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Filler characterization, mechanical properties, x-ray diffraction and crosslink density analysis of starch/natural rubber biopolymer composites

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Starch fillers were extracted from three plant sources namely amora tuber, Tacca lentopeteloides; sweet potato, Ipomoea batatas; yam starch, *Dioscorea rotundata* and their particle size, pH, amylose, and amylopectin percentage decomposition determined accordingly. The starch was introduced into natural rubber in liquid phase (through gelatinization) by the latex compounding method and compounded according to standard method. The prepared starch/natural rubber composites was characterized by Instron Universal testing machine (UTM) for tensile mechanical properties. The composites was further characterized by x-ray diffraction and crosslink density analysis. The particle size determination showed that amora starch granules has the smallest particle size (156 × 47 μm) followed by yam starch (155 × 40 μm) and then sweet potato starch (153 × 46 μm) with the biggest particle size. The pH test also revealed that amora starch has a near neutral pH of 6.9, yam 6.8, and sweet potato 5.2 respectively. Amylose and amylopectin determination showed that yam starch has a higher percentage of amylose (29.68), followed by potato (22.34) and then amora starch with the lowest value (14.86) respectively. The tensile mechanical properties testing revealed that yam starch produced the best tensile mechanical properties followed by amora starch and then sweet potato starch. The structure, crystallinity/amorphous nature of the product composite was confirmed by x-ray diffraction, while the nature of crosslinking was confirmed by swelling test in toluene solvent using the Flory-Rehner approach. The increasing values of crosslink density in the starch/rubber composite is a clear evidence of good interfacial adhesion between the starch fillers and the rubber, hence good dispersion of starch fillers in the rubber. This research has rendered a workable strategy for enhancing interfacial interaction between a hydrophilic filler (Starch) and hydrophobic polymeric matrix (natural rubber) yielding moderately good tensile mechanical properties with prospects for the rubber processing industry. The studied fillers can partially replace carbon black as natural rubber fillers with reduced cost, no risk to human health and also an environment friendly approach.

Key words: Natural rubber, fillers, starch, amylose, amylopectin, crosslink density, mechanical properties.

INTRODUCTION

Starch obtained from natural renewable resources has many advantages such as low cost, abundant supply and environment friendliness. Ways to find out its other potential applications on a big scale in the rubber

processing industry has attracted much interest (Zhang et al., 2006). In an effort to reduce dependence on expensive conventional fillers like carbon black, and silica, much research effort have been exerted on developing starch-based natural rubber polymer materials with moderately good tensile mechanical properties (Zhang et al., 2006). During the last few decades, the effects of different types of fillers on natural rubber have been studied, in search of improvements on its physical and tensile mechanical properties (Ichazo et al., 2007). Several organic fillers such as starch, ground wood waste, bamboo, white rice husk, saw dust have been used as fillers and fibres for plastics and elastomers, (Ichazo et al., 2006). Starch is a biopolymer consisting of amylose and amylopectin, present in most plants and in considerable amounts (Ichazo et al., 2006, 2007).

More so according to patent literature, starch has been creatively used as a filler in tyre compounds to reduce cost and improve properties (USP, 1997, 2001, 2002, 2003). Goodyear Tyre Company has utilized a new starch-based material called BioTRED to partially replace the conventional carbon black and silica in producing a new brand, GT3 tyres to reduce tyre weight, rolling resistance and simultaneously decrease the energy consumption and cut down the emission of carbon (iv) oxide in car automobiles (Zhang et al., 2004).

The mechanical properties of starch-filled rubbers actually depend on many factors among which the most important ones are the dispersion level, interfacial strength and the type of starch (Zhang et al., 2004). However the polar surface, high melting point and large particle size of starch make it difficult to finely disperse starch in a rubber matrix by direct blending method (Zhang et al., 2004). Therefore incorporation of starch into rubber by direct blending results in a micro-composite with large, dispersed particles, which causes significant deterioration of the mechanical properties of the vulcanizates, especially the resistance to abrasion and tear properties (USP, 2002; Sandstrom, 2001). Report was also found that the amylose content of starch can affect mechanical properties of starch-filled rubber blends (Buchanan et al., 1971) in Rouilly et al. (2004). Differences in amylose content have been reported to result in differences on starch physicochemical properties (Schoch and Maywald, 1968; Jane et al., 1999). More so amylose plays a role in the pasting properties of starch thus affecting the starch viscoelastic properties (Charles et al., 2004; Novelo-Cen and Bentacur- Ancona, 2005). Also natural rubber latex has been found to contain natural stabilizers (that is, proteins and lipids) which should help compatibilization with natural starch (Rouilly et al., 2004; Bhatnagar, 2004).

In order to utilize all the above mentioned advantages of starch and prevent unlikely and undesirable properties in a starch/natural rubber composites, prepared by direct bending, a workable strategy was adopted in the study introducing natural starch into natural rubber without any modification by coupling agents or compatibilizers and using a latex compounding method (LCM) after having gelatinized the starch to reduce its particle size in order to achieve a fine dispersion in the rubber matrix. Thus this research has used starch fillers with good outcome and have also rendered a workable strategy for enhancing interfacial interaction between a hydrophilic filler (starch) and hydrophobic polymeric matrix (natural rubber) yielding polymer composite with moderately good mechanical properties and hence the starch fillers used can partially replace carbon black and silica in tyre products pending further investigations.

More so, for many years carbon black has been the most widely used reinforcing fillers in the rubber industry. However owing to its origin from petroleum and high cost, its use in rubber products not only consumes energy, but its also harmful to the environment and human health .The finely divided particles of carbon black causes easy lung damage and occupational illnesses such as cancer and kidney diseases over prolonged exposure for rubber compounding operators in the rubber processing industry, (Li and Ryong, 2012; Stelescu et al., 2017). Therefore it is highly expedient and of paramount importance to develop new cheap, clean, renewable, biodegradable and environment friendly reinforcing fillers for natural rubbers with application in tyre treads for automobile tyre production.

EXPERIMENTAL

Local varieties of starch, crops, sweet potato, and yam were purchased from agricultural food market in Mubi, Adamawa State, Nigeria while the amora crop (a wild variety of Irish potato) was harvested from a farmland in Paiko village, Gwagwalada Area Council FCT, Abuja. Natural rubber latex (dry rubber content, 30%) was obtained from Rubber Research Institute of Nigeria (RRIN), Iyanomo, Benin, Edo State, Nigeria. Coagulant and compounding agents were purchased from the chemical market as well as the toluene for the swelling test.

Starch extraction

Starch was extracted from the plant source by the method adopted from Vasanthan (2001).

Particle size and pH determination

Particle size of the starch granules were determined using

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Table 1. A typical formulation of the blended starch/rubber composites in part per hundred rubber (Phr).

Ingredients	Contents (Phr)
Natural rubber	100
Starch filler	10,20,30
Zinc oxide	5
Stearic acid	2
MBT	3
TMQ	1
Sulphur	3
Carbon black	30

Ro Control A: Natural rubber + carbon black filler (30 Phr)
NRO Control B: Natural rubber without any filler
Key: MBT -Mercaptobenzothiazole (Captax), TMQ –Tetramethylquinoline.
Source: Uzoh, Buba et al (2018).

Olympus electric binocular microscope at four objectives lens magnification. The pH was also determined using Medfielddigital pH meter at 1 g in 25 cm^3 of distilled water.

Amylose/Amylopectin percentage composition

The amylose and amylopectin percentage composition in the starch samples were determined using High Performance Liquid Chromatography (HPLC) according to the Standard Methods of Association of Official Analytical Chemists (AOAC, 2010) 18th Edition, Washington DC.

Gelatinization procedure

The gelatinized starch was prepared according to the method adopted from Zhang et al. (2004).

The preparation of starch/natural rubber coagulum

The gelatinized starch and the rubber latex were mixed and stirred vigorously for thirty minutes then about 10% of calcium chloride aqueous solution was added to co-coagulate the rubber latex and gelatinized starch mixture. The coagulum was then washed several times with water and dried in a dry oven at 80°C for 18 h period (Zhang et al., 2004). A control sample, Ro, (containing only natural rubber) was also prepared.

Compounding procedure

The recipe for the preparation of the starch/natural rubber blend is given in Table 1. The compounding procedure was done in accordance with the American Society for Testing and Material (ASTM). Designation D3184-80 compounding was done on two-roll mill. The control sample, Ro was compounded with carbon black at 30 (Phr) part per hundred rubber. After the two-roll compounding, the rubber blend was taken to the hydraulic press for vulcanization at 140°C for about five minutes. Tests samples of dumbbell shapes were punched out from the vulcanized rubber sheets for mechanical testing on Instron Universal Testing Machine (UTM).

Characterization of starch-filled natural rubber composites

Tensile mechanical property test

Tensile mechanical properties of the prepared starch/natural rubber composites were determined using a Lloyd Instruments Machine, Instron Universal Testing Machine, Model EZ20 according to ASTM D412 at a speed rate of 500 mm/min using three dumbbell test piers.

X-ray diffraction analysis

X-ray diffraction (XRD) analysis was performed using a Schimadzu 6000 model X-ray diffractometer using a Cu target at 40 kV, 30 mA, run between 0° to 120° theta (θ) Bragg angle (Scam range) at a rate of between 2 to 10° degree per min.

Swelling tests

Swelling tests on the starch/natural rubber vulcanisates were performed in toluene in accordance with ISO 1817. The cured test pieces (round-shaped pieces) were weighed in a weighing balance and swollen in toluene until equilibrium which took 72 h at room temperature. The samples were taken out from the liquid, the toluene was removed from the sample surfaces and the weight was determined. The samples were then dried in the oven at 60°C until constant weights were obtained. The swelling results were used to calculate the molecular weight between two crosslink (Mc) and the degree of crosslink density (Vc) applying Flory-Rehner Equation (Flory and Rehner,1943).

$$M_c = \frac{-P_p V_s V_r^{\frac{1}{3}}}{\ln(1-r) + V^r + xVr^2}$$

(1)

$$V_r = \frac{1}{1 + Q_m}$$

(2)

$$V_c = \frac{1}{2M_c}.$$

(3)

Table 2. Particle size, pH, amylose and amylopectin composition of the starch samples.

S/N	Source of Starch	Particle Size (µm)	pH	Amylose (%)	Amylopectin (%)
1	Amora	156×47	6.9	14.86	84.97
2	Potato	153×46	5.2	22.34	77.64
3	Yam	155×40	6.8	29.68	70.29

Source: Uzoh, Buba et al (2018).

Table 3. Physical and mechanical properties of amora, starch-filled natural rubber.

Composite Starch/natural rubber composite (Phr)	Tensile strength (MPa)	Elongation at break	Hardness (IRHD)
R_O	87.6	6807	66-68
10	13.3	4850	56-57
20	8.9	2883	52-53
30	57.4	10007	47-48
NRo	21.9	17069	20-21

Key:R_O = Natural rubber with carbon black filler (Control sample A) Phr = Part per hundred rubber.
NR_O = Natural rubber without any filler (Control B) IRHD = International rubber hardness degree.
Source: Uzoh, Buba et al (2018).

Where p, is the rubber density (p of NR = 0.92 g/cm^3) $V_S$is the molar volume of the toluene (V_S=106.4 cm^3/mol) V_r is the volume fraction of the polymer in the swollen specimen, Q_m is the weight increase of the blends in toluene and x is the interaction parameter of the rubber network solvent (x, NR = 0.393). Other swelling parameters determined include the mol% uptake of solvent (Q_t), swelling index percentage.

$$Q_t = \frac{\left(\frac{w_2 - w_1}{M_s}\right) \times 100}{w_1}$$

(4)

$$Swelling\ index\ \% = \frac{w_2 - w_1}{w_1} \times 100$$

(5)

Hardness test: Type A durometer gauge was used to measure the hardness of the rubber composite in international rubber hardness degree (IRHD) according to ASTM D2240.

RESULTS AND DISCUSSION

The results of tests and measurements are presented in Table 2.

**Discussion on particle size analysis, pH, amylose and amylopectin composition of the starch samples**

The particle size of amora starch is the smallest with 156 × 47 µm while the largest among the sample is sweet potato starch with 153 × 46 µm followed by yam starch (155 × 40 µm). These results are comparable to those previously reported by Vasiljevic (1999); Farhat et al. (1999) and Zaidul et al. (2007). The particle size of starch is one of the most important characteristics which may influence other physicochemical properties such as swelling power, paste clarity and water-binding capacity (Singh et al., 2003). Amylose and amylopectin percentage compositions of the starch samples are also presented in Table 2. Yam starch has the highest percentage composition of amylose (29.68) while amora starch has the lowest percentage amylose composition (14.86) (Figure 1a and b). The amylopectin percentage compositions are 70.29 and 84.97 for yam and amora respectively. The pH of the starch samples are as follows: amora 6.9, sweet potato 5.2 and yam 6.8 respectively (Tables 3 to 5, Figures 2a and b, 3a and b).

Discussion on the effect of different starch fillers on the tensile strength, elongation at break and hardness of the starch filled natural rubber composites

The tensile strength of rubber or a rubber-like material (elastomer) indicates how much force or stress a rubber material can withstand before breaking. It is typically expressed as the amount of force in mega Pascal (MPa) in metric unit. It is important for a material that is going to be stretched or under tension from external force.

According to Table 3, the tensile strength of amora starch filled natural rubber are 13.3, 8.9 and 57.4 MPa at 10, 20 and 30 Phr respectively with their corresponding percentage elongation at break at 4850,
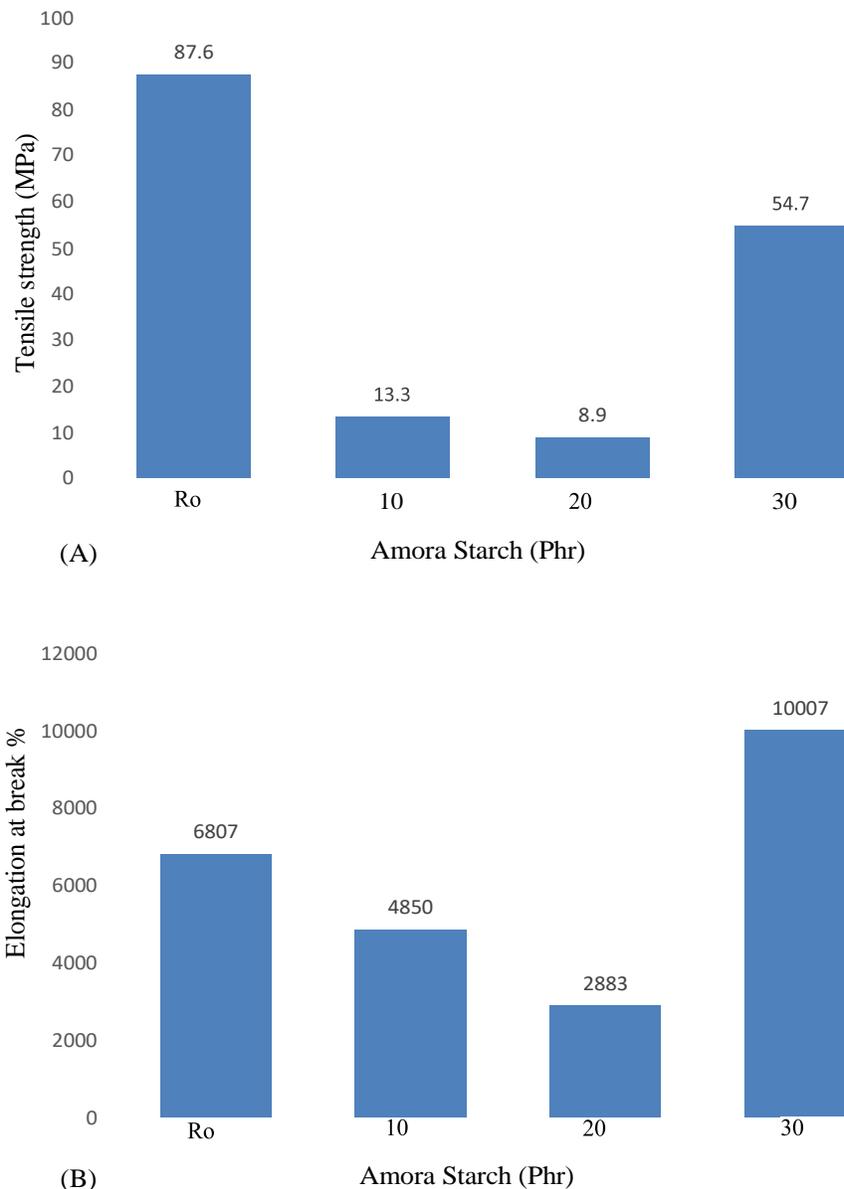
Figure 1. (a) Effect amora starch on tensile strength, (b) Effect of amora starch on elongation at break.
Source: Uzoh, Buba et al (2018).

2883 and 10007, while the control sample (R_O) with elongation at break of 6807. The starch loading at 10 and 20 Phr did not improve the tensile strength and percentage (%) elongation at break of the natural rubber under study with reference to the control sample Ro. But however at 30 Phr starch loading, significant increase was observed at 57.4 MPa with percentage elongation at break at 10007 when compared to the values of the control sample Ro. The tensile strength value of the control sample (Ro) reinforced with 30 Phr carbon black was still higher than the values of amora starch filled rubber composite which suggest that finely divided

carbon black has the value of 87.6 MPa and percentage carbon black is a better reinforcing filler than amora starch although amora starch filled natural rubber produced a high value of % elongation at break than the control sample, Ro.

The effects of the starch fillers on the tensile strength and percentage elongation at break of the starch/natural rubber composites were displayed on two histograms in figure 1a and figure 1b giving a pictorial illustration of the explained phenomena above. The introduction of potato starch fillers into natural rubber matrix as displayed in Table 4 produced improvement in tensile mechanical

Table 4. Physical and mechanical properties of potato starch filled rubber composite.

Starch/natural rubber composite (Phr)	Tensile strength (MPa)	Elongation at break	Hardness (IRHD)
R_O	87.6	6807	66-68
10	11.5	3137	54-55
20	12.9	3490	58-59
30	35.7	6654	52-53
NR_O	21.9	17069	20-21

Key: R_O = Natural rubber with carbon black filler (Control Sample A) Phr = Part per hundred rubber.
NR_O = Natural rubber without any filler (control B) IRHD = International rubber hardness degree
Source: Uzoh, Buba et al (2018).

Table 5. Physical and mechanical properties of yam starch filled natural rubber.

Composite Starch/natural rubber composite (Phr)	Tensile strength (MPa)	Elongation at break	Hardness (IRHD)
R_O	87.6	6807	66-68
10	9.5	2544	54-55
20	76.1	11274	57-58
30	80.4	12440	60-61
NR_O	21.9	17069	20-21

Key: R_O = Natural rubber with carbon black filler (Control Sample A) Phr = Part hundred rubber
NR_O = Natural rubber without any filler (control B) IRHD = International rubber hardness degree.
Source: Uzoh, Buba et al (2018).

properties at 30 Phr with 35.7 MPa and 6654 as tensile strength and percentage elongation at break. These values are lower than the values when compared to that of yam starch-rubber composite (80.4 MPa and 12440) and the values in the control sample, R_O (87.6 MPa and 6807).

According to Table 5, the reinforcing activity of yam starch started at 20 Phr loading and increased further at 30 Phr loading giving 76.1 and 80.4 MPa as tensile strength respectively and 11274 and 12440 as percentage elongation at break respectively though still below the tensile strength value of the control sample, R_O (87.6MPa). This reinforcing activity of yam starch, potato starch and amora starch might be attributed to the high percentage of amylose in their starch structure which supports the observation made by Buchanan et al. (1971) in Rouilly et al. (2004) that the amylose content of starch can affect mechanical properties in starch/natural rubber blends. The amylose content is highest in yam starch (29.68) followed by sweet potato (22.34) and then amora starch (14.86). However Buchanan's observation is not a general rule or principle and does not apply to every situation of increased value of amylose contents.

However another phenomenon emerged here which superimposes the effect of high amylose content in starch with near neutral pH such as in amora starch with low amylose (14.86) and with a near neutral pH (6.9) which produces a better reinforcing activity more than sweet potato with a higher amylose content (22.34) but with an acidic pH (5.2). Yam starch and amora starch has almost the same pH value (6.8) and (6.9) respectively. More so in all the rubber composite samples in Tables 3 to 5, hardness values increase as expected as increased filler content increases hardness in rubbers, (Stelescu et al., 2017). More so the observed effect of Starch fillers on the tensile strength and elongation at break was displayed on histograms in Figures 1a and b, 2a and b, and 3a and b respectively. As expected, increase filler loading reduces elongation at break as compared to unfilled rubber, NR_O (Control B) (Figure 4a, b and c).

X-ray diffractograms

From the x-ray diffractogram on Figure 4a, the control sample, R_O was not completely in amorphous state but has some crystalline region which was indicated by the diffuse diffraction peaks at 17°, 21° and 34° with the 17° (shoulder peak) and 34° weak peak appearing around 800 intensity (counts). The 21° peak was displayed at around 990 intensity indicating higher
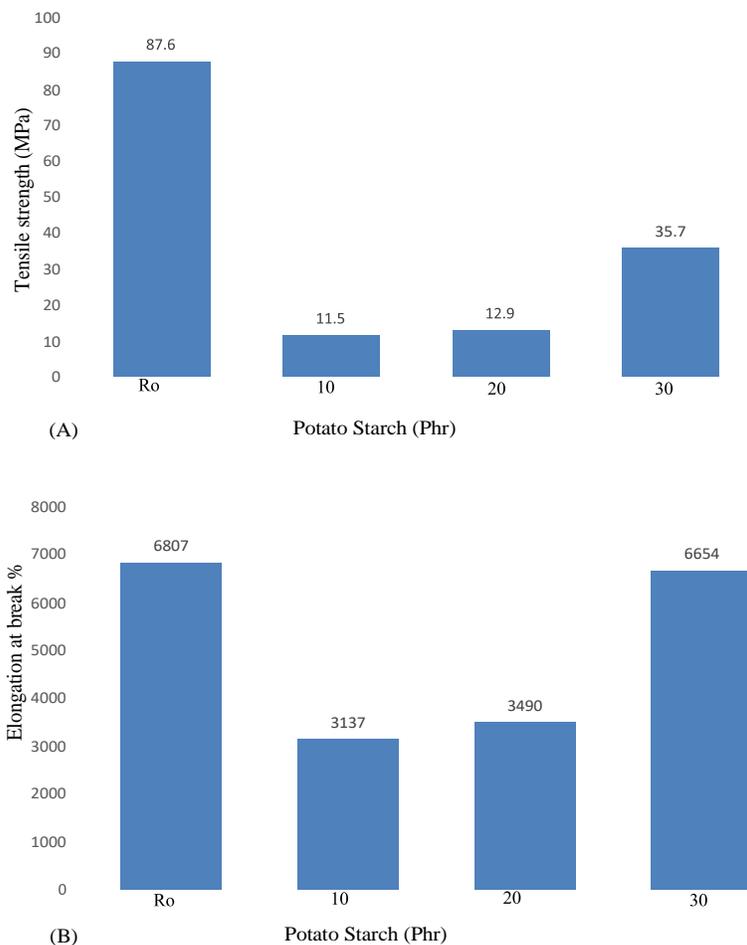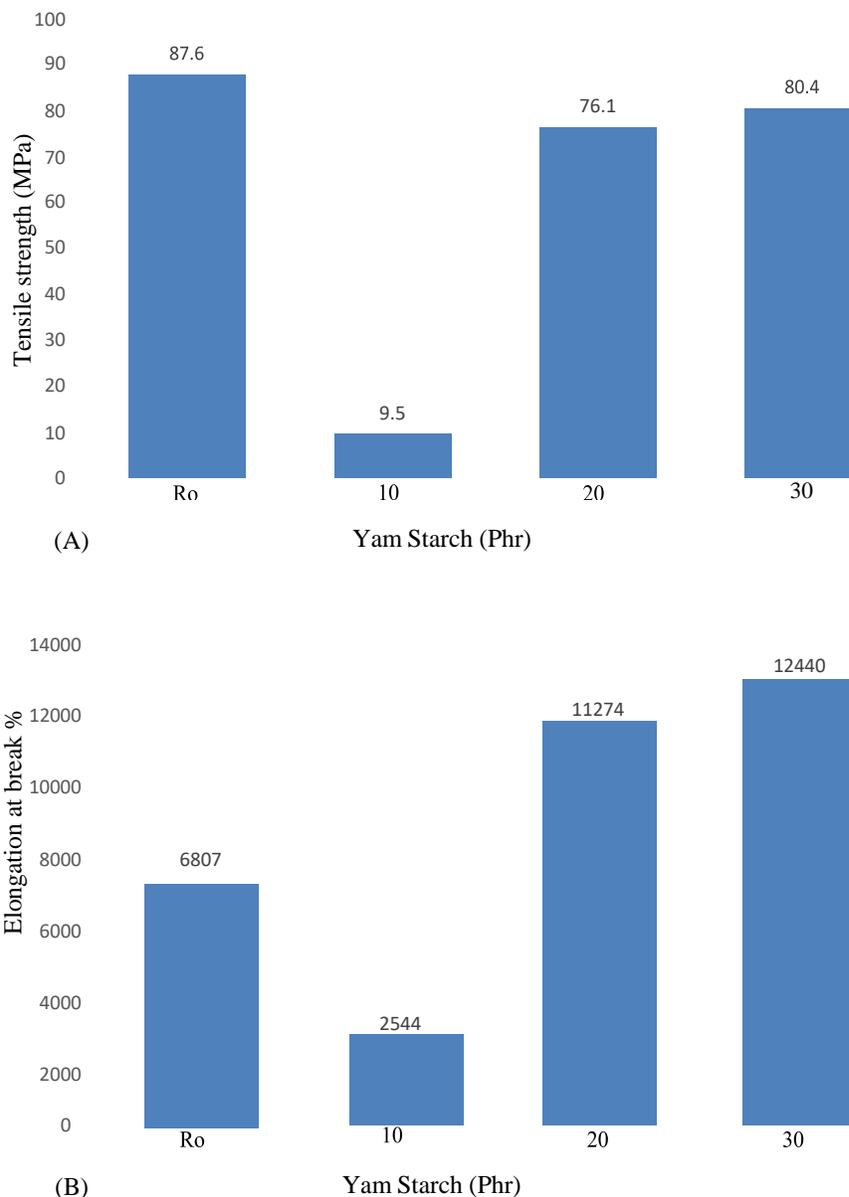
Figure 2. (a) Effect of potato starch on tensile strength, (b) Effect of amora starch on elongation at break.
Source: Uzoh, Buba et al (2018).

concentration.

According to Figure 4b, the diffraction pattern of sample A3, displayed a sharp broad diffraction peak at 21° at about 1450 intensity (counts) and a shoulder peak at26° with intensity of about 750. From the diffractogram of sample A3, the broad peak of 21° of intensity 1450 compared to 990 intensity at 21° peak for the control sample, Ro shows that the presence of starch filler (semi crystalline polymer) has increased the region of crystallinity in the rubber matrix and also in concentration (30 Phr) as indicated by the higher intensity at 1450 of the amora starch natural rubber composite. From the diffraction pattern of sample P3 (Potato starch/natural rubber composite in Figure 4c, it was seen that four sharp diffraction peaks at 15°, 17°, 18° and 23° with intensities at 1000, 1400 and 1450 were observed and two diffuse peaks at 33° and 38° with 800 counts (intensity). These four sharp peaks exhibited by the diffraction pattern of sample P3 is very similar to the diffraction pattern exhibited by starch x-ray diffractogramin a published work

of Zhang et al. (2004) and they also correspond to four out of five crystalline region of A- type starch (Campas-Baypoli et al., 2002). This behaviour suggest that there was recrystallization of the starch granules of the potato starch after gelatinization process and probably during the processing of the rubber material. More so, the peaks also appeared at higher intensities of 1000, 1400, and 1450 indicating higher concentrations at the crystalline region of the starch, natural rubber composite. Diffraction pattern of sample Y3 (Yam starch/natural rubber composite) displayed four (4) diffraction peaks at 15°, 17°, 18° and 23° at intensities of 600, 700, and 800, a shoulder peak of 27° at 500 intensity and two sharp and narrow diffraction peaks of 50° and 60° with intensities of 1450 and 1400 respectively. Fortunately three out of these four diffraction peak correspond to the crystalline region of A-type starch (Campas-Baypoli et al., 2002). More so the diffraction peaks at 50° and 60° in the diffractogram of sample Y3 are sharper and narrower with higher intensities showing an increase in the degree

Figure 3. (a) Effect of yam starch on tensile strength, (b) Effect of yam starch on the elongation at break.
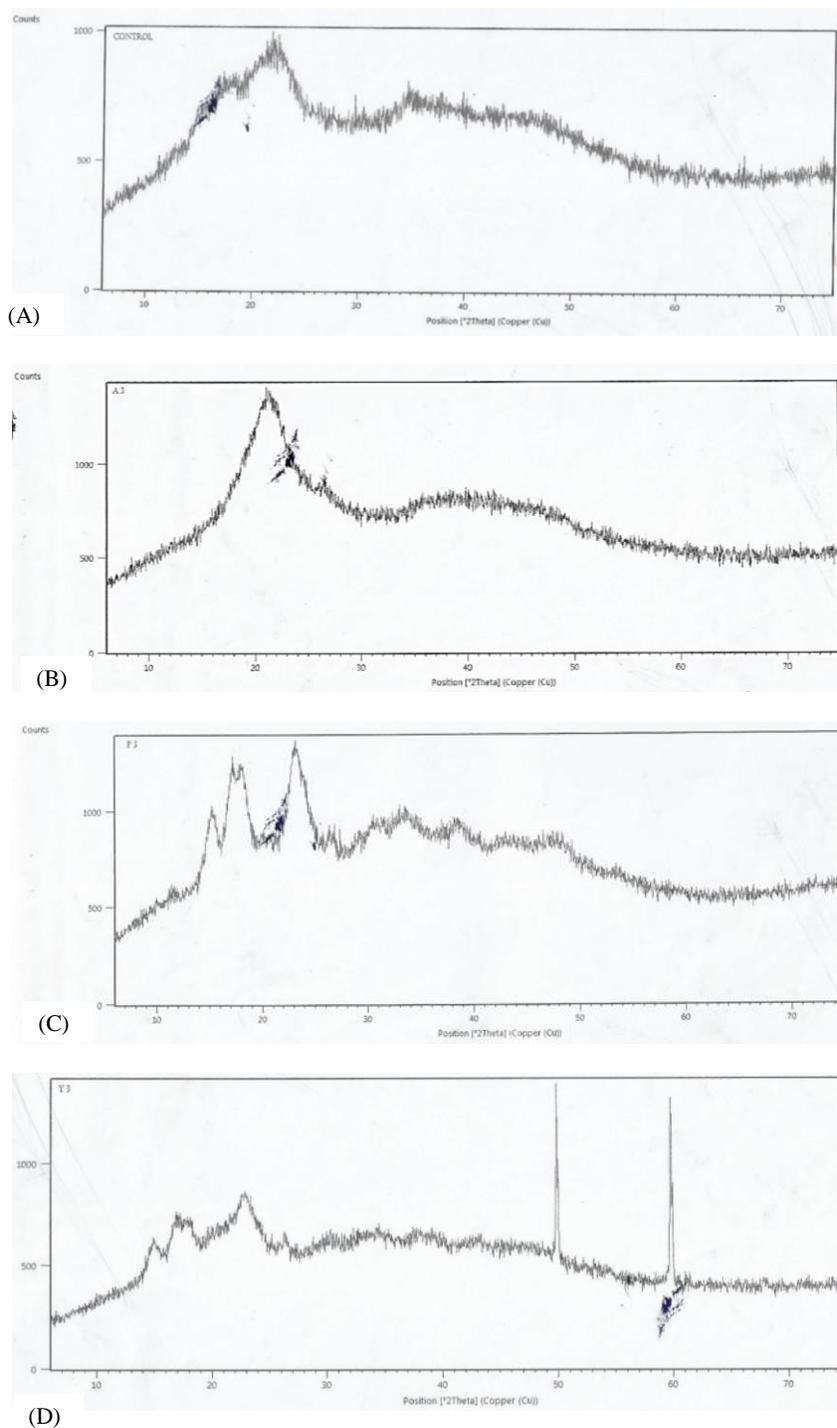Source: Uzoh, Buba et al (2018).

of crystallinity and a tendency to higher tensile strength Montane et al. (1998) and Mostafa et al. (2015). This behaviour also suggest that there could be recrystallization of the starch during the processing or that gelatinization procedure didn't remove the crystalinity of starch completely. This is also very similar to that observed in potato starch- natural rubber composite. This behaviour of the yam starch filler in the natural rubber composite accounts for the higher tensile strength produced by its reinforcing action on the rubber composite among all the starch fillers under study, as evidenced by the high amylose content in yam starch

filler, (Rouilly et al., 2004).

Swelling test parameters

According to Table 6, sample P3 potato starch/natural rubber composite has the highest value in Mol% uptake of the solvent Qt, swelling index %, swelling coefficient α, and molecular weight between two crosslinks, Mc while it has the lowest value in the degree of crosslink density, Vc. The control sample, Ro has the lowest value of mol % uptake of solvent Qt, swelling index %, swelling

Figure 4. (a) Control Sample Ro, (b) Amora starch/natural rubber composite, (c) Potato starch/natural rubber composite, (d) Yam starch/natural rubber.
Source: Uzoh, Buba et al (2018).

coefficient (α), and molecular weight between two crosslinks, Mc, but has the highest value in the degree of crosslink density, Vc. It was also observed that the higher molecular weight between two crosslinks, the lesser the value of the degree of crosslink density, Vc, and vice versa. In other words, the degree of crosslink density, Vc in $mol.cm^{-3}$ is inversely proportional to the molecular weight between two crosslinks, Mc that is, by a constant 0.5 which was applicable to all the samples at 30 Phr starch loading in toluene solvent irrespective of the type

Table 6. Swelling parameters in toluene for starch/natural rubber composites with maximum tensile mechanical properties.

S/N	Starch/ Rubber Composite (30 Phr)	Mol.% uptake of the solvent Qt	Swelling Index %	Swelling coefficient α	Degree of crosslink density Vc (Mol/cm^{-3})	Molecular Wight between two crosslink (Mc)
1	A3	2.40	221	2.55	44.0×10-5	113.62
2	P3	2.83	260	3.00	37.5 × 10-5	133.34
3	Y3	2.61	240	2.77	41.1 × 10-5	121.78
4	R0	1.49	137	1.58	51.9 × 10-5	96.36

Key: Phr = Part per hundred rubber
A$_3$ = Amora starch/natural rubber composite.
P$_3$ = Potato starch/natural rubber crop composite.
Y$_3$ = Yam starch/natural rubber composite
R$_O$ = Control sample.
Source: Uzoh, Buba et al (2018).

Table 7. Effect of amora starch filler on swelling parameter in toluene.

S/N	Starch/ rubber composite (30 Phr)	Mol.% uptake of the solvent Qt	Swelling Index %	Swelling coefficient α	Molecular Wight between two crosslink (Mc)	Degree of crosslink density Vc (Mol/cm^{-3})
1	A1	3.44	316.7	3.66	149.41	33.5 ×10-5
2	A2	3.32	305.3	3.52	151.1	33.1×10-5
3	A3	2.4	221	2.55	113.62	44.0 × 10-5

Key: A$_1$=Amora Starch at 10 Phr
A$_2$=Amora Starch at 20 Phr
A$_3$=Amora Starch at 30 Phr.
Source: Uzoh, Buba et al (2018).

of starch, so that for a particular starch/natural rubber composite. From Table 7, it was observed that as the starch filler loading increased from 10, 20 and to 30 Phr, Mol.% uptake of solvent, swelling index %, and swelling coefficient decreased in values. However at a highest value of molecular weight between two crosslinks, Mc 151.10, the degree of crosslink density is at the lowest value of 33.1×10^{-5} mol.cm^{-3} while at the lowest value of molecular weight between two crosslink, Mc at 113.62, the degrees of crosslink density is at the highest value of 440×10^{-5} mol.cm^{-5}. From observations on swelling test results, increase in molecular weight between two crosslinks (Mc) leads to lower values in degree of crosslink density (Vc). Also increasing starch loading from 10Phr to 30Phr leads to lower molecular weight between two crosslink, Mc and hence a higher value of degree of crosslink density, Vc in mol.cm^{-3}. The decrease in swelling parameters in the composites is caused by restrictions to swelling caused by the filler rubber- adhesion in the composites. More so the increase values of the crosslink density as the filler loading increased from 10 to 30 Phr suggest evidence of strong interfacial adhesion betweenthe starch filler and the rubber matrix, hence good dispersion of the starch

filler in the rubber matix (Cao et al., 2017; Wang et al., 2018).

Conclusion

Compounding rubber latex with gelatinized starch and directly co-coagulating the mixture with an electrolyte produced a starch natural rubber composite with moderately good mechanical properties. In an aqueous state, the rubber latex particles generally smaller than 100nm has been mixed with gelatinized starch uniformly giving starch/rubber composites with a fine dispersion. This research study has rendered a strategy for enhancing interfacial interaction between hydropholic starch filler and hydrophobic natural rubber through the latex compounding method (LCM) producing moderately good tensile mechanical properties. Effect of amylose in starch/natural rubber composites in enhancing mechanical properties was highlighted as well as the effect of near neutral pH in the reinforcing action of starch fillers. As evidence by the reinforcing action of amora filler with pH, 6.9. Moreso, with the good physical and mechanical properties exhibited by

the starch/rubber composite in comparison with the control A, Ro(carbon black filled rubber), amora, potato and yam starch filler has the potential to partially replace carbon black as filler in rubber tyres pending further investigation. Finally an environment friendly starch fillers has been developed that can help produce the risk to human health in terms of occupational illnesses caused by carbon black and silica to compounding operators in the rubber industry.

CONFLICT OF INTERESTS

ACKNOWLEDGMENTS

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