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ORIGINAL RESEARCH ARTICLE

THERMOPLASTIC STARCH BIOCOMPOSITE PRODUCED FROM CORN (*Zea mays*): COMPARISON OF PLASTICIZERS EFFECTS ON MECHANICAL PROPERTIES

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ABSTRACT

The excessive reliance on plastic materials made from fossil-fuel based and its ineffective waste management leads to environmental pollution due to their non-biodegradable nature. This study examined the production and testing of bio-composites from Gmelina Arborea wastes and thermoplastic starches as a polymer matrix. Particles of G. Arborea sawdust were obtained from a local sawmill while dry powdered corn starches were sourced from the chemicals market, Ojota, Lagos, Nigeria. Bio-composites were produced by mixing 40 g of corn starch, glycerol/sorbitol, and fractions of prepared G. Arborea (650µm particle sizes) fibre at 0 to 20% (wt/wt of fibre to polymer matrix) were added. The slurries were poured into a mould in accordance with ASTM D638 for tensile strength test samples. The results revealed that the thermoplastic starches, particle sizes, plasticizers, and wood fibre loading significantly affected the tensile strengths, tensile modulus, elongation at break of the bio-composites. Composites produced exhibited tensile strengths values range of 0.35 to 1.75 MPa, modulus of elasticity of 3.10 to 22 MPa, while there was a gradual reduction in elongation at break with a range of 127.4% to 111%. Sorbitol-corn starch generally recorded higher tensile properties of between 1.65 to 1.75 MPa tensile strength than those produced with glycerol plasticizer which ranged between 0.35 to 0.45 MPa as fibre loading increases. It is clear that sorbitol-plasticized bio-composite gives higher values in mechanical strengths when compared to that produced from glycerol

1.0 Introduction

The consistent and astronomical anthropogenic effect on the environment cannot be overlooked as these affect the world's ecosystem through pollution causing factors introduced into the environment. The anthropogenic effect resulting from human activities possess great risks to life, as some of these factors leads to environmental pollution. Humans' activities over the years depended on fossil fuel as a resource for the production of many products resulting in degradation of the environment. Plastics and other products produced from hydrocarbon resources tend to cause pollution in the environment because majority of these products are not biodegradable. An increase in world population resulted in the geometrical increment in the needs for plastic materials for usage resulting in large quantities of these plastics finding their way onto landfills and the ocean has evidence had shown an increase in micro-plastics in the ocean ecosystem (Lusher, 2015). The use of plastic materials is on the high side as human's disposition to its usage is high, also couple with the population boom in the world, the tonnes of used plastics create challenges of disposal as many are either ship to landfills or indiscriminately disposed into waterbodies (Lusher, 2015). This phenomenon has been a major environmental challenge which requires holistic approach to developing materials that can replace petrol-based plastics example of which can be termed as green composite (Wattanakornsiri *et al*., 2011). An example of such a material is a termed green composite that can be biodegraded in the environment (Wattanakornsiri *et al*., 2011). It is important that such composite materials should utilized biodegradable renewable polymers as

matrices and fibre as fillers to the matrix. Composite material produced from biodegradable constituent is expected to be biodegradable when disposed of in the environment. Biodegradable composite materials assist in the reduction of the long effect of plastic pollution in the environment leading to a reduction in the greenhouse gas effect (Mohanty *et al*., 2005). Biodegradable composite materials break down easily in the environment forming compost when returned to the soil (Huang and Netravali, 2007). These materials can easily replace petrol-based materials leading to a major shift in the material selection that is environmentally friendly (Mohanty *et al*., 2002). Sources of polymer matrices abound but researchers have looked into the use of starch as a source for matrix and this is due to its naturally renewable nature, relatively cheap, and abundance (Teixeira *et al*., 2009, Sarifuddin *et al*., 2012). In order for starch to be used as a polymer matrix, it undergoes thermal modification into thermoplastic starch by the addition of a plasticizer into the starch-water mixture and agitated on a high heating element (Averous and Boquillon, 2004). Another alternative environmentally friendly material are natural fibres such as wood fibres (hardwood, wood dust and softwood) or other wide varieties of natural plant fibres, such as flax, pineapple, bamboo, hemp, cotton, kenaf, coir (Ochi, 2011) which can be mixed with polymer matrices to produce a biodegradable material. Puglia *et al*. (2003) reported that the mechanical properties of thermoplastic starch increases with the addition of natural fibres. Averous *et al*., (2001) researched into the use of wheat starch as the polymer matrix in the development of thermoforming packaging material. Wattanakornsiri *et al*. (2011) studied the effect on mechanical properties of the thermoplastic starch by addition of recycled paper cellulose fibres. Flax and ramie fibre serves as a source of filler in the development of green biocomposite (Wattanakornsiri and Tongnunui, 2014).
Development of an environmentally friendly composite from naturally biodegradable material sources may be a means of curtailing the menace of plastic pollution. This research work is aimed at the production of bio-composite from thermoplastic corn starch (matrix) with the use of *Gmelina Arborea* fibre as fillers, looking at the fibre loading effect as well as that of the plasticizers on the mechanical properties.

2.0 Materials and Methodology

2.1 Materials

The materials used in the production of the bio-composites were sourced from different markets. Chemicals such as powdered corn starch, glycerol, sorbitol were sourced from chemical market in Lagos, vegetable oil from local food store while the sawdust of *Gmelina Arborea* was sourced from Bodija International Market Sawmill in Ibadan. The remaining equipment were obtained at the laboratory in which the research was carried out.

Digital weighing balance, flat bottom flask, beaker, an electric stove, stirrer, moulds, powdered corn starch, glycerol, sorbitol, water, *Gmelina arborea* particles, vegetable oil, were the materials and equipment used.

2.2 Fibre Preparation

Gmelina Arborea particles was obtained from the sawmill at Bodija Market Sawmill Ibadan, Oyo State. The particles were sieved through some set of sieve sizes range and 650µm sieve size was eventually selected for the research (Figure 1a). These were then treated with a 5% NaOH solution for 4 hours and air-dried until 12% moisture content was obtain.

2.3 Production of Bio-Composites

The bio-composites test samples were produced in the laboratory in batches, with the first batch been the glycerol plasticized biocomposite which was fabricated by mixing known dried weight of corn starch with distilled water, glycerol and the filler (*Gmelina Arborea* fibre) following the method reported by Adefisan and Eyeowa, (2015).The mixture was heated on a hotplate with continuous stirring until gelatinization occurred. This procedure was repeated to obtain bio-composite plasticized with sorbitol by replacing glycerol with sorbitol plasticizer.

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The *Gmelina Arborea* fibres of 650µm particle sizes were added to the mixture at varying proportion of 0, 5, 10, 15 and 20% wt/wt of fibre to matrix according to Johar and Ahmad (2012). Neat bioplastic was produced following the procedure above without the addition of fibre that is no fibre load, to serve as a basis for comparison; the dried sample of which is shown in Figure 1b. The slurries of the produced biocomposite were poured into a dump-belled mould (Figure 1c) of standard procedures (ASTM D638-14) with specimen dimension of length overall 165 mm, width overall 19 ± 6.4mm, the width of narrow section 13 ± 0.5mm, length of narrow section 57 ± 0.5mm, gage length 50 ± 0.25mm, the distance between grips 115 ± 5mm, a radius of fillet 76 ± 1mm and thickness 3.2 ± 0.4mm. The mould was smeared with vegetable oil (to allow easy de-moulding) and the de-mould sample were dried at room temperature for 3weeks.

2.4 Mechanical Test

2.4.1 Tensile test

The biocomposite tensile test samples were produced in triplicates in accordance with ASTM D638-14. The test was carried out at the Centre for Energy Research and Development (C.E.R.D) at Obafemi Awolowo University, Ile Ife using the Universal Testing Machine INSTRON 3369- Load Cell Capacity 50 KN, manufactured in USA (Figure 1d), and operated with a crosshead speed of 5 mm/mins in accordance to Sarifuddin *et al.* (2012).

2.4.2 Modulus of Elasticity

The Modulus of Elasticity or Young's Modulus of the test samples was derived from the result output of the Universal Testing Machine UTM, making use of the relationship as expressed below;

$$\text{Young's Modulus} = \frac{Tensile\ Stress}{Tensile\ Strain}$$

2.4.3 Elongation at Break

The Elongation at break expresses the capability of natural plant fiber to resist changes of shape without crack formation. It is derived from the ratio of change in length with that of the initial length and it is expressed in percentage.

2.5 Statistical Analysis

The results obtained from the machine output (excel spreadsheet) of the test carried out were subjected to analysis using the Duncan Multiple Range Test at $p < 0.05$ significant level. This test is carried out in order to reveal the significant difference between the thermoplastic starch, plasticizers and wood fibre loading on the mechanical properties of the bio-composites.

Figure 1: (a) sieving of sawdust, (b) bioplastic, (c) test sample, (d) tensile test on machine

Figure 1 (a) shows the researcher sieving the sawdust in order to obtain the required particle size, while (b) shows the neat bioplastic produced and (c) and (d) shows the tensile test samples of the biocomposite produced and the material undergoing deformation on the test machine, respectively.

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3.0 Results and Discussion

3.1 Tensile Strength

The results of the tensile strengths of the bio-composites are shown in Table 1 and Figure 2. The tensile strengths ranged from 0.35 to 0.45 MPa and 1.65 to 1.75 MPa for the corn-thermoplastic bio-composites plasticized with glycerol and sorbitol respectively. These values compared favourably with the findings of Yauri and Abbot, (2014). Figure 2 shows a gradual increase in the tensile strength of the bio-composite plasticized with glycerol but these strengths cannot be compared favourably to that of sorbitol as biocomposites produced with sorbitol as plasticizers performed better than that of glycerol. The tensile strength value of sorbitol plasticized biocomposite is far greater than that of glycerol. Generally, the addition of *G. Arborea* particles (5 – 20%) increases the tensile strength of the bio-composites produced. The fibre 650 µm addition generally record higher tensile strengths, suggesting that the fibre particle provided good reinforcement which is possibly due to higher tensile strength required to detach fibres from the matrices (Luddee *et al*., 2014). It is evident that composites plasticized with sorbitol generally recorded higher tensile strengths than those made with glycerol. This may be attributed to the effective plasticization of glycerol over sorbitol thermoplastic starches which results in the reduction in the frictional forces between the starch molecules, brittleness of the composites and the reduction in tensile strength (Vieira *et al.,* 2011). Statistically, there exists a significant difference ($p < 0.05$) between the values of tensile strength obtained for glycerol and sorbitol. Similarly, the fibre loading has a significant effect on the tensile strength of the produced bio-composites. Overall sorbitol plasticized bio-composites give a higher tensile strength.

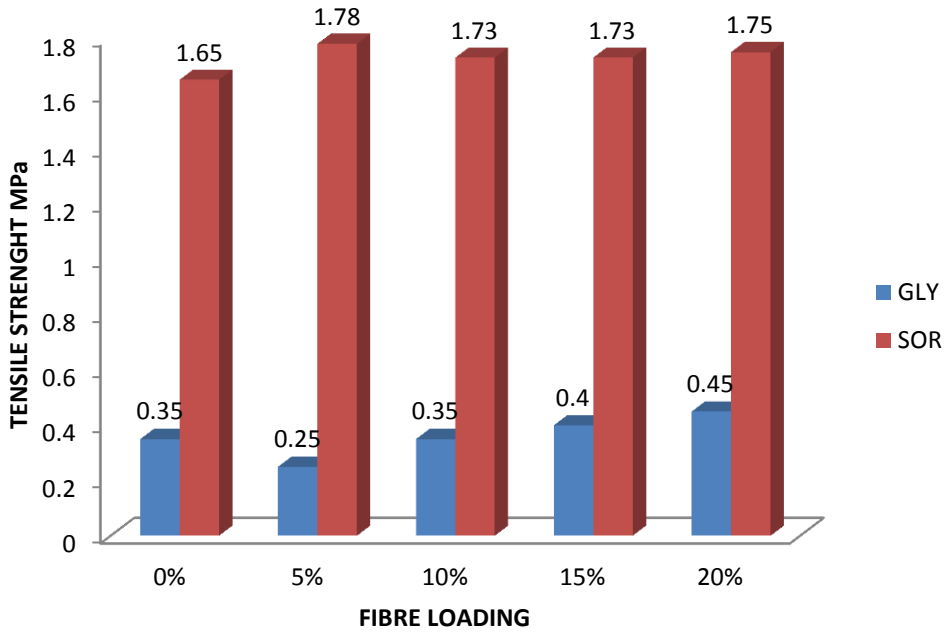

Figure 2: Variation of Tensile Strengths of produced bio-composite with Fibre Loading.

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Table 1: Duncan's Multiple Range Test of Effects of Plasticizer and Fiber Loading on Tensile Strength of Bio-composite.

Parameters	Tensile Strength (MPa)
Plasticizers	
Glycerol	0.41^{B}
Sorbitol	2.17^{A}
Fibre Loading (%)	
0	1.38^{A}
5	1.32^{AB}
10	1.33^{AB}
15	1.23^{AB}
20	1.19^{B}

Means with the same letters are not statistically different ($P < 0.05$)

3.2 Tensile modulus of Elasticity

The tensile modulus of elasticity of the bio-composites made from corns thermoplastic starch fall between 3.10 to 22 MPa. In specific terms, the values of the tensile modulus of elasticity for glycerol plasticized biocomposite increases gradually from 3.1 to 5 MPa. Although there is a slight increment in the modulus of elasticity between 0% to 5% fibres loading, the increment exhibits a much higher value as the fibre loading increases with the highest value obtained at 20% fibre content as shown in Figure 2. Sorbitol plasticized bio-composite also shows an increase in tensile modulus as the fibre loading percent increases with the highest value obtained at 20% fibre loading at 22 MPa. These values show same pattern as obtained by Boudjema and Bendaikha (2015). The gradual increase in the modulus of elasticity as the fibre loading increases can be attributable to the increased fibre-fibre interaction with the reduction in the thermoplastic starch matrices. Bio-composite produced from sorbitol-corn starch thermoplastic exhibited a high value of modulus of elasticity than that obtained for glycerol plasticized bio-composite. The result obtained from the tensile properties tested shows similarity with those reported by other researchers for lignocellulose fibre mixture with thermoplastic starches resulting in bio-composite (Ramírez *et al*., 2011). The explanation for this phenomenon is the compatibility of the chemical and structural composition of the matrix and the fibre of the plant used as reinforcement resulting in the improvement of the mechanical properties of the bio-composite (Guimaraes et al., 2010; Averous and Boquillon, 2004).

Statistical analyses (Duncans' multiple range test $P < 0.05$) (Table 2) revealed a significant difference between the plasticizers on the modulus of elasticity of the bio-composites produced showing evidently that fibre loading exerts an effect on the tensile modulus as well.

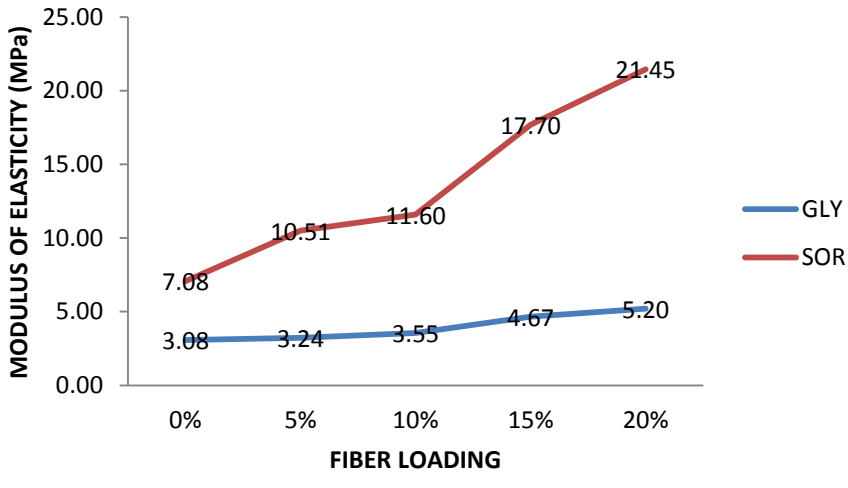

Figure 2: The effect of fibre loading on the Modulus of Elasticity of Corn-Plasticized Biocomposite.

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Table 2: Duncan's Multiple Range Test of Effects of Plasticizer and Fiber Loading on Modulus of Elasticity of TPS - Bio-composite.

Parameters	Tensile Modulus (MPa)
Plasticizer	
Glycerol	3.0^{B}
Sorbitol	16.8^{A}
Fibre Loading (%)	
0	6.6^{D}
5	7.1^{D}
10	9.6^{C}
15	11.3^{B}
20	15.1^{A}

Means with the same letters are not statistically different ($P < 0.05$)

3.3 Elongation at Break

The elongation at break for different fiber contents is presented in Figure 3.

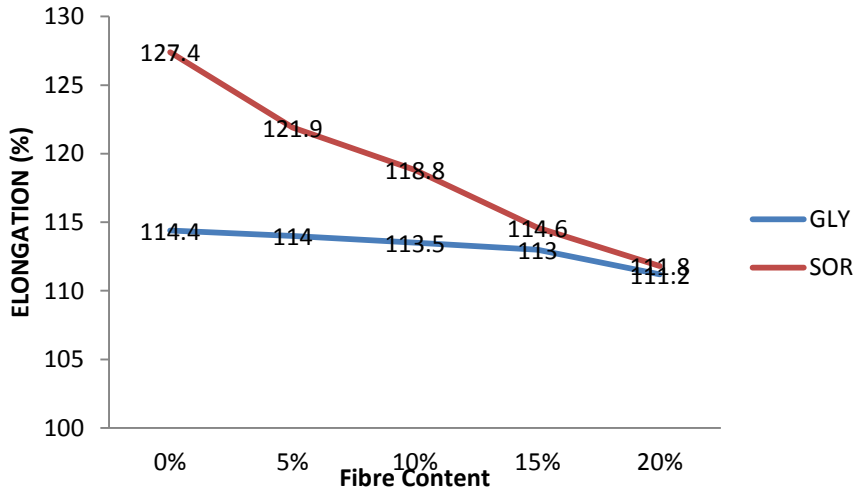

Figure 3: Effect of fiber content on elongation at break of biocomposite.

As shown from Figure 3, the elongation at break decreased monotonically as the fibre loading increases from 0% to 20% that is from 114.4% to 111.2% and 127.8% to 111.2% for the corn-glycerol and corn-sorbitol bio-composites respectively. These observations are in conformity with the reports of Talib *et al.* (2010). The incorporation of 100 % TPS matrix elongated the bio-composites due to its elasticity. As the *Gmelina Arborea* fibre loading increases, the elasticity of Thermoplastic starch (TPS) was reduced by the increment in the fibre. The rigid interface between the matrix and the fibre which is responsible for level of deformability reduces as fibre loading increases thereby leading to a reduction seen in the elasticity of TPS, this assertion agrees with Rozman *et al.* (2005). At higher fibre loading, the domination of fibre-matrix interaction diminished being replaced by filler-filler interaction. This effect is because fibre incorporation imparts rigidity and restrains the deformation on the matrix. This often leads to an inevitable decrease in the degree of the ductility of the material (Belhassen *et al.*, 2009).

It is evident from the result obtained in this work that tensile properties of bio-composite produced are in tandem with other research work reported by researchers in the area of lignocellulose fibres or cellulose fibres used as fillers in TPS (Torres *et al.*, 2007; Woehl *et al.*, 2010). This phenomenon is due to the compatibility between matrix and reinforced fibres structurally and chemically. Due to the chemical nature of both starch and fibre a linkage is formed at the cellulose chain which tends to influence the performance and mechanical properties of the bio-composite (Ma *et al.*, 2005; Mo *et al.*, 2010).

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4.0 Conclusion

This study shows that bio-composite can be produced with the incorporation of *G. Arborea* fibre particles in a corn-thermoplastic starch matrix plasticized with sorbitol and glycerol, with the test samples achieving a moisture content of 12%. The different plasticizers used in this work affected the tensile properties of bio-composites produced with that of sorbitol plasticized bio-composite having a higher influence. A steady increase in the tensile strength and modulus of elasticity as the fibre loading increases while the elongation at break reduces with increase in fibre content was witnessed. The fibre used in this work affected the mechanical properties of the bio-composites having a greater influence on the resulting outcome, which is as a result of a good interfacial bonding between the thermoplastic starch matrix and fibre giving it strength in the range that can favourably replace petrol based-plastics materials. Further studies should be carried out in the area of moisture content to determine the effect if it were to be varied.

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