2014-1431, -1462

IN THE UNITED STATES COURT OF APPEALS
FOR THE FEDERAL CIRCUIT

THE DOW CHEMICAL COMPANY,

Plaintiff-Cross-Appellant,

v.

NOVA CHEMICALS CORPORATION (CANADA)
AND NOVA CHEMICALS INC. (DELAWARE),

Defendants-Appellants.

Appeals from United States District Court for the
District of Delaware in No. 1:05-cv-00737-LPS,
Judge Leonard P. Stark.

BRIEF FOR DEFENDANTS-APPELLANTS NOVA CHEMICALS CORPORATION (CANADA) AND NOVA CHEMICALS INC. (DELAWARE)

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UNITED STATES COURT OF APPEALS FOR THE FEDERAL CIRCUIT

DOW CHEMICAL V NOVA CHEMICALS

Appeal No. 2014-1431

Certificate of Interest

Counsel for NOVA Chemicals Corporation (New Brunswick, Canada) and NOVA Chemicals Inc. (Delaware) certify the following (use "None" if applicable; use extra sheets if necessary):

1. The full name of every party or amicus represented by me is:

   NOVA Chemicals Corporation
   NOVA Chemicals Inc.

2. The name of the real party in interest (if the party named in the caption is not the real party in interest) represented by me is:

   NOVA Chemicals Corporation
   NOVA Chemicals Inc.

3. All parent corporations and any publicly held companies that own 10 percent or more of the stock of the party or amicus curiae represented by me are:

   The following entities own, directly or indirectly, more than 10% of the stock of NOVA Chemicals Inc.: International Petroleum Investment Company (IPIC), a sovereign wealth fund of the Emirate of Abu Dhabi; IPIC Holdings GmbH; NOVA Chemicals Holding GmbH; NOVA Chemicals Corporation; NOVA Petrochemicals Ltd.; NOVA Chemicals (Canada) Ltd. - NOVA Chimie (Canada) Ltd.; NC Holdings USA Inc. The following entities own, directly or indirectly, more than 10% of the stock of NOVA Chemicals Corporation: IPIC; IPIC Holdings GmbH; NOVA Chemicals Holding GmbH

4. The names of all law firms and the partners or associates that appeared for the party or amicus now represented by me in the trial court, the earlier appeal (No. 2010-1526), or are expected to appear in this court are:

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TABLE OF CONTENTS

Page

CERTIFICATE OF INTEREST i

TABLE OF CONTENTS iii

STATEMENT OF RELATED CASES xi

STATEMENT OF JURISDICTION 1

I. STATEMENT OF THE ISSUES 2

II. STATEMENT OF THE CASE 3

A. Preliminary Statement 3

B. Nature of the Case, Course of Proceedings, and Disposition Below 5

C. Statement of Facts 7

1. The Patent Claims at Issue and the District Court's Claim Construction 7

2. Facts Relating to Indefiniteness under *Nautilus* 9

a. The Required Location on the Stress-Strain Curve for Measuring "Slope of Strain Hardening" Is Undisclosed and Indeterminate 10

i. The Missing Figure and Disclosure 12

ii. Dow Employed Multiple Conventions for Measuring Slope of Strain Hardening that Were Different from Dow's Expert's Approach 13

iii. Dr. Hsiao's "Maximum" Slope Location Was Arbitrarily Selected for this Litigation 15

iv. Dr. Hsiao's Testimony Regarding the Location of the Relevant Slope Was Unsubstantiated 16

b. The Units Required for Measuring the "Slope of Strain Hardening" Are Undisclosed and Indeterminate 17

i. The Claims Fail to Specify the Units Employed....17

ii. The Specifications Fail to Specify the Units Employed....18

iii. Dow's Contemporaneous Testing Documents Fail To Establish Standard Measurement Units 20

iv. "Instron" Tensile Testers Fail to Provide a Single Set of Default Units....20

v. SHC Values Determined for the Accused Products Vary, Making Infringement Turn on the Units Employed....21

c. Dow Relied on Expert Testimony Unsupported by the Practice in the Art 21

d. The District Court's Decision Denying NOVA JMOL on Indefiniteness 22

e. The Federal Circuit's Decision on Indefiniteness....22

3. Facts Establishing That Dow Failed To Prove Entitlement To Lost Profits....24

a. Procedural History and Background 24

b. Capacity 25

i. Dow's Ability To Produce ELITE Products 25

ii. Incumbency 28

iii. Dow's Actual Practices and Practical Market Constraints Existing During the Supplemental Damages Period 30

c. Dow's Profit Margin on ELITE Used Lower Estimated Costs Instead of Its Higher Actual Costs....33

III. SUMMARY OF THE ARGUMENT....35

IV. ARGUMENT....36

A. Standards of Review....36

B. The Intervening *Nautilus* Decision Is Controlling in this Appeal, Requiring that the April 13, 2014 Supplemental Damages Judgment Be Vacated....37

1. Notwithstanding This Court's Prior Definiteness Opinion, the Definiteness Issue Must Be Reconsidered under the Intervening Law Exception to the Law of the Case....37

2. *Nautilus* Changed the Standard for Definiteness....40

3. Judge Farnan's Jury Instruction and the Dow Panel Majority's Decision Conflict with *Nautilus*....43

4. Dow's Claims Are Indefinite as a Matter of Law Under the New *Nautilus* Test....44

C. The District Court Clearly Erred in Finding that Dow Had Satisfied the "Capacity" Prong of *Panduit*....54

1. Dow's Hypothetical "Potential" Capacity Theory Is Legally Erroneous....54

2. Mr. Hoffman's "Waterfall" Theory Is Speculative, Unrealistic, and Contrary To Actual Market Conditions and Dow's Responses to those Conditions During the Supplement Damages Period....57

a. Dow Lacked Excess ELITE Production Capacity 58

b. Dow Made Less ELITE and More HDPE 58

c. Dow Could Not Acquire More Octene in 2010 59

d. Dow Deferred Production of ELITE Products 59

e. Dow Had Limitations on How Much HDPE It Could Free Up In Certain Months 60

3. Dow's Incumbency Argument Is Unsupported by Law 60

D. The District Court Committed Legal Error and Thereby Abused Its Discretion in Adopting Dow's Proposed Profit Margin When Mr. Hoffman's Reliance on Estimated Ethylene Costs Admittedly Overstated Dow's Margin on ELITE 62

V. CONCLUSION 65

TABLE OF AUTHORITIES

Page(s)

Federal Cases

Amado v. Microsoft Corp.,
517 F.3d 1353 (Fed. Cir. 2008) 37-38

Andrew Corp. v. Gabriel Electronics, Inc.,
785 F. Supp. 1041 (D. Me. 1992) 63

Biosig Instruments, Inc. v. Nautilus, Inc.,
715 F.3d 891 (Fed. Cir. 2013), *rev'd*, 134 S. Ct. 2120 (2014) 41

Carella v. Starlight Archery & Pro Line Co.,
804 F.2d 135 (Fed. Cir. 1986) 62

Datamize, LLC v. Plumtree Software, Inc.,
417 F.3d 1342 (Fed. Cir. 2005) 41

Dow Chem. Co. v. Nova Chems. Corp. (Canada),
458 Fed. App'x 910 (Fed. Cir.),
cert. denied, 133 S. Ct. 544 (2012) ..... xi, 6, 19, 22-23, 36, 43-46, 48-52

Every Penny Counts, Inc. v. Wells Fargo Bank, N.A.,
No. 11cv2816, 2014 WL 869092 (M.D. Fla. Mar. 5, 2014) 42-43

Exxon Research & Eng'g Co. v. United States,
265 F.3d 1371 (Fed. Cir. 2001) 22-23, 41, 43-44, 49

Fresenius USA, Inc. v. Baxter Int'l, Inc.,
721 F.3d 1330 (Fed. Cir. 2013) 38-39

Gargoyles, Inc. v. United States,
113 F.3d 1572 (Fed. Cir. 1997) 54

Gindes v. United States,
740 F.2d 947 (Fed. Cir. 1984) 39

Grain Processing v. Am. Maize-Products Co.,
185 F.3d 1341 (Fed. Cir. 1999) 61

Harper v. Virginia Department of Taxation,
509 U.S. 86 (1993).....37

Hearing Components, Inc. v. Share Inc.,
600 F.3d 1357 (Fed. Cir. 2010) 41

Hebert v. Lisle Corp.,
99 F.3d 1109 (Fed. Cir. 1996) 62

Home Sav. of Am. FSB v. United States,
399 F.3d 1341 (Fed. Cir. 2005) 37

Honeywell Int'l, Inc. v. ITC,
341 F.3d 1332 (Fed. Cir. 2003) 46

Insituform Techs., Inc. v. Cat Contr., Inc.
10 Fed. App'x 871 (Fed. Cir. 2001) 39

Joy Techs., Inc. v. Flakt, Inc.,
954 F. Supp. 796 (D. Del. 1996).....64

Marconi Wireless Tel. Co. v. United States,
320 U.S. (1943).....38

Mendenhall v. Barber-Greene Co.,
26 F.3d 1573 (Fed. Cir. 1994) 39

Nautilus, Inc. v. Biosig Instruments, Inc.,
134 S. Ct. 2120 (2014)..... 2-3, 9, 35, 37-44, 49, 53-54, 65

Nova Chems. Corp. v. Dow Chem. Co.,
133 S. Ct. 544 (2012).....23

Nova Chems. Corp. (Canada) v. Dow Chem. Co.,
Civil Action No. 13-1601-LPS (D. Del.).....xi

Oiness v. Walgreen Co.,
88 F.3d 1025 (Fed. Cir. 1996) 64

Panduit Corp. v. Stahlin Bros. Fibre Works, Inc.,
575 F.2d 1152 (6th Cir. 1978) 54-55

Polaroid Corp. v. Eastman Kodak Co.,
No. 76-1634-MA, 1990 WL 324105 (D. Mass. Oct. 12, 1990),
amended on recons., 1991 WL 4087 (D. Mass. Jan. 11, 1991) 55-56

Seattle Box Co. v. Industrial Crating and Packing, Inc.,
756 F.2d 1574 (Fed. Cir. 1985) 37

Simmons Co. v. Grier Bros. Co.,
258 U.S. 82 (1922)..... 38-39

Smith International, Inc. v. Hughes Tool Co.
No. CV 72-1231-HLH, 1986 WL 4795 (C.D. Cal. Mar. 12, 1986),
vacated on other grounds, 839 F.2d 663 (Fed. Cir. 1988) 56

SmithKline Diagnostics, Inc. v. Helena Labs. Corp.,
926 F.2d 1161 (Fed. Cir. 1991) 36-37

Sulzer Textil A. G. v. Picanol N.V.,
358 F.3d 1356 (Fed. Cir. 2004) 38

United Carbon Co. v. Binney & Smith Co.,
317 U.S. 228 (1942).....42

Wechsler v. Macke Int'l Trade,
486 F.3d 1286 (Fed. Cir. 2007) 54

White v. Murtha,
377 F.2d 428 (5th Cir. 1967) 39

Wordtech Sys., Inc. v. Integrated Networks Solutions, Inc.,
609 F.3d 1308 (Fed. Cir. 2010) 57

Federal Statutes

28 U.S.C. § 1295(a)(1).....1

28 U.S.C. § 1331.....1

28 U.S.C. § 1338(a) 1

28 U.S.C. § 2107.....1

28 U.S.C. § 2201-02 1

35 U.S.C. § 112 ..... 36, 40-41

35 U.S.C. § 284 24

35 U.S.C. 271(f) 38

Rules

Federal Rule of Appellate Procedure Rule 4(a) 1

Federal Rule of Civil Procedure Rule 50 24

Federal Rule of Civil Procedure 59(e) 24

STATEMENT OF RELATED CASES

This Court previously heard another appeal from the same civil action. *Dow Chem. Co. v. Nova Chems. Corp. (Canada)*, 458 Fed. App'x 910 (Fed. Cir.), *cert. denied*, 133 S. Ct. 544 (2012). The panel consisted of Judge Linn, then-Judge Prost, and Judge Reyna. The appeal was docketed as No. 2010-1526, and the opinion issued on January 24, 2012.

Nova Chems. Corp. (Canada) v. Dow Chem. Co., Civil Action No. 13-1601-LPS (D. Del.), is a district court case that could be directly affected by this Court's decision in the pending appeal.

STATEMENT OF JURISDICTION

The lower court had jurisdiction pursuant to 28 U.S.C. §§ 1331, 1338(a), and 2201-02.

This Court has jurisdiction pursuant to 28 U.S.C. § 1295(a)(1). A timely joint notice of appeal was filed on April 23, 2014 in accordance with 28 U.S.C. § 2107 and Federal Rule of Appellate Procedure 4(a).

I. STATEMENT OF THE ISSUES

1. Whether, in light of *Nautilus, Inc. v. Biosig Instruments, Inc.*, 134 S. Ct. 2120 (2014), this Court's prior decision upholding the validity of the patents-in-suit must be reconsidered under the intervening law exception to the law of the case and the supplemental damages judgment vacated as a matter of law because the claims of the patents-in-suit, read in light of the specification and the prosecution history, fail to inform those skilled in the art with reasonable certainty about the scope of the invention and therefore are indefinite given undisputed evidence that

- the patents-in-suit are missing a key figure that would have specifically informed the public (a) where the slope of the strain hardening curve for a claimed parameter should be measured and (b) what units should be used in that measurement;
- whether the accused products fall within the claims depends upon an arbitrary selection of undisclosed, nonstandardized test parameters and measurement units, particularly given the various options available to and used by Dow as of the filing date; and
- Dow relied upon conclusory extrinsic testimony that was not supported by the intrinsic record?

2. Whether the district court's finding that Dow had the hypothetical capacity to meet excess demand in support of a supplemental award of lost profits was clearly erroneous when, *inter alia*, Dow was admittedly operating all of its plants at full capacity, had made no provision for the production of additional

substitute products, and in fact had affirmatively decreased or deferred production of some products covered by its patents during the supplemental damages period?

3. Whether the district court's determination of Dow's profit margin was an abuse of discretion when it was based on Dow's estimated costs in the face of undisputed evidence of Dow's higher actual costs?

II. STATEMENT OF THE CASE

A. Preliminary Statement

The Supreme Court's intervening change in the law of indefiniteness compels this Court to reconsider its prior definiteness decision. The *Nautilus* Court rejected the legal test used by the panel majority to uphold the validity of Dow's patent claims, namely, that such claims were definite so long as some meaning could be ascribed to a claim, even if reasonable people might disagree about that meaning. The *Nautilus* Court, however, replaced that test with one that determines whether the claims, read in light of intrinsic evidence, fail to inform, with reasonable certainty, those skilled in the art about the scope of the invention.

Dow's claims center on a Dow-coined new parameter in the art called "slope of strain hardening coefficient" ("SHC") that Dow defined in its patent specifications as being based on a measured "slope of strain hardening" value. The intrinsic evidence (the patents themselves) states that the particular location used in determining the "slope of strain hardening" is disclosed by Figure 1.

Dow, however, failed to include this key figure in its applications (hereinafter "the missing figure"), and there was no established convention at the time its applications were filed for determining where the slope of strain hardening for SHC should be measured or its units of measurement.

It is undisputed that whether the accused products fall within the claimed SHC range depends upon the particular location on the stress-strain curve selected for determining the "slope of strain hardening" and the units employed in that measurement, critical variables that cannot be objectively determined from the patents themselves or from the prior art. The fact that Dow never used the maximum slope location approach advocated by Dow's expert at trial and in fact its scientists used multiple alternative locations and units itself further demonstrates that the scope of the claims cannot be determined with reasonable certainty.

If the Court invalidates Dow's claims and reverses, it need not consider NOVA's remaining challenges to the supplemental damages award. If it does address them, the Court should vacate the lost profits portion of the supplemental damages judgment because the district court erred in finding that Dow possessed the necessary capacity to manufacture additional ELITE products to replace NOVA's products sold in the relevant 2010-2011 time period. Dow's plants were admittedly operating at full capacity during the supplemental damages period.

Dow's expert testified in 2013 to a theoretical hindsight "plan" to shift capacity among its plants to make more ELITE products. No such steps were taken by Dow in 2009, 2010, or 2011. Dow in fact reduced or deferred some production of its ELITE products during the relevant time period in favor of increasing the production of products that do not incorporate the patented technology. The district court's adoption of Dow's hypothetical construct rather than the evidence of what actually occurred in the market was legal error, and its finding of capacity was clear error.

Moreover, the district court's adoption of Dow's profit margin was an abuse of discretion because it was based on Dow's projected cost estimates rather than Dow's *actual costs*, which undisputedly ranged from 4 to 10 cents per pound higher. The use of Dow's estimated costs instead of Dow's actual costs overstates Dow's profit margin on ELITE products by approximately 11 million dollars.

B. Nature of the Case, Course of Proceedings, and Disposition Below

Plaintiff-Appellee The Dow Chemical Company (herein referred to as "Dow") is the original assignee of U.S. Patent Nos. 5,847,053 ("the '053 patent) (A300-312), and U.S. Patent No. 6,111,023 ("the '023 patent) (A313-327). Dow filed suit on October 21, 2005 alleging that Defendants-Appellants NOVA Chemicals Corporation and NOVA Chemicals Inc. (collectively "NOVA")

infringed the patents-in-suit by importing and selling NOVA's SURPASS polyethylene ("PE") products.[1] (A205 at #1.)

A jury trial was conducted in 2010. (*See* A261 at #573 - A264 at #585.) The jury found the patents-in-suit infringed and not proven to be invalid, and assessed damages in the amount of $61,770,994.60. (A446-49.) A bench trial on standing was held the following day. (*See* A264 at #585.) On June 18, 2010, Judge Farnan entered a judgment in Dow's favor. (A442.)

On July 30, 2010, Judge Farnan denied NOVA's motion to dismiss for lack of standing. (A432-41; A430.) He simultaneously issued orders and opinions denying, *inter alia*, NOVA's renewed motion for JMOL on the issue of invalidity. (A409-10; A411-23.) Judge Farnan retired the next day, and the case was assigned to Judge Stark.

In Nova's prior appeal, a panel majority affirmed on the issues of standing, indefiniteness, and infringement. *Dow Chem. Co.*, 458 Fed. App'x at 911. Judge Reyna dissented on standing and did not reach the merits of the other two issues. *Id*. at 922.

[1] Dow markets its version of the patented product under the name "ELITE." ELITE and SURPASS are film-grade linear low density polyethylene ("LLDPE") products, and are used in a multitude of film applications such as food packaging, stretch film, and heavy duty shipping sacks. (A9 at ¶¶1-3.) Both parties also sell lower grade, high density polyethylene ("HDPE") products that fall outside the scope of the patents.

On remand, the district court, *inter alia*, granted NOVA's motion for summary judgment of no willful infringement. (A99.) The district court held a bench trial on supplemental damages. (A280 at #743-44.)

On March 28, 2014, the district court granted supplemental damages in the form of lost profits and reasonable royalties, but denied Dow's request for enhanced damages. (A3; A4-29.) On April 14, 2014, the district court entered a final judgment awarding Dow (1) $30,450,896 in supplemental damages up through and including April 4, 2014; (2) (3) an additional $2,711 per day in prejudgment interest from April 5, 2014 to April 14, 2014; and (3) post-judgment interest as provided by law and its costs of the action. (A1-2.) The royalty rate found by the district court for the supplemental damages period was 2.11%, a 20% increase over the rate implicitly awarded by the jury due to NOVA's new status as an adjudicated infringer. (A25.)

C. Statement of Facts

1. The Patent Claims at Issue and the District Court's Claim Construction

The claims at issue are claims 6, 7, 10, and 12 of the '053 patent (A300-12) and claims 1, 2, 5, and 8 of the '023 patent (A313-27).[2] Claim 6 of the '053 patent, the only asserted independent claim of that patent, recites, in relevant part

"[a]n ethylene polymer composition comprising … (A) at least one

[2] The asserted dependent claims do not raise any unique issues in this appeal.

homogeneously branched linear ethylene/α-olefin interpolymer having … (vi) **a slope of strain hardening coefficient greater than or equal to 1.3** ...." (A312 at col. 16:4-25 (emphasis added).)

Claim 1, the only independent claim of the '023 patent, covers a somewhat different ethylene polymer. (A325 at col. 15:59 - col. 16:39.) The claims of the '023 patent likewise recite compositions comprising polyethylenes with a component A having a "slope of strain hardening coefficient ["SHC"] greater than or equal to 1.3."

After a *Markman* hearing, the district court construed the phrase "slope of strain hardening coefficient greater than or equal to 1.3" to mean "the slope of strain hardening multiplied by the melt index raised to the 0.25 power must be greater than or equal to 1.3." (A476.) The court did not specify where the "slope of strain hardening" was to be measured, or what units were involved in that determination. (A492-99.)

The court found "the patent's failure to explicitly set forth the units for the slope of strain hardening coefficient and include a figure exemplifying the manner for computing the slope of strain hardening" to be "troubling." (A498.) It noted that "NOVA has elicited testimony from numerous Dow engineers who, after reviewing the patents-in-suit, expressed an inability to calculate the slope of strain hardening," including that of a named inventor who "was even unable to explain

how to carry out this calculation after reviewing the relevant portions of the specification." *Id.*; *see also* A3273-74; A3419-20; A3609. The district court subsequently denied NOVA's motion for summary judgment of indefiniteness, but reiterated that "many of the troubling questions regarding indefiniteness … remain, and require resolution by the jury on a full factual record." (A404-05, A408.)

Over Nova's objections (A12638-40; A3783; A3827-28; A4082-83), Judge Farnan provided jury instructions that, *inter alia*, permitted the jury to find the claims to be definite "***even if the conclusion may be one over which reasonable persons will disagree***." (A12702 (emphasis added).) Using these instructions, the jury found Dow's claims not indefinite.

2. Facts Relating to Indefiniteness under *Nautilus*[3]

NOVA's indefiniteness defense stems from the missing figure in the patent specifications bearing on the claimed SHC parameter and the failure to designate any units for that parameter, neither of which would have been known with reasonable certainty by one of ordinary skill in the art at the time. Dr. Lai, a named inventor, admitted that SHC was a "key parameter" and one of "two key things that led to [his] patent." (A1443-44.) Dow also relied upon SHC to

[3] *See also* NOVA's briefs in the prior appeal. Reference is generally made herein to the '053 specification, although parallel citations to the virtually-identical '023 specification exists.

distinguish prior art during prosecution and at trial. (*See, e.g.*, A8394-95; A4020-21.)

The SHC parameter was a new construct of Dow—not previously known, defined, or used in the art. (A1436-38; A2543; A3347; A3493.) According to the patents-in-suit, determining SHC requires at least three interdependent steps:

> (1) The tensile properties of a test sample are tested by stretching the test sample on an Instron Tensile Tester at a constant rate to generate a "stress/strain" tensile curve. (A307, col. 6:24-28.)
>
> (2) "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." (A307 at col. 6:26-28 (emphasis added).)
>
> (3) SHC is calculated by multiplying the material's "slope of strain hardening" by the material's "melt index" ("I_2") raised to the 0.25 power. (A307, col. 6:45-50; *see also* A476; A492.)

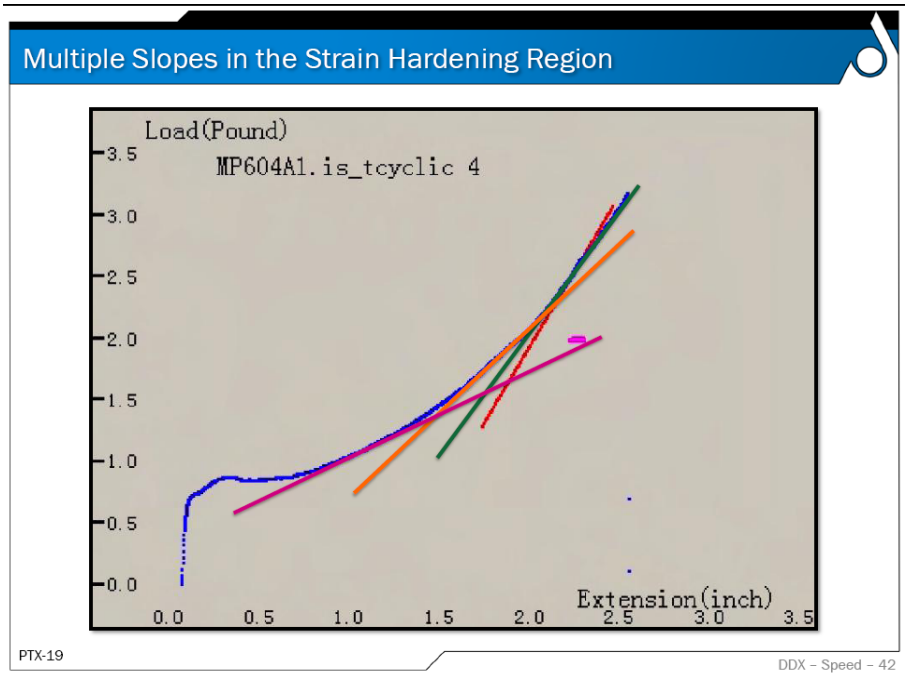
a. The Required Location on the Stress-Strain Curve for Measuring "Slope of Strain Hardening" Is Undisclosed and Indeterminate

The patents provide a basic description of the "strain hardening region" of the stress/strain curve obtained for the test sample:

> ***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 done 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.*** .... ***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.

(A307, col. 6:29-43 (emphasis added).) The patents further state that "[t]he 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." (A307, col. 6:26-29.) Dow filed its applications without the missing figure showing the location on the stress-strain curve "used to calculate the slope of strain hardening" and failed to provide any other disclosure supplying this critical information.

Because the strain hardening region is generally curved (A2584), there is no unique location for a "parallel line" and no single slope value within the strain hardening region. (A2583-84.) Indeed, numerous possible slopes were identified at trial, shown below by several straight lines drawn parallel to various sections of the curved strain hardening region:



(A3349-51; A3523-25; A6830.)

The particular location within the strain hardening region used to measure the slope is critical to determining an SHC value, as the resultant SHC value may be above or below the 1.3 value claimed depending on where the slope is measured. (A1468; A3522-23.) Indeed, the dependency of SHC on slope location is well illustrated in Dow's internal data.[4]

i. The Missing Figure and Disclosure

As noted above, the patent specifications rely upon Figure 1 to "show[] the various stages of the stress/strain curve used to calculate the slope of strain hardening." (A307, col. 6:40-44.) **This figure is missing.** (A1406; A2559.) An inventor, Dr. Lai, could offer no explanation for how the specified figure came to be missing, saying that he had "no idea" and "no idea what's going on." (A1407.)

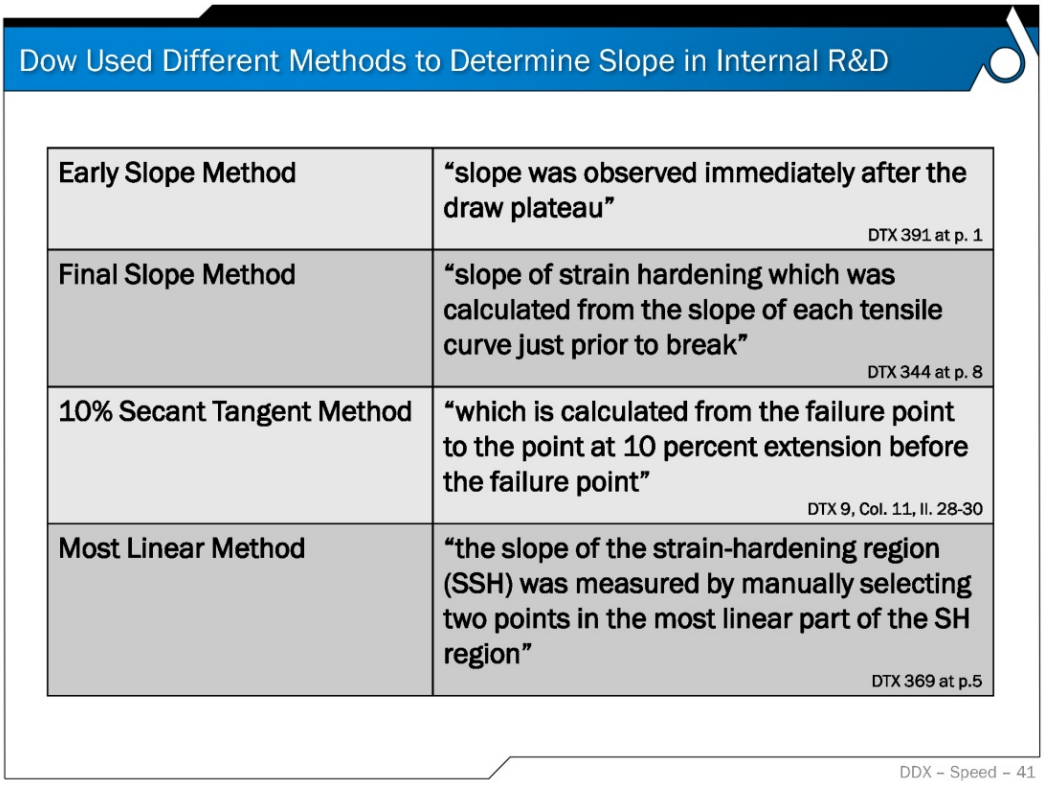
Although the patents-in-suit do contain two other figures, one of which is also denominated "Figure 1" and referenced elsewhere in the specification (A303-04; A307, col. 6:60-67), neither specifies a particular location on a stress-strain curve for determining the slope of strain hardening. (A1406, A1408-09; A2559.) Nor do the patents-in-suit otherwise specify a location, such as "maximum slope,"

[4] For example, one internal Dow R&D document identifies two distinct slope measurement locations: "S.S.H. (psi) = the slope of strain hardening, i.e., the slope of the tensile curve just prior to break" and "S.S.H._b = slope of strain hardening (higher value) immediately after the draw plateau." (A6831, col. B, rows 6, 14.) For the same underlying data, the values for "S.S.H" (reported in column H) and "S.S.H._b" (reported in column L) differ by as much as seven times. (A6832, row 44 (409.129 vs. 2388.19), row 45 (730.46 vs. 1588.09), row 46 (207.216 vs. 1579.36), row 47 (489.799 vs. 2303.58).)

within the "strain hardening region" where the "parallel line" for the slope is to be located. (A1403; A1420; A2550.)

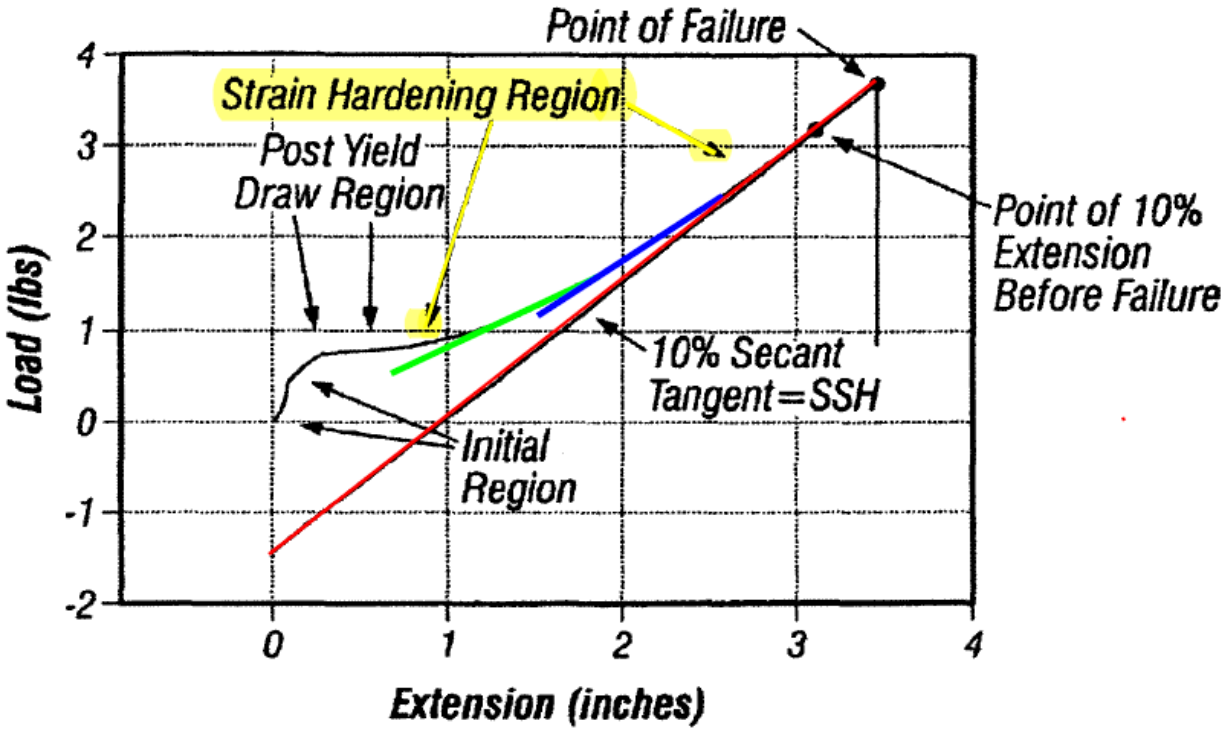
ii. Dow Employed Multiple Conventions for Measuring Slope of Strain Hardening that Were Different from Dow's Expert's Approach

Dow's internal files and later disclosures contain multiple definitions for where the slope of strain hardening for SHC should be determined. For example, as summarized in the following trial demonstrative (A6829), Dow utilized at least four different conventions to determine slope:



(See also A3348-49; A6627, col. 11:8-11; A6662; A6704; A6769; A6831, cells 6-B, 10-D, 14-B.) None of these various possible approaches was disclosed in the patents-in-suit.

The type of stress-strain curve that should have been included in the patents but was inexplicably absent can be illustrated by one such curve (defined as a load extension curve) depicted in a subsequently-issued but related Dow patent[5] not in suit:



(A6618 (Fig. 2) (color added); A6626 at col. 10:64 - A6627 at col. 11:4.) The two arrows extending from the label "Strain Hardening Region" identify the extended, curved region occurring after a plateau ("post yield draw region"). Unlike the patents-in-suit, this later figure shows a specific location ("10% secant tangent = [Slope of Strain Hardening]") and unambiguous units (pound per inch) for

[5] Later-filed, co-pending U.S. Patent No. 6,723,398 shares inventors in common with the patents-in-suit and included a claim of priority thereon. (A6622 at col. 1:5-9.)

determining Dow's SHC parameter. No such figure was included in the patents-in-suit.[6]

iii. Dr. Hsiao's "Maximum" Slope Location Was Arbitrarily Selected for this Litigation

Dow did not rely at trial upon intrinsic evidence—or even prior art—to specify the relevant slope location. Instead, Dow's expert, Dr. Hsiao, used his own mathematically-defined "maximum slope" in the strain hardening region based on a computer algorithm he developed for this litigation. (A2387-88; A2436-37; A2583; A2648; A3521-23). Dr. Hsiao admitted that the patents-in-suit make no reference to using "maximum slope" to determine slope of strain hardening. (A2550.) Significantly, while the patents dictate that the slope line must be "parallel" to the strain hardening region (A307 at col. 6:27-29), lines generated by Dr. Hsiao's "maximum" slope location approach inconsistently cross the stress/strain curve. (*See, e.g.*, A5689 (lower curve - showing Dr. Hsiao's red slope line crossing the blue data curve); A2582-83; A2646-47.)

Dr. Hsiao's "maximum" slope location approach will yield values higher than four other slope locations used by Dow scientists at that time, except for limited situations such as where the "strain hardening region" is a linear (straight

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6 The need for designating a particular section for determining the slope of strain hardening (SSH) is readily apparent, since one can clearly discern different slopes for the added green and blue colored lines (*not found in the original Figure 2*) as compared to the 10% secant tangent line used by Dow (designated here in red) as the parallel line in the '398 patent. (*See* A3349-51; A3523-25; A6830.)

line) curve. (A1595-98; A2564; A2566-70; A2579-82; A6745; A6763 (Fig. 14).) In other words, when locations other than Dr. Hsiao's "maximum" slope are used or a larger number of data points than his algorithm considered, the result can be lower SHC values. (A2569-70; A2580-84; A3521-23.)

In fact, if one applied Dr. Hsiao's "maximum" slope location approach to the data in a declaration about the prior art filed by Dow during prosecution of the patents-in-suit, it would have yielded a prior art SHC value greater than 1.3 and therefore would not have provided the point of novelty that Dow argued to the PTO. (A3511-17.)

iv. Dr. Hsiao's Testimony Regarding the Location of the Relevant Slope Was Unsubstantiated

Dow's reliance on the "maximum slope" location approach is supported only by reference to Dr. Hsiao's unsubstantiated testimony regarding the alleged knowledge of a person skilled in the art. (A2517; A2565-66.) Indeed, Dr. Hsiao freely admitted that "the only thing [the jury] can rely on is [his] word that that's what a person of ordinary skill in the art would have done[.]" (A2518.)

Dr. Hsiao admitted that Dow never used the "maximum slope" location approach to determine where the slope of strain hardening for SHC should be measured (A1403-05), and that he was not made privy to Dow's various other internal approaches at the time he did his analysis (A2565). Nor did Dr. Hsiao interview anyone who had actually practiced the invention using the slope of strain

hardening at the time of the invention in 1993 to see how it had been practiced. (A2565.)

Dr. Hsiao also conceded that his opinions were not supported by "a single reference that used the term maximum slope as the way to calculate slope of strain hardening" or "a single document showing that a person skilled in the art actually would have used maximum slope[.]" (A2517.) Nor were his opinions supported by any reference showing where the slope of strain hardening was to be measured in and around 1993, the time of the invention. (A2565.) Moreover, he acknowledged that "[t]here are no documents that [the jury] can rely on to support [his] testimony that a person of ordinary skill would have used the maximum slope[.]" (A2518.)

b. The Units Required for Measuring the "Slope of Strain Hardening" Are Undisclosed and Indeterminate

i. The Claims Fail to Specify the Units Employed

None of the claims of the patents-in-suit recites any specific units for the recited "slope of strain hardening" or the claimed "1.3" value of SHC. (A312; A325-26.) Moreover, Dr. Hsiao admitted at trial that "there are lots and lots of possible units for slope of strain hardening[.]" (A2593.) He also admitted that one has to "decide on units of [slope of strain hardening] in advance to get a meaningful number for" SHC "because the units for the slope term fundamentally

matter[.]" (A2588; *see also* A2519-20.) Consequently, he acknowledged that SHC "would be higher or lower depending on what units you use[.]" (A2589.)

If any inference can be drawn from the claims, it is that the SHC units would be metric and not the pounds per inch English units assumed by Dr. Hsiao. The claims recite density in terms of grams per cubic centimeter (g/cm^3), and melt index in terms of grams per 10 minutes (A312), the two parameters relied on by Dow's expert Dr. Soares to prove infringement. Moreover, the district court's claim construction of the phrase "slope of strain hardening coefficient of greater than or equal to 1.3" is defined in part in terms of "melt index" (A476), which is expressly denominated in metric units. (A307 at col. 6:40; A312 at col. 16:14-15.)

ii. The Specifications Fail to Specify the Units Employed

The patent specifications also do not identify particular units for the slope of strain hardening. To the contrary, as admitted by Dow's expert, "it's just silent, it doesn't give you units for slope of strain hardening[.]" (A2615.) Indeed, the patents-in-suit do not even specify between metric or English units, much less specifying the units of pounds per inch assumed by Dow's witnesses. (A1412; A1447-49; A2584-1585; A2597; A2602-03.) Since SHC was a newly-developed Dow parameter (A1436-38; A2543; A3347), there was no discussion of the units for SHC in the prior art.

The specification further lacks any consistent blaze marks pointing to specific units. While the patents-in-suit refer to a "draw rate" for the tensile test of 1 inch per minute, this does not define SHC or slope units. Dr. Hsiao admitted that both he and Dow mixed English or metric draw rate units with different systems of stress and strain units. (A2602; A2610; *see also* A3291-92.) Although the panel majority understood there to be no evidence that English and metric units could not be combined, 458 Fed. App'x at 920, as explained by Dr. Fuller and uncontroverted by Dow, it would have been both uncommon and highly irregular to mix English units for one term with metric units for another term in a single equation. (A3499-500; A3576-77.) Likewise, only metric units are used in the sole table and figure containing SHC data. (A308 (Table 1); A303 (Fig. 1).)

Moreover, contrary to Dr. Hsiao's assertion that a person of ordinary skill would use pounds per inch slope units based on the disclosure of the patents-in-suit, Dow's later U.S. Patent No. 6,506,866 (A6842-69) expressly recites the identical SHC claimed parameter and reports metric megapascal ("MPa") units – not English lbs/inch– for the slope of strain hardening coefficient. (A2612-14; A6867 at col. 50:Table 4 ("Slope SH Mpa").) All three patents share a virtually identical section entitled "Determination of the Slope of Strain Hardening Coefficient." (*Compare* A307 at col. 6:6-50 *with* A6846 at col. 8:36 - A6847 at col. 9:9; *see also* A2612-14.)

iii. Dow's Contemporaneous Testing Documents Fail To Establish Standard Measurement Units

Since SHC is a Dow-coined new parameter, the only non-litigation documentary evidence of slope of strain hardening units are internal Dow documents, which reflect a variety of possible units. (*See, e.g.*, A1477; A1539-42; A3355; A6649 (MPa); A6674 (MPa); A6816 (ksi v. strain); A6650 (psi); A6770 (psi); A6831 (psi); A6745 (lb/in).)

iv. "Instron" Tensile Testers Fail to Provide a Single Set of Default Units

Dow's expert relied on extrinsic evidence of alleged machine default units in a software user manual for an Instron tensile tester, and inferred from this that pounds per inch units were the appropriate measurement units. (*See, e.g.*, A2401-02; A2407; *see also* A1632; A2773.) However, Dr. Hsiao admitted that the default Instron units were not limited to English units, and that they could also be either metric or SI units (an accepted scientific form of the metric system). (A2685-86; *see also* A3489-90.)

Significantly, no particular Instron model or manual is referenced in the patent specifications. (A307 at col. 6:25-26; A2671.) Moreover, the manual (A5788-6614) on which Dow relies for alleged default units is not an instrument manual but an optional software manual. Dow's own expert, Dr. Hsiao, "[did] not believe that a person skilled in the art would look at an optional software package,

where the patent does not require or even mention any device besides the Instron Tensile Tester itself." (A2684.) In fact, Dr. Hsiao admitted that "there is no reason ... to look at the [1990 Instron Software Manual (A5788-6614)] to see what units there may or may not be in there[.]" (A2685.)

v. SHC Values Determined for the Accused Products Vary, Making Infringement Turn on the Units Employed

The Dow employee assisting Dr. Hsiao (Mr. Reicheck) actually measured slope of strain hardening on Dow's replicates of the accused products in ***metric*** units. (A2590; A2592; A2679-80; A3593.) It is undisputed that these measurements yielded non-infringing SHC values (less than 1.3) for all product samples. Thus, where SHC is measured using the actual raw data metric units generated by Dow (kilograms per millimeters (kg/mm) or kilograms per millimeter squared (kg/mm^2)), there was no infringement. (A3518-21; *see also* A2592-93; A3491-506; A6827; A6828.) The patents do not direct against using these units. (A3521.) Dr. Hsiao converted the actually-measured ***metric*** units to lbs/in and only then obtained SHC values ≥ 1.3. (A2592-93.)

c. Dow Relied on Expert Testimony Unsupported by the Practice in the Art

Dr. Hsiao's opinion regarding units—that one "would conclude that you should use pounds for load and inches for extension or elongation" (A2400-01)—is not supported by or based on any documents evidencing how SHC is to be

measured. To the contrary, documents reflecting Dow's internal SHC practice for its newly-coined parameter were not even considered by Dr. Hsiao in forming his opinions. (A2594.) Nor did Dr. Hsiao consider what units persons of ordinary skill actually used at the time of the invention. (A2593-94.)

d. The District Court's Decision Denying NOVA JMOL on Indefiniteness

The district court did not separately discuss the issues of indefiniteness and written description in its opinion or comment upon any of the controlling trial evidence summarized above. Rather, it summarily concluded that "the jury's verdict indicates that the jury found Dr. Hsiao to be more credible than Dr. Fuller" and that there was "ample evidentiary support" in Dr. Hsiao's opinion "to support the jury's verdict." It therefore denied NOVA's motion for JMOL on invalidity. (A417-18.)

e. The Federal Circuit's Decision on Indefiniteness

The panel majority affirmed the district court's ruling that the patents-in-suit were not indefinite. *Dow*, 458 Fed. App'x at 917. In so doing, it stated that, to be indefinite, "a claim term must be such that 'no narrowing construction can properly be adopted' to interpret the claim." *Id*. (*quoting Exxon Research & Eng'g Co. v. United States*, 265 F.3d 1371, 1375 (Fed. Cir. 2001)).

Although the panel majority acknowledged that SHC was a new Dow construct not previously known in the art and that the patents-in-suit omitted "for

whatever reason" a Figure 1 that could be used in determining the slope of the strain hardening, it held that Dow had established that one of ordinary skill in the art would have known that the slope of the hardening curve would have to be measured at its maximum value. 458 Fed. App'x at 918-19. In particular, it held that claims are not indefinite even if some experimentation is required to determine the exact scope of the claims and "the mere fact that the slope may be measured in more than one way does not make the claims of the patent invalid." *Id*. at 920 (citing *Exxon*, 265 F.3d at 1375).

In addition, the panel majority held that one of ordinary skill in the art would realize that the slope of strain hardening should be determined in English units despite the fact that the claimed equation for SHC incorporates the melting index reported in metric units. *Id*. at 920. It again reiterated that the "test for indefiniteness is not whether the scope of the patent claims is easy to determine, but whether 'the meaning of the claims is discernible, even though the task may be formidable and the conclusion may be one over which reasonable persons will disagree ....'" *Id*. (*quoting Exxon*, 265 F.3d at 1375).

NOVA filed a petition for *certiorari* on, *inter alia*, the indefiniteness issue, which was subsequently denied. *Nova Chems. Corp. v. Dow Chem. Co.*, 133 S. Ct. 544 (2012).

3. Facts Establishing That Dow Failed To Prove Entitlement To Lost Profits

a. Procedural History and Background

The jury returned its verdict on June 15, 2010. (A446-49.) The jury determined that certain NOVA polymers infringed at least one claim of either the '053 or '023 patents, and awarded Dow $61,770,994.60 in total damages—$57,447,024.98 in lost profits and $4,323,969.62 in reasonable royalties. (*Id*.) The jury award covered Dow's damages sustained between March 1, 2002 and December 31, 2009. (*Id*.) The jury's award was approximately 90% of the lost profits that Dow had requested. (A22 at n.5.) Although Dow had sought a reasonable royalty rate of 10% (A25; A14065), as noted by the district court, the jury's reasonable royalty award works out to an implicit reasonable royalty rate of 1.755%. (A22 n.5; A25.)

NOVA filed a Rule 50 motion for judgment arguing, in part, that Dow failed to prove at the 2010 trial that it was entitled to lost profits damages. (A259 at #554.) On July 30, 2010, the district court denied NOVA's motion. (A411-23.) NOVA did not separately challenge the damages award in its prior appeal.

On July 2, 2010, Dow moved under 35 U.S.C. § 284 and Federal Rule of Civil Procedure 59(e) to amend the jury's verdict to include an accounting of infringing sales of NOVA's SURPASS products that were not considered by the jury in awarding damages. (A260 at #558-59.) The district court granted Dow's

request for a supplemental accounting. (A424.). The supplemental damages proceeding covered "Defendants' January 1, 2010 to October 15, 2011 sales of SURPASS products adjudicated to infringe."[7] (A13074.)

b. Capacity

i. Dow's Ability To Produce ELITE Products

NOVA's SURPASS sales were slightly under 140 million pounds per year during the supplemental damages period. (A14009.)

ELITE is a co-polymer comprised of ethylene and octene. (A12156-59; A14069.) Historically, Dow has made approximately 500 million pounds of ELITE annually. (A12 at ¶13; A12079; A12235-37; A14066; A13964-69.) Dow's dual reactor ELITE-capable production lines (*a.k.a.* "trains") were operating at full capacity in 2010 and most of 2011, but only about half of Dow's capacity on those trains was used to manufacture ELITE in those years; the remaining capacity was used to manufacture DOWLEX[8] and HDPE. (A12-13 at ¶13; A12235-37; A14067; A13964-69.) The district court found that Dow had the equipment to produce 500 million pounds of additional ELITE per year during the supplemental damages period. (A13 at ¶13; A12237.)

[7] October 15, 2011 was the date that the patents-in-suit expired.

[8] DOWLEX is another Dow LLDPE product. (A13 at n.3.) It is a co-polymer comprised of ethylene and either octene or hexene. (A12020.) Production of DOWLEX does not require an ELITE-capable (or dual-reactor) train. (A13 at n.3.)

Dow engages in an annual production planning session to determine what volumes of high value, high margin products like ELITE and DOWLEX Dow can expect to sell in the following year. (A13 at ¶14.) These planning sessions are completed at a fourth-quarter meeting at which sales managers discuss realized sales and project ELITE's increased sales for the next year. (A13 at ¶14; A12099-100, A12204-05.) Dow focuses on its current year's sales, uses its yearly historical sales figures as a starting point to determine its projected ELITE production for the upcoming year, and then adjusts its projections to account for Dow's expected yearly growth in ELITE sales. (A13 at ¶14; A12099-100; A12204-05; A14005.) Dow's strategy is to load its solution reactors with high value, high margin products like ELITE and DOWLEX, which has a profit margin at least as high as that of ELITE. (*Compare* A12388-89 *with* A14015.) Dow then loads any remaining capacity with lower margin, commodity-grade products like HDPE (as long as it would be profitable to do so). (A13 at ¶15; A12209.)

The district court found that Dow could have manufactured additional ELITE products with its North American assets in 2010 and 2011 by planning differently. (A13 at ¶16; A12216; A12238-39; A12244.) If Dow had been selling more ELITE in 2009, the district court concluded Dow would have planned to produce and sell more ELITE in 2010, and would have contracted for more raw

materials (e.g., octene) to allow it to manufacture the additional volume that Dow planned to sell. (A13-14 at ¶16; A12205-06; A12239; A12241-43.)

Based on the foregoing, the district court accepted that Dow would have planned to produce and sell less of its low margin, commodity-grade HDPE products (A14 at ¶16; A12238-39; A12244), notwithstanding the fact that Dow's damages expert, Mr. Hoffman, admitted that "very different circumstances surround[ed] the Dow plant capacity during the jury phase and the supplemental damages phase." (A12353.) Throughout the 2002-2009 original damages period, Dow had excess production capacity in its ELITE production trains. (A14056.) However, in 2010-2011, Dow's plants were "running flat out" with essentially no excess ELITE capacity during nearly the entire supplemental damages period. (A12349-53; *see also* A13098; A12084; A12247; A12238; A13964-69.) Mr. Hoffman conceded that Dow had zero "excess Elite capacity" at its Fort Saskatchewan plant in 2010-2011 and just 42 million pounds of "excess Elite capacity" at Dow's Plaquemine facility in late 2011. (A12348-50.)

Accordingly, for the supplemental damages period, Dow advanced a new lost profits theory, one which posited that Dow would have forgone some HDPE sales and shifted other HDPE manufacturing to facilities with a one-reactor production train, while the two-reactor facilities that are necessary for ELITE production (the Fort Saskatchewan and Plaquemine plants) would be freed up to

produce additional ELITE products. (A21; A12075-76; A12080-81; A12327-28.) The district court found that Dow could have met additional demand by allocating more of its capacity to its high value ELITE products, and less of its capacity to non-strategic, commodity-grade products like HDPE, consistent with the strategy discussed above. (A14 at ¶17; A12216.) However, this model, which Dow's witness Mr. Hoffman termed his "waterfall" method, was characterized by NOVA's damages expert as a speculative accountant's model that did not provide a reasonable degree of certainty regarding alleged lost profits. (A12467-68.)

ii. Incumbency

The district court found that it was important to be the "incumbent" supplier (*i.e.*, the manufacturer with customers to whom it was already selling) because converters and customers tend to stay with their incumbent supplier to avoid having to qualify an alternative resin. (A10 at ¶4; A12140; A12143-44.) Qualifying a PE resin is a demanding, time-consuming process in which suppliers work with converters to ensure that particular PE products are engineered to work with the converters' manufacturing equipment and to deliver the properties required by the converters and the converters' customers. (A10 at ¶5; A12144-47.) The process for qualifying an alternative resin typically takes as long as 18 months. (A10 at ¶5; A12195.)

The district court determined that, but for NOVA's infringement, Dow would have begun the supplemental damages period in the position of incumbent supplier and therefore Dow would have planned for, and produced, the additional ELITE products. (A16 at ¶23; A22; A12099-100; A12204-05.) The district court also found that Dow had historically been able to procure the necessary volume of octene (a critical material used in the manufacture of ELITE) to meet the growth in demand for its ELITE products (A14 at ¶18; A12218-19) and that, if Dow needed to manufacture additional ELITE in the supplemental damages period, Dow could have and would have procured additional octene (A14 at ¶18; A12221). It therefore concluded that Dow would have maintained (rather than actually reduced) its contract levels for octene in 2009. (A14 at ¶18; A22; A12289-90.)

But notwithstanding the district court's finding that Dow was the incumbent, Dow's own expert witness conceded that, while his analysis assumed that Dow was the incumbent for NOVA's customers as of January 1, 2010, Dow "sure wasn't" the incumbent in fact. (A12378.) On the contrary, Mr. Hoffman admitted that "there were no actual Dow sales they would have been walking away from." *Id*.

iii. Dow's Actual Practices and Practical Market Constraints Existing During the Supplemental Damages Period

Inconsistent with Mr. Hoffman's "waterfall" theory, rather than increasing production of ELITE and proportionally reducing HDPE production, Dow actually elected to make less ELITE and began making more HDPE at times during the supplemental damages period. (A12446-48; A13716; A13745.) Indeed, contemporaneous Dow documents tout the importance of selling HDPE to its "high profit" food and beverage industry customers. (A12171; A12104; A12109; A12111; A12121; A13932; A13939.) Another reason Dow increased, at times, its HDPE production was because its octene supply needed to manufacture ELITE and DOWLEX was "critically low" and it could not reach its target octene levels in 2010. (A12261-64; A12275-76; A13845; A13312; A13314; A13890; A13344; A12445-46; A12115.)

Dow's own witness testified that Dow could not have bought or imported more octene because Dow had long term contracts, and it would have been "very difficult" to obtain more octene in 2010, a year of supply disruption. (A12263-65; A12276-79.) Mr. Obregon, Dow's Global Business Director for Solution and Gas Phase PE, did not know what the people managing the octene levels at Dow were actually doing to respond to the octene shortage. (A12273-275.) Other than

buying more octene, he didn't know what else they could do. (A12275-76.) But Dow, he admitted, could not have bought more octene at that time. (A12276.)

Even if sufficient additional octene was available to Dow, it does not follow that customers would have purchased ELITE. The record shows that customers could have purchased and, in a number of cases, in fact did purchase acceptable alternatives from third parties such as Exxon, Chevron Phillips, and Lyondell Basell rather than buying Dow's ELITE product. (A12516-21; A13168-71.) Moreover, the stop-gap option of converting from octene-based DOWLEX to hexene-based DOWLEX proposed by Dow to free up octene for ELITE would not have been acceptable to all of Dow's customers. (A12267.) For example, Dow's customer Paragon objected strongly to this forced conversion and stopped buying from Dow altogether. (A12386-87; A13140 ¶2.)

When Dow was presented with an actual opportunity to make additional ELITE sales during the supplemental damages period, Dow "postponed production of [a] new grade [of ELITE 5400G requested by a customer, 'AEP,'] until [the] end of July" 2011 due to "[t]ight capacity at [Dow's] Plaquemine" production facility. (A13190; A12439-40.) As Dow's 30(b)(6) witness, Mr. Bunker, admitted, Dow did not forgo making some other polymer to make the ELITE resin requested by AEP (A12439-40), but instead deferred AEP's order until the end of

July 2011 when Dow finally had some unused production capacity. (A12349; A12466-67; A14004.)

Mr. Hoffman's "waterfall" theory also failed to address Dow's actual practice of production planning on a monthly basis. (*See, e.g.*, A12227-28; A14057; A12476.) His scenario would require Dow to free up 10 million pounds of actual HDPE production for each month Dow's plants were running at capacity in order to make 10 million pounds of ELITE. (A12477.) However, in a number of months—September 2010, and February, May, June, and July 2011—Dow did not have enough HDPE production to free up in order to make room for enough ELITE to cover its pro-rata share of NOVA's SURPASS sales. (A12476-78; A14037; A14057.) Because Mr. Hoffman's analysis was based on cumulative or annualized HDPE production, it ignored the monthly limitation in Dow's capacity. (A12586.)

Dow places reliance on the fact that, but for NOVA's infringement, it would have been the incumbent in the 2002-2009 time period, and therefore would have made provisions in 2009 to purchase sufficient octene before the octene shortage developed. Dow, however, took no steps in 2008, 2009, or even prior to or after the jury trial in mid-2010 to plan a shift of HDPE production to non-ELITE plants or to secure more octene to make additional ELITE product so that it would have excess ELITE stockpiled or already in the pipeline to immediately sell to NOVA's

customers in the event that Dow prevailed in the jury trial. Such plans were made only in Mr. Hoffman's hypothetical construct for the supplemental damages trial.

c. Dow's Profit Margin on ELITE Used Lower Estimated Costs Instead of Its Higher Actual Costs

NOVA argued below that Dow used incorrect estimates to calculate Dow's raw material costs and, as a result, that Dow's profit margin was artificially and improperly inflated. The district court did not provide any substantial analysis on this point, and simply chose Dow's method of calculating the profit margin. (*See* A23-24.)

For calculating lost profits, Dow's profit margin per pound needs to be based on the incremental cost of goods to produce additional ELITE polyethylene polymers. The single largest component of these incremental costs was Dow's costs for the ethylene raw material. (A12479.)

Dow's purported profit margin for ELITE, however, were not based on Dow's actual Standard Costs. (A12374-76.) As admitted by Mr. Hoffman, "[t]here are a bunch of things in Dow's Standard Costs from its P&L statements that are not in the [quarterly product cost estimate ('PCE')] numbers [he] used to calculate damages in this case." (A12376.)

The Standard Cost of trade value used in Dow's profit and loss statement reflects Dow's *actual* costs for the ethylene raw materials, while the PCE values used by Mr. Hoffman are Dow's quarterly *projected cost estimates* (i.e.,

guesstimates) that do not reflect actual costs. (A12376-77; A12459-61; A12480-83; A14058-59; A12368-70.) Indeed, Mr. Hoffman admitted that "Dow's Standard Costs on its profits and loss statement include actual ethylene costs," the principal cost component of ELITE, "whereas the [PCEs] that [he] relied on do not[.]" (A12376-77; *see also* A12479.) For the calculation of lost profits, Mr. Hoffman also conceded that Dow's incremental cost of goods sold should be based on Dow's actual ethylene costs: "that would be ideal." (A12371-72.)

By using PCEs instead of Dow's Standard Costs, Mr. Hoffman understated Dow's costs and thereby overstated Dow's profit margin on ELITE. (A12488; A14061-64.) For example, looking at 2010 data, Mr. Hoffman's average PCE unit cost was 35.5 cents per pound ("cpp"), while Dow's actual ELITE Standard Cost was 41 cpp. (A12489-91; A12495-96; A14060; A14016; A13289.)

As explained by the unrebutted analysis of Nova's damages expert, Mr. Tanger, Mr. Hoffman understated Dow's ELITE costs at 4-10 cpp, depending on the production plant and year. (A12558; A14063.) Given the volume of LLDPE material at issue, the consequence of Mr. Hoffman's error is to overstate the ELITE profit component in Dow's lost profits contentions by $11.3 million. (A12479-80; A12491-98; A14061-64.) This is roughly one-third of the total amount of supplemental damages awarded in this case.

III. SUMMARY OF THE ARGUMENT

In light of *Nautilus*, this Court must reconsider its prior definiteness decision under the intervening law exception to the law of the case, hold Dow's patents invalid, and vacate the supplemental damages judgment because the claims of the patents-in-suit, read in light of the specification and the prosecution history, fail to inform, with reasonable certainty, those skilled in the art about the scope of the invention. Dow's patent applications were filed with a missing figure that would have specifically informed the public (1) where on the slope of the strain hardening curve SHC should be measured and (2) what particular units should be used in that measurement. As a result, the patents-in-suit fail to give proper notice as to what materials are within the scope of Dow's claims. Whether the accused products fall within the claims depends entirely upon an arbitrary selection of undisclosed, nonstandardized slope location measurements and measurement units, particularly given the various options reasonably available to and used by Dow as of the filing date. Moreover, the only support for Dow's and thus the Court's construction of the SHC determination is conclusory extrinsic expert testimony. This is inappropriate when it was not supported by the intrinsic record at the time that the patent applications were filed, as required by *Nautilus*.

If the Court rules in NOVA's favor in light of *Nautilus*, it need not consider the merits of NOVA's supplemental damages issues. If it does address them, the

district court's finding that Dow possessed the necessary capacity to manufacture ELITE products was clearly erroneous, requiring the lost profits component of the supplemental damages award to be vacated in light of undisputed evidence that Dow's plants were already operating at full capacity. Dow failed to show any actual planning for additional ELITE production during the supplemental damages period. In fact, Dow actually reduced or deferred some production of its ELITE products during that period in favor of increasing the production of non-infringing products that did not require the use of octene.

The district court committed legal error and thereby abused its discretion when it adopted Dow's inflated profit margin because it was based on Dow's projected cost estimates in the face of undisputed evidence of Dow's higher actual costs.

IV. ARGUMENT

A. Standards of Review

Whether claims are considered indefinite in accordance with 35 U.S.C. § 112, ¶2, is a question of law reviewed *de novo*. *Dow*, 458 Fed. App'x at 917.

The amount of the patentee's damages determined in a bench trial is a finding of fact reviewed under the clearly erroneous standard. *SmithKline Diagnostics, Inc. v. Helena Labs. Corp.*, 926 F.2d 1161, 1164 (Fed. Cir. 1991). The clear error standard governs the trial court's findings about the general type of

damages to be awarded (e.g., lost profits), their appropriateness (e.g., foreseeability), and rates used to calculate them. *Home Sav. of Am. FSB v. United States*, 399 F.3d 1341, 1347 (Fed. Cir. 2005). However, certain subsidiary decisions are discretionary with the court (e.g., the choice of an accounting method for determining profit margin) and are reviewed under the abuse of discretion standard. *SmithKline Diagnostics,* 926 F.2d at 1164. "Abuse of discretion may be established by showing that the district court either made an error of law, or a clear error of judgment, or made findings which were clearly erroneous." *Seattle Box Co. v. Industrial Crating and Packing, Inc.*, 756 F.2d 1574, 1581 (Fed. Cir. 1985).

B. The Intervening *Nautilus* Decision Is Controlling in this Appeal, Requiring that the April 13, 2014 Supplemental Damages Judgment Be Vacated

1. Notwithstanding This Court's Prior Definiteness Opinion, the Definiteness Issue Must Be Reconsidered under the Intervening Law Exception to the Law of the Case

In *Harper v. Virginia Department of Taxation*, 509 U.S. 86, 97 (1993), the Supreme Court held "[w]hen this Court applies a rule of federal law to the parties before it, that rule is the controlling interpretation of federal law and ***must be given full retroactive effect in all cases still open on direct review*** and as to all events, regardless of whether such events predate or postdate our announcement of the rule." [Emphasis added.] *See also Amado v. Microsoft Corp*., 517 F.3d 1353, 1364 (Fed. Cir. 2008) (remanding the case so the district court could apply the

intervening *AT&T* decision to its post-verdict damages determination); *Sulzer Textil A. G. v. Picanol N.V.*, 358 F.3d 1356, 1368 (Fed. Cir. 2004) ("*Festo II* thus represented a change of law" and "is the new law that governs the appeal.")

Between the time that the parties filed their notices of appeal in this case and the filing of this brief, the Supreme Court decided *Nautilus, Inc. v. Biosig Instruments, Inc.*, 134 S. Ct. 2120 (2014). As discussed below, the *Nautilus* decision changed the law of indefiniteness as enunciated by the district court in its jury instruction and as applied by this Court in the prior appeal on this issue. Under *Nautilus*, the patents-in-suit are invalid as a matter of law, and no valid judgment therefore exists to support any award of supplemental damages to Dow.[9]

Although the district court's interlocutory holding of non-invalidity was appealed to and decided by this Court, "the decision was not final until the conclusion of the accounting." *Fresenius USA, Inc. v. Baxter Int'l, Inc.*, 721 F.3d 1330, 1342 (Fed. Cir. 2013) (*quoting Marconi Wireless Tel. Co. v. United States*, 320 U.S. 1, 47-48 (1943)), *cert. denied*, 2014 U.S. LEXIS 3538 (U.S. May 19, 2014). As discussed in *Fresenius*, the Supreme Court in *Simmons Co. v. Grier Bros. Co.*, 258 U.S. 82, 91 (1922), found error in the Third Circuit's refusal to consider an intervening Supreme Court decision on the claims' validity during the

[9] Unlike *Amado*, where there was an open factual question as to whether Microsoft's products were within the scope of 35 U.S.C. 271(f), 517 F.3d at 1364, there are no facts in dispute here that would require a remand on indefiniteness.

accounting phase because the Third Circuit's appellate mandate had not ended the case. 721 F.3d at 1342; *see also Mendenhall v. Barber-Greene Co.*, 26 F.3d 1573, 1581 (Fed. Cir. 1994) ("Absent a final judgment ending the litigation, the issue of liability is not barred from reconsideration.") Here, unlike in *Simmons*, the *Nautilus* decision issued too late for the district court to reopen and reconsider its indefiniteness decision on remand. Nonetheless, this Court must do so at this stage, as new law "even after a final decision on appeal is applicable in a pending case, so long as our mandate ending the litigation has not yet issued." *Fresenius*, 721 F.3d at 1346.

The law of the case doctrine is subject to recognized exceptions where "controlling authority has since made a contrary decision of the law applicable to the issues" or the earlier ruling was "clearly erroneous and would work a manifest injustice." *Gindes v. United States*, 740 F.2d 947, 950 (Fed. Cir. 1984) (quoting *White v. Murtha*, 377 F.2d 428, 432 (5th Cir. 1967)). When, as here, an intervening decision has changed the applicable law, this Court has relied upon such intervening changes as a basis for reconsideration. *See*, *e.g.*, *Mendenhall*, 26 F.3d at 1573 (Fed. Cir. 1994) (permitting liability to be reconsidered because "intervening controlling authority on the relevant issue of law[] would qualify as an exceptional circumstance under any articulated test"); *see also Insituform Techs., Inc. v. Cat Contr., Inc.* 10 Fed. App'x 871, 878 (Fed. Cir. 2001) (Court

reconsiders prior holding, as the "law of the case doctrine does not apply because *Festo* was an intervening decision that changed the law upon which the district court relied and the law upon which we relied on in *Insituform I* and *Insituform II"*), *vacated on other grounds*, 535 U.S. 1108 (2002).

2. *Nautilus* Changed the Standard for Definiteness

The Supreme Court in *Nautilus*[10] unanimously held that the previous Federal Circuit threshold requiring only that a claim be "amenable to construction" and not "insolubly ambiguous" was insufficient to satisfy the definiteness requirement of 35 U.S.C. § 112. 134 S. Ct. at 2129-30.

The disputed claim term "spaced relationship" at issue in *Nautilus* referred to the distance between a reference electrode and common electrode on the handle bars of an exercise machine. Nautilus moved for summary judgment, arguing that the term "spaced relationship," as construed, was indefinite under section 112, ¶2. The district court granted Nautilus' motion. The district court concluded those words in the claim "did not tell [the court] or anyone what precisely the space should be," or even supply "any parameters" for determining the appropriate spacing. *Id*. at 2127.

The Federal Circuit reversed and remanded. The panel majority stated that a claim is indefinite "only when it is 'not amenable to construction' or 'insolubly

[10] NOVA filed an amicus brief in support of Nautilus, Inc.'s position.

ambiguous'" and, under that standard, found the claims to be definite. *Biosig Instruments, Inc. v. Nautilus, Inc.*, 715 F.3d 891, 898 (Fed. Cir. 2013) (quoting *Datamize, LLC v. Plumtree Software, Inc.*, 417 F.3d 1342, 1347 (Fed. Cir. 2005)).

The Supreme Court, however, stated that "[i]t cannot be sufficient that a court can ascribe *some* meaning to a patent's claims; the definiteness inquiry trains on the understanding of a skilled artisan at the time of the patent application, not that of a court viewing matters *post hoc*." *Nautilus*, 134 S. Ct. at 2130 (emphasis in original). It noted that the expressions "insolubly ambiguous" and "amenable to construction," which had permeated the Federal Circuit's decisions concerning section 112, ¶2, left "courts and the patent bar at sea without a reliable compass." *Id*. at 2130 & n.9 (*citing, inter alia, Exxon*, 265 F.3d at 1375) ("If a claim is insolubly ambiguous, and no narrowing construction can properly be adopted, we have held the claim indefinite.") and *Hearing Components, Inc. v. Share Inc.*, 600 F.3d 1357, 1366 (Fed. Cir. 2010) ("the definiteness of claim terms depends on whether those terms can be given any reasonable meaning")).

In particular, the Supreme Court noted that the "amenable to construction" or "insolubly ambiguous" formulations have bred lower court confusion "for they lack the precision § 112, ¶2 demands." *Nautilus*, 134 S. Ct. at 2130. As an example, it cited a recent case where, even though the district court found a claim term "'lacks definiteness,' because it ***might mean several different things and 'no***

informed and confident choice is available among the contending definitions,'" the district court nonetheless concluded "that 'the extent of the indefiniteness ... falls far short of the 'insoluble ambiguity' required to invalidate the claim.'" *Id.* at 2129-30 n.8 (*quoting Every Penny Counts, Inc. v. Wells Fargo Bank, N.A.*, No. 11cv2816, 2014 WL 869092, *4 (M.D. Fla. Mar. 5, 2014) (emphasis added).

In place of the "insolubly ambiguous" standard, the Supreme Court instead articulated a "reasonable certainty" standard for claim definiteness: "[W]e hold that a patent is invalid for indefiniteness if its claims, read in light of the specification delineating the patent, and the prosecution history, fail to inform, with reasonable certainty, those skilled in the art about the scope of the invention." *Nautilus*, 134 S. Ct. at 2124. The Court stressed that, despite any inherent limitations of language, "a patent must be precise enough to afford clear notice of what is claimed"; anything less "would diminish the definiteness requirement's public-notice function and foster the innovation-discouraging 'zone of uncertainty.'" *Id*. at 2129, 2130 (*quoting United Carbon Co. v. Binney & Smith Co.*, 317 U.S. 228, 236 (1942)). The Court also noted that "the patent drafter is in the best position to resolve the ambiguity." *Id*. at 2129 (internal quotation marks omitted).

3. Judge Farnan's Jury Instruction and the Dow Panel Majority's Decision Conflict with *Nautilus*

As evidenced from the language in the jury instruction, Judge Farnan and the jury clearly relied on the indefiniteness standard from *Exxon* rejected by *Nautilus*. *Compare* A12702 ("If the meaning of the claim is discernible, it is definite, even though the task may be formidable and even if the conclusion may be one over which reasonable persons will disagree.") *with Exxon* 265 F.3d at 1375 ("If the meaning of the claim is discernible, even though the task may be formidable and the conclusion may be one over which reasonable persons will disagree, we have held the claim sufficiently clear to avoid invalidity on indefiniteness grounds."). An instruction which permits "reasonable people" to disagree about the scope of a claim à la *Exxon* is the antithesis of a standard that requires the intrinsic evidence to inform those skilled in the art about the scope of the invention "with reasonable certainty." *Nautilus*, 134 S. Ct. at 2123.

While the panel majority did not expressly use the words "amenable to construction" or "insolubly ambiguous" in its opinion, it was at the time bound to follow *Exxon* and likewise held that the test for indefiniteness was whether "'the meaning of the claims is discernible, even though the task may be formidable and the conclusion may be one over which reasonable persons will disagree ....'" *Dow*, 458 Fed. App'x at 920 (*quoting Exxon*, 265 F.3d at 1375). As in *Every Penny Counts*, the panel majority did not find Dow's claim language to lack sufficient

definiteness despite the fact that no informed and confident choice is available among the various places where the slope of the stress-strain curve could be measured or the various units that could be used in the SHC equation.

Compounding the problem, the panel majority also expressly relied upon *Exxon* to hold that, "[t]o be indefinite, a claim term must be such that 'no narrowing construction can properly be adopted' to interpret the claim." *Id*. at 917 (quoting *Exxon*, 265 F.3d at 1375). This is the identical language of *Exxon* cited by the Supreme Court with disapproval in footnote 9 of the *Nautilus* opinion, and is merely an alternate way of framing the question as being "amenable to construction" versus "insolubly ambiguous." Such a holding is at odds with the Supreme Court's rejection of the notion that it is sufficient that "a court can ascribe ***some*** meaning to a patent's claims." *Nautilus*, 134 S. Ct. at 2130 (emphasis added).

4. Dow's Claims Are Indefinite as a Matter of Law Under the New *Nautilus* Test

Dow's claims are invalid as a matter of law under *Nautilus* because the intrinsic evidence fails to inform, with reasonable certainty, those skilled in the art about the scope of the invention. Just as the district court in *Nautilus* found the term "spaced relationship" "did not tell [the court] or anyone what precisely the space should be," or even supply "any parameters" for determining the appropriate spacing, *Nautilus* 134 S. Ct. at 2127, Dow's patents do not tell anyone specifically

where on a stress-strain curve the slope of the curve should be measured or what units should be used for SHC. The district court acknowledged that such omissions were "troubling." (A498; A405.)

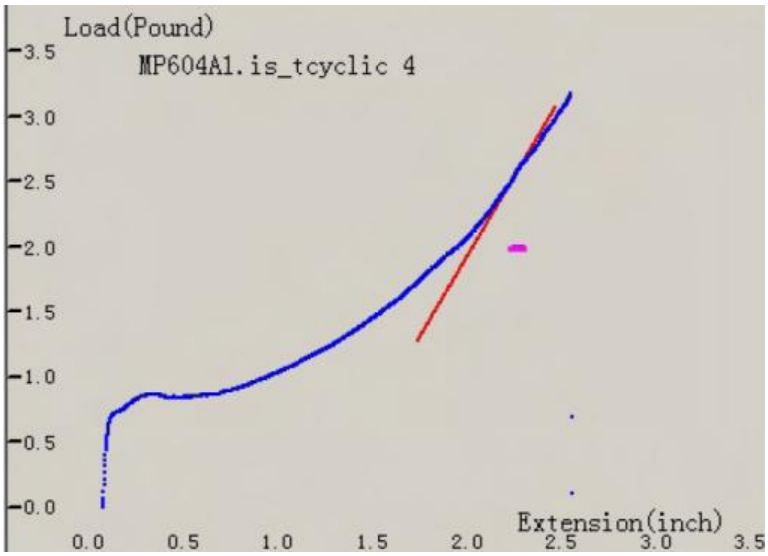
First, as acknowledged by the panel majority, SHC was "a new Dow construct, not previously se in the art ...." 458 Fed. App'x at 918; *see also* Sections II.C.2. (intro) and II.C.2.b.ii., *supra*. Accordingly, it would be impossible for a skilled artisan at the time to know with any certainty where on the curve the slope is to be measured or what units are to be used.

The only sources for determining this new SHC parameter are the patent specifications, which in turn refer the reader to the missing Figure 1 "show[ing] the various stages of the stress/strain curve used to calculate the slope of strain hardening." (A307, col. 6:40-44; *see also* A1406; A2559.) The four corners of Dow's patents are otherwise silent as to where the SHC slope location is to be determined and what measurement units should be employed.

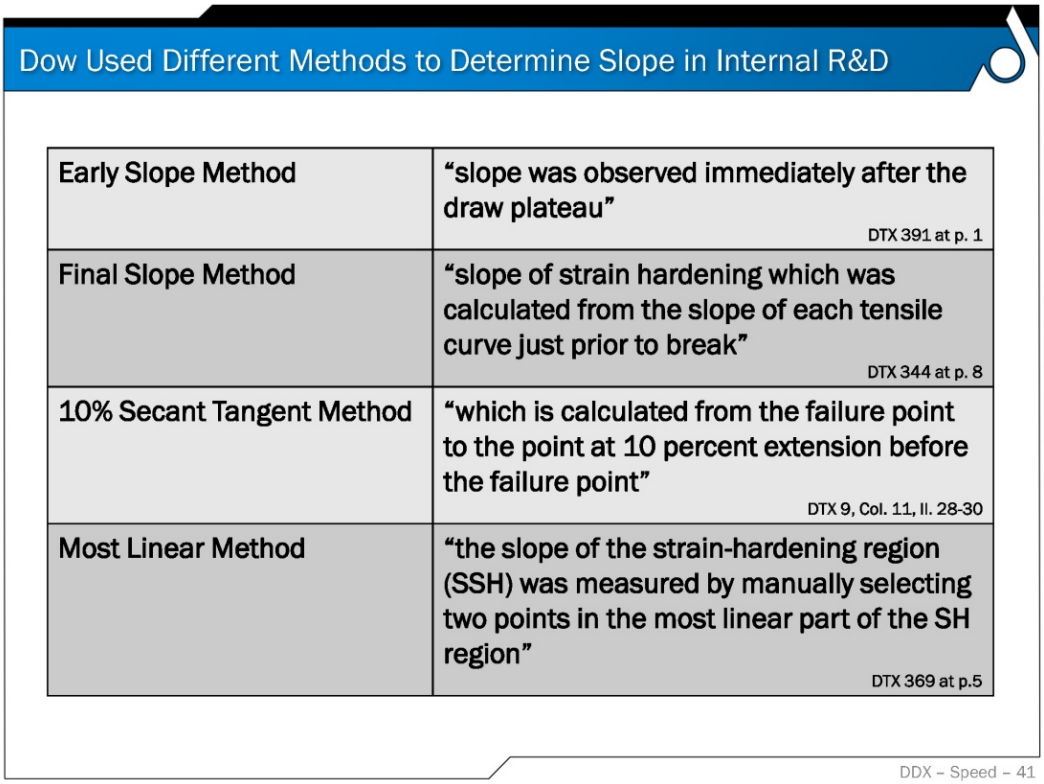
Second, as acknowledged by the panel majority, the "strain hardening region" of a stress-strain curve generated for the claimed polymer would "reveal some curvature (when graphed)" and "such curves do not have a single slope." 458 Fed. App'x at 919. Dow's expert, Dr. Hsiao, conclusorily testified that "one of ordinary skill in the art would know that the slope of the hardening curve would have to be measured at its maximum value, which reflects the best tensile

performance of the material." *Id*. Yet "best" tensile performance is itself a subjective determination that depends upon the customer's desired tensile properties for the final product made with the claimed polymer and varies from product to product. (A2384-85; A3351-54.) The record, however, not only fails to show with reasonable certainty that the "maximum" slope location approach was disclosed in the patents, or universally recognized and employed in the field, but it is devoid of any evidence whatsoever that anyone had ever actually used Dr. Hsiao's litigation-induced "maximum slope" location approach to measure slope.

As discussed in Section II.C.2.a.iii-iv, *supra*, Dr. Hsiao admitted that his opinion regarding the use of a "maximum" slope location was not disclosed in the patent or supported by any prior art reference. (A2517-18; A2550; A2565; A2632.) *See Honeywell Int'l, Inc. v. ITC*, 341 F.3d 1332, 1340 (Fed. Cir. 2003) (expert testimony on indefiniteness is insufficient where the ***intrinsic evidence*** fails to provide "guidance as to what one of ordinary skill in the art would interpret the claim to require."). Moreover, Dr. Hsiao admitted that certain of his red "maximum" slope lines were "not parallel to the strain hardening region as a whole" (A2582-83, A2646-47), as the patents-in-suit require. (A307 at col. 6:26-28.) Indeed, as shown below, his "parallel" line actually intersects the curve. (A5689; A2645.)



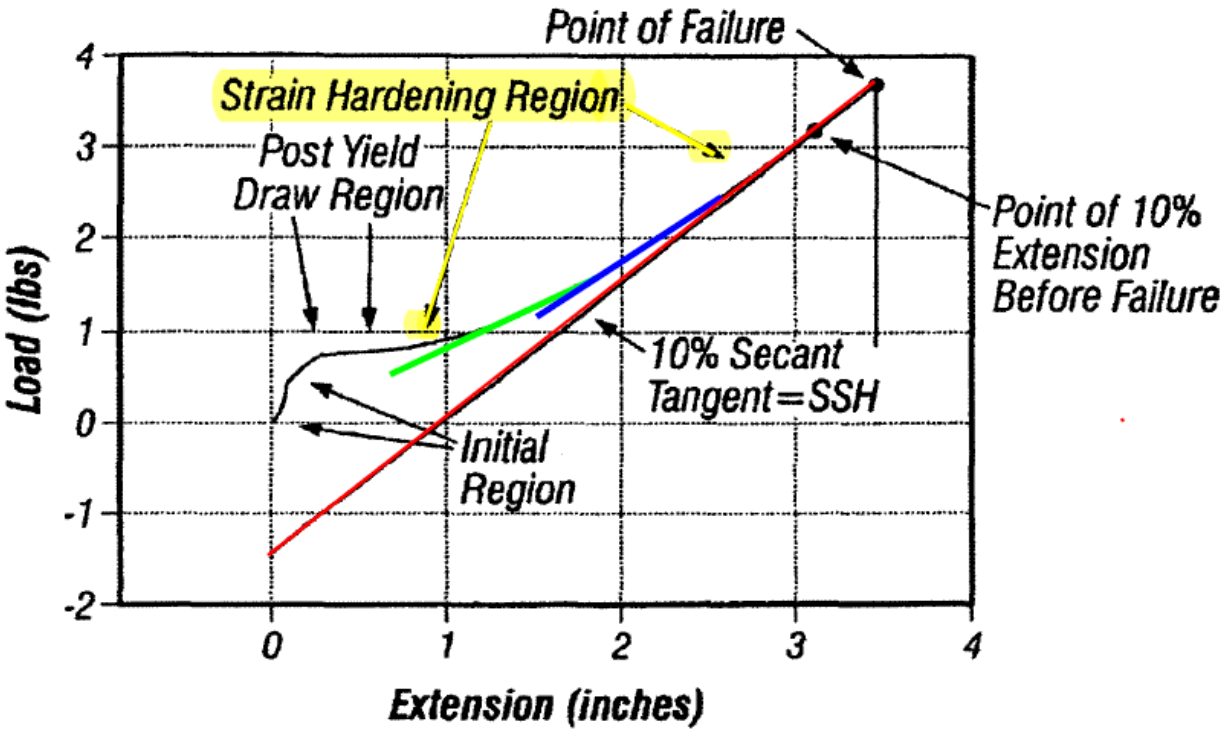
Nor is there any evidence that Dow itself ever used Dr. Hsiao's "maximum" slope location approach. Rather, the record reveals that Dow internally used four other, entirely different conventions to determine slope of strain hardening. (A2517-18; A2564-68; A2579-82; *see also* Sections II.C.2.a.ii. and II.C.2.a.iv., *supra*.)



It does not matter, as the panel majority opined, whether or not these Dow approaches "necessarily involved practicing the claimed method." 458 Fed. App'x at 919. They establish that there was no universally accepted way to measure slope of the strain hardening region of stress-strain curves generated for polymers in general, let alone for the specific claimed polymer at issue. Moreover, since SHC was a Dow-coined new parameter, Dr. Hsiao could not possibly know whether "one of ordinary skill in the art would know that the maximum slope of the stress/strain curve was the appropriate value *for calculating the SHC coefficient*." *Id.* (emphasis added).

Of course, one skilled in the art could determine the "maximum slope" location on a stress-strain curve if the artisan knew that such an approach was required by the patents-in-suit. But that begs the question whether Dr. Hsiao's "maximum" slope location approach was the location dictated by the patents. Ultimately, the panel majority concluded that "the mere fact that the slope may be measured in more than one way does not make the claims of the patent invalid." *Id*. at 920 (citing *Exxon*, 265 F.3d at 1375). The panel's reliance on *Exxon*, governing precedent at the time, is understandable, but nonetheless conflicts with *Nautilus* and constitutes legal error. Given that multiple other approaches were known and used by Dow, combined with the omission of the promised Figure 1, it cannot be said with reasonable certainty that the inventors informed those in the field to use Dr. Hsiao's "maximum" slope location approach.

Third, perhaps the most compelling evidence of a lack of certainty in the interpretation of the slope of strain hardening is Figure 2 in Dow's later related, co-pending '398 patent. (A6618 (Fig. 2); A6626 at col. 10:64 - A6627 at col. 11:4.) Unlike the patents-in-suit, this patent contains a Figure 2 showing "the various regions of a typical tensile curve ... and *the particular region* used to determine the slope of strain hardening [(SSH)]." (A6622 at col. 2:45-49 (emphasis added).)



The panel majority did not address this figure in its opinion. However, as can be seen, the strain hardening region comprises a substantial portion of the entire stress-strain curve and the slope of that region is highly variable. Rather than disclosing Dr. Hsiao's unsubstantiated "maximum" slope location approach, the figure instead discloses the use of the 10% secant tangent location to determine slope, one of the four locations that Dow had used internally at the time, and specifies the units involved (pounds per inch). Moreover, it "depict[s] the specific point along the curve at which the slope should be measured," something the panel majority posited that the omitted Figure 1 of the patents-in-suit would not necessarily suggest. 458 Fed. App'x at 919 n.11. This is further evidence that, not only was there no standard convention for measuring the slope of strain hardening

but that, when slope can be realistically measured in more than one way and the patent is silent on the question, reasonable uncertainty in fact exists as to how the slope of strain hardening is to be measured.

Fourth, the panel majority's opinion concedes that the patents-in-suit do not specify particular units for the SHC coefficient. *Id.* at 920. Its opinion improperly relies on extrinsic evidence to conclude that the slope of strain hardening should be measured and reported in all English units. However, the Instron Tensile Tester software user manual relied upon was not incorporated by reference into the patents-in-suit, and even Dr. Hsiao admitted that a skilled artisan would not look at this software user manual to determine the units. (A2684.) Moreover, while the Instron Tensile Tester could report output measurement in English units, the machines also had a setting to permit the operator to report the output measurement in either metric or SI units. (A2685-86; *see also* A3489-90.)

The record likewise reveals a lack of a standard convention in the field, as evidenced by Dow's internal work (not considered by Dr. Hsiao) and at least one other Dow patent employing other various units in addition to pounds per inch. (*See* Sections II.C.2.b.ii-iii and II.C.2.c., *supra*.) Dow's own infringement tests were measured by Dow in metric units, which yielded non-infringing SHC values, and only converted after-the-fact by Dow's expert to "lb/in" to yield values ≥ 1.3. (*See* Sections II.C.2.b.v, *supra*.)

Finally, the intrinsic evidence shows that the equation for SHC uses, at least in part, metric units for melt index, making it reasonable for a skilled artisan to similarly use metric units for the slope of strain hardening. Indeed, although the panel majority asserted that no evidence supported NOVA's position, 458 Fed. App'x at 920, NOVA had noted Dr. Fuller's testimony that it would have been both "uncommon" and "highly irregular" to mix English and metric units in a single equation. (A3499-500; A3576-77; NOVA's January 7, 2011 Brief at 31.) While the panel majority noted the reference to English units in Table 3 of the patents-in-suit, *id.*, that table reported the tensile properties of the ***final film***, not the ***Component A*** recited in the claims. Table 1 of the patents is the only table relevant to SHC and component A, and it refers ***only*** to ***metric*** units.

Moreover, even Table 3 would not mandate what specific English units should be used in measuring the slope of strain hardening. There is simply no way under the circumstances to divine with reasonable certainty if pounds per inch are required (as Dow maintains), if the stress units of pounds per square inch (psi) are required (as appear in Table 3), or if some other type of English units are required, creating a hopeless zone of uncertainty about the scope of Dow's claims.

Although the panel majority referred to many of the above factors supporting indefiniteness but still affirmed the district court's contrary finding, its

holding was based on a significantly less rigorous standard than that required by *Nautilus* and which – NOVA submits – would not pass muster under *Nautilus*.

In summary, these are seriously flawed patents. This is not a situation where some degree of uncertainty must be tolerated because of the inherent imprecision of claim language in the art. In fact, this is actually a stronger indefiniteness case than that before the *Nautilus* Court. Rather than having to decide the meaning of an ambiguous term such as "spaced relationship," Dow's patents plainly direct the reader to a Figure 1 to provide explicit guidance as to the particular location on the stress-strain curve for determining the "slope of strain hardening" and the units employed in that measurement. However, because the relevant Figure 1 is missing, essential information about this Dow-coined new parameter was left out of the patents.

Each of the reasons discussed above provides a sufficient basis to hold Dow's claims indefinite. Indeed, the diversity of Dow's own practices in measuring the slope of strain hardening and in the use of units demonstrates that skilled artisans could reasonably interpret the patent claim differently, revealing a substantial "zone of uncertainty" regarding the scope of the claimed invention. *Nautilus*, 134 S. Ct. at 21. Cumulatively, they more than establish reasonable uncertainty about the scope of Dow's patents. This zone of uncertainty inherent in Dow's defective patents thus fails to provide reasonable claim clarity as mandated

by *Nautilus*. Accordingly, this Court should overturn the judgment of no invalidity in light of the *Nautilus* decision.

C. The District Court Clearly Erred in Finding that Dow Had Satisfied the "Capacity" Prong of *Panduit*

If the Court rules in NOVA's favor in light of *Nautilus*, it need not consider the merits of NOVA's supplemental damages issues. If it does do so, to obtain lost profits damages, Dow must prove it had sufficient manufacturing capacity to cover NOVA's sales. *See Wechsler v. Macke Int'l Trade*, 486 F.3d 1286, 1293-94 (Fed. Cir. 2007) (denying claim for lost profits where patentee had not met burden of establishing third *Panduit* factor); *Gargoyles, Inc. v. United States*, 113 F.3d 1572, 1577 (Fed. Cir. 1997) (finding lack of capacity to meet the demand "was dispositive of the lost profits issue"). The district court's finding that Dow had met its burden on the capacity issue is clearly erroneous.

1. Dow's Hypothetical "Potential" Capacity Theory Is Legally Erroneous

The district court committed error by giving tacit legal approval of and dispositive effect to Dow's theoretical and speculative preference to make and promote the sale of a high value, high margin product like ELITE at the expense of other lower margin products, despite the particular market conditions existing at the time (e.g., no excess capacity, the octene shortage, etc.) and the real-life decisions that Dow actually made during the supplemental damages period (e.g.,

electing to make more HDPE and reduce or defer some ELITE production). All businesses "desire" to make more sales of high margin products at the expense of sales of low margin products. But if mere desire were enough, a patentee would meet its *Panduit* burden of proving sufficient capacity in every case. The law clearly requires more.

An argument similar to Dow's was unsuccessfully advanced by the patentee in *Polaroid Corp. v. Eastman Kodak Co.*, No. 76-1634-MA, 1990 WL 324105 (D. Mass. Oct. 12, 1990), *amended on recons.*, 1991 WL 4087 (D. Mass. Jan. 11, 1991). As in this case, the patentee's production facilities were running at essentially full capacity for the damages period. 1990 WL 324105, at *40 ("The seed of this dispute is that Polaroid's in-place manufacturing capacity was not sufficient to satisfy the combined demand which came fast and furious in the first three years of the infringement."). The *Polaroid* court rejected the contention that the patentee adequately proved that it could have made the sales by showing that it "theoretically could have taken steps to produce the combined volume." *Id.* Such a "could have" argument "renders the manufacturing capability requirement [under *Panduit Corp. v. Stahlin Bros. Fibre Works, Inc.*, 575 F.2d 1152 (6th Cir. 1978)] meaningless. Any patentee can concoct a scenario to achieve the necessary result." *Id*.

In holding that the patentee could have offset only a fraction of the infringing sales by expanding its production capacity, the *Polaroid* court assumed that Polaroid "would have labored to meet the demand to the best of its ability," but looked at concrete factual considerations about what "Polaroid reasonably and realistically could have done and what they would have done" as distinct from what might have been theoretically possible. *Polaroid*, 1990 WL 324105, at *41-42, *44-45. *Polaroid* is similar to *Smith International, Inc. v. Hughes Tool Co.*, where the court found the patentee to have insufficient manufacturing capacity where it "was strained to meet the sales actually available to it," with inventory levels that "rarely if ever exceeded its own minimum requirements of the level desirable for proper service of the market." No. CV 72-1231-HLH, 1986 WL 4795, at *18 (C.D. Cal. Mar. 12, 1986), *vacated on other grounds*, 839 F.2d 663 (Fed. Cir. 1988).

As in those cases, the market forces constrained Dow's flexibility to satisfy market demand for ELITE products. The district court's decision to ignore such real world evidence in favor of the self-serving, speculative testimony of Dow's Global Business Director that Dow would have "planned differently" is reversible error.

2. Mr. Hoffman's "Waterfall" Theory Is Speculative, Unrealistic, and Contrary To Actual Market Conditions and Dow's Responses to those Conditions During the Supplement Damages Period

For the supplemental damages period, Mr. Hoffman presented a new lost profits theory, whereby Dow would shift the production of some products between three of its plants and forgo production of other HDPE products in order to produce additional ELITE products at its plants capable of making those products. (A12327-28.) Mr. Hoffman's "waterfall" theory, however, is no more than an accountant's model, divorced from Dow's actual constraints and practices, and does not provide a reasonable degree of certainty regarding alleged lost profits. (A12467-68.) It is too abstract and speculative to legally support Dow's lost profits claims. *See Wordtech Sys., Inc. v. Integrated Networks Solutions, Inc.*, 609 F.3d 1308, 1321-22 (Fed. Cir. 2010) (awarding new trial where damages were based on speculation or guesswork).

Dow offered no proof of any actual consideration or planning for the drastic shifts in production postulated by Mr. Hoffman, dislocation of customers, shifts in product transportation costs, and the like. Moreover, as shown below, the record is replete with facts which demonstrate that Dow would not and indeed often could not have made the changes theorized by Mr. Hoffman.

a. Dow Lacked Excess ELITE Production Capacity

In contrast to the original damages period, for nearly the entire supplemental damages period, Dow's plants were running "flat out" and lacked excess ELITE production capacity. (A12349-53; *see also* A13098; A12084; A12247; A12238; A13964-69; A13971.) Mr. Hoffman admitted that Dow had zero "excess Elite capacity" for its Fort Saskatchewan plant in 2010-2011 and just 42 million pounds of "excess Elite capacity" at its Plaquemine facility in late 2011. (A12348-50.)

b. Dow Made Less ELITE and More HDPE

Contrary to Mr. Hoffman's "waterfall" theory, changed circumstances in 2010 compelled Dow to shift to making *less* ELITE and *more* HDPE at times during the supplemental damages period. (A12446-48; A13716; A13745.) Dow did so in part because its octene supply was "critically low" and it could not reach its target octene levels. (*See* Section II.C.3.b.iii, *supra*.) Yet, Dow's octene constraint is nowhere addressed by Mr. Hoffman's "waterfall" theory. (A12476-77; A14005.) Dow now describes its HDPE sales—which Mr. Hoffman would reduce—as inconsequential but Dow's actual contemporaneous documents show the importance of HDPE sales to its high profit food and beverage customers. (*See* Section II.C.3.b.iii, *supra*.)

c. Dow Could Not Acquire More Octene in 2010

Mr. Obregon's assertions that Dow could have renegotiated its octene contracts or switched customers to hexene resins (A12224; A12290) are unsupported. Mr. Obregon also admitted that Dow could not have bought or imported more octene because Dow had long term contracts, and it would have been "very difficult" to obtain more octene in 2010, a year of supply disruption due to force majeure. (A12263-65; A12276-79.) Mr. Obregon admitted that some DOWLEX customers would not have accepted a hexene "alternative" to DOWLEX (A12267), and in fact Dow's forced conversion from octene-based to hexene-based DOWLEX so frustrated Paragon's ability to make a profit that it stopped buying Dow's products. (A12386-87; A13140 ¶2.)

Dow's assertion that it would have taken steps to acquire the necessary amounts of octene is also belied by the fact that it took no steps in 2008, 2009, or prior to or after the jury trial in mid-2010 to secure more octene to make additional ELITE product in order to be ready to satisfy customer demand for LLDPE in the event that Dow prevailed at trial.

d. Dow Deferred Production of ELITE Products

In actuality, Dow did not forgo HDPE production to make space for additional ELITE, as assumed by Mr. Hoffman's "waterfall" theory. Instead, when faced with a clear opportunity to sell more ELITE, Dow "postponed production" of

a new ELITE grade requested by AEP until the end of July 2011 due to "[t]ight capacity at [Dow's] Plaquemine" production facility. (A13190; A12439-40.)

e. Dow Had Limitations on How Much HDPE It Could Free Up In Certain Months

Mr. Hoffman also failed to address production planning on a monthly basis, as Dow actually does. (*See*, *e.g.*, A12227-28; A13978; A12476.) However, for five months in the relevant time period Dow had insufficient HDPE production that could be freed up to make room for enough ELITE to match Dow's pro-rata market share of NOVA's SURPASS sales. (A12476-78; A14057.)

3. Dow's Incumbency Argument Is Unsupported by Law

The district court adopted Dow's argument that, but for NOVA's infringement, Dow would have begun the supplemental damages period in the position of the incumbent supplier and therefore would have planned for, and been able to produce, the additional ELITE products. (A16 at ¶23; A22-23.) Dow had maintained below that incumbency has significant value because Dow "would have built up relationships with those customers over years of sales but for Nova's infringement," (*See, e.g.*, A13094; A12035; A12143; A12147.) The reality, however, is that NOVA, not Dow, was the incumbent. Indeed, Dow's own damages expert, Mr. Hoffman, conceded that Dow "sure wasn't" the incumbent in fact and that "there were no actual Dow sales they would have been walking away from." (A12378.)

Dow had the burden below of proving that it had incumbent status, a novel theory that heretofore has never been recognized by any court. Given that it takes customers up to 18 months to qualify a new resin, there is simply no basis in either fact or logic from which to conclude that Dow could possibly have acquired any significant number of NOVA's customers during the 21.5 month supplemental damages period. Indeed, the record contains uncontroverted evidence that companies such as Paragon (a large NOVA customer) could have purchased, and in a number of cases, in fact did purchase acceptable alternatives from third parties rather than buying Dow's ELITE product. (A12516-21; A13168-71.) In the case of Paragon, Dow's forced conversion from octene-based to hexene-based DOWLEX so frustrated Paragon's ability to make a profit that it affirmatively stopped buying Dow's products. (A12386-87; A13140 ¶2.) Consequently, there is no basis in fact to conclude that, even in a "but for" world, that Dow would have been the incumbent during the supplemental damages period.

The sole case cited by the district court in alleged support of the incumbency theory is also inapt. *Grain Processing v. Am. Maize-Products Co.*, 185 F.3d 1341 (Fed. Cir. 1999), was not a supplemental damages case and did not address the issue of incumbency in a post-trial accounting setting. In fact, *no lost profits were awarded* in that case. *Id.* at 1356.

Accordingly, the award of lost profits should be vacated as a matter of law and the case should be remanded for the recalculation of reasonable royalties at the 2.11% rate found by the district court.

D. The District Court Committed Legal Error and Thereby Abused Its Discretion in Adopting Dow's Proposed Profit Margin When Mr. Hoffman's Reliance on Estimated Ethylene Costs Admittedly Overstated Dow's Margin on ELITE

"Damage awards cannot be based upon speculation or optimism, but must be established by evidence." *Hebert v. Lisle Corp.*, 99 F.3d 1109, 1119 (Fed. Cir. 1996). As this Court noted in *Carella v. Starlight Archery & Pro Line Co.*, 804 F.2d 135, 141 (Fed. Cir. 1986), "the amount of detailed testimony and documentation regarding computations of the patentee's lost profits will vary with the size and complexity of the patentee's company and with the extent to which that information is challenged or contradicted by the infringer."

In this case, it is not disputed that Mr. Hoffman calculated Dow's profit margin based on Dow's quarterly product cost ***estimate*** "PCE" values. (A12374-76; A12504-05; *see also* A13289; A14015-16.) In particular, he admitted that the PCEs he relied on did not include such costs. (A12376-77; *see also* A12479.) This is significant because Dow's cost for the ethylene raw material was the single largest component of its incremental costs (A12479), and because ethylene cost varies depending on plant location (A12563).

Mr. Hoffman could have investigated Dow's financial records, as he did in *Andrew Corp. v. Gabriel Electronics, Inc.*, 785 F. Supp. 1041, 1050-51 (D. Me. 1992). But he did not, despite being challenged by NOVA for failing to use Dow's actual standard costs.[11] Consequently, the district court's reliance on Mr. Hoffman's guesstimates is entitled to no deference and constitutes reversible legal error.

By using PCEs that contained inaccurate and low estimated ethylene cost figures, Mr. Hoffman understated Dow's costs and overstated Dow's profit margin on ELITE. (A12488.) For example, looking at 2010 data, Mr. Hoffman's average PCE unit cost was 35.5 cents per pound ("cpp") while Dow's actual ELITE standard cost was 41 cpp. (A12489-91; A12495-96; A14060; A13289.)

As explained by Mr. Tanger's unrebutted analysis, Mr. Hoffman understated Dow's Elite costs at 4-10 cpp, depending on the production plant and year. (A12558; A14063.) This equates to an $11.3 million overstatement[12] in Mr.

[11] Although NOVA did not object to Mr. Hoffman's use of quarterly PCEs in the original damages trial, that was because Dow did not produce documentation of Dow's actual ethylene costs in response to NOVA's discovery requests, and so NOVA lacked knowledge of Dow's higher actual costs in the earlier trial.

[12] Although Dow contends that a portion of this $11.3 million overstatement should be subject to an offset, a point upon which the district court made no findings, that argument necessarily admits that Mr. Hoffman's analysis was flawed and unreliable.

Hoffman's margin for ELITE, calculated from PCEs, as compared to the margin calculated using Dow's actual standard costs. (A12479-80; A12495-98; A14064.)

There can be no question that Dow's actual standard costs are the best available evidence on the question. When the accuracy of Mr. Hoffman's calculations were called into question, it was incumbent upon Dow to revise its proofs to employ its own actual standard cost data, which it failed to do.

This Court need not decide whether a patentee should be prohibited in all cases from relying on projected cost estimates. However, when, as here, the actual raw material standard costs are readily known to and in the possession of the patentee, affirmatively introduced and relied upon by the infringer at trial, and are shown to materially vary from projected cost estimates, it is a clear abuse of discretion for the court to award lost profits to the patentee based on such lower cost estimates. *See, e.g., Joy Techs., Inc. v. Flakt, Inc.*, 954 F. Supp. 796, 806 (D. Del. 1996) (denying lost profits claim because patentee's expert's calculations contained mathematical and logical errors significantly overstating lost profits); *Oiness v. Walgreen Co.*, 88 F.3d 1025, 1033 (Fed. Cir. 1996) (reversing and reducing lost profit award where patentee's damage theory rested on faulty assumptions and inaccurate estimates).

V. CONCLUSION

For the foregoing reasons, NOVA respectfully requests that this Court reverse the July 30, 2010 validity judgment, hold the asserted claims of the patents-in-suit invalid under *Nautilus*, vacate the April 13, 2014 supplemental damages award, and remand for further proceedings. Alternatively, NOVA requests that the Court vacate the supplemental award of lost profits and remand for a recalculation of reasonable royalty damages.

Respectfully submitted,

July 24, 2014

/s/ Darrel C. Karl

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ADDENDUM

IN THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF DELAWARE

THE DOW CHEMICAL COMPANY,	)	
	)	
Plaintiff,	)	
	)	
v.	)	C.A. No. 05-737 (LPS)
	)	
NOVA CHEMICALS CORPORATION (CANADA) and NOVA CHEMICALS INC. (DELAWARE),	)	
	)	
Defendants.	)	

~~[PROPOSED]~~ ORDER OF FINAL JUDGMENT

It is hereby ORDERED AND ADJUDGED that, consistent with this Court's March 28, 2014 Order and Memorandum Opinion (D.I. 760; D.I. 761) granting the request of Plaintiff, The Dow Chemical Company for supplemental damages but denying its requests for a finding of willful infringement and enhanced damages, FINAL JUDGMENT is hereby entered in favor of Plaintiff and against Defendants Nova Chemicals Corporation, and Nova Chemicals Inc. Plaintiff is awarded supplemental damages in the amount of $27,676,974. Additionally, Plaintiff is awarded prejudgment interest in the amount of $2,773,922 through April 4, 2014, plus an additional $2,711 per day from April 5, 2014 through the date of this ORDER.

THEREFORE, Plaintiff shall recover from the Defendants the sum of Thirty Million Four Hundred Fifty Thousand Eight Hundred and Ninety-Six Dollars ($30,450,896) up through and including April 4, 2014, plus additional prejudgment interest as provided by this ORDER, postjudgment interest as provided by law and its costs of action.

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April 4, 2014
8147766.1

SO ORDERED this 14th day of April, 2014.

UNITED STATES DISTRICT JUDGE

IN THE UNITED STATES DISTRICT COURT

FOR THE DISTRICT OF DELAWARE

THE DOW CHEMICAL COMPANY, Plaintiff, v. NOVA CHEMICALS CORPORATION (CANADA), AND NOVA CHEMICALS, INC. (DELAWARE) Defendants.	:	Civ. No. 05-737-LPS

ORDER

At Wilmington this 28th day of March, 2014, for the reasons stated in the Memorandum Opinion issued this same date,

IT IS HEREBY ORDERED that:

1. Dow's request for supplemental damages is GRANTED. The parties will calculate appropriate damages based on the Court's opinion, plus an appropriate amount of prejudgment and postjudgment interest.

2. NOVA's motion for stay of entry and execution of judgment (D.I. 746) is DENIED.

3. The parties shall meet and confer and shall submit, no later than April 4, 2014, a proposed form of final judgment consistent with the Court's rulings issued today.

Leonard P. Stark

UNITED STATES DISTRICT JUDGE

IN THE UNITED STATES DISTRICT COURT

FOR THE DISTRICT OF DELAWARE

THE DOW CHEMICAL COMPANY, Plaintiff, v. NOVA CHEMICALS CORPORATION (CANADA), AND NOVA CHEMICALS, INC. (DELAWARE) Defendants.	:	Civ. No. 05-737-LPS

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March 28, 2014
Wilmington, Delaware.

STARK, U.S. District Judge:

Pending before the Court is the issue of post-trial damages for the period of continued infringement from January 1, 2010 through October 15, 2011 ("Supplemental Damages Period") by Defendants, NOVA Chemicals Corporation (Canada) and NOVA Chemicals Inc. (Delaware) (together, "NOVA" or "Defendant"), of two of Plaintiff Dow Chemical Company's ("Dow" or "Plaintiff") patents. The Court held a two-day bench trial on this issue in April and May, 2013. Also pending is NOVA's motion for stay of entry and execution of judgment. (D.I. 746) For the reasons explained below, the Court finds that Dow is entitled to lost profits and reasonable royalties for the Supplemental Damages period, but is not entitled to enhanced damages. The Court also denies NOVA's motion for a stay of entry and execution of judgment.

BACKGROUND

A. The Parties, their Products, and the Patents

1. Plaintiff Dow is a chemicals company that manufactures plastics, including polyethylene ("PE") products, which it then sells to customers who convert the PE product into items such as stretch film or shampoo bottles. (D.I. 749 at 3)

2. Plaintiff is the owner of the patents-in-suit: U.S. Patent Nos. 5,847,053 and 6,111,023. (D.I. 1) The patents-in-suit expired on October 15, 2011. (D.I. 731 at 2, ¶1)

3. Dow manufactures and sells ELITE, a type of plastic product that is both stronger and thinner than other types of plastic, and is also an embodiment of the patents-in-suit. (D.I. 573 at 114-15)

4. Defendant NOVA manufactures chemicals, including PE and other products. (D.I. 14 at 2)

5. NOVA manufactures and sells SURPASS, a PE product that competes directly with Dow's ELITE. (D.I. 748 at 2; D.I. 573 at 179) NOVA also manufactures and sells XJS, an off-grade product that does not meet the target grade specifications, which has to be sold for a lower price. (D.I. 578 at 99)

B. The Jury Trial

6. Dow filed suit against NOVA for infringement of the patents-in-suit on October 21, 2005. (D.I. 1)

7. The Court held a jury trial in June 2010.

8. On June 15, 2010, the jury returned a verdict finding that NOVA's SURPASS and XJS products infringed one or more claims of the patents-in-suit. (D.I. 526) The jury awarded Dow $61,770,994.60 in total damages: $57,447,024.98 for lost profits and $4,323,969.62 in reasonable royalties. These damages were for NOVA's infringement of the patents-in-suit from March 2002 through December 31, 2009. (*Id.*)

9. The jury did not award Dow the full amount of damages it had sought. With respect to lost profits, Dow's expert, Creighton Hoffman, opined that Dow would have captured 80% of NOVA's sales, but the jury apparently applied only a 72% capture rate. (314-15, 377-78; *see also* 530, 536 (testimony of Dow expert Chelton Tanger))[1] As for a reasonable royalty, the

[1]All citations refer to the transcript from trial on supplemental damages held on April 30 (D.I. 743) and May 1, 2013 (D.I. 744), unless indicated otherwise (*e.g.*, D.I. 746). The Court's citations to the trial transcript are intended to refer both to the testimony itself as well as any admitted exhibits discussed by the witness in that portion of the transcript.

jury rejected Dow's proposed rate of 10% and instead applied an effective royalty rate of 1.755%. (346)

C. Post-Trial Proceedings

10. On July 30, 2010, the Court denied NOVA's motions for judgment as a matter of law and a new trial related to the jury's lost profits award. (D.I. 609) Generally, the Court concluded that "the jury was entitled to credit Mr. Hoffman's testimony" and, in particular, that "Mr. Hoffman's testimony and the related evidence on lost profits [supported] the jury's lost profits award." (*Id.* at 9) The Court further concluded that the evidence presented at the 2010 trial sufficiently established the absence of non-infringing alternatives that were acceptable to NOVA's customers who purchased NOVA's infringing SURPASS products. (*Id.*)

11. Also on July 30, 2010, the Court denied Dow's motion for a permanent injunction (D.I. 556), thereby permitting NOVA to continue to sell SURPASS and XJS products (D.I. 603). Among other things, the Court found that there were "substantial issues for appeal" and that NOVA customers would be injured by an injunction. (*Id.* at 2-3) After the denial of the permanent injunction, NOVA adopted and implemented a remedial policy not to sell the infringing SURPASS products to new accounts. (414)

12. NOVA filed an appeal. (D.I. 614, D.I. 617) On December 22, 2010, the Court stayed proceedings in this Court during the pendency of the appeal. (D.I. 634) On April 4, 2012, the Federal Circuit affirmed the judgment for Dow in all respects. (D.I. 637, D.I. 638) On June 7, 2012, the Court lifted its stay. (D.I. 652)

13. On August 20, 2012, the Court granted NOVA's motion for summary judgment of no willful infringement. (D.I. 674)

14. On October 29, 2012, the Supreme Court denied NOVA's petition for a writ of certiorari. *See Nova Chemicals Corp. v. Dow Chem. Co.*, 133 S. Ct. 544 (2012).

D. Bench Trial

15. As the parties were unable to agree on the amount of supplemental damages to which Dow is entitled, the Court scheduled a bench trial. (D.I. 652, D.I. 711)

16. At the pretrial conference on April 12, 2013, the Court, among other things, denied NOVA's motion to stay the damages proceeding or, in the alternative, stay entry and execution of judgment during the pendency of ongoing reexamination proceedings of the patents-in-suit by the U.S. Patent and Trademark Office ("PTO"). (D.I. 727, D.I. 741, D.I. 759) The Court's denial was without prejudice to renew the stay request with respect to entry and execution of the judgment in connection with briefing that would follow the bench trial on damages. (D.I. 741)

17. The Court held a bench trial on supplemental damages on April 30 and May 1, 2013. The parties completed post-trial briefing on July 18, 2013. (D.I. 747-751, D.I. 754)

18. NOVA renewed its motion for a stay on June 3, 2013. (D.I. 746) After briefing was completed on NOVA's motion on July 18, 2013 (D.I. 747, D.I. 752), the parties advised the Court of subsequent developments on October 24, 2013, November 6, 2013, and February 12, 2014 (D.I. 755, D.I. 756, D.I. 758), including the PTO's issuance of Office Actions rejecting all claims of both patents-in-suit (D.I. 755).

FINDINGS OF FACT[2]

[2]Each of the Court's findings of fact, in this section and throughout this Opinion, are based on the Court's assessments of what was demonstrated by a preponderance of the credible evidence, including the testimony of witnesses. *See* Fed. R. Civ. Proc. 52(a); *Minco, Inc. v.*

A. Dow's Polyethylene Products and Manufacturing Processes

1. Dow sells a variety of polyethylene ("PE") products into the marketplace through direct transactions with customers and through distribution channels. (67-68) The direct customers, called converters, receive PE product from Dow and then convert the PE product into rigid items such as shampoo bottles, or flexible film items such as stretch film. (68-69)

2. Dow's PE products include high-density PE, also known as HDPE, and linear low density PE, also called LLDPE. (68-69) HDPE is used to manufacture rigid types of product such as buckets, pales, milk bottles, shampoo bottles, and butter tubs. (*Id.*) Because of the product's low density, LLDPE can be used in a variety of flexible film products like trash bags, stretch wrap, and meat wrap. (*Id.*)

3. Dow's patented products are LLDPE products sold by Dow under the brand name ELITE. Dow considers ELITE to be one of its premium products. (69) ELITE falls under a subclass of LLDPE products referred to as mLLDPE, whereby "m" refers to the metallocene catalyst used in the production of ELITE, which causes the polymer to have different properties than the type of polymers made with other catalysts. (130) ELITE is sold into the Industrial and Consumer Packaging Market and the Food and Specialty Packaging Market. (130-31) ELITE is used in a variety of product applications, including stretch film and heavy-duty shipping sacks. (69-71, 131-32)

B. Polyethylene Qualification and the Value of Incumbency

Combustion Engineering, Inc., 95 F.3d 1109, 1118 (Fed. Cir. 1996) ("The patent holder bears the burden of proving the amount of the award...by a preponderance of the evidence.").

4. It is valuable for a supplier of PE, such as Dow or NOVA, to be the incumbent supplier of PE products to converters or customers because converters and customers tend to stay with their incumbent supplier to avoid having to qualify an alternative resin. (143-44) Former Dow marketing director Mr. Christopher Paul Thomson testified, "incumbency is a very powerful position, something we worked very hard to get, and we worked very hard . . . to maintain that position." (143)

5. Qualifying a PE resin is a demanding, time-consuming process in which suppliers work with converters to ensure that particular PE products are engineered to work with the converters' manufacturing equipment and deliver the properties required by the converters and the converters' customers. (144-47) NOVA's Quality Leader, Dr. Paul Tas, explained that the qualification process consists of eight steps: an initial qualification trial, a scale-up trial, testing of the film product manufactured by the converter, evaluating the results of such testing, evaluation by the converter's customers (the end users), a second scale-up trial, potential negotiations over product specifications, and then full acceptance. (189-95) Each trial alone takes three to fourth months. (192-95) Moreover, according to Dr. Tas, full acceptance can be delayed by "[a]nother three, fourth months" if the customer wants to negotiate the product specification after the scale-up trials. (195) Hence, the process for qualifying an alternative resin at a customer typically takes as long as 18 months. (195)

C. The Market for Dow's ELITE Products in the Supplemental Damages Period

6. In the Supplemental Damages Period, ELITE was Dow's premium linear low-density film product. (129) This was also true during the period prior to 2010. (130) Dow

positioned ELITE in the marketplace as having a superior overall balance of excellent physical properties and processability as compared to other products. (131) In 2010-2011, Dow targeted its ELITE product to the same customers that it had targeted in the period considered by the jury, 2002 to 2009 – that is, customers that valued processability and superior physical properties. (131)

7. In the Supplemental Damages Period, Dow's primary competitor for the ELITE film grades was NOVA's SURPASS. (132, 156) This was also true during the period prior to 2010 considered by the jury. (156) According to Mr. Thomson, NOVA's SURPASS and Dow's ELITE products were the most comparable products on the market in terms of processability and balance of physical properties at all pertinent times. (161)

8. In the Supplemental Damages Period, there were PE product offerings in the mLLDPE space other than ELITE and SURPASS. Those products included Exxon's EXCEED, LyondellBasell's STARFLEX, ChevronPhillip's MPACT/MARFLEX, and Westlake's MXSTEN. (156-57) Of all the products sold in the mLLDPE space during the Supplemental Damages Period, only SURPASS and ELITE were made using a dual-reactor, solution process; only ELITE and SURPASS could offer the benefits of the patented invention: the overall balance of excellent physical properties and excellent processability. (157-58, 313; PDEM_SD2205)

9. According to Mr. Thomson, the competitive dynamics between EXCEED, STARFLEX, MPACT and MXSTEN on the one hand, and ELITE and SURPASS on the other, did not change in the 2010-2011 time frame as compared to the 2002 to 2009 period considered by the jury. (159, 160-61) Thomson also testified that what customers found acceptable in the mLLDPE market did not change in 2010 and 2011 as compared with 2009. (161)

D. Dow's Strategy for Selling Polyethylene Products

10. Dow refers to its solution PE sales strategy as "Selling Out and Selling Up," which has been implemented into Dow's business strategy – including at the sales and production levels – since at least the early 2000s. (137-38)

11. "Selling out" refers to Dow's strategy of selling out its manufacturing capacity to fully utilize its reactors. (207-08)

12. Dow also focuses on "selling up" – that is, selling as much of its high value, high margin products as possible. (83-85; PTX 2035 at 19) As Dow strives to first "sell up" higher margin products like ELITE, it necessarily reduces sales of lower margin products like commodity-grade HDPE that is being displaced by the resources (such as manufacturing capacity) it invests in production of higher margin products. (83-86) Dow's commodity grade HDPE products, which Dow refers to as non-strategic products, are sold primarily on a transaction-by-transaction basis; that is, they are not generally subject to a long-term supply agreements. (102, 209)

E. Dow's Strategy for Manufacturing Additional ELITE

13. Dow's eight solution polyethylene trains have the capacity to produce over three billion pounds of polyethylene each year and over one billion pounds of ELITE each year. (77, 202-03, 234-35; PDEM_SD2106) Historically, Dow has made only about 500 million pounds of ELITE annually. (79, 235-37; PDEM_SD2304; PTX2042) Dow's ELITE-capable trains were fully utilized in 2010 and nearly fully utilized in 2011, but only about half of Dow's capacity on those trains was used to manufacture ELITE in those years; the remaining capacity was used to

manufacture DOWLEX[3] and HDPE. (235-37; PDEM_SD2305; PTX2042) Thus, Dow had the ability to produce another 500 million pounds of additional ELITE per year during the Supplemental Damages Period. (237)

14. Dow engages in an annual production planning session to determine what volumes of high value, high margin products like ELITE Dow can expect to sell in the following year. This planning session is completed at an annual, fourth-quarter meeting at which sales managers discuss realized sales and project ELITE's increased sales for the next year. (99-100, 204-05) Dow focuses on its current year's sales, and uses its yearly historical sales figures as a starting point to determine its projected ELITE production for the upcoming year, and then adjusts its projections to take into account the fact that Dow has experienced consistent year-over-year growth of ELITE since the product was launched. (*Id.*; PTX2515)

15. Dow's strategy is to load its solution reactors with high value, high margin products like ELITE and high margin DOWLEX, and then load any remaining capacity with lower margin, commodity-grade, non-strategic products like HDPE (as long as it would be profitable to do so). (209)

16. Dow could have manufactured additional ELITE products on its North American assets in 2010 and 2011. (216, 238) As explained by Mr. Jose Obregon, Dow's Global Business Director for Solution- and Gas-Phase PE, Dow could have accomplished this by planning differently. (238-39, 244) If Dow had been selling more ELITE in 2009, Dow would have planned to produce and sell more ELITE in 2010, and would have planned to contract for more

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[3]DOWLEX is another premium LLDPE that has different properties than ELITE (which is classified as an mLLDPE). (D.I. 749 at 3) Production of DOWLEX does not require an ELITE-capable (or dual-reactor) train. (D.I. 748 at 15)

raw materials to allow it to manufacture the additional volume Dow planned to sell. (205-06, 239, 241-43) As a result, Dow would have planned to produce and sell less of its low margin, commodity-grade, non-strategic HDPE products. (238-39, 244)

17. Dow could have met additional demand by allocating more of its capacity to its high value ELITE products, and less of its capacity to non-strategic, commodity-grade products like HDPE, consistent with the strategies discussed above. (216)

18. Dow has historically been able to procure the necessary volume of octene to meet the growth in demand for its ELITE products. (218-19) If Dow needed to manufacture an additional 100 million pounds of ELITE in the Supplemental Damages Period, Dow would have needed to purchase an additional 4.5 kilotons of octene. (221) Given that Dow purchases about 350 kilotons of octene annually, Dow could have and would have procured an additional 4.5 kilotons if needed to meet demand for ELITE, including by maintaining (rather than reducing) its contract levels for octene in 2009. (289-90)

F. Computation of Damages

19. At the jury trial in 2010, the jury awarded Dow $61,770,994.60 in total damages: specifically, $57,447,024.98 in lost profits and $4,323,969.62 in reasonable royalties. (303; PDEM_SD2402; D.I. 526) This was equal to more than 90% of the lost profits and 17.5% of the reasonable royalties Dow requested. (303; PDEM_SD2402) These damages were for NOVA's infringement of the patents-in-suit from March 2002 through 2009. (303)

20. The jury's lost profits award of $57,447,024.98 was 17.07% of NOVA's infringing sales of SURPASS in the 2002 to 2009 period of $336,505,027. (PTX556 at Supp. Ex. C-4; 534) If one applies this effective rate of 17.07% to the approximately $173 million in

infringing SURPASS sales during the Supplemental Damages Period (*see* PTX2572 at Revised Updated Suppl. Ex. D-9), the damages NOVA would owe Dow would be $29.5 million for SURPASS, plus damages for XJS sales.

21. Dow's expert Mr. Hoffman determined the amount of supplemental damages resulting from NOVA's continued infringement during the Supplemental Damages Period to be $30,962,780. (303-06, 345; PDEM_SD2404; PDEM_SD2437; PTX2572 at Revised Updated Suppl. Exs. I & J) Mr. Hoffman calculated damages separately for the period from January 1, 2010 until July 30, 2010 (the date judgment was entered), and the period from July 31, 2010 until October 15, 2011 (the date the patents expired). (305) Damages during the former period, adjusted for the jury award, totaled $7,621,670, while damages for the latter period totaled $23,341,110, which was not adjusted for the jury award. (*Id.*) Mr. Hoffman applied the jury's implicit royalty rate of 1.755% for the period January 1, 2010 through July 30, 2010, and applied a 10% royalty rate for the period July 31, 2010 to October 15, 2011. (305-06; PDEM_SD2405)

22. Mr. Hoffman's methodology for calculating Dow's damages in the Supplemental Damages Period was virtually the same as the methodology he had used at the 2010 jury trial. (303-04) Mr. Hoffman made certain adjustments to his calculation for the Supplemental Damages Period that were not necessary for his calculation of damages for the 2002 to 2009 period. For instance, for the Supplemental Damages Period, he deducted Dow's profits on sales of HDPE that Dow would have foregone to make additional ELITE. (*Id.*; PDEM_SD2403) This deduction was not necessary to calculate Dow's lost profits during the 2002-2009 period because, unlike during most of the Supplemental Damages Period, Dow had substantial empty plant capacity between 2002 and 2009. (303)

23. The jury's award to Dow of substantial lost profits damages indicates that the jury determined that Dow would have been the incumbent supplier for the bulk of NOVA's infringing sales in the 2002-2009 time period. (316-17) Mr. Hoffman concluded from the evidence presented that Dow would have, in the Supplemental Damages Period, maintained the business the jury determined Dow would have captured in the 2002-2009 period but for NOVA's infringement. The evidence established that there were no relevant material changes in NOVA's SURPASS customers or competitive dynamics in the Supplemental Damages Period from what had existed in the 2002 to 2009 period considered by the jury. (317-318; PDEM_SD2413; PTX2572 at Revised Updated Supplemental Exhibit L-1; 539)

24. Mr. Hoffman assessed Dow's supplemental lost profits by examining the *Panduit* factors: demand for the patented product; absence of acceptable, non-infringing alternatives; manufacturing capacity; and calculation of lost profits. (308-10)

a. As in the 2002 to 2009 period, demand continued to exist in the Supplemental Damages Period for the patented product. NOVA's sales of the infringing SURPASS product reached record levels in the Supplemental Damages Period. (310; PDEM_SD2408; PTX2572 at Revised Updated Suppl. Exs. H & D-7) In this latter period, NOVA sold about 246 million pounds of infringing SURPASS, and generated about $173 million in sales. (PTX2572 at Revised Updated Suppl. Ex. H; PDEM_SD2408)

b. As in the 2002 to 2009 period, there were no non-infringing alternatives in the Supplemental Damages Period that were acceptable to those customers who purchased the infringing SURPASS products. (341-42) No new relevant products came onto the market during the Supplemental Damages Period. (*Id.*) There were no products on the market, other

than SURPASS, that offered the same overall balance of excellent physical properties and processability as the patented ELITE products. (*See id.*) While NOVA's sales director testified that a trend existed to utilize multilayer structures in film products, allowing products with inferior processability (such as Exxon's EXCEED) to become more acceptable (410-11), this trend started in the 1990's and grew exponentially in the early 2000s – the exact time frame considered by the jury at the first trial. (299, 416-19) This "trend" is not a change that differentiates the Supplemental Damages Period from the 2002-2009 period.

c. As in the 2002 to 2009 period, Dow had adequate manufacturing capacity, including the capability to manufacture about 500 million pounds of additional ELITE annually, in the Supplemental Damages Period. (238)

d. As in the 2002 to 2009 period, Mr. Hoffman calculated Dow's lost profits for the Supplemental Damages Period to a reasonable degree of certainty, the only difference being that in the Supplemental Damages calculation he deducted the profit on HDPE that Dow would not have made in the "but for" world in order to make more ELITE. (326-27) Just as he did during the first trial, Mr. Hoffman calculated Dow's ELITE component costs based on Dow's quarterly standard cost sheets that Dow prepares in the normal course of business, which are referred to as product cost estimates ("PCEs"), and which are updated every quarter. (335-36) NOVA's expert, Mr. Tanger, also relied on Dow's PCEs at the first trial, as well as in his first expert report relating to the Supplemental Damages Period. (*Id.*; 549)

25. Dow seeks a reasonable royalty of $3,008,453 on NOVA's sales of infringing SURPASS not subject to lost profits, and infringing XJS sales during the Supplemental Damages Period based on a rate of at least 10%. (D.I. 749 ¶ 74 (citing 345; PDEM_SD2436; PTX2572 at

Revised Updated Suppl. Exs. I-3, I-4, J-3, & J-4) By contrast, NOVA insists that Dow is entitled only to the reasonable royalty rate of 1.755%, consistent with the jury's 2010 verdict. (D.I. 751 ¶ 77)

DISCUSSION

I. Dow's Request for Supplemental Damages

A. Legal Standards

The Court's power to award damages not found by a jury is based on statute. *See* 35 U.S.C. § 284 ("When the damages are not found by a jury, the court shall assess them."). The statute gives no guidance for awarding damages, so the Court must use its discretion in the matter. *See Grain Processing Corp. v. American Maize-Products Co.*, 185 F.3d 1341, 1349 (Fed. Cir. 1999) ("The district court must exercise sound discretion to determine the amount of damages, based upon its underlying factual findings and legal conclusions"); *Devex Corp. v. Gen. Motors Corp.*, 667 F.2d 347, 357 (3d Cir. 1981) ("[T]he awarding of damages involves a subtle judicial function; the statute allows considerable latitude (to the fact-finding court).").

To recover damages based on lost profits, the patent owner has the burden to show a reasonable probability that "but for" the infringement, it would have made additional sales of and additional profits on the patented product. *See Grain Processing*, 185 F.3d at 1349; *Wechsler v. Macke Intern. Trade, Inc.*, 486 F.3d 1286, 1293 (Fed. Cir. 2007). In awarding damages, the Court must rely on hypothetical economic projections. "The 'but for' inquiry therefore requires a reconstruction of the market, as it would have developed absent the infringement" to determine the amount of lost profits. *Grain Processing*, 185 F.3d at 1350.

Courts tasked with defining the parameters of the "but for" world of lost profits have articulated four elements the patent owner must prove to recover. "[T]he patent owner must prove

(1) a demand for the patented product, (2) an absence of acceptable noninfringing substitutes, (3) the manufacturing and marketing capability to exploit the demand, and (4) the amount of profit the patent owner would have made." *Siemens Medical Solutions USA, Inc. v. Saint-Gobain Ceramics & Plastics, Inc.*, 637 F.3d 1269, 1287 (Fed. Cir. 2011) (citing *Panduit Corp. v. Stahlin Bros. Fibre Works, Inc.*, 575 F.2d 1152, 1156 (6th Cir. 1978)). After the patent owner establishes a reasonable probability of "but for" causation, the burden shifts to the infringer to show that this conclusion is unreasonable. *See Grain Processing*, 185 F.3d at 1349.

B. Dow Proved It Should Receive Lost Profits for the Supplemental Damages Period

In the 2010 infringement trial, the jury found that Dow proved the *Panduit* factors for lost profits recovery. The Court finds each of these elements were also proven by a preponderance of the evidence for the Supplemental Damages period.

1. Continued demand for the patented product

Both Dow and NOVA continued to sell their ELITE and SURPASS products during the Supplemental Damages period. NOVA does not contend that Dow failed to prove continued demand for the patented product.

2. Absence of acceptable non-infringing alternatives

To prove that there are no acceptable noninfringing alternatives, the patent owner "must show either that (1) the purchasers in the marketplace generally were willing to buy the patented

product for its advantages, or (2) the specific purchasers of the infringing product purchased on that basis." *Standard Havens Prods. v. Gencor Indus., Inc.,* 953 F.2d 1360, 1373 (Fed. Cir. 1991). It follows that "if purchasers are motivated to purchase because of particular features available only from the patented product, products without such features – even if otherwise competing in the marketplace – would not be acceptable noninfringing substitutes." *Id.*

NOVA argues that the market for polyethylene was changing after 2009: customers were finding products "acceptable" that may not have been considered acceptable before (517-21); processability, the main selling point for the ELITE and SURPASS products, was becoming less important to customers (441-42); and competition was increasing in the market for multi-layer resins (170-71). However, the evidence shows that while these trends did exist in the Supplemental Damages Period, they likewise existed in the 2002 to 2009 period considered by the jury, and indeed began in the 1990s. (299, 416) Moreover, qualifying polyethylene products is a time-consuming and demanding process, often taking as long as eighteen months, which prevents customers from easily moving their business among competitors. (195) The Court finds no material changes in the market for acceptable alternatives took place between the time of the jury trial and the time now under consideration (until October 15, 2011). NOVA failed to show that any new products had been introduced into the market during the Supplemental Damages period which would qualify as non-infringing alternatives.

3. Marketing and manufacturing capability to meet demand

NOVA argues that Dow had no manufacturing capacity during the Supplemental Damages Period, and since there was no excess ELITE capacity there could be no lost profits. While Dow's manufacturing trains were fully utilized during the Supplemental Damages Period

(349-52), Dow was producing HDPE, a lower-grade product than ELITE, and a product having a lower profit-margin (141, 209). The Court is persuaded that, but for NOVA's infringement, Dow could have and would have been utilizing its production facilities to the full extent necessary for manufacturing ELITE instead of HDPE.

The evidence at trial showed that Dow's business model, throughout the 2000s and through the Supplemental Damages Period, was to optimize production of high profit-margin products like ELITE over other, less profitable products. (235-37) HDPE was just one example of a less profitable product, which Dow used as a capacity filler to prevent production trains from sitting idle.[4] (*Id.*) Given the opportunity, Dow showed it would "sell up" its plants by decreasing sales of HDPE and other non-strategic products while increasing sales of high margin products like ELITE. (141)

Dow had the capability to forgo its HDPE manufacturing and prioritize its production of ELITE, which would result in higher profits for the company. Unlike ELITE, which was sold as part of a long-term contract agreement (94-96, 102), Dow sold HDPE on a transaction-by-transaction basis (102, 209). The Court found persuasive the testimony presented by Dow outlining its business practice of displacing HDPE production to free up capacity for ELITE or other high profit-margin products. (141) Under this "waterfall" production system, HDPE manufacturing would be moved to facilities with a one-reactor production train, while the two-reactor facilities which are necessary for ELITE production (at The Fort and Plaquemine) would be freed up to produce ELITE products. (75-76, 80-81)

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[4]Dow's expert accounted for the profits from sales of HDPE during the Supplemental Damages period as filler to keep the production lines at full capacity, and deducted those sales from his lost profits calculation. (303-04)

The Court further agrees with Dow that but for NOVA's infringement, Dow would have begun the Supplemental Damages Period in the position of incumbent supplier (for the NOVA sales it would have captured). Dow would have planned for, and produced, the additional ELITE products. (99-100, 204-05) Furthermore, the Court is unconvinced by NOVA that Dow would have been defeated by a shortage of octene and finds that Dow had access to the octene necessary to fulfill the demand for ELITE in the but for scenario.

4. Amount of profit the patent owner would have made

Dow has proven its lost profits by multiplying NOVA's total volume of SURPASS sold by the capture rate of sales by the profit margin per pound on ELITE. While NOVA contests Dow's estimates on both the capture rate and the profit margin, the Court finds that Dow met its burden to prove lost profits under the *Panduit* factors, subject to the limitations described below.

a. Capture Rate

As part of its lost profits calculations, Dow asserts that but for NOVA's infringement, Dow would have captured 80% of the SURPASS sales (in other words, that 80% of NOVA's sales would have instead been sales of Dow's ELITE products). Dow presented the same 80% estimate to the jury at the 2010 trial, but the jury reduced Dow's requested damages award, implying a capture rate of no lower than 72%.[5] (314-16) As the Court is persuaded that the record warrants essentially extending the jury's award to cover the Supplemental Damages Period, the Court deems it most appropriate to use the 72% capture rate, rather than Dow's

[5]The jury award was approximately 90% of Dow's requested lost profits amount, and approximately 17% of its requested royalties amount. (305)

requested 80% capture rate. The remaining 28% of infringing SURPASS sales will be subject to a reasonable royalty, as discussed below.

NOVA argues that the jury award fully compensates Dow for any injury, including lost customer relationships, and so there would be no captured sales in the "but for" world during the Supplemental Damages period. (D.I. 750 at 5) NOVA's theory is that the jury award essentially acts to license all infringing sales from 2002 to 2009, and so NOVA rightfully became the incumbent for these sales at the start of the Supplemental Damages Period. (*Id.* at 6) The Court disagrees. *See Grain Processing Corp. v. Am. Maize-Prods. Co.*, 185 F.3d 1341, 1350 (Fed. Cir. 1999) (stating that but for inquiry "requires a reconstruction of the market as it would have developed absent the infringing product, to determine what the patentee would . . . have made"). NOVA's approach would give it, an adjudicated infringer, a benefit of substantially reduced damages solely due to the happenstance that the jury trial took place during, and not after, infringement. (*See generally* D.I. 754 at 2) The Court will not exercise its discretion in NOVA's favor on this basis.

b. Profit Margin

Dow estimates an incremental profit per pound of $0.37 (329; PDEM_SD2427), while NOVA contends these estimates understate ELITE costs by 4-10 cents per pound ("cpp") and overstate Dow's profit margin (D.I. 750 at 25). NOVA's contention is based on its reliance on Dow's Profit and Loss Statements' "Standard Cost - Trade" line item, while Dow's estimates are based on ELITE component costs from Dow's quarterly standard cost sheets, referred to as product cost estimates ("PCEs"). (D.I. 750 at 24) During the jury trial, Dow's expert used the PCE estimates and the same methodology (335-37) – as did, notably, NOVA's expert (549).

Dow contends that the "Standard Cost - Trade" estimates include fixed costs, which result in a $0.04 to $0.07 increase in cost per pound over the PCEs. NOVA contends, by contrast, that PCEs are only estimates and that the calculation of lost profits should be based on actual cost. (371-72)

The Court sides with Dow on this dispute. Accordingly, the Court will apply Dow's proposed profit margin, $0.37 cents per pound. (329; PDEM_SD2427)[6]

5. Reasonable royalties

The Court has discretion in determining a reasonable royalty based on all the evidence and facts presented, including the results of a hypothetical negotiation between the patent owner and the infringer, at the appropriate time. *See Georgia-Pacific Corp. v. U.S. Plywood Corp.*, 318 F.Supp. 1116, 1120 (S.D.N.Y. 1970). "The primary inquiry, often complicated by secondary ones, is what the parties would have agreed upon, if both were reasonably trying to reach an agreement. . . . It requires consideration not only of the amount that a willing licensee would have paid for the patent license but also of the amount that a willing licensor would have accepted." *Id.* at 1121.

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[6]The Court has considered all of NOVA's arguments, even those not explicitly discussed in this Opinion. For instance, the Court evaluated each of the specific criticisms NOVA and its expert leveled on Dr. Hoffman's analysis, but finds these criticisms – individually and collectively – do not render Hoffman's analysis unreliable. Additionally, the Court overrules NOVA's renewed objections to certain portions of Hoffman's testimony at trial as being beyond the scope of Hoffman's pretrial disclosures, as the Court agrees with Dow that none of the challenged testimony on which Dow seeks to rely was anything other than a reasonable elaboration on the opinions Hoffman disclosed prior to trial, particularly as they are generally responses to criticisms from NOVA's expert to which Hoffman had no prior opportunity to respond. (*See* D.I. 754 at 9, 15)

Both parties agree that the royalty rate of 1.755% awarded by the jury should be applied to the period up through July 30, 2010, the date on which the Court denied Dow's request for a permanent injunction to halt further sales of NOVA's SURPASS. The parties disagree as to the appropriate royalty rate for the remainder of the Supplemental Damages Period. NOVA asserts that the jury's 1.755% royalty rate should apply for the entire Supplemental Damages period. (D.I. 750 at 34) Dow contends, instead, that the 1.755% should be increased nearly six-fold to a 10% royalty rate. (D.I. 748 at 26) The Court is not persuaded to adopt either proposed royalty rate.

In exercising its discretion based on the particular facts proven here, the Court concludes that an appropriate royalty rate for the period of July 31, 2010 through October 15, 2011 is 2.11%, which is a 20% increase over the jury's rate of 1.755%. The Court reaches this conclusion based primarily on the evidence that no material changes took place between the 2002 to 2009 period and the Supplemental Damages Period other than NOVA changed from being an alleged infringer to being an adjudicated infringer, a factor that would have reduced NOVA's relative bargaining power in the hypothetical negotiations after trial. The Court has also factored in that former Judge Joseph J. Farnan, Jr., who presided over the 2010 jury trial, denied Dow's motion for a permanent injunction.

C. Damages for Infringing Sales of XJS

In addition to reasonable royalties from NOVA's sales of infringing SURPASS products, Dow is entitled to reasonable royalties on the sales of infringing XJS products. During the Supplemental Damages period, NOVA sold approximately 12.5 million pounds of infringing XJS products.

(PTX2572 at D-10) Dow requests $68,212 for the period prior to July 30, 2010, which is adjusted to reflect the jury's award (at a rate of 1.755%). (D.I. 748 at 25; PDEM_SD2437) On the remaining sales during the Supplemental Damages period, the Court applies the same 2.11% royalty rate as above. Since NOVA had a net revenue of $3,715,761 on XJS resins between July 30, 2010 and October 15, 2011 (PTX2572 at D-10), the additional royalties for this period are $78,403. Therefore, the total amount of royalties for the infringing XJS sales between January 1, 2010 and October 15, 2011 is $146,615.

D. The Court Will Not Award Enhanced Damages

Dow requests enhanced damages for NOVA's willful post-judgment infringement during the sixteen months between when the Court denied Dow's request for a permanent injunction and the expiration of the patents-in-suit. Dow argues that the Court may enhance a supplemental damages award upon a finding that the adjudicated infringer's continued infringement in the face of an adverse jury verdict and judgment was willful. The Court, however, agrees with NOVA that the grant of NOVA's motion for summary judgment of no willfulness effectively decided this issue in NOVA's favor. (D.I. 674)

The Court is not persuaded it should revisit the question. Dow's reliance on *SynQor, Inc. v. Artesyn Techs., Inc.,* 709 F.3d 1365 (Fed. Cir. 2013), is unavailing. *SynQor* shows that a Court may award enhanced damages even in the absence of a prior finding of willful infringement.

Here, rather than no finding at all regarding willfulness, there is a finding of ***no*** willful infringement.

E. Interest

The parties appear to be in agreement that Dow is entitled to prejudgment interest, at the prime rate compounded quarterly, for the Supplemental Damages Period, with the exception of December 11, 2012 through April 13, 2013 (due to the Court's continuance of trial at Dow's request). (D.I. 711 at 3; D.I. 748 at 30) The parties appear to further agree that Dow is entitled to postjudgment interest as authorized by 28 U.S.C. § 1961. (D.I. 748 at 30; DI 750 at 2)

II. NOVA's Motion for Stay of Entry and Execution of Judgment

NOVA requests the Court stay entry of judgment, or at least stay execution of judgment, until after completion of the ongoing reexaminations of the patents-in-suit. NOVA argues that it risks substantial and irreparable harm by paying the supplemental damages being awarded by the Court, as the PTO may eventually invalidate the patents-in-suit. NOVA has already paid the damages awarded by the jury in the 2010 trial and does not contend that it would be permitted to recover that payment even if the PTO invalidates the patents-in-suit. Rather, NOVA distinguishes the supplemental damages, on the grounds that there is a fundamental distinction between an executed judgment and an as-yet unexecuted judgment. NOVA further argues that, at a minimum, the Court should require NOVA to place funds in escrow sufficient to pay the amount of supplemental damages being awarded, which would purportedly not cause any prejudice to Dow.

NOVA's arguments implicate potentially difficult questions. If the patents-in-suit are invalid, NOVA cannot infringe them, and should not have to pay Dow anything further. But as

of today the patents-in-suit remain valid, and they will be so until at least after any PTO proceedings are completed and any appeal to the Federal Circuit is completed (or the time for such appeal expires without any appeal being filed). Additionally, and weighing heavily against granting the requested stay, NOVA waited until very late in this litigation to initiate the reexaminations and to seek a stay. NOVA filed its petitions for reexamination of the patents-in-suit on December 17, 2012, more than seven years after this case was filed and more than two years after the jury's verdict on infringement and invalidity. Under the circumstances, the Court believes the most appropriate exercise of its discretion is to deny the requested stay.

The Court has discretion to stay litigation pending reexamination. *See Smarter Agent, LLC v. Mobilerealtyapps.com, LLC*, 889 F.Supp.2d 673, 674 (D. Del. 2012) (citing *Ethicon, Inc. v. Quigg*, 849 F.2d 1422, 1426-27 (Fed. Cir. 1988)). In considering a stay request, the Court must weigh the competing interests of the parties by considering: "(1) whether a stay will simplify the issues and trial of the case, (2) whether discovery is complete and a trial date has been set, and (3) whether a stay would unduly prejudice or present a clear tactical disadvantage to the non-moving party." *Smarter Agent*, 889 F.Supp.2d at 674. The Court has considered these factors and find they disfavor a stay: given that trial is completed and the Court has ruled on the parties' disputes, no decision from the PTO could have any simplifying effect; the proceedings in this Court are essentially completed, while the reexaminations are ongoing; and a stay would unduly prejudice Dow, which has litigated this case for close to a decade, and prevailed at two trials and on appeal, and is entitled to be paid damages as compensation for all the harm it has suffered.

The Court will deny NOVA's motion for a stay.

CONCLUSION

For the foregoing reasons, Dow's request for lost profits and reasonable royalty damages will be granted. Dow's request for enhanced damages will be denied. NOVA's request for a stay of entry and execution of judgment will be denied.

Case: 14-1431 Document: 24 Page: 108 Filed: 07/24/2014

US005847053A

United States Patent [19]

Chum et al.

[11] **Patent Number: 5,847,053**

[45] **Date of Patent: Dec. 8, 1998**

Dow v. Nova 05-737 (JJF) (D.Del.)
Dow Trial Exhibit
PTX - 1

[54] **ETHYLENE POLYMER FILM MADE FROM ETHYLENE POLYMER BLENDS**

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

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

[21] Appl. No.: **834,050**

[22] Filed: **Apr. 11, 1997**

Related U.S. Application Data

[63] Continuation of Ser. No. 544,497, Oct. 18, 1995, Pat. No. 5,677,383, which is a continuation of Ser. No. 378,998, Jan. 27, 1995, abandoned, which is a continuation of Ser. No. 54,379, Apr. 28, 1993, abandoned, which is a continuation-in-part of Ser. No. 776,130, Oct. 15, 1991, Pat. No. 5,272,236.

[51] **Int. Cl.**^{6} **C08L 23/08**

[52] **U.S. Cl.** **525/240**

[58] **Field of Search** 525/240

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Primary Examiner—David W. Wu

[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

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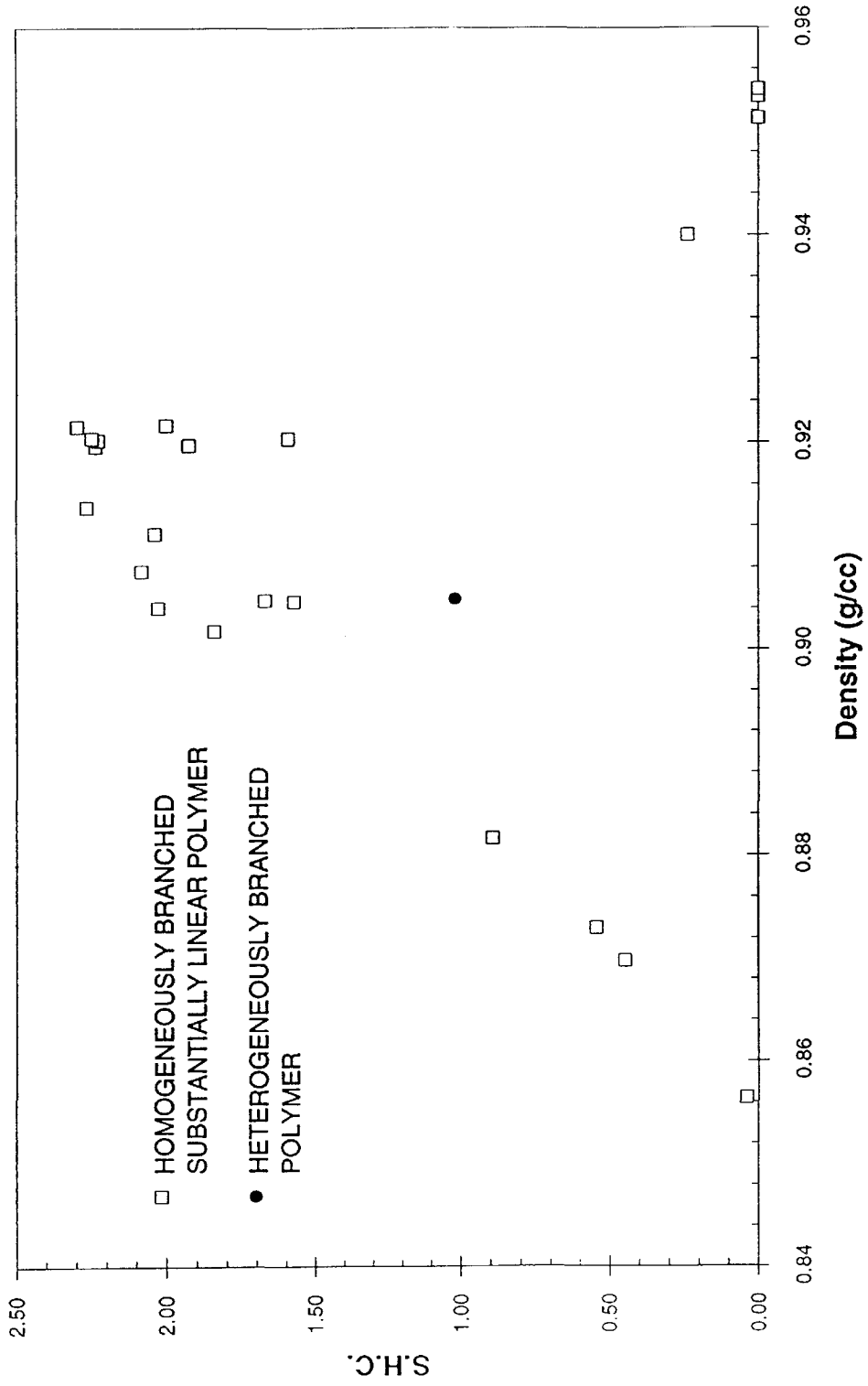
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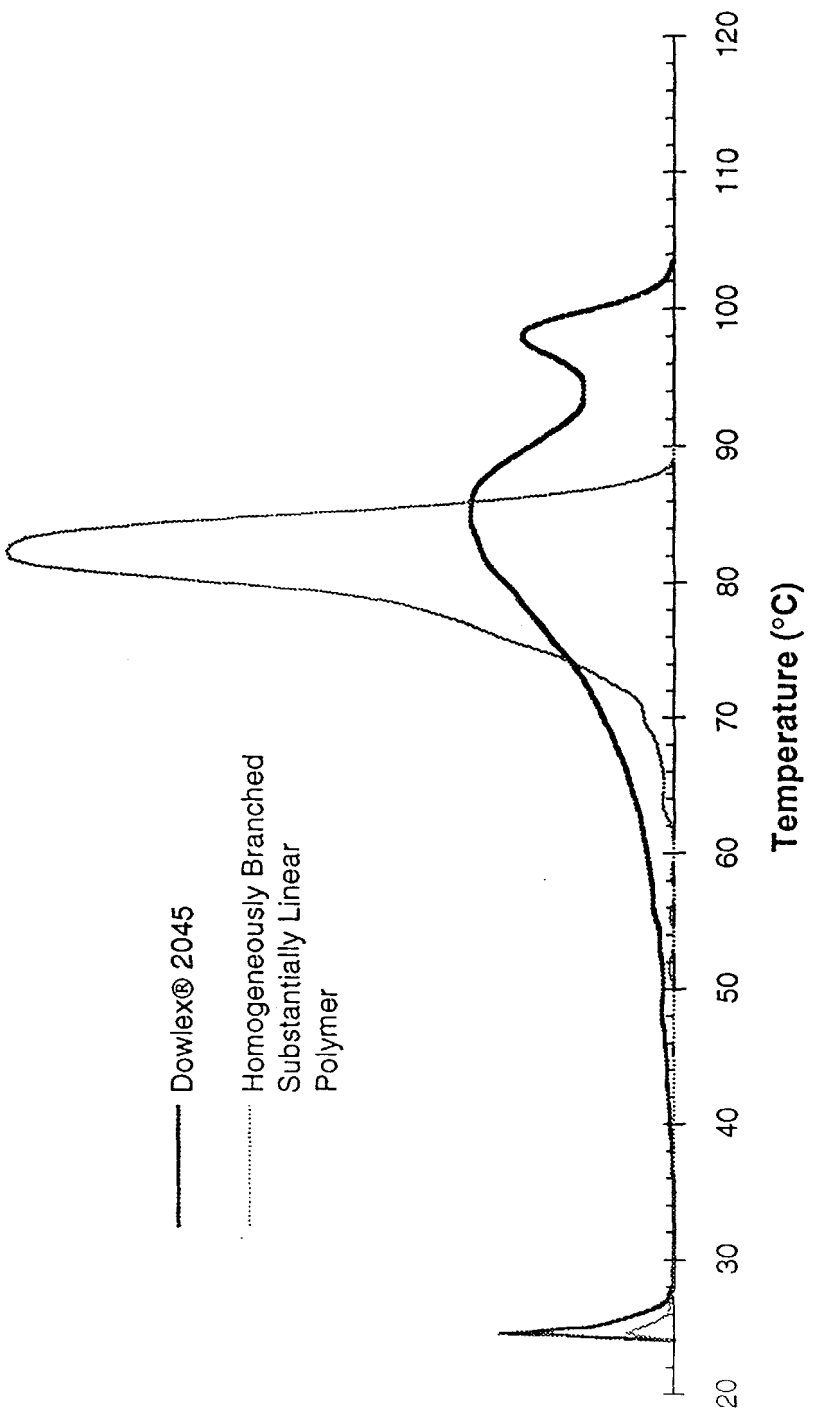
FIGURE 1



Case: 14-1431 Document: 24 Page: 112 Filed: 07/24/2014

Figure 2

Analytical Temperature Rising Elution Fractionation (ATREF)
Comparison of SCBD



ETHYLENE POLYMER FILM 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, filed Oct. 18, 1995, now issued U.S. Pat. No. 5,677,383, which was a Rule 1.60 continuation application of application Ser. No. 08/378,998, filed Jan. 27, 1995, now abandoned, which was a Rule 1.62 continuation application of application Ser. No. 08/054,379, filed Apr. 28, 1993, now abandoned, which was a continuation-in-part application of Ser. No. 07/776,130, filed Oct. 15, 1991, now issued U.S. Pat. No. 5,272,236, the disclosures of each of which are incorporated herein in their entirety by reference. This application is also related to application Ser. No. 08/475,737, filed Jun. 7, 1995, now abandoned; application Ser. No. 07/939,281, filed Sep. 2, 1992, now issued U.S. Pat. No. 5,278,272; and application Ser. No. 08/510,527, filed Aug. 2, 1995, now abandoned which is a continuation application of application Ser. No. 08/010,958, filed Jan. 29, 1993, now abandoned; the disclosures of each of which are 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 homo-

geneously 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 now U.S. Pat. No. 5,272,236. 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 Ser. No. 07/776,130 now U.S. Pat. No. 5,272,236 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: $M_w=\Sigma w_i*M_i$, 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 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:

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

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	2.61	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, now abandoned 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 now abandoned describes, inter alia, interpolymerizations 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. No. 4,340,563, U.S. Pat. No. 4,663,220, U.S. Pat. No. 4,668,566, or U.S. Pat. No. 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*, Jun. 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 add (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$\geq$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 clamping 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 clamped 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 clamp=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 24, 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, now abandoned 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, now U.S. Pat. No. 5,250,612 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/l-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, now U.S. Pat. No. 5,250,612 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	473
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
Frostline 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 now abandoned) 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 now U.S. Pat. No. 5,250,612) 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. 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 homogeneously branched linear ethylene/α-olefin interpolymer having:

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

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

(vi) a slope of strain hardening coefficient greater than or equal to 1.3; 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 heterogeneously branched linear ethylene polymer having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3.

2. The film of claim **1** wherein the homogeneously branched linear ethylene interpolymer has a slope of strain hardening coefficient greater than or equal to 1.5.

3. The film of claim **1** wherein the homogeneously branched linear ethylene/α-olefin interpolymer is an interpolymer of ethylene with at least one C_3–C_{20} α-olefin.

4. The film of claim **1** wherein the homogeneously branched linear ethylene/α-olefin interpolymer is a copolymer of ethylene and a C_3–C_{20} α-olefin.

5. The film of claim **4** wherein the homogeneously branched linear ethylene/α-olefin copolymer is a copolymer of ethylene and 1-octene.

6. 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 homogeneously branched linear ethylene/α-olefin interpolymer having:

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

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

(vi) a slope of strain hardening coefficient greater than or equal to 1.3; 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 heterogeneously branched linear ethylene polymer having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3.

7. The composition of claim **6** wherein the homogeneously branched linear ethylene/α-olefin interpolymer has a slope of strain hardening coefficient greater than or equal to 1.5.

8. The composition of claim **6** wherein the homogeneously branched linear ethylene/α-olefin interpolymer is an interpolymer of ethylene with at least one C_3–C_{20} α-olefin.

9. The composition of claim **6** wherein the homogeneously branched linear ethylene/α-olefin interpolymer is a copolymer of ethylene and a C_3–C_{20} α-olefin.

10. The composition of claim **9** wherein the homogeneously branched linear ethylene/α-olefin interpolymer is a copolymer of ethylene and 1-octene.

11. The composition of claim **6** wherein the heterogeneously branched ethylene polymer is a copolymer of ethylene and a C_3–C_{20} α-olefin.

12. The composition of claim **11** wherein the heterogeneously branched ethylene polymer is a copolymer of ethylene and 1-octene.

13. The film of claim **9** or the composition of claim **6**, wherein the density of the at least one homogeneously branched linear ethylene/α-olefin interpolymer is in the range from about 0.905 g/cm^3 to about 0.925 g/cm^3 and the I_2 melt index is in the range of from about 0.001 g/10 minutes to less than about 1 g/10 minutes.

14. The film of claim **1** wherein the heterogeneously branched linear ethylene polymer is an interpolymer of ethylene with at least one C_3–C_{20} α-olefin.

15. The film of claim **14** wherein the heterogeneously branched linear ethylene polymer is a copolymer of ethylene and a C_3–C_{20} α-olefin.

16. The film of claim **15** wherein the heterogeneously branched linear ethylene polymer is a copolymer of ethylene and 1-octene.

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Case: 14-1431 Document: 24 Page: 121 Filed: 07/24/2014

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United States Patent [19]

Chum et al.

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

Dow v. Nova 05-737 (JJF) (D.Del.)
Dow Trial Exhibit
PTX - 2

[54] **FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS**

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[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

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[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

EXHIBIT
PDX 244
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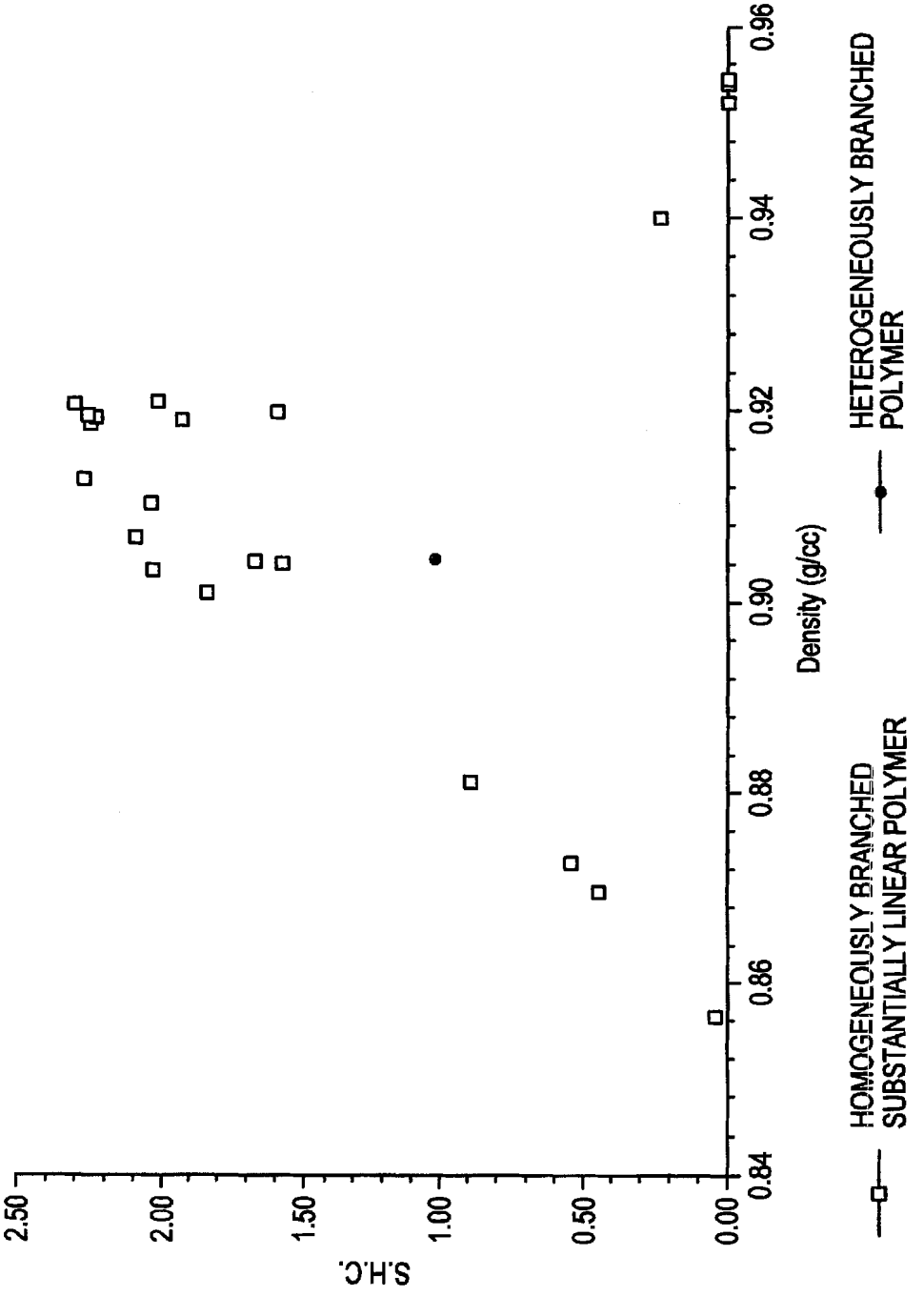
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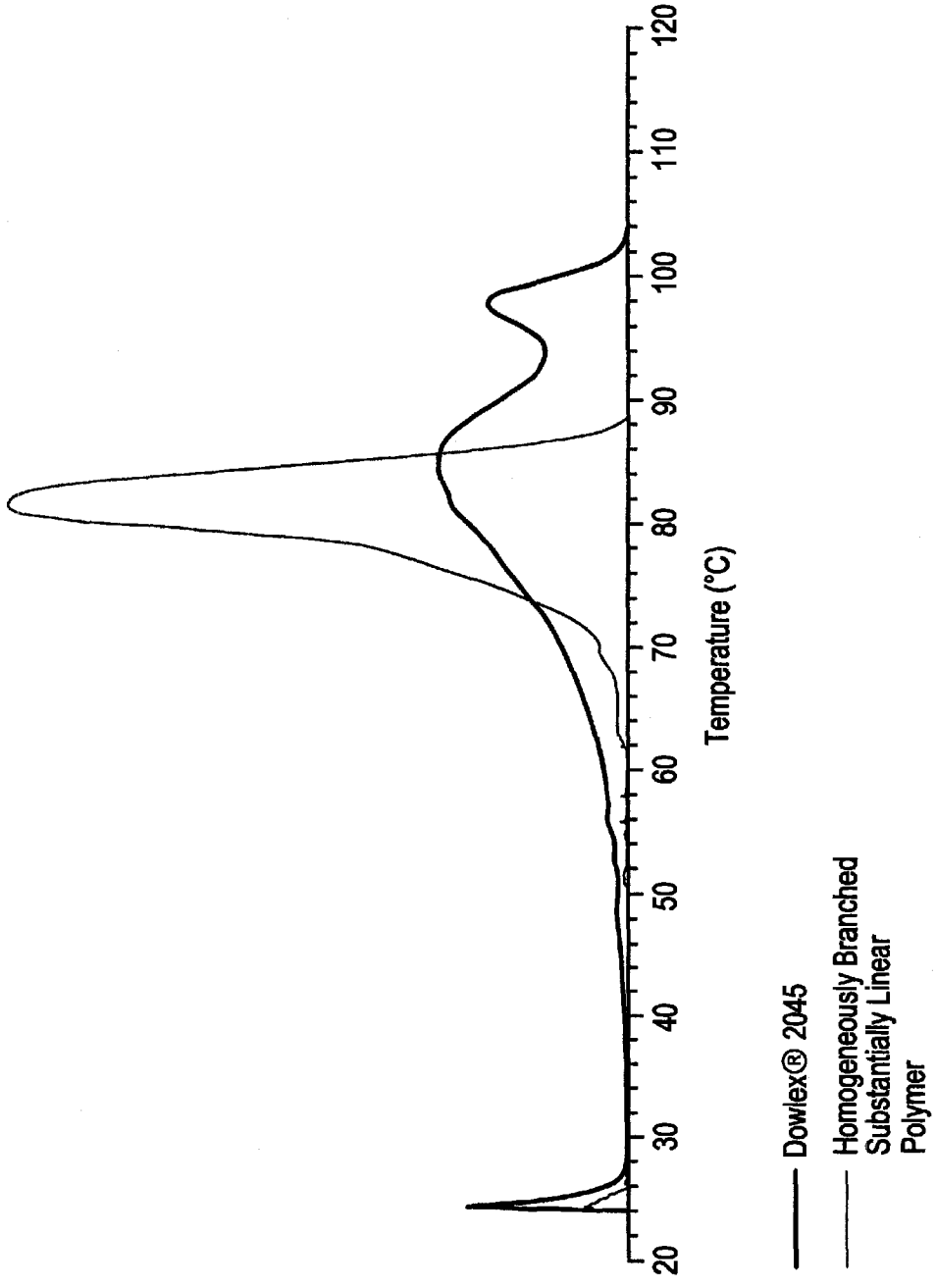
Case: 14-1431 Document: 24 Page: 124 Filed: 07/24/2014

FIG. 1



Case: 14-1431 Document: 24 Page: 125 Filed: 07/24/2014

FIG. 2



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.

Case: 14-1431 Document: 24 Page: 127 Filed: 07/24/2014

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

Case: 14-1431 Document: 24 Page: 128 Filed: 07/24/2014

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

Case: 14-1431 Case: 14-1431 Document: 24 Page: 129 Filed: 07/24/2014

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.

Case: 14-1431 Case: 14-1431 Document: 24 Page: 130 Filed: 07/24/2014
Case: 14-1431 CASE PARTICIPANTS ONLY Document: 23 Page: 130 Filed: 07/24/2014

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

Case: 14-1431 Document: 24 Page: 131 Filed: 07/24/2014

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 Elrnendorf 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.

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.

* * * * *

UNITED STATES PATENT AND TRADEMARK OFFICE

CERTIFICATE OF CORRECTION

PATENT NO. : 6,111,023
APPLICATION NO. : 08/927393
DATED : August 29, 2000
INVENTOR(S) : Pak-Wing S. Chum et al.

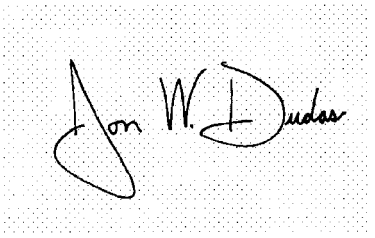
It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

"Related U.S. Application Data"

"(63) This application is a Rule 1.60 continuation application of 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.

Signed and Sealed this

Twenty-ninth Day of August, 2006



JON W. DUDAS
Director of the United States Patent and Trademark Office

CERTIFICATE OF SERVICE

I hereby certify that the foregoing **BRIEF FOR DEFENDANTS-APPELLANTS NOVA CHEMICALS CORPORATION (CANADA) AND NOVA CHEMICALS INC. (DELAWARE)** was filed via operation of the Court's CM/ECF system. Copies of the Confidential brief were served upon registered counsel via electronic means on this 24th day of July, 2014.

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CERTIFICATE OF COMPLIANCE

This brief complies with the type-volume limitation of Federal Rule of Appellate Procedure 32(a)(7)(B). The reply brief contains 13,992 words, excluding the parts of the brief exempted by Federal Rule of Appellate Procedure 32(a)(7)(B)(iii).

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