Biodegradable Paper Sheeting as Agricultural Covering with Incorporation of Bamboo Pulp Sludge

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This paper reports the manufacturing process for biodegradable paper sheeting with incorporation of bamboo paper sludge, fibers of poplar woods, and viscose fibers by wet-laid nonwoven technology. The best process conditions included a basis weight of 30 g/m^2, a bamboo paper sludge content of 10 wt%, and a polyvinyl alcohol concentration of 4 wt%. The burst strength, tearing resistance, tensile properties, resistance to water, and degradation rate were 220.65 kPa, 60.00 N, 46.10 N, 153 Pa, and 56.18%, respectively, under the best process conditions. The biodegradable paper sheeting can satisfy the demand for replacement of agricultural plastic sheeting used for such purposes as moisture retention of soil and promotion of plant growth.

Keywords: Bamboo pulp sludge; Biodegradable; Paper mulch film

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INTRODUCTION

The production and usage of plastic agricultural sheeting in China is the highest in the world. The usage of plastic agricultural sheeting has reached 1 million tons and has covered 206.7 billion square meters annually in China since 2009 (Zhang *et al*. 2011). With the continuing development of China's agriculture, the demands for agricultural sheeting and/or mulch materials are growing. Agricultural sheeting products from polyvinyl chloride (PVC) and from polyethylene (PE) are widely used for agriculture and can shorten the growth cycle of crops and increase production by 20%, which brings great economic benefits (Sheng 2009). However, there are disadvantages. For example, the agricultural sheeting that cannot be degraded will make the soil infertile and give rise to airborne dust (Sheng 2009). To reduce entrainment of dust into the atmosphere and to achieve sustainable agriculture development, research on biodegradable sheeting should be conducted.

Bamboo pulp sludge is a byproduct of pulping and papermaking wastewater treatment and is a renewable biomass raw material. However, it is difficult to dispose of, and the cost of treatment is high. Not only can the development and utilization of bamboo pulp sludge solve the problem of secondary pollution of the environment, but it can also produce certain economic effects. At present, the comprehensive utilization of bamboo pulp sludge has the following values: burning, fertilizer application, soil improvement, and related uses (Cong *et al*. 2011; Wang *et al.* 2009; Zhang *et al.* 2011). The authors' laboratory also has conducted related work, such as research on wood-based panels and MDF (Wang 2011; Zhang 2011; Pang 2008). However, there is almost no literature dealing with biodegradable paper sheeting made from bamboo pulp sludge. In the present work, bamboo paper sludge, fibers of poplar wood, and viscose fibers (man-made fibers made from cotton and other natural fibers) were used to make biodegradable paper sheeting by wet-laid nonwoven technology through purification, mixing, forming the pulp suspension into paper, and surface sizing. The properties of the agricultural paper were tested to obtain the best process conditions that make full use of bamboo pulp sludge and reduce airborne dust pollution.

EXPERIMENTAL

Materials

Bamboo paper sludge was provided by the GuiZhou bamboo paper industry in Guizhou province, China. The poplar wood fibers were provided by the Guangda artificial board factory in Hefei, Anhui province, China. Viscose fibers were provided by the Antai Engineering Materials Company Ltd. (Laiwu, Shandong province, China), polyvinyl alcohol (PVOH) by Guangfu Fine Chemical Research Institute (Tianjin, China), and polyacrylamide by Yurun Chemical Technology Company Ltd. (Guangzhou, Guangdong province, China).

Methods

Pretreatment

Bamboo pulp sludge provided with a moisture content of 40 to 50% was dried in an oven at 103 ± 2 °C to a final moisture content of 12.5%. The dried material was filtered through a 100 mesh (0.150 mm) screen in a sieve. Poplar wood fibers were blanched first with a 6% hydrogen peroxide (H_2O_2) and 4.5% sodium hydroxide (NaOH) solution (pH 10.5 to 11.0) at a 3:1 (by volume) concentration of solution to fibers. After 60 min, the fibers turned white and were washed with water until the pH was neutral. The wet fibers were dried in the oven (the temperature was 103 ± 2 °C) until the moisture content was 12.5%. Viscose fibers were cut into lengths of 1 to 2 mm. All the materials should be cleaned, such as stone, glass and grass. Figure 1 shows the materials used to make the paper mulch film.

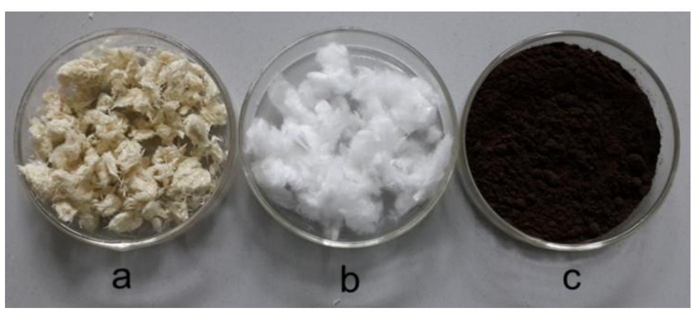

Fig. 1. The materials used to make paper sheeting; (a) poplar wood fibers after blanching, (b) viscose fibers, and (c) bamboo paper sludge

Manufacturing

Manufacturing of the paper sheeting was performed according to the parameters set forth in Table 1. The quantities of bamboo paper sludge, poplar wood fibers, and viscose fibers were calculated by area density, and the manufacturing was randomly assigned to three treatments. The poplar wood fibers and the viscose fibers were mixed at a 1:1 ratio (Factor A), and then added to the appropriate concentration of bamboo paper sludge (Factor B) and polyacrylamide (1 wt‰) dispersing agent in a beaker to make a uniform liquid suspension (Fig. 2). To manufacture paper, the liquid suspension was poured into a sheeting mold (Fig. 3). After the water had wicked, the paper sheet was dried in an oven for 2 h at 60 °C and then carefully removed. For surface sizing, the paper specimen was placed on the sheeting mold and the PVOH solution (Factor C) was brushed onto the surface to fully saturate the material. The surface sizing strengthened the paper as well as filled the holes in the paper sheeting. After air drying, the sized paper sheeting was characterized.

Table 1. Orthogonal Experimental Plan

No.	Factor A Area density (g/m^2)	Factor B Bamboo paper sludge content (wt%)	Factor C PVOH concentration (wt%)
1	30	5	2
2	40	10	4
3	50	15	6

Fig. 2. Uniform liquid suspension of fibers, sludge, and polyacrylamide

Fig. 3. Liquid suspension poured into sheeting mold

Fig. 4. Surface sizing of dried paper with PVOH

Property Testing

Tests to determine burst strength, tearing resistance, tensile properties, and resistance to water were performed in accordance with GB/T 454-2002 (2002), GB/T 455-2002 (2002), GB/T 22898-2008 (2008), and GB/T 4744-1997 (1997). The paper sheeting and PE agricultural sheeting were both tested.

Degradation Testing

The agricultural sheeting (both the paper sheeting and PE sheeting) was buried in soil (moisture content of 20%) to a depth of 10 cm for 7, 14, 21, and 49 days to test the degradation rate (DR) of the film, which was calculated according to Eq. 1:

$$DR = \frac{W_0 - W_1}{W_0} \times 100\% \tag{1}$$

where W_0 refers to the quantity before degradation and W_1 refers to the quantity after degradation. The content of carbon (C%) and nitrogen (N%) in bamboo pulp sludge and soil were tested by a Euro EA 3000 carbon and nitrogen analyzer (EuroVector, Milan, It).

Morphology and Topography

The surface topography of the paper sheeting was analyzed using a SMZ-168-BL stereo microscope (Motic, Hong Kong) which was magnified by 50x. The eyepiece was 10x and the diameter of the field of view 4.6 mm.

RESULTS AND DISCUSSION

Properties

Table 2 provides a summary of the properties of the paper sheeting, which is shown in Fig. 5. Composite strength, which combines burst strength with tearing resistance, was used to describe the properties of the paper mulch film. The composite strength of sample 2 was found to be the highest. Moreover, the burst strength and tensile properties of sample 2 were both higher than the average value. The tearing resistance was lower than the average value which did not have a significant effect on the properties.

Fig. 5. Paper sheeting

Table 2. Properties of Paper Sheeting

No.	Factors			Thickness	Tearing Resistance	Burst Strength	Tensile Properties	Resistance to Water	Composite Strength*
	A	B	C	mm	N	kPa	N	Pa	
1	1	1	1	0.30	41.33	86.63	18.16	141.33	1.995
2	1	2	2	0.38	60.00	220.65	46.10	153.00	3.835
3	1	3	3	0.33	54.00	164.26	39.77	326.67	3.139
4	2	1	2	0.44	61.33	189.60	26.53	283.33	2.696
5	2	2	3	0.47	72.00	229.48	40.02	296.50	3.213
6	2	3	1	0.39	56.00	127.81	20.69	299.00	2.115
7	3	1	3	0.58	81.55	325.58	71.83	517.33	3.259
8	3	2	1	0.47	68.00	106.57	22.56	276.00	1.703
9	3	3	2	0.51	80.53	261.18	47.37	306.33	2.901
Average	-	-	-	0.43	63.86	190.19	37.00	288.83	2.762

*Composite Strength Index = (Burst Strength Index × Tearing Resistance Index)$^{1/2}$
A, area density; B, bamboo paper sludge content; C, PVA concentration

To confirm the influence of these factors on the sheeting and to enhance the reliability of the results, we conducted range analyses on the properties of the sheeting. Results, which are shown in Table 3, indicate the following:
(1) The relative influence of different factors for thickness and tearing resistance were in the order Factor A ＞ Factor C ＞ Factor B. The properties increased with increasing basis weight (mass per unit of area). Sheeting having a higher basis weight will have more fibers per unit of area. Also, the thickness, burst strength, and tearing resistance will increase. If the basis weight is too high, the cost of the mulch film will be excessive.
(2) The influence or factors affecting tensile properties and burst strength were in the order Factor C ＞Factor A ＞Factor B. The thickness will increase when the PVOH concentration is increased because the sheeting surface will then contain high amounts of PVOH. High amounts of PVOH will also increase the cost. In addition, the resistance to water will be poor and lead to a softening of the sheeting upon exposure to moisture.

The best process parameters as listed in Table 1 were A1, B2, C2 (area density of 30 g/m^2, bamboo paper sludge content of 10 wt%, and PVA concentration of 4 wt%). The burst strength, tearing resistance, tensile properties, and resistance to water were 220.65 kPa, 60.00 N, 46.10 N, and 153 Pa, respectively, under these conditions.

Table 3. Range Analysis of the Sheeting Properties

	Thickness/mm			Tearing Resistance/N			Bursting Strength/kPa			Tensile Properties/N		
	A	B	C	A	B	C	A	B	C	A	B	C
K1	1.0	1.3	1.2	155.3	184.2	165.3	471.54	601.80	321.00	104.0	116.5	61.4
K2	1.3	1.3	1.3	189.3	200.0	201.9	546.88	556.69	671.43	87.3	108.7	120.0
K3	1.6	1.2	1.4	230.1	190.5	207.5	693.33	553.26	719.32	141.8	107.8	151.6
k1	0.3	0.4	0.4	51.8	61.4	55.1	157.18	200.60	107.00	34.7	38.8	20.5
k2	0.4	0.4	0.4	63.1	66.7	67.3	182.29	185.56	223.81	29.1	36.2	40.0
k3	0.5	0.4	0.5	76.7	63.5	69.2	231.11	184.42	239.77	47.3	35.9	50.5
R	0.2	0.0	0.1	24.9	5.3	14.1	73.93	16.18	132.77	18.2	2.9	30.1

Table 4 compares PE sheeting and paper sheeting that was under the optimal process parameters (*i.e.*, area density of 30 g/m^2, bamboo paper sludge content of 10 wt%, and PVA concentration of 4 wt%). The properties of the paper sheeting were better than that of the PE sheeting, to include higher tearing resistance, burst strength, tensile properties, and resistance to water.

Table 4. Comparison of PE Film and Paper Sheeting

	Thick-ness (mm)	Area Density (g/m^2)	Tearing Resistance (N)	Burst Strength (kPa)	Tensile Properties (N)	Resistance to Water (Pa)	Composite Strength*
PE sheeting	0.02	16	2.12	107.87	10.00	-	2.053
Paper sheeting	0.38	30	60.00	220.65	46.10	153.00	3.835

*Composite Strength Index = (Burst Strength Index × Tearing Resistance Index)$^{1/2}$

The paper sheeting had a higher composite strength, so it may be able to satisfy the demand for agricultural plastic mulch film used for agricultural production. Moreover, the resistance to water is a new function to the sheeting, which could increase the breathability of soil and crops, in contrast to the non-breathable nature of a PE film.

Degradation

The contents of carbon (C%) and nitrogen (N%) in the bamboo pulp sludge and soil are listed in Table 5. The nitrogen content and carbon content decreased in the course of exposure to the soil. It is possible that the mulch film contained bamboo pulp sludge. The microorganisms in the bamboo pulp sludge might expend the nitrogen. The content of nitrogen in the soil is a factor of soil fertility. So the decreasing nitrogen content indicated that the degradation of agricultural paper sheeting would not improve soil fertility. Because the degradation reduced the fertility of the soil, the choice of soil in which to bury the paper sheeting must be considered.

Table 6 shows the quantitative changes of paper sheeting and PE film during degradation. The quantity of PE sheeting changed little, while the quantity of paper sheeting exhibited a large change. Figure 6 shows the degradation change curve of paper sheeting in the soil as calculated from the degradation rate. The slope of the curve was high until 21 days, where it leveled off and the degradation increased rapidly. The degradation tended to increase with time and reached 56.18% after 49 days. The paper sheeting degraded completely in 100 days. However, the ordinary PE mulch film did not change in the soil after 49 days. These results showed that the paper sheeting, relative to PE mulch, had a high degradability.

The main factors affecting the degradation of sheeting are as follows (Huang *et al.* 2007; Li *et al*. 2009):

(1) Water and humidity. Viscose fibers and PVOH have large numbers of hydrophilic groups. With the seeping of water, the dissolved hydrophilic groups in the paper sheeting began to dissolve out, and the internal composition of polymer materials changed, which increased the rate of decomposition.

(2) Microbes. Bamboo pulp sludge, cellulose fibers, and polyvinyl alcohol are rich in carbon and can increase water and carbon in the soil with the help of microbes that provide adequate nutrients which in turn help the growth of microorganisms. These microorganisms can reduce the crystallinity of cellulose under anaerobic and aerobic conditions and decompose cellulose into stable small molecules (CO_2 and H_2O) (Tang *et al*. 2010; Zhang *et al*. 2007). Poplar wood fiber primarily contains cellulose, hemicellulose, and lignin. Fungi in the soil can secrete a variety of enzymes that decompose these materials into simple carbohydrates. Based on the interaction of all of these factors, the degradation rate of mulch film can be accelerated (Zhang *et al*. 2012).

Table 5. Contents of Carbon (C%) and Nitrogen (N%) in the Bamboo Pulp Sludge and Soil

Element	Nitrogen (N%)	Carbon (C%)
Bamboo pulp sludge	3.27	35.68
The soil before degradation	0.296	6.272
The soil after degradation	0.242	5.536

Table 6. Quantity Change of Paper Sheeting and PE Film during the Degradation

Degradation Days>>	7		21		35		49	
	Before	After	Before	After	Before	After	Before	After
PE Mulch Film	0.404g	0.405g	0.558g	0.560g	0.890g	0.889g	1.302g	1.303g
Paper Mulch Film	0.402g	0.388g	0.605g	0.386g	0.952g	0.526g	1.275g	0.558g

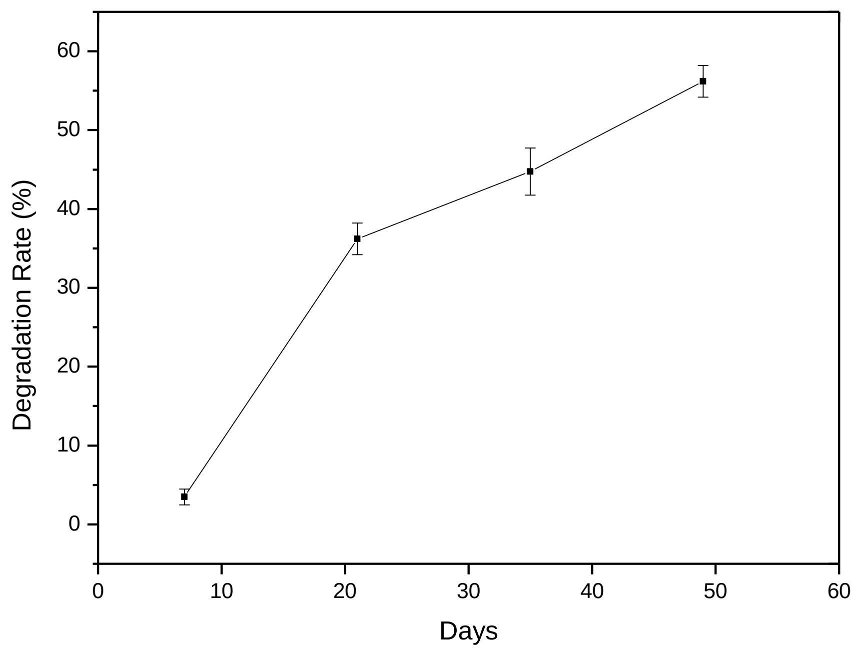

Fig. 6. Degradation change curve of paper sheeting

Morphology and Topography

Figure 7 shows the paper sheeting before degradation, as observed by stereo microscopy at a magnification of 50x. Bamboo paper sludge, poplar wood fibers, and viscose fibers intertwine to form a stable structure. The holes and the gaps between the mulch film fibers were stabilized after brush-application of the PVOH (①) which binds the fibers together. The molecular structure of PVOH contains many hydroxyl groups, which may be superimposed onto hydroxyl groups in the viscose fibers to enhance the strength of the sheeting and provide a smooth surface film (Zhang *et al.* 2012).

The paper sheeting after 21 days and 35 days of degradation is presented in Figs. 8 and 9. It can be seen that the decomposition process of the PVOH took place evenly and the fibers began to degrade in Fig. 8. As shown in Fig. 9, the surface of the paper sheeting began to have small holes. The paper sheeting after 49 days of degradation is presented in Fig. 10. The surface of the sheeting appears uneven, with cracks and holes (②). The surface glue between the fibers appears to have disintegrated and the surface area became larger, which led to the appearance of holes in the sheeting. The holes grew larger with degradation time and the sheeting appears to have a poor structure, with the overall structure of the sheeting damaged. These results demonstrate that the macromolecules in the paper were broken by large numbers of microbes, and the cellulose and polyvinyl alcohol underwent natural degradation (Zhang *et al.* 2012). The function of microbes was randomly assigned to four parts. The surface of the paper sheeting adsorbed microbes, followed by seepage of the glue into the soil. Then the fibers began to swell after absorbing water from the soil (Huang *et al.* 2007). Finally, the structure of the cellulose became fluffy. With the progress of degradation time and the presence of microbes, the internal structure of the paper mulch film became fragile. Finally, the paper mulch film degraded into the soil.

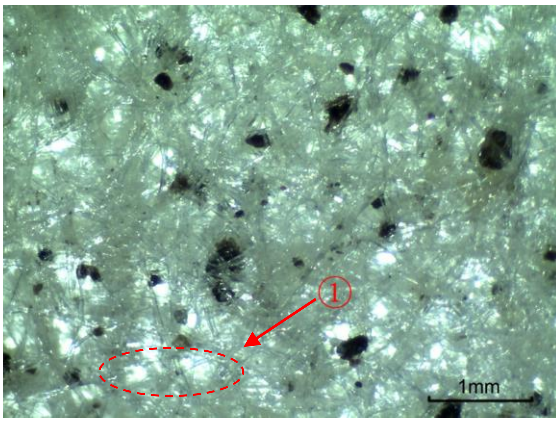

Fig. 7. The paper mulch film before degradation in the soil

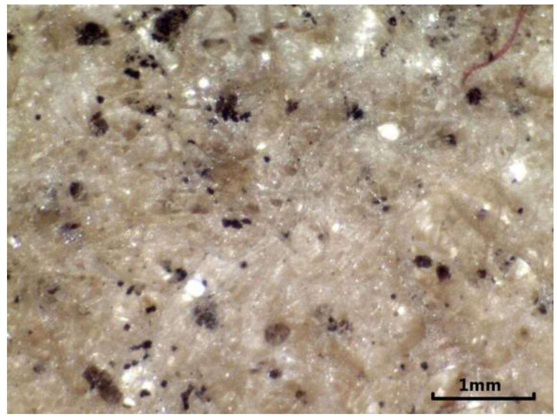

Fig. 8. The paper mulch film after 21 days of degradation

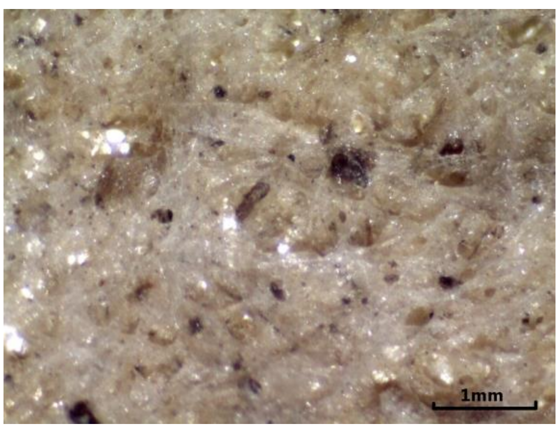

Fig. 9. The paper mulch film after 35 days of degradation

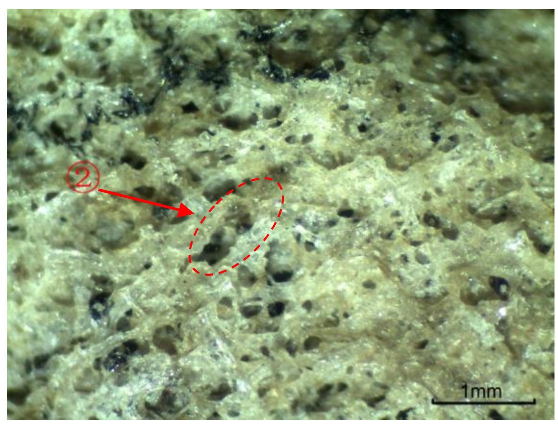

Fig. 10. The paper mulch film after 49 days of degradation

CONCLUSIONS

1. The best process conditions for the paper sheeting for use as a biodegradable agricultural covering included an area density of 30 g/m^2, a bamboo paper sludge content of 10wt%, and a PVOH concentration of 4wt%.

2. The burst strength, tearing resistance, tensile properties, and resistance to water were 220.65 kPa, 60.00 N, 46.10 N, and 153 Pa, respectively, under the best process

conditions. The paper sheeting can satisfy the demand for agricultural sheeting used for promotion of agricultural production.

3. The degradation rate reached 56% when the mulch was buried in the soil for 49 days. The mulch had good degradability and made practical use of bamboo pulp sludge, a solid waste.

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