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Ecological adhesive based on cassava starch: a sustainable alternative to replace urea-formaldehyde (UF) in particleboard manufacture

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ABSTRACT

This study assessed the properties of particleboard produced from the replacement of urea formaldehyde with cassava starch. Cassava starch (CS) and urea-formaldehyde (UF) were mixed at percentages of 100/0, 90/10, 70/30, 50/50, and 0/100, respectively. The mixture was added to the wood particles at 15% adhesive content based on the weight of oven-dried particles. Particleboards of sizes 20 mm × 300 mm × 300 mm and a targeted density of 600 kg m^{-3} were manufactured by hot-pressing at 170 °C, compacting pressure 3.5 MPa for 8 min. Physical and mechanical properties (density, thickness swelling, water absorption, static bending, and hardness) were tested. The physical and mechanical properties also increased in the highest proportions of UF. Comparatively, manufactured with 50% CS + 50% UF, it performed better in all the evaluated properties. Thus, it could adopt the latter mixed ratio in the industry to reduce the quantity and cost of UF and its environmental emissions. The results obtained were adequate and in conformity with international standards. Hence, it could mix cassava starch and urea formaldehyde adhesives in different ratios to get desirable properties. It is possible to conclude that cassava starch could be considered a sustainable alternative adhesive.

Keywords: Sustainability; Composites; Bioadhesive; Physical and Mechanical Properties.

1. INTRODUCTION

Commercial particleboard is a composite panel product of wood particles such as sawdust, wood chips, sawmill shavings, or other agricultural wastes bound together with a synthetic adhesive. Adhesive accounts for up to 32% of manufacturing costs in the glue-wood composite industry. Synthetic adhesives based on petroleum have been used for a long time and are known to have excellent performance, good working properties, and economic satisfaction [1, 2].

Urea-formaldehyde (UF) is the dominant resin in adhesive production for applications in different wood industry sectors [3]. These adhesives are used mainly due to their excellent performance in panels, low cost, high reactivity, and fast curing. However, due to their low resistance to water and other weather conditions, their application is limited to the manufacture of components for indoor environments [4, 5]. Melamine-urea-formaldehyde (MUF) resin is another material widely used to manufacture panels. UF resin has a shorter manufacturing time, lower pressure rates, low temperature, low labor cost, and lower electricity in panel production [6]. Despite the biological effects presented by these synthetic agents, negative factors need to be considered when using these materials. According to ZHOU *et al.* [4], components manufactured using UF adhesives tend to release formaldehyde during their lifetime, which is highly harmful to human health and the environment. MAZAHERI *et al.* [7] expose that the amount of formaldehyde present in indoor air is directly related to factors such as the sources of formaldehyde in the building, ventilation, temperature, and food.

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The growing concern for human health and the environment has motivated the development of environmentally friendly products, including manufacturing natural labels for wood products, vegetal fibers, or agro-industrial products [8–10]. These environmental impacts could be lessened by developing environmentally friendly materials manufactured from renewable sources [11–14].

Adhesives for making wood products can be classified and identified according to their renewable raw material. They can be classified into tannin, wood, vegetable, lignin, soy, starch, and bioadhesives [15]. Among these natural materials, starch has been widely studied as a constituent for producing natural adhesives and biocomposites. Characterizing the properties of biocomposites reinforced with cellulose nanocrystals extracted from rice husk with cassava starch matrix, KARGARZADEH *et al*. [16] found an increase in tensile properties, storage modulus, thermal stability, and a decrease in water absorption.

In their research with biocomposites reinforced with cellulose extracted from *Posidonia oceanica* biomass and corn starch matrix, BENITO-GONZÁLEZ *et al.* [17] found improvements in mechanical properties and water barrier. REINALDO *et al.* [18] evaluated biocomposites with grape skin, acerola waste, and cassava starch matrix. The authors found that adding acerola residue reinforcement with cassava starch promoted improvements in the antioxidant properties of the biomaterial, thus making it an exciting alternative for packaging production. JOVANOVIĆ *et al.* [19] improved hydrolytic and thermal stability in the biocomposites with cellulosic reinforcement and starch-modified urea-formaldehyde matrix.

The cassava crop for starch production is ranked the fifth most produced globally and the third among tropical regions' most consumed food sources. After harvesting, the cost of cassava may increase due to processing to make starch, contributing positively to the growth of rural economies [20, 21]. Cassava starch is amorphous and has good mechanical properties [1, 2, 22]. Therefore, it could achieve superior properties by blending urea-formaldehyde with it. Starch is a natural material with thermoplastic properties when subjected to plasticizers, high temperatures, and shear. Because this raw material has thermoplastic properties that are very close to those of synthetic polymers, it is possible to use the exact processing mechanisms in starch [23].

Cassava starch is a natural and abundant material. It is low-cost and has good gelatinization, rheological, and solubilization properties. It is used in several applications and can be modified to vary its chemical, physical, and enzymatic properties [20–22]. Therefore, blending it with UF would reduce the environmental challenges associated with the use of only UF and other synthetic adhesives. This study seeks to assess the characteristics of particleboard produced with cassava starch and urea-formaldehyde adhesives.

2. MATERIALS AND METHODS

2.1. Raw material

Cassava starch and urea-formaldehyde matrices and sawdust reinforcement from *Ceiba pentandra* were used to carry out the study. Fresh cassava tubers were obtained, washed, peeled, and milled to obtain cassava dough. The solid mass produced with cassava starch was diluted with water at a temperature of 26 ± 2 °C to form a solution. After that, the solution was strained with 1 mm wire mesh and stood for 24 hours to allow the starch to settle. The water was decanted to obtain the cassava starch. The starch was air-dried for ten days and then ground to get the powdered starch.

The properties of the cassava starch, moisture content, ash content, and solid content were determined using standard laboratory procedures. The moisture content was determined using ASTM D 1037 [24], the ash content was determined using ASTM D 1102 [25], and the solid content was determined using the processes described by UMEMURA *et al.* [26].

For making the particleboard, urea-formaldehyde (UF) adhesive with a ratio of 1:1, 65% solid content, a specific gravity of 1.266 g.cm^{-3} at 30 °C, a viscosity of 230 mPa.s at 30 °C, pH of 7.5, and a gel time of 65 seconds at 100 °C was combined with cassava starch. *Ceiba pentandra* sawdust was obtained from a timber processing company in Ghana.

2.2. Preparation of the adhesive

A blending of the adhesives, cassava starch (CS), and urea-formaldehyde (UF) were mixed at a percentage of 100:0 90:10, 70:30, 50:50, and 0:100 (CS: UF). 2% ammonium chloride (NH4Cl) was added to the resultant blend as a curing catalyst (hardener). The pH of the blended adhesives was determined by the method described by NASIR *et al.* [27], which determines the acidity or alkalinity of biomass materials and their compatibility with adhesives. 50 g of each biomass material and the blended adhesives were soaked in 200 ml distilled water in a 1 L beaker. Five replicates of each solution were boiled for 10 minutes, cooled, and decanted before the pH was measured using a pH meter (Hanna Instrument HI 4522).

2.3. Particleboard manufacture

The *Ceiba pentandra* particle size ranged from 0.5 mm to 1.5 mm, and each dried to a moisture content of 4% and was thoroughly mixed with the blended adhesives. 2% ammonium chloride was added as a hardener. The particles were mixed with the resin in an aluminum container. Subsequently, they were cold-pressed to form a blanket with a layer of 80 mm. The process was carried out using an aluminum mold measuring 300 mm × 300 mm.

Five replicates for each combination were produced. To carry out the hot-pressing process, a 20 mm thick metal stop was used to ensure that the plates produced had the same thickness. The mat was then pressed with the following conditions: temperature 170 °C; pressure 3.5 MPa; time 8 minutes; closing rate 3-4 mm min^{-1}; target thickness 20 mm; and compacting time 15 minutes. The pressure was defined empirically to guarantee the dimensions established for the manufactured panels. The produced particleboards were then trimmed and conditioned for six days in a climate-controlled room having a temperature of 20 °C ± 2 °C and a relative humidity of 62% ± 2% before they were sawn into various sizes for further studies, five replicates from each treatment.

2.4. SEM analysis

The surfaces of particleboard specimens of sizes 5 mm × 10 mm × 10 mm were evaluated using a scanning electron microscope (SEM). The specimens were sputter-coated with a thin film of gold, mounted on an aluminum stub using carbon tape, and then analyzed with a Phenom ProX desktop SEM with EID at an accelerating current of 15 kV and a magnification range of 1300× to 1500×.

2.5. Physical properties

The apparent density of the biomass panels was determined using the methodology established by WIDYORINI *et al.* [28]. This was done by filling a 50 L cylindrical container to the brim and weighed. The measurement was repeated five (5) times for each biomass material.

The water absorption (WA) and thickness swelling (TS) tests were conducted according to ASTM D1037 [24]. The specimens' initial weights were recorded, after which they were soaked in distilled water for 2 and 24 hours. Thickness swelling was calculated from the difference in a specimen's thickness before and after immersion in distilled water for 2 and 24 hours. The swelling was measured using an electronic digital caliper with a precision of 0.01 mm.

2.6. Mechanical properties

The static bending modulus of elasticity and modulus of rupture of the particleboards were determined by ASTM D 1037 [24]. Specimens 20 mm × 50 mm × 250 mm were prepared from the particleboards produced. A Universal Testing Machine (model Inspekt 50-1; Am Gründchen 1, 01683 Nossen, Germany) operated with a load cell capacity of 50 kN was used for the test. The loading rate applied was 4 mm/min.

The hardness of the particleboards was determined under ASTM D 1037 [24]. The particleboards were laminated to obtain the given thickness and subsequently cut into dimensions 25 mm × 75 mm × 150 mm, as specified by the standard. Janka ball test was used to determine the hardness using the "Instron" universal testing machine (Instron Model 4482, Norwood, MA, USA) with the hardness test fixture.

The steel ball with a diameter of 11.28 mm was driven into the specimen, and the load necessary to force it into the sample to a depth of 5.6 mm by the steel ball was recorded automatically by the "Instron" machine as the failure load.

2.7. Statistical analyses

A one-way analysis of variance (ANOVA) was used to determine the difference in the properties evaluated of the particleboards. The adhesives in different proportions (Pure CS, 90% CS + 10% UF, 70% CS + 30% UF, 50% CS + 50% UF, and Pure UF) levels, respectively, were compared by Tukey's multiple tests at $p \leq 0.05$.

3. RESULTS AND DISCUSSION

3.1. Adhesive properties

The results showed that cassava starch (CS) presented values of 76.72 ± 2.63 (%) for solid content, 0.34 ± 0.07 (%) for ash content, 10.13 ± 0.14 (%) for moisture content, and a pH of 7.54 ± 0.14. The urea-formaldehyde (UF) adhesive showed values of 51.93 ± 6.51 (%) for solids content, 2.73 ± 0.81 (%) for ash content, and a pH of 8.30 ± 0.51. In this study, a few characteristics of cassava starch were verified for purity for manufacturing particleboards. The percentage of ash content determined the purity of starch. The lower the ash content, the purer the cassava starch. The results show that the cassava starch with ash content (0.30%) was purer than starch obtained from oil palm in other studies [29].

The maximum moisture content of starch for commercial purposes is 13% [29–31]. In this study, the moisture content for CS was 10.13%. Hence, it is within the range of commercial starch. Moisture and the solid content of adhesives are of the most significant importance to the viscosity and rheological properties of the adhesives during hot pressing [29, 32]. The adhesives were all alkaline (basic) on the pH scale. The characteristics were compatible with the alkalinity of the biomass materials and their affinity to adhesives for the study; thus, the CS is expected to form a good mix with UF in bonding with the biomass materials.

3.2. SEM analysis

Bonding between the adhesive mix ratios and the biomass particles was characterized with SEM graphs of the cross-sections of the particleboard specimens. The analysis of the surfaces of the manufactured particleboards is shown in Figure 1. The micrographs indicate adequate bonding in the adhesive mix of the various percentages, and particles were well-embedded in the varied mixed adhesive ratios. It was apparent that the adhesives dispersed evenly into the spaces, and the biomass was bonded more tightly. Therefore, the strength and water resistance of the particleboards were enhanced. Similar results were obtained by YE *et al.* [32], which could contribute to the performance of the physical and mechanical test results.

3.3. Density

The density of particleboards reduces with the corresponding reduction in the percentage of CS in the mix (Figure 2). Cassava starch, with excellent viscosity and solid content [30], can fix itself in the matrix with a higher aspect ratio and bond the particles during curing processing. However, with the lower viscosity and solid content, UF causes the mix to flow rather than bond with the particles. The inability of UF with lower solid content and viscosity to fill the interparticle voids is caused by the biomass's lower aspect ratio (large surface area). As a result, there is poor interfacial bonding between the particles and the UF adhesive.

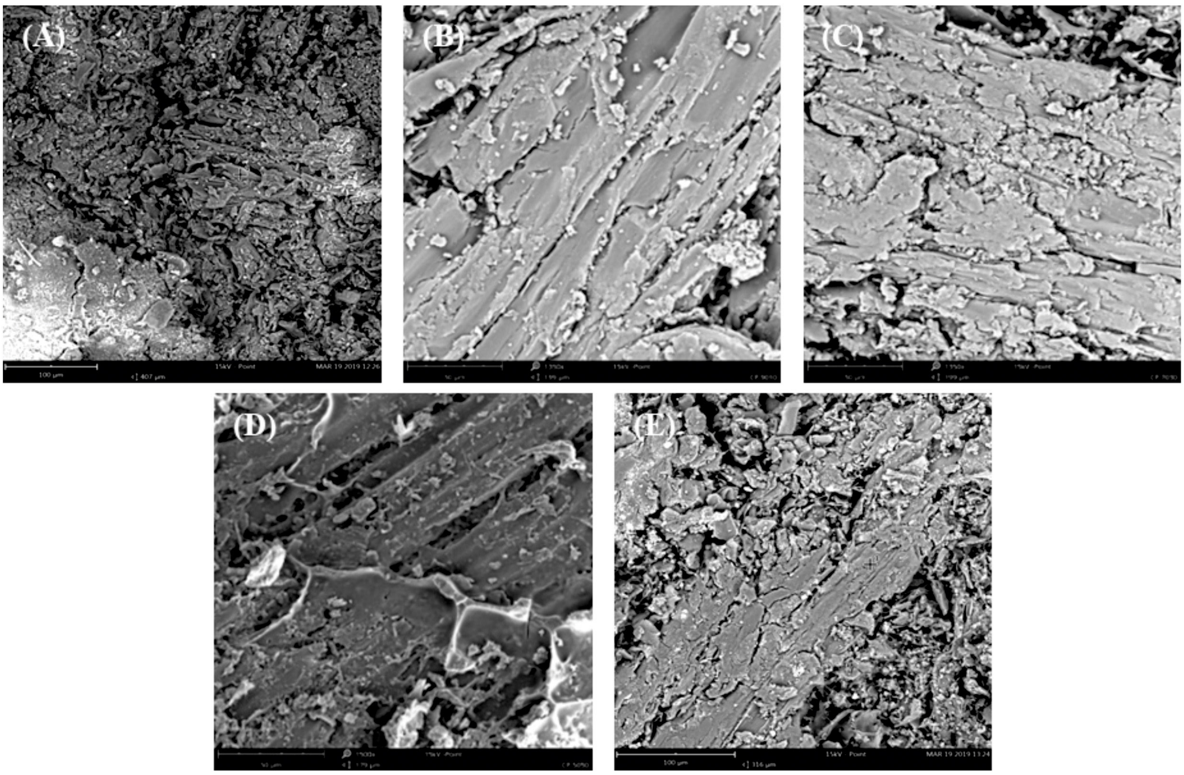

Figure 1: Scanning electron micrographs with a magnification range of 1300× to 1500× of the manufactured particleboards using varied adhesive mixed ratios. The figures show smooth surfaces between particles and adhesives (A, B, C, D, and E, representing the manufacture of chipboard with 100%, 90%, 70%, 50%, and 0% cassava starch, respectively).

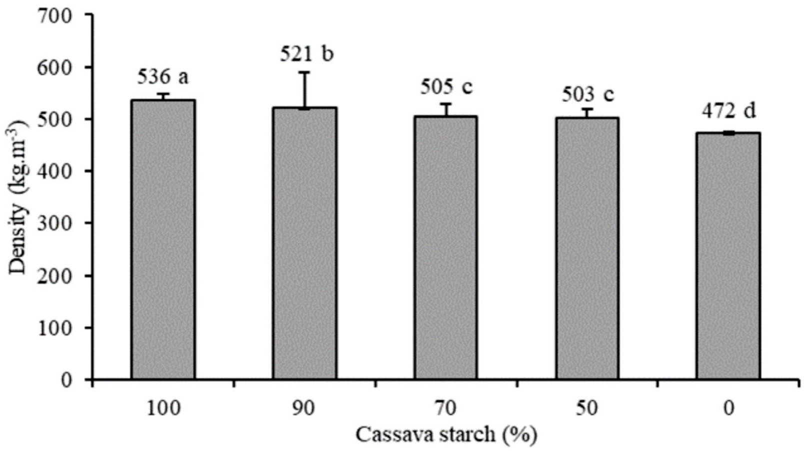

Figure 2: The density of the manufactured particleboards was produced using varied adhesive combinations with cassava starch and urea-formaldehyde.

The moisture content of the biomass materials ranged from 3.61% to 3.67%, similar to that used by HUANG and SUN [33]. Such moisture content improves adhesive spread, curing, and proper circulation of heat in the mat during pressing and consolidates particles and adhesives to ensure the plasticity of the composite board [34, 35].

3.4. Water absorption (WA)

The results for water absorption in periods of 2 and 24 hours of immersion showed that the increase in the addition of urea-formaldehyde (UF) resin to the biomass reduced the water absorption property of the material (Figure 3A). QIN *et al.* [36] studied the interaction of melamine-urea-formaldehyde (MUF) resin with poplar and eucalyptus wood. They found a reduction in water absorption that may be due to the adhesive penetrating the cell wall and filling the empty spaces in the wood, thus occupying the cavities responsible for absorbing moisture. According to ZHANG *et al.* [37], the reduction in water and humidity absorption due to the impregnation of the adhesive in the wood is not only related to the filling of the fibers, lumen, vessels, and empty spaces of the cell wall but also to exchange hydrophilic groups present in amorphous regions and the outer surface of crystalline regions.

The mixed adhesive ratio used in manufacturing the particleboards for the study produced similar or better WA means than other researchers who used adhesives UF, MF, or PRF. In a study to evaluate the quality of particleboard manufactured with wood from Sequoia sempervirens and *Pinus taeda* using UF as an adhesive, DIMITRIOU *et al.* [38] indicated that 24-hour water absorption of the particleboards manufactured ranged from 26.3% to 55.4%. Also, LIU *et al.* [5] recorded 30% to above 80% and 60% to above 90% for 2 hours and 24 hours WA, respectively, from assessing the properties of Sago particleboards resinated with UF and PF resins. This shows that the adhesive ratio mix is applicable.

The analyses verified that adding 10, 30, and 50% of urea-formaldehyde (UF) resin in the mixture reduced the water absorption property compared to the treatment composed exclusively of cassava starch (CS). In reducing water absorption on the phenomenon in particleboard, the adhesive promotes a reduction in porosity and a decrease in empty spaces in the panels. In his research with chipboard reinforced with sago particles and UF and PF resins, TAY *et al.* [39] concluded that the increase in the samples of both resins promoted a reduction in the percentages of water absorption and swelling in thickness due to factors such as the reduction of the hygroscopicity generated by the interaction between the constituents of the resins and the hydroxyls present in the sago and by reducing the existing pores in the biomass. ONG *et al.* [40] also state that wood has porosity on the surface, and the filling of these pores can increase the interactions of bonds between the components that make up the wood.

3.5. Thickness swelling (TS)

Thickness swelling of the panel is influenced by the quality and distribution of the adhesive [41]. Adhesive type, therefore, significantly affects the TS of manufactured particleboard [42]. Although CS can produce a rich foam that fills the interparticle space, thus improving the density (Figure 3B) of the particleboards, due to its affinity to water, this causes speedy expansion followed by collapse when it is removed from the confinement of the press

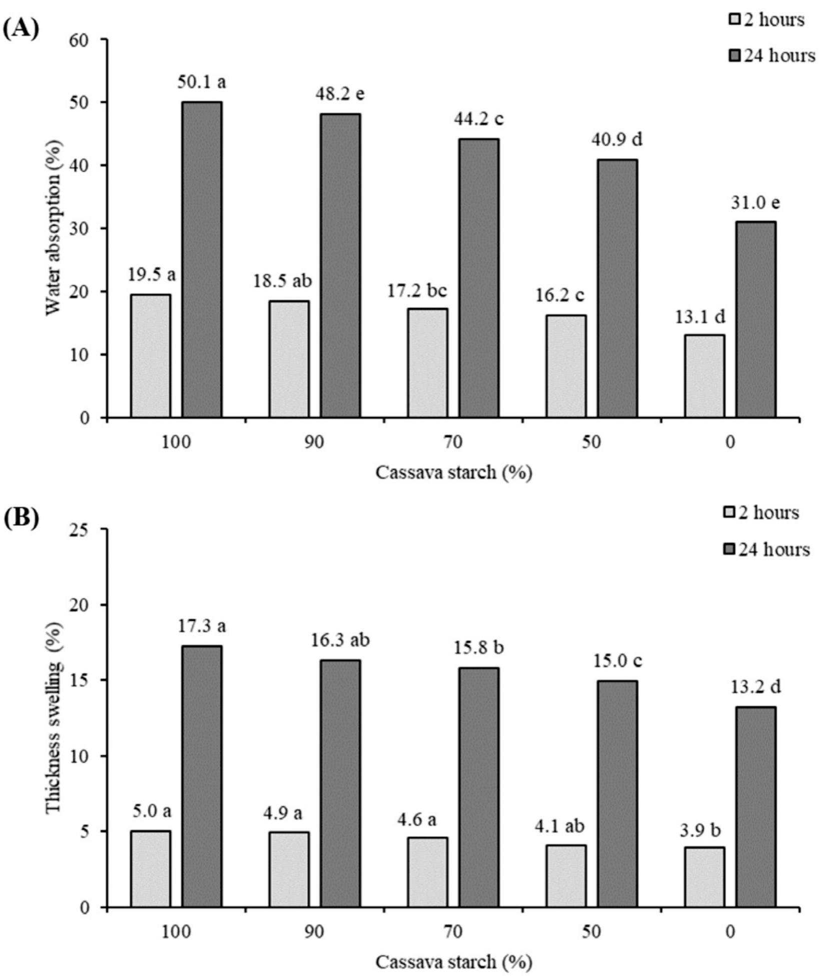

Figure 3: The particleboards' water absorption (A) and thickness swelling (B) were produced using varied adhesive combinations with cassava starch and urea-formaldehyde.

platens. This results in the bursting of the foam cell walls, creating interior cracks that weaken the particleboards during immersion. Thus, the particleboards increase in TS [43].

The thickness swelling increase in the UF resin also decreased the swelling property of the manufactured material. This decrease may be due to the reduction in hygroscopicity caused by the chemical constituents of the resin interacting with the biomass used. According to KOCAEFE *et al.* [44], thermoset adhesives such as phenol-formaldehyde (PF), melanin formaldehyde (MF), and urea formaldehyde, when impregnated in lignocellulosic materials, form a water-insoluble adhesive on the inner structural part of the cell wall. When mixed with starch, SULAIMAN *et al.* [45] expose that adding UF resin modifies the starch, improves its granule properties, and contributes positively to the resistance of manufactured panels.

3.6. Mechanical properties

It was verified that the UF adhesive recorded the highest MoE values for all the treatments compared to the other adhesive combinations. The results indicate that a higher amount of UF encourages stronger interfacial bonding between fibers in the particleboard. Thus, prolonging the ability of the particleboard to withstand resistance to deformation during bending [2, 35]. MIRSKI *et al.* [46] also explain that the particles' dimensions in the external layers directly influence the MoE property during bending. The presence of long, thin particles in the external layers tends to increase the panels' resistance and durability. The panels' resistance properties and modulus of rigidity depend largely on the external layers. Hence, the MoE values obtained were higher than the required limit values given by ANSI A208.1 [47] and JIS A5908 [48] standards (1550 MPa) for low-density particleboards (Figure 4A). According to COSTA *et al.* [49], the mechanical properties of the modulus of rupture and elasticity are fundamental for measuring resistance and analyzing the elastic performance of materials subjected to loads.

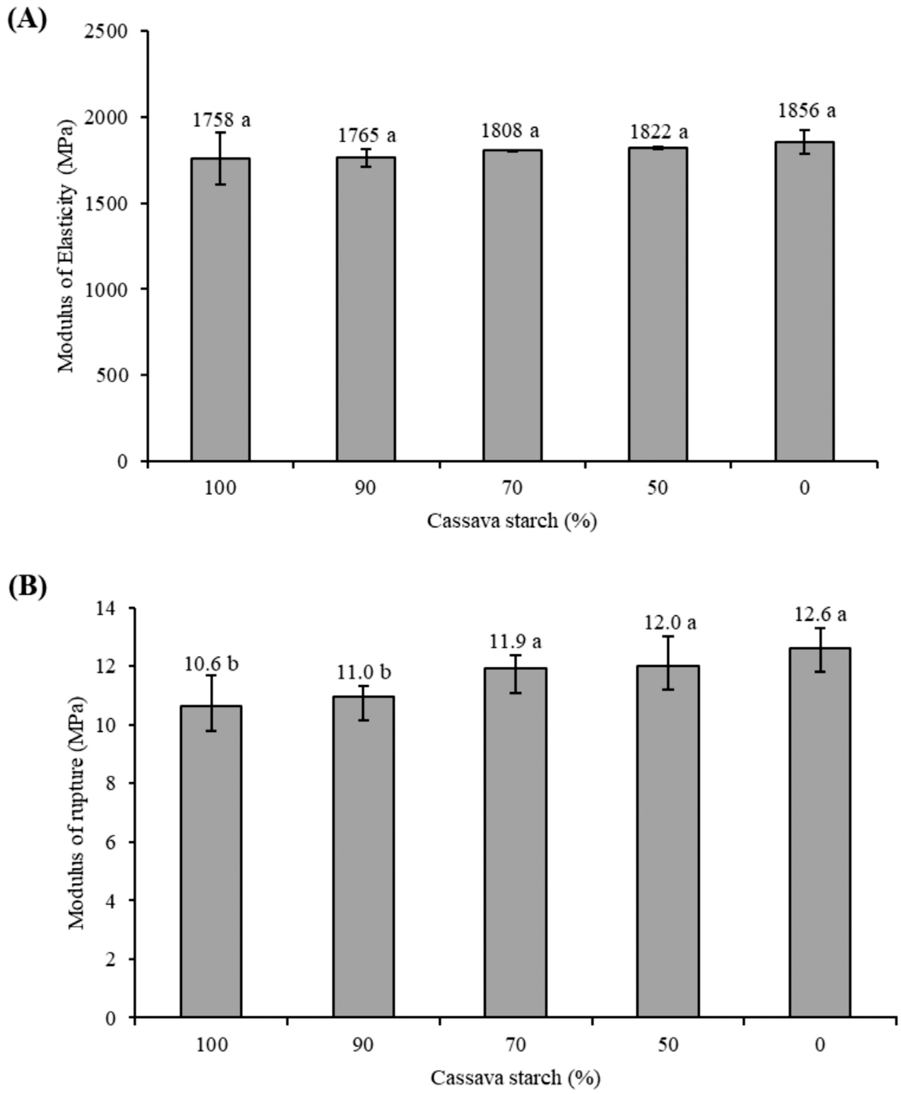

Figure 4: Modulus of elasticity (A) and modulus of rupture (B) for the particleboards produced using varied adhesive combinations with cassava starch and urea-formaldehyde.

MoR values recorded for all the adhesive combinations were higher than the required MoR limits set by the ANSI A208.1 [47] for construction and other fitments (Figure 4B). These results showed that the presence of UF adhesive significantly positively affects bending strength, especially at UF loading of percentages used. This may be due to UF resin's effectiveness in covering the particles' surfaces, enhancing chemical bonding through hydrogen bonds and UF + US covalent bonds [24, 25]. Thus improving the strength of wood before rupture (breaks) during bending.

A hardness test measured the particleboard's resistance to indentation, surface scratching, or the percentages used (Figure 5). This may be due to UF resin's effectiveness in covering the particles' surfaces, enhancing chemical bonding through hydrogen bonds and UF + US covalent bonds [24, 25]. Thus improving the strength of wood before rupture (breaks) during bending. According to SCOUGALL-VILCHIS *et al.* [50] and COMBA *et al.* [51], hardness's mechanical property indicates a given material's resistance to indentation. This property can be directly influenced by the load's size, weight, volume, and chemical composition of the resin used in the material manufacturing process [50, 51].

Hardness tests to measure the resistance of the particleboard to indentation, surface scratching, or the mean values for hardness were obtained by YEMELE *et al.* [52]. Their study evaluated the panels made from extracted black spruce and trembling aspen bark. The same authors recorded mean hardness values of 2.8 to 6.5 MPa for low-density particleboards. The higher the mean hardness value, the higher the hardness. All the particleboards manufactured met the hardness requirements (2.8 MPa) of the ANSI A208.1 [47] and JIS A5908 [48] standards for furniture and other fitments.

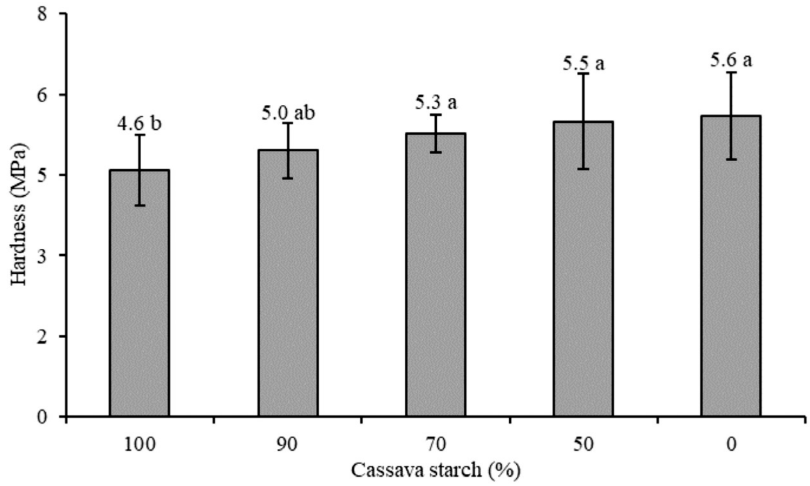

Figure 5: Hardness properties of the manufactured particleboards were produced using varied adhesive combinations with cassava starch and urea-formaldehyde.

The UF addition to the adhesive mix showed different mechanical strength on the particleboards than CS. UF showed low impact strength at 10%. The poor dispersion of particles in the matrix led to weak stress transfer when the load was applied [53]. Thus, LAIVENIECE and MOROZOVS [54] confirm that higher fiber loading caused difficulties for the matrix in flowing through and led to weak stress transfer. Hence, better strength properties with 30% and 50% UF. The result is like that of LIU *et al.* [5], who observed that the higher the percentage of UF adhesive in the blending, the higher the strength properties.

A lower percentage of UF in the mixed adhesive ratio provided lower mean values of MoE, MoR, and hardness. That increase in the quantity of UF in the mixed provided higher mean values. The reason may be that the adhesive mix containing more UF is more reactive [55]. Also, UF's cyclic structure greatly impacts the stability of the resulting linkages. The three amine groups ensure a three-dimensional cross-linked molecular structure when fully cured [55].

Consequently, the mechanical properties using an adhesive mix ratio with higher UF were better. Using a low percentage of formaldehyde-based adhesive did not substantially affect their mechanical properties. Nevertheless, the particleboards produced using the UF partial replacement adhesive were higher than the acceptable standard values by ANSI A208.1 [47] and JIS A5908 [48] standards.

4. CONCLUSION

The interaction between urea-formaldehyde and cassava starch has remarkable synergistic properties, with 50% of cassava starch being the optimal inclusion value. Water absorption, thickness swelling, density, and moisture content obtained from this experiment with panels made with less cassava starch are within the acceptable levels required in the particleboard industry.

This result presents cassava starch-modified urea-formaldehyde as an adhesive with relatively poor moisture uptake and formaldehyde emission against the traditional stern and brittle resin. Cassava starch could produce adhesives for the particleboard industry, especially in formulating biomass particleboard.

The manufactured particleboards using a percentage-varied adhesive ratio are feasible. The particleboards studied are strong enough to meet the requirements for application, including partitioning materials, ceiling, tabletops, and other fitments.

5. ACKNOWLEDGMENTS

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