

TENSILE AND BIODEGRADABLE PROPERTIES OF EXTRUDED SORGHUM FLOUR FILLED HIGH DENSITY POLYETHYLENE FILMS

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ABSTRACT

The objective of this study was to produce sorghum flour (SF) filled high density polyethylene (HDPE) films through extrusion process. Maleic anhydride-graft-polyethylene (MA-g-PE) was used as a compatibilizer to improve the interfacial bonding. Tensile properties for both compatibilized and uncompatibilized films were investigated. The extruded films were also subjected to degradation studies with the agencies of exposure to soil microorganisms and immersion in water. Tensile strength and elongation at break of the films decreased with increase in SF content whereas the Young's modulus increased with increase in SF content. However, on addition of MA-g-PE, all these properties improved indicating good interfacial adhesion between the SF and HDPE. Biodegradation studies showed that water uptake, and weight loss increased with SF content as well as immersion and burial time, and was found to reduce on addition of MA-g-PE. These results support the application of bio-films as environmentally friendly materials.

Keywords: Sorghum flour, high density polyethylene, maleic anhydride-graft-polyethylene, tensile properties, water absorption, soil burial

INTRODUCTION

The numerous applications of synthetic polymers in almost all areas of human endeavours are due to their versatility. This versatility arises from the fact that these polymers are made from low cost petrochemical feed stocks and their ease of processing into various products (Arutchelvi et al., 2008). Moreover, these polymers possess functional properties such as good tensile and tear strength, good barrier properties to oxygen and aromatic compounds and heat seal ability (Tharanathan, 2003). These property attributes have triggered increased production and utilization of these polymers on daily basis. However, these attributes have been overshadowed by their non-degradable nature which leads to their accumulation in the environment. They have become a serious source of environmental pollution affecting both plants and animal lives since not easily degraded by microorganisms.

The quest for sustainable and environmentally friendly materials necessitated the development of biodegradable polymers from renewable resources. Again, discussions on climate changes in the recent times have stressed the need for clean environment and utilization of renewable resources thus minimizing the dependence on dwindling natural resources such as wood and oil (Kesfur, 2010). In this regard, biodegradable polymers with controllable lifetimes have been considered as promising alternatives to non-degradable ones (Albertson, 1980; Albertson et al., 1994).

Starch is a biopolymer of great interest and is obtained from renewable resources. It has many advantages which include abundant supply, environmental friendliness and inexpensive (Takashi, 1986). Starch has different proportions of amylose and amylopectin depending on the source from about 10-20 % amylose and 80-90 % amylopectin (Ramesh et al., 1999). Its

hydrophilicity allows the internal interaction and morphology of starch to be readily changed by water molecules thus affecting the dimensional and mechanical properties. However, the hydrophilicity of starch can be used to improve the degradation rate of some hydrophobic polymers. Starch by itself is poor in processability with its end products having poor dimensional stability and mechanical properties (Choi et al., 1999). One of the ways to overcome these limitations is by blending starch with synthetic polymers such as polyethylene, polylactic acid, polypropylene (St-Pierre et al., 1997).

Combining a biological polymer such as starch, protein, or lipids and a hydrocarbon polymer such as polyethylene, polypropylene raises the issue of how biodegradation will proceed in the environment (Obuz et al., 2001).

Sorghum is a cereal crop grown for food and non-food applications. It is the fourth most widely used cultivated cereal grain crop after wheat, rice and maize. Starch from later cereal crops have be found useful in the blending of synthetic polymers. Sorghum flour (SF) is used greatly as a feed ingredient. The plant is a raw material for feed, food, fuel and industry. It can be separated into a number of different kernel and straw fractions for various industrial applications such as particle board, paper, adhesives, detergents, sugars and plastics through solvent-cast process (Obuz et al., 2001). To our knowledge, the production and use of sorghum flour films through extrusion pathway has not been adequately reported in this area.

The objective of this study therefore was to investigate the production of sorghum flour filled polyethylene films through extrusion process and evaluate the tensile and biodegradable properties of the resultant starch/PE composites.

MATERIAL AND METHODS

Materials

Grain Sorghum seeds were bought from Dawanau International Grains Market Kano Nigeria. The grains were soaked, and washed thoroughly with distilled water to remove impurities and finally sun dried. These grains were pulverized subsequently to fine powder, sieved to 300 μm mesh size to obtain to obtain sorghum flour (SF). The sorghum flour was oven dried at 80° C for 12 h and stored in desiccators to ward off from further moisture contamination.

High density polyethylene (HDPE) granules were purchased from Indorama Petrochemical Company, Ltd, Eleme Rivers State, Nigeria. The density of HDPE is 0.946 g/cm^3 and has a melt flow index (MFI) of 6 $\text{g}/10$ min. Maleic anhydride- graft- polyethylene (MA-g-PE) been used as the compatibilizing agent. It is a product of Sigma-Aldrich Chemicals, Germany and was used as received.

Preparation of Blend Sample

Laboratory size Co-rotating twin extruder Haake, Model Rhemix CTW 100P was used to compound SF and HDPE. The screw speed was 40 rpm and temperature range varied from 130 to 150°C. Filler level content of 0-25 wt. % was used in the blends. The blends were placed in a hot press preheating chamber at 150°C for 10 min followed by compression for about 4 min at the same temperature. Samples were manually removed from the moulds after cooling.

In another set of sample preparation, SF, HDPE and MA-g-PE were measured, fed into the extruder and processed as described above. MA-g-PE used was 5 wt.% based on filler loading.

Tensile Properties

Tensile properties were investigated according to ASTM D882 in a universal testing machine (Shimadzu series) using blend samples obtained. Conditioning and measuring of tensile properties were performed at the same room temperature, $23\pm 5^\circ\text{C}$ at a rate of 50 mm/min and average values of tensile strength, elongation at break and modulus were determined.

Water Absorption Study

Test samples of SF/HDPE blends of various formulations were cut into 2.0 cm x 4.0 cm. Each sample was weighed, dried in an oven at 80°C for 24 h prior to immersion in distilled water at room temperature. The water absorption was investigated by weighing the samples at regular intervals. The surfaces of the blends were wiped dry with filter paper, re-weighed to the nearest 0.001g and calculated for the percentage of water absorption. Results were recorded every week for 9 weeks. The percentage of water absorption was calculated using;

$$\% M_t = [(W_s - W_o)/W_o] \times 100 \dots\dots (1)$$

Where, M_t = total water absorption, W_s = weight of sample after immersion in water, W_o = original weight of sample before immersion.

Soil Burial Test

Kaur and Guatam (2010) method of soil burial test was employed. SF/HDPE blend samples of definite dimensions (2.5 cm x 5.0 cm) were placed under soil surface to a depth of 15 cm. The P^H and temperature of the soil were maintained at 7 and $32\pm 5^\circ\text{C}$ respectively. Samples were removed, washed with distilled water and then dried over night in an oven at 80°C before weighing. The test was conducted every 7 days for a period of 9 weeks.

RESULTS AND DISCUSSION

Tensile Properties

A biodegradable polymer must withstand normal stress encountered during its application. Tensile tests were conducted to investigate the tensile properties of both uncompatibilized and compatibilized SF/HDPE and CSF/HDPE films respectively. Figures 1-3 show the effect of filler concentration on the tensile strength, elongation at break and modulus of the films. The tensile strengths of SF/HDPE and CSF/HDPE films are shown in Figure 1. It can be seen that the tensile strength of the neat HDPE was approximately 20 MPa; however the tensile strength of HDPE decreased by 13 % on addition of 5 wt. % sorghum filler. This observation suggests that the filler is non-reinforcing filler. This is in agreement with the claims of Billmeyer (1971), who stated that non-reinforcing fillers produced reducing effect on the strength of polymer by weakening the rigidity of the polymers.

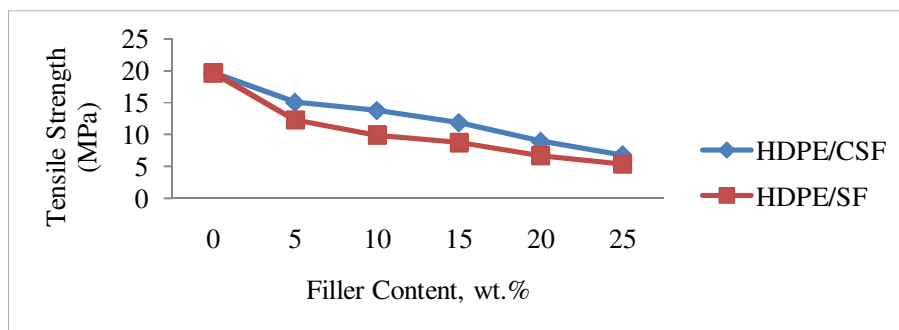


Figure 1. Plot of Tensile Strength versus Filler Content of HDPE/SF Blends

This behaviour has been described in similar studies and has been explained by the increase of the interfacial area with filler content (Piva et al., 2004; Lee et al., 2004). As this area increases, the worsening bond between them decreases the tensile strength. It is observed from the results that with increasing filler content, the reducing effect became more pronounced at 25 wt. % filler content. It implies that the effect of sorghum filler reduces the strength of HDPE instead of improving it.

The relationship between the tensile strength and the filler content in the presence of the compatibilizing agent is also shown in Figure 1. Incorporation of 5 wt. % MA-g-PE based on filler content showed an increase in the strength of the SF/HDPE films for all filler content, indicating an improved positive stress transfer at the interface, showing an increment of 25 % in tensile strength. The improvement of tensile strength would be due to the chain entanglement between HDPE and polyethylene part of MA-g-PE molecule and strong ester linkage formed between maleic anhydride part of MA-g-PE and hydrophilic sorghum filler so that the incompatible part between the hydrophilic sorghum starch and hydrophilic HDPE could be linked and strengthened

Figure 2 shows the effect of sorghum filler on elongation at break of the SF/HDPE films. The elongation at break of the filled polymer decreased progressively with increase in filler content when compared with the neat HDPE. It was seen that on addition of 5 wt. % sorghum filler, the elongation at break of the neat polymer decreased by 40 %.

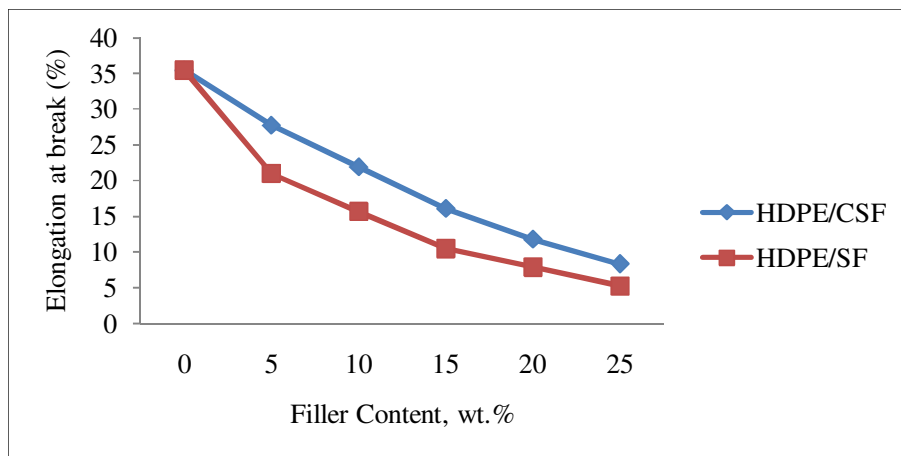


Figure 2. Plot of Elongation at Break versus Filler Content of HDPE/SF Blends

The reduction in elongation at break at higher filler content has been observed for similar systems (Ismail et al., 2002; Otay et al., 1980). This also supports the claim of Billmeyer (1971) that inclusion of non-reinforcing fillers to polymer melt weakens the polymer chain. The reduction might be due filler accumulation in adjacent polymeric chains which could reduce the mobility of the chains (Raymond and Charles, 1981).

The effect of MA-g-PE compatibilizer on the elongation at break was investigated (Fig. 2). Similar to the tensile strength, the elongation at break was found to improve on addition of compatibilizer, showing a 33 % increment on elongation at break. The reasons earlier advanced for tensile strength apply.

Young's modulus determination showed that increasing filler content had a tendency to increase the films stiffness, as presented in Figure 3. It was evident that stiffness increased from 61 MPa to more than 134 MPa. The increase in Young's modulus with the increase in sorghum filler addition shows that HDPE has inherent stiffness of the filler. Young's modulus is known to be a less sensitive variation of interfacial adhesion than the tensile

strength which greatly is linked with interfacial failure behaviour (Doan et al., 2006). The Young’s modulus of the SF/HDPE films in the presence of MA-g-PE was observed to be significant despite a possible lack of homogeneity in the dispersion of the filler from 61 MPa to over 157 MPa. This may be attributed to the enhancement of interfacial adhesion between the matrix and the filler.

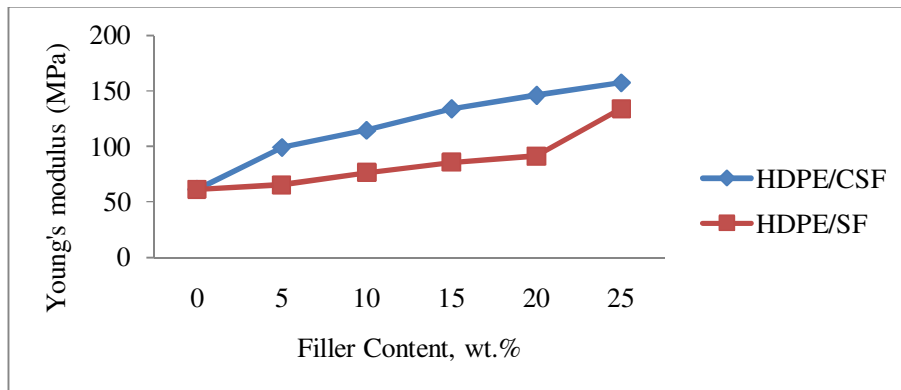


Figure 3. Plot of Young’s Modulus versus Filler Content of HDPE/SF Blends

Water Absorption Study

Water absorption tendency is one of the major limitations to the utilization of starch-based materials and any improvement in water resistance is therefore highly important (Wang et al., 2008). Figures 4 and 5 show the water uptake for the compatibilized and uncompatibilized SF/HDPE blends. From the figures, the water uptake for both SF/HDPE/MA-g-PE and SF/HDPE blends increased with increasing immersion time and sorghum flour content. This may be attributed to the hydrophilic character of the filler and by virtue of the abundant hydroxyl groups present which are available for interaction with water molecules. Therefore, a higher filler content led to a higher amount of water being absorbed (Kale et al., 2007). Neat HDPE did not show any measurable water uptake due to its hydrophobic structure. It has noticed from the figure that the rate of water uptake decreased with immersion time after attainment of plateau. This could be due to the changes in the concentration gradient across the two materials and some starch particles may have leached away from the blend films. Initial water molecules adsorbed to starch particles are strongly bonded to the hydroxyl groups as in a hydrate. When all of the available hydroxyl groups are used up, further water uptake is held loosely (Willet, 1994).

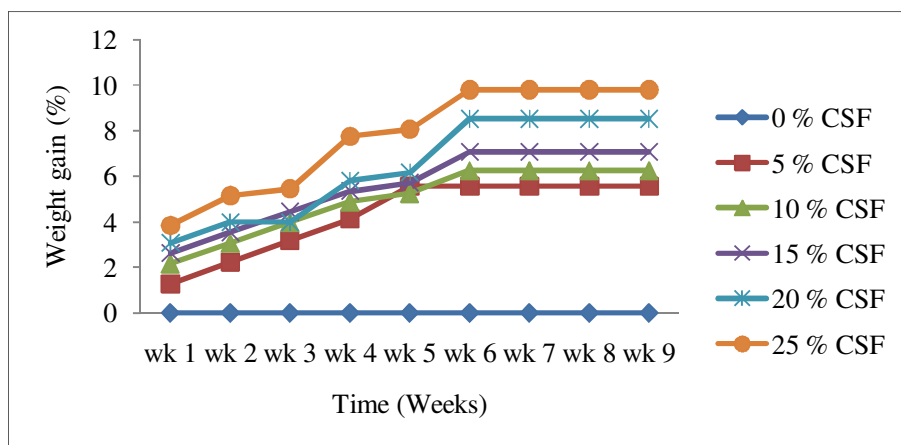


Figure 4. Weight Gain versus Time of HDPE/CSF Blends

The compatibilized, SF/HDPE blends films exhibited lower water uptake as compared to the uncompatibilized blend films. This is probably due to the bonding which is formed between the SF-MA-g-PE and HDPE-MA-g-PE with improved interface adhesion that reduced the water penetration into the blends.

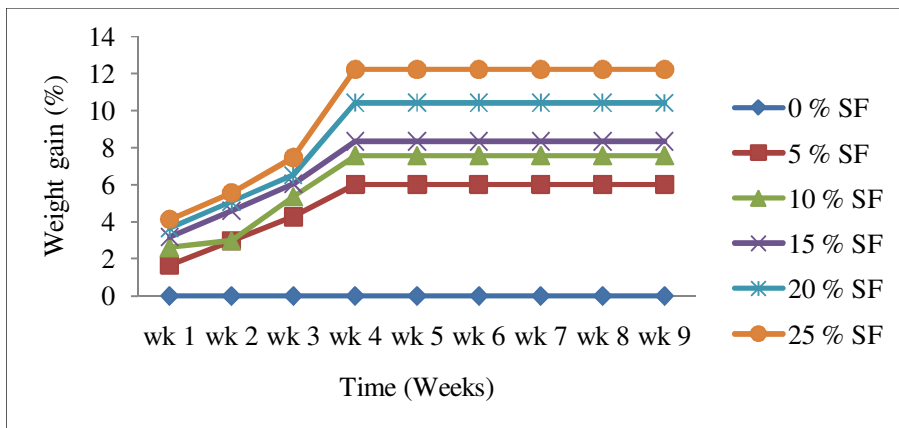


Figure 5. Weight Gain versus Time of HDPE/SF Blends

Soil Burial Test

The percentage weight loss of SF/HDPE blend films for both compatibilized and uncompatibilized blends is presented in Figures 6 and 7. The natural soil burial test is well known to be a slow process. It is interesting to note that the burial test reflects real-life conditions better than any other test. From the figures, it can be seen that the percentage weight loss of the blends increased with increasing sorghum flour addition. The weight loss of sorghum flour mainly involves hydrolytic depolymerisation of starch materials to lower molecular weight compounds, finally yielding monomeric glucose units by microorganisms (Kim et al., 2005). As the content of sorghum flour increased, localized micropores are created in the blends, major deterioration of cellulose and wood- based lignocellulosic materials caused by microorganisms giving rise to increase in the rate of degradation (Tserki et al., 2003; Ratto et al., 1999; Ishiaku et al., 2002; Hon and Shiriash, 1991). Thus, this explains the observed high weight loss of HDPE blends filled with sorghum flour. However, the SF/HDPE blends with the addition of the MA-g-PE compatibilizer caused less percentage weight loss than that of the SF/HDPE blends without the compatibilizer due to the improved phase compatibility between the SF and HDPE.

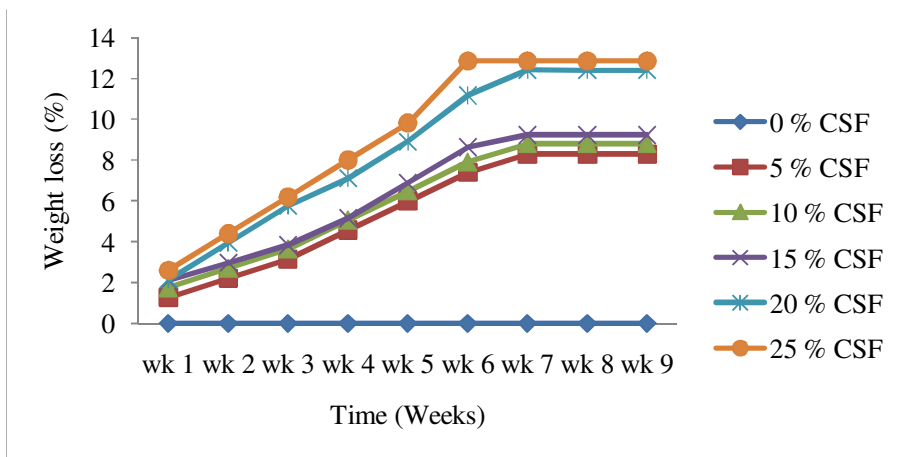


Figure 6. Weight Loss versus Time of HDPE/CSF Blends

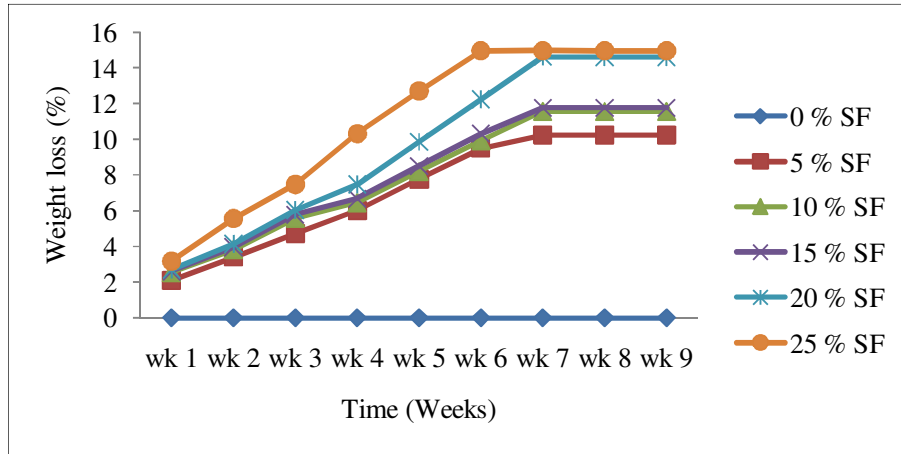


Figure 7. Weight Loss versus Time of HDPE/SF Blends

CONCLUSIONS

The sorghum flour behaved as filler in the blend system. As was the case with other fillers, sorghum flour filler addition decreases the tensile properties of extruded blend films due to weak interfacial bonding between filler and matrix polymer. Therefore, MA-g-PE is required to enhance the compatibility between the matrix and sorghum flour. Tensile properties of SF/HDPE blends are enhanced on addition of MA-g-PE due to better interfacial adhesion between SF and HDPE. Water absorption study and soil burial test show that the incorporation of MA-g-PE into the blends reduced the water uptake and weight loss of SF/HDPE blends films respectively.

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