BIODEGRADABLE NONWOVENS BASED ON COTTON AND POLYESTERS Ovidiu Chiparus, Ioan Negulescu and Yan Chen Louisiana State University Baton Rouge, LA Mary Warnock University of Arkansas Fayetteville, AR Larry Wadsworth University of Tennessee Knoxville, TN

Abstract

The main objective of the paper was to prepare cellulosics/polyester biodegradable nonwoven flexible sheets containing cotton and at least 50% bagasse coarse fibers, and to determine their most relevant end-use properties.

Introduction

Approximately 22% of the textile fibers produced in US are used for industrial applications, which includes a multitude of products, such as low and high resins composites (ATMI, 1995). However, the abundance of recyclable fiber and agricultural residues/byproducts coupled with the dwindling supply of some natural resources have created the need to develop alternative structural composites and materials. Agricultural residues and by products from major U.S. agricultural commodities (sugarcane, soybeans, wheat, corn, etc.) potentially could be used to produce a multitude of value-added non-food products, ranging from fibers, films, plastics, and composites to resins, finishing agents, and auxiliaries. However, most often than not, they are discarded or used for only low-value applications. For example, the crushed stalks (bagasse) remaining from traditional sugar cane processing are used in-house as a fuel in mill processes and for other low value applications such as mulch and inexpensive ceiling tiles (Paturau 1989). Development of value-added products from the waste or low-value materials could allow mills to migrate to cleaner burning fuels and provide economic benefits as cane producers compete in a freer trade environment. To this aim, researchers in Louisiana, a state with significant sugarcane products (Collier et al. 1995, 1997 and 1998). In line with these efforts, the scope of the present work was to produce novel value-added materials based on bagasse and biodegradable synthetic polymers, such as flexible nonwoven composites.

Materials, Methods and Results

Cellulosic Fibers

Bagasse fibers have been blended with cotton fibers in a 70:30 weight ratio, carded and lightly needled to form a web.

Synthetic Polymer

The polymer used as an adhesive for binding the cellulosic webs was poly(tetramethylene adipate-co-terephthalate), referred also as Eastar BioCopolymer. It is a biodegradable random copolyester of adipic acid, terephthalic acid and butanediol (Haile 2001):

 $HOOC-(CH_2)_4-COOH + HOOC-C_6H_4-COOH + HO-(CH_2)_4-OH \Rightarrow \equiv Eastar BioCopolyester$

The polyester pellets have been transformed in a melt blown nonwoven fabric (57 g/m²) at the TANDEC unit from the University of Tennessee in Knoxville, TN.

Formation of Composite Nonwovens

To obtain the nonwoven composites shown in Fig. 1, the cellulosics webs and the melt blown polyester nonwoven sheets (in a 70:30 wt. ratio) have been sandwiched and pressed into a flexible bonded sheet for one minute at 150°C and 8.62 Pa (12,500 psi).

Characteristics

Bagasse content: ~54% Initial sandwich density: 0.089 g/cm³ (88 oz/ft³) Density after pressing: 0.55 g/cm³ (548 oz/ft³)

Possible End-Uses

A number of disposable and limited use applications are foreseen, including sound proofing materials, seed mats and other seasonal agricultural nonwovens.

Thermal Analysis

Thermogravimetric analysis of the composite nonwoven and its components was carried out both in air and inert atmosphere (nitrogen). Weight loss curves (TG) in nitrogen are shown in Figure 2. A comparison of derivatives (DTG) is presented in Figure 3. A shift to higher temperature of the DTG peak of cellulosics from the composite nonwoven suggests a chemical interaction between cotton/bagasse and polyester during hot-pressing (trans-esterification?). Maximum loss data are listed in Table 1.

Determination of Nonwoven Composition from TG Data

The composition of the nonwoven (X, Y) can be determined from thermogravimetric data using the following equations:

$$X + Y = 100$$

 $X600 + Y600 = Z600$

and

$$x_{600} = [100 X_{600}]/X$$

 $y_{600} = [100 Y_{600}]/Y$

where:

X = % cellulosics Y = % polyester X₆₀₀ = 100 -%X at 600°C Y₆₀₀ = 100 -%Y at 600°C Z₆₀₀ = %(X+Y)at 600°C

and

 $x_{600} = 100$ -%Weight loss of pure cellulosics at 600°C $y_{600} = 100$ -%Weight loss of pure polyester at 600°C

Taking from thermograms: $x_{600} = 22.7\%$, $y_{600} = 3.0\%$ and $Z_{600} = 15.4\%$ and solving the equations one obtains: X = 63% and Y = 37%.

Determination of Nonwoven Composition from DTG Data

The nonwoven content of water (2-4%, depending upon the composition) may affect the weight loss at a certain temperature (e.g., 600° C). Therefore, the composition (X, Y) can be determined more precisely from DTG data using the ratio of peak values (or of values at any other temperature) for individual components. For example, the ratio of DTG data at 326°C will determine the X (bagasse and cotton), while the ratio of data at 388°C will give the content of the Eastar BioCopolymer, Y:

<u>At 326°C</u>:

$$X = [(0.775 \%)^{\circ}C):(1.14 \%)^{\circ}C)]*100 = 67.98\%$$

It follows that

Y = 100 - 67.98 = 32.02%

<u>At 388°C</u>:

 $Y = [(0.799 \%)^{\circ}C):(2.49 \%)^{\circ}C)]*100 = 32.09\%$

which gives

$$X = 100 - 32.09 = 67.91\%$$

This value for X agrees very well with the content determined at 326° C and the initial ratio of cellulosics and BioCopolyester (X = 70% and Y = 30%). The loss of water during pressing and a diminished moisture regain of the cellulosics from the pressed nonwoven lowered the cellulosics content correspondingly.

Dynamo-Mechanical Analysis of Nonwoven Composites

The major thermal transitions observed in DMA spectra of composites were related to the synthetic polyester. Glass transition, Tg, as defined by the loss elastic modulus E", was seen at negative temperatures, Depending on the frequency used during the measurement, between -25 and -12° C. The melting was also dependent on the frequency and was observed around $75\pm5^{\circ}$ C, the higher the frequency used, the higher the melting peak (Figure 4). It should be mentioned that the glass transition at a certain frequency observed in the composite material was 20°C higher than the Tg of the pure polyester observed at the same frequency, suggesting some kind of interaction between the polyester and cellulosic fibers during hot-pressing (trans-esterification?).

Biodegradation of the Bonding Eastar BioCopolymer and of Composite Nonwovens

The Eastar BioCopolymer is designed to perform for the useful life of a manufactured item and then fully degrade within a composting environment. In a time frame comparable to cellulosics (paper), this aliphatic-aromatic polyester fully degrades to CO₂, water and biomass. Within 12 weeks in an active composting site, an article made from this copolymer typically becomes invisible to the naked eye and completely biodegrades within six month (Haile 2001). In the present work, duplicate samples of melt blown Eastar BioCopolymer and composite nonwoven with bagasse and cotton mats have been buried and for 1, 2 and 3 weeks and analyzed thereafter for lost of mechanical strength. The results are shown in Figures 5 and 6.

Conclusions

Elastic composite nonwoven sheets have been successfully prepared by hot pressing from bagasse/cotton mats sandwiched between Eastar BioCopolymer melt blown nonwoven fabrics.

A chemical interaction took place between cellulosics and polyester during hot pressing as evidenced by the shift of Tg and of peak temperatures of DTG curves.

The soil burial method clearly evidenced the starting of the biodegradation process by the diminution of the breaking strength of test samples after three weeks.

References

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Table 1. Thermogravimetric analysis of the composite nonwoven and its components in nitrogen.

	Weight loss at	100 – TG at 600°C	DTG at maximum loss
Material	600°C (TG) %	%	%/°C
Composite	84.6	15.4	0.775 @ 326°C
nonwoven			0.799 @ 388°C
Bagasse/	77.3	22.7	1.14 @ 326°C
Cotton web			0.09 @ 388°C
Eastar	97.0	3.0	0.05 @ 326°C
BioCopolymer			2.49 @ 388°C



Figure 1. Layered composite nonwovens made of bagasse/cotton webs and Eastar Bio Copolymer melt blown nonwovens before (top) and after hot-pressing (bottom).



Figure 2. Comparison of weight loss during drying and thermal decomposition in nitrogen (5C/min) of composite nonwoven and parent materials.



Figure 3. Derivatives of weight loss curves (DTG) in nitrogen for nonwoven and its components.



Figure 4. Variation with temperature of the loss modulus E" of composite nonwovens.



Figure 5. Mechanical properties non-compost of soil degraded samples: Young modulus.



Figure 6. Mechanical properties non-compost of soil degraded samples: stress at break.