is submitted with this Disclaimer.

RECEIVED
FEB 15 2000
TC 1700 MAIL ROOM

(b) U.S. Patent No. 5,677,383:

[☒] The original documents are recorded at reel 8459, frame 0434.

[☐] The original documents have been submitted to Assignment Branch for recording. A copy of the assignment document(s) is submitted with this Disclaimer.

(3) On behalf of the assignee, the undersigned disclaims the terminal part of any patent granted on the Present Application which would extend beyond the expiration date for the full statutory term of U.S. Patent No. 5,677,383 and hereby agrees that any patent so granted on the above-identified application shall be enforceable only for and during such period that the legal title to said patent shall be the same as the legal title to U.S. Patent No. 5,677,383. This agreement runs with any patent granted on the Present Application and is binding upon the grantee, its successors or assigns.

(4) The undersigned does not disclaim any terminal part of any patent granted on the Present Application prior to the expiration date of the full statutory term of U.S. Patent No. 5,677,383 in the event that U.S. Patent No. 5,677,383 later: expires for failure to pay a maintenance fee, is held unenforceable, is found invalid, is statutorily disclaimed in whole or terminally disclaimed under 37 CFR 1.321, has all claims canceled by a reexamination certificate, or otherwise expires or is terminated prior to the expiration of its full statutory term, except for the separation of legal title stated above.

(5) Please charge $110.00 to our Deposit Account No. 04-1512. If this estimate is incorrect, please charge or credit our account accordingly. Two duplicate copies of this sheet are enclosed.

RECEIVED
FEB 16 2000
TC 1700 MAIL ROOM

Respectfully submitted,

Osborne K. McKinney
Registration No. 40,084
Phone: (409) 238-7889

Date: 02-03-00
2301 Brazosport Blvd., B-1211
Freeport, Texas 77541

OKM/man

OIPE
FEB 10 2000
PATENT & TRADEMARK OFFICE

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant (s): **Pak-Wing S. Chum, et al.**

Serial No.:**08/927,393** Group Art Unit: **1713**

Filed:**August 27, 1997** Examiner: **Harlan, R.**

For: **FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS**

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: ASSISTANT COMMISSIONER FOR PATENTS, WASHINGTON, DC 20231, ON:

February 3, 2000
DATE OF DEPOSIT

MaryAnn Navarrette
PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

MaryAnn Navarrette
SIGNATURE OF PERSON SIGNING CERTIFICATE

2-3-00
DATE OF SIGNATURE

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

TERMINAL DISCLAIMER

The undersigned registered patent attorney or agent states that:

(1) The undersigned is an attorney or agent of record in Application Serial No.08/927,393, filed on August 27, 1997, for "FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS" (the Present Application).

(2) The undersigned has reviewed the evidentiary assignment documents and, to his/her best knowledge and belief, The Dow Chemical Company, a Delaware corporation, is the sole and common assignee of the following patent properties:

(a) The Present Application:

[☒] The original documents are recorded at reel 8459, frame 0434.

[☐] The original documents have been submitted to Assignment Branch for recording. A copy of the assignment document(s) is submitted with this Disclaimer.

RECEIVED
FEB 16 2000
TC 1700 MAIL ROOM

(b) U.S. Patent No. 5,677,383:

[☒] The original documents are recorded at reel 8459, frame 0434.

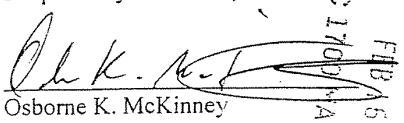
[☐] The original documents have been submitted to Assignment Branch for recording. A copy of the assignment document(s) is submitted with this Disclaimer.

(3) On behalf of the assignee, the undersigned disclaims the terminal part of any patent granted on the Present Application which would extend beyond the expiration date for the full statutory term of U.S. Patent No. 5,677,383 and hereby agrees that any patent so granted on the above-identified application shall be enforceable only for and during such period that the legal title to said patent shall be the same as the legal title to U.S. Patent No. 5,677,383. This agreement runs with any patent granted on the Present Application and is binding upon the grantee, its successors or assigns.

(4) The undersigned does not disclaim any terminal part of any patent granted on the Present Application prior to the expiration date of the full statutory term of U.S. Patent No. 5,677,383 in the event that U.S. Patent No. 5,677,383 later: expires for failure to pay a maintenance fee, is held unenforceable, is found invalid, is statutorily disclaimed in whole or terminally disclaimed under 37 CFR 1.321, has all claims canceled by a reexamination certificate, or otherwise expires or is terminated prior to the expiration of its full statutory term, except for the separation of legal title stated above.

(5) Please charge $110.00 to our Deposit Account No. 04-1512. If this estimate is incorrect, please charge or credit our account accordingly. Two duplicate copies of this sheet are enclosed.

Respectfully submitted,



Osborne K. McKinney
Registration No. 40,084
Phone: (409) 238-7889

RECEIVED
FEB 15 2000
TC 1700 MAIL ROOM

Date: 02-03-00
2301 Brazosport Blvd., B-1211
Freeport, Texas 77541

OKM/man

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office
Address: COMMISSIONER OF PATENTS AND TRADEMARKS
Washington, D.C. 20231

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.
08/927,393	08/27/97	CHUM	P 40121BU

EXAMINER	
HARI AN, R	
ART UNIT	**PAPER NUMBER**
1713	13

IM22/0413

THE DOW CHEMICAL COMPANY
B-1211 PATENT DEPT
FREEPORT TX 77541

DATE MAILED: 04/13/00

Please find below and/or attached an Office communication concerning this application or proceeding.

Commissioner of Patents and Trademarks

PTO-90C (Rev. 2/95)
*U.S. GPO: 1996-404-496/40510

1- File Copy

Notice of Allowability

Application No.	Applicant(s)
08/927,393	Chum, et al.
Examiner	**Group Art Unit**
Robert D. Harlan	1713

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance and Issue Fee Due or other appropriate communication will be mailed in due course.

☒ This communication is responsive to 02/03/2000

☒ The allowed claim(s) is/are 36-51

☐ The drawings filed on _____ are acceptable.

☐ Acknowledgement is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d).

☐ All ☐ Some* ☒ None of the CERTIFIED copies of the priority documents have been

☐ received.

☐ received in Application No. (Series Code/Serial Number) _____ .

☐ received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

*Certified copies not received: _____

☒ Acknowledgement is made of a claim for domestic priority under 35 U.S.C. § 119(e).

A SHORTENED STATUTORY PERIOD FOR RESPONSE to comply with the requirements noted below is set to EXPIRE **THREE MONTHS** FROM THE "DATE MAILED" of this Office action. Failure to timely comply will result in ABANDONMENT of this application. Extensions of time may be obtained under the provisions of 37 CFR 1.136(a).

☐ Note the attached EXAMINER'S AMENDMENT or NOTICE OF INFORMAL APPLICATION, PTO-152, which discloses that the oath or declaration is deficient. A SUBSTITUTE OATH OR DECLARATION IS REQUIRED.

☒ Applicant MUST submit NEW FORMAL DRAWINGS

☐ because the originally filed drawings were declared by applicant to be informal.

☒ including changes required by the Notice of Draftsperson's Patent Drawing Review, PTO-948, attached hereto or to Paper No. 8 .

☐ including changes required by the proposed drawing correction filed on _____ , which has been approved by the examiner.

☐ including changes required by the attached Examiner's Amendment/Comment.

Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the reverse side of the drawings. The drawings should be filed as a separate paper with a transmittal lettter addressed to the Official Draftsperson.

☐ Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Any response to this letter should include, in the upper right hand corner, the APPLICATION NUMBER (SERIES CODE/SERIAL NUMBER). If applicant has received a Notice of Allowance and Issue Fee Due, the ISSUE BATCH NUMBER and DATE of the NOTICE OF ALLOWANCE should also be included.

Attachment(s)

☐ Notice of References Cited, PTO-892

☒ Information Disclosure Statement(s), PTO-1449, Paper No(s). 3 & 6

☐ Notice of Draftsperson's Patent Drawing Review, PTO-948

☐ Notice of Informal Patent Application, PTO-152

☐ Interview Summary, PTO-413

☐ Examiner's Amendment/Comment

☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material

☒ Examiner's Statement of Reasons for Allowance

DETAILED ACTION

1. Two duplicate copies of the Terminal Disclaimer have been entered into the record.

Response to Arguments

2. The rejection of claims 36-51 under 35 U.S.C. 102(b) as anticipated by or, in the alternative, under 35 U.S.C. 103(a) as obvious over WO 90/03414 (hereinafter "WO '414") is withdrawn. However, the reason for the withdrawal is not for the arguments the Applicant outlines in the Response (Paper No. 11).

3. The Applicant argues the claimed invention is not anticipated by WO '414 because the recited slope of strain hardening coefficient is not an inherent property. The Applicant further argues there is ample evidence of record that indicates not all homogeneously branched ethylene polymers have a slope strain hardening coefficient equal to or greater than 1.3, thus the recited requirement of the present invention is clearly not an inherently property. The Applicant has overstated the rejection. The Examiner agrees that not all homogeneously branched ethylene polymers have a slope strain hardening coefficient equal to or greater than 1.3. The examiner contends, as stated in the First Office Action, although WO '414 fails to show the value of the slope of strain hardening coefficient of polymer '013 and a temperature rising elution fractionation (TREF) showing a linear polymer portion of '006, *in view of other substantially similar physical*

properties and processes for making the ethylene polymer, the examiner has a reasonable basis to believe that it inherently possesses said slope of strain hardening coefficient and said linear polymer portion. The Examiner argues if the process for make the blend in the instant invention and WO '414 are substantially similar (i.e. selection of catalysts, cocatalysts, temperature, pressure, etc.), then the properties on one blend (WO '414) is inherent in the properties of the other blend (instant invention), therefore, necessitating a rejection under 35 USC 102/103. See In re Fitzgerald et al., 205 USPQ 594 (CCPA 1980). The Examiner apologizes for any misunderstandings due to the First Office Action.

4. The Applicant further argues the present invention is nonobvious over WO '414 because the present invention provides unexpected results and comparative runs that are necessarily and consistently inferior in regard to impact and tear performance. The Markovich declaration filed on 27 Aug. 1997 has been reconsidered but deemed not to be persuasive since the comparative test do not show unexpected results of the claimed polymer blends. The Markovich declaration reports comparative results of polymer '013 but not the blend as disclosed in WO '414. Second, in the 28/50/72 blends the toughness properties of the comparative runs is not necessarily inferior to that of the inventive runs. The Applicant selectively points out properties that are superior, but does not compare the properties as a whole to show unexpected results. The results of the Markovich declaration are inconclusive at best.

Reasons for Allowance

5. The Examiner effectively withdraws the rejection of claims 36-51 under 35 U.S.C. 102(b) as anticipated by or, in the alternative, under 35 U.S.C. 103(a) as obvious over WO 90/03414 (hereinafter "WO '414") because upon further examination of Application 08/927,393 the blends disclosed in the claimed invention and the blends disclosed in WO '414 have a polyolefin prepared by different processes. The Applicant discloses that the homogeneously branched ethylene/alpha ethylene interpolymer can also be a linear ethylene/ alpha olefin interpolymer as describe in USP 3,645,992 (Elston) and the substantially linear/alpha olefin interpolymers of the present invention are herein defined as in copending application serial number 07/776,130, now U.S. Patent No. 5,272,236. According to Elston, the homogeneously branched ethylene/alpha ethylene interpolymer is prepared by using a Ziegler-Natta catalyst. Blends comprising at least one polymer prepared from a Ziegler-Natta catalyst is known in the prior art. However, blends comprising at least one polymer prepared from a catalyst with constrained geometry about the metal atom as described in U.S. Patent No. 5,272,236 is not disclosed by WO '414. WO '414 discloses bridged metallocenes with a transition metal sandwiched between two cyclopentadienyl rings. It is know in the prior art that these two types of catalysts produce polyolfeins with different properties.

6. As of the date of the Notice of Allowability, the Examiner as not located or identified any references that can be used in combination or individually that would render instant claims 36-51 anticipated or obvious to a person of ordinary skill in the art.

Conclusion

7. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Robert D. Harlan whose telephone number is (703)306-5926. The examiner can normally be reached on weekdays from 9 am to 6 pm.

8. If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, David Wu, can be reached on (703)308-2450. The fax phone number for the organization where this application or proceeding is assigned is (703)306-3429.

R. Harlan
3 April 2000

DAVID W. WU
SUPERVISORY PATENT EXAMINER
TECHNOLOGY CENTER 1700

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office

NOTICE OF ALLOWANCE AND ISSUE FEE DUE

IM22/0413

THE DOW CHEMICAL COMPANY
B-1211 PATENT DEPT
FREEPORT TX 77541

APPLICATION NO.	FILING DATE	TOTAL CLAIMS	EXAMINER AND GROUP ART UNIT	DATE MAILED
08/927,393	09/27/97	016	HARLAN, R 1713	04/13/00

First Named Applicant	35 USC 154(b) term ext. =
CHUM,	0 Days.

TITLE OF INVENTION: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

ATTY'S DOCKET NO.	CLASS-SUBCLASS	BATCH NO.	APPLN. TYPE	SMALL ENTITY	FEE DUE	DATE DUE
40121BU	525-240.000	L15	UTILITY	NO	$1210.00	07/13/00

THE APPLICATION IDENTIFIED ABOVE HAS BEEN EXAMINED AND IS ALLOWED FOR ISSUANCE AS A PATENT. PROSECUTION ON THE MERITS IS CLOSED.

THE ISSUE FEE MUST BE PAID WITHIN THREE MONTHS FROM THE MAILING DATE OF THIS NOTICE OR THIS APPLICATION SHALL BE REGARDED AS ABANDONED. THIS STATUTORY PERIOD CANNOT BE EXTENDED.

HOW TO RESPOND TO THIS NOTICE:

I. Review the SMALL ENTITY status shown above.
If the SMALL ENTITY is shown as YES, verify your current SMALL ENTITY status:

A. If the status is changed, pay twice the amount of the FEE DUE shown above and notify the Patent and Trademark Office of the change in status, or
B. If the status is the same, pay the FEE DUE shown above.

If the SMALL ENTITY is shown as NO:

A. Pay FEE DUE shown above, or

B. File verified statement of Small Entity Status before, or with, payment of 1/2 the FEE DUE shown above.

II. Part B-Issue Fee Transmittal should be completed and returned to the Patent and Trademark Office (PTO) with your ISSUE FEE. Even if the ISSUE FEE has already been paid by charge to deposit account, Part B Issue Fee Transmittal should be completed and returned. If you are charging the ISSUE FEE to your deposit account, section "4b" of Part B-Issue Fee Transmittal should be completed and an extra copy of the form should be submitted.

III. All communications regarding this application must give application number and batch number.
Please direct all communications prior to issuance to Box ISSUE FEE unless advised to the contrary.

IMPORTANT REMINDER: Utility patents issuing on applications filed on or after Dec. 12, 1980 may require payment of maintenance fees. It is patentee's responsibility to ensure timely payment of maintenance fees when due.

PATENT AND TRADEMARK OFFICE COPY

PTOL-85 (REV. 10-96) Approved for use through 06/30/99. (0651-0033)

*U.S. GPO: 1998-437-639/80023

#14 MR B

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant (s): Pak-Wing S. Chum et al.

Serial No.: 08/927,393 Group Art Unit: 1713

Filed: August 27, 1997 Examiner: R. Harlan

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS



I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE. WITH SUFFICIENT POSTAGE. AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO...ASSISTANT COMMISSIONER OF PATENTS AND TRADEMARKS, WASHINGTON, D.C. 20231,

On July 10, 2000
DATE OF DEPOSIT

Margaret Greene
PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

Margaret Greene
SIGNATURE OF PERSON SIGNING CERTIFICATE

July 10, 2000
DATE OF SIGNATURE

Assistant Commissioner for Patents
Washington, D.C. 20231

Attn: Official Draftsman

Sir:

TRANSMITTAL OF FORMAL DRAWING(S)

Responsive to the Office Action dated April 13, 2000, enclosed find the drawing(s) required for the understanding of the subject matter of the invention in compliance with 35 USC §113, in response to request in Paper No. 8, mailed September 30, 1999.

Respectfully submitted,

Osborne K. McKinney
Registration No. 40,084
Phone: 979-238-7889

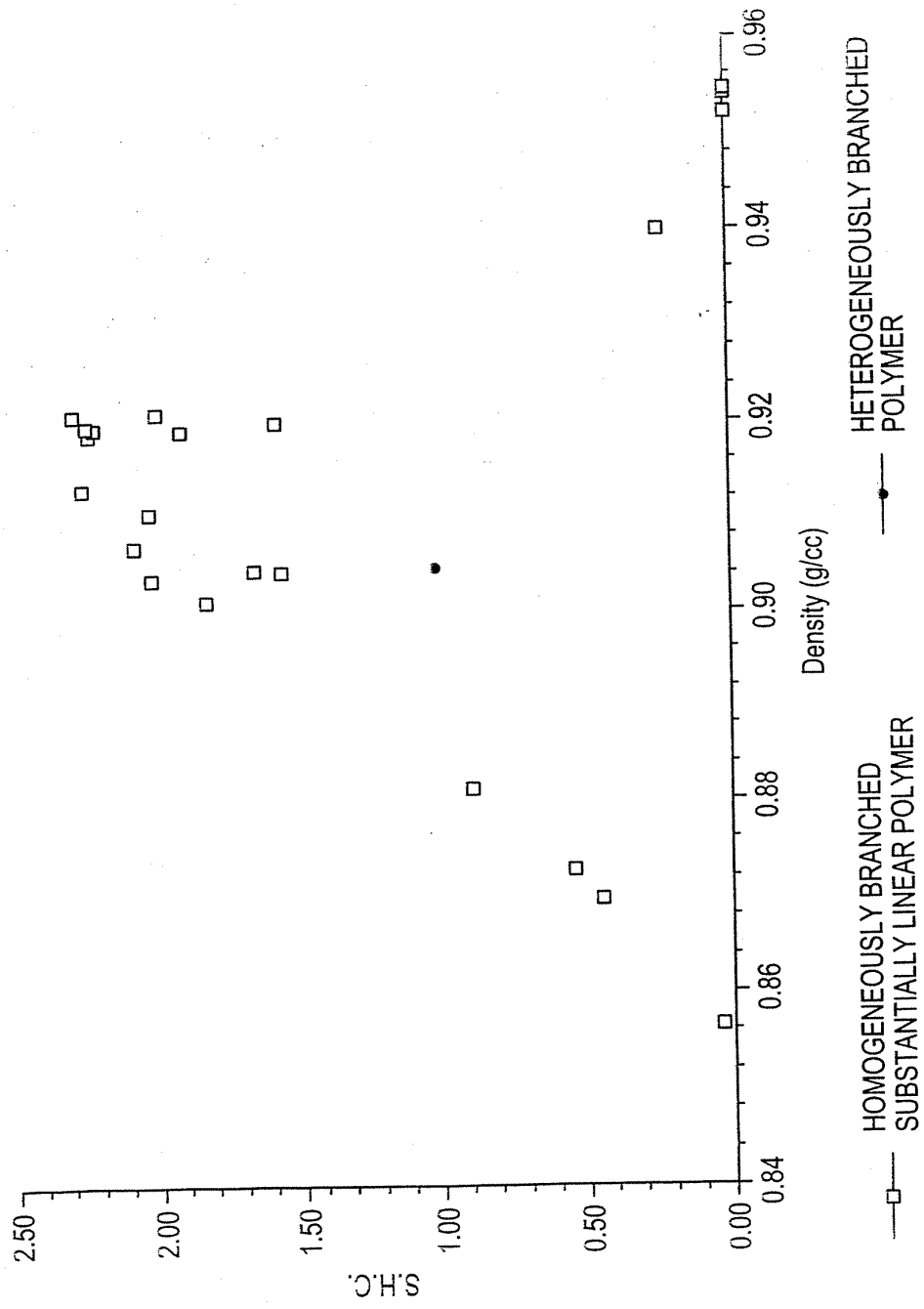
2301 Brazosport Blvd.
Freeport, Texas 77541

OKM/mfg

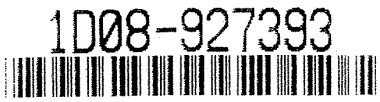
40121BU

04/13/00

FIG. 1


S.H.C.
2.50
2.00
1.50
1.00
0.50
0.00
0.84
0.86
0.88
0.90
0.92
0.94
0.96
Density (g/cc)
HOMOGENEOUSLY BRANCHED
SUBSTANTIALLY LINEAR POLYMER
HETEROGENEOUSLY BRANCHED
POLYMER

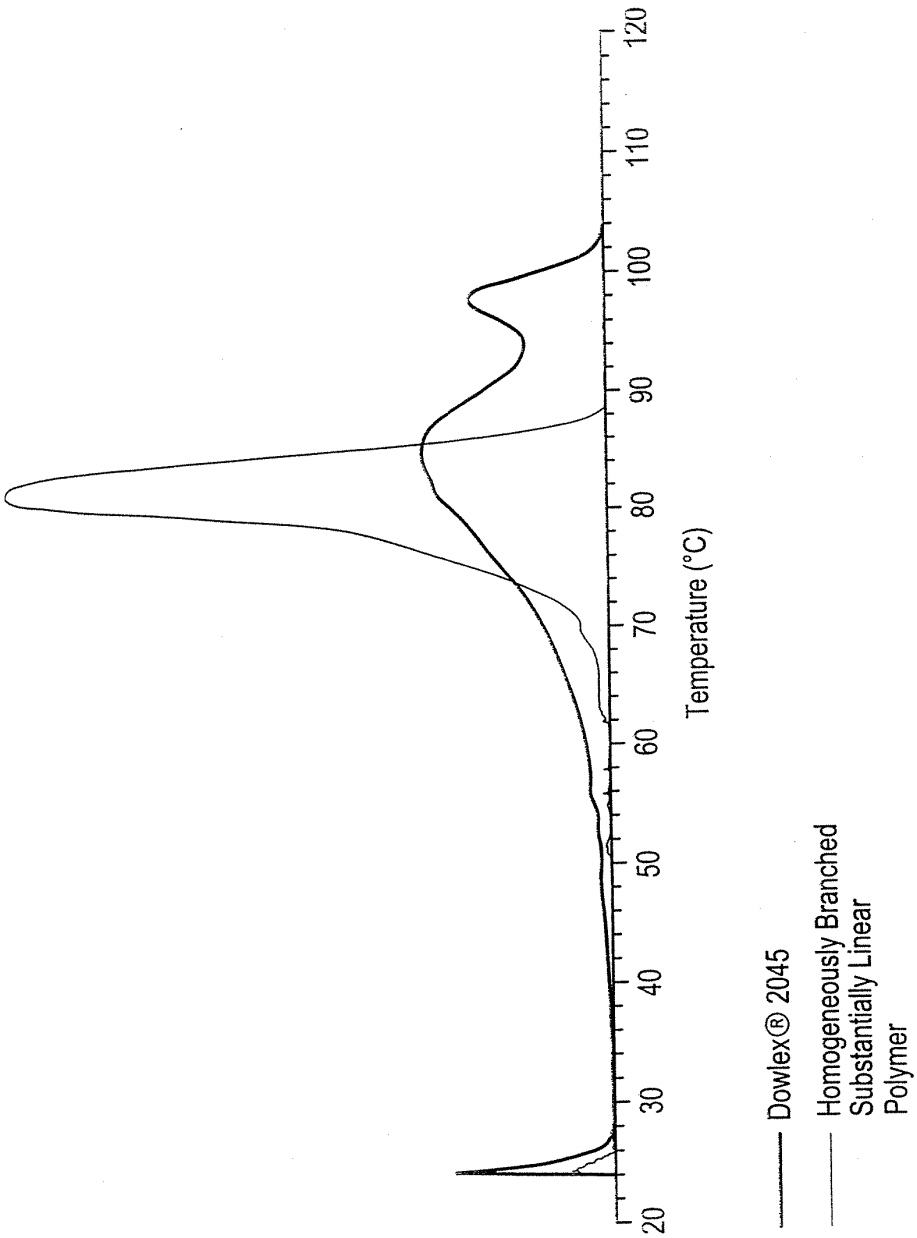
611102



4012180
Serial No.: 08/927,393
Group Art Unit: 1713
Attorney: Osborne K. McKinney
Phone: (979) 238-2889
Sheet 1 of 2



FIG. 2


20
30
40
50
60
70
80
90
100
110
120
Temperature (°C)
Dowlex® 2045
Homogeneously Branched
Substantially Linear
Polymer


OIPE
JUL 13 2000
PATENT & TRADEMARK OFFICE

PART B—ISSUE FEE TRANSMITTAL

Complete and mail this form, together with applicable fees, to: **Box ISSUE FEE**
Assistant Commissioner for Patents
Washington, D.C. 20231

OIPE JUL 18 2000 PATENT & TRADEMARK OFFICE

$-13

MAILING INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE. Blocks 1 through 4 should be completed where appropriate. All further correspondence including the Issue Fee Receipt, the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address, and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

Note: The certificate of mailing below can only be used for domestic mailings of the Issue Fee Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing.

Certificate of Mailing

I hereby certify that this Issue Fee Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Box Issue Fee address above on the date indicated below.

Heather Terry (Depositor's name)
Heather Terry (Signature)
July 13, 2000 (Date)

CURRENT CORRESPONDENCE ADDRESS (Note: Legibly mark-up with any corrections or use Block 1)

IM22/0413

THE DOW CHEMICAL COMPANY
B-1211 PATENT DEPT
FREEPORT TX 77541

APPLICATION NO.	FILING DATE	TOTAL CLAIMS	EXAMINER AND GROUP ART UNIT		DATE MAILED
08/927,393	08/27/97	016	HARLAN, R	1713	04/13/00

First Named Applicant: CHUM, 35 USC 154(b) term ext. = 0 Days.

TITLE OF INVENTION: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

ATTY'S DOCKET NO.	CLASS-SUBCLASS	BATCH NO.	APPLN. TYPE	SMALL ENTITY	FEE DUE	DATE DUE
1 40121BU	525-240.000	L15	UTILITY	NO	$1210.00	07/13/00

1. Change of correspondence address or indication of " Fee Address" (37 CFR 1.363). Use of PTO form(s) and Customer Number are recommended, but not required.

☒ Change of correspondence address (or Change of Correspondence Address form PTO/SB/122) attached. The Dow Chemical Company
P.O. Box 1967
Midland, MI 48641-1967

☐ "Fee Address" indication (or "Fee Address" Indication form PTO/SB/47) attached.

2. For printing on the patent front page, list (1) the names of up to 3 registered patent attorneys or agents OR, alternatively, (2) the name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1 _____
2 _____
3 _____

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)
PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. Inclusion of assignee data is only appropriate when an assignment has been previously submitted to the PTO or is being submitted under separate cover. Completion of this form is NOT a subsitiitue for filing an assignment.

(A) NAME OF ASSIGNEE: The Dow Chemical Company

(B) RESIDENCE: (CITY & STATE OR COUNTRY) Midland, MI USA

Please check the appropriate assignee category indicated below (will not be printed on the patent)

☐ individual ☒ corporation or other private group entity ☐ government

4a. The following fees are enclosed (make check payable to Commissioner of Patents and Trademarks):

☐ Issue Fee
☐ Advance Order - # of Copies _____

4b. The following fees or deficiency in these fees should be charged to:

DEPOSIT ACCOUNT NUMBER 04-1512
(ENCLOSE AN EXTRA COPY OF THIS FORM)

☒ Issue Fee
☐ Advance Order - # of Copies _____

The COMMISSIONER OF PATENTS AND TRADEMARKS IS requested to apply the Issue Fee to the application identified above.

(Authorized Signature) Reg # 40084 (Date) 07-10-00

NOTE: The Issue Fee will not be accepted from anyone other than the applicant; a registered attorney or agent; or the assignee or other party in interest as shown by the records of the Patent and Trademark Office.

Burden Hour Statement: This form is estimated to take 0.2 hours to complete. Time will vary depending on the needs of the individual case. Any comments on the amount of time required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, D.C. 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND FEES AND THIS FORM TO: Box Issue Fee, Assistant Commissioner for Patents, Washington D.C. 20231

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

08927393
07/13/2000 CVORACH1 00000101 041512
01 FC:142 1210.00 CH

TRANSMIT THIS FORM WITH FEE

PTOL-85E (REV.10-96) Approved for use through 06/30/99. OMB 0651-0033 Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE




1F08-927393

Cof-C

#15

IAP76 JUN 16 2006 TRADEMARK OFFICE

*Attorney Docket No.*40121 BU

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Patent No.:6,111,023

Application No.: 08/927,393

Issue Date: August 29, 2000

Inventors: Pak-Wing S. Chum et al.

Certificate JUN 20 2006 of Correction

Request FOR CORRECTION

Certificate of Correction Branch
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

This is a Request for Correction pursuant to MPEP § 1480 to correct a mistake in the above-mentioned U.S. Patent, incurred through the fault of the U.S. Patent Office, which mistake is clearly disclosed in the records of the U.S. Patent Office.

Please note that although the printed copy of the above U.S. Patent includes the claim of priority, however, the PAIR System of the U.S. Patent Office fails to include such claim of priority. In addition, the front page of the above U.S. patent also fails to include such information.

Furthermore, attached please find a copy of the Email instruction that the Patentee has received from the U.S. Patent Office in connection with this error.

Finally, attached please find a copy of the above U.S. Patent.

Accordingly, the patentee respectfully request the correction of the above-mentioned error.

Should there be any questions, please contact me.

Respectfully submitted,

Ray Ashburg
Attorney for Applicant
Reg. No. 53,956
THE DOW CHEMICAL CO.
2030 Dow Center
Midland, Michigan 48674
Phone No:(979) 238-4262

US006111023A

United States Patent [19]

Chum et al.

[11] Patent Number: 6,111,023

[45] Date of Patent: *Aug. 29, 2000

[54] FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

[75] Inventors: **Pak-Wing Steve Chum**; **George W. Knight**, both of Lake Jackson; **Ronald P. Markovich**, Friendswood; **Shih-Yaw Lai**, Sugar Land, all of Tex.

[73] Assignee: **The Dow Chemical Company**, Midland, Mich.

[*] Notice: This patent is subject to a terminal disclaimer.

[21] Appl. No.: **08/927,393**

[22] Filed: **Aug. 27, 1997**

Related U.S. Application Data

[63] Continuation of application No. 08/544,497, Oct. 18, 1995, Pat. No. 5,677,383, which is a continuation of application No. 08/378,998, Jan. 27, 1995, abandoned, which is a continuation of application No. 08/054,379, Apr. 28, 1993, abandoned, which is a continuation-in-part of application No. 07/776,130, Oct. 15, 1991, Pat. No. 5,272,236, which is a continuation-in-part of application No. 07/939,281, Sep. 2, 1992, Pat. No. 5,278,272.

[51] **Int. Cl.**[7] **C08L 23/06**

[52] **U.S. Cl.** **525/240**; 525/242; 525/320

[58] **Field of Search** 525/240, 242, 525/320; 526/160, 161, 352

[56] **References Cited**

U.S. PATENT DOCUMENTS

2,983,704	5/1961	Roedel .	
3,179,720	4/1965	Hillmer .	
3,231,636	1/1966	Synder et al. .	
3,340,328	9/1967	Brindell et al. .	
3,491,073	1/1970	Marinak .	
3,645,992	2/1972	Elston .	
3,914,342	10/1975	Mitchell .	
3,998,914	12/1976	Lillis et al. .	
4,205,021	5/1980	Morita et al. .	
4,230,831	10/1980	Sakurai et al. .	
4,263,422	4/1981	Lowery, Jr. et al. .	
4,320,088	3/1982	Nicco .	
4,330,639	5/1982	Matsuura et al. .	
4,346,834	8/1982	Mazumdar .	
4,374,227	2/1983	Michie, Jr. .	
4,405,774	9/1983	Miwa et al. .	
4,429,079	1/1984	Shibata et al. .	
4,438,238	3/1984	Fukushima et al. .	
4,461,873	7/1984	Bailey et al. .	
4,469,752	9/1984	Yoshimura et al. .	
4,510,303	4/1985	Oda et al. .	
4,530,914	7/1985	Ewen et al. .	
4,542,199	9/1985	Kaminsky et al. .	
4,659,685	4/1987	Coleman, III et al. .	
4,668,752	5/1987	Tominari et al. .	
4,701,432	10/1987	Welborn, Jr. .	
4,752,597	6/1988	Turner .	
4,770,912	9/1988	Furrer et al. .	
4,786,688	11/1988	Thiersault et al. .	
4,789,714	12/1988	Cozewith et al. .	
4,801,652	1/1989	Mizutani et al. .	
4,804,714	2/1989	Olive .	
4,828,906	5/1989	Nishimura et al. .	
4,837,262	6/1989	Jeon et al. .	
4,935,474	6/1990	Ewen et al. .	
4,937,299	6/1990	Ewen et al. .	
4,939,217	7/1990	Stricklen .	
4,981,760	1/1991	Naito et al. .	
4,987,212	1/1991	Morterol et al. .	
5,001,206	3/1991	Bashir et al. .	
5,026,798	6/1991	Canich .	
5,028,663	7/1991	Chung .	
5,032,651	7/1991	McDaniel et al. .	
5,041,501	8/1991	Shirodkar .	
5,047,468	9/1991	Lee et al. .	
5,055,438	10/1991	Canich .	
5,077,255	12/1991	Welborn .	
5,082,902	1/1992	Gurevitch et al. .	
5,084,540	1/1992	Albizzati et al. .	
5,091,228	2/1992	Fujii et al. .	
5,177,147	1/1993	Spenadel et al. .	
5,189,106	2/1993	Morimoto et al. .	
5,206,075	4/1993	Hodgson, Jr. .	
5,210,142	5/1993	Kale et al. .	
5,218,071	6/1993	Tsutsui et al. .	
5,266,392	11/1993	Land et al. .	
5,272,016	12/1993	Ralph .	
5,272,236	12/1993	Lai et al. .	
5,278,272	1/1994	Lai et al. .	
5,350,807	9/1994	Pettijohn et al. .	
5,374,700	12/1994	Tsutsui et al. .	
5,376,439	12/1994	Hodgson et al. .	
5,395,471	3/1995	Obijeski et al. .	
5,395,810	3/1995	Shamshoum et al. .	
5,408,004	4/1995	Lai et al. .	
5,444,145	8/1995	Brant et al. .	
5,464,905	11/1995	Tsutsui et al. .	
5,519,091	5/1996	Tsutsui et al. .	
5,530,065	6/1996	Farley et al. .	
5,656,696	8/1997	Yamamoto et al. .	
5,663,236	9/1997	Takahashi et al. .	
5,677,383	10/1997	Chum et al.	525/240

FOREIGN PATENT DOCUMENTS

2008315	7/1990	Canada .
0 374 695	6/1990	European Pat. Off. .

(List continued on next page.)

OTHER PUBLICATIONS

Modern Methods of Polymer Characterization, pp. 103–112, (1991) "Measurement of Long–Chain Branch Frequency in Synthetic Polymers", by Alfred Rudin.

(List continued on next page.)

Primary Examiner—David W. Wu
Assistant Examiner—R. Harlan

[57] **ABSTRACT**

Fabricated articles made from formulated ethylene polymer compositions are disclosed. Films made from such formulated compositions have surprisingly good impact and tensile properties, and an especially good combination of modulus and toughness. The ethylene polymer compositions have at least one homogeneously branched substantially linear ethylene/α-olefin interpolymer and at least one heterogeneously branched ethylene polymer. The homogeneously branched substantially linear ethylene/α-olefin interpolymer has a density from about 0.89 to about 0.92 g/cm^3 and a slope of strain hardening coefficient greater than or equal to about 1.3.

16 Claims, 2 Drawing Sheets

FOREIGN PATENT DOCUMENTS

0 416 815	3/1991	European Pat. Off. .
0 436 328	7/1991	European Pat. Off. .
0 447 035	9/1991	European Pat. Off. .
0 503 791	9/1992	European Pat. Off. .
0 572 034	12/1993	European Pat. Off. .
0 598 626	5/1994	European Pat. Off. .
0 662 989	7/1995	European Pat. Off. .
0 735 090	2/1996	European Pat. Off. .
53-75278	7/1978	Japan .
131516	6/1984	Japan .
62-121709	6/1987	Japan .
63-328197	10/1990	Japan .
53-31229	12/1993	Japan .
60-16880	1/1994	Japan .
1233599	5/1971	United Kingdom .
WO 87/03610	6/1987	WIPO .
WO 90/03414	9/1989	WIPO .
WO 90/03414	4/1990	WIPO .
WO 93/00400	1/1993	WIPO .
WO 93/03093	2/1993	WIPO .
WO 93/08221	4/1993	WIPO .
WO 93/13143	7/1993	WIPO .
WO 94/00500	1/1994	WIPO .
WO 94/03538	2/1994	WIPO .
WO 94/06857	3/1994	WIPO .
WO 94/12568	6/1994	WIPO .
WO 94/17112	8/1994	WIPO .
WO 95/13321	5/1995	WIPO .

OTHER PUBLICATIONS

The Journal of Chemical Physics, vol. 17, No. 12, Dec. (1949), pp. 1301–1314, "The Dimensions of Chain Molecules Containing Branches and Rings", by Bruno H. Zimm and Walter H. Stockmayer.

Antec 93—Be In That Number, New Orleans, May 9–13, (1993), vol. II, "Dow Constrained Geometry Catalyst Technology (CGCT): New Rules For Ethylene α–Olefin Interpolymers–Controlled Rheology Polyolefins", pp. 1188–1192, by S. Lai and G.W. Knight.

Journal of Rheology, (1986), pp. 340–341, 344–345, 348–349, 352–353, 356–357, "Wall Slip in Viscous Fluids and Influence of Materials of Construction", by A.V. Ramamurthy.

Rheometers for Molten Plastics, (1982), pp. 97–99, by John Dealy.

Polymer Engineering and Science, vol. 17, No. 11, Nov. (1977), pp. 769–774, "Correlation of Low Density Polyethylene Rheological Measurements with Optical and Processing Properties", by M. Shida, R.N. Shroff, and L.V. Cancio.

"A Review of High Resolution Liquid ^{13}Carbon Nuclear Magnetic Resonance Characterizations of Ethylene–Based Polymers", *JMS–Rev. Macromol. Chem. Phys.*, C29(2&3), pp. 201–317, (1989) by James C. Randall.

ACS Symposium Series, No. 142, pp. 94–118, (1980) "Characterization of Long–Chain Branching in Polyethylenes Using High–Field Carbon–13 NMR", by J.C. Randall.

SPE Regional Technical Conference, Quaker Square Hilton, Akron, Ohio, Oct. 1–2, (1985), pp. 107–119, "The Role of Comonomer Type and Distribution in LLDPE Product Performance", by L.D. Cady.

Journal of Polymer Science: Polymer Physics Edition, vol. 20, pp. 441–455 (1982), "Determination of Branching Distributions in Polyethylene and Ethylene Copolymers", by L. Wild, T.R. Ryle, D.C. Knobeloch, and I.R. Peat.

Antec 93, pp. 58–62, "Flexomer Polyolefins, A Unique Class of Ethylene Copolymers for Low Temperature Film Applications", by D.C. Eagar, G.E. Ealer, S.A. Bartocci and D.M. Kung.

Worldwide Metallocene MetCon '94, May 25–27, (1994), "Improved Processing and Performance Balance of Polyethylene Resins Using Metallocene Catalyst Technology", by Mark A. Wendorf.

Specialty Plastics Conference 1990–The Raw Materials Scenario for PE and PP Film Applications and Markets, Dec. 3–4, "High Value Added Film Using an Olefin Based Elastomer", by M. Tanaka.

Packaging Technology and Engineering, Apr. 1994, pp. 34–37, "Single–Site Catalysts Produce Tailor–Made, Consistent Resins", by David F. Simon.

Polyolefin Modification with EXACT™ Plastomers, SPE, Feb. 21–24, 1993, pp. 539–564, by T.C. Yu and G.J. Wagner.

Research Disclosure 37652, Aug. 1995.

Research Disclosure 36210, Jun. 1994.

Research Disclosure 37644, Aug. 1995.

Plastics World, LLDPE Blends Perk Up Performance of PE Films, pp. 40–43, Dec. 1982.

MetCon '93, May 26–28, 1993, Houston, Tx, "The Dfistinguished Features of Metallocene–Based Polyolefins", pp. 235–243, 240, by Akira Todo.

EXACT™ Polymers—For Targeted Performance in Film Packaging, product brochure published by Exxon Chemical Company Oct. 1992.

Journal of Polymer Science: Part B: Polymer Physics, "An Analytical Technique for Measuring Relative Tie–Molecule Concentration in Polyethylene", vol. 29, pp. 1047–1055 (1991) by A. Lustiger and N. Ishikawa.

Journal of Materials Science, "Molecular Topology in Ethylene Copolymers Studied by Means of Mechanical Testing", vol. 23, pp. 415–421 (1988) by R. Seguela and F. Rietsch.

Polymer, "Tensile Drawing Behaviour in a Linaer Low–Density Polyethylene: Changes in Physical and Mechanical Properties", vol. 27, pp. 532–536 (1986) by R. Seguela and F. Rietsch.

Makromol. Chem., Macromol. Symp., "Slow Crack Growth in Polyethylene—A Review", vol. 41, pp. 55–67 (1991) by Norman Brown, Xici Lu, Yan–Ling Huang, and Ruzheng Qian.

Journal of Polymer Science: Part B: Polymer Physics, "Fatigue Crack Propagation in High–Density Polyethylene", vol. 29, pp. 371–388 (1991) by J.T. Yen and J. Runt.

Macromolecules, "Sequence and Branching Distribution of Ethylene/1–Butene Copolymers Prepared with a Soluble Vanadium–Based Ziegler–Natta Catalyst", vol. 25, pp. 2820–2827 (1992) by N. Kuroda, Y. Nishikitani, K. Matsuura and N. Ikegami.

Polymer Communications, "Morphological Location of Ethyl Branches in 13C–Enriched Ethylene/1–Butene Random Copolymers", vol. 29, pp. 258–260 Sep. (1988) by D. C. McFaddin, K.E. Russell and E. C. Kelusky.

Journal of Polymer Science: Polymer Chemistry Edition, "The Preparation and Characterization of Homogeneous Copolymers of Ethylene and 1–Alkenes", vol. 22, pp. 1383–1392 (1984) by B.K. Hunter, K.E. Russell, M.V. Scammell and S.L. Thompson.

Polymer Engineering and Science, "Characterizing Polyethylene–Based Blends with Temperature Rising Elution Fractionation (TREF) Techniques", vol. 27, No. 20, pp. 1562–1571, Mid–Nov. (1987) by Eric C. Kelusky, Clay T. Elston and Ron E. Murray.

"Exxpol Technology For Targeted Polymer Performance," (brochure from/by Exxon Chemical Company with an unknown date).

SPO '93, "Exact Polymers Unique Properties for Value–Added Applications" by F.J. Steininger.

FIG. 1

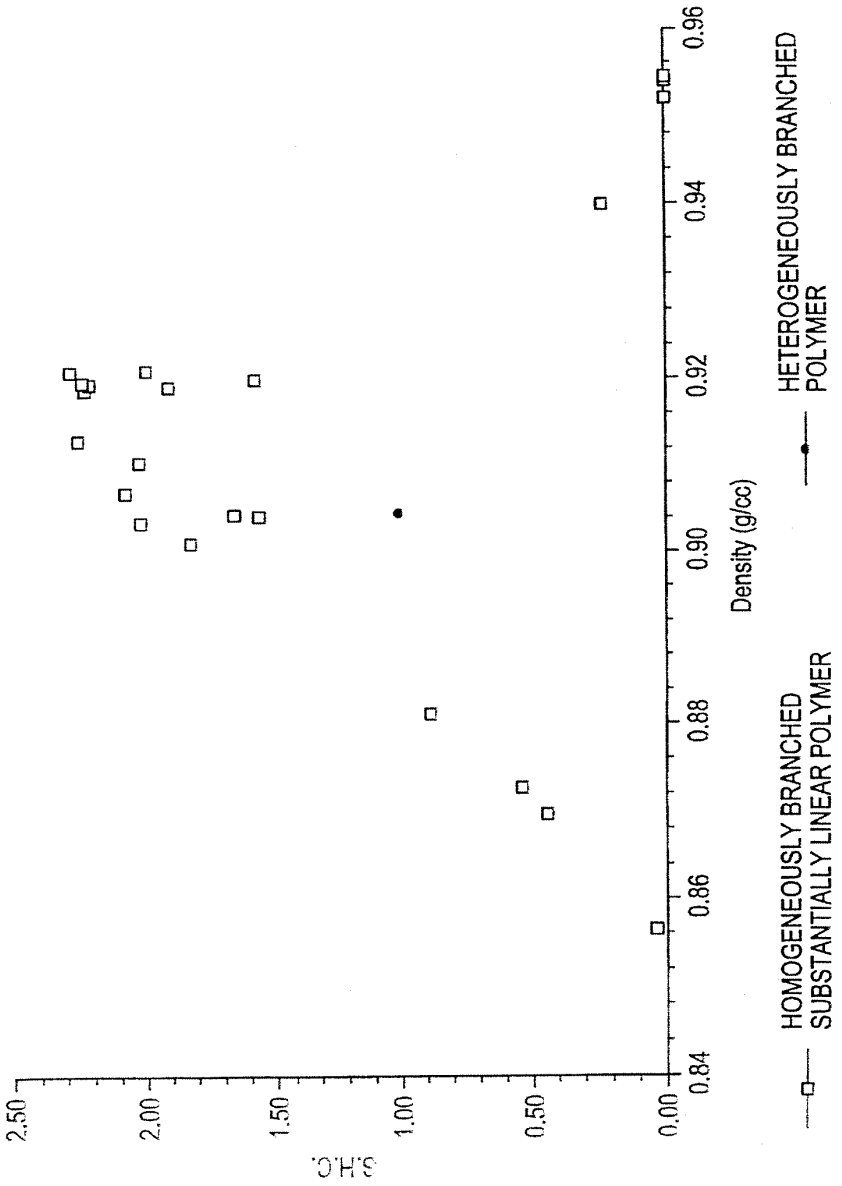
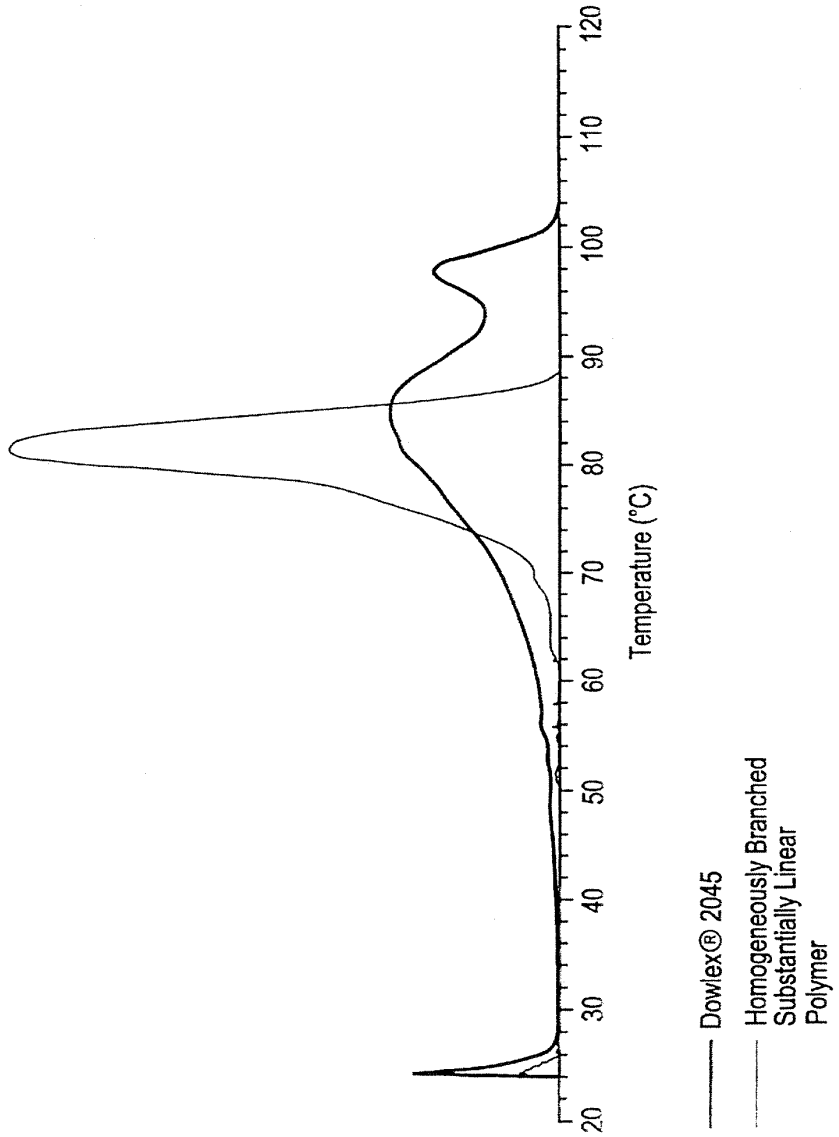

2.50
2.00
1.50
1.00
0.50
0.00
0.84
0.86
0.88
0.90
0.92
0.94
0.96
Density (g/cc)
HETEROGENEOUSLY BRANCHED POLYMER
HOMOGENEOUSLY BRANCHED SUBSTANTIALLY LINEAR POLYMER

FIG. 2


120
110
100
90
80
70
60
50
40
30
20
Temperature (°C)
Dowlex® 2045
Homogeneously Branched
Substantially Linear
Polymer

FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is a Rule 1.60 continuation application of application Ser. No. 08/544,497, now U.S. Pat. No. 5,677,383, filed Oct. 18, 1995 as a Rule 1.60 continuation application of abandoned application Ser. No. 08/378,998, filed Jan. 27, 1995 as a Rule 1.62 continuation application of abandoned application Ser. No. 08/054,379, filed Apr. 28, 1993 as a continuation-in-part application of application Ser. No. 07/776,130, filed Oct. 15, 1991, now U.S. Pat. No. 5,272,236 and as a continuation-in-part application of application Ser. No. 07/939,281, filed Sep. 2, 1992, now U.S. Pat. No. 5,278,272. This application is also related to application Ser. No. 08/834,050, filed Apr. 11, 1997, now issued U.S. Pat. No. 5,847,053. The disclosures of each of the above are incorporated herein, in their entireties, by reference. This application is also related to pending application Ser. No. 08/010,958, filed Jan. 29, 1993 in the names of Brian W. S. Kolthammer and Robert S. Cardwell and entitled Ethylene Interpolymerizations, the disclosure of which is incorporated herein by reference.

FIELD OF THE INVENTION

This invention relates to compositions comprising specific ethylene/α-olefin polymer blends. The polymer blends preferably comprise,:

(A) at least one homogeneously branched substantially linear ethylene/α-olefin interpolymer having specific processing characteristics, blended together with

(B) a heterogeneously branched ethylene polymer.

Such compositions are particularly useful in film applications (e.g., high strength thin gauge packaging film or heat sealable packaging film).

BACKGROUND OF THE INVENTION

Thin film products fabricated from linear low density polyethylene (LLDPE) and/or high density polyethylene (HDPE) are widely used for packaging applications such as merchandise bags, grocery sacks, and industrial liners. For these applications, films with high tensile strength, as well as high impact strength, are desired because film producers can down gauge their film products and still retain packaging performance.

Previous attempts were made to optimize film tensile strength and yield strength by blending various heterogeneous polymers together on theoretical basis. While such blends exhibited a synergistic response to increase the film yield strength, the film impact strength followed the rule of mixing, often resulting in a "destructive synergism" (i.e., the film impact strength was actually lower than film made from one of the two components used to make the blend).

For example, it is known that while improved modulus linear polyethylene resin can be produced by blending high density polyethylene with a very low density polyethylene (VLDPE), the impact strength of the resin blend follows the rule of mixing.

There is a continuing need to develop polymers which can be formed into fabricated articles (e.g., film) having these combinations of properties (e.g., improved modulus, yield strength, impact strength and tear strength). The need is especially great for polymers which can be made into film which can also be down gauged without loss of strength properties, resulting in savings for film manufacturers and consumers, as well as protecting the environment by source reduction.

Surprisingly, we have now discovered that film can have synergistically enhanced physical properties, when the film is made from a blend of at least one homogeneously branched ethylene/α-olefin interpolymer and a heterogeneously branched ethylene/α-olefin interpolymer.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows the relationship between the density and the slope of strain hardening coefficient for homogeneously branched substantially linear ethylene/α-olefin interpolymers used in the compositions disclosed herein, in comparison with a heterogeneously branched ethylene/α-olefin copolymer.

FIG. 2 shows the short chain branching distribution (as measured by analytical temperature rising elution fractionation (ATREF)) for a homogeneously branched substantially linear ethylene/1-octene copolymer used in the invention, in comparison with Dowlex™ 2045 (a heterogeneously branched ethylene/1-octene copolymer made by The Dow Chemical Company).

SUMMARY OF THE INVENTION

Formulated ethylene/α-olefin compositions have now been discovered to have improved physical and mechanical strength and are useful in making fabricated articles. Films made from these novel compositions exhibit surprisingly good impact and tensile properties, and an especially good combination of modulus, yield, ultimate tensile, and toughness (e.g., Dart impact).

The compositions comprise (A) from about 10 percent (by weight of the total composition) to about 95 percent (by weight of the total composition) of at least one homogeneously branched substantially linear ethylene/α-olefin interpolymer having:

(i) a density from about 0.89 grams/cubic centimeter (g/cm^3) to about 0.92 g/cm^3,

(ii) a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8,

(iii) a melt index (I_2) from about 0.001 grams/10 minutes (g/10 min) to about 10 g/10 min,

(iv) no linear polymer fraction, and

(v) a single melting peak as measured using differential scanning calorimetry; and

(B) at least one heterogeneously branched ethylene polymer having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3.

In another aspect, the compositions comprise from about 10 percent (by weight of the total composition) to about 95 percent (by weight of the total composition) of:

(A) at least one homogeneously branched linear ethylene/α-olefin interpolymer having:

(i) a density from about 0.89 grams/cubic centimeter (g/cm^3) to about 0.92 g/cm^3,

(ii) a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8,

(iii) a melt index (I_2) from about 0.001 grams/10 minutes (g/10 min) to about 10 g/10 min,

(iv) no linear polymer fraction, and

(v) a single melting peak as measured using differential scanning calorimetry; and

(B) at least one heterogeneously branched ethylene polymer having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3.

Preferably, both the homogeneously branched substantially linear ethylene/α-olefin interpolymer and the homogeneously branched linear ethylene/α-olefin interpolymer each have a slope of strain hardening coefficient greater than or equal to about 1.3.

DETAILED DESCRIPTION OF THE INVENTION

The homogeneously branched ethylene/α-olefin interpolymer is preferably a homogeneously branched substantially linear ethylene/α-olefin interpolymer as described in pending U.S. Ser. No. 07/776,130. The homogeneously branched ethylene/α-olefin interpolymer can also be a linear ethylene/α-olefin interpolymer as described in U.S. Pat. No. 3,645,992 (Elston), the disclosure of which is incorporated herein by reference.

The substantially linear ethylene/α-olefin interpolymers are not "linear" polymers in the traditional sense of the term, as used to describe linear low density polyethylene (e.g., Ziegler polymerized linear low density polyethylene (LLDPE)), nor are they highly branched polymers, as used to describe low density polyethylene (LDPE).

The substantially linear ethylene/α-olefin interpolymers of the present invention are herein defined as in copending application U.S. Ser. No. 07/776,130 and in copending application entitled "Elastic Substantially Linear Olefin Polymers" filed Sep. 2, 1992 in the names of Shih-Yaw Lai, George W. Knight, John R. Wilson and James C. Stevens.

The homogeneously branched ethylene/α-olefin interpolymers useful for forming the compositions described herein are those in which the comonomer is randomly distributed within a given interpolymer molecule and wherein substantially all of the interpolymer molecules have the same ethylene/comonomer ratio within that interpolymer. The homogeneity of the interpolymers is typically described by the SCBDI (Short Chain Branch Distribution Index) or CDBI (Composition Distribution Branch Index) and is defined as the weight percent of the polymer molecules having a comonomer content within 50 percent of the median total molar comonomer content The CDBI of a polymer is readily calculated from data obtained from techniques known in the art, such as, for example, temperature rising elution fractionation (abbreviated herein as "TREF") as described, for example, in Wild et al, *Journal of Polymer Science, Poly. Phys. Ed.,* Vol. 20, p. 441 (1982), in U.S. Pat. No. 4,798,081 (Hazlitt et al.), or in U.S. Pat. No. 5,089,321 (Chum et al.) the disclosures of all of which are incorporated herein by reference. The SCBDI or CDBI for the linear and for the substantially linear olefin polymers of the present invention is preferably greater than about 30 percent, especially greater than about 50 percent. The homogeneous ethylene/α-olefin polymers used in this invention essentially lack a measurable "high density" fraction as measured by the TREF technique (i.e., the homogeneously branched ethylene/α-olefin polymers do not contain a polymer fraction with a degree of branching less than or equal to 2 methyls/1000 carbons). The homogeneously branched ethylene/α-olefin polymers also do not contain any highly short chain branched fraction (i.e., the homogeneously branched ethylene/α-olefin polymers do not contain a polymer fraction with a degree of branching equal to or more than about 30 methyls/1000 carbons).

The substantially linear ethylene/α-olefin interpolymers for use in the present invention typically are interpolymers of ethylene with at least one C_3–C_{20} α-olefin and/or C_4–C_{18} diolefins. Copolymers of ethylene and 1-octene are especially preferred. The term "interpolymer" is used herein to indicate a copolymer, or a terpolymer, or the like. That is, at least one other comonomer is polymerized with ethylene to make the interpolymer. Ethylene copolymerized with two or more comonomers can also be used to make the homogeneously branched substantially linear interpolymers useful in this invention. Preferred comonomers include the C_3–C_{20} α-olefins, especially propene, isobutylene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene, and 1-decene, more preferably 1-butene, 1-hexene, 4-methyl-1-pentene and 1-octene.

The term "linear ethylene/α-olefin interpolymer" means that the interpolymer does not have long chain branching. That is, the linear ethylene/α-olefin interpolymer has an absence of long chain branching, as for example the linear low density polyethylene polymers or linear high density polyethylene polymers made using uniform (i.e., homogeneous) branching distribution polymerization processes (e.g., as described in U.S. Pat. No. 3,645,992 (Elston)) and are those in which the comonomer is randomly distributed within a given interpolymer molecule and wherein substantially all of the interpolymer molecules have the same ethylene/comonomer ratio within that interpolymer. The term "linear ethylene/α-olefin interpolymer" does not refer to high pressure branched (free-radical polymerized) polyethylene which . is known to those skilled in the art to have numerous long chain branches. The branching distribution of the homogeneously branched linear ethylene/α-olefin interpolymers is the same or substantially the same as that described for the homogeneously branched substantially linear ethylene/α-olefin interpolymers, with the exception that the linear ethylene/α-olefin interpolymers do not have any long chain branching. The homogeneously branched linear ethylene/α-olefin interpolymers comprise ethylene with at least one C_3–C_{20} α-olefin and/or C_4–C_{18} diolefin. Copolymers of ethylene and 1-octene are especially preferred. Preferred comonomers include the C_3–C_{20} α-olefins, especially propene, isobutylene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene, and 1-decene, more preferably 1-butene, 1-hexene, 4-methyl-1-pentene and 1-octene.

Both the homogeneously branched substantially linear and linear ethylene/α-olefin interpolymers have a single melting point, as opposed to traditional heterogeneously branched Ziegler polymerized ethylene/α-olefin copolymers having two or more melting points, as determined using differential scanning calorimetry (DSC).

The density of the homogeneously branched linear or substantially linear ethylene/α-olefin interpolymers (as measured in accordance with ASTM D-792) for use in the present invention is generally from about 0.89 g/cm^3 to about 0.935 g/cm^3, preferably from about 0.905 g/cm^3 to about 0.925 g/cm^3, and especially from about 0.905 g/cm^3 to less than about 0.92 g/cm^3.

The amount of the homogeneously branched linear or substantially linear ethylene/α-olefin polymer incorporated into the composition varies depending upon the heterogeneously branched ethylene polymer to which it is combined. However, about 50 percent (by weight of the total composition) of the homogeneous linear or substantially linear ethylene/α-olefin polymer is especially preferred in the novel compositions disclosed herein.

The molecular weight of the homogeneously branched linear or substantially linear ethylene/α-olefin interpolymers for use in the present invention is conveniently indicated using a melt index measurement according to ASTM

D-1238, Condition 190° C./2.16 kg (formerly known as "Condition (E)" and also known as I_2). Melt index is inversely proportional to the molecular weight of the polymer. Thus, the higher the molecular weight, the lower the melt index, although the relationship is not linear. The lower melt index limit for the homogeneously branched linear or substantially linear ethylene/α-olefin interpolymers useful herein is generally about 0.001 grams/10 minutes (g/10 min). The upper melt index limit for the homogeneously branched linear or substantially linear ethylene/α-olefin interpolymers is about 10 g/10 min, preferably less than about 1 g/10 min, and especially less than about 0.5 g/10 min.

Another measurement useful in characterizing the molecular weight of the homogeneously branched linear or substantially linear ethylene/α-olefin interpolymers is conveniently indicated using a melt index measurement according to ASTM D-1238, Condition 190° C./10 kg (formerly known as "Condition (N)" and also known as I_{10}). The ratio of the I_{10} and I_2 melt index terms is the melt flow ratio and is designated as I_{10}/I_2. Generally, the I_{10}/I_2 ratio for the homogeneously branched linear ethylene/α-olefin interpolymers is about 5.6. For the homogeneously branched substantially linear ethylene/α-olefin interpolymers used in the compositions of the invention, the I_{10}/I_2 ratio indicates the degree of long chain branching, i.e., the higher the I_{10}/I_2 ratio, the more long chain branching in the interpolymer. Generally, the I_{10}/I_2 ratio of the homogeneously branched substantially linear ethylene/α-olefin interpolymers is at least about 6, preferably at least about 7, especially at least about 8 or above. For the homogeneously branched substantially linear ethylene/α-olefin interpolymers, the higher the I_{10}/I_2 ratio, the better the processability.

Other additives such as antioxidants (e.g., hindered phenolics (e.g., Irganox® 1010 made by Ciba Geigy Corp.), phosphites (e.g., Irgafos® 168 also made by Ciba Geigy Corp.)), cling additives (e.g., PIB), antiblock additives, pigments, fillers, and the like can also be included in the formulations, to the extent that they do not interfere with the enhanced formulation properties discovered by Applicants.

MOLECULAR WEIGHT DISTRIBUTION DETERMINATION

The molecular weight distribution of the linear or substantially linear olefin interpolymer product samples is analyzed by gel permeation chromatography (GPC) on a Waters 150C high temperature chromatographic unit equipped with three mixed porosity columns (Polymer Laboratories 10^3, 10^4, 10^5, and 10^6), operating at a system temperature of 140° C. The solvent is 1,2,4-trichlorobenzene, from which 0.3 percent by weight solutions of the samples are prepared for injection. The flow rate is 1.0 milliliter/minute and the injection size is 200 microliters. A differential refractometer is being used as the detector.

The molecular weight determination is deduced by using narrow molecular weight distribution polystyrene standards (from Polymer Laboratories) in conjunction with their elution volumes. The equivalent polyethylene molecular weights are determined by using appropriate Mark-Houwink coefficients for polyethylene and polystyrene (as described by Williams and Word in *Journal of Polymer Science,* Polymer Letters, Vol. 6, (621) 1968, incorporated herein by reference) to derive the following equation:

$$M_{polyethylene}=a^*(M_{polystyrene})^b.$$

In this equation, a=0.4316 and b=1.0. Weight average molecular weight, M_w, is calculated in the usual manner according to the following formula: $M_w=R\ w_i*M_i$, where w_i and M_i are the weight fraction and molecular weight, respectively, of the i^{th} fraction eluting from the GPC column.

For both the homogeneously branched linear and substantially linear ethylene/α-olefin polymers, the molecular weight distribution (M_w/M_n) is preferably from about 1.8 to about 2.8, more preferably from about 1.89 to about 2.2 and especially about 2.

DETERMINATION OF CRITICAL SHEAR STRESS AND CRITICAL SHEAR RATE

Gas extrusion rheometry (GER) is described by M. Shida, R. N. Shroff and L. V. Cancio in *Polymer Engineering Science,* Vol. 17, no. 11, p. 770 (1977), and in "Rheometers for Molten Plastics" by John Dealy, published by Van Nostrand Reinhold Co. (1982) on page 97, both publications of which are incorporated by reference herein in their entirety. All GER experiments are performed at a temperature of 190° C., at nitrogen pressures between 5250 to 500 psig using a 0.0296 inch diameter, 20:1 L/D die. An apparent shear stress vs. apparent shear rate plot is used to identify the melt fracture phenomena. According to Ramamurthy in *Journal of Rheology,* 30(2), 337–357 1986, above a certain critical flow rate, the observed extrudate irregularities may be broadly classified into two main types: surface melt fracture and gross melt fracture.

Surface melt fracture occurs under apparently steady flow conditions and ranges in detail from loss of specular gloss to the more severe form of "sharkskin." In this disclosure, the onset of surface melt fracture is characterized at the beginning of losing extrudate gloss at which the surface roughness of extrudate can only be detected by 40× magnification. The critical shear rate at onset of surface melt fracture for the substantially linear olefin polymers is at least 50 percent greater than the critical shear at the onset of surface melt fracture of a linear olefin polymer having about the same I_2 and M_w/M_n.

Gross melt fracture occurs at unsteady flow conditions and ranges in detail from regular (alternating rough and smooth, helical, etc.) to random distortions. For commercial acceptability, (e.g., in blown film products), surface defects should be minimal, if not absent. The critical shear rate at onset of surface melt fracture (OSMF) and onset of gross melt fracture (OGMF) will be used herein based on the changes of surface roughness and configurations of the extrudates extruded by a GER. Preferably, the critical shear stress at the OGMF and the critical shear stress at the OSMF for the substantially linear ethylene polymers [described] used herein is greater than about 4×10^6 dyne/cm^2 and greater than about 2.8×10^6 dyne/ cm^2, respectively.

DETERMINATION OF THE SLOPE OF STRAIN HARDENING COEFFICIENT

The slope of strain hardening is measured by compression molding a plaque from the polymer to be tested. Typically, the plaque is molded at about 177° C. for 4 minutes under almost no pressure and then pressed for 3 minutes under a pressure of about 200 psi. The plaque is then allowed to cool at about 8° C./minute while still under 200 psi pressure. The molded plaque has a thickness of about 0.005 inches. The plaque is then cut into a dogbone shaped test piece using a steel rule die. The test piece is 0.315 inches wide and 1.063 inches long. The start of the curved portion of the dogbone shape begins at 0.315 inches from each end of the sample and gently curves (i.e., tapers) to a width of 0.09 inches. The curve ends at a point 0.118 inches from the start of the curve

such that the interior portion of the dogbone test piece has a width of 0.09 inches and a length of 0.197 inches.

The tensile properties of the test sample is tested on an Instron Tensile Tester at a crosshead speed of 1 inch/minute. The slope of strain hardening is calculated from the resulting tensile curve by drawing a line parallel to the strain hardening region of the resulting stress/strain curve. The strain hardening region occurs after the sample has pulled its initial load ((i.e., stress) usually with little or no elongation during the initial load) and after the sample has gone through a slight drawing stage (usually with little or no increase in load, but with increasing elongation (i.e., strain)). In the strain hardening region, the load and the elongation of the sample both continue to increase. The load increases in the strain hardening region at a much lower rate than during the initial load region and the elongation also increase, again at a rate lower than that experienced in the drawing region. FIG. 1 shows the various stages of the stress/strain curve used to calculate the slope of strain hardening. The slope of the parallel line in the strain hardening region is then determined.

The slope of strain hardening coefficient (SHC) is calculated according to the following equation:

$$SHC=(\text{slope of strain hardening})*(I_2)^{0.25}$$

where I_2=melt index in grams/10 minutes.

For both the homogeneously branched linear and substantially linear ethylene/α-olefin interpolymers used in the invention, the SHC is greater than about 1.3, preferably greater than about 1.5.

Surprisingly, the slope of strain hardening coefficient reaches a maximum for the linear or the substantially linear ethylene/α-olefin polymers at a density from about 0.89 g/cm^3 to about 0.935 g/cm^3. Heterogeneous ethylene/α-olefin polymers, in contrast, do not behave in the same manner. FIG. 1 graphically depicts the relationship between the density of the homogeneously branched substantially linear ethylene polymers and ethylene/α-olefin polymers as a function of their slope of strain hardening coefficient, and includes a heterogeneously branched ethylene/1-octene copolymer (polymer W** in Table 1) for comparison purposes. Table 1 displays the data of FIG. 1 in tabular form:

TABLE 1

Polymer	Melt Index (I_2) (g/10 min)	Density (g/cm^3)	I_{10}/I_2	SHC*
A	1	0.8564	7.36	0.004
B	1.03	0.8698	7.46	0.45
C	0.57	0.873	7.22	0.54
D	1.01	0.8817	7.36	0.89
E	1.06	0.9018	7.61	1.84
F	2.01	0.9041	8.07	2.03
G	0.77	0.9047	9.01	1.57
H	9.82	0.9048	7.03	1.67
I	4.78	0.9077	7.18	2.08
J	3.13	0.9113	7.67	2.04
K	2.86	0.9139	7.87	2.27
L	1.08	0.9197	8.07	2.24
M	0.96	0.9198	9.61	1.93
N	0.99	0.9203	9.09	2.23
O	1.11	0.9204	10.15	1.59
P	1.06	0.9205	9.08	2.25
Q	1.12	0.9216	8.94	2.3
R	30.74	0.9217	6.27	2
S	31.58	0.94	6.02	0.24
T	0.97	0.9512	12.11	0
U	0.97	0.9533	10.5	0
V	0.92	0.954	7.39	0
W**	0.8	0.905	8.7	1.02

*SHC = Slope of Strain Hardening Coefficient

**A comparative heterogeneously branched ethylene/1-octene copolymer

THE HETEROGENEOUSLY BRANCHED ETHYLENE POLYMER

The ethylene polymer to be combined with the homogeneous ethylene/α-olefin interpolymer is a heterogeneously branched (e.g., Ziegler polymerized) interpolymer of ethylene with at least one C_3–C_{20} α-olefin (e.g., linear low density polyethylene (LLDPE)).

Heterogeneously branched ethylene/α-olefin interpolymers differ from the homogeneously branched ethylene/α-olefin interpolymers primarily in their branching distribution. For example, heterogeneously branched LLDPE polymers have a distribution of branching, including a highly branched portion (similar to a very low density polyethylene), a medium branched portion (similar to a medium branched polyethylene) and an essentially linear portion (similar to linear homopolymer polyethylene). The amount of each of these fractions varies depending upon the whole polymer properties desired. For example, linear homopolymer polyethylene has neither branched nor highly branched fractions, but is linear. A very low density heterogeneous polyethylene having a density from about 0.9 g/cm^3 to about 0.915 g/cm^3 (such as Attane® copolymers, sold by The Dow Chemical Company and Flexomer® sold by Union Carbide Corporation) has a higher percentage of the highly short chain branched fraction, thus lowering the density of the whole polymer.

Heterogeneously branched LLDPE (such as Dowlex® sold by The Dow Chemical Company) has a lower amount of the highly branched fraction, but has a greater amount of the medium branched fraction. FIG. 2 graphically depicts the relative amounts of these various fractions (as measured using temperature rising elution fractionation) for Dowlex® 2045 (a heterogeneously branched ethylene/1-octene copolymer having a melt index (I_2) of about 1 g/10 min, a density of about 0.92 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 7.93 and a molecular weight distribution (M_w/M_n) of about 3.34), as compared with a homogeneously branched substantially linear ethylene/1-octene copolymer having a melt index (I_2) of about 1 g/10 min, a density of about 0.92 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 10.5 and a molecular weight distribution (M_w/M_n) of about 2.18. Note that the homogeneously branched polymer has a single relatively narrow peak at an elution temperature of about 85° C., while the Dowlex® 2045 polymer has a broad branching distribution, as represented by the breadth of elution temperatures over which the polymer fractions elute. Dowlex® 2045 also has a distinct peak at an elution temperature of about 98° C., indicating the "linear" fraction of the whole polymer. Increasing the fraction of the polymer which has the beneficial properties, without concomitantly increasing other fractions has not been demonstrated here-to-fore.

Preferably, however, the heterogeneously branched ethylene polymer is a heterogeneously branched Ziegler polymerized ethylene/α-olefin interpolymer having no more than about 10 percent (by weight of the polymer) of a polymer fraction having a SHC≧1.3.

More preferably, the heterogeneously branched ethylene polymer is a copolymer of ethylene with a C_3–C_{20} α-olefin, wherein the copolymer has:

(i) a density from about 0.93 g/cm^3 to about 0.965 g/cm^3,

(ii) a melt index (I_2) from about 0.1 g/10 min to about 500 g/10 min, and

(iii) no more than about 10 percent (by weight of the polymer) of a polymer fraction having a SHC≧1.3.

The heterogeneously branched ethylene/α-olefin interpolymers and/or copolymers also have at least two melting peaks as determined using Differential Scanning Calorimetry (DSC).

THE FORMULATED COMPOSITIONS

The compositions disclosed herein can be formed by any convenient method, including dry blending the individual components and subsequently melt mixing or by pre-melt mixing in a separate extruder (e.g., a Banbury mixer, a Haake mixer, a Brabender internal mixer, or a twin screw extruder).

Another technique for making the compositions in-situ is disclosed in pending U.S. Ser. No. 08/010,958, entitled Ethylene Interpolymerizations, which was filed Jan. 29, 1993 in the names of Brian W. S. Kolthammer and Robert S. Cardwell, the disclosure of which is incorporated herein in its entirety by reference. U.S. Ser. No. 08/010,958 describes, inter alia, interpolyrmerizations of ethylene and C_3–C_{20} alpha-olefins using a homogeneous catalyst in at least one reactor and a heterogeneous catalyst in at least one other reactor. The reactors can be operated sequentially or in parallel.

The compositions can also be made by fractionating a heterogeneous ethylene/α-olefin polymer into specific polymer fractions with each fraction having a narrow composition (i.e., branching) distribution, selecting the fraction having the specified properties (e.g., SHC≧1.3), and blending the selected fraction in the appropriate amounts with another ethylene polymer. This method is obviously not as economical as the in-situ interpolymerizations of U.S. Ser. No. 08/010,958, but can be used to obtain the compositions of the invention.

FABRICATED ARTICLES MADE FROM THE NOVEL COMPOSITIONS

Many useful fabricated articles benefit from the novel compositions disclosed herein. For example, molding operations can be used to form useful fabricated articles or parts from the compositions disclosed herein, including various injection molding processes (e.g., that described in *Modern Plastics Encyclopedia*/89, Mid October 1988 Issue, Volume 65, Number 11, pp. 264–268, "Introduction to Injection Molding" by H. Randall Parker and on pp. 270–271, "Injection Molding Thermoplastics" by Michael W. Green, the disclosures of which are incorporated herein by reference) and blow molding processes (e.g., that described in *Modern Plastics Encyclopedia*/89, Mid October 1988 Issue, Volume 65, Number 11, pp. 217–218, "Extrusion-Blow Molding" by Christopher Irwin, the disclosure of which is incorporated herein by reference), profile extrusion, calandering, pultrusion (e.g., pipes) and the like. Rotomolded articles can also benefit from the novel compositions described herein. Rotomolding techniques are well known to those skilled in the art and include, for example, those described in *Modern Plastics Encyclopedia*/89, Mid October 1988 Issue, Volume 65, Number 11, pp. 296–301, "Rotational Molding" by R. L. Fair, the disclosure of which is incorporated herein by reference).

Fibers (e.g., staple fibers, melt blown fibers or spunbonded fibers (using, e.g., systems as disclosed in U.S. Pat. Nos. 4,340,563, 4,663,220, 4,668,566, or 4,322,027, all of which are incorporated herein by reference), and gel spun fibers (e.g., the system disclosed in U.S. Pat. No. 4,413,110, incorporated herein by reference)), both woven and nonwoven fabrics (e.g., spunlaced fabrics disclosed in U.S. Pat. No. 3,485,706, incorporated herein by reference) or structures made from such fibers (including, e.g., blends of these fibers with other fibers, e.g., PET or cotton)) can also be made from the novel compositions disclosed herein.

Film and film structures particularly benefit from the novel compositions described herein and can be made using conventional hot blown film fabrication techniques or other biaxial orientation processes such as tenter frames or double bubble processes. Conventional hot blown film processes are described, for example, in *The Encyclopedia of Chemical Technology,* Kirk-Othmer, Third Edition, John Wiley & Sons, New York, 1981, Vol. 16, pp. 416–417 and Vol. 18, pp. 191–192, the disclosures of which are incorporated herein by reference. Biaxial orientation film manufacturing process such as described in a "double bubble" process as in U.S. Pat. No. 3,456,044 (Pahlke), and the processes described in U.S. Pat. No. 4,352,849 (Mueller), U.S. Pat. No. 4,597,920 (Golike), U.S. Pat. No. 4,820,557 (Warren), U.S. Pat. No. 4,837,084 (Warren), U.S. Pat. No. 4,865,902 (Golike et al.), U.S. Pat. No. 4,927,708 (Herran et al.), U.S. Pat. No. 4,952,451 (Mueller), U.S. Pat. No. 4,963,419 (Lustig et al.), and U.S. Pat. No. 5,059,481 (Lustig et al.), the disclosures of each of which are incorporated herein by reference, can also be used to make film structures from the novel compositions described herein. The film structures can also be made as described in a tenter-frame technique, such as that used for oriented polypropylene.

Other multi-layer film manufacturing techniques for food packaging applications are described in *Packaging Foods With Plastics,* by Wilmer A. Jenkins and James P. Harrington (1991), pp. 19–27, and in "Coextrusion Basics" by Thomas I. Butler, *Film Extrusion Manual: Process, Materials, Properties* pp. 31–80 (published by TAPPI Press (1992)) the disclosures of which are incorporated herein by reference.

The films may be monolayer or multilayer films. The film made from the novel compositions can also be coextruded with the other layer(s) or the film can be laminated onto another layer(s) in a secondary operation, such as that described in *Packaging Foods With Plastics,* by Wilmer A. Jenkins and James P. Harrington (1991) or that described in "Coextrusion For Barrier Packaging" by W. J. Schrenk and C. R. Finch, *Society of Plastics Engineers RETEC Proceedings,* June 15–17 (1981), pp. 211–229, the disclosure of which is incorporated herein by reference. If a monolayer film is produced via tubular film (i.e., blown film techniques) or flat die (i.e., cast film) as described by K. R. Osborn and W. A. Jenkins in "Plastic Films, Technology and Packaging Applications" (Technomic Publishing Co., Inc (1992)), the disclosure of which is incorporated herein by reference, then the film must go through an additional post-extrusion step of adhesive or extrusion lamination to other packaging material layers to form a multilayer structure. If the film is a coextrusion of two or more layers (also described by Osborn and Jenkins), the film may still be laminated to additional layers of packaging materials, depending on the other physical requirements of the final film. "Laminations Vs. Coextrusion" by D. Dumbleton (Converting Magazine (September 1992), the disclosure of which is incorporated herein by reference, also discusses lamination versus coextrusion. Monolayer and coextruded

films can also go through other post extrusion techniques, such as a biaxial orientation process.

Extrusion coating is yet another technique for producing multilayer film structures using the novel compositions described herein. The novel compositions comprise at least one layer of the film structure. Similar to cast film, extrusion coating is a flat die technique. A sealant can be extrusion coated onto a substrate either in the form of a monolayer or a coextruded extrudate.

Generally for a multilayer film structure, the novel compositions described herein comprise at least one layer of the total multilayer film structure. Other layers of the multilayer structure include but are not limited to barrier layers, and/or tie layers, and/or structural layers. Various materials can be used for these layers, with some of them being used as more than one layer in the same film structure. Some of these materials include: foil, nylon, ethylene/vinyl alcohol (EVOH) copolymers, polyvinylidene chloride (PVDC), polyethylene terephthalate (PET), oriented polypropylene (OPP), ethylene/vinyl acetate (EVA) copolymers, ethylene/acrylic acid (EAA) copolymers, ethylene/methacrylic add (EMAA) copolymers, LLDPE, HDPE, LDPE, nylon, graft adhesive polymers (e.g., maleic anhydride grafted polyethylene), and paper. Generally, the multilayer film structures comprise from 2 to about 7 layers.

EXAMPLE 1

Seventy five percent (by weight of the total composition) of a homogeneously branched substantially linear ethylene/1-octene copolymer having I_2 of about 1 g/10 min, density of about 0.91 g/cm^3, I_{10}/I_2 of about 10, M_w/M_n of about 2, and SHC of about 1.81 is dry blended and then melt blended with 25 percent (by weight of the total composition) of a heterogeneously branched ethylene/1-octene copolymer having I_2 of about 1 g/10 min, density of about 0.935 g/cm^3, I_{10}/I_2 of about 7.8, and M_w/M_n of about 3.4. The heterogeneously branched ethylene/1-octene copolymer has a fraction of about 5 percent (by weight of the heterogeneously branched copolymer) having a SHC≧1.3. The dry blend is tumble blended in a 50 gallon drum for about 1 hour.

The melt blend is produced in a ZSK 30 twin screw extruder (30 mm screw diameter) and is then fabricated into film. The final blended composition has a density of about 0.919 g/cm^3.

The blended composition is then fabricated into blown film having a thickness of about 1 mil on an Egan Blown Film Line having a 2 inch diameter screw, a 3 inch die and at a 2.5 inch blow up ratio (BUR), as described in Table 2. For all film samples in Examples 1, 2,4, and 6 and for comparative examples 3, 5, and 7, the targeted gauge is about 1 mil, using a blow-up ratio (BUR) of 2.5:1, a LLDPE screw design is used, a die gap of 70 mils is used, and a lay flat of about 11.875 inches is used.

Film properties are measured and reported in Table 3 with other examples of the invention and with comparative examples. Dart impact (type A) of the films is measured in accordance with ASTM D-1709-85; tensile strength, yield, toughness, and 2% secant modulus of the films is measured in accordance with ASTM D-882; Elmendorf tear (type B) is measured in accordance with ASTM D-1922; PPT tear is measured in accordance with ASTM D-2582; Block is measured in accordance with ASTM D-3354.

Puncture is measured by using an Instron tensiometer Tensile Tester with an integrator, a specimen holder that holds the film sample taut across a circular opening, and a rod-like puncturing device with a rounded tip (ball) which is attached to the cross-head of the Instron and impinges perpendicularly onto the film sample. The Instron is set to obtain a crosshead speed of 10 inches/minute and a chart speed (if used) of 10 inches/minute. Load range of 50% of the load cell capacity (100 lb. load for these tests) should be used. The puncturing device is installed to the Instron such that the damping unit is attached to the lower mount and the ball is attached to the upper mount on the crosshead. Six film specimens are used (each 6 inches square). The specimen is damped in the film holder and the film holder is secured to the mounting bracket. The crosshead travel is set and continues until the specimen breaks. Puncture resistance is defined as the energy to puncture divided by the volume of the film under test. Puncture resistance (PR) is calculated as follows:

$$PR=E/((12)(T)(A))$$

where

PR=puncture resistance (ft-lbs/in^3)

E=energy (inch-lbs)=area under the load displacement curve

12=inches/foot

T=film thickness (inches), and

A=area of the film sample in the damp=12.56 in^2.

EXAMPLE 2

Seventy five percent (by weight of the total composition) of a homogeneously branched substantially linear ethylene/1-octene copolymer having I_2 of about 0.5 g/10 min, density of about 0.915 g/cm^3, I_{10}/I_2 of about 11, M_w/M_n of about 2.4, and SHC of about 2.265 is dry blended and then melt blended (as described in Example 1) with 25 percent (by weight of the total composition) of a heterogeneously branched ethylene/1-octene copolymer having I_2 of about 1 g/10 min, density of about 0.935 g/cm^3, I_{10}/I_2 of about 7.8, and M_w/M_n of about 3.4. The heterogeneously branched ethylene/1-octene copolymer has a fraction of about 5 percent (by weight of the heterogeneously branched copolymer) having a SHC≧1.3. The final blended composition has a density of about 0.92 g/cm^3.

Blown film is made as described in Table 2 and film properties are measured and reported in Table 3 with other examples of the invention and with comparative examples.

COMPARATIVE EXAMPLE 3

A heterogeneously branched ethylene/1-octene copolymer having I_2 of about 1 g/10 min, density of about 0.92 g/cm^3, I_{10}/I_2 of about 7.93, and M_w/M_n of about 3.34 is made into film as described in Example 1. The heterogeneously branched ethylene/1-octene copolymer has a fraction of about 36 percent (by weight of the heterogeneous copolymer) having a SHC≧1.3. The entire heterogeneous ethylene/1-octene copolymer has a SHC of about 1.5.

Blown film is made as described in Table 2 and film properties are measured and reported in Table 3 with other examples of the invention and with comparative examples.

EXAMPLE 4

Example 4 is an in-situ blend made according to U.S. Ser. No. 08/010,958, wherein the homogeneously branched substantially linear polymer is made in a first reactor and is an ethylene/1-octene copolymer having a melt index (I_2) of about 0.5 g/10 min., and a density of about 0.9054 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 8.27 and a molecular weight distribution (M_w/M_n) of about 1.979 and comprises about

50% (by weight of the total composition). A heterogeneously branched ethylene/1-octene copolymer is made in a second reactor operated sequentially with the first reactor and has a melt index (I_2) of about 1.5 g/10 min., and a density of about 0.944 g/cm^3 and comprises the remaining 50% (by weight of the total composition). The total composition has a melt index (I_2) of about 1 g/10 min., a density of about 0.9248 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 7.22 and a molecular weight distribution (M_w/M_n) of about 2.641. This composition is made into blown film as described in Table 2 and the resultant film properties are reported in Table 3.

COMPARATIVE EXAMPLE 5

Comparative Example 5 is an ethylene/1-octene copolymer made according to U.S. Ser. No. 07/773,375, filed Oct. 7, 1991, the disclosure of which is incorporated herein by reference. About 15% (by weight of the total composition) is made in a first reactor, with the remaining portion of the composition polymerized in a second sequentially operated reactor. Both reactors utilize Ziegler type catalysts and make heterogeneously branched polymers. The total composition has a melt index (I_2) of about 0.56 g/10 min., a density of about 0.9256 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 9.5 and a molecular weight distribution (M_w/M_n) of about 4.35. This composition is also made into blown film as described in Table 2 and the resultant film properties are reported in Table 3.

EXAMPLE 6

Example 6 is an in-situ blend made according to U.S. Ser. No. 08/010,958, wherein the homogeneously branched substantially linear polymer is made in a first reactor and is an ethylene/1-octene-copolymer having a fractional melt index (I_2), a density of about 0.906 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 8–10 and a molecular weight distribution (M_w/M_n) of about 2.2 and comprises about 43% (by weight of the total composition). A second heterogeneously branched ethylene/1-octene copolymer is made in a second reactor operated sequentially with the first reactor and has a melt index (I_2) of about 0.85 g/10 minutes, a density of about 0.938 g/cm^3 and comprises the remaining 57% (by weight of the total composition). The total composition has a melt index (I_2) of about 0.53 g/10 minutes, a density of about 0.9246 g/cm^3 a melt flow ratio (I_{10}/I_2) of about 7.83, and a molecular weight distribution (M_w/M_n) of about 2.8. This composition is made into blown film as described in Table 2 and the film properties are reported in Table 3.

COMPARATIVE EXAMPLE 7

Comparative Example 7 is an ethylene/1-octene copolymer made according to U.S. Ser. No. 07/773,375, filed Oct. 7, 1991, the disclosure of which is incorporated herein by reference. About 25% (by weight of the total composition) is made in a first reactor, with the remaining portion of the composition polymerized in a second sequentially operated reactor. Both reactors utilize Ziegler type catalysts and make heterogeneously branched polymers. The total composition has a melt index (I_2) of about 0.49 g/10 min., a density of about 0.9244 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 10 and a molecular weight distribution (M_w/M_n) of about 4.78. This composition is also made into blown film as described in Table 2 and the resultant film properties are reported in Table 3.

COMPARATIVE EXAMPLE 8

Comparative example 8 is a heterogeneously branched ethylene/1-octene copolymer having a melt index (I_2) of about 1 g/10 minutes, a density of about 0.9249 g/cm^3, a melt flow ratio (I_{10}/I_2) of about 8 and a molecular weight distribution (M_w/M_n) of about 3.5.

Blown film is made as described in Table 2 and film properties are measured and reported in Table 3 with other examples of the invention and comparative examples.

TABLE 2

	Ex. 1	Ex. 2	Comp. Ex. 3	Ex. 4	Comp. Ex. 5	Ex. 6	Comp. Ex. 7	Comp. Ex. 8
Zone 1A (° F.)	300	300	300	300	300	300	300	300
Zone 1B (° F.)	450	451	475	474	475	475	475	474
Zone 1C (° F.)	450	450	475	475	475	475	475	475
Zone 2A (° F.)	450	450	475	474	475	475	475	475
Zone 2B (° F.)	450	450	455	475	475	475	475	475
Zone 2C (° F.)	450	450	475	475	475	475	475	475
Zone 3 (° F.)	451	452	474	477	477	476	476	474
Zone 4 (° F.)	450	450	473	475	475	475	475	475
Zone 5 (° F.)	450	450	475	475	475	475	475	475
Melt temp. (° F.)	475	477	515	501	502	499	499	497
Blower Air temp. (° F.)	47.3	45.7	57	44.4	86.5	47.6	NA	47.3
Chill Water temp. (° F.)	39	37.6	51.1	38.3	86.8	40	38.7	40.5
Extruder Die press. (psi)	2843	3427	1321	1874	1763	2883	2525	1952
Nozzle press. (in.)	3.2	4.5	4.38	4.4	4.9	4.6	4.6	4.3
Amps	27.3	33.1	37.7	39.9	40.2	50.1	42.6	38.6
Extruder speed (rpm)	27.6	28.8	21.5	23.1	21.1	21.5	22.1	21.7
Nip Roll speed (rpm)	33.1	36.9	39	39.8	36.2	37	36	37.8
Output (lbs/hr)	31	NR*	38.3	39	NR*	36	36	36
Frost line height (in.)	12.5	9	13	12	12	10.5	11	10.5

*NR = Not recorded

TABLE 3

	Ex. 1	Ex. 2	Comp. Ex. 3	Ex. 4	Comp. Ex. 5	Ex. 6	Comp. Ex. 7	Comp. Ex. 8
Yield (MD*) (psi)	1605	1595	1643	2040	2243	1973	1810	1782
Tensile (MD*) (psi)	8522	9525	7444	7794	7931	9325	8455	4928
Toughness (MD*) (ft-lbs/in^3)	1689	1773	1439	1671	1519	NR	NR	NR
Yield (CD**) (psi)	1530	1489	1706	2267	2407	1997	1809	1832
Tensile (CD**) (psi)	6252	7603	5807	7079	7458	7153	6326	4598
Toughness (CD**) (ft-lbs/in^3)	1282	1599	1358	1656	1495	NR	NR	NR
Elmendorf B(MD*) (grams)	288	216	334	317	194	320	398	297
Elmendorf B (CD**) (grams)	621	566	413	630	664	640	621	527
PPT Tear (MD*) (lbs.)	6.79	6.18	5.99	6.2	6.5	6.2	6.2	5.3
PPT Tear (CD**) (lbs.)	7.44	7.42	6.46	6.8	8.1	7.0	7.5	6.1
Dart Impact A (grams)	708	610	354	410	186	412	186	164
Puncture (ft-lbs/in^3)	316	349	251	231	256	250	227	237
Film Block g.	75	33	87	32	17	11.8	17	22
Film Gradient Density (g/cm^3)	0.9145	0.9153	0.9155	0.9205	0.9218	0.9198	0.9201	0.9207
Film Gauge (low) (mils)	0.9	0.9	0.9	0.85	0.8	0.98	0.95	1.05
Film Gauge (high) (mils)	1.2	1.05	1.1	0.95	1	1.08	1.05	1.15

*MD = Machine direction
**CD = Cross direction
NR = Not Recorded

In general, films made from the novel formulated ethylene/α-olefin compositions exhibit good impact and tensile properties, and an especially good combination of tensile, yield and toughness (e.g., toughness and dart impact). Further, films from the example resins exhibited significant improvements over films made from the comparative resins in a number of key properties.

For example, comparing examples 1 and 2 with comparative example 3, the data show films produced from the melt blends (examples 1 and 2) exhibited significantly higher values for the following film properties: dart impact, MD tensile, CD tensile, MD toughness, CD toughness MD ppt tear, CD ppt tear, CD Elmendorf tear B, puncture and significantly lower block.

Comparing example 4 to comparative example 5, the data show films produced from the in-situ blend (made according to U.S. Ser. No. 08/010,958) exhibited significantly higher values for the following film properties: dart impact, MD toughness and CD toughness.

Comparing example 6 to comparative examples 7 (an ethylene/1-octene copolymer made according to U.S. Ser. No. 07/773,375) and 8 (an heterogeneously branched ethylene/1-octene copolymer), the data show films produced from the in-situ blend (made according to U.S. Ser. No. 08/010,958) exhibited significantly higher values for the following film properties: dart impact, md yield, cd yield, md tensile, cd tensile, cd Elmendorf tear b and puncture and significantly lower block.

We claim:

1. An ethylene polymer composition comprising

(A) from about 10 percent (by weight of the total composition) to about 95 percent (by weight of the total composition) of at least one ethylene interpolymer having:

(i) a density from about 0.89 grams/cubic centimeter (g/cm^3) to about 0.935 g/cm^3,

(ii) a melt index (I_2) from about 0.001 grams/10 minutes (g/10 min.) to about 10 g/10 min.,

(iii) a slope of strain hardening coefficient greater than or equal to 1.3, and

(iv) a Composition Distribution Branch Index (CDBI) greater than 50 percent; and

(B) from about 5 percent (by weight of the total composition) to about 90 percent (by weight of the total composition) of at least one ethylene polymer characterized as having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3 and comprising a linear polymer fraction, as determined using a temperature rising elution fractionation (TREF) technique.

2. The composition of claim 1 wherein the ethylene interpolymer (A) has a slope of strain hardening coefficient greater than or equal to 1.5.

3. The composition of claim 1 wherein the ethylene interpolymer (A) is further characterized as having a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8, as determined using gel permeation chromatography.

4. The composition of claim 1 wherein the ethylene interpolymer (A) is further characterized as having no high density polymer fraction, as determined using a temperature rising elution fractionation technique.

5. The composition of claim 1 wherein the ethylene interpolymer (A) is further characterized as having a single melting point, as determined using differential scanning calorimetry.

6. The composition of claim 1 wherein the ethylene interpolymer (A) is further characterized as having a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8, as determined using gel permeation chromatography, and no high density polymer fraction, as determined using a temperature rising elution fractionation technique.

7. The composition of claim 1 wherein the ethylene interpolymer (A) is further characterized as having a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8, as determined using gel permeation chromatography, and a single melting point as determined using differential scanning calorimetry.

JUN 9 A 2008

8. The composition of claim 1 or claim 3 wherein the ethylene interpolymer (A) is further characterized as having a no high density polymer fraction, as determined using a temperature rising elution fractionation technique, and a single melting point, as determined using differential scanning calorimetry.

9. A film made from an ethylene polymer composition, wherein the composition comprises

(A) from about 10 percent (by weight of the total composition) to about 95 percent (by weight of the total composition) of at least one ethylene interpolymer having:

(i) a density from about 0.89 grams/cubic centimeter (g/cm^3) to about 0.935 g/cm^3,

(ii) a melt index (I_2) from about 0.001 grams/10 minutes (g/10 min.) to about 10 g/10 min.,

(iii) a slope of strain hardening coefficient greater than or equal to 1.3, and

(iv) a Composition Distribution Branch Index (CDBI) greater than 50 percent; and

(B) from about 5 percent (by weight of the total composition) to about 90 percent (by weight of the total composition) of at least one ethylene polymer characterized as having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3 and comprising a linear polymer fraction, as determined using a temperature rising elution fractionation (TREF) technique.

10. The film of claim 9 wherein the ethylene interpolymer (A) has a slope of strain hardening coefficient greater than or equal to 1.5.

11. The film of claim 9 wherein the ethylene interpolymer (A) is further characterized as having a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8, as determined using gel permeation chromatography.

12. The film of claim 9 wherein the ethylene interpolymer (A) is further characterized as having no high density polymer fraction, as determined using a temperature rising elution fractionation technique.

13. The film of claim 9 wherein the ethylene interpolymer (A) is further characterized as having a single melting point, as determined using differential scanning calorimetry.

14. The film of claim 9 wherein the ethylene interpolymer (A) is further characterized as having a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8, as determined using gel permeation chromatography, and no high density polymer fraction, as determined using a temperature rising elution fractionation technique.

15. The film of claim 9 wherein the ethylene interpolymer (A) is further characterized as having a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8, as determined using gel permeation chromatography, and a single melting point as determined using differential scanning calorimetry.

16. The film of claim 9 or claim 11 wherein the ethylene interpolymer (A) is further characterized as having a no high density polymer fraction, as determined using a temperature rising elution fractionation technique, and a single melting point, as determined using differential scanning calorimetry.

* * * * *