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Formulation and Characterization of Green Adhesive Using Agricultural and Plastic Waste Materials as Composites

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Abstract
The formulation and characterization of green adhesives using agro and plastic waste materials as composite for domestic and industrial utilizations was carried out. Preparation of adhesive filler (Rice Husk Ash) and formulation of adhesive binders (Polystyrene waste and Arabic gum) were conducted. Physicochemical investigation on carbonized rice husk, rice husk ash and arabic gum were performed. A synthetic adhesive (Sample A) served as a control. Some mechanical properties and FTIR were carried out. Adhesive samples formulated and the standard were labeled A, B, C, D, E, F and G respectively based on varied quantities of fillers and binders. Two important mechanical properties shear force and shear strength were investigated and compared with the standard synthetic adhesive (Sample A). The results showed higher values of both shear strength and shear force in all the green adhesive formulated. The results revealed the shear force values of 460.00 N, 50.00 N, 353.00 N, 365.00 N, 530.00 N,127.00 N and 440.00 N for samples A, B, C, D, E, F and G respectively. The shear strength values of sample A, B, C, D, E, F and G were 306.66 MPa, 33.33 MPa, 235.33 MPa, 243.33 MPa, 353.33 MPa 84.67 MPa, and 293.33 MPa respectively. It was observed from the investigation that the highest mechanical property was found in sample E with a shear force of 530.00 N and shear strength of 353.33 MPa. The lowest mechanical property was observed in sample B with a shear force of 50.00 N and shear strength of 33.33 MPa. The results of FTIR revealed the spectrum of the different adhesives samples in the form of various values. All the samples have peaks between 3316 cm^{-1} to 3361 cm^{-1} and at 1640 cm^{-1}. Three (03) functional groups were identified in all the samples. This indicated the existence of long chains in the compound. Thus, long chains are imperative in the formation of bonds by the adhesive. Based on the finding of the work, the suitability and workability of the products was investigated and proved to be effective.
Keywords: Adhesive, Bond strength, Shear force, Shear strength and Binders.

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I. Introduction

Adhesive materials have been used successfully in a variety of applications for centuries. Today adhesives are more important than ever in our daily lives and their usefulness is increasing rapidly. Materials are generally defined as adhesives by what they do (Robert, 2002). Almost any organic polymer and even many inorganic materials can function as adhesives in some situation. Whether they are organic or inorganic, all adhesives share common traits (bond) in performing their functions (Buba and Kabiru, 2015).

Agricultural waste management is part of the ecological cycle in which everything is cycled and recycled such that an independent relationship is maintained in the eco-system (Shehrawat and Nitu, 2012). Rice husk (RH) for example is one of the most widely available agricultural wastes in many rice producing countries around the world. Approximately 600 million tons of rice paddy is produced each year. RH is a great environmental threat causing damage to land and surrounding area where it is dumped (Ajay *et al.,* 2012). Only a little portion of the rice husk produced is utilized in a meaningful way, the remaining part is burnt into ashes or dumped as a solid waste with little being used in animal feed formulation. Its proper use will therefore eliminate waste disposal problem experienced by rice milling industries. It will provide an alternative use that will consequently improve the economic base of Nigeria among the committee of developing nations of the world (Alao *et al*., 2015). The most important relatively non-adhesive additive in an adhesive formulation is the filler (Ohoke and Igwebike-Ossi, 2015). Fillers are generally added in the adhesive formulation to provide some of these simultaneous benefits, increase thermal and electrical conductivity, reduce shrinkage and stress during cure, improve bond strength, improve flow properties, extend pot life and reduce cost (Licari and Dale, 2011).

Gum Arabic (GA) or *acacia gum* is the exudates from the Acacia senegal and *Acacia seyal* trees, belonging to *Leguminosae* family. It is a complex, branched heteropolysaccharide, either neutral or slightly acidic and composed of 1,3-linked β-D-galactopyranosyl units (Seema and Arun, 2015). Gum Arabic is certainly the most well-known of all gum types. European merchants who imported it from Arab ports such as Jedda and Alexandria (Eqbal and Aminah, 2013) coined the term ‘gum arabic’.

In view of the above, in the effort to formulate and convert waste to useful material, this research is expected to find an adhesive of high quality and strength which is environmentally friendly and will reduce the risk of health related diseases. The rice husk and the polystyrene wastes would be converted from waste to wealth. The present research is aimed at recycling of polystyrene and rice husk wastes into filler and binder in order to reduce environmental pollution and health hazard in the society.

II. Materials And Methods

Collection and Identification of Samples

Waste expanded polystyrenes (EPS) were collected around homes and refuse dumps whereas rice husk was collected at rice milling locations all within Bauchi metropolis. The polystyrene wastes, gum arabic and the rice husks were identified in the Departments of Chemistry and Crop Production Department of Abubakar Tafawa Balewa University, Bauchi. The Topbond glue was purchased from a vendor in Muda Lawal Market, Bauchi.

Preparation of Adhesive Filler from Agro-Waste Materials

The rice husk was first washed severally with clean water to remove stone, sand and dust contaminants. This was rinsed with distilled water to remove the metal ions that may be contained as impurities in the previous water. The washed rice husk was spread on a stainless tray and the broken pieces of rice grains present were removed by handpicking. The product was dried in a dust free environment. The dried rice husk was carbonize (d (100 ^{0}C) in a stainless steel pot.

The carbonized rice husk was weighed (765.92g) into crucibles and placed in a muffle furnace. The temperature of the muffle furnace was set at 650 ^oC and left for 4 hours. After 4 hours, white rice husk ash in a granular form was obtained. The rice husk ash was allowed to cool and reweighed again. The rice husk ash was ground in a ceramic mortar and pestle to obtain rice husk ash powder of smaller particle size. The powdered ash was sieved through a 63 μm mesh (Ohoke and Igwebike, 2015).

Preparation of Wood Adhesive

The method of Ohoke *et al.,* (2015) was modified for the compounding of wood adhesive. The following reagents were used for the formulation of wood adhesive: Filler (Rice husk ash, calcium carbonate and sodium hydroxide), plasticizer (glycerol), surfactant (Sweet potato juice), curing agent (benzoyl peroxide), antifoam (paraffin oil) and preservative (sodium benzoate), binder (polyvinyl acetate, gum arabic and plastic waste).

Table 1: Formulation of Wood Adhesive (Mixing Ratio of Components in Wood Adhesive Preparation)

Sample	Component	Wt (%)	Filler	Surfactant (ml)	Antifoam	Curing Agent	Plasticizer	Preservative
B	AG	70	20	2.0	2.0	2.0	2.0	2.0
C	EPW	70	20	2.0	2.0	2.0	2.0	2.0
D	EPW/AG	70	20	2.0	2.0	2.0	2.0	2.0
E	EPW/AG	70	20	2.0	2.0	2.0	2.0	2.0
F	PVA/AG	70	20	2.0	2.0	2.0	2.0	2.0
G	EPW/AG	70	20	2.0	2.0	2.0	2.0	2.0

NOTE: Filler D =RHA/ $CaCO_3$, Filler E = RHA, G = $CaCO_3$, EPW = Expanded Polystyrene Waste, RHA = Rice Husk Ash, AG = Arabic Gum and PVA = Polyvinyl Acetate.

Mechanical Properties of Adhesive Samples

The thickness/depth of the occupied gum (0.1 mm); the surface area for every sample (1.5 mm^2) and width of the occupied surface of the gum (15mm).Shear strength of compression molded sheet was measured in accordance with ASTM D638 on universal testing machine (Nr-261, Lloyds Instrument, UK). Crosshead speed for testing was maintained at 50 mm/min. Shore D hardness was determined in accordance with ASTM D2240 on shore ‘’D’’ hardness tester (International Equipments, Mumbai, India). Both the tests were performed at ambient conditions of 25^0 C and 75% relative humidity.

Determination of the Bond Strength of the Adhesive

The method of Ochigbo *et al*. (2010) was used with little modification prior to application of the adhesives. The surface of the substrate/adherends was pretreated by smoothening with sand paper followed by air-blasting to remove accompanying dust. After the surface treatment, the adherend/substrate pair was joined following the application of the film of the adhesive on the measured area. Two sets of test samples were made for each adherend/substrate set, one for drying in the air and the second for drying in an oven condition at a constant temperature (50 ^{0}C) for comparative purposes. The shear strength was determined from the minimum amount of the load that would result in failure at the adhesive joint and was calculated by the expression below;

$$\text{Shear Strength} = \frac{\text{Average Force (N)}}{\text{Surface Area (mm}^2\text{)}}$$

Fourier Transform Infrared Spectroscopy (FTIR)

The FTIR spectra were recorded using the range of 4,000cm^{-1} to 650 cm^{-1}.The FTIR of pure sample was run prior to the running of FTIR of the samples. The FTIR spectra were recorded with a PerkinElmer, Spectra GX equipment, USA. A 1-2 wt % solution of hot melt adhesive (HMA) was dissolved in chloroform and scanned with a resolution of 2 cm^{-1} in the scan range of 450-4000 cm^{-1} FTIR of the pure solvent was run prior to running the FTIR of the sample to enable the automatic subtraction of the baseline from the sample peak (Pravin *et al*.,2014).

III. Results

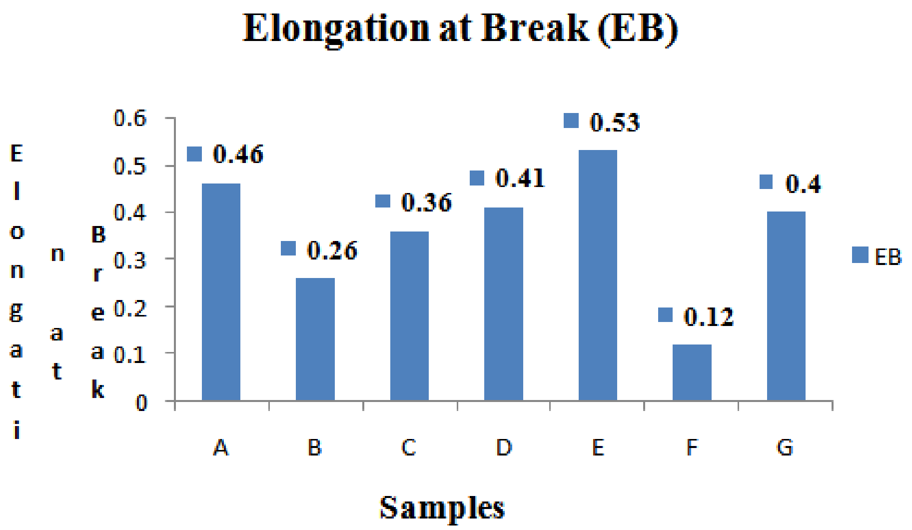

Figure 1: Elongation at break of Adhesive Samples

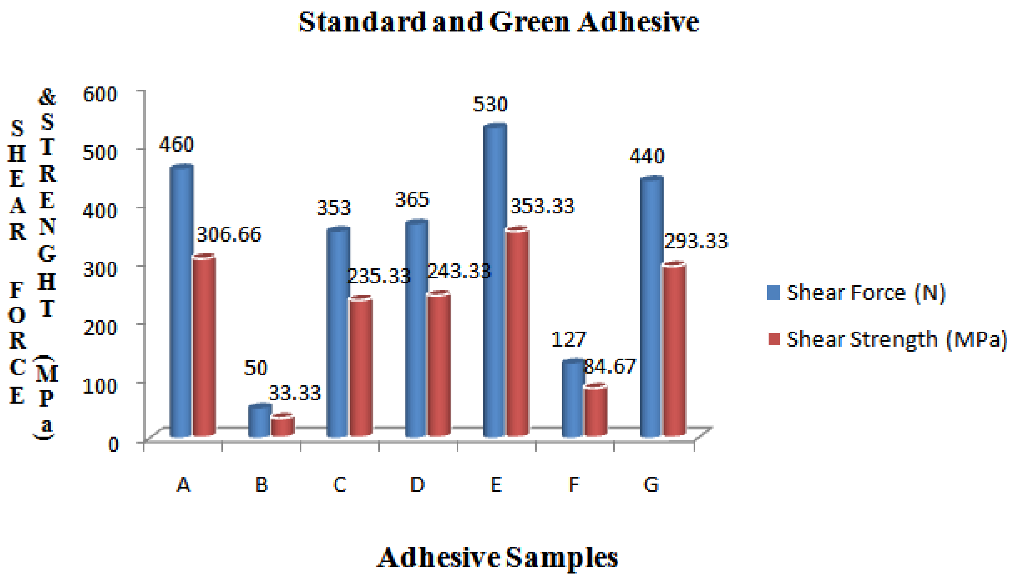

Figure 2: Shear Force and Shear Strength Values of Standard and Green Adhesive Formulated

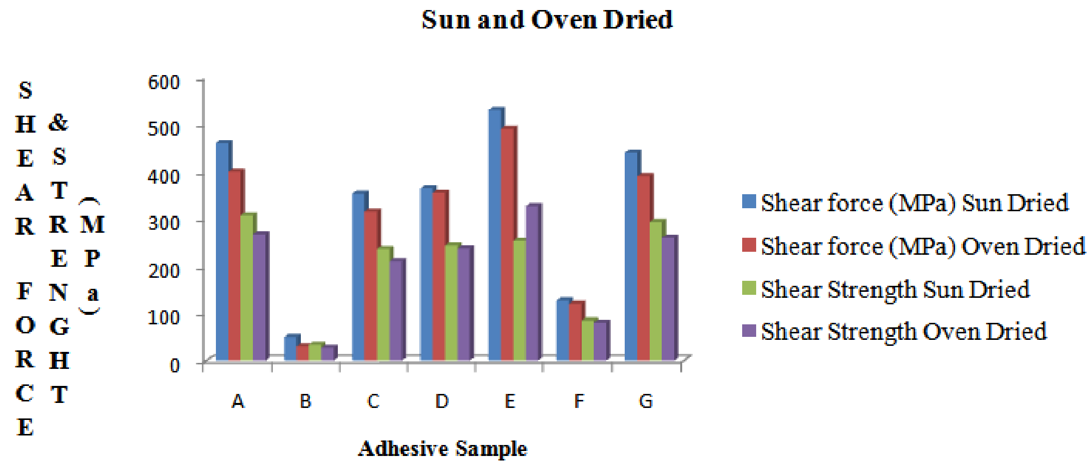

Figure 3: Shear Force and Shear Strength (Sun and Oven Dried)

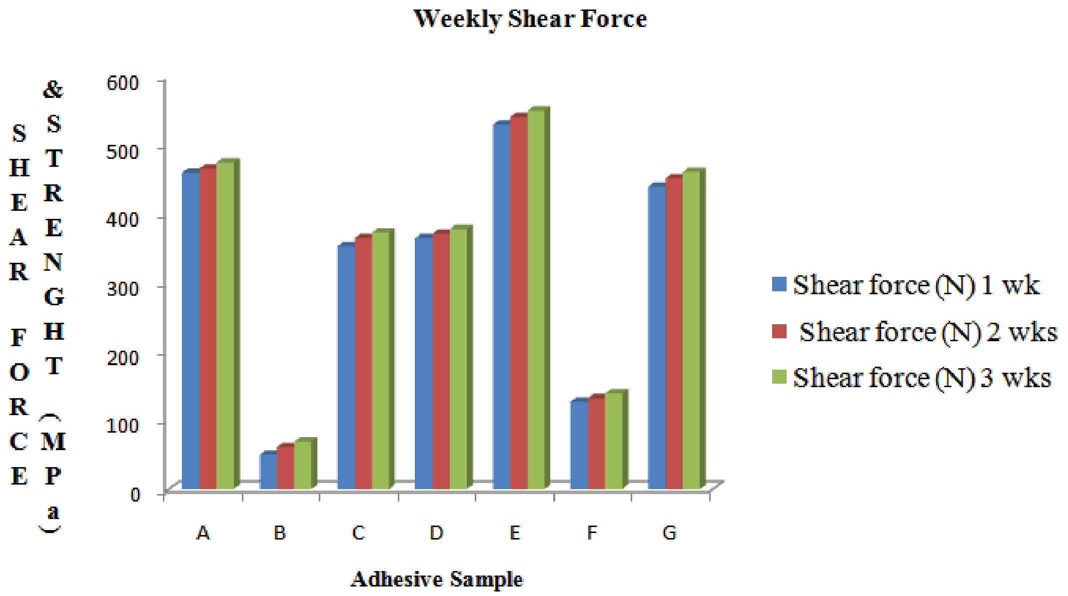

Figure 4: Weekly Shear Force of Adhesive Samples

Table 2: IR Absorption Band and Functional Group in the Adhesive Samples

Sample	IR Absorption Band (Cm^{-1})	Functional Group
A	3361	O-H Stretching band (Broad and Strong)
	2921	C – H Aliphatic Saturated
	2117	C≡C stretch
	1640	C=C Stretching of Aromatic ring
	1436	C – H of – CH_2 – bending
	1376	C – H of CH_3 – bending
	1238	C – O Stretching
	1026	Vibration – C – OH Stretch (Primary alcohol)
	948	C – H bend out of plane
B	3324	O-H Stretching band (Broad and Strong)
	2140	C≡C Stretch
	1640	C=C Stretching of Aromatic ring
	1419	Vinyl C-H bend (In the plane)
	1261	O-H in plane bend (1^0 and 2^0 –alcohol)
	1011	C-OH Stretch (Primary alcohol)
C	3316	O-H Stretching band (Broad and Strong)
	2124	C≡C Stretching
	1640	C=C Stretch of Aromatic ring
	1019	C-OH Stretch (Primary alcohol)
D	2106	C≡C Stretching
	1640	C=C Stretching of Aromatic ring
E	3335	O-H Stretching band (Broad and Strong)

	2113	C≡C Stretching vibration
	1640	C=C Stretch of Aromatic ring
	1417	C-H determination of $SP^3 – CH_3$
	1045	Vibration C-OH Stretch (Primary alcohol)
F	3339	O-H Stretching band (broad and strong)
	2110	C≡C Stretching
	1640	C=C Stretch of Aromatic ring
	1007	Vibration C-OH Stretch (Primary alcohol)
G	3335	O-H Stretching band (Broad and Stro)
	1640	C=C Stretch of Aromatic ring
	1030	Skeletal C-C vibration C-OH Stretch (Primary alcohol)

IV. Discussion

Preparation of Adhesive Filler

Rice husk ash (RHA) is a general term describing all types of ash produced from burnt rice husks (Deepa *et al*., 2013). The rice husk after pretreatment was subjected to100^0 C to get carbonized rice husk (CRH) and the value was 765.92 g. The carbonized rice husk was subjected to 650 ^{0}C for four hours and got a white rice husk ash (RHA) and the value reduced drastically to 224.35 g. The drastic reduction in weight and the white nature of RHA indicated the level of silica content in it and therefore good for adhesive formulation (Ajay *et al., 2012).*

Shear Force and Shear Strength Values of the Adhesives

The shear force and shear strength values observed in Table 6 were investigated with both standard and the green adhesives formulated. It was revealed that the shear force of all the samples were; A (460.00 N), B (50.00 N), C (353.00 N), D (365.00 N), E (530.00 N), F (127.00 N) and G (440.00 N) respectively. In another development, the shear strength of all the samples are; A (306.66 MPa), B (33.33MPa), C (235.33MPa), D (243.33MPa), E (353.33MPa), F (84.67MPa) and G (293.33 MPa) respectively. It is clear from the investigation that the highest shear strength was observed in sample E with a shear force (530N) and shear strength (353.33 MPa). The least values in shear force and shear strength were observed in sample B (50N and 33.33MPa) and sample F (127N and 84.67 MPa). The study was supported by Ohoke *et al.*,2015 and Lara *et al*.,2018.

Spectrum of Various Formulated Adhesive Samples using FTIR

The results of the FTIR were shown in Table 2 revealed the spectrum of different adhesive samples in the forms of various values. The curves were useful in predicting the functional group through various standard FTIR tables (Ghotkar, 2012). The seven samples are dominated in the spectra by the strong and broad O-H stretching vibrations at 3361 cm^{-3} to 3316 cm $^{-3}$.The C = C stretch of aromatic ring modes are observed at 1640 cm^{-3} in all the samples. These indicated the existence of long chains in the compound. Thus long chains are imperative in the formation of bonds by the adhesive. The similarities between the FTIR spectra in the samples are due to their common functional group (Alkyl group, alcohol and aromatic ring). These results are in accordance with those of Hindi *et al.*, (2017).The FTIR transmittance curves were obtained for both the standard and the formulated samples.

Bond Strength of Adhesive under Sun and Oven Dried Conditions

In Figure 3, the shear force and shear strength values were observed under sun and oven dried conditions. The adhesive samples labeled A, B, C, D, E, F and G with shear force and shear strength values under sun dried were found to be, 460 MPa and 306.66 MPa, 50 MPa and 33.33 MPa, 353 MPa and 235.33 MPa, 365 MPa and 243.33 MPa, 530 MPa and 353.33 MPa, 127 MPa and 84.67 MPa, 440 MPa and 260.00 MPa respectively. On the other hand, the samples under oven dried conditions has shear force and strength values as 400 MPa and 266.67 MPa, 30 MPa and 26.67 MPa, 315 MPa and 210.00 MPa, 355 MPa and 236.67 MPa, 490 MPa and 326.67 MPa, 120 MPa and 80.00 MPa, 390 MPa and 260.00 MPa respectively. It was observed that there is a significant reduction in terms of the values of shear force and strength under oven condition when compared with the samples under sun dried condition. The general lower values of bond strength attained against oven dried conditions might be due to the possibility that the oven conditions impact degradation effect on the polymer of the adhesive which lead to partial destruction of its mechanical and consequently, reduced bond strength as observed (Amine *et al*, 2010).

In Figure 4, the weekly shear force of all the adhesive samples under investigation shown that there is a significant increase in all the samples weekly. The longer the time after application, the wider the difference in bond strength as reported by Ohoke *et al.*, (2015) between acrylic/PVA/NR-$CACO_3$ and acrylic/PVA/NR-RHA.

V. Conclusion

The roles of rice husk ash as a filler, polystyrene waste and Gum Arabic as binders were established. They have proved to be vital ingredients in the formulation of green Adhesives. The physico-chemical property and the mechanical property revealed shear force of sample A,B,C,D,E,F and G to be 460 N, 50 N, 353 N, 365 N, 530 N,127 N and 440 N respectively. In another development, the shear strength was 306.66MPa, 33.33MPa, 235.33 MPa, 243.33 MPa, 353.33 MPa, 84.67 MPa and 293.33 MPa respectively. The FTIR peaks of all the seven (07) samples were between 3316 cm^{-3} to 3361 cm^{-3} (strong and broad vibrations) and 1640 cm^{-3} for the C = C stretching modes of aromatic ring. The similarities between the observed spectra are due to their common functional groups.

The adhesive binders and the fillers used in the research have demonstrated a significant usage in terms of the workability and applicability of the formulated Adhesive. The research revealed the bonding strength of the formulated adhesives that indicated a significant development and can compete with the standard adhesive produced and sells in the market. The research also supports the facts that henceforth, Arabic gum, polystyrene waste and rice husk ash should be used in the formulation of Adhesive so as to reduce environmental pollution and health effect.

The advantages of the formulated adhesives are their low cost, environmentally friendly, biocompatibility and relative widespread availability. Therefore, it was concluded that, the adhesives formulated will compete favourably interms of bond strength and workability of the product both domestically and in the industry

VI. Recommendations

The following recommendations were made based on the research findings.

i. The Adhesive Industry should consider the uses of Rice Husk Ash (filler), Arabic gum and polystyrene waste (binders) in the formulation of Adhesive.
ii. It is also recommended that further research on Adhesive sample for thermal, Scanning Electron Microscope (SEM) and rheological properties should be conducted.
iii. The Federal Government should emphasize and recognize the use of environmentally friendly materials in the Adhesive Industry.
iv. Federal Government and universities should commercialize Adhesive research findings to Adhesive Industries for suitability and applicability to the nation economic development.
v. Government should encourage and setup small-scale industries for the formulation of adhesive from agro and plastic waste materials that can test the feasibility and marketability of the product.

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