

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

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
28 December 2000 (28.12.2000)

PCT

(10) International Publication Number  
**WO 00/78127 A1**

(51) International Patent Classification<sup>7</sup>: **A01G 9/02**,  
13/00, A01N 25/00, 25/34, A23G 3/00, A61K 38/43, C08J  
9/00, C08K 5/13, C08L 9/00, C09K 11/00, D02G 3/00

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(21) International Application Number: PCT/US00/17232

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(22) International Filing Date: 22 June 2000 (22.06.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:  
09/337,580 22 June 1999 (22.06.1999) US  
09/338,209 22 June 1999 (22.06.1999) US

(81) Designated States (*national*): AE, AG, AL, AM, AT, AU,  
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ,  
DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR,  
HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR,  
LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ,  
NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM,  
TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(63) Related by continuation (CON) or continuation-in-part  
(CIP) to earlier applications:

US 09/337,580 (CIP)  
Filed on 22 June 1999 (22.06.1999)  
US 09/338,209 (CIP)  
Filed on 22 June 1999 (22.06.1999)

(84) Designated States (*regional*): ARIPO patent (GH, GM,  
KE, LS, MW, MZ, SD, SL, SZ, TZ, 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).

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Published:  
— With international search report.

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For two-letter codes and other abbreviations, refer to the "Guid-  
ance Notes on Codes and Abbreviations" appearing at the begin-  
ning of each regular issue of the PCT Gazette.

(54) Title: TEXTURIZED CELLULOSIC AND LIGNOCELLULOSIC MATERIALS AND COMPOSITIONS AND COMPOS-  
ITES MADE THEREFROM

(57) Abstract: Texturized cellulosic or lignocellulosic materials (e.g., texturized polycoated paper), and compositions and compos-  
ites made therefrom, are disclosed.

WO 00/78127 A1

TEXTURIZED CELLULOSIC AND LIGNOCELLULOSIC MATERIALS  
AND COMPOSITIONS AND COMPOSITES MADE THEREFROM

Cross Reference to Related Applications

This application claims priority from USSN 09/337,580, filed 22 June 1999,  
5 entitled, "Texturized Cellulosic and Lignocellulosic Materials and Compositions and  
Composites Made Therefrom," which is a continuation-in-part of USSN 08/961,863,  
filed 31 October 1997, entitled, "Cellulosic Fiber Composites," and from USSN  
09/338,209, filed 22 June 1999, entitled, "Texturized Fibrous Materials from Poly-  
Coated Paper and Compositions and Composites Made Therefrom," which is a  
10 continuation-in-part of USSN 08/921,807, filed 2 September 1997, entitled, "Poly-  
Coated Paper Composites."

Background of the Invention

The invention relates to texturized cellulosic or lignocellulosic materials (e.g.,  
15 texturized poly-coated paper) and compositions and composites made from such  
texturized materials.

Cellulosic and lignocellulosic materials are produced, processed, and used in  
large quantities in a number of applications. For example, paper coated with a polymer  
(i.e., poly-coated paper) is used to make a variety of food containers, including  
20 individual-serving size juice cartons and boxes for frozen foods. Once used, these  
cellulosic and lignocellulosic materials are usually discarded. As a result, there is an  
ever-increasing amount of waste cellulosic and lignocellulosic material.

Summary of the Invention

25 In general, the invention features texturized cellulosic or lignocellulosic  
materials (e.g., texturized poly-coated paper) and compositions and composites made  
therefrom.

In one embodiment, the invention features a process for preparing a texturized  
fibrous material. The process involves shearing a cellulosic or lignocellulosic material  
30 having internal fibers (e.g., flax, hemp, cotton, jute, rags, finished or unfinished paper,  
poly-coated paper, paper products, or byproducts of paper manufacturing such as pulp  
board, or synthetic cellulosic or lignocellulosic materials such as rayon), to the extent  
that the internal fibers are substantially exposed, resulting in texturized fibrous material.

The cellulosic or lignocellulosic material can be, for example, a woven material such as a woven fabric, or a non-woven material such as paper or bathroom tissue. The exposed fibers of the texturized fibrous material can have a length/diameter (L/D) ratio of at least about 5 (e.g., at least about 5, 10, 25, 50, or more). For example, at least  
5 about 50% of the fibers can have L/D ratios of this magnitude.

In another embodiment, the invention features a texturized fibrous material that includes a cellulosic or lignocellulosic material (e.g., poly-coated paper) having internal fibers, where the cellulosic or lignocellulosic material is sheared to the extent that the internal fibers are substantially exposed.

10 The texturized fibrous material can, for example, be incorporated into (e.g., associated with, blended with, adjacent to, surrounded by, or within) a structure or carrier (e.g., a netting, a membrane, a flotation device, a bag, a shell, or a biodegradable substance). Optionally, the structure or carrier may itself be made from a texturized fibrous material (e.g., a texturized fibrous material of the invention), or of a  
15 composition or composite of a texturized fibrous material.

The texturized fibrous material can have a bulk density less than about 0.5 grams per cubic centimeter ( $\text{g}/\text{cm}^3$ ), or even less than about  $0.2 \text{ g}/\text{cm}^3$ .

Compositions that include the texturized fibrous materials described above, together with a chemical or chemical formulation (e.g., a pharmaceutical such as an  
20 antibiotic or contraceptive, optionally with an excipient; an agricultural compound such as a fertilizer, herbicide, or pesticide; or a formulation that includes enzymes) are also within the scope of the invention, as are compositions that include the texturized fibrous materials and other liquid or solid ingredients (e.g., particulate, powdered, or granulated solids such as plant seed, foodstuffs, or bacteria).

25 Composites that include thermoplastic resin and the texturized fibrous materials are also contemplated. The resin can be, for example, polyethylene, polypropylene, polystyrene, polycarbonate, polybutylene, a thermoplastic polyester, a polyether, a thermoplastic polyurethane, polyvinylchloride, or a polyamide, or a combination of two or more resins.

30 In some cases, at least about 5% by weight (e.g., 5%, 10%, 25%, 50%, 75%, 90%, 95%, 99%, or about 100%) of the fibrous material included in the composites is texturized.

The composite may include, for example, about 30% to about 70% by weight resin and about 30% to about 70% by weight texturized fibrous material, although proportions outside of these ranges may also be used. The composites can be quite strong, in some cases having a flexural strength of at least about 6,000 to 10,000 psi.

5 The composites can also include inorganic additives such as calcium carbonate, graphite, asbestos, wollastonite, mica, glass, fiber glass, chalk, talc, silica, ceramic, ground construction waste, tire rubber powder, carbon fibers, or metal fibers (e.g., stainless steel or aluminum).

10 The inorganic additives can represent about 0.5% to about 20% of the total weight of the composite.

The composite can be in the form of, for example, a pallet (e.g., an injection molded pallet), pipes, panels, decking materials, boards, housings, sheets, poles, straps, fencing, members, doors, shutters, awnings, shades, signs, frames, window casings, backboards, wallboards, flooring, tiles, railroad ties, forms, trays, tool handles, stalls,  
15 bedding, dispensers, staves, films, wraps, totes, barrels, boxes, packing materials, baskets, straps, slips, racks, casings, binders, dividers, walls, indoor and outdoor carpets, rugs, wovens, and mats, frames, bookcases, sculptures, chairs, tables, desks, art, toys, games, wharves, piers, boats, masts, pollution control products, septic tanks, automotive panels, substrates, computer housings, above- and below-ground electrical casings, furniture, picnic tables, tents, playgrounds, benches, shelters, sporting goods,  
20 beds, bedpans, thread, filament, cloth, plaques, trays, hangers, servers, pools, insulation, caskets, bookcovers, clothes, canes, crutches, and other construction, agricultural, material handling, transportation, automotive, industrial, environmental, naval, electrical, electronic, recreational, medical, textile, and consumer products. The  
25 composites can also be in the form of a fiber, filament, or film.

The terms "texturized cellulosic or lignocellulosic material" and "texturized fibrous material" as used herein, mean that the cellulosic or lignocellulosic material has been sheared to the extent that its internal fibers are substantially exposed. At least about 50%, more preferably at least about 70%, of these fibers have a length/diameter  
30 (L/D) ratio of at least 5, more preferably at least 25, or at least 50. An example of texturized cellulosic material (i.e., texturized newsprint) is shown in Fig. 1. An example of texturized poly-coated paper is shown in Fig. 2.

The texturized fibrous materials of the invention have properties that render them useful for various applications. For example, the texturized fibrous materials have absorbent properties, which can be exploited, for example, for pollution control. The fibers are generally biodegradable, making them suitable, for example, for drug or  
5 chemical delivery (e.g., in the treatment of humans, animals, or in agricultural applications). The texturized fibrous materials can also be used to reinforce polymeric resins.

Those composites that include texturized fibrous material and resin are strong, light-weight, and inexpensive. The raw materials used to make the composites are  
10 available as virgin or recycled materials; for example, they may include discarded containers composed of resins, and waste cellulosic or lignocellulosic fiber (e.g., discarded containers composed of poly-coated paper).

Poly-coated paper can be difficult to recycle because the paper and the polymer layers generally cannot be separated. In the present invention, both the paper and the  
15 polymer portions are utilized, so there is no need to separate the two. Poly-coated paper including one or more layers of aluminum can similarly be used. The invention thus helps to recycle discarded post-consumer containers, while at the same time producing useful products.

Other features and advantages of the invention will be apparent from the  
20 following detailed description, and from the claims.

#### Brief Description of the Drawing

Fig. 1 is a photograph of a texturized newspaper, magnified fifty times.

Fig. 2 is a photograph of texturized poly-coated paper, magnified fifty times.

25 Fig. 3 is a photograph of a half-gallon polyboard juice carton.

Fig. 4 is a photograph of shredded half-gallon polyboard juice cartons.

Fig. 5 is a photograph of texturized fibrous material prepared by shearing the shredded half-gallon polyboard juice cartons of Fig. 4.

#### Detailed Description of the Invention

30 Examples of cellulosic raw materials include paper and paper products such as newsprint and effluent from paper manufacture, and poly-coated paper. Examples of

lignocellulosic raw materials include wood, wood fibers, and wood-related materials, as well as materials derived from kenaf, grasses, rice hulls, bagasse, cotton, jute, other stem plants (e.g., hemp, flax, bamboo; both bast and core fibers), leaf plants (e.g., sisal, abaca), and agricultural fibers (e.g., cereal straw, corn cobs, rice hulls, and coconut hair). Aside from virgin raw materials, post-consumer, industrial (e.g., offal), and processing waste (e.g., effluent) can also be used as fiber sources.

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, virgin waste poly-coated paper (e.g., edge trimmings, surplus, misprinted stock) can be 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 waste and recycling streams. Poly-coated paper can be made of a polymer (e.g., polyethylene) and paper, and, in some cases, one or more layers of aluminum. Poly-coated paper that includes one or more layers of aluminum foil is commonly used for airtight liquid storage. Used, waste, or scrap poly-coated paper (e.g., post-consumer waste, industrial offal) can also be purchased from brokers of this material.

#### Preparation of Texturized Fibrous Material

If scrap cellulosic or lignocellulosic materials are used, they should be clean and dry. The raw material can be texturized using any one of a number of mechanical means, or combinations thereof. One method of texturizing includes first cutting the cellulosic or lignocellulosic material into 1/4- to 1/2-inch pieces, if necessary, using a standard cutting apparatus. Counter-rotating screw shredders and segmented rotating screw shredders such as those manufactured by Munson (Utica, NY) can also be used, as can a standard document shredder as found in many offices.

The cellulosic or lignocellulosic material is then sheared with a rotary cutter, such as the one manufactured by Sprout, Waldron Companies, as described in Perry's Chem. Eng. Handbook, 6th Ed., at 8-29 (1984). Although other settings can be used, the spacing between the rotating knives and bed knives of the rotary cutter is typically set to 0.020" or less, and blade rotation is set to 750 rpm or more. The rotary cutter can be cooled to 100°C or lower during the process, for example, using a water jacket.

The texturized material is passed through a discharge screen. Larger screens (e.g., up to 6 mm) can be used in large-scale production. The cellulosic or lignocellulosic feedstock is generally kept in contact with the blades of the rotary cutter until the fibers are pulled apart; smaller screens (e.g., 2 mm mesh) provide longer residence times and more complete texturization, but can result in lower length/diameter (L/D) aspect ratios. A vacuum drawer can be attached to the screen to maximize and maintain fiber length/diameter aspect ratio.

The texturized fibrous materials can be directly stored in sealed bags or may be dried at approximately 105°C for 4-18 hours (e.g., until the moisture content is less than about 0.5%) immediately before use. Fig. 1 is an SEM photograph of texturized newspaper.

#### Uses of Texturized Fibrous Material

Texturized fibrous materials and compositions and composites of such fibers with other chemicals and chemical formulations can be prepared to take advantage of the material's properties. The materials can be used to absorb chemicals, for example, potentially absorbing many times their own weight. Thus, the materials could, for instance, be used to absorb spilled oil, or for clean-up of environmental pollution, for example, in water, in the air, or on land. Similarly, the material's absorbent properties, together with its biodegradability, also make them useful for delivery of chemicals or chemical formulations. For example, the materials can be treated with solutions of enzymes or pharmaceuticals such as antibiotics, nutrients, or contraceptives, and any necessary excipients, for drug delivery (e.g., for treatment of humans or animals, or for use as or in animal feed and/or bedding), as well as with solutions of fertilizers, herbicides, or pesticides. The materials can optionally be chemically treated to enhance a specific absorption property. For example, the materials can be treated with silanes to render them lipophilic.

Compositions including texturized materials combined with liquids or particulate, powdered, or granulated solids can also be prepared. For example, texturized fibrous materials can be blended with seeds (i.e., with or without treatment with a solution of fertilizer, pesticides, etc.), foodstuffs, or bacteria (e.g., bacteria that digest toxins). The ratio of fibrous materials to the other components of the

compositions will depend on the nature of the components and readily be adjusted for a specific product application.

In some cases, it may be advantageous to associate the texturized fibrous materials, or compositions or composites of such materials, with a structure or carrier  
5 such as a netting, a membrane, a flotation device, a bag, a shell, or a biodegradable substance. Optionally, the structure of carrier may itself be made of a texturized fibrous material (e.g., a material of the invention), or a composition or composite thereof.

#### Composites of Texturized Fibrous Material and Resin

10 Texturized fibrous materials can also be combined with resins to form strong, lightweight composites. Materials that have been treated with chemicals or chemical formulations, as described above, can similarly be combined with biodegradable or non-biodegradable resins to form composites, allowing the introduction of, for example, hydrophilic substances into otherwise hydrophobic polymer matrices.  
15 Alternatively, the composites including texturized fibrous materials and resin can be treated with chemicals or chemical formulations.

The texturized cellulosic or lignocellulosic material provides the composite with strength. The composite may include from about 10% to about 90%, for example from about 30% to about 70%, of the texturized cellulosic or lignocellulosic material by  
20 weight. Combinations of texturized fibrous materials can also be used (e.g., texturized poly-coated paper blended with texturized wood-related materials or other cellulosic or lignocellulosic fibers).

The resin encapsulates the texturized cellulosic or lignocellulosic material in the composites, and helps control the shape of the composites. The resin also transfers  
25 external loads to the fibrous materials and protects the fiber from environmental and structural damage. Composites can include, for example, about 10% to about 90%, more preferably about 30% to about 70%, by weight, of the resin.

Examples of resins that are suitably combined with texturized fibers include polyethylene (including, e.g., low density polyethylene and high density polyethylene),  
30 polypropylene, polystyrene, polycarbonate, polybutylene, thermoplastic polyesters (e.g., PET), polyethers, thermoplastic polyurethane, PVC, polyamides (e.g., 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 g/10 min, and more preferably less than 1 g/10 min.

The resins can be purchased as virgin material, or obtained as waste materials, and can be purchased in pelletized or granulated form. One source of waste resin is  
5 used polyethylene milk bottles. If surface moisture is present on the pelletized or granulated resin, however, it should be dried before use.

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

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

The composites can also include inorganic additives such as calcium carbonate, graphite, asbestos, wollastonite, mica, glass, fiber glass, chalk, silica, talc, ceramic, ground construction waste, tire rubber powder, carbon fibers, or metal fibers (e.g.,  
20 aluminum, stainless steel). When such additives are included, they are typically present in quantities of from about 0.5% up to about 20-30% by weight. For example, submicron calcium carbonate can be added to the composites of texturized fibrous material and resin to improve impact modification characteristics or to enhance composite strength.

25

#### Preparation of Compositions

Compositions containing the texturized cellulosic or lignocellulosic materials and chemicals, chemical formulations, or other solids can be prepared, for example, in various immersion, spraying, or blending apparatuses, including, but not limited to,  
30 ribbon blenders, cone blenders, double cone blenders, and Patterson-Kelly "V" blenders.

For example, a composition containing 90% by weight texturized cellulosic or

lignocellulosic material and 10% by weight ammonium phosphate or sodium bicarbonate can be prepared in a cone blender to create a fire-retardant material for absorbing oil.

5        Preparation of Composites of Texturized Fiber and Resin

Composites of texturized fibrous material and resin can be prepared as follows. A standard rubber/plastic compounding 2-roll mill is heated to 325-400°F. The resin (usually in the form of pellets or granules) is added to the heated roll mill. After about 5 to 10 minutes, the coupling agent is added to the roll mill. After another five minutes,  
10       the texturized cellulosic or lignocellulosic material is added to the molten resin/coupling agent mixture. The texturized material 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  
15       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  
20       as a twin-screw extruder equipped with co-rotating screws. The resin and the coupling agent are introduced at the extruder feed throat; the texturized cellulosic or lignocellulosic material or materials 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 can be, for example,  
25       pelletized by cold strand cutting.

Alternatively, the mixture can first be prepared in a mixer, then transferred to an extruder.

In another embodiment, the composite can be formed into filaments for knitting, warping, weaving, braiding, or in the manufacture of non-wovens. In a further  
30       embodiment, the composite can be made into a film.

### Properties of the Composites of Texturized Fibrous Material and Resin

The resulting composites include a network of fibers, encapsulated within a resin matrix. The fibers form a lattice network, which provides the composite with strength. Since the cellulosic or lignocellulosic material is texturized, the amount of surface area available to bond to the resin is increased, in comparison to composites prepared with un-texturized cellulosic or lignocellulosic material. 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.

### Uses of the Composites of Texturized Fibrous Material and Resin

The resin/fibrous material composites can be used in a number of applications. The composites are strong 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 often stored outdoors for extended periods of time, wine staves, rowboats, furniture, skis, and oars. Many other uses are contemplated, including panels, pipes, decking materials, boards, housings, sheets, poles, straps, fencing, members, doors, shutters, awnings, shades, signs, frames, window casings, backboards, wallboards, flooring, tiles, railroad ties, forms, trays, tool handles, stalls, bedding, dispensers, staves, films, wraps, totes, barrels, boxes, packing materials, baskets, straps, slips, racks, casings, binders, dividers, walls, indoor and outdoor carpets, rugs, wovens, and mats, frames, bookcases, sculptures, chairs, tables, desks, art, toys, games, wharves, piers, boats, masts, pollution control products, septic tanks, automotive panels, substrates, computer housings, above- and below-ground electrical casings, furniture, picnic tables, tents, playgrounds, benches, shelters, sporting goods, beds, bedpans, thread, filament, cloth, plaques, trays, hangers, servers, pools, insulation, caskets, bookcovers, clothes, canes, crutches, and other construction, agricultural, material handling, transportation, automotive, industrial, environmental, naval, electrical, electronic, recreational, medical, textile, and consumer products. Numerous other applications are also envisioned. The composites may also be used, for example, as the base or carcass for a veneer product. Moreover, the composites can be, for example,

surface treated, grooved, milled, shaped, imprinted, textured, compressed, punched, or colored. The surface of the composites can be smooth or rough.

5 The following examples illustrate certain embodiments and aspects of the present invention and not to be construed as limiting the scope thereof.

### Examples

#### Example 1

10 A 1500 pound skid of virgin, half-gallon juice cartons made of polycoated white kraft board was obtained from International Paper. One such carton is shown in Fig. 3. Each carton was folded flat.

The cartons were fed into a 3 hp Flinch Baugh shredder at a rate of approximately 15 to 20 pounds per hour. The shredder was equipped with two rotary blades, each 12" in length, two fixed blades, and a 0.3" discharge screen. The gap  
15 between the rotary and fixed blades was 0.10".

A sample of the output from the shredder, consisting primarily of confetti-like pieces, about 0.1" to 0.5" in width and about 0.25" to 1" in length, is shown in Fig. 4. The shredder output was fed into a Thomas Wiley Mill Model 2D5 rotary cutter. The rotary cutter had four rotary blades, four fixed blades, and a 2 mm discharge screen.  
20 Each blade was approximately 2" long. The blade gap was set at 0.020".

The rotary cutter sheared the confetti-like pieces across the knife edges, tearing the pieces apart and releasing a finely texturized fiber at a rate of about one pound per hour. The fiber had an average minimum L/D ratio of between five and 100 or more. The bulk density of the texturized fiber was on the order of 0.1 g/cc. A sample of  
25 texturized fiber is shown in Fig. 5 at normal magnification, and in Fig. 2 at fifty-fold magnification.

#### Example 2

30 Composites of texturized fiber and resin were prepared as follows. A standard rubber/plastic compounding 2-roll mill was heated to 325-400°F. The resin (usually in the form of pellets or granules) was added to the heated roll mill. After about 5 to 10 minutes, the resin banded on the rolls (i.e., it melted and fused on the rolls). The

coupling agent was then added to the roll mill. After another five minutes, the texturized cellulosic or lignocellulosic material was added to the molten resin/coupling agent mixture. The cellulosic or lignocellulosic fiber was added over a period of about 10 minutes.

5 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 composite contained the following ingredients:

10	<u>Composition No. 1</u>	
	<u>Ingredient</u>	<u>Amount(g)</u>
	High density polyethylene <sup>1</sup>	160
	Old newspaper <sup>2</sup>	240
	Coupling agent <sup>3</sup>	8
15	<sup>1</sup> Marlex 16007	
	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
	<sup>3</sup> FUSABOND® 100D	

20 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:

25	Flexural strength (10 <sup>3</sup> psi)	9.81 (ASTM D790)
	Flexural modulus (10 <sup>5</sup> psi)	6.27 (ASTM D790)

A second composite contains the following ingredients:

	<u>Composition No. 2</u>	
	<u>Ingredient</u>	<u>Amount(g)</u>
30	High density polyethylene <sup>1</sup>	160
	Old magazines <sup>2</sup>	240
	Coupling agent <sup>3</sup>	8
	<sup>1</sup> Marlex 16007	
	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
35	<sup>3</sup> FUSABOND® 100D	

The properties of Composition No. 2 are as follows:

5	Flexural strength ( $10^3$ psi) Flexural modulus ( $10^5$ psi)	9.06 (ASTM D790) 6.78 (ASTM D790)
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A third composite contains the following ingredients:

Composition No. 3

	Ingredient	Amount(g)
10	HDPE <sup>1</sup>	160
	Fiber paper <sup>2</sup>	216
	3.1 mm texturized kenaf	24
	Coupling agent <sup>3</sup>	8
	<sup>1</sup> Marlex 16007	
15	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
	<sup>3</sup> FUSABOND® 100D	

The properties of Composition No. 3 are as follows:

20	Flexural strength ( $10^3$ psi) Flexural modulus ( $10^5$ psi)	11.4 (ASTM D790) 6.41 (ASTM D790)
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A fourth composite contains the following ingredients:

Composition No. 4

	Ingredient	Amount (g)
25	SUPERFLEX® CaCO <sub>3</sub>	33
	Fiber <sup>2,4</sup>	67
	HDPE (w/3% compatibilizer) <sup>1,3</sup>	100
	<sup>1</sup> Marlex 16007	
30	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
	<sup>3</sup> FUSABOND® 100D	
	<sup>4</sup> Virgin polycoated milk cartons	

The properties of Composition No. 4 are as follows:

5	Flexural strength ( $10^5$ psi)	8.29 (ASTM D790)
	Ultimate elongation (%)	<5 (ASTM D638)
	Flexural modulus ( $10^5$ psi)	10.1 (ASTM D790)
	Notch Izod (ft-lb/in)	1.39 (ASTM D256-97)

A fifth composite contains the following ingredients:

Composition No. 5		
	<u>Ingredient</u>	<u>Amount (parts)</u>
	SUPERFLEX® CaCO <sub>3</sub>	22
	Fiber <sup>2,4</sup>	67
	HDPE (w/3% compatibilizer) <sup>1,3</sup>	100
15	<sup>1</sup> Marlex 16007	
	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
	<sup>3</sup> FUSABOND® 100D	
	<sup>4</sup> Virgin polycoated milk cartons	

The properties of Composition No. 5 are as follows:

20	Flexural strength ( $10^5$ psi)	8.38 (ASTM D790)
	Ultimate elongation (%)	<5 (ASTM D638)
	Flexural modulus ( $10^5$ psi)	9.86 (ASTM D790)
	Notch Izod (ft-lb/in)	1.37 (ASTM D256-97)

A sixth composite contains the following ingredients:

Composition No. 6		
	<u>Ingredient</u>	<u>Amount (parts)</u>
30	ULTRAFLEX® CaCO <sub>3</sub>	33
	Fiber <sup>2,4</sup>	67
	HDPE/compatibilizer <sup>1,3</sup>	100
	<sup>1</sup> Marlex 16007	
	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
35	<sup>3</sup> FUSABOND® 100D	
	<sup>4</sup> Virgin polycoated milk cartons	

The properties of Composition No. 6 are as follows:

5	Flexural strength ( $10^5$ psi)	7.43 (ASTM D790)
	Ultimate elongation (%)	<5 (ASTM D638)
	Flexural modulus ( $10^5$ psi)	11.6 (ASTM D790)
	Notch Izod (ft-lb/in)	1.27 (ASTM D256-97)

A seventh composite contains the following ingredients:

10	<u>Composition No. 7</u>	
	<u>Ingredient</u>	<u>Amount (pbw)</u>
	HDPE (w/3% compatibilizer) <sup>3,5</sup>	60
	Kraftboard <sup>2</sup>	40
15	<sup>1</sup> Marlex 16007	
	<sup>2</sup> Texturized using rotary cutter with 2 mm mesh	
	<sup>3</sup> FUSABOND® 100D	
	<sup>4</sup> Virgin polycoated milk cartons	
	<sup>5</sup> HDPE with melt-flow index <1	

The properties of Composition No. 7 are as follows:

25	Flexural Strength ( $10^5$ psi)	7.79 (ASTM D790)
	Ultimate elongation (%)	<5 (ASTM D638)
	Flexural Modulus ( $10^5$ psi)	7.19 (ASTM D790)

An eighth composite contains the following ingredients:

30	<u>Composition No. 8</u>	
	<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
35	<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 properties of Composition No. 8 are as follows:

	Tensile modulus ( $10^5$ psi)	8.63 (ASTM D638)
	Tensile strength at break (psi)	6820 (ASTM D638)
5	Ultimate elongation (%)	<5 (ASTM D638)
	Flexural Strength (psi)	12,200 (ASTM D790)
	Flexural modulus ( $10^5$ psi)	6.61 (ASTM D790)

A ninth composite contains the following ingredients:

10

Composition No. 9

<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

15

<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. 9 are as follows:

20

	Tensile modulus ( $10^5$ 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)
25	Flexural modulus ( $10^5$ psi)	6.50 (ASTM D790)

A tenth composite contains the following ingredients:

Composition No. 10

	<u>Ingredient</u>	<u>Amount (g)</u>
30	High density polyethylene <sup>1</sup>	160
	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

35

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

The properties of Composition No. 10 are as follows:

5	Tensile modulus ( $10^5$ psi)	7.08 (ASTM D638)
	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)

10 An eleventh composite contains the following ingredients:

Composition No. 11

	<u>Ingredient</u>	<u>Amount (g)</u>
	High density polyethylene <sup>1</sup>	160
	Poly-coated paper <sup>2</sup>	240
15	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® MB 100D

20 The properties of Composition No. 11 are as follows:

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

Other embodiments are within the claims.

30 What is claimed is:

### Claims

1. A composite comprising (a) a texturized fibrous material comprising a  
cellulosic or lignocellulosic material having internal fibers, (b) a thermoplastic resin,  
5 and (c) an inorganic additive, wherein said cellulosic or lignocellulosic material is  
sheared to the extent that the internal fibers are substantially exposed.
2. The composite of claim 1, wherein the resin is selected from the group  
consisting of polyethylene, polypropylene, polystyrene, polycarbonate, polybutylene,  
10 thermoplastic polyesters, polyethers, thermoplastic polyurethane, polyvinylchloride,  
and polyamides.
3. The composite of claim 1, wherein the cellulosic material comprises poly-  
coated paper.  
15
4. The composite of claim 3, wherein the poly-coated paper comprises  
polyethylene and paper.
5. The composite of claim 4, wherein the poly-coated paper further comprises  
20 one or more layers of aluminum.
6. The composite of claim 1, wherein said cellulosic or lignocellulosic material  
is selected from the group consisting of flax, hemp, cotton, jute, rags, paper, paper  
products, and byproducts of paper manufacturing.  
25
7. The composite of claim 1, wherein the cellulosic or lignocellulosic material  
is pulp board.
8. The composite of claim 1, wherein the cellulosic or lignocellulosic material  
30 is a synthetic material.
9. The composite of claim 1, wherein the cellulosic or lignocellulosic material

is a non-woven material.

10. The composite of claim 1, wherein at least about 50% of the fibers have a length/diameter ratio of at least about 5.

5

11. The composite of claim 1, wherein at least about 50% of the fibers have a length/diameter ratio of at least about 25.

12. The composite of claim 1, wherein at least about 50% of the fibers have a length/diameter ratio of at least about 50.

10

13. The composite of claim 1, wherein the inorganic additive is selected from the group consisting of calcium carbonate, graphite, asbestos, wollastonite, mica, glass, fiber glass, chalk, talc, silica, ceramic, ground construction waste, tire rubber powder, carbon fibers, and metal fibers.

15

14. The composite of claim 1, wherein the inorganic additive comprises from about 0.5% to about 20% of the total weight of the composite.

20

15. The composite of claim 1, wherein said composite is in the form of a pallet.

16. The composite of claim 15, wherein said pallet is injection molded.

17. The composite of claim 1, wherein said composite is in the form of an article selected from the group consisting of panels, pipes, decking materials, boards, housings, sheets, poles, straps, fencing, members, doors, shutters, awnings, shades, signs, frames, window casings, backboards, wallboards, flooring, tiles, railroad ties, forms, trays, tool handles, stalls, bedding, dispensers, staves, wraps, totes, barrels, boxes, packing materials, baskets, straps, slips, racks, casings, binders, dividers, walls, indoor and outdoor carpets, rugs, wovens, and mats, frames, bookcases, sculptures, chairs, tables, desks, art, toys, games, wharves, piers, boats, masts, pollution control products, septic tanks, automotive panels, substrates, computer housings, above- and

30

below-ground electrical casings, furniture, picnic tables, tents, playgrounds, benches, shelters, sporting goods, beds, bedpans, thread, cloth, plaques, trays, hangers, servers, pools, insulation, caskets, bookcovers, clothes, canes, crutches, and other construction, agricultural, material handling, transportation, automotive, industrial, environmental,  
5 naval, electrical, electronic, recreational, medical, textile, and consumer products.

18. The composite of claim 1, wherein said composite is in the form of a fiber, filament, or film.

10 19. A composition comprising a texturized fibrous material comprising (a) a cellulosic or lignocellulosic material having internal fibers and (b) a chemical or chemical formulation, wherein said cellulosic or lignocellulosic material is sheared to the extent that the internal fibers are substantially exposed.

15 20. The composition of claim 19, wherein the chemical formulation comprises a pharmaceutical composition.

21. The composition of claim 19, wherein the chemical formulation an agricultural compound.

20

22. The composition of claim 19, wherein the chemical formulation comprises an enzyme.

23. A composition comprising (a) a texturized fibrous material comprising a  
25 cellulosic or lignocellulosic material having internal fibers and (b) a liquid, wherein said cellulosic or lignocellulosic material is sheared to the extent that the internal fibers are substantially exposed.

24. A composition comprising (a) a texturized fibrous material comprising a  
30 cellulosic or lignocellulosic material having internal fibers and (b) particulate, powdered, or granulated solid, wherein said cellulosic or lignocellulosic material is sheared to the extent that the internal fibers are substantially exposed.

25. The composition of claim 24, wherein the solid comprises plant seed.

26. The composition of claim 24, wherein the solid comprises a foodstuff.

5

27. The composition of claim 24, wherein the solid comprises bacteria.

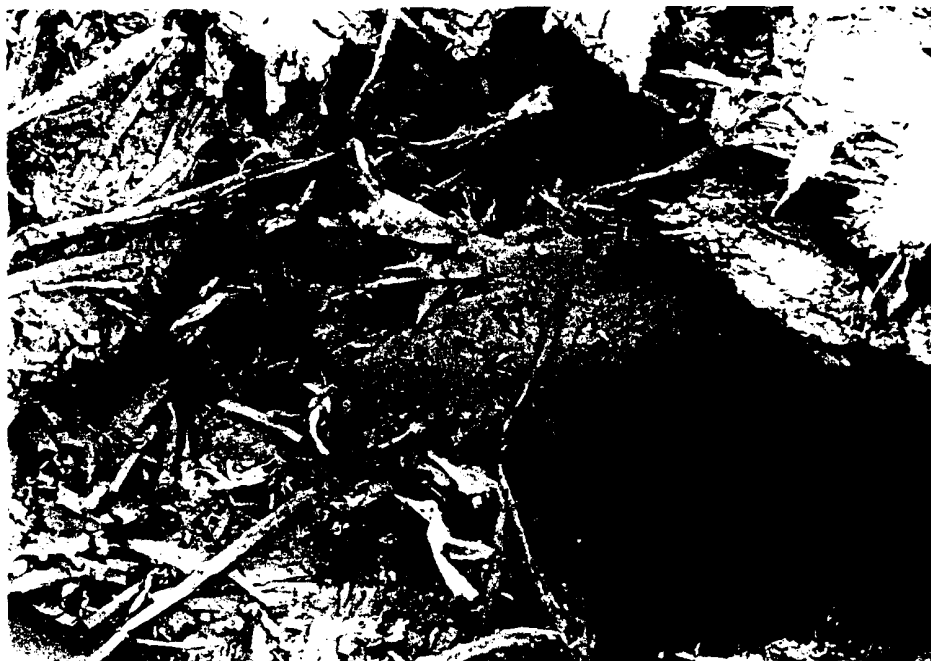


FIG. 1



FIG. 1

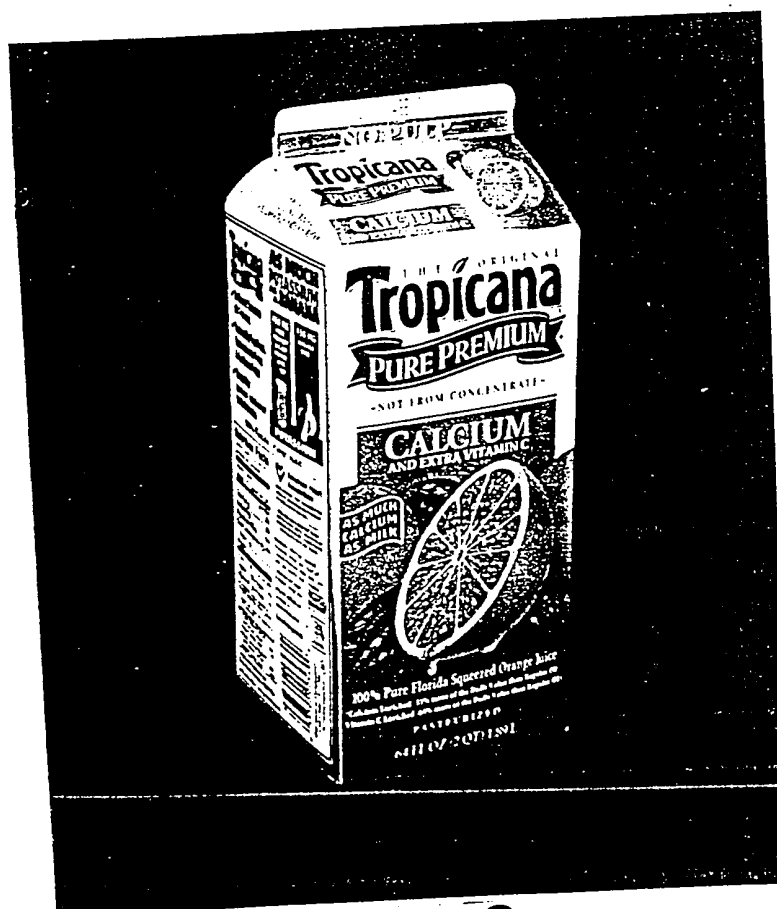


FIG. 3



FIG. 34

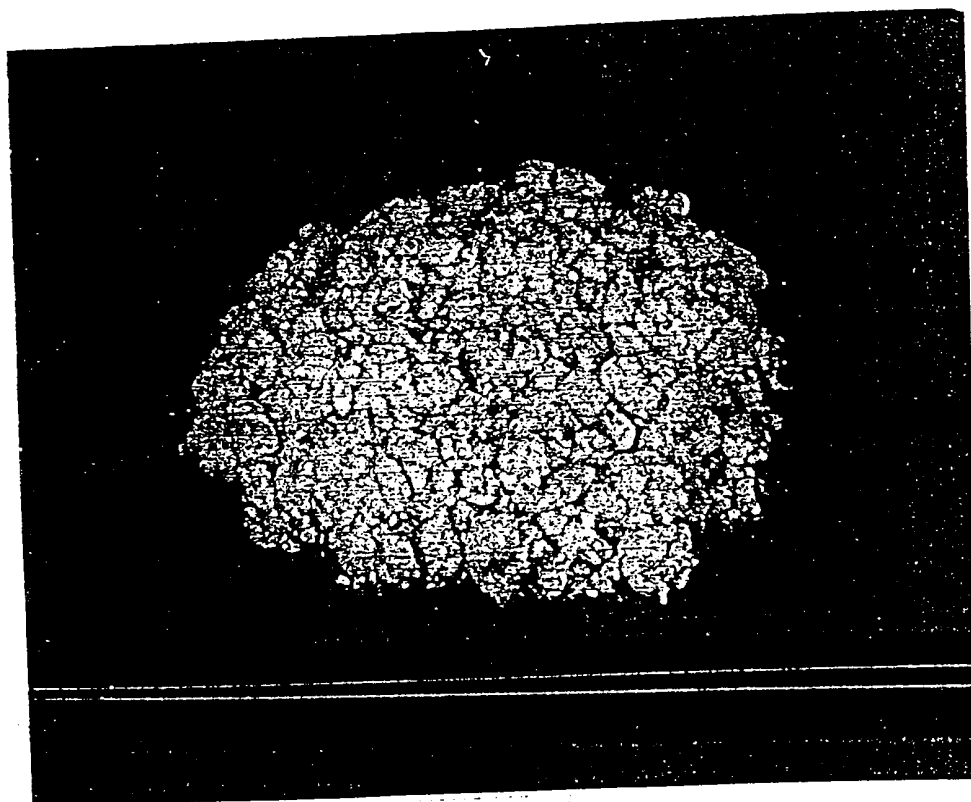


FIG. 45

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US00/17232

**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(7) : Please See Extra Sheet.

US CL : Please See Extra Sheet.

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 47/26, 65.5; 111/200; 424/405, 411, 413, 414, 94.1; 426/5; 428/364, 374; 523/129; 524/13, 14, 76

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

NONE

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4,244,847 A (POSIVIATA et al) 13 January 1981, see entire document.	1-27
Y	US 4,791,020 A (KOTKA) 13 December 1988, see entire document.	1-27
Y	US 5,516,472 A (LAVER) 14 May 1996, see entire document.	1-27



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

07 AUGUST 2000

Date of mailing of the international search report

24 AUG 2000

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# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US00/17232

## A. CLASSIFICATION OF SUBJECT MATTER:

IPC (7):

A01G 9/02, 13/00; A01N 25/00, 25/34; A23G 3/00; A61K 38/43; C08J 9/00; C08K 5/13; C08L 9/00; C09K 11/00;  
D02G 3/00

## A. CLASSIFICATION OF SUBJECT MATTER:

US CL :

47/26, 65.5; 111/200; 424/405, 411, 413, 414, 94.1; 426/5; 428/364, 374; 523/129; 524/13, 14, 76