
Formulation and Performance Evaluation of Wood Adhesives Produced with Rice Husk Ash as New Filler

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To cite this article:

Ohoke Francis Okemini, Igwebike-Ossi Clementina Dilim. Formulation and Performance Evaluation of Wood Adhesives Produced with Rice Husk Ash as New Filler. *American Journal of Applied Chemistry*. Vol. 3, No. 2, 2015, pp. 33-39. doi: 10.11648/j.ajac.20150302.11

Abstract: This study investigated the applicability of rice husk ash (RHA) as filler in wood adhesives containing a blend of ethylene acrylic resin, poly vinyl acetate resin and natural rubber solution. Rice husk, obtained from Rice Mill Industries in Abakaliki, Nigeria, was washed, dried and heated to char (carbonized) on a gas stove until there was no emission of fumes. The rice husk char obtained was incinerated under controlled conditions in a muffle furnace at 650°C for four (4) hours. The RHA obtained was ground with ceramic mortar and pestle to reduce the particle size, sieved with a standard 63µm sieve and then used as filler in acrylics/PVA/NR wood adhesive. Filler levels in the adhesives were varied from 0 to 16%. The bond strength and thermal resistance of the prepared adhesives were determined and compared with that produced with CaCO₃ as well as a popular brand in the Nigerian market, Top Bond, used as reference standard. The result showed that after application, there was a general increase in bond strength with time for both CaCO₃ and RHA adhesives. The highest bond strength was obtained at a filler level of 12% for both fillers. At this level, RHA adhesive had higher bond strength of 170.3 KPa than CaCO₃ adhesive which had 167.8 KPa. RHA-filled adhesives were found to be more thermally stable than those of CaCO₃ and comparable to the reference standard.

Keywords: Filler, Rice Husk Ash, Wood Adhesive, Polyvinyl Acetate, Ethylene Acrylics, Natural Rubber

1. Introduction

An adhesive is any non-metallic material that is capable of joining bodies together by surface adhesion and internal strength without the structure of the bodies undergoing significant changes [1]. The technology of adhesives and their uses date back several millennia. Practical ways of uniting articles to provide a joint with resistance to an applied stress have been explored and developed to a useful level by entirely empirical methods. These are applied in several instances such as plywood manufacturing, securing cartons and packages, constructing of aircrafts, affixing postage stamps and manufacturing of carpets [2].

For optimum performance, an adhesive before cure should have good wettability, rheology, evaporative properties, flexibility characters and minimum shrinkage during cure [3]. The ability to wet a surface (wettability) is related to the ease with which a liquid spreads on a solid surface and is essential in maximizing coverage and minimizing voids in the bondlines [4]. Cured adhesives should have good bond strength and thermal stability. Properties like thermal and

electrical conductivity can be imparted to adhesives when necessary by the incorporation of conductive filler materials. Materials used as adhesives are polymers, which occur either naturally or are synthetic. Some examples in the natural category include cement (inorganic polymer based on silicates), glues, or pastes, waxes, natural resins, gums and asphaltic pitches as hot melt adhesives. Others in the synthetic category are silicone, epoxy, urethane and phenolic resins to mention but a few. These can also be grouped according to the adherends among which is wood adhesive.

The most important relatively non-adhesive additive in an adhesive formulation is the filler. Fillers are generally added in adhesive formulations to provide some of these simultaneous benefits [3]: increase thermal and electrical conductivity, increase thermal stability, reduce coefficient of thermal expansion, reduce shrinkage and stress during cure, improve bond strength, improve flow properties, extend pot life, reduce cost [3][5]. Substances often used as fillers include: silver flake, aluminium nitride, composite clay, zinc

Component	WA0C	WA4C	WA6C	WA8C	WA10C	WA12C	WA14C	WA16C
CaCO ₃	0.00	4.00	6.00	8.00	10.00	12.00	14.00	16.00
Curing agent	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Anti-oxidant	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Preservative	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
	100	100	100	100	100	100	100	100

Table 3. Formulation of acrylics/PVA/NR wood adhesive using varied wt% of RHA filler.

Component	WA0R	WA4R	WA6R	WA8R	WA10R	WA12R	WA14R	WA16R
Acrylics/PVA/NR	94.50	90.50	88.50	86.50	84.50	82.50	80.50	78.50
Surfactant	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Plasticizer	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Antifoam	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
RHA	0.00	4.00	6.00	8.00	10.00	12.00	14.00	16.00
Curing agent	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Anti-oxidant	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Preservative	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00

Key: WA4C = wood adhesive with 4% of CaCO₃; WA4R = wood adhesive with 4% of RHA

A total of sixteen (16) samples of wood adhesive were produced. The first eight (8) samples (WA-C) was formulated with acrylics/PVA/NR binder and CaCO₃ filler while the second eight samples (WA-R) were formulated with acrylics/PVA/NR binder and rice husk ash filler. All the samples were formulated with variations in the filler levels from 0 – 16wt% at the expense of the binder. The ratios of other components were kept constant. The quality parameters of each formulated sample were compared to that of Top Bond, a popular commercial brand of wood adhesive in the Nigerian market.

2.2.4. Tested Wood Adhesive Quality Parameters

(i). Bond Strength

A simple, effective improvised technique [26] was employed. A wood sample was cut to 200mmx45mm bonding surface. The adhesive was applied on only a section (60mmx45mm) of the wood surface leaving 140mmx45mm of the bonded wood free on both sides. After the curing process, one end of the bonded wood was firmly attached to the surface of a table by means of a clamp. On the other end of it, standard weights were gradually applied via a loop. The force was gradually increased until at a point when pull-off between the two surfaces was noticed. The value of the force (x) was regarded as the adhesive bond strength expressed in N/27000 mm² [26].

The value of x was converted to Y expressed in Kilopascals (1000N/m²) using the following formula:

$$Y = 37.04x$$

$$\text{Where } x = \frac{N}{27000\text{mm}^2} \text{ (measured)}$$

$$Y = 1000 \frac{N}{\text{m}^2} \text{ (Kilopascals)}$$

37.04 = constant

(ii). Thermal Stability Determination

The thermal stability of the adhesive was determined to find out the degree to which its bond strength can withstand temperature variations [3][27]. Two pairs of bonded wood samples were prepared. One pair was kept in an electric oven (DHG9033A) at the temperature of 98 - 100°C for 2 hours while the other pair was kept in a refrigerator (at 0 -2°C) for 2 hours. The bond strength of each pair was determined and recorded. The process was repeated for 3 hours and 4 hours inspection intervals. Thermal stability test was conducted on the 16 samples and also on Top Bond.

4. Results and Discussion

4.1. Results

4.1.1. pH of Acrylics/PVA/NR Wood Adhesives Filled with Either CaCO₃ or RHA

Table 4. pH of the formulated wood adhesives and TB.

Wt% of filler	Acrylics/PVA/NR adhesives	
	CaCO ₃	RHA
0	6.7	6.7
4	6.7	6.7
6	6.7	6.7
8	6.7	6.7
10	6.7	6.7
12	6.7	6.7
14	6.8	6.7
16	6.8	6.8
TB	6.6	6.6

4.1.2. Bond Strength of Wood Adhesives

Table 5. Bond strength (KPa) of CaCO₃-filled, RHA-filled acrylic/PVA/NR and TB wood adhesives.

% filler	2 hours		1 day		7 days	
	CaCO ₃	RHA	CaCO ₃	RHA	CaCO ₃	RHA
0	93.00±0.06	93.00±0.04	140.00±0.05	140.00±0.02	165.90±0.01	165.90 ±0.04
4	93.30±0.04	93.30±0.04	140.40±0.06	140.70±0.03	166.60±0.01	167.04±0.04
6	93.40±0.01	93.80±0.02	140.40±0.01	140.70±0.02	166.60±0.01	167.04±0.01
8	93.70±0.11	94.60±0.04	140.70±0.01	141.10±0.02	167.04±0.02	167.80±0.02
10	94.10±0.06	95.00±0.04	140.70±0.03	141.90±0.04	167.04 ±0.05	169.80±0.01
12	94.40±0.06	95.80±0.03	141.10±0.04	142.20±0.03	167.80±0.01	170.30±0.06
14	94.40±0.01	95.80±0.01	141.10±0.04	142.20±0.05	167.40 ±0.07	169.30 ±0.01
16	94.10±0.02	95.00±0.02	140.70±0.10	141.90±0.01	167.04±0.03	168.50 ±0.02
TB	95.80±0.02	95.80±0.06	142.60±0.11	142.60±0.01	169.60±0.03	169.60 ±0.05

4.1.3. Thermal Stability of Wood Adhesives

Table 6. Bond strength (KPa) of CaCO₃-filled and RHA-filled Acrylics/PVA/NR adhesives at room temp, 98 - 100°C and 0 - 2°C.

Wt% of filler	CaCO ₃ -filled Acrylics/PVA adhesives			RHA-filled Acrylics/PVA adhesives		
	98 – 100°C	Room temp	0 -2°C	98 – 100°C	Room temp	0 – 2°C
0	162.2±0.15	165.9±0.06	140.8±0.04	162.9±0.09	167.1±0.16	154.8±0.10
4	162.2±0.01	166.7±0.02	140.8±0.04	163.3 ±0.03	167.4±0.11	155.6±0.01
6	163.0±0.07	166.7±0.02	141.1±0.03	164.1±0.08	168.5±0.09	155.6±0.01
8	163.3±0.09	167.1±0.03	141.1±0.07	164.1 ±0.09	168.9±0.05	156.7 ±0.02
10	164.1±0.01	167.1±0.03	142.6±0.01	165.9 ±0.01	169.6 ±0.15	157.1 0.07
12	164.5±0.01	167.8±0.04	142.6±0.03	165.9±0.01	170.0±0.01	157.1±0.01
14	163.7±0.01	167.4±0.03	143.0 ±0.09	165.6± 0.01	169.6 ±0.01	157.1±0.06
16	163.7±0.04	167.1±0.02	142.6 ±0.02	165.6±0.06	169.3±0.10	1563±0.03
TB	165.9±0.05	169.6±0.06	161.4±0.01	165.9±0.14	169.6±0.02	161.4±0.03

Graphical illustrations

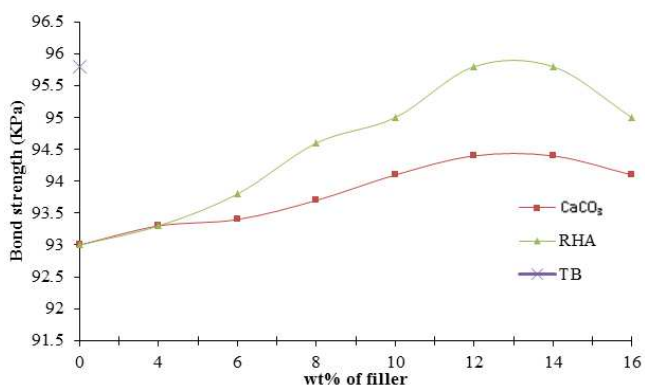


Fig. 1a. Plots of bond strength (KPa) versus wt% of filler for CaCO₃ and RHA filled.

Acrylics/PVA/NR adhesives (2hours after application).

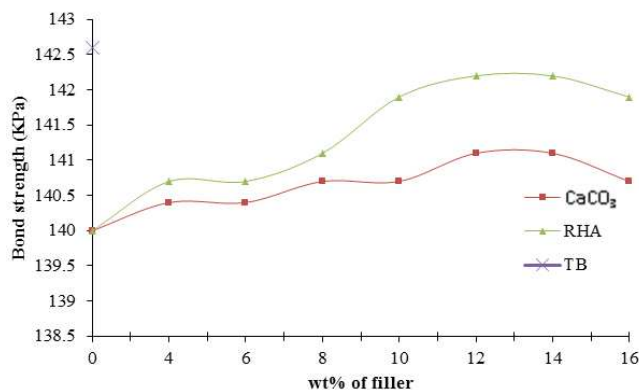


Fig. 1b. Plots of bond strength (KPa) versus wt% of filler for CaCO₃ and RHA filled.

Acrylics/PVA/NR adhesives (1 day after application)

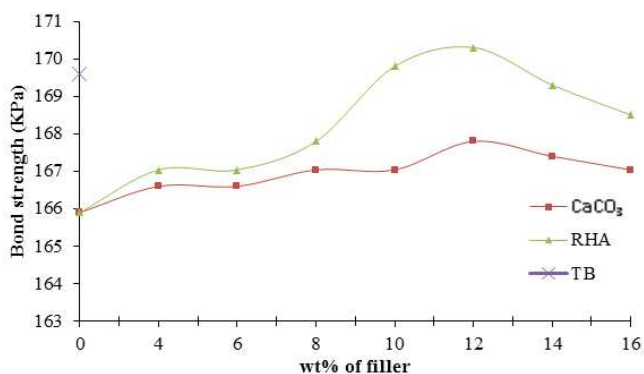


Fig. 1c. Plots of bond strength (KPa) versus wt% of filler for CaCO_3 and RHA filled.

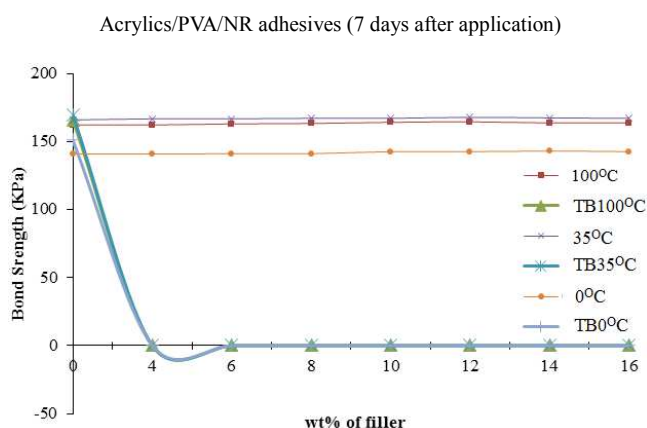


Fig. 2a. Plots of bond strength (KPa) versus wt% of filler for CaCO_3 filled Acrylics/PVA/NR.

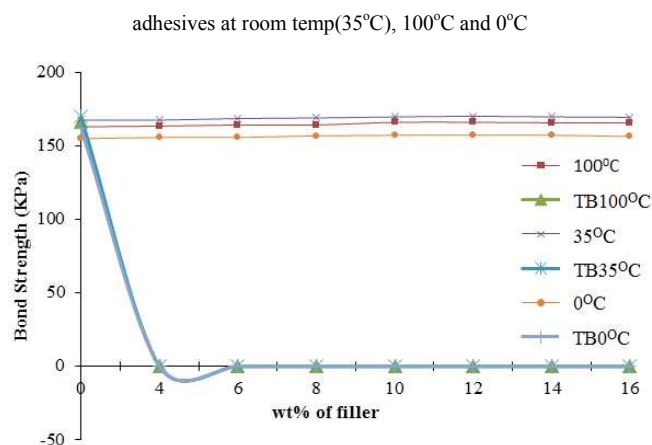


Fig. 2b. Plots of bond strength (KPa) versus wt% of filler for RHA filled Acrylics/PVA/NR.

adhesives at room temp(35°C), 100°C and 0°C

4.2. Discussion

4.2.1. pH

Table 4 shows the pH values of CaCO_3 or RHA filled acrylics/PVA/NR wood adhesives. The pH of pure acrylics/PVA/NR sample was 6.7 but the addition of 14%w/w filler, changed the pH of acrylics/PVA/NR- CaCO_3 sample to 6.8 while that of acrylics/PVA/NR-RHA changed

to 6.8 when the filler was increased to 16%w/w. In all, RHA filled samples were found to maintain more stable pH than the CaCO_3 filled samples. These findings were not unconnected with the pH difference of the filler materials.

4.2.2. Bond Strength of Wood Adhesive

It is evident from Table 5 and Fig. 1a, that after 2 hours of application, both acrylics/PVA/NR- CaCO_3 acrylics/PVA/NR-RHA have the same bond strength at 4wt% filler level. However, there was difference in bond strength in favour of acrylics/PVA/NR-RHA at filler levels of 6wt% and above. The same trend of bond strength difference was observed after 24 hours and 7 days of application. The reverse was the case after 24 hours and 7 days of application. It can also be observed that the longer the time after application, the wider the difference in bond strength between acrylics/PVA/NR- CaCO_3 and acrylics/PVA/NR-RHA. The set-to-touch time [28] of acrylics/PVA/NR-RHA was shorter than that of acrylics/PVA/NR- CaCO_3 . It may be expected that acrylics/PVA/NR-RHA should coalesce faster, as expressed by Ogban (2007), thereby limiting its flow into the wood pores. But the reverse was observed. This can be explained by the fact that the plasticizing effect of toluene (the NR solvent) and even the flexibilizing effect of rubber itself helped the adhesive to penetrate deep into the wood pores. Deeper penetration of the adhesive into the wood pores is responsible for stronger interlock between adherends. This was thought to be the reason behind the trend of higher bond strength of acrylics/PVA/NR-RHA over acrylics/PVA/NR- CaCO_3 .

Again, the pozzolanic (cementitious) property of RHA which gives synergy to the binding property of the binding agents was observed to increase with time. This was responsible for the observed reversal of increase in bond strength from acrylics/PVA/NR- CaCO_3 to acrylics/PVA/NR-RHA with RHA showing superior performance to CaCO_3 . Compared to CaCO_3 filler, RHA has lower specific gravity and higher surface area/volume ratio. The greater number of particles per unit volume of RHA in adhesive reduces void spaces in the adhesive thereby increasing the number of bonds per unit volume which was reflected in increase in bond strength.

4.2.3. Thermal Stability

The ability of an adhesive to maintain high bond strength even across wide variations of temperature is important. This is because the bonded wood, during use, will definitely be exposed to fluctuation in environmental conditions which may affect the coefficient of thermal expansion of the adherends as well as the adhesive [19]. Table 6 and Fig. 2a and Fig. 2b, show the response of bond strength of the prepared adhesive samples to temperature when the bonded pair was subjected to 100°C and 0°C temperature for 4 hours. The thermal stability of the adhesives was determined by evaluating the difference in bond strength at room temperature and at lower and higher temperatures.

In comparison, both acrylics/PVA/NR- CaCO_3 and acrylics/PVA/NR-RHA adhesives were found to maintain

more stable bond strength at higher temperature than at lower temperature (Table 6, Fig 2a and Fig 2b). However, on both sides of the temperature difference, acrylics/PVA/NR-RHA adhesive recorded lower difference in bond strength. This established the fact that acrylics/PVA/NR-RHA has higher thermal stability than acrylics/PVA/NR-CaCO₃.

4.3. Conclusion

In conclusion, the applicability of RHA as filler in adhesives has been established. RHA has lower specific gravity than CaCO₃ hence there will be a greater number of RHA particles dispersed in the adhesive which results in higher viscosity of RHA-filled adhesives. The pozzolanic (cementitious) property of RHA has synergistic effect on the binding property of the binding agents thus acrylics/PVA/NR-RHA adhesive has higher bond strength and thermal stability than the CaCO₃-filled samples. Comparison of the physical properties of both RHA and CaCO₃-filled adhesives with Top Bond reveals that RHA-filled adhesives have values more comparable to Top Bond than the CaCO₃-filled samples. Above all, maximum contribution to bond strength was achieved by incorporating between 8wt% and 12wt% RHA. Beyond 12wt% filler level, the bond strength declines for both RHA and CaCO₃.

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