

Original Research Article

EFFECT OF COMPOSITION VARIATION ON THE TENSILE STRENGTH OF POLYLACTIC ACID REINFORCED WITH NATURAL POLYMERS FOR BIODEGRADABLE BIOMEDICAL IMPLANTS

ABSTRACT (ARIAL, BOLD, 11 FONT, LEFT ALIGNED, CAPS)

Aims: This research focuses on developing a blend of natural polymers suitable for biomedical implants with standard mechanical (tensile) properties, compatible and consumable with human body

Study design: *This research developed a polymer composite from a blend of the three natural polymer materials with different compositions that can adequately replace non-biodegradable implants.*

Place and Duration of Study: The research was conducted at the department of industrial and production engineering and department of metallurgy and material engineering federal university of technology akure Nigeria between 2017 and 2018.

Methodology: such PLA was blended with starch and chitosan which are the two most abundant natural polymers using compression molding machine. The mechanical (tensile) property test carried out which give information on the strength and stiffness of the composite using the ASTM D3039. also the effect of composition variation of each of the constituent polymer was analyzed using multiple regression analysis..

Results: The results shows similarity with the mechanical (tensile) properties of cortical bone which is the major bone in human body that is susceptible to fracture. The result of the regression analysis revealed a negative 43.129 effect of PLA on the tensile stress; positive 10.43 effect of starch on tensile stress; negative 0.596 combine effect PLA and starch on the tensile and the effect of chitosan we have a NA value which implies that the coefficient is not quantifiable.

Conclusion: Combining PLA and starch with chitosan to obtain biodegradable implant materials represents good alternative means to reduce use of synthetic or metallic materials. Consequently, it will cause a paradigm shift from the heavy dependence on non-biodegradable metallic materials to lower cost natural polymers.

Keywords: *[Biodegradable, Chitosan, Composite, Compression molding, Poly-Lactic acid, Starch]*

1. INTRODUCTION

A biomedical implant is a device aimed at replacing an organ or tissue in the body as a result of damage to the latter or to give a better performance than the natural one can produce. A biomaterial is defined as any natural or synthetic substance engineered to interact with biological systems in order to direct medical treatment. (Bret D. Ulery et. al., 2011). The

23 increase in need for biomedical implants over the years has equally spurred an increase in
24 research in the area of materials that can be safely introduced into the body system without
25 causing harm.

26 The advent of tissue engineering has been motivated by the challenge of producing tissue
27 substitutes that can restore the structural features and physiological functions of natural
28 tissues in vivo (Vanolen et al., 2010; Freed et al., 2009). The interest in using the bio-
29 absorbable polymer is mainly because they are degraded by hydrolysis; thereby preventing
30 the patient from undergoing a second surgery to remove the device, reducing cost and
31 trauma. Biopolymers are used as replacements for damaged tissue and or stimulate its
32 regeneration. Among the various bio-reabsorbable polymers are the poly (α -hydroxy acids).
33 These polymers are considered bio-reabsorbable due to its good biocompatibility and its
34 decomposition products are eliminated from the body via metabolic pathways. Poly (lactic
35 acid) (PLA) is a highly versatile, biodegradable, aliphatic polyester derived from 100%
36 renewable resources (Drumright et al., 2000). It has extensive applications in biomedical
37 fields, including suture, bone fixation material, drug delivery microsphere, and tissue
38 engineering (Nampoothiri et al., 2010). Because of these properties PLA has been widely
39 studied for use in medical applications.

40 Chitin is also a major component for this research. Chitin, extracted primarily from shellfish
41 sources, is a unique biopolymer based on the N-acetyl-glucosamine monomer. It is a co-
42 polymer of N-acetyl-glucosamine and N-glucosamine units randomly or block distributed
43 throughout the biopolymer chain depending on the processing method used to derive the
44 biopolymer. When the number of N-acetyl-glucosamine units is higher than 50%, the
45 biopolymer is termed chitin. Chitin, together with its derivatives, has been shown to be useful
46 as a wound dressing material, drug delivery vehicle and an essential candidate for tissue
47 engineering.

48 Starch, the final component of the polymer blend is abundant naturally and is easily
49 synthesized for the composite. Starches from various plant sources, such as wheat, corn,
50 rice and potato have received extensive attention in relation to structural and
51 physicochemical properties. Starch is widely used in food, pharmaceutical and biomedical
52 applications because of its biocompatibility, biodegradability, non-toxicity, and abundant
53 sources. The role of starch for tissue engineering of bone, bone fixation, carrier for the
54 controlled release of drugs and hormones; and as hydrogels has already been recognized
55 (Won et al., 1997; Chakraborty et al., 2005). Starch-based biodegradable bone cements are
56 highly advantageous because they can provide for immediate structural support and, as they
57 degrade from the site of application, allow the ingrowth of new bone for complete healing of
58 bone fracture (Domb et al., 1996; Pereira et al, 1998).

59 The major concern in this study are the mechanical properties of the material and
60 consumability of the material by the body. This informed the choice of the material; poly
61 lactic acid, chitin and starch are all consumables and the effect on the body is minimal on the
62 long-run. Also the need for affordable biomedical implants is one of the basis of the
63 research. The process of making implants affordable can easily be achieved if the base
64 materials can be naturally obtained.

65 **2. MATERIAL AND METHODS**

66 **2.1 Materials**

67 The material selection for this research is governed by the desire to shift focus from
68 synthetic polymers to natural polymers for the synthesis of biomedical materials. The natural

69 polymers are readily available and easily produced therefore, optimizing their properties to
 70 those suitable for biomedical implants will greatly reduce the cost of biomedical implants.
 71 The polymers to be used for this research include chitin which is extracted from crab bones,
 72 starch which is produced from various foods and PLA which can be produced from corn
 73 starch, cassava roots. Starch has adhesive properties which can make it a suitable binder
 74 for the composite. Also, the combination of chitin and calcium carbonate produces a tougher
 75 and less brittle structure which is a useful property for the blend.

76 2.2. Composition Variation

77 The PLA will range from 92% to 98% of the composition, starch and chitosan will range from
 78 2% to 8%. The PLA serves as the matrix, the chitosan is a reinforcement and the starch is a
 79 binder. The composition variation for research is shown in table 1

80 **Table 1 Percentage Composition for the experiment**

Percentage composition			
S/N	PLA (%)	Starch (%)	Chitosan (%)
1	92	8	0
2	92	6	2
3	92	4	4
4	92	2	6
5	92	0	8
6	94	6	0
7	94	4	2
8	94	2	4
9	94	0	6
10	96	2	2
11	96	4	0
12	96	0	4
13	98	2	0
14	98	1	1
15	98	0	2

81

82 2.3. Compression Molding Process

The essence of choosing compression molding (figure 1) is because it is a suitable method for producing high strength plastic and polymer materials. It is a closed molding process with high pressure application. The compression molding process is a method of molding in which a preheated polymer is placed into an open, heated mould cavity. The mould is closed with a top plug and pressure is applied to force the material to contact all areas of the mould. Throughout the process heat and pressure are maintained until the polymer has cured. While the compression molding process can be employed with both thermosets and thermoplastics, today most applications use thermoset polymers. Advanced composite thermoplastics can also be compression molded with unidirectional tapes, woven fabrics, randomly orientated fiber mat or chopped strand. Compression molding is a high-volume, high-pressure plastic molding method that is suitable for molding complex, high-strength objects. And with its short cycle time and high production rate, many organizations in the automotive industry have chosen compression molding to produce parts.

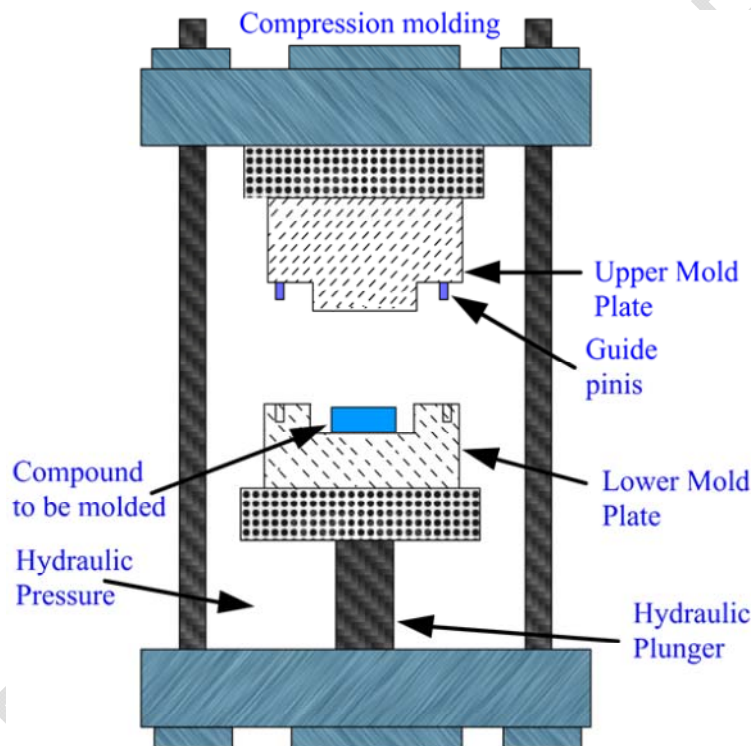


Fig.1.Compression molding machine

2.4. Experimental Design for the Compression Molding Process

The composite will be produced by compression molding, the compression molding machine has a variety of control options that determine the final properties of the molded sample. The parameters that need to be controlled include Mould Temperature, Holding Pressure and Holding time. The temperature used for the mould was 750C and the mould pressure used was 1.5 metric tons with a holding time of 15mins. The composition variation was based on after previous experiment from Romana et. al., 2017 and pre - trial experiment during this research The percentages of chitin and starch was varied between 8% to 0% at interval of plus or minus 2%

2.5. Mechanical Property Test

The mechanical (tensile) properties of the samples was tested. Using American standard for material test, the tensile, test method ASTM D3039 is selected with standard dimension. The test method is used for reinforced or unreinforced materials including high modulus composites and for materials that do not fail within the limits. This is shown in figure 2

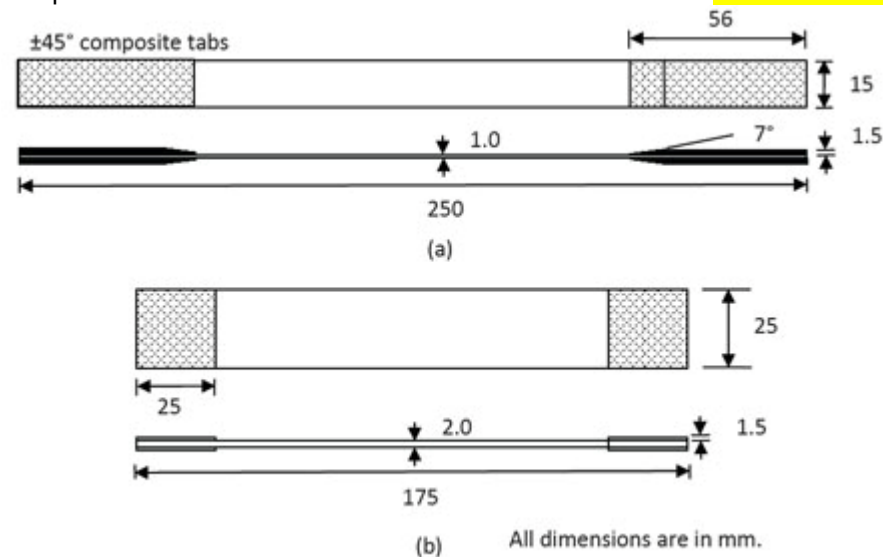


Fig 2. Polymer composite specimen for tensile test (ASTM D3039) www.astm.org

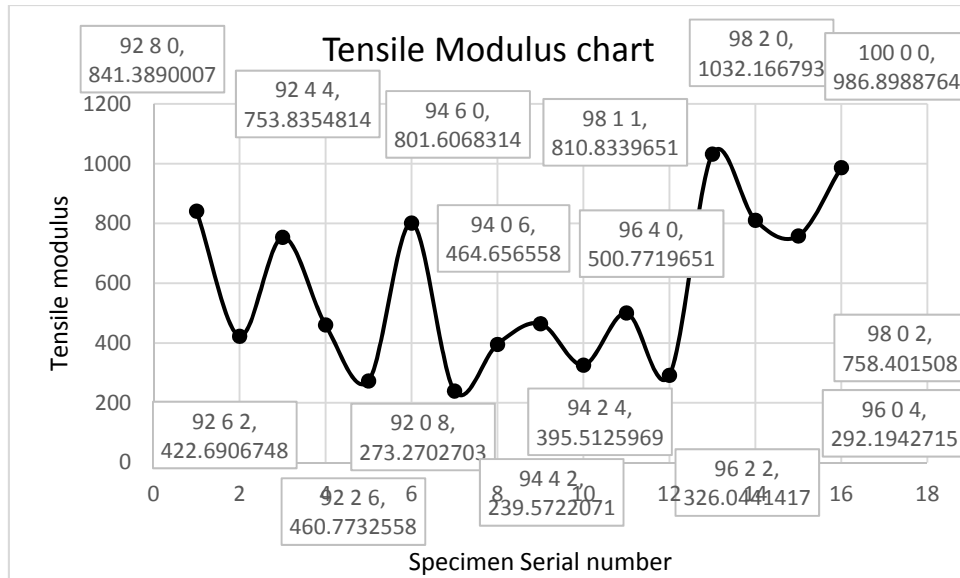
3. RESULTS AND DISCUSSION

The results from the various tensile tests conducted on the samples show some important properties about the behavior of the composite under different kinds of load. The results of the tests comparing them to the equivalent properties of packaging materials are discussed in the following sections. The maximum tensile stress the withstood by a single sample is 16.09328MPa (Fig. 3) which was gotten form the sample with 98% PLA and 2% Chitosan when compared with the control sample of 100% PLA with tensile strength of 25.47186 MPa (Table. 2). This Tensile strength is close to the strength of the neat PLA used showing that a further research into a favorable composition or with a compatibilizer will further increase the strength of the composite. The graph of strain Energy to strain at break point shows significant similarities with the graphs that compare the tensile stress which validates the tensile stress values deduced from the experiment. The tensile properties of the polymer composite is shown in table 2 and 3 respectively

Table 2 Tensile properties of all the samples

Percentage composition				Max tensile stress (MPa)	Tensile strain	Tensile modulus (MPa)	Percentage elongation
S/N	PLA	STARCH	CHITOSAN				
1	92	8	0	12.54511	0.01491	841.389001	1.491
2	92	6	2	5.57529	0.01319	422.690675	1.319
3	92	4	4	9.94309	0.01319	753.835481	1.319
4	92	2	6	4.75518	0.01032	460.773256	1.032
5	92	0	8	3.13441	0.01147	273.27027	1.147
6	94	6	0	11.03011	0.01376	801.606831	1.376
7	94	4	2	4.39615	0.01835	239.572207	1.835
8	94	2	4	4.08169	0.01032	395.512597	1.032
9	94	0	6	6.12882	0.01319	464.656558	1.319
10	96	2	2	5.98291	0.01835	326.044142	1.835
11	96	4	0	7.46651	0.01491	500.771965	1.491
12	96	0	4	2.34632	0.00803	292.194271	0.803
13	98	2	0	13.61428	0.01319	1032.16679	1.319
14	98	1	1	10.6949	0.01319	810.833965	1.319
15	98	0	2	16.09328	0.02122	758.401508	2.122
16	100	0	0	25.47186	0.02581	986.898876	2.581

144
 145 From the figure 3 it can see the composition with PLA: STARCH ratio of 98:2 having a
 146 tensile strength of 16Mpa and a tensile modulus of 758.401 MPa and a percentage
 147 elongation of 2.1%, this indicates that the composite is more flexible and still maintains its
 148 strength to a great extent compared to the neat PLA having a tensile modulus of 986.9 MPa.
 149 And a percentage elongation of 2.5%. This gives this composite a fairly large amount of
 150 applications as a biomedical material (Khan et. al., 2014; Somasondaram et.al., 2017)
 151 such as cancellous bone scaffold (Dhivyaa et. al., 2018) and other tissue
 152 repairs.



153

154 **Fig 3 Graph Showing the Tensile modulus of all the samples**

155 3.1 Statistical Analysis with Multiple Regression

156 The data was analyse using multiple regression analysis to understand the significant
 157 interactions between the variables and its corresponding effect on the Tensile strength. The
 158 result is presented in table 3 and equation 1

159 **Table 3. Tensile stress regression analysis output**

Coefficients: (1 Not Defined Because Of Singularities)		
	Estimate	Pr(> T)
(Intercept)	4335.9879	0.0578
PLA	-43.1285	0.0585
Starch	10.4308	0.6164
Chitin	Na	Na
PLA: Chitin	-0.4941	0.0508
PLA: Starch	-0.5965	0.0237*
Starch: Chitin	-0.1291	0.4591

160

161 Im (formula = MAX. TENSILE. STRESS. Mpa. ~ PLA + STARCH + CHITIN + PLA * CHITIN
162 + STARCH * PLA + STARCH * CHITIN)

163 Multiple R-squared: 0.6948, Adjusted R-squared: 0.5252

164 Fitted regression equation:

$$\begin{aligned} \mu_{(y|x)} = & 4335.988 - 43.129 \times \text{PLA} + 10.43 \times \text{STARCH} + \text{NA} \times \text{CHITOSAN} \\ & 0.494 \times \text{PLA} \times \text{CHITOSAN} - 0.596 \times \text{PLA} \times \text{STARCH} - 0.129 \times \text{STARCH} \times \text{CHITOSAN} \end{aligned} \quad (1)$$

167 The analysis shows the effect of PLA on the tensile strength dependent on CHITOSAN and
168 STARCH. The analysis output shows the p-value for PLA:CHITOSAN and STARCH:
169 CHITOSAN which are .0508 and .0459, which implies that this interaction is not statistically
170 significant, therefore it is not include it in the model. While for PLA:STARCH there is a
171 statistically significant interaction that as occur between this two variables.

172 Therefore the fitted regression model would be reduced to;

$$\mu_{(y|x)} = 4335.988 - 43.129 \times \text{PLA} + 10.43 \times \text{STARCH} + \text{NA} \times \text{CHITOSAN} - 0.596 \times \text{PLA} \times \text{STARCH} \quad (2)$$

175 The above equation 2 shows a negative 43.129 effect of PLA on the TENSILE STRESS
176 adjusting or controlling for STARCH and CHITOSAN. The estimated effect of STARCH on
177 TENSILE STRESS to be 10.43 increase, adjusting PLA and CHITOSAN. Also the estimated
178 significant interaction effect between PLA and STARCH to be negative .596. The effect of
179 CHITOSAN result in NA value which implies that the coefficient is not quantifiable, and it
180 could be due to exact collinearity. Also the R-squared value is 0.6948 which implies that
181 approximately 69% of variation in maximum tensile stress can be explained by the
182 experiment and the model derivable from the experiment. The conclusion of the results is
183 that for the tensile strength, the percentage composition of starch has a significant effect on
184 the maximum tensile stress the material can withstand than chitosan.

185 4. CONCLUSION

186
187 This paper has shown the effect on composition variation on the tensile property of the
188 composite and also propose the use of the composite as a replacement for synthetic in
189 tissue repair and replacement. Poly lactic acid - Starch – chitin composite was fabricated
190 and the tensile properties analyzed and compared with tissue material tensile properties
191 from literature. The finding shows that this material can be used for a number of replacement
192 and repair implant material. Combining PLA and starch with chitosan to obtain
193 biodegradable implant materials represents good alternative means to reduce use of
194 synthetic polymer or metallic materials. The blending of these polymers in adequate
195 proportions with the incorporation of some compatibilizers on further research will yield high
196 performance material which can meet several biomedical requirements. The use of this
197 composite which is obtained from natural compounds will reduce the use of non-
198 biodegradable implants on the body. Consequently, it will cause a paradigm shift from the
199 heavy dependence on non-biodegradable metallic materials to lower cost natural polymers.
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