



(72) MEDOFF, MARSHALL, US

(72) LAGACE, ARTHUR, US

(71) XYLECO, INC., US

(51) Int.Cl.<sup>7</sup> B32B 9/04

(30) 1997/09/02 (08/921,807) US

(54) **COMPOSITES DE PAPIER MULTICOUCHE**

(54) **POLY-COATED PAPER COMPOSITES**

(57) La présente invention concerne des composites résine et papier multicouché. L'invention concerne également des procédés permettant l'élaboration de ces composites.

(57) Composites of poly-coated paper and a resin, and methods for forming the composites, are disclosed.

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification <sup>6</sup> : <b>B32B 9/04</b></p>	<p><b>A1</b></p>	<p>(11) International Publication Number: <b>WO 99/11453</b></p> <p>(43) International Publication Date: 11 March 1999 (11.03.99)</p>
<p>(21) International Application Number: PCT/US98/17753</p> <p>(22) International Filing Date: 27 August 1998 (27.08.98)</p> <p>(30) Priority Data: 08/921,807                      2 September 1997 (02.09.97)                      US</p> <p>(71) Applicant (for all designated States except US): XYLECO, INC. [US/US]; 90 Addington Road, Brookline, MA 02146 (US).</p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): MEDOFF, Marshall [US/US]; 90 Addington Road, Brookline, MA 02146 (US). LAGACE, Arthur [US/US]; 21 Prospect Park, Newtonville, MA 02146 (US).</p> <p>(74) Agent: NABINGER, Robert, C.; Fish &amp; Richardson P.C., 225 Franklin Street, Boston, MA 02110-2804 (US).</p>		<p>(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p><b>Published</b> <i>With international search report.</i></p>
<p>(54) Title: POLY-COATED PAPER COMPOSITES</p>		
<p>(57) Abstract</p> <p>Composites of poly-coated paper and a resin, and methods for forming the composites, are disclosed.</p>		

- 1 -

POLY-COATED PAPER COMPOSITESBackground of the Invention

The invention relates to composites of poly-coated paper and a resin.

5 Paper coated with a polymer (poly-coated paper) is used in a number applications. For example, poly-coated paper is used to make a variety of food containers, including individual-serving size juice cartons and boxes for frozen foods.

10 Resins are also used in a variety of applications, for example, in food packaging. Food containers made of poly-coated paper or resins are typically used once, then discarded. As a result, there is an ever-increasing amount of waste poly-coated paper and resins.

15 Summary of the Invention

In general, the invention features composites of texturized poly-coated paper and a resin.

The invention features a composite including at least about 2% by weight, more preferably at least about  
20 5% by weight, texturized poly-coated paper, and a resin, such as a thermoplastic resin. The composite may also include cellulosic or lignocellulosic fiber.

The invention also features a composite that includes polyethylene and at least about 50% by weight  
25 texturized poly-coated paper.

The invention further features composites, including poly-coated paper and resin, that have flexural strengths of at least about 3,000 psi, or tensile strengths of at least about 3,000 psi.

30 In addition, the invention features a process for manufacturing a composite; the process includes shearing poly-coated paper to form texturized poly-coated paper, then combining the texturized poly-coated paper with a resin. A preferred method includes shearing the poly-

- 2 -

coated paper with a rotary knife cutter. The invention also features a process for manufacturing a composite; the process includes shearing poly-coated paper and combining the poly-coated paper with a resin.

5           The term "texturized poly-coated paper" as used herein, means that the paper has been sheared to the extent that the internal fibers are substantially exposed. At least about 50%, more preferably at least about 70%, of these fibers, as well as the external  
10 polymer fibers, have a length/diameter (L/D) ratio of at least 10, more preferably at least 25, or at least 50. An example of texturized poly-coated paper is shown in Fig. 1.

          The composites of the present invention are  
15 strong, light-weight, and inexpensive. The raw materials used to make the composites are readily available; for example, they may include discarded containers composed of resins, and discarded containers composed of poly-coated paper.

20           Poly-coated paper can be difficult to recycle because for many applications, the paper and the polymer layers must be separated. In the present invention, both the paper and the polymer portions are utilized, so there is no need to separate the two. The invention thus helps  
25 to recycle discarded post-consumer containers, while at the same time producing useful materials.

          Other features and advantages of the invention will be apparent from the description of the preferred embodiments thereof, and from the claims.

### 30           Brief Description of the Drawing

          Fig. 1 is a photograph of an embodiment of texturized poly-coated paper, magnified 50 times.

### Description of the Preferred Embodiments

- 3 -

A preferred composite includes texturized poly-coated paper and a resin.

The texturized poly-coated paper provides the composite with strength. The composite may include from 5 about 30% to about 90%, more preferably from about 50% to about 70%, of the texturized poly-coated paper by weight. Examples of poly-coated paper include materials having layers of polymer and paper, and materials having layers of polymer, paper, and aluminum. A preferred poly-coated 10 paper is one having layers of polymer and paper.

Poly-coated paper is available in a variety of forms. For example, whole sheets of virgin poly-coated paper can be purchased from International Paper, New York. Alternatively, scraps of poly-coated paper can be 15 obtained from International Paper or other paper manufacturers. Used poly-coated paper, in the form of discarded food and beverage containers, can be gathered from various sources, including refuse bins. Used poly-coated paper can also be purchased from brokers of this 20 material. If used poly-coated paper is included in the composites, it should be thoroughly washed before it is used. Scrap poly-coated paper is preferred, as it is less expensive than full sheets.

The resins encapsulate the texturized poly-coated 25 paper and help control the shape of the composite. The resins also transfer the external loads to the poly-coated paper and protect the poly-coated paper from environmental and structural damage. Preferred composites include about 20% to about 60%, more 30 preferably about 30% to about 50%, by weight of the resins.

Examples of resins include polyethylene (including, e.g., low density polyethylene and high density polyethylene), polypropylene, polystyrene, 35 polycarbonate, polybutylene, thermoplastic polyesters,

- 4 -

polyethers, thermoplastic polyurethane, PVC, Nylon, and other resins. It is preferred that the resins have a low melt flow index. Preferred resins include polyethylene and polypropylene with melt flow indices of less than 3  
5 g/10 min, and more preferably less than 1 g/10 min.

The resins may be purchased as virgin material, or obtained as scrap or waste materials, and are usually purchased in pelletized form. Preferably, the resins are obtained as scrap or waste resins, as these materials are  
10 less expensive. A preferred source of resin is used polyethylene milk bottles.

The composites also include coupling agents. The coupling agents help to bond the hydrophilic fibers of the poly-coated paper to the hydrophobic resins.  
15 Examples of coupling agents include maleic anhydride modified polyethylenes, such those in the FUSABOND® (available from Dupont, Delaware) and POLYBOND® (available from Uniroyal Chemical, Connecticut) series. A preferred coupling agent is a maleic anhydride modified  
20 high density polyethylene such as FUSABOND® MB 100D.

The composites can also include cellulosic or lignocellulosic fibers. These fibers provide extra strength to the composite. The quantity of fiber which is incorporated into the composites can vary, depending  
25 on the desired physical and mechanical properties of the finished products. Preferred composites contain about 5% to about 50%, more preferably about 10% to about 30%, by weight of the cellulosic or lignocellulosic fiber. Examples of such fibers include paper and paper products,  
30 wood, wood fibers, and wood-related materials, as well as materials derived from kenaf, grasses, rice hulls, bagasse, cotton, jute, and other cellulosic or lignocellulosic materials. Preferred fibers include jute and kenaf. The fibers may have, for example, a L/D ratio  
35 of at least 10, or at least 25 or 50.

- 5 -

The composites can also contain additives known to those in the art of compounding, such as plasticizers, lubricants, antioxidants, opacifiers, heat stabilizers, colorants, impact modifiers, photostabilizers, flame retardants, biocides, and antistatic agents.

#### Preparation of starting materials

If scrap poly-coated paper is used, it should be cleaned and dried. The poly-coated paper must then be texturized before it is combined with the resin. The poly-coated paper can be texturized using any one of a number of mechanical means, or combinations thereof. During the texturizing process, the polymer layers are sheared away from the paper layers, thus exposing the paper fibers. A preferred method of texturizing includes first cutting the poly-coated paper into 1/4- to 1/2-inch pieces using a standard paper-cutting apparatus. These pieces are then sheared with a rotary cutter, such as the one (available from Sprout, Waldron Companies) described in Perry's Chem. Eng. Handbook, 6th Ed., at 8-29 (1984). The texturized material is then passed through a 2 mm mesh screen. The texturized poly-coated paper can be stored in sealed bags. It should be dried at approximately 105°C for 4-18 hours (until the moisture content is less than about 0.5%) immediately before use. The Figure is an SEM photograph of the texturized poly-coated paper.

The resin may purchased in a pelletized or granulated form and used without further purification or drying. If surface moisture is present on the pelletized or granulated resin, however, it should be dried before use.

If cellulosic or lignocellulosic fibers are used, they can be texturized, using the process described above.

- 6 -

Preparation of composites

The composites can be prepared as follows. A standard rubber/plastic compounding 2-roll mill is heated to 325-400°C. The resin (usually in the form of pellets or granules) is added to the heated roll mill. After about 10 minutes, the coupling agent is added to the roll mill. After another five minutes, the texturized poly-coated paper is added to the molten resin/coupling agent mixture. The texturized poly-coated paper is added over a period of about 10 minutes.

The composite is removed from the roll mill, cut into sheets and allowed to cool to room temperature. It is then compression molded into plaques using standard compression molding techniques.

Alternatively, a mixer, such as a Banbury internal mixer, is charged with the ingredients. The ingredients are mixed, while the temperature is maintained at less than about 190°C. The mixture can then be compression molded.

In another embodiment, the ingredients can be mixed in an extruder mixer, such as a MARIS (Turin) TM 85 extruder equipped with co-rotating screws. The resin and the coupling agent are introduced at the extruder feed throat; the poly-coated paper (and cellulosic or lignocellulosic fiber, if used) are introduced about 1/3 of the way down the length of the extruder into the molten resin. The internal temperature of the extruder is maintained at less than about 190°C. At the output, the composite is pelletized by cold strand cutting.

Alternatively, the mixture can first be prepared in a mixer, then transferred to an extruder for the extrusion and pellet-cutting steps.

In another embodiment, the composite can be formed into filaments for knitting, warping, weaving, and

- 7 -

braiding, and to make non-wovens. In a further embodiment, the composite can be made into film.

#### Properties of the composite

The resulting composites include a network of  
5 fibers, encapsulated within a resin matrix. The exposed  
fibers form a lattice network, which provides the  
composite with strength. Since the poly-coated paper is  
texturized, the amount of surface area available to bond  
to the resin is increased, in comparison to composites  
10 prepared with un-texturized poly-coated paper. The resin  
binds to the surfaces of the exposed fibers, creating an  
intimate blend of the fiber network and the resin matrix.  
The intimate blending of the fibers and the resin matrix  
further strengthens the composites. Cellulosic or  
15 lignocellulosic fibers may also be added to strengthen  
the composite further.

#### Uses

The poly-coated paper/resin composites can be used  
in a number of applications. The composites are strong  
20 and light weight; they can be used, for example, as wood  
substitutes. The resin coating renders the composites  
water-resistant, so they may be used in outdoor  
applications. For example, the composites may be used to  
make pallets which are stored outdoors for extended  
25 periods of time. The composite also may be used, for  
example, as the base or carcass for a veneer product.  
Moreover, the composites when found can be surface  
treated, grooved, milled, shaped, imprinted, textured,  
compressed, punched, colored, etc. The surface may be  
30 made smooth or roughed.

#### Examples

The following examples were prepared as follows.  
A standard rubber/plastic compounding 2-roll mill was

- 8 -

heated to 325-400°C. The resin (usually in the form of pellets or granules) was added to the heated roll mill. After about 10 minutes, the resin banded on the rolls (i.e., it melted and fused to the rolls). The coupling agent was then added to the roll mill. After another five minutes, the poly-coated paper was added to the molten resin/coupling agent mixture. The poly-coated paper was added over a period of about 10 minutes.

The composite was then removed from the roll mill, cut into sheets, and allowed to cool to room temperature. Batches of about 80 g each were compression molded into 6" x 6" x 1/8" plaques using standard compression molding techniques.

One composition contains the following ingredients:

Composition No. 1

<u>Ingredient</u>	<u>Amount (g)</u>
High density polyethylene <sup>1</sup>	160
Poly-coated paper <sup>2</sup>	240
Coupling agent <sup>3</sup>	8

<sup>1</sup> Marlex 6007, melt flow index 0.65 g/10 min, commercially available from Phillips

<sup>2</sup> Texturized using rotary cutter with 2 mm mesh

<sup>3</sup> POLYBOND® 3009, commercially available from Uniroyal

Chemical

The plaques were machined into appropriate test specimens and tested according to the procedures outlined in the method specified. Three different specimens were tested for each property, and the mean value for each test was calculated.

The properties of Composition No. 1 are as follows:

Tensile modulus (10 <sup>5</sup> psi)	8.63 (ASTM D638)
Tensile strength at break (psi)	6820 (ASTM D638)
Ultimate elongation (%)	<5 (ASTM D638)

WO 99/11453

PCT/US98/17753

- 9 -

Flexural Strength (psi)	12,200 (ASTM D790)
Flexural modulus (10 <sup>5</sup> psi)	6.61 (ASTM D790)

Another composition contains the following ingredients:

5 Composition No. 2

<u>Ingredient</u>	<u>Amount (g)</u>
High density polyethylene <sup>1</sup>	160
Poly-coated paper <sup>2</sup>	240
Coupling agent <sup>3</sup>	8

10 <sup>1</sup> Scrapped milk jugs, melt flow index approximately 0.8 g/10 min

<sup>2</sup> Texturized using rotary cutter with 2 mm mesh

<sup>3</sup> POLYBOND® 3009

The properties of Composition No. 2 are as follows:

15 Tensile modulus (10 <sup>5</sup> psi)	7.38 (ASTM D638)
Tensile strength at break (psi)	6500 (ASTM D638)
Ultimate elongation (%)	<5 (ASTM D638)
Flexural Strength (psi)	11,900 (ASTM D790)
20 Flexural modulus (10 <sup>5</sup> psi)	6.50 (ASTM D790)

A third composition is as follows:

Composition No. 3

<u>Ingredient</u>	<u>Amount (g)</u>
High density polyethylene <sup>1</sup>	160
25 Poly-coated paper <sup>2</sup>	240
Coupling agent <sup>3</sup>	8

<sup>1</sup> Scrap milk jugs, melt flow index approximately 0.8 g/10 min

<sup>2</sup> Texturized using rotary cutter with 2 mm mesh

30 <sup>3</sup> FUSABOND® MB 100D, commercially available from DuPont

The properties of Composition No. 3 are as follows:

Tensile modulus (10 <sup>5</sup> psi)	7.08 (ASTM D638)
---------------------------------------	------------------

WO 99/11453

PCT/US98/17753

- 10 -

Tensile strength at break (psi)	6480 (ASTM D638)
Ultimate elongation (%)	<5 (ASTM D638)
Flexural Strength (psi)	10,200 (ASTM D790)
Flexural modulus ( $10^5$ psi)	5.73 (ASTM D790)

5 A fourth composition contains the following ingredients:

Composition No. 4

<u>Ingredient</u>	<u>Amount (g)</u>
High density polyethylene <sup>1</sup>	160
10 Poly-coated paper <sup>2</sup>	240
Coupling agent <sup>3</sup>	8

<sup>1</sup> Marlex 6007, melt flow index 0.65 g/10 min

<sup>2</sup> texturized using rotary cutter with 2 mm mesh

<sup>3</sup> FUSABOND<sup>®</sup> MB 100D

15 The properties of Composition No. 4 are as follows:

Tensile modulus ( $10^5$ psi)	7.17 (ASTM D638)
Tensile strength at break (psi)	6860 (ASTM D638)
Ultimate elongation (%)	<5 (ASTM D638)
20 Flexural Strength (psi)	12,200 (ASTM D790)
Flexural modulus ( $10^5$ psi)	7.50 (ASTM D790)

Other embodiments are within the claims.

What is claimed is:

- 11 -

Claims

1. A composite comprising poly-coated paper and a resin, wherein at least about 2% by weight of the poly-coated paper is texturized.
- 5       2. The composite of claim 1, wherein at least about 5% by weight of the poly-coated paper is texturized.
3. The composite of claim 1, wherein the poly-coated paper comprises polyethylene and paper.
- 10       4. The composite of claim 3, wherein the poly-coated paper further comprises aluminum.
5. The composite of claim 1, wherein the resin is a thermoplastic resin.
6. The composite of claim 5, wherein the  
15 thermoplastic resin is polyethylene.
7. The composite of claim 5, wherein the thermoplastic resin is polypropylene.
8. The composite of claim 1, wherein the  
20 composite comprises about 50% to about 70% by weight poly-coated paper and about 30% to about 50% by weight resin.
9. The composite of claim 1, wherein the composite further comprises lignocellulosic fiber.
- 25       10. The composite of claim 1, wherein the composite further comprises cellulosic fiber.

- 12 -

11. A composite comprising poly-coated paper and polyethylene, wherein at least about 50% by weight of the poly-coated paper is texturized.

12. A composite comprising poly-coated paper and  
5 a resin, wherein the composite has a flexural strength of at least 3000 psi.

13. The composite of claim 12, wherein the composite has a flexural strength of at least 6000 psi.

14. The composite of claim 12, wherein the  
10 composite has a flexural strength of at least 10,000 psi.

15. A composite comprising poly-coated paper and a resin, wherein the composite has a tensile strength of at least about 3,000 psi.

16. The composite of claim 15, wherein the  
15 composite has a tensile strength of at least 5,000 psi.

17. The composite of claim 15, wherein the composite has a tensile strength of at least 6,000 psi.

18. A process for manufacturing a composite comprising shearing poly-coated paper to form texturized  
20 poly-coated paper, and combining the texturized poly-coated paper with a resin.

19. The process of claim 18, wherein the resin is a thermoplastic resin.

20. The process of claim 18, wherein the step of  
25 shearing the poly-coated paper comprises shearing with a rotary knife cutter.

- 13 -

21. A process for manufacturing a composite, the process comprising shearing poly-coated paper and combining the poly-coated paper with a resin.



**FIG. 1**