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(12) # EUROPEAN PATENT APPLICATION

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(54) **Process for preparing a fibril-reinforced polyolefin film and olefin polymer film so prepared.**

(57) A fibril reinforced olefin polymer is made of a web of randomly-dispersed olefin polymer fibrils which are heated under an applied pressure to form a fused polymer film having a small percentage of unmelted fibrils therein. The film has a higher tensile strength than a solvent cast film prepared from the same olefin polymer.

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TITLE MODIFIED
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Fibril-reinforced olefin polymer film

Olefin polymer films, particularly ethylene polymer films, are used in commerce in large volume. By reason of their volume usage, the art is continuously seeking (1) olefin polymer films having improved properties and (2) more economical processes for the manufacture of olefin polymer films.

According to this invention there is provided a process for preparing a fibril-reinforced polyolefin comprising:

(a) forming a web of randomly dispersed olefin polymer fibrils,

(b) heating the web to a temperature at which said fibrils melt,

(c) subjecting the web to pressure throughout step (b) so as to form a compressed melted film, and

(d) cooling the compressed melted film of step (c) to form a self-supporting film;

the olefin polymer included in said fibrils having a weight average molecular weight of at least one million; the duration of heating step (b) being such that a portion of the fibrils are incompletely melted and retain their fibril identity in the fused olefin polymer film.

Preferably the web is laid down from an aqueous dispersion of the olefin polymer fibrils and the web is melted by being passed through heated calender rolls.

The olefin polymer may be a polymer of ethylene or propylene.

The olefin polymer fibrils employed in the practice of the invention constitute a recognized class of materials known and described in the art. Such fibrils are prepared by precipitating an olefin polymer from an organic solvent solution thereof under conditions of high shear. Representative U.S. patents disclosing the preparation of such fibrils include the following:

U.S. 2,933,782	U.S. 3,031,519
U.S. 3,341,242	U.S. 3,740,333

U.S. 3,743,272 U.S. 3,800,091
U.S. 3,885,014 U.S. 3,891,499
U.S. 4,010,229 U.S. 4,013,751

When examined with the naked eye, the olefin polymer fibrils employed in the invention resemble in appearance natural fibres such as cotton and staple fibres of solution spun polymers such as nylon and the like. When examined microscopically, however, fibrils have grossly irregular shapes, including segments which appear to be film-like in nature. See for example Fig. 4 of U.S. 3,891,499. It's believed possible that these particular physical characteristics may play a role in the function of the olefin polymer fibrils in the present invention.

The majority of the olefin polymer fibrils employed in the practice of the invention are small in size, with the largest dimension not exceeding about 10 mm. The fibrils have a large surface to weight ratio of at least about 2 m^2/gram as determined by nitrogen absorption/desorption methods.

The olefin polymer fibrils employed in the invention are prepared from an ethylene or propylene polymer having a weight average molecular weight of at least about 1 million and preferably at least about 1.5 million. Such polymers will have an intrinsic viscosity of at least 3.5, preferably at least 5.0, and most especially at least 10.0. A listing of suitable olefin polymers and olefin polymer mixtures is set forth in U.S. 4,013,751, which description is incorporated herein by reference.

The web of randomly-dispersed olefin polymer fibrils employed in the invention can be prepared by numerous techniques known and reported in the art. The preferred method is to prepare a water-laid web by the techniques known and used in the paper making arts.

The fused film is prepared by heating the web to a temperature above the melting point of the olefin polymer

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and subjecting the web to pressure to compress the melted polymer to form a fused film. The pressure required and the duration of the required heating cycle will depend upon a multiplicity of factors including the thickness and bulk density of the web, the melting point of the olefin polymer, and the heat transfer characteristics of the press surfaces. The proper combination of temperature and pressure to be employed for a given web in a particular press can be readily established through a few routine experiments.

In pressing a given web at a fixed temperature and a fixed pressure, it is noted that the fibrils at the surface of the web melt first. With the passage of time, the fibrils in the interior of the web melt. If the heating is continued for a sufficient period of time, all of the fibrils melt and a homogeneous fused film is obtained. At intermediate points in the heating cycle, it is noted that the film, while fused, will contain thin fibre-like sections of unmelted fibrils. Film containing such a structure has a tensile strength greater than solvent cast film prepared from the olefin polymer from which the fibrils were prepared. Accordingly, the temperature, pressure and cycle time should be controlled so that the fused film contains some incompletely melted fibrils.

A desirable feature of the invention is that the fused film can be prepared at high rates of speed at low cost in a continuous process. The random web of fibrils is laid down continuously on a paper making machine. The web then is passed over drying rolls and passed through a stack of heated calender rolls.

The following example is set forth to illustrate more clearly the principles and practice of this invention to those skilled in the art. Where parts or percentages are referred to, they are parts or percentages by weight unless otherwise noted.

Example 1

Part A A lot of fibrils was prepared following the procedure of Example 1 of U.S. 4,013,751. The ethylene polymer from which the fibrils were prepared had a weight average molecular weight of about 1,500,000. The hydrocarbon-wet fibrils were refined with 91% isopropanol to displace the hydrocarbon. The fibrils then were filtered and pressed to expel the maximum quantity of isopropanol.

Part B Water-laid sheets were prepared from the fibrils of Part A employing an experimental size paper making machine. The dried sheets had a basis weight of about 60 lbs. per ream. An 8" x 8" sheet was pressed for 5 minutes in a platen press under a pressure of 40,000 psig at a plate temperature of 300°F (149°C). The resulting film was well fused and generally transparent. When viewed in strong light, a few fine fibres were noted in the film. The film had a tensile strength of 4200 psi.

Part C A hydrocarbon solution was prepared from the ethylene polymer employed in Part A and a film was cast therefrom. The film had a tensile strength of 2900 psi.

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Claims:

1. A process for preparing a fibril-reinforced polyolefin comprising:

(a) forming a web of randomly dispersed olefin polymer fibrils,

(b) heating the web to a temperature at which said fibrils melt,

(c) subjecting the web to pressure throughout step (b) so as to form a compressed melted film, and

(d) cooling the compressed melted film of step (c) to form a self-supporting film;

the olefin polymer included in said fibrils having a weight average molecular weight of at least one million; the duration of heating step (b) being such that a portion of the fibrils are incompletely melted and retain their fibril identity in the fused olefin polymer film.

2. A process according to claim 1, in which the web is laid down from a aqueous dispersion of the olefin polymer fibrils and the web is melted by being passed through heated calender rolls.

3. A process according to claim 1 or claim 2, in which the olefin polymer is an ethylene polymer.

4. A process according to claim 1 or claim 2, in which the olefin polymer is a propylene polymer.

5. A process according to claim 1, substantially as hereinbefore described.

6. An olefin polymer film whenever prepared by the process claimed in any of claims 1 to 5.

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EUROPEAN SEARCH REPORT

Application number
EP 79 30 1268

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	FR - A - 2 128 750 (CROWN ZELLERBACH) * Page 1, lines 36-40; page 2, lines 1-26; page 3, lines 19-29; page 5, lines 11-14 and 27-30; example 2; page 11, lines 16-23; claims 1,2,3,7,9 * -- FR - A - 2 265 806 (MONTEDISON) * Page 2, lines 13-35; page 4, lines 19-23; example 1; claims 1,4,6 * -- FR - A - 2 173 928 (GULF) * Page 1, lines 1-9; claim 1 * ----	1-6 1-6 1-6	D 21 H 5/20 C 08 J 5/18
			TECHNICAL FIELDS SEARCHED (Int.Cl.3)
			D 21 H 5/20

CATEGORY OF CITED DOCUMENTS

X: particularly relevant
A: technological background
O: non-written disclosure
P: intermediate document
T: theory or principle underlying the invention
E: conflicting application
D: document cited in the application
L: citation for other reasons

&: member of the same patent family, corresponding document

X The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
The Hague	05-12-1979	Mme. ELSEN-DROUOT

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