Vol 22, No 1 (2015): Journal of Faculty of Engineering & Technology (JFET) [available online]

Synthesis & Characterization of cotton fiber reinforced starch/PVA biodegradable composite films

Wasif Razzaq[*†], Atif Javaid[*]

Abstract

The focus of this work is to study the effect of the cotton fibers reinforcement on the starch/ Poly (vinyl alcohol) (PVA) blend films. The biodegradable composite films were prepared by starch, PVA and natural cotton fibers using solution casting method. The starch/PVA/cotton fiber films samples were analyzed for the tensile, chemical and morphological properties. Experimental results showed that the incorporation of the short cotton fibers into the starch/PVA matrix caused a reasonable improvement in of the tensile modulus (18.6 times), tensile strength (2.07 to 4.38MPa), elongation (104.56 to 8.82%) and impact strength (28.4%). Chemical and structure analysis were done by Fourier Transform Infrared Spectroscopy (FTIR) and surface morphology were studied by optical microscope. The obtained results showed that addition of cotton fibers enhanced both the mechanical and surface properties of the starch/PVA blend film

Keywords: Bio degradable, Composites, Starch, Poly(vinyl alcohol), Cotton fiber, Mechanical characterization

[*] *Department of Polymer and Process Engineering, University of Engineering and Technology, G. T. Road, Lahore 54890, Pakistan

[†] **Corresponding Author.** Email Address**:** wasifrazzaq@hotmail.com.
Tel: 03214263273

1 Introduction

In last two decades there has been an enormous increase in the preparation of the food and commodity packaging plastics products. The use of the polyolefin based polymers caused significant environmental issues. Since the waste generated from such polymers take up thousands of years to degrade so it causes the environmental pollution [1].Moreover in case of the food packaging the migration of the chemicals have made the food contaminated and may cause the risk of the consumers safety [2]. In order to find out the solution of the petroleum based plastic waste, many researchers had done a lot of work to produce the renewable environmental friendly biodegradable products which have comparable properties and lower cost [3, 4, 5].

From the family of renewable natural polymers, starch is one of the attractive polymer because of its huge availability, low cost and has the potential to produce from many renewable resources [6]. Starch is a carbohydrate polymer that can be produce from variety of resources such as potato, corn, maize, rice and wheat [7].The composition of starch is based on the linear and branched chain molecule. It consist of amylose, a linear molecule having molecular weight between 103 to 106 and amylopectin which is a branched structure molecule [8]. Structural studies of native starch indicated that it has granular structure with crystallinity ranges from 15 to 45% from different resources [9]. One of the disadvantage of the starch is its poor mechanical properties which restrict its application as compare with other petroleum based polymers [10].

In order to improve the mechanical properties of the starch, numerous attempts have been made to blend starch with other synthetic biodegradable polymers. PVA is one of the excellent synthetic biodegradable polymer having good mechanical properties [11]. PVA is a water soluble polymer which produced by the hydrolysis of the polyvinyl acetate. PVA molecules have the -OH bonds in their backbone which made it biodegradable [12]. Because of the biodegradability, solubility in water and excellent film forming abilities, PVA has increasing blend with starch to optimize the mechanical and biodegradable properties of the composite films [13].

Since both starch and PVA having crystalline nature due to which it is not easy to made blend of starch and PVA so their processing have made easier by addition of plasticizer which act as gelatinization agent [14, 15].Different studies have been made for the plasticization of the starch and PVA with different plasticizers such as water, glycerol and sorbitol and among all these plasticizer the best results was observed with glycerol as plasticizer [16, 17, 18].

From environmental prospect, the use of the natural fibers gaining popularity in biodegradable products because these fibers are easily producible with low cost, environmental friendly and have comparable properties with conventional materials [19, 20]. Among different natural fibers, cotton fibers are most widely used natural vegetable fibers. They excessively used in order to produce apparel home furnishing and automotive products [21]. Cotton fibers are eco-friendly, biodegradable and light weight fibers with reasonable strength [22]. Different studies have been reported to use cotton fibers as good biodegradable reinforcing material [23, 24].

Starch/PVA composites films have large applications as packaging and agricultures films. Moreover control drug delivery systems and blood carrying bags are also important applications of starch/PVA films in the medical field [25].

2 Experimental

2.1 Materials

Laboratory grade starch soluble was bought from the BDH Company. PVA (Lab grade with degree of hydrolysis ≥ 98% and ash content ≤ 1.0% and Laboratory grade Glycerin was purchased from the Merck Chemicals. Cotton fibers was taken from the National Textile University, Faisalabad, Pakistan.

2.2 Preparation of the composite film

Films were prepared by solution casting method. Starch and PVA was taken in the form of solid powder and cotton fibers were cut into short fibers as much as possible by manual chopping. Solution of PVA and starch (50:50) was prepared by adding 6g PVA and 6g starch in 150ml water in a beaker and set the beaker on water bath with continuous stirring at 80°C. Chopped cotton fibers were added with 2, 4, 6, 8 and 10% by weight in the solution at the start of the mixing. After thirty minutes, glycerin was added as plasticizer and further mix the solution for 10 minutes and then the homogenous mixture was poured on flat glass plate which was already placed on balanced surface to get uniform thickness of the film. Film was dried for 48 hours in closed place at ambient condition and after that it cured in oven at 40°C for 30min. Finally the film peeled off form the plate and then use for the characterization.

2.3 Mechanical Testing

Universal testing Machine of Model TIRA-2810 was used for characterization of tensile properties of the composite film. Tensile strength, tensile modulus and elongation properties were measured using ASTM D882. Sample strips were cut according to the ASTM dimension and the cross head speed was adjusted at 0.5mm/min. For each concentration five samples cut from film which were tested and then mean values was reported in results.

2.4 Impact Testing

Impact testing was done to find out the mean failure energy for at different concentration. Impact testing was done using ASTM 1709-03. Square shape samples were cut in the dimension indicated by ASTM and adjusted in the clamps of impact tester. Drop weight of impact tester has uniform load of 2lb. Mean failure energy was calculated using ASTM-5628 with formula

$$\text{Mean Failure Energy} = w*h*f$$

Where

w = drop weight

h= Mean failure height

f = factor of conversion into joules = 9.80665×10^{-3}

2.5 Fourier Transform Infrared spectroscopy

FTIR of JASCO with model FT/IR-4100 was used in order to characterize the chemical composition of the composite film. Standard source of light was utilized and reference pellets of KBr was used. Wave number range was adjusted from 400 to 4000cm^{-1}. Sample was prepared by mixing approx.7g specimen powder with KBr and formed the pellet using hydraulic pressure

2.6 Morphological properties

Morphology of the surface was studied using optical microscope. Specimen images at different magnification e.g. 5x, 10x was taken.

3 Results and discussions

3.1 Tensile Modulus

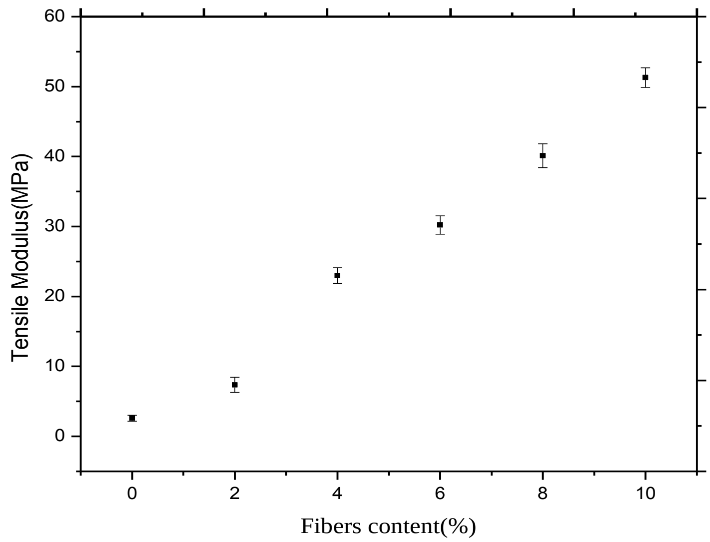


Figure 1 Effect of cotton fiber content on Tensile Modulus of starch/PVA film

Effect of the cotton fibers on the tensile modulus of the starch/PVA blend film was shown in Fig 1. Cotton fibers was used as reinforcement in the starch/PVA blend film and it was observed that the tensile modulus of PVA/starch/cotton fibers composite films increases as the concentration of fibers increases from 2 to 10%. The increase in the tensile modulus is from 2.6 to 51.2 MPa for maximum level of loading. A reasonable increase in the tensile modulus was due to the better adhesion of the cotton fibers with the matrix interface of the starch and PVA blend because of the similar chemical nature of the starch, PVA and cotton fibers [26, 27]

3.2 Tensile Strength and Elongation

Figure 2 indicates the effect on the tensile strength (MPa) and elongation at break (%) of the starch/PVA/cotton fibers composite films by varying the content of cotton fibers and it was found that tensile strength of the starch/PVA blend was improved from 2.1MPa to 4.4MPa for incorporation of 6% cotton fibers and tensile strength decreases on further addition of cotton fibers

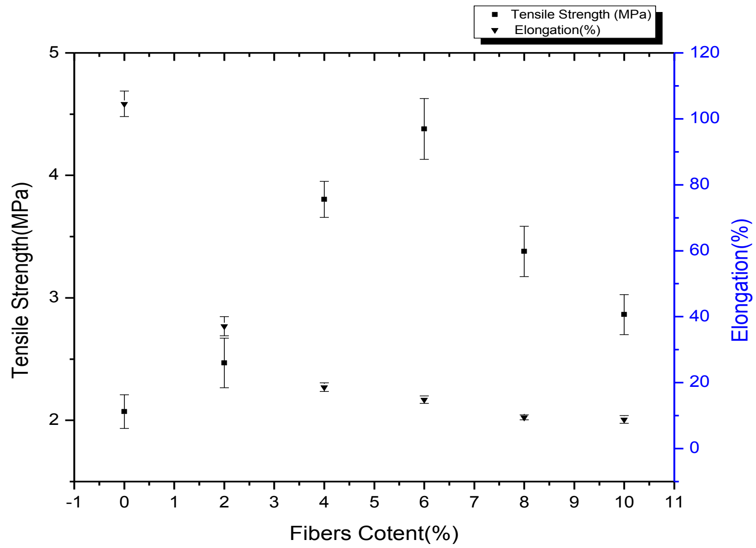


Figure 2 Effect of cotton fibers content on Tensile strength (MPa) and Elongation (%) of starch/PVA film

This increase in the tensile strength may be due to phase compatibility and similar chemical structure of the cotton fibers reinforcement with matrix phase which cause proper adhesion of two phases and helps to transfers load to fibers easily and strengthen the structure of the film [26]. On the other hand decrease in tensile strength was observed when the fibers concentration increases from 6 to 10%. This decrease may be due the reason that high concentrations of the cotton fibers can produce micro cracking in the films due to its brittle nature which reduces the strength of the film [28].

It was also observed that elongation at break decreases from 104.56 to 8.82% (10.9 times) with increasing contents of the cotton fibers. This was due to the reason that cotton fibers have large cellulose contents which makes fibers structure very brittle and stiff and when these fibers were added into the film increases the stiffness of the film and hence decrease elongation.

3.3 Impact Strength

Toughness or impact strength of the material actually depend upon the interfacial strength and inter-laminar structure of the composite films [29]. Figure 3 shows the results of the starch/PVA films with different cotton fibers loading and found an increase in 26% of the means failure energy for 10% fibers loading. The continuous increase in mean failure energy indicates that fibers have proper interfacial association with the matrix, gives reasonable support to the matrix and acts as stress transferring medium [30]

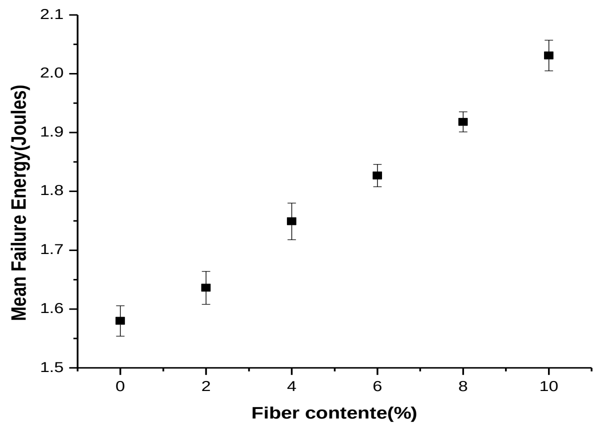


Figure 3 Impact strength of the Starch/PVA composite film at different cotton fibers content

3.4 FTIR Analysis

The FTIR result of the starch/PVA composite filled with cotton fibers shown in the figure 4. There was a large transmittance found at the peak 11 with wavenumber 3416.28cm^{-1} which is due to the -OH vibration stretching present in starch [31]. A slight transmittance was observed at peal 10 with wavenumber 2929.34cm^{-1} which arises due to the C-H group present in the starch and PVA [32]. Second large absorption was observed at peak 8 having wavenumber 1628.59cm^{-1} and this was due to the carbonyl C=O group stretch. CH_2 symmetric bending of PVA was observed at peak 7 with wavenumber 1454.06cm^{-1} [33]. Transmittance of 1249.65 cm^{-1} may be due to variations in CH mainly present in the PVA and also a little starch. Finally, the number of peaks around 600-1050 cm^{-1} due to the stretching of CO and OH groups, and this can be seen in the spectra of most materials [34]. Transmittances at different wavenumbers was found to be slight higher than the true value of the pure groups. The reason for this slight increase was may be the growth of the bonding among starch, PVA, glycerol and the cotton fibers. These bonding create better adhesion which may cause the wavenumber to increase.

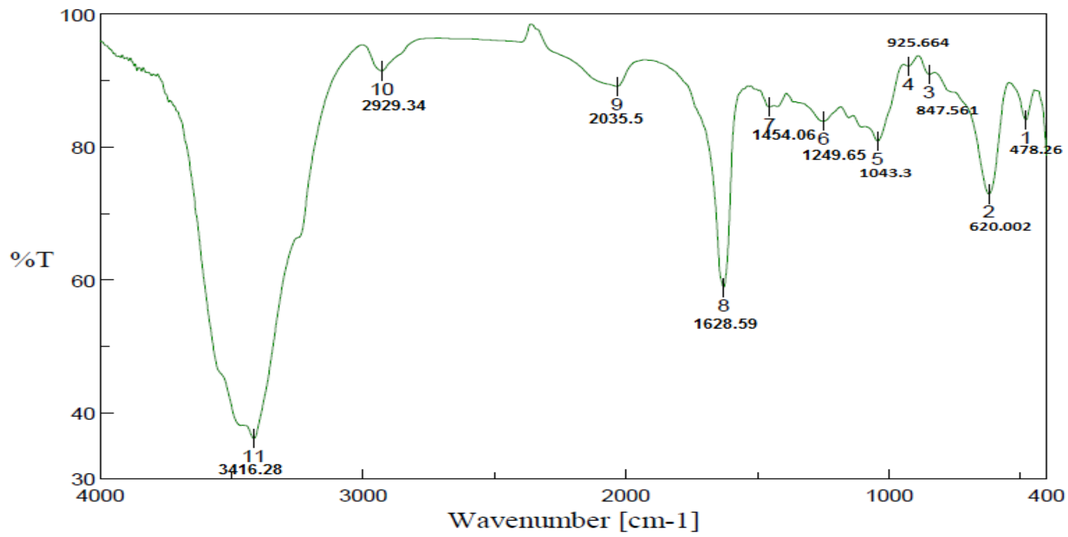
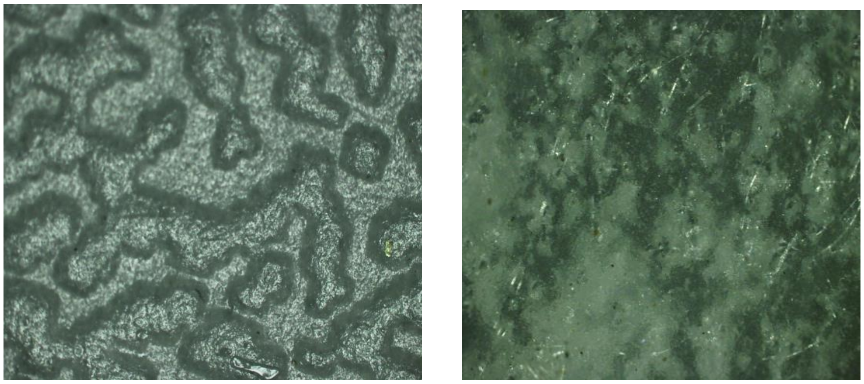


Figure 4 . FTIR spectral curve for the transmittance of starch/PVA composite film

3.5 Morphological Properties

Morphological properties of starch / PVA composite films were examined using an optical microscope image. Figures 5(a,b) show the appearance of the surface of the composite films at 10x magnification and gave us an overview of changes in the surface appearance of PVA / starch films reinforced with cotton fibers. Surface of the starch / PVA composite films were not very smooth. The figure 5(a) represents an optical image of starch and PVA film without a fibrous reinforcement and can be clearly seen that the starch and PVA are present in the form of dispersions, and this may be due to the reason that the PVA and starch were not fully soluble in each other and therefore when PVA concentration reaches up to 50% in blend dispersion start to appear at the surface. Figure 5(b) showing the optical image of the surface of PVA/starch composite film reinforced with the cotton fibers. It was observed that smoothness in the surface slightly better for this case and fibers are spread smoothly into the materials. This may be due to the better bond compatibility of the cotton fibers within the composite films and this increase the solubility of starch/PVA and gives less dispersion



(a) (b)

Fig 5 (a) Optical image of starch/PVA film, (b) optical image of the starch/PVA film reinforced with cotton fibers

Conclusion

PVA/Starch films reinforced with cotton fibers were produce using solution casting method with glycerin as plasticizer. Addition of cotton fibers increases the tensile modulus (2.61 to 51.29 MPa) and tensile strength (2.07 to 4.38MPa) of the PVA/Starch blend film. Optimum condition of loading regarding tensile strength was observed at 6% after which tensile strength start to decrease but tensile modulus still increase. Improvement in the impact (26%) and elongation (10.9 times) properties were also observed with the addition of cotton fibers. FTIR analysis of the starch/PVA composite films verified the strong interactions of the OH groups of the starch, PVA and cotton fibers. Mechanical and chemicals improvements were observed as a result of good interaction of the starch/PVA matrix with cotton fibers due to similar chemical structure. These results endorse the recommendation that cotton fibers are good reinforcement for the starch/PVA matrix and result in the form of a good biodegradable composite.

References

[1] Azahari N. A. , Othman N. and Ismail H., *Journal of Physical Science*, **22,** 15(2011).

[2] Barnes K. A., Sinclair R., and Watson D.*Chemical migration and food contact materials*, Woodhead Publishing, *122(*2007).

[3] Bertuzzi M. A., Armada M. & Gottifredi J. C., *Journal of Food Engineering,* **82**, 17(2007).

[4] Larotonda F. D. S., Matsui K. N., Sobral P. J. A. & Laurindo J. B., *Journal of Food Engineering*, **71**, 394(2005).

[5] Mali S., Sakanaka L. S., Yamashita F. & Grossmann M. V. E, *Carbohydrate Polymers,* **60**, 283(2005).

[6] Tang S., Zou P., Xiong H. & Tang H., *Carbohydrate Polymer,* **3**, 521(2007).

[7] Dufrense A. & Vignon M. R, *Macromolecules,* **31**, 2693(1998).

[8] Forssell P. M., Mikkilä J. M., Moates G. K. & Parker R., *Carbohydrate Polymers,* **34**, 275(1997).

[9] Forssell P. M., Mikkilä J. M., Moates G. K. & Parker R., *Carbohydrate Polymers,* **34**, 275(1997).

[10] Funke U., Bergthaller W. & Lindhauer M. G., *Polymer Degradation and stability,* **59**, 293(1998).

[11] Huang X. & Netravali A., *Composite Science Technology,* **69**, 1009(2009).

[12] Pavol A., Darina K., Miroslav K., Dusan B. & Barbora S. , *Polymer Degradation and stability*, **3**, 413(2002).

[13] Xuegang L., Jiwei L., Xiaoyan L., *Carbohydrate Polymers,* **90**,1595(2012).

[14] Chen P., Yu L., Simon G. P., Liu X. X., Dean K., & Chen L., *Carbohydrate polymers,* **83**, 1975(2011).

[15] Liu P., Xie F. W., Li M., Liu X. X., Yu L., Halley P. J. et al, *Carbohydrate Polymers,* **85**, 180(2011).

[16] Kalichevsky M.T. and Blanshard J.M.V, *Carbohydrate Polymers*, **20**, 107(1993).

[17] Ma X., Yu J. and Kennedy J.F., *Carbohydrate Polymers*, **62**, 19(2005).

[18] Teixeira E.D.M., Pasquini D., Curvelo A.A.S., Corradini E.,Belgacem M.N. and Dufresne A., *Carbohydrate Polymers,* **78**, 422(2009).

[19] Bavan D.S., Mohankumar G.C, *Hindwai Publishing Corporation. Journal of Engineering*, 1(2013).

[20] Rijswijk K. V.,Brouwer W.D., Beukers A.,, *"Application of Natural Fiber Composites in the Development of Rural Societies".*

[21] Shahedifar V., Rezadoust A.M., *Journal of Reinforced Plastics and Composites*, **3**, 681(2013).

[22] Vijaykumar C. & Piyush P.G., *International Journal Metallurgical & Materials,* **3**, 15(2013).

[23] Muller D.H., Krobjilowski A., Mussig J., in *National Cotton Council of 01-13, National Cotton Council of*, USA 2001-01-12–2001-01-13, 2001.

[24] Jorg Mussig, *Journal of Polymer and Environment*, **16**, 94(2008).

[25] Kunhua Yao, Jie Cai, Miao Liu, Yan Yu, Hanguo Xiong, Shanwen Tang, Shiyong Ding, *Carbohydare Polymer, Elsevier,* 86, 1784(2011)

[26] Prachayawarakorn J., Sangnitidej P.,Boonpasith P., *Carbohydrate Polymers,* **81**, 425(2010).

[27] Amnuay W., Katavut P., Supranee K., Pichan S.,and Claudio M., *Songklanakarin Journal of Science and Technology*, **4**, 461(2011).

[28] Alomayri T., Shaikh F.U.A. , Low I.M.,*Elsevier,* **50**, 1(2013).

[29] Mishra S., Mohanty A. K., Drzal L. T., Misra M., Parija S., Nayak S. K., Tripathy S. S, *Composites Science and Technology,* **63**, 1377(2003).

[30] Zhong J.B., Lv J. Wei C., *Express polymer letters*, **1**, 681(2007).

[31] Cao X., Chen Y., Chang P., Muir A., Falk G., *Express Polymer*, **2**, 502(2008).

[32] Khalil H., Ismail H., Rozman H., Ahmad M., *European Polymer*, **37**, 1037(2001).

[33] Le Troedec M., Sedan D., Peyratout C. et al, *Composite Part A*, **39**, 514(2008).

[34] Nacos M, Katapodis P, Pappas C. et al, *Carbohydrate polymers*, **66**, 126(2006).