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## COMPARATIVE STUDY OF SUCROSE PLASTICISED BIOPLASTIC DERIVATIVES WITH SELECTED PETRO-PLASTICS

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### ABSTRACT

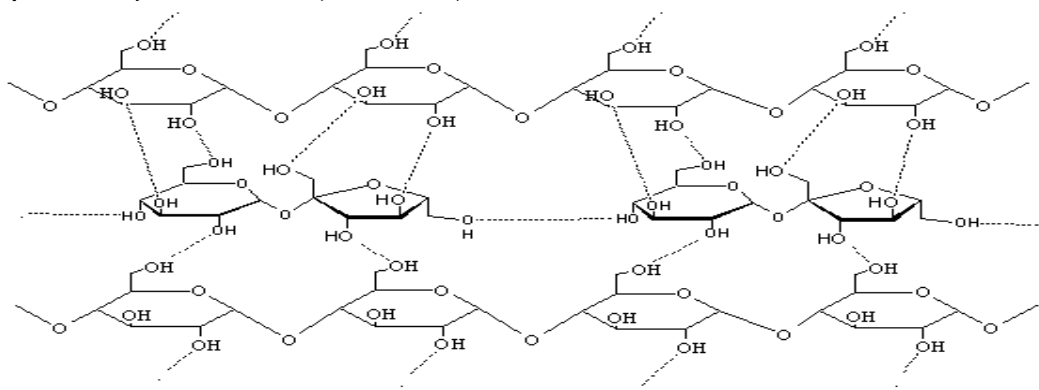
*The major setback with most bioplastics is their inherent inability to compete with Petro-plastics in terms of high production cost, and their poor mechanical properties like low tensile strength and percentage extension. This study explores the availability and affordability of mango starch as raw material for bioplastic production and compared some of its mechanical properties with High density Polyethylene (HDPE), Low density polyethylene (LDPE), polyvinyl chloride (PVC), and Polyurethane (PU). Mango starch was used to synthesize bioplastic derivatives, with variable levels of sucrose as plasticiser, aqueous HCl concentration and Carboxymethyl cellulose (CMC) as additive and the mechanical properties of the derived biofilms were measured and compared with the selected Petro-plastics films. It was observed that B1 thermoplastic derivatives have the higher young modulus of 5.658 GPa than that obtained for PVC (4.682 GPa), and PU (3.771 GPa) but show no significant difference and significantly higher than that of HDPE (0.049 GPa), and LDPE (0.063 GPa) ( $p < 0.05$ ). B2 and B3 indicated a young modulus that is significantly lower than PU and PVC, but showed a young modulus that is higher than LDPE and HDPE with no significant difference ( $p < 0.05$ ). The FTIR spectra indicate that hydrogen bond was formed in the bulk matrix of the bioplastic derivatives at a band region of 3600 -600  $\text{cm}^{-1}$  wavenumber with broad discrete peaks.*

**Keywords:** Petro-plastics; Bioplastics; Mechanical Properties; Plasticization; Mango Starch

### INTRODUCTION

The question that is most asked is "why do we prefer Petro-plastic over bioplastic, even with all the advantages", the reason is not farfetched from the limitation usually associated with its processability derived from their mechanical properties, and physicochemical properties in which water and gas barrier are the major factors. This has resulted in a sudden surge of investigations into bioplastics to improve these properties and make them more versatile, robust, and desirable in a competitive polymer market especially in single-use polymer applications. Since they are non-biodegradable and non-compostable, Petro-plastics has resulted to problem of environmental pollution both inland and marine, has resulted to decimation of marine life as a result of ingestion and entanglement (UNEP, 2015) and with the recent alarming investigation by Orb Media in 2017 reveal the presence of microplastics i.e.

Petro-plastic fragments in drinking water around the world, that pose a great concern for public health (Kosuth *et al.*, 2018; Medrano *et al.*, 2019), so the environmental unfriendliness is no more in question. Varda *et al.*, (2014), asked in his research titled "Can a starch-based Plastic be an option of Environmental Friendly Plastic" as against other bioplastics like PHA, PLA, PHB, and PCL, PEG and Petro-plastic, and the factor of production cost comes to play. Although starch is not the only renewable raw material from which bioplastic can be made, as protein-based bioplastic has also shown excellent properties when applied in an injection mould (Felix *et al.*, 2016). But starch is still the most affordable, and readily available raw biopolymer that can be easily converted into bioplastic by acidic hydrolysis and non-volatile plasticizer which are also very affordable, also available Varda *et al.*, (2014).



**Scheme 1: Proposed Hydrogen Bond interaction between the Matrix of Starch and sucrose (SUC)**

The process involves in the preparation of polyhydroxylalkanoate PHA from microorganism is very tedious and costly in time and resources when compared to thermoplastic starch. Amongst all the biobased-biodegradable bioplastics, thermoplastic starch represents a single most feasible alternative to replace Petro-plastic since its properties can be tailored to specific with the help of additives such as plasticizer and flexibility. Most of the bioplastic found today in the market are made from starch, and represent about 85-90% of bioplastics in the global market (Bastioli, 2000). The most widely investigated and used polyol in thermoplastic starch production is probably glycerol, and this may be due to its high safety profile in food and biomedical applications, inexpensive, and non-volatile nature (Mekonnen *et al.*, 2013). Mechanical properties and water barrier are two properties of polymer that influence the choice and end application of these materials both domestically and industrially. The use of Ammonium salt and urea as a plasticizer in thermoplastic starch has showed improved mechanical property than HDPE and produced a very transparent bioplastic under hot compression mould which is also recyclable (Abbot *et al.*, 2012). The drive to switch to sustainable plastic from the conventional is not yet a global practice, like the most countries, especially sub-Saharan African is yet to key into the potential of renewable material as a source of raw a materials for bioplastic production may be due to its dependence on the food source for starch needed for bioplastic production. But starch derived from mango seed kernel is a promising alternative for raw biomaterial for application in bioplastic production, as the seed are discarded after consumption and treated as Agricultural waste though compostable still added to the pollution problem. and will not required additional cultivation of mango plant on

scarce land, since it exists in abundance in most countries in the world (Fowomola, 2010), and (Kittiphon, 2012), since other plant sources such as corn, cassava, may require the land expanse of land could result in possible deforestation (Abbot *et al.*, 2012). Therefore, this study will involve the use of starch extracted from waste Mango seed kernel, plasticized with Sucrose and reinforced with CMC in order to synthesize thermoplastic starch comparable with Petro-plastic in mechanical property. The stress, strain, percentage extension, and young modulus will be measured and compared for significant differences with the selected Petro-plastic films of PU, PVC, LDPE, and HDPE.

## MATERIALS AND METHODS

Mango starch powder and chemicals used was obtained from laboratory faculty of pharmaceutical science of Usmanu Danfodiyo University Sokoto and was used in this study without further modification or purification.

### a. Preparation of bioplastic derivative films

With some modification, the method described by Wissinger *et al.*, 2016 was adopted. To a 50 cm<sup>3</sup> beaker, 13.00 cm<sup>3</sup> of distilled water was added; follow by 1.25 g starch power, 2.0 cm<sup>3</sup> of HCl was added, followed by sucrose, and carboxymethyl cellulose (CMC) as shown in table 1 below. The mixture was placed on a magnetic stirrer, and heated and stirred slowly on a hot plate until it gets to 70°C. The mixture started out white in colour and change to transparent or translucent and thickens. Once the initial white colour of the starch has completely changed and the mixture had thickened, 2.0 cm<sup>3</sup> of NaOH<sub>(aq)</sub> was added to neutralized the acid (same concentration with HCl<sub>(aq)</sub> acid ) and removed from the heat. The hot mixture was slowly cast into a labelled Petri-dish and was dried in the oven for 12 hours at 70 °C.

**Table 1: The Composition of Sucrose Plasticised Bioplastic Derivatives (sample)**

Sample code	Plasticizer Type	Plasticizer (%)	CMC (%)	HCl (M)
B1	SUC	180	30	0.1
B2	SUC	80	70	0.1
B3	SUC	80	30	0.1

B: bioplastic derived from sucrose, SUC = sucrose, CMC: carboxymethyl cellulose

**b. Mechanical properties Determination**

**i. Tensile Strength (Stress)**

The mechanical properties were ascertained using a locally fabricated testing machine, according to the ASTM D882-09 standard method. A dumbbell-shaped template was made from flat iron bar to cut the test bioplastic derivatives into a dumbbell shape to length, thickness, width measured using a Vernier calliper and micrometer screw gauge respectively; the bioplastic specimen was examined to find the area free of defects such as small tears, ridges, air bubbles, curves, etc. A portion of the sample was cut out using the template.

The sample was carefully placed, secured and fastened at both ends of the dumbbell on the clamp of the tensile strength measuring machine. At one end of the movable clamp, a string was attached, and the other end of the string, a plastic container was attached. Gradually water was added to the container in very small quantity until the sample breaks around the middle, and the weight of the water added plus container was recorded as the weight that broke the sample, also the elongation at the point of breaking was recorded.

The cross-sectional area in square meters was calculated by converting the thickness and width from millimeters to meters. Tensile Strength (Stress) was calculated using the equation (i):

$$\text{Tensile Strength (Stress)} = \frac{\text{Force (N)}}{\text{Area (m}^2\text{)}} \text{----- (i)}$$

Convert tensile strength from Pa to GPa. Where 1GPa = 1 x 10<sup>9</sup> Pa

**ii. Elongation at Break and Strain**

The elongation at break (ε) is the maximum elongation that the specimen can reach when pulled, before failure. It is measured as the ratio between the original length of the specimen and

length at the moment of break. It expresses the capability of a material to resist changes of shape without crack formation.

The strain is the maximum extension the bioplastic can stretch under tension, while, Elongation at break is an indication of bioplastics flexibility and is expressed as a percentage. Percentage Elongation at break was calculated as follows using equation (ii), and (iii):

$$\text{Strain} = \frac{\text{Extension(m)}}{\text{Initial Length (m)}} \text{----- (ii)}$$

$$\% \text{ Elongation at break } (\epsilon) = \frac{\text{Extension}}{\text{Initial Length}} \times 100 \text{ -- (iii)}$$

**iii. Young Modulus**

This is the ratio of stress to strain, and this property defines the ultimate application of polymer. This property was calculated using the equation (iv) shown below;

$$\text{Young Modulus} = \frac{\text{Stress(GPa)}}{\text{Strain}} \text{----- (iv)}$$

**c. Determination of Hydrogen-Bond formation with Fourier Transformed Infrared Spectroscopy (FT-IR) analysis**

The sample was dried and IR spectra recorded with an Agilent-FTIR spectrometer, at 27°C from 650 to 4000 cm<sup>-1</sup> range, background scanned at 64, sample scanned at 32, and the resolution was 4, and fitted with a compatible PC running recommended operating system as described by Maulida *et al.*, (2016)

**d. Statistical Analysis**

The outcome of the experiment was analyzed for the significant difference using analysis of variance (ANOVA). The multiple comparisons were performed by Fisher LSD method with 95% confidence interval using Minitab(R) statistical software (version 17). The statistical significant difference was declared at p < 0.05.

**RESULTS AND DISCUSSION**

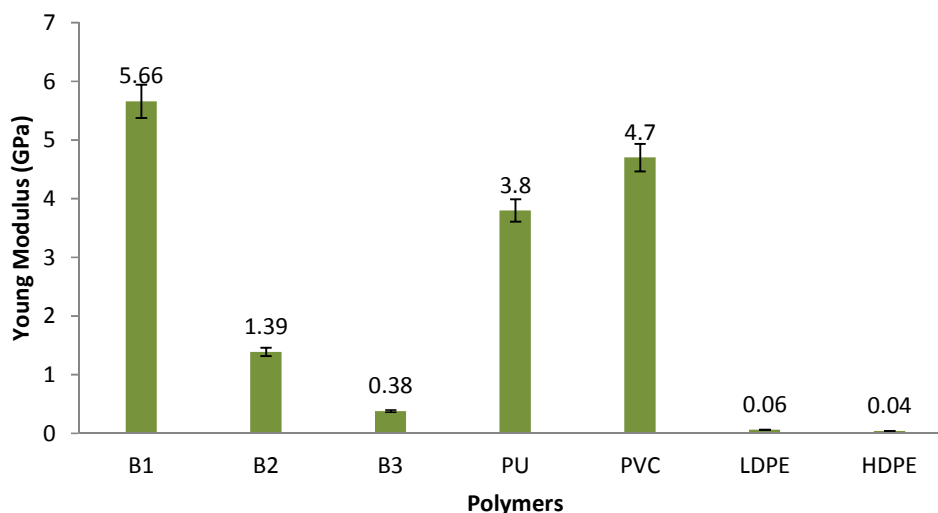
The result from Figure 1 and Table 1 shows that B1 has a significantly high Young Modulus for thermoplastic starch, compared with that of PU, PVC, LDPE, HDPE, all measured under the same condition. B1 indicates a young modulus that is significantly higher than B2 and B3, while B2 has a higher young modulus than B3 but are not significantly different ( $p < 0.05$ ). It was observed that B1 thermoplastic derivatives have the higher young modulus of 5.658 GPa than that obtained for PVC (4.682 GPa), and PU (3.771 GPa) but show no significant difference and significantly higher than that of HDPE (0.049 GPa), and LDPE (0.063 GPa) ( $p < 0.05$ ). B2 and B3 indicated a young modulus that is significantly lower than PU and PVC, but showed a young modulus that is higher than LDPE and HDPE with no significant difference ( $p < 0.05$ ). This implies that the bioplastic B1 is stiff, strong, and possess mechanical strength that share similarities with the PVC and PU and could be used in similar applications with some medication perhaps in physical properties. B2

and B3 are suitable for similar application like packaging materials as HDPE and LDPE since they show even higher young modulus. The higher strength of sucrose plasticized bioplastics (B1 and B2) could be attributed to the chemical structure and composition of sucrose. Sucrose is polyols, disaccharides, these multiple hydroxyl groups present contributes immensely in the intermolecular cross-linking with the starch biopolymer chain, thereby improving the overall strength of the bioplastics. Hydrogen bonding plays a vital role in the overall strength of the bioplastic, as a gives a more composite biopolymer, and this strength improves as the numbers of hydroxyl group increases, and this observation in higher young modulus, is a result of higher adhesion due to similarity in the matrix and fiber polarities of the reactants been carbohydrate all through i.e. starch, sucrose, and CMC as suggested by Van de-Velde and Piekens (2002). Ultimately the young modulus as observed increased with higher amount of sucrose.

**Table 2: The result for Mechanical Property of Bioplastic derivatives and the Standard Petro-plastics**

Sample	Stress (mpa)		Strain (cm)		Young modulus (GPa)		Elongation (%)	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
B1	934.215	63.314	0.166	0.012	5.658	0.796	17.302	0.419
B2	447.002	51.147	0.330	0.055	1.397	0.372	32.95	5.490
B3	271.654	64.444	0.708	0.141	0.384	0.055	70.80	2.410
PU	363.778	120.952	0.106	0.040	3.771	1.655	10.556	0.429
PVC	257.949	99.273	0.059	0.016	4.682	2.482	5.897	1.554
LDPE	146.123	31.274	2.908	1.963	0.063	0.003	290.754	1.441
HDPE	44.426	8.453	0.933	0.288	0.049	0.006	93.302	1.057

KEY: PU = Polyurethane, PVC = Polyvinylchloride, LDPE = low density polyethylene, HDPE = High density polyethylene.



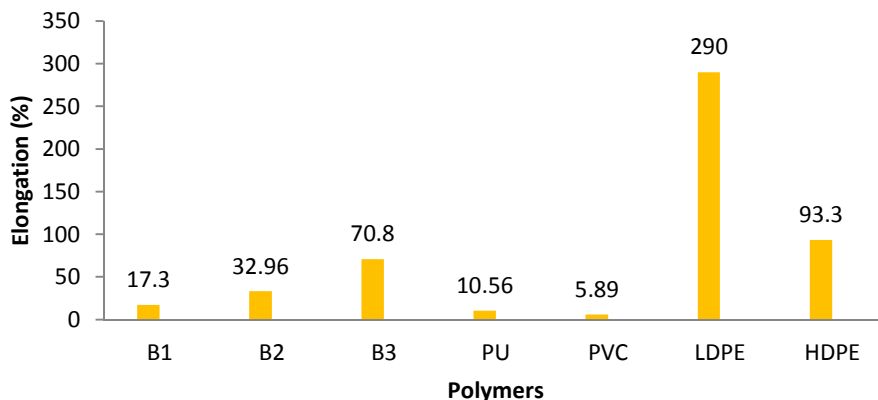
**Figure 1: Compares Young modulus of Bioplastic derivatives and Standard Petro-plastics**

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Compared to bioplastic filled with nanofiller Zinc Oxide as reinforcement produced a material with diminishing young modulus with glycerol as plasticizer (Harunyah *et al.*, 2017) which is inconsistent with this investigation when CMC and sucrose as plasticiser. The results obtained by Eterigho *et al.*, 2017, show that blending with PVA at 80% produces bioplastic with young modulus of 000.31 GPa, while this study obtained young modulus of 0.5199 GPa at 0 %, 0.4863 GPa at 30 %, 1.3971 GPa at 70 % CMC, this goes to show that CMC it is a better reinforcement material when compared to PVA and ZnO nanofillers, this can be attributed to a high degree of Hydrogen bond and intermolecular interaction within the matrix biocomposite. According to Khurmi and Sedha (2012), the range of obtainable young modulus for Petro-plastics (HDPE, LDPE, PU, or PVC e.t.c.) is between 0.07 to 4.00 GPa, which is in agreement with the bioplastic derivatives, and the standard Petro-plastic film obtained for this studies for sample B1 that show a young modulus of 5.66 GPa that is significantly higher than 4.68 GPa of PVC film observed for this study and agrees with values stated by Khurmi and Sdha (2012).

Figure 2 and Table 1, shows the percentage elongation of the sucrose plasticised thermoplastic mango starch compared with petro-polymers which includes PU, PVC, LDPE, and HDPE. The results show that the percentage elongation of the bioplastic derivatives increases significantly when sucrose (80 to 180 %) 17.30 % in B1 to 70.80 % in B3 and CMC (70 to 30 %) in B2 and B3 was significantly decreased from 32.96 to 70.80 % ( $p < 0.05$ ). but varies from the petro-polymer PU and PVC has significant difference in the percentage elongation of 10.56 and 5.89 %, are significantly different from

LDPE and HDPE. LDPE showed the highest percentage elongation of 290.8 %, and PVC is lowest at 5.9 % ( $p < 0.05$ ). It was observed that has the highest derived percentage elongation is 70.80 % for B3 which has is significant difference from all other bioplastics derivatives ( $p < 0.05$ ). Sucrose derived bioplastics behave more like composite and show little elongation and only breaks at high tensile stress and have lower percentage elongation, but higher young modulus i.e. displaying an inverse relationship. From Table 3, shows the FTIR spectra corresponding to bioplastic derivatives, and starch. The bioplastic derivative spectra corresponding to the band of starch, CMC and sucrose i.e., bands at 920 and 1148  $\text{cm}^{-1}$  (Mendes *et al.*, 2016). it was observed that the presence of H-bonding network has resulted to more broaden and discrete low-frequency peaks as seen the region of 3600 – 3000  $\text{cm}^{-1}$  wavenumber in spectra obtained for all the bioplastic derivatives, compared to hydrogen bond observed in the starch spectrum to have less discrete peak and a shift of the stretching frequency in the region of 3500- 3070  $\text{cm}^{-1}$ . Starch biopolymer spectrum show in a region of 970 and 1200  $\text{cm}^{-1}$  and this was observed in all derivatives of bioplastics. The presence of carboxylate ( $-\text{COO}-$ ) in the CMC gives the strong bands at about 1646.30, 1422.55 and 1360  $\text{cm}^{-1}$  which is line with absorption band obtained by Jiang *et al.*, (2011) but is almost absent in starch. The symmetrical stretching vibration for  $-\text{CH}_2-$  was observed with an intense peak at 2929.97  $\text{cm}^{-1}$  in all the variety of bioplastic derivatives also observed in the unplasticized starch but at less broaden peaks but is similar to 2914 observed by Mendes *et al.*, (2016).



**Figure 2: Compares Percentage Elongation of Bioplastic derivatives and Standard Petro-plastics**

**Result of FTIR Analysis for Sucrose Plasticised Bioplastic derivatives.**

**Table 3: FTIR Spectra of Pristine Mango Starch, and sucrose plasticised bioplastic derivatives**

Functional Group	Wave number (cm <sup>-1</sup> )			
	Mango	B1	B2	B3
O - H	3260.84	3336.85	3340.58	3342.62
C - H	2937.84	2925.64	2931.60	2984.17
C = O	1646.77	1650.34	1649.55	1668.46
C - O	1041.12	1037.16	1028.98	1075.49

**CONCLUSION**

Result has shown that thermoplastic starch with higher comparable mechanical properties to PVC, and PU, HDPE, and LDPE could be derived from Mango starch plasticised with sucrose and reinforced with CMC. The derived sucrose plasticised bioplastics have an edge of better the rate of biodegradation in case it is used as a single-use-plastic compared to petro-plastic which are the major contributors to environmental pollution which have negatively affected land and marine ecosystems (UNEP, 2015). Therefore, it can be concluded that

sucrose plasticized bioplastics could be applied in the production of biodegradable bioplastics cups, spoon, planting bags plate and other single-use that requires high tensile materials, these bioplastics derivatives could be applied in production of packaging bag, mulching bag and other single-use purposes with some modification. Therefore, mango starch could be utilized to synthesize bioplastic derivatives that are affordable, available, with suitable mechanical properties that are significantly comparable with some Petro-plastics.

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**Appendix**

Table 4: Basic Statistical Analysis for Young Modulus of Sucrose plasticised Bioplastics and selected petroplastics

<b>Runs</b>	<b>B1</b>	<b>B2</b>	<b>B3</b>	<b>PU</b>	<b>PVC</b>	<b>LDPE</b>	<b>HDPE</b>
1	5.6240	1.4161	0.3278	4.0258	3.9412	0.0640	0.0428
2	4.8791	1.0155	0.4372	5.2840	7.4501	0.0592	0.0547
3	6.4700	1.7596	0.3862	2.0036	2.6557	0.0657	0.0491
<b>Mean (GPa)</b>	5.6577	1.3971	0.3837	3.7711	4.6823	0.0630	0.0488
<b>SD</b>	0.7960	0.3724	0.0547	1.6550	2.4816	0.0033	0.0060
<b>SE</b>	0.6336	0.1387	0.0030	2.7390	6.1585	0.0000	0.0000

SD: Standard deviation, SE: Standard error

Table 5: One Analysis of Variance for Young Modulus using the Fisher LSD Method and 95% Confidence

<b>Factor</b>	<b>N</b>	<b>Mean (GPa)</b>	<b>SD</b>	<b>95% CI</b>	<b>Grouping</b>
B1	3	5.658	0.796	(4.202, 7.113)	A
B2	3	1.397	0.372	(-0.059, 2.853)	B
B3	3	0.3837	0.0547	(-1.0719, 1.8394)	B
PU	3	3.771	1.655	(2.316, 5.227)	A
PVC	3	4.68	2.48	(3.23, 6.14)	A
LDPE	3	0.06297	0.00337	(-1.39266, 1.51859)	B
HDPE	3	0.04887	0.00595	(-1.40676, 1.50449)	B

N: Experimental run, SD: Standard Deviation, CI: Confidence Interval.

Pooled SD = 1.17551. Means that do not share a letter are significantly different.

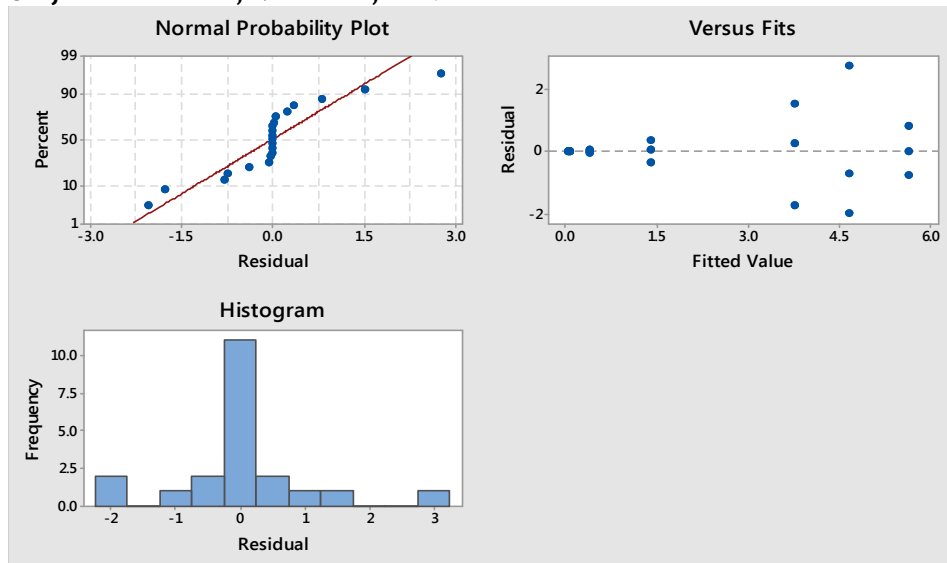


Figure3: Residual Plots for Young Modulus of Sucrose plasticised Bioplastics and Selected Petroplastics

Table 6: Basic Statistical Analysis for Percentage Elongation of Sucrose plasticised Bioplastics and selected Petro-plastics

Runs	B1	B2	B3	PU	PVC	LDPE	HDPE
1	16.8831	31.5068	72.2047	10.1429	5.0000	291.8292	94.3770
2	17.7215	39.0244	68.0253	10.5238	5.0000	291.3158	93.2650
3	17.3023	28.3333	72.1832	11.0000	7.6923	289.1167	92.2631
<b>Mean (%)</b>	17.3023	32.9549	70.8044	10.5556	5.8974	290.7539	93.3017
<b>SD</b>	0.4192	5.4906	2.4068	0.4295	1.5544	1.4409	1.0575
<b>SE</b>	0.1757	30.1472	5.7926	0.1844	2.4162	2.0762	1.1182

SD: Standard deviation, SE: Standard error

Table 7: One Analysis of Variance for Percentage Elongation using the Fisher LSD Method and 95% Confidence

Factor	N	Mean (%)	SD	95% CI	Grouping
<b>B1</b>	3	17.302	0.419	( 14.272, 20.332)	A
<b>B2</b>	3	32.95	5.49	( 29.92, 35.98)	B
<b>B3</b>	3	70.80	2.41	( 67.77, 73.83)	C
<b>PU</b>	3	10.556	0.429	( 7.526, 13.586)	D
<b>PVC</b>	3	5.897	1.554	( 2.867, 8.927)	E
<b>LDPE</b>	3	290.754	1.441	(287.724, 293.784)	F
<b>HDPE</b>	3	93.302	1.057	( 90.272, 96.332)	G

N: Experimental run, SD: Standard Deviation, CI: Confidence Interval.

Pooled SD = 2.44688. Means that do not share a letter are significantly different.



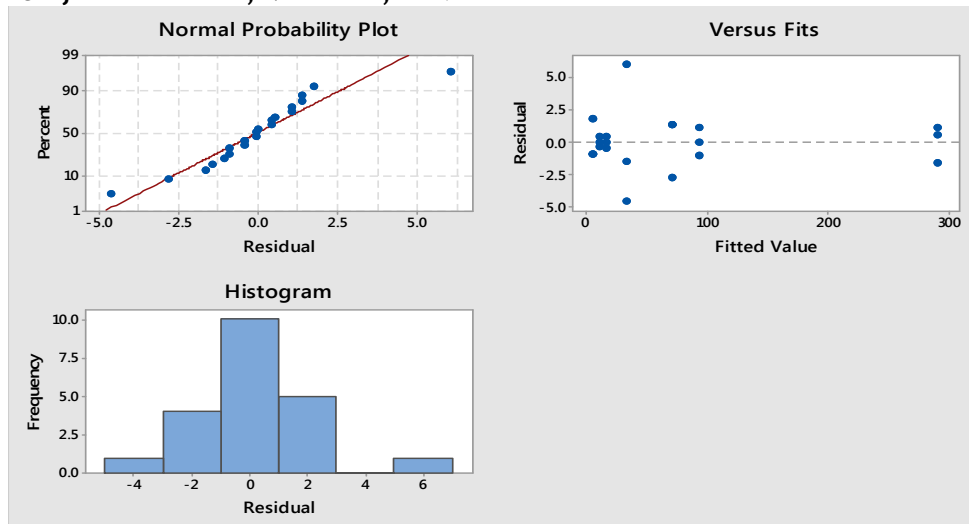


Figure 4: Residual Plots for Young Modulus of Sucrose plasticised Bioplastics and Selected Petroplastics

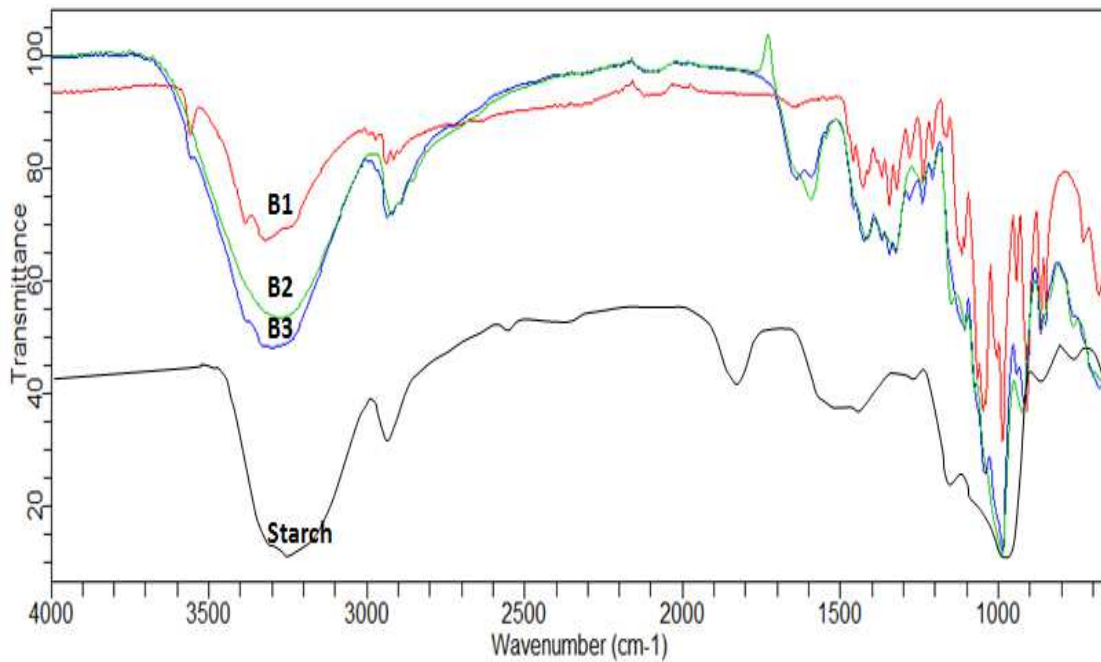


Figure 5: FTIR spectra for sucrose Plasticised Bioplastics (B1, B2, and B3) and Pristine Mango Starch