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

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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 $CaCO_3$ 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 $CaCO_3$ 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 $CaCO_3$ adhesive which had 167.8 KPa. RHA-filled adhesives were found to be more thermally stable than those of $CaCO_3$ 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 oxide, carbon black, hydrated alumina, calcium carbonate, fused silica, silicates and boron nitride. Wood fibres, oil palm fibres, pineapple leaf fibres, banana leaf fibres and a host of other plant derived fibres have been reported as adhesive fillers [6 - 9].

Researchers, in their effort to convert wastes to useful materials, have in the recent years been investigating the use of agro wastes (plant and animal by-products) as filler materials in both thermoplastic and adhesive composites. Several reports have been made on the improvement of the rheology, the adhesion and mechanical properties of adhesives via the incorporation of agro wastes as filler materials [10 - 14]. Rice husk is generated in large quantities in the rice producing areas of the world during the process of rice milling. The domestic and industrial demand for rice husk is extremely low, thus the large volumes of unutilized rice husk in these areas constitute an environmental nuisance [15]. Therefore the use of rice husk ash for industrial application will go a long way in minimizing the accumulation of rice husk in the environment.

Filler applications of rice husk ash (RHA) in plastics, elastomers and other polymer composites have been reported. RHA has been reported as excellent filler in natural rubber and epoxidized natural rubber [16 - 18]. Incorporation of RHA in polypropylene composites has been reported to improve the tensile modulus and flexural modulus of polypropylene/ethylene-propylene-diene terpolymer [5], increase the degree of crystallinity of polypropylene by small margin [19], improve the adhesion and phase continuity in the polypropylene composites[20] and enhances thermal stability of epoxy foams[21]. RHA has been reported as adsorbent in water treatment and vegetable oil treatment [22], as an additive to high performance building blocks, concrete and mortar [23], as extender in textured paint [15] and red oxide primer [24] and as a good flatting extender in cellulose matt paint [25].

In this work, the effects of RHA on the bond strength of acrylics/poly(vinyl acetate)/natural rubber wood adhesives were investigated and compared with adhesives formulated using $CaCO_3$ as filler.

2. Experimentals

2.1. Materials

i. Binder: Polyvinyl acetate (PVA) and ethylene acrylic emulsions were purchased from Joytex Paint and Industrial Chemicals Limited, Enugu; rubber latex was purchased from Rubber Research Institute, Nekede, Imo State, Nigeria.

ii. Rubber Solution: Rubber latex was allowed to coagulate into a white mass. This white mass was shredded and left under the sun for 24 hours to dry. 4g of rubber was then dissolved in 100ml of toluene to form rubber solution.

iii. Commercial Filler: The commercial filler, $CaCO_3$, used in this study was obtained from Glisco Laboratory Chemicals Limited, Enugu, Nigeria.

2.2. Methods

2.2.1. Preparation of Rice Husk Ash Filler

Rice husk was obtained from a rice mill in Abakaliki, Ebonyi State, Nigeria. The rice husk was first washed severally with clean water to remove stone, sand and dust contaminants. It was again 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 broken rice grains present were removed by handpicking. It was then left to dry in a dust free environment.

The dried rice husk was burnt to char using a stainless steel pot. This stage of burning is called carbonization. Carbonized rice husk was weighed in crucibles and fed into a muffle furnace using the same crucibles; the temperature of the furnace was set at 650^{o}C and left for 4 hours. After 4 hours, white rice husk ash in a granular form was obtained. The rice husk ash-containing crucibles were removed from the furnace, allowed to cool and weighed again. The rice husk ash was ground in a ceramic mortar and pestle to obtain rice husk ash powder of smaller particle size. The ground ash was sieved with standard sieve of 63µm mesh.

2.2.2. Compounding of Wood Adhesive

The following reagents were used for the preparation of wood adhesive: Binder (polyvinyl acetate, acrylics and rubber solution), Filler (Calcium carbonate and Rice husk ash), Curing agent (sulphur), Plasticizer (glycerol), Surfactant (sodium laurylsulphate), Antifoam (paraffin oil) and Preservative (sodium benzoate).

2.2.3. Mixing Ratio of Components in Wood Adhesive Formulations

Table 1. *Typical formulation used for the production of wood adhesives.*

Ingredient	Wt%
Acrylics/PVA/NR (8:1:1)	94.50
Surfactant	1.00
Plasticizer	1.00
Antifoam	1.00
Filler	0.00 - 16.00
Curing agent	1.00
Anti-oxidant	1.00
Preservative	0.50
	100.00

Table 2. *Formulation of acrylics/PVA/NR wood adhesive using varied wt% of $CaCO_3$ filler.*

Component	WA0C	WA4C	WA6C	WA8C	WA10C	WA12C	WA14C	WA16C
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

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 $CaCO_3$; 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 $CaCO_3$ 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 $N/27000\ mm^2$ [26].

The value of x was converted to Y expressed in Kilopaschals ($1000N/m^2$) using the following formula:

$$Y = 37.04x$$

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

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

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^{o}C for 2 hours while the other pair was kept in a refrigerator (at 0 -2^{o}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 $CaCO_3$ or RHA

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

	Acrylics/PVA/NR adhesives	
Wt% of filler	$CaCO_3$	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 $CaCO_3$-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 $CaCO_3$-filled and RHA-filled Acrylics/PVA/NR adhesives at room temp, 98 - 100^{o}C and 0 - 2^{o}C.*

Wt% of filler	$CaCO_3$-filled Acrylics/PVA adhesives			RHA-filled Acrylics/PVA adhesives		
	98 – 100^{o}C	Room temp	0 -2^{o}C	98 – 100^{o}C	Room temp	0 – 2^{o}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 illusrtations

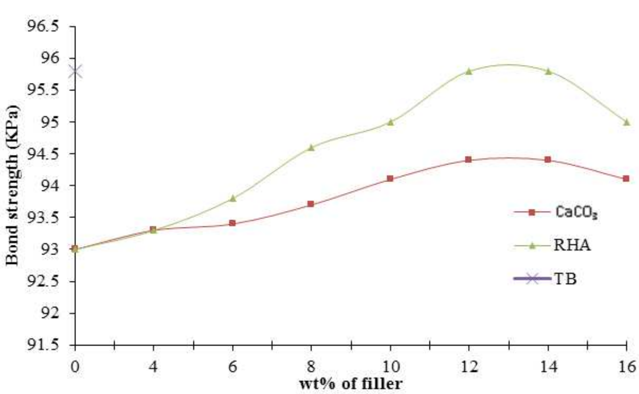

Fig. 1a. *Plots of bond strength (KPa) versus wt% of filler for $CaCO_3$ and RHA filled.*

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

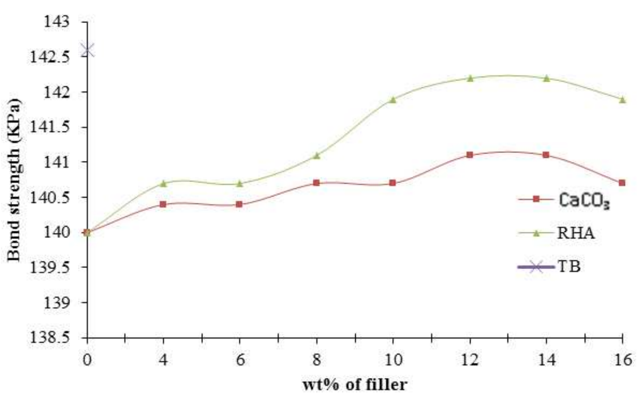

Fig. 1b. *Plots of bond strength (KPa) versus wt% of filler for $CaCO_3$ and RHA filled.*

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

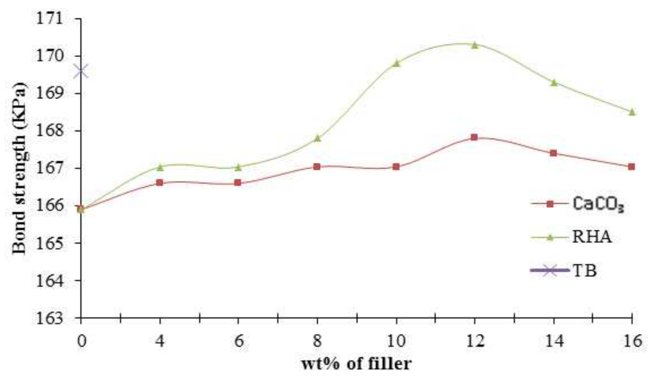

Fig. 1c. Plots of bond strength (KPa) versus wt% of filler for $CaCO_3$ and RHA filled.

Acrylics/PVA/NR adhesives (7 days after application)

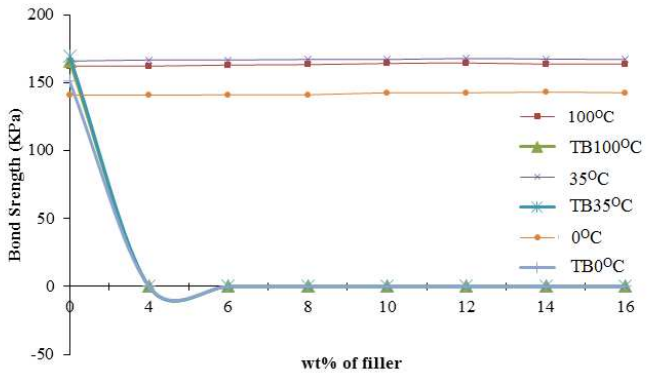

Fig. 2a. Plots of bond strength (KPa) versus wt% of filler for $CaCO_3$ filled Acrylics/PVA/NR.

adhesives at room temp(35ºC), 100ºC and 0ºC

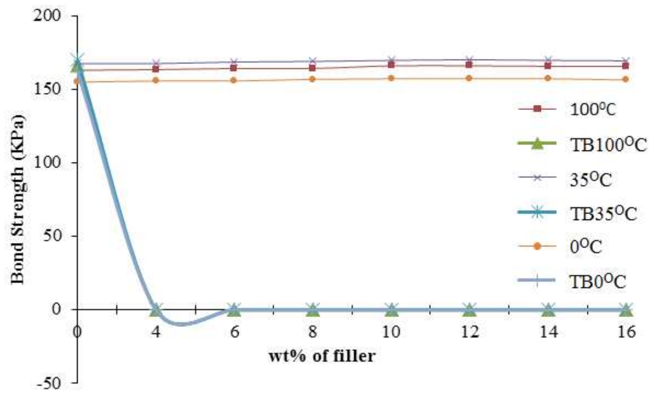

Fig. 2b. Plots of bond strength (KPa) versus wt% of filler for RHA filled Acrylics/PVA/NR.

adhesives at room temp(35ºC), 100ºC and 0ºC

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 to16%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^{o}c and 0^{o}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-$CaCO_3$.

4.3. Conclusion

In conclusion, the applicability of RHA as filler in adhesives has been established. RHA has lower specific gravity than $CaCO_3$ 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 $CaCO_3$-filled samples. Comparison of the physical properties of both RHA and $CaCO_3$-filled adhesives with Top Bond reveals that RHA-filled adhesives have values more comparable to Top Bond than the $CaCO_3$-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 $CaCO_3$.

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