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**Development of Next Generation Biodegradable Mulch Nonwovens
to Replace Polyethylene Plastic**

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ABSTRACT

The purpose of this research is to develop the next generation of low-cost, biodegradable mulch nonwoven fabrics to replace polyethylene plastic, without the drawbacks of the current biodegradable textile and film options. Plastic mulch offers a number of benefits to farmers, such as weed control, disease protection, and optimization of soil temperature and moisture, but there are a number of emerging economic and environmental issues regarding the use and disposal of polyethylene plastic. The cost of polyethylene mulch is increasing, due to rising petroleum prices, and this trend can only be expected to continue. In addition, polyethylene plastic cannot be tilled into the soil after use and must be extracted from the field. Removal is time consuming, and there are significant labor costs. Once it is removed, the used plastic is typically sent to a landfill, or else it is buried or burned on the farm. Recycling options are limited and sustainable alternatives are needed.

We are developing biodegradable mulch fabrics from renewable resources using polylactic acid (PLA) that is given a treatment to make it truly biodegradable on controlled schedules. The current PLA mulch films that are available are “compostable” under specific conditions, but will generally not biodegrade in soil. PLA is made from lactic acid, a fermentation byproduct derived from corn, wheat, rice, or sugar beets. The PLA mulch nonwovens are designed so that they are durable throughout the growing season, and then rapidly degrade in field with the application of our treatment. By using a mulch fabric that degrades in the field, farmers will save significant labor and disposal costs, help conserve resources, and decrease pollution.

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BACKGROUND

Plastic mulches have been used in horticultural cropping systems since the early 1960's for weed control, disease protection, and optimization of soil temperature (Lamont, 2005), but there are a number of drawbacks with their use. First, the cost of polyethylene mulch is increasing, due to rising petroleum prices, and this trend can only be expected to continue. Second, polyethylene plastic cannot be tilled into the soil after use and must be extracted from the field. Removal can be a time-consuming process, and there are significant labor costs. Third, and most importantly, disposal of the used plastic mulch has become a real problem. There are significant fees charged by landfills that will accept used mulch film, but often this is not even an option, because numerous state and local governments now prohibit agricultural plastics from being buried in landfills.

Many farmers dispose of the plastic by open burning, and some researchers estimate that ~50% of used agricultural plastics are burned on-farm (Levitan, 2005). There are no disposal fees with this method, but the environmental cost is high, because open burning of waste releases high levels of toxic pollutants such as heavy metals, dioxins, and particulates into the air (Lemieux, 1997). And even though open burning is illegal in many areas, the tongue-in-cheek phrase “it doesn't make smoke if you burn it at night” is routinely heard. Controlled incineration or recycling of the used plastic might one day become more viable options, but these alternatives still require that the mulch be removed from the field and transported great distances, generally, at the growers' expense. Many operations now bury or pile the used mulch film on their own land; however, this practice is not sustainable and may also be also restricted in the not too distant future. In Europe, for instance, farmers are fined for piling used agricultural plastic on their farms. This situation is only going to get worse, and the agricultural community is in desperate need of a dependable, sustainable alternative to polyethylene plastic mulch.

A number of alternative mulch materials have been developed in the past, but there have been significant problems with application, durability, and predictability. Photodegradable plastics are unpredictable and incomplete degradation is problematic (Greer and Dole, 2003). Crop canopy can slow or inhibit degradation, and areas that are buried by the soil do not degrade. The remaining material must still be removed by hand, or else small fragments litter the field. Paper mulches have been in use for a long time, but they are difficult to lay and lack reliability. Weber (2003) evaluated biodegradable paper mulches for weed suppression in the establishment year of matted-row strawberries. While the mulches did suppress some weeds, the rate of degradation was too unpredictable. For example, black planters paper quickly degraded where the edges were covered with soil, and large sections were blown away by the wind, whereas other paper was still intact after 26 weeks.

Biodegradable starch-based plastics appear to be an improvement over the paper and photodegradable films, but these materials are expensive and the most widely used product (Mater-Bi[®], Novamont SpA, Novara, Italy) is imported from Italy. Rangarajan and Ingall (2006) calculated that biodegradable plastic costs 3 to 3.5 times more per acre when compared to polyethylene plastic. In addition, unpredictability still haunts the biodegradable plastic mulches. Breakdown of the bioplastics is often very rapid, and does not correlate with film thickness (Rangarajan and Ingall, 2005). These materials also have the reputation of “breaking up, not

down”, and pieces of the mulch are often blown around the farm, creating an eyesore and polluting nearby waterways.

We are developing a dependable, biodegradable mulch fabric from renewable resources using polylactic acid (PLA). PLA is made from lactic acid, a fermentation byproduct derived from corn (*Zea mays*), wheat (*Triticum* spp.), rice (*Oryza sativa*), or sugar beets (*Beta vulgaris*). When polymerized, the lactic acid forms an aliphatic polyester with the dimmer repeat unit shown below:

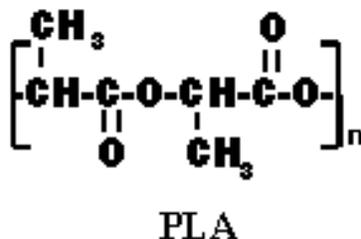


Figure 1. PLA polymer repeat unit

Unlike other synthetic fiber materials with vegetable sources (e.g., cellulose), PLA is well suited for melt spinning into fibers. Compared to the solvent-spinning process required for synthetic cellulosic fibers, melt spinning allows PLA fibers to be made with both lower financial cost and lower environmental cost, and allows the production of fibers with a wider range of properties. Like poly(ethylene terephthalate) polyester (PET), PLA polymer needs to be dried before melting to avoid hydrolysis during melt extrusion, and fibers from both polymers can be drawn (stretched) to develop useful tensile strength. The PLA molecule tends to assume a helical structure which allows it to crystallize readily. Also the lactide dimer occurs in three forms: the L form, which rotates polarized light in a clockwise direction; the D form, which rotates polarized light in a counter-clockwise direction; and the Meso form, which is optically inactive. During polymerization, the relative proportions of these forms can be controlled, resulting in relatively broad control over important polymer properties. The combination of a thermoplastic “natural” fiber polymer, unique polymer morphologies, and control over the isomer content in the polymer enables the manufacturer to engineer a relatively broad spectrum of properties in the fiber (Dugan, 2001 and Khan, et al.1995).

Fiber-grade PLA is not biodegradable in its “as-extruded” state. Instead, it must first be hydrolyzed before it becomes biodegradable. To achieve hydrolysis of PLA at significant levels, both a relative humidity at or above 98% and a temperature at or above 60°C are required. Once these conditions are met, degradation occurs rapidly (Dugan 2001 and Lunt, 2000). However, control over the content and arrangement of the three stereoisomers allows the polymerization of PLA in which the melt temperature can be controlled between about 120°C and 175°C and the low melting temperature polymers are essentially completely amorphous. The more amorphous polymer should be more assessable to enzymes and microbes in the soil and those which are added as treatments for easier biodegradation at ambient soil conditions. This will require working with the PLA manufacturer to produce polymer which is more susceptible to biodegradation, but will produce fabric and film which have the required strength for use in mulching.

The biodegradable fabric mulch we produce will also have a number of advantages over biodegradable and polyethylene plastic mulches. First, the fiber-web structure of the fabric will make it less brittle than bioplastic when it degrades, and fragmentation and unwanted dispersal will be reduced. Second, the mulch fabric will have superior tensile properties, and will be more reliable when it is applied to the soil using mulch-laying equipment. In our experience, biodegradable films have broken off the roll a number of times during application. Third, the fabric will be water permeable and allow the infiltration of rainwater to help maintain soil moisture. Plastic mulches are impermeable to water, and there can be significant runoff of water and agrichemicals after rainfall (Rice et al., 2001). Fourth, microscopic open-spaces between the fibers in the fabric will provide increased accessibility for the enzyme and microbial products and help facilitate degradation when needed.

Polylactic acid has been used to make a number of different products, and factors that control its stability and degradation rate have been well documented. Both the L-lactic acid and D-lactic acid forms produced during fermentation can be used to produce PLA (Hartmann, 1998). The beauty with PLA is that the rate of degradation can be controlled by altering factors such as the proportion of the D and L forms, the molecular weight, or the degree of crystallization (Drumright et al., 2000). For instance, Hartmann (1998) found that unstructured PLA samples will rapidly degrade to lactic acid within weeks, whereas a highly crystalline material can take months to years to fully degrade. This flexibility and control makes PLA a highly advantageous starting material for the production of agricultural mulch fabrics, where the material is intended to be degraded in the field after a specific time period (Drumright et al., 2000).

Polylactic acid is broken down into smaller molecules through a number of different mechanisms, and the final breakdown products are CO₂ and H₂O. Degradation is influenced by temperature, moisture, pH, and enzyme and microbial activity, yet is not affected by ultraviolet light (Drumright et al., 2000; Lunt, 2000). In some early work that evaluated PLA degradation for biomedical applications, Williams (1981) found that **bromelain**, pronase, and proteinase K could accelerate the rate of PLA breakdown. More recently, Hakkarainen et al. (2000) incubated 1.8 mil thick samples of PLA at 86°F in a mixed culture of microorganisms extracted from compost. After 5 weeks of incubation, the compost-treated film had degraded to a fine powder, whereas the untreated control remained intact. It is important to note that this study used only the L form, and degradation rates will differ based on the ratio of the D and L forms. Regardless, the work by Hakkarainen et al. (2000) illustrates that application of a broad range of readily available microorganisms from compost can accelerate breakdown.

Many of these microorganisms already exist in fertile agricultural soils, and the stability of the mulch fabric will need to be tested under a range of soil conditions. To our knowledge a 100% PLA fabric mulch has not yet been tested in the field, and the majority of the research relating to PLA stability has been focused on food packaging and surgical materials (e.g. sutures and staples). It is well known that humidity, temperature, and moisture will influence the rate of PLA breakdown, yet the PLA degradation studies so far were either performed in liquid culture in vitro, or else in active composting operations above 140°F (Drumright et al., 2000; Hakkarainen et al., 2000; Lunt, 2000; Williams, 1981). Degradation was rapid when PLA was composted at 140°F, with nearly 100% biodegradation achieved in 40 days (Drumright et al., 2000), but

stability below 140°F when the fabric is in contact with soil organic matter remains to be determined. Thus to accurately gauge breakdown as it relates to agricultural applications, it will be important to test the fabric mulch on soils with different moisture and organic matter/microbe levels.

Our multi-disciplinary team has considerable expertise with the use of PLA in a variety of applications. Spunlaid and meltblown non-wovens using PLA were first researched by Dr. Larry Wadsworth at the University of Tennessee (Khan et al., 1995). Other team members are evaluating PLA- and starch-based biodegradable films for the production of warm-season vegetables and strawberries grown as annuals. The results from our work should have a remarkable impact on horticultural crop production. The PLA fabric mulch we develop will be a reliable and economically comparable alternative to polyethylene plastic. It has the potential to decrease pollution, conserve natural resources, and decrease labor and disposal costs associated with mulch removal.

EXPERIMENTAL

In an attempt to test the efficacy of a number of different compost, enzyme and microbial treatments to accelerate degradation of the fabric, tests were carried out in greenhouses with a constant overhead misting of 6 x 6 inch pots containing soil with the fabrics placed on top of the soil. We cut 6 in. x 6 in. sections of the nominal 30 g/m² 100% PLA spunbond (SB) fabric and placed them on top of sieved, moist field soil in the pots. The 15-inch wide sample roll of SB PLA used in this study was provided by Oerlikon, Neumag Nonwovens. The compost, enzyme and microbial treatments listed below were applied to the surface of the mulch fabric (3 replicates per treatment) at the levels specified (typically in gallons per acre or tons per acre). After four weeks exposure during August 2008, the specimens were removed and representative specimens were analyzed for fiber breakage and morphological changes by scanning electron microscopy (SEM) and other representative samples were tested for weight, thickness, peak tensile load and peak elongation. Since the treated fabric specimens were rather small for this preliminary study at 6 x 6 in., only two 1 in. x 6 in. specimens in the machine direction (MD) were tested on each treated sample, although six 1 in. x 6 in. specimens were tested in the MD with the non-treated control PLA fabric.

Treatments and Rates:

- Pineapple juice (25 gal/acre)
- Molasses QC (25 gal/acre)
- Biotron Bxi (5 gal/acre)
- Biocat (5 gal/acre)
- Promot MZM (5 gal/acre)
- Biodynamic (5 gal/acre)
- Biocat (5gal/acre)+ Pineapple Juice (PJ) (25 gal/acre)
- Biodynamic (5gal/acre) + PJ (25 gal/acre)
- Compost Tea Catalyst (5 gal/acre) + PJ (25 gal/acre)
- Dairy Manure Compost (10 tons/acre) + PJ (25 gal/acre)
- Biozome (5 gal/acre) + PJ (25 gal/acre)
- Pond Clarifier (5 gal/acre) + PJ (25 gal/acre)

- Biotran Bxi (5 gal/acre) + Molasses (25 gal/acre)
- Promot MZM (5 gal/acre) + Molasses (25 gal/acre)

OBSERVATIONS

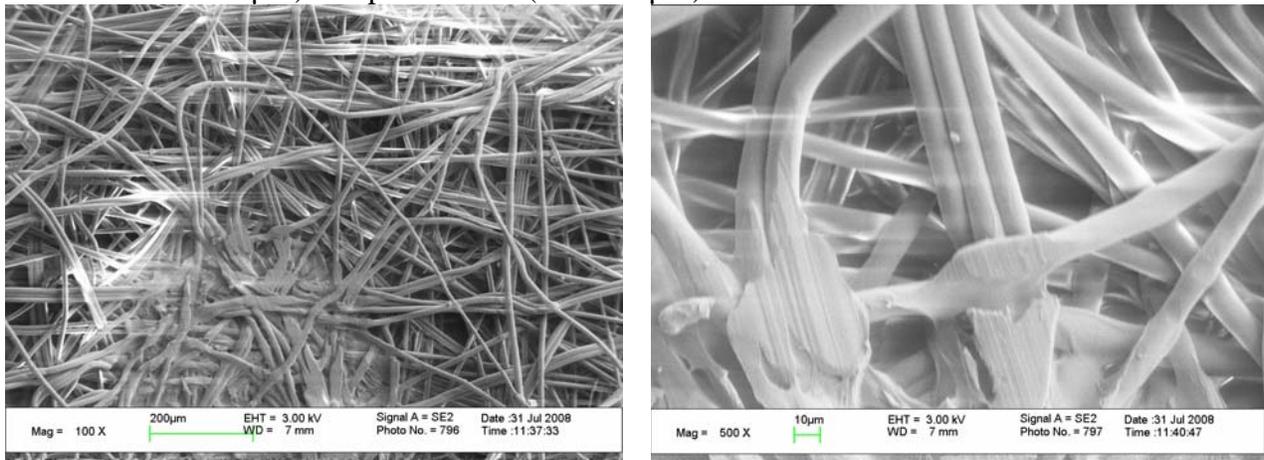
After 4 weeks exposure in the greenhouse, the nominal 30 g/m² (gsm) 100% PLA SB specimens were removed from the pots and examined. It was generally obvious that the specimens showed little degradation and that they should have been exposed for 8 to 12 weeks before testing. Nevertheless to obtain some preliminary data for this paper, representative PLA spunbond specimens were subjected to physical tested and some were examined by SEM. SEM photomicrographs of the control (as-received) SB at 100X, 500X and 1000X ,are shown in Figure 2.

The diamond shaped bonding pattern of the control (as received) 30 gsm SB nonwoven fabric can be seen in Figure 2a at 100X magnification. The fibers are partially melted on the bonded area and the continuous PLA fibers are not bonded between bonded areas. Typically, only about 15% of a SB fabric is bonded in discreet areas. In the upper right hand corner of Figure 2a, part of another bonded area can be seen, and the continuous fibers can be seen to travel from one bonded area to another. The combination of strong, well-drawn fibers (during the SB process) and the continuous filaments which each travel through several bonded areas contribute to the high strength and flexibility of SB fabrics at much lower weights compared to short-fiber (staple fiber) nonwovens and to conventional woven fabrics. In Figure 2b, at 500X, the flattened fibers in the bonded area can be seen and the SB fibers in the non-bonded area are very round, and also at 1000X (Figure 2c), the PLA SB fibers in focus are quite round and have little residue on the surface. The average fiber diameter as determined from SEM analyses of the control SB fabric was 11.4 micrometers (µm). As noted in Table 1, the weight of the non-treated control PLA SB was 29.2 gsm, the thickness was 0.140 mm, the average peak tensile load was 3.83 lbs (determined from 6 specimens) and the peak elongation was 11.1%. The standard deviation of the peak force was 1.05 from the 6 tests with a range of 2.2 to 5.15 lbs.

As shown in Figure 3, the SEM photomicrographs at 100X and 500X of the specimen 2S1, which was exposed to pineapple juice (PJ) for 4 weeks, appear much the same as the corresponding SEMs of the non-treated control fabric, with the exception that particles of soil or residues are visible. In Table the weights and thickness values of the treated fabrics generally appear to be about the same as the control fabric, except some of the higher thickness values may possibly attributed to residue from the soil attached to the specimens. Similar observations are made from the 100x and 500X SEMs in Figures 4 and 5 of the PLA specimen treated with Biocat + PJ and the specimen treated with Promot MZM + molasses, respectively. Residue particles on the surface of the treated fibers are magnified in SEM photomicrograph at 1000X (Figure 5c).

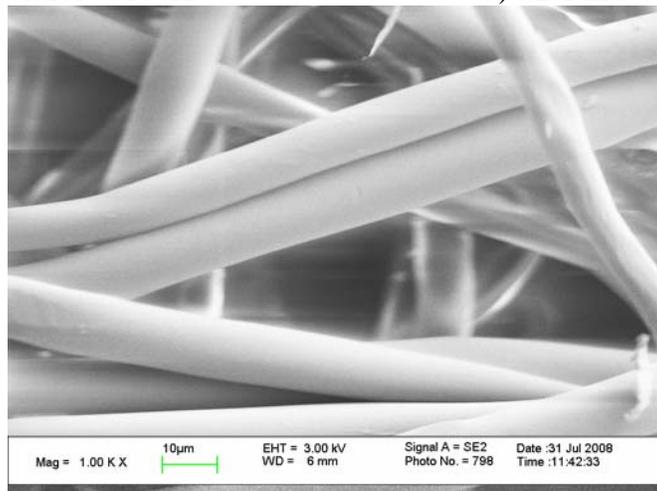
In Table I each average peak tensile load shown represents the mean of only one, or two or three specimens. Thus the peak force of the pineapple juice treatment was the average of only two tests of one specimen (3S2), the Biotron Bxi treatment was the average of two tests on each of two specimens (4S2 and 4S3), and the average peak force for the Biocat treatment was the average of two tests on each of three specimens (5S1, 5S2, and 5S3), etc. For a good statistical comparison of means, typically six tests are conducted on six 1in. x 6in. test strips in both the

machine direction (MD) and in the cross machine direction (CD) of the fabric to be tested. However, the small swatches of PLA fabrics treated in this preliminary study were much too small to allow enough testing for making sound statistical comparisons. Only the non-treated control PLA SB was evaluated with six tensile tests in the MD, and as was noted above, the standard deviation was 1.05 with the range of peak force values obtained being from 2.2 to 5.15 lbs. In the future the specimens treated should be a minimum of 12 in. wide and 24 in. long. Also the level of treatments should probably be at least doubled and the treatment time increased to 10 to 12 weeks. In addition to accelerate any biodegradation in the soil, one-half of the treated specimens should be buried one inch below the soil, although mulches are typically placed on top of the ground and only covered with soil on the edges to hold the material down during windy conditions. Nevertheless, there is an indication that most of the treatments may have reduced the strength of the 100% PLA SB and more extensive study seems warranted. In the next study, 30 and 60 gsm 100% PLA nonwoven fabrics made by both SB and meltblown (MB) processes will be evaluated and each specimen treated will be at least 12 in. x 15in. to enable sufficient physical testing and characterization. Although MB fabrics are much weaker than SB fabric, MB fiber diameters are even smaller and should be more accessible to degradation (mean diameters of 2 to 6 μm) compared to SB (10 to 15 μm).



a) SB PLA Control at 100X

b) SB PLA Control at 500X

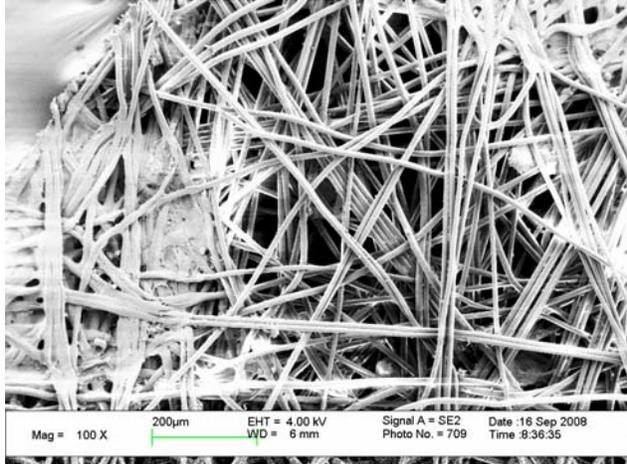


c) SB PLA Control at 1000X

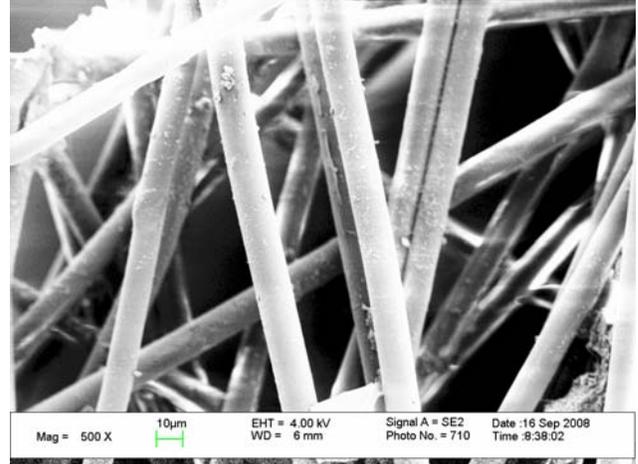
Figure 2. SEMs of As-Received Control 100% PLA Spunbond fabric at 100X, 500X and 1000X.

Table 1. Effects of treatments after four weeks exposure on weight, thickness, peak force and peak elongation of spunbond PLA fabric.

Treatment	Spec No.	Weight (g/m ²)	Thickness (mm)	Peak Force (Lbs)		Peak Elongation (%)
				Avg	S. Dev.	
No Treatment		29.2	0.140	3.83	1.05	11.1
Pineapple Juice	2S1	30.0	0.171	2.33	-	8.6
Molasses	3S2	31.0	0.148	2.78	-	11.4
Biotron Bxi	4S2, 4S3	31.2	0.178	2.97	0.35	11.6
Biocat	5S1, 5S2, 5S3	30.9	0.214	2.55	0.53	12.5
Promot MZM	6S1, 6S2, 6S3	30.1	0.234	2.79	0.29	14.4
Biodynamic	7S3	32.0	0.201	3.5	-	12.9
Biocat + Pineapple Juice (PJ)	11S1, 11S2, 11S3	31.8	0.243	3.07	0.76	11.9
Biodynamic + PJ	13S1, 13S2, 13S3	30.5	0.178	2.82	0.73	11.2
Compost Tea Cat + PJ	14S1, 14S2	29.4	0.203	2.60	0.12	12.2
Dairy Manure Compost + PJ	15S1, 15S3	27.0	0.141	2.74	0.14	14.0
Biozome + PJ	16S1, 16S3	32.0	0.197	2.94	0.13	11.5
Pond Clarifier + PJ	17S1, 17S2, 17S3	30.0	0.258	2.67	0.25	11.3
Biotran Bxi + Molasses	18S1, 18S2, 18S3	30.5	0.185	2.27	0.47	15.3
Biocat + Molasses	19S1, 19S2, 19S3	29.5	0.167	2.44	0.54	11.3
Promot MZM + Molasses	20S1, 20S2, 20S3	30.2	0.230	2.54	0.14	12.0

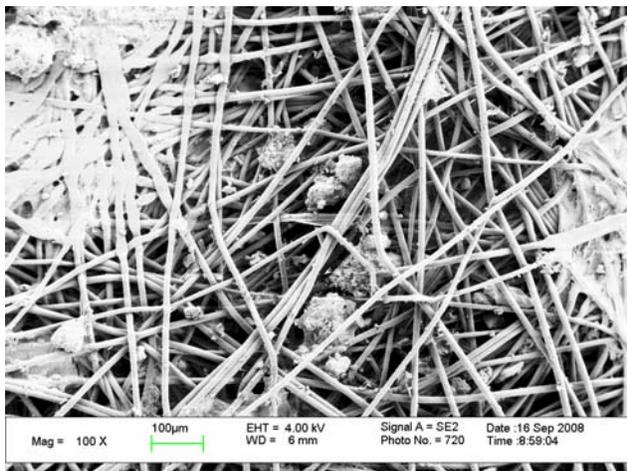


a) SB Specimen 2S1 (PJ) at 100X

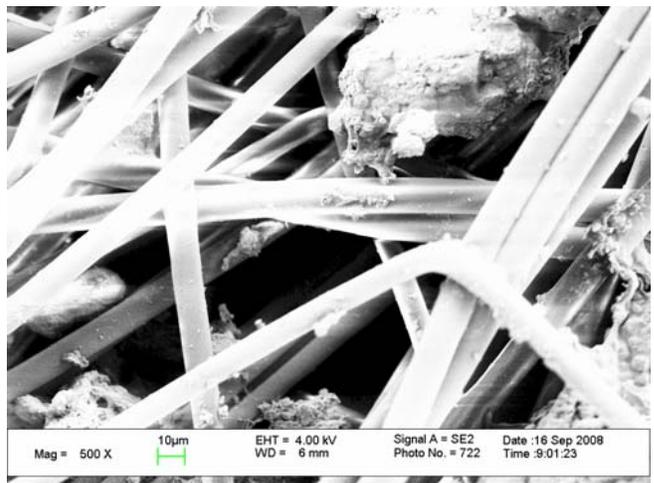


b) SB Specimen 2S1 (PJ) at 500X

Figure 3. SEM photomicrographs of PLA Specimen 2S1 at 100X and 500X after 4 weeks exposure to Pineapple Juice (PJ) on top of soil.

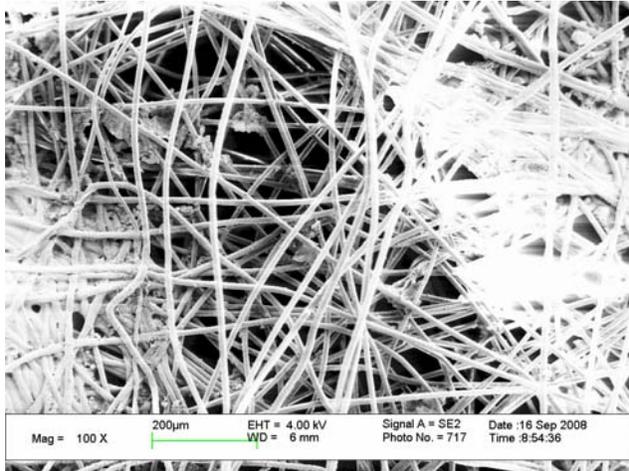


a) SEM of 11S2 (Biocat + PJ) at 100X

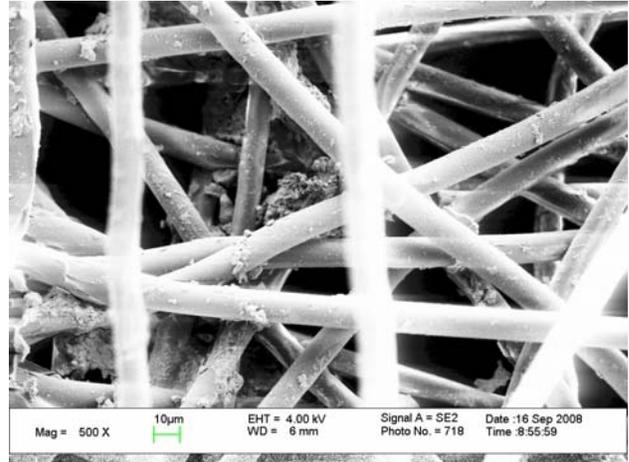


b) SEM of 11S2 (Biocat + PJ) at 500X

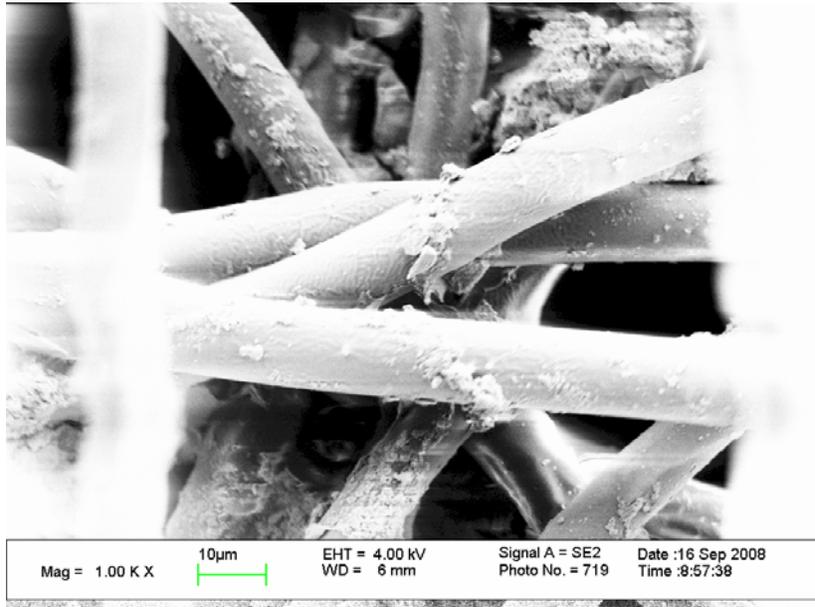
Figure 4. SEM photomicrographs of PLA Specimen 11S2 at 100X and 500X after 4 weeks exposure to Biocat + PJ on top of soil.



a) SEM of 20S3 (Promot MZM + Molasses) at 100X



b) SEM of 20S3 (Promot MZM + Molasses) at 500X



c) SEM of 20S3 (Promot MZM + Molasses) at 1000X

Figure 5. SEM photomicrographs of PLA Specimen 20S3 at 100X , 500X and 1000X after 4 weeks exposure to Biocat + PJ on top of soil.

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