EFFECT OF PLASMA POLYMERIZATION ON LYOCELL FABRIC/PLA COMPOSITES

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1 General Introduction

Today, composites are used as the materials for aircraft, automotive, architectural interior and other structural and functional applications. Generally polymer matrix based on fossil fuel is utilized for fabrication of these composites. Scarcity and environmental problems related to the use of these petroleum-based plastics make it crucial to develop alternative materials that are biodegradable. Composites with bioplastics constituents are looked into as the alternatives. However, poor interfacial adhesion between these bioplastics matrix and the reinforcement fibers leads to poor mechanical strength, poor durability and also rapid degradation when in contact with moisture. Plasma polymer coating may be considered as a solution for these shortcomings of biocomposites. In this study, plasma coating modifies the hydrophilic lyocell fabric into a hydrophobic surface. The hydrophobic surface of this cellulose based fabric is expected to enhance the interfacial adhesion with the bioplastics matrix, polylactic acid (PLA) in particular, which in turn increases the mechanical strength of the biocomposite and increases its ability to resist moisture. In order to study and optimize the effect of plasma polymer coating on Lyocell fabric for the biocomposite, mechanical testing, SEM microstructural observations and evaluation on the moisture resistance property of Lyocell fabric/PLA composite are carried out.

2 Experimental

2.1 Materials

Polylactic Acid (PLA) used was from Green Chemical, GC9000R, and has specific weight of ~ 1.24 g/cm^3, diameter of ~3 mm and melting point of 110^{o}C. Lyocell fabric (Yu Kwang, Acetate Taffeta) used was classified as semisynthetic fiber of cellulose system. Acrylic acid monomer was procured from Junsei Chemicals. The organofunctional silanes were from Dow Chemicals.

2.2 Plasma polymerization

The distance between electrodes was 10mm. Helium carrier gas containing monomer filled the plasma chamber (Fig. 1). Investigation was conducted to determine optimal conditions to generate plasma glow. Four types of electrode size (width of 10, 15, 20, and 25mm) were studied and the corresponding optimal plasma conditions for plasma polymerization on the fabric (area of 2m^2) were determined.

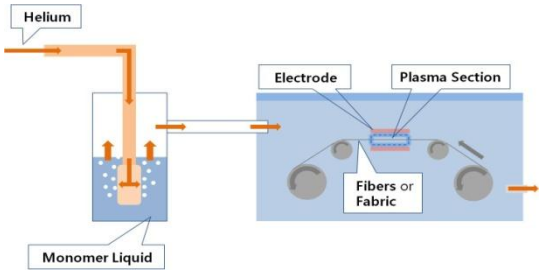


Fig.1. Plasma polymerization schematic

2.3 Silane Treatments

Three types of silane coupling agents, i.e. aminoethylaminopropyltrimethoxysilane (APS), 3-methacryloxypropyltrimethoxysilane (MPS), and 3-glycidoxy-propyltrimethoxysilane (GPS) were used respetively. Silane treatment on the fabric was conducted by process depicted in Fig. 2. In summary, the silane was mixed into a solution containing methanol and distilled water with composition of 9:1,

previously acidified to pH of 4.0-4.5 through acetic acid addition. Then, hydrolysis was promoted by stirring for 5 minutes. After the fabric was dipped into the silane solution for 10 minutes, it was washed with distilled water and dried in a vacuum oven for 24 hours.

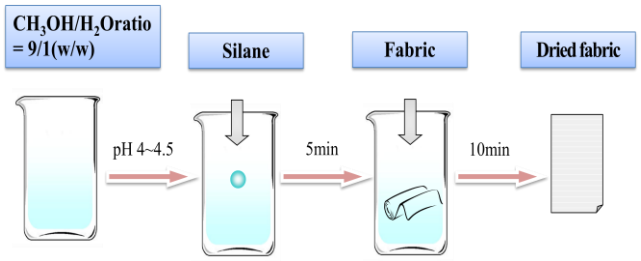


Fig.2. Silane treatment schematic

2.4 Specimen preparation

Lyocell fabric and PLA films were alternatively stacked before the fabrication in a hot press at 140℃ (a value determined from differential scanning calorimeter analysis of neat PLA) and 49.05KPa pressure for 5 minutes (Fig. 3)

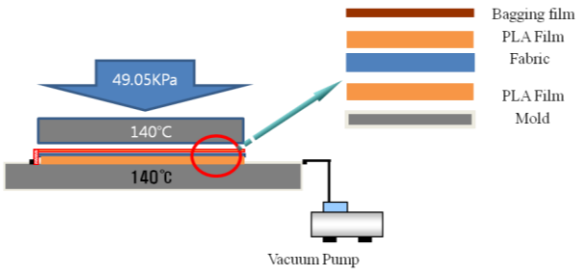


Fig.3. Lyocell fabric/ PLA composite manufacturing process

2.5 Characterization and testing

Tensile test was conducted according to ASTM D3039 in a controlled ambient condition. Selected samples were further characterized using scanning electron microscope (SEM). Evaluation of moisture absorption of plasma polymerized composites was also conducted according to ASTM D5229.

3 Results and Discussion

3.1 Plasma conditions determination

The particular optimal conditions for electrodes of different sizes were determined (Table 1). The results are used for the settings on subsequent plasma polymerization.

Table 1. Optimal conditions to generate plasma and Plasma polymerization on fabric ($2m^2$)

Electrode size (mm)	Frequency (kHz)	Voltage (kV)	Time (min)
10	10	3.5	32
15	5	2.3	21
20	5	3.4	16
25	5	2.7	13

Tensile testing on plasma polymerized lyocell fabric/PLA composites using optimal plasma conditions for each electrode size was conducted. The composites were of similar tensile strength of 30-32MPa (Fig. 4). This shows consistency of plasma polymerization. In addition, the strength was higher than that of non treated composite (tensile strength of 22.91MPa). This indicates, that the larger electrode size provides faster plasma polymerization.

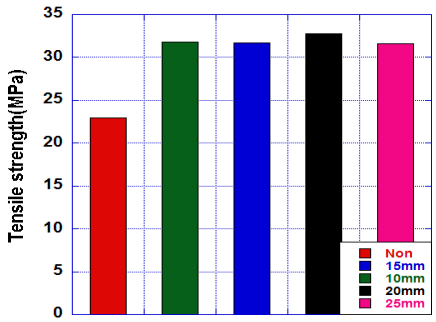


Fig.4. Size of the electrode mechanical strength results

3.2 Evaluation of moisture absorbance

Non treated composite showed the highest tendency of moisture absorption. Overall, the plasma polymerized composite was showing the least moisture absorption.

Plasma polymerization on the fiber affected the composite's moisture absorption behavior. The

finding is in agreement with analysis on previous experiments on other natural fibers which reported that the plasma polymerization helped reducing the water absorption tendency of natural fiber composites [6].

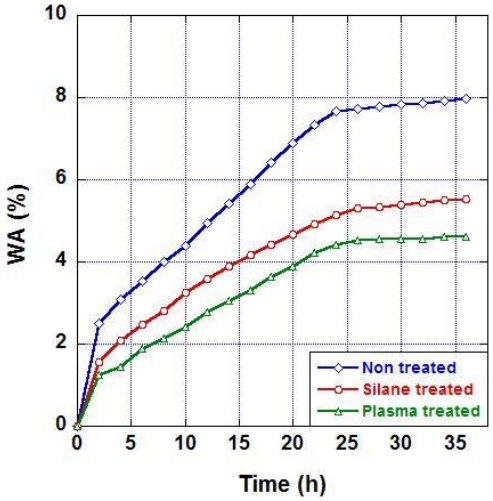


Fig.5. Water adsorption

3.2 Strength of plasma polymerized lyocell fabric/PLA composites

Results of tensile test on the silane treated and plasma polymerized composites are displayed on Fig. 6, including the strength of non treated one. It was found that there is increase in tensile strength of the composites as the results of silane treatment and plasma polymerization on the lyocell fabrics (44.04% and 52.77%) increase, respectively, compared to non treated composite). The improved strength of silane treated lyocell fabric/PLA composite was likely due to reaction between the OH group in PLA of the sizing materials and the OH group in silane treated fabric. The acrylic acid precursor used for plasma polymerization caused the surface of lyocell fabric more hydrophylic.

Further analysis was conducted to determine the effect of plasma polymerization time to the strength of composite. It was found that there is optimum plasma polymerization time (Fig. 7), where longer polymerization time no further increased strength of the corresponding composite.

Observation on the composite's fractured surface indicates adhesion improvement on silane treated and plasma polymerized lyocell fibers and the PLA matrix (Fig. 8). The silane treatment and plasma polymerization improved the compatibility of the lyocell with PLA matrix and led to increased composites' strength.

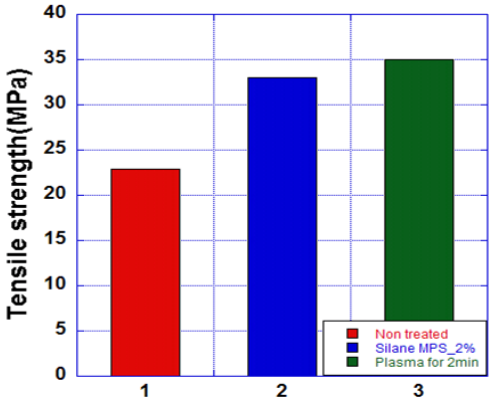


Fig.6. Tensile strength of Lyocell/PLA composites

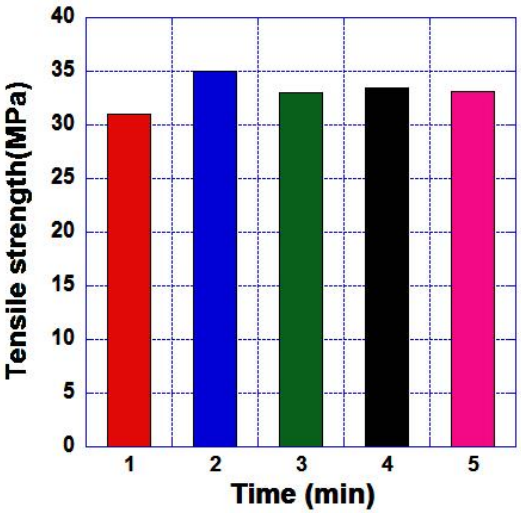
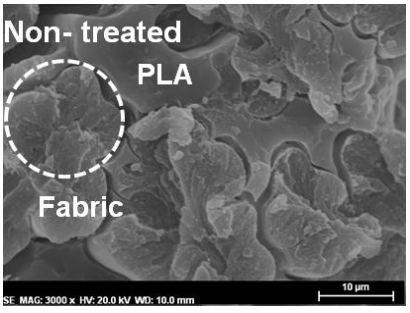


Fig.7. Mechanical properties of plasma polymeri-zation time



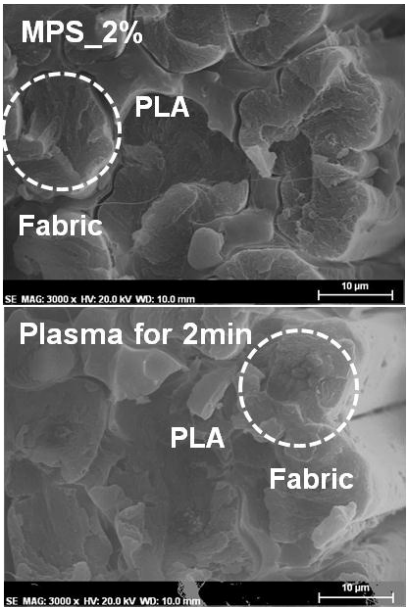


Fig.8. SEM images of Lyocell fabric/PLA composites' fracture surfaces
(a) Non-treated
(b) Silane MPS_2%
(b) plasma polymerized for 2 minutes.

4 Conclusions

Plasma polymerization conditions on lyocell fabric were determined for four types of electrode size. The change of electrode size from 10mm to 25mm helped reducing the process time of up to 59.37%. The plasma polymerization modified the fabric surface, leading to increased strength of the composite. Plasma polymerization reduced the tendency of moisture absorption of the composite.

References

[1] H. Takagi and BS Kim, "Inherent Multi-Functions of Green Composites," CJJCC-9, China, Sept., 2010

[2] J. B. Moon, "Influence of Silane Coupling Agent Treatments on Physical Properties of Rubbery Materials", Elastmer, Vol.36, No.4, 2001, pp.237-245

[3] S. J. Park, J. S. Jin, J. R. Lee, Y. K. Kim, "Effect of Silane Coupling Agent Treatment on Interfacial Adhesion of Glass Fiber-reinforced Composites", J. Korean Ind. Eng. Chem. Vol.11, No.3, 2000, pp.285-289

[4] Gellerstedt F, Gatenholm P, Surface properties of lignocellulosic fibers bearing carboxylic group, Cellulose 1999; 6(2):103-121

[5] H. Yasuda, Plasma Polymerization, Academic Press, Orlando, Florida, 1985

[6] BS Kim, MN Mguyen, BS Hwang and S Lee "Effect of Plasma treatment on the mechanical properties of natural fiber/PP composites" HPSM IV, P159-166, V97, 2008.