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## Effect of Barley Husk Fibre on the Mechanical and Barrier Properties of Polyvinyl Alcohol Biodegradable Films

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### Abstract

Poly vinyl alcohol (PVA) composite films were prepared using barley husk fibre (BHF) with different loadings of 5, 10, 15, 20 and 25 wt %. The composite films were subjected to mechanical properties (tensile strength, % elongation and Young's modulus) and barrier properties (Water vapour transmission rate and water vapour permeability). All the films were prepared using solution casting method. It was found that the mechanical properties of the composite films were strongly dependent on the loading of BHF in PVA matrix. PVA/BHF composite films showed better mechanical and barrier properties at lower BHF loading (up to 10 wt %). The highest tensile strength observed was 28 MPa with BHF loading of 10 wt %. Morphology of the composite films revealed that the surface become rough at higher BHF loading. Based on the biodegradability and low cost of natural fillers, the produced composite films may offer a very promising bio-composite material for packaging applications.

**Keywords** - Mechanical properties, Barrier Properties, Filler loading, Barley husk fibre, Poly vinyl alcohol.

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### Introduction

Now a days, a great deal of attention has been paid to the application of biodegradable polymers, such as starch, lignin, cellulose, chitin and its derivatives to replace the conventional non- biodegradable petroleum-based plastics that cause serious environmental problems due to its non-biodegradability (Zuchowska *et al.*, 1998). For a better alternative, it is required to make blends of synthetic polymers with degradable polymers. Generally synthetic polymers are blended with starch (Okaya *et al.*, 1992; Fanta *et al.*, 1992; Shibata *et al.*, 2002) or natural low cost fillers (Yang *et al.*, 2004; Digabel *et al.*, 2004; Chiellini *et al.*, 2004) to make them environmentally friendly. Polyvinyl alcohol (PVA) is one of a few completely biodegradable synthetic polymers (Dweiri *et al.*, 2004). It is prepared by hydrolysis of polyvinyl acetate. But, the cost of PVA is a limiting factor for application into the market, where PVA has to compete with low-cost synthetic thermoplastic materials such as polyethylene, polypropylene, poly (vinyl chloride), in packaging applications. Therefore, polymers containing natural fillers such as barley husk fibre (BHF) are of great interest because it is low cost and renewable. BHF is a lignocellulosic agro waste which contains 39 % cellulose and can be used to reinforce polymers. The presence of lignocellulosic fillers in the composites enhanced the degradability of the resultant composite. In this work, PVA based biodegradable composite films were prepared with lignocellulosic agro waste and (Ramaraj *et al.*, 1994) properties such as tensile, elongation and Young's modulus, water vapour permeability and water vapour transmission were evaluated. These studies were performed to assess film properties for packaging applications.

### Experimental

#### Materials

Poly vinyl alcohol (average molecular weight of 115000, degree of hydrolysis 80–90%, ash 0.75%),

calcium chloride, and sodium chloride were purchased from LobaCheme Limited, Mumbai (India). Barley was obtained from the local market of Jalandhar.

#### *Pre treatment of barley husk*

Barley was mildly ground to separate the BHF. The separated BHF was washed 2-3 times with double distilled to remove the dust and other water soluble impurities. Finally, the BHF was ground to 0.5-1 mm in size.

#### *Preparation of composite films*

PVA was made into solution by dissolving known amount PVA in hot water. This was blended with 5, 10, 15, 20 and 25 wt % of barley husk fibre (BHF). The solid content of PVA and BHF were made into a homogeneous solution by constant stirring at a temperature of approximately 80–90 °C for 30 min. Thereafter the solution was poured into a glass Petri dish. After drying the solution at 45 °C for 24 hours, the films were removed from the Petri dishes.

#### *Mechanical properties*

Dumbbell specimens (Type IV) were cut from the film according to ASTM D638. The tensile tests were carried out by using a Tensile Tester EZ 20 kN Lloyd instrument at a strain rate of 20 mm/min. The specimens were conditioned at 25 °C for 2 days before testing. Five replicates were tested for each sample, and the results were averaged to obtain mean values and standard deviations.

#### *Water vapour transmission rate (WVTR) and water vapour permeability (WVP)*

ASTM method E96-80 was used to measure WVTR and WVP of the films. Each film sample was sealed over a circular opening (Area=0.002826 m<sup>2</sup>) in a permeation cell that was conditioned at 25 °C in a desiccators. Anhydrous calcium chloride (0% RH) was placed inside the cell while a saturated sodium chloride solution (75% RH) was placed in the desiccators. Due to the vapour pressure gradient across the film, the water vapour continuously diffuses through the film and the water vapour transmission rate can be determined from the weight gained by the permeation cell. The change in the weight of the cell was measured every 24 h over a week.

The change in the weight of the cell was plotted as a function of time and the slope of each line was calculated by linear regression. The water vapour transmission rate (WVTR) was calculated from the slope (g/s) and the cell area (m<sup>2</sup>). After these permeation tests, film thickness was measured and the WVP (g·Pa<sup>-1</sup>h<sup>-1</sup> m<sup>-1</sup>) was calculated. WVTR and WVP are determined using equation (1) and equation (2).

$$\text{WVTR} = \frac{1}{A} \left( \frac{\Delta W}{t} \right) \quad (1)$$

$$\text{WVP} = \frac{\text{WVTR} \times x}{P \times (R_1 - R_2)} \quad (2)$$

Here,  $\Delta W$  is the weight change of the cell (g);  $t$  is the time (h);  $A$  is the test area (m<sup>2</sup>);  $P$  is the saturation vapour pressure of water (3.169 × 10<sup>3</sup> Pa at 25 °C);  $R_1$  and  $R_2$  are the relative humidity (RH) values in the desiccators and the permeation cell (expressed in fraction). Here,  $x$  is the thickness of film (m).

#### *Scanning electron microscopy (SEM)*

Effect of modification on the surface morphology of natural PVA/BHF composite films was conducted by using JEOL JSM-6100 (Tokyo, Japan) scanning electron microscope. Samples were gold plated and observed at a magnification range of 200 to 4000 X.

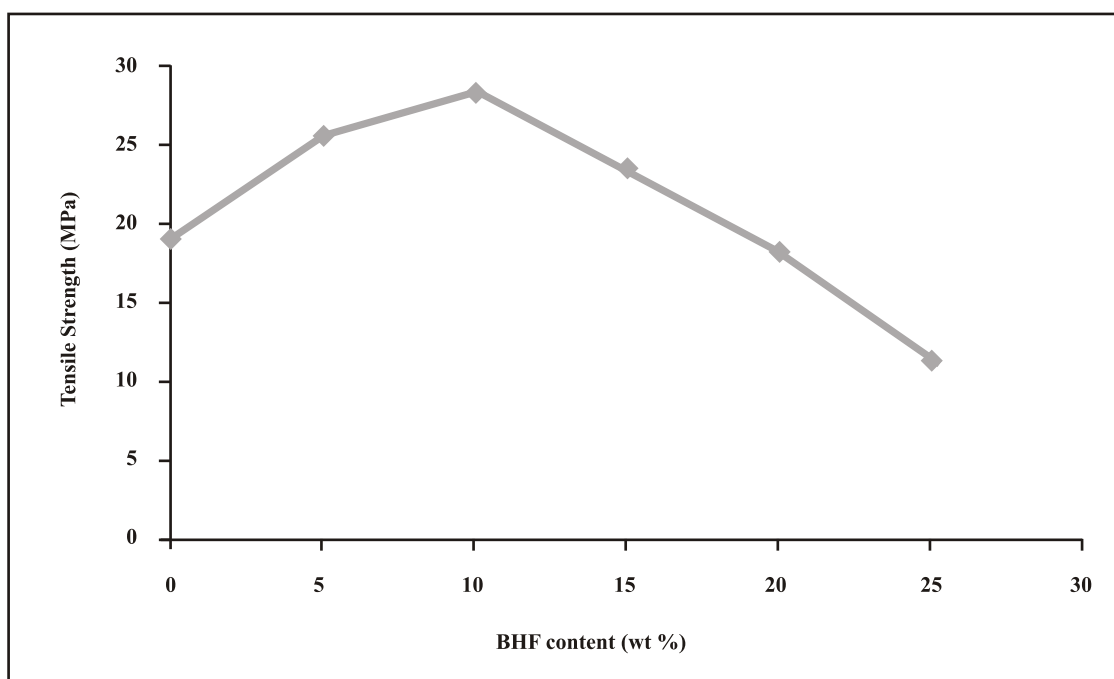
## Results and Discussion

Different composite films were prepared with poly vinyl alcohol matrix, reinforced with barley husk fibre with loading. Tensile strength, % elongation and Young's modulus, water vapour transmission rate and water vapour permeability were evaluated.

### Mechanical properties

#### *Effect of BHF loading on tensile strength*

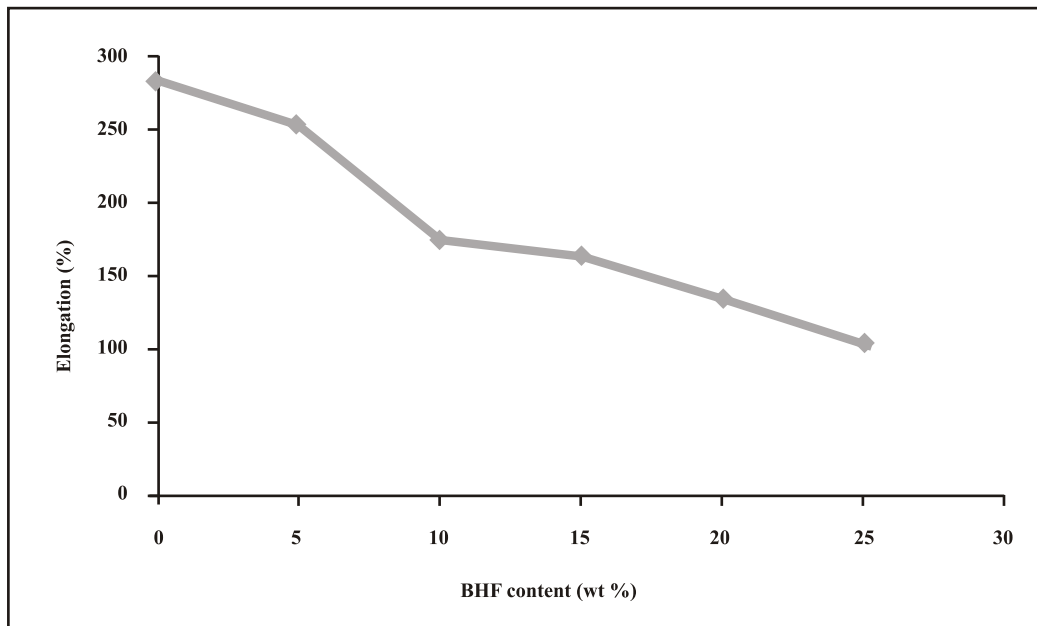
The mechanical properties of a natural fibre composite depend on the orientation and distribution of the fibres or fillers in the matrix. Figure 1 shows the tensile strength versus BHF loading graph for PVA reinforced with BHF fillers. The tensile strength for pure PVA was 19 MPa. It was observed that the tensile strength increased by 32 % with increase in BHF loading from 0 wt % to 10 wt %. The gradual increase in tensile strength could be attributed to the strengthening between the interface of matrix and filler materials. This could only happen when there is proper miscibility between BHF and the PVA matrix (Ramaraj et al., 2006; Sapalidis et al., 2007). The proper miscibility result into less voids and so better stress transfer. As the BHF loading increases, there is an increase in interfacial area, this increase in interfacial area result into poor interfacial bonding and inefficient stress transfer between filler and matrix polymer. The poor bonding resulted into lower tensile properties.



**Figure 1:** Effect of BHF loading on the tensile strength of the PVA/BHF composite films.

#### *Effect of BHF loading on % elongation*

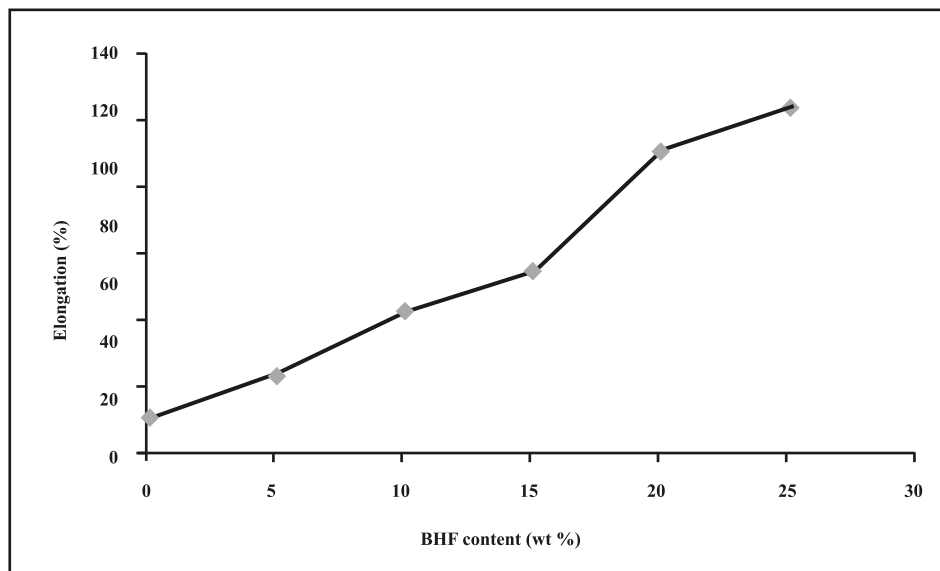
Figure 2 shows the effect of BHF loading on the % elongation of composite films. It is evident from figure 2 that as the BHF loading increased from 5 wt % to 25 wt %, the elongation decreased from 243 % to 98 %. Neat poly vinyl alcohol had elongation of 271 % but elongation decreased with BHF loading. Elongation at break decreases with the increase in loading because higher BHF loading leads easy crack initiation (Ku et al., 2011).



**Figure 2 :** Effect of BHF loading on the % elongation of the PVA/BHF composite films.

*Effect of BHF loading on Young's modulus*

Figure 3 shows that with increase in BHF loading from 5 wt % to 25 wt %, Young's modulus increased. Neat PVA has a Young's modulus of 19.62 Mpa but with the addition of BHF the Young's modulus increased. The increase in Young's modulus as the BHF content increased was an indication that BHF had more stiffness than PVA matrix (Chiellini *et al.*, 2001).



**Figure 3 :** Effect of BHF loading on the Young's modulus of the PVA/BHF composite films.

*Water vapour transmission rate (WVTR) and water vapour permeability (WVP)*

WVTR is the measure of the ease of moisture to penetrate and pass through the material. WVTR of the neat PVA film and the PVA/BHF films were calculated and are presented in Table 1. The WVTR ranges

between 3.538 and 6.21 ( $10^{-1} \text{ g/m}^2\text{h}$ ) and is significantly affected by the addition of BHF. The WVTR increased due to the high sensitivity of the BHF towards moisture. The moisture could act as a plasticizer, which causes greater mobility of the BHF and PVA. Thus, it will increase the interchain distance and facilitate the transport of water molecules through the films. The WVP of the films also showed the same trend (Table 2). The WVP of the composite films increased with increase in the content of BHF. This was due to the hydrophilic nature of the BHF which favours the water absorption. Hence, WVP increased with BHF loading.

**Table 1:** Water vapour transmission rate for PVA/BHF composite films.

PVA (wt %)	BHF (wt %)	WVP ( $10^{-1} \text{ gm}^2\text{h}^{-1}$ )
100	0	3.53
95	5	3.99
90	10	4.43
85	15	4.83
80	20	5.57
75	25	6.21

**Table 2:** Water vapour permeability for PVA/BHF composite fil.

PVA (wt %)	BHF (wt %)	WVP ( $10^{-7} \text{ g pa}^{-1} \text{ h}^{-1} \text{ m}^{-1}$ )
100	0	0.52
95	5	0.58
90	10	0.65
85	15	0.71
80	20	0.82
75	25	0.91

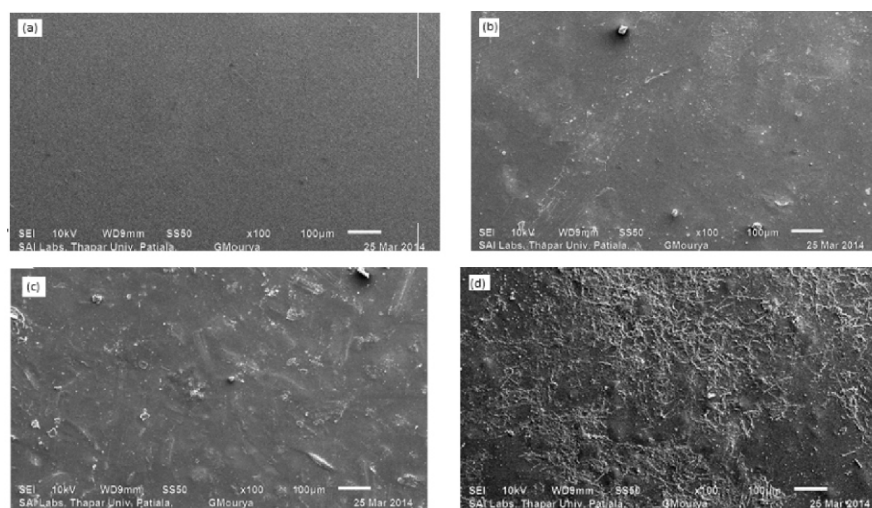
#### Scanning electron microscopy (SEM)

The surface morphology of PVA/BHF composite films was investigated by SEM and is shown in Figure 4. Surface micrograph of pure PVA showed smooth and uniform surface (Figure 4a). The micrograph of composite film reinforced with 5 wt % of BHF loading also showed homogenous surface as BHF particles were uniformly dispersed in the matrix (Figure 4b). But, with increase in BH loading from 5 wt % to 25 wt %, improper dispersion and agglomeration of BHF particles appeared within PVA matrix (Figure 4(c-d)). The surface of the composite films reinforced with BHF loading (15 wt % and 25 wt %) demonstrated aggregation of BHF and matrix (Figure 4(c-d)). This was due to improper adhesion between the BHF particles and PVA matrix at higher particle size and higher loading of BHF.

#### Conclusion

The effect of BHF loading on the mechanical and barrier properties of the PVA/BHF composite films were studied. The result showed that higher mechanical and barrier properties were obtained with smaller BHF loading and vice versa. The PVA/BHF composite films showed enhancement in tensile strength and Young's modulus. SEM analysis showed rough surface with higher BHF loading. So, natural fillers can be used as an effective reinforcement for 9 polymeric films. In general, PVA/BHF composite films could be considered as a potential source of low cost alternative for packaging applications.





**Figure 4 :** SEM images of the PVA composite films; (a) neat PVA ; (b) 5 wt % BHF, (c) 15 wt % BHF, (d) 25 wt % BHF.

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