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EVALUATION OF ENVIRONMENTALLY FRIENDLY PLYWOOD MADE USING GLUTARALDEHYDE-MODIFIED STARCH AS THE BINDER

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Abstract: *A standard resin system used in wood-based panel preparation was formaldehyde-based glue, which was believed to have fatal effects on the environment and human health due to the release of free formaldehyde. Therefore, the study's objective was to evaluate the physical and mechanical properties of experimental plywood made using glutardialdehyde-modified starch as environmentally friendly binder. Two gelatinisation methods were used: sodium hydroxide (NaOH) and hot water. Plywood was manufactured using starch modified with 10, 20, and 30% glutardialdehyde. The physical testing revealed that the plywood density value was around 0.34 to 0.38 g/cm^3, the moisture content ranged between 19.34 and 23.67%, water absorption was between 24.56 and 71.60%, and thickness swelling between 1.18 and 7.27%. It was found that plywood made using NaOH-gelatinized glutardialdehyde-modified starch binder has higher density and lower moisture content, which are advantages for this composite type. In mechanical testing, it was found that plywood made using NaOH-gelatinized glutardialdehyde modified starch binder has higher overall strength than plywood using hot water-gelatinised glutardialdehyde modified starch binder, measured by their bending strength and tensile shear strength. The MOR values of manufactured plywood were found ranging from 124.88 to 179.32 N/mm^2, MOE from 8027.50 to 10603.90 N/mm^2, and tensile shear strength from 651.64 to 1385.16 N/mm^2. The highest glutardialdehyde: starch ratio, which is 30% of glutardialdehyde used based on the weight of starch, showed better overall plywood physical and mechanical properties. The results showed that glutardialdehyde-modified starch could be used as an alternative binder in plywood manufacturing.*

Key words: *plywood, composite, starch, binder.*

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1. Introduction

Composite materials made of wood elements are known as wood-based composite. The art of veneering and using adhesives to attach decorations to wood was discovered by the ancient Egyptians in 3500 BC [6].There are various forms of wood element in wood composites as to different wood composite products to be produced. Yang et al. [24] stated in their study that the standard resin system used in wood-based panel preparation was the three-formaldehyde glue, which mainly consists of Urea-Formaldehyde (UF) resin adhesive, Phenolic-Formaldehyde resin adhesive (PF), and Melamine-Formaldehyde (MF) resin adhesive. Formaldehyde is produced by the oxidation of methanol. However, formaldehyde has adverse effects on the environment and on human health due to the release of free formaldehyde.

It is sorted differently from other pollutants due to its high danger level and it is also known as a potential human carcinogen substance [5]. Thus, it is crucial to develop an environmentally friendly alternative wood adhesive from renewable materials, besides limited oil reserves, expanding the adhesive wood market, and the hazardous issues linked up with formaldehyde-based adhesives [10].

As an alternative, starch has become a substitute for conventional resin systems. Due to its potential, physical and chemical methods could be tested on starch to improve its properties, such as changing its solubility and viscosity and related properties to fulfill the requirements of different application areas of interest. Chemical modification plays a vital role in starch-based adhesive preparation [20].

Starch is typically modified to create better properties of the composites, such as higher bending strength. Wang et al. [23] synthesized an environmentally-friendly starch-based wood adhesive by the graft polymerization of vinyl acetate monomer onto waxy com starch where the shear strength of the grafted starch adhesive increased compared with the blend of commercial polyvinyl acetate (PVAc)/gelatinised starch. Previous researchers conducted studies to enhance the properties of starch as an adhesive by the addition of various chemicals as well as using certain modifications. Various studies have been done to use starch as an adhesive. Corn starch-tannin adhesive [16], glutardialdehyde-modified corn starch–urea formaldehyde resin [1], corn starch-tannin mixed with phenol-formaldehyde resin [15], epichlorohydrin-modified rice starch [22], citric acid modified starch [2], corn starch mixed with sodium dodecyl sulfate [14], and cross linked resorcinol-formaldehyde modified starch with reinforcement of *S. spontaneum* fibres [13] are a few examples.

Currently, there is a lack of investigations concerning the use of glutardialdehyde as a starch-modifying agent for binder production in plywood composite production. This study investigates the better solution to overcoming the issue arising from using formaldehyde-based resin as binder by exploring the possibility of using a starch-based binder and its effect on the properties of the plywood produced. This work aims to determine the properties of plywood using glutardialdehyde modified starch as binder and to study the impact of different glutardialdehyde: starch ratio as starch modifiers on the plywood properties.

2. Materials and Methods

2.1. Sample and Raw Material Preparation

Wood veneers with a dimension of 200 mm x 200 mm were obtained from a local company in Jeli, Kelantan, Malaysia. Glutardialdehyde solution (25% concentration), sodium hydroxide (NaOH) pellets, and corn starch of analytical grade were obtained from Sigma Aldrich.

2.2. Binder Preparation

100 g of corn starch were dissolved in 300 ml of 1M sodium hydroxide (NaOH) solution. The mixture was continuously stirred, followed by adding glutardialdehyde solution. Three different amounts of glutardialdehyde were added, which were 10, 20, and 30% based on the weight of the starch. Stirring was done until resinification was attained and starch became well mixed [3]. Another type of binder with three different percentages of glutardialdehyde was made by substituting NaOH solution with hot distilled water as the control binder. The list of prepared binders is shown in Table 1.

2.3. Plywood Making

Plywood is produced by stacking plies of wood veneers using glues where the layers are rotated up to 90 degrees to one another. In this study, three layers of plywood were produced using rubber wood veneers. Plywood of a final dimension of at least 200 mm x 200 mm was prepared by applying 500gm^{-2} binder on the veneer, cold-pressed at 5 MPa until the binder dried [18]. Six types of plywood were made based on binders listed in Table 1. The parameters involved are different gelatinization method and different amount of glutaraldehyde in the binder formulation.

List of prepared samples Table 1

Sample code	Gelatinisation method	Glutaraldehyde percentage (w/w)
Na10	NaOH	10
Na20	NaOH	20
Na30	NaOH	30
HW10	Hot water	10
HW20	Hot water	20
HW30	Hot water	30

2.4. Evaluation of Plywood Properties

All the test methods followed the Japanese Agricultural Standard Association [12] requirements. The sample size was 50 mm x 50 mm for the density and moisture content test. Water absorption and the thickness swelling test was conducted with soaking time of 1hr, 2 hrs, 4hrs, 12 hrs, and 24 hrs. The plywood was cut into a sample size of 163 mm x 50 mm for the bending strength test. The test specimen was placed with the surface veneer side up. The bending test was conducted using a Testometric M350-5 CT tensile testing machine. The tensile shear strength test was done using 80 mm x 25 mm test pieces, according to the Japanese Agricultural Standard Association [12] requirements. Before testing, two notches were made manually on the upper and lower surface of the test sample using a handsaw. Six replicate samples were used for each test to ensure the reliability of the obtained results. All the data collected were analysed by Analysis of variance (ANOVA), significant at a 95% confidence level.

3. Results and Discussion

3.1. Binder Solid Content Analysis

The mean values of the solid content for the NaOH binder were 24.43, 24.81, and 24.99% for Na10, Na20, and Na30, respectively. For hot water, gelatinised, glutardialdehyde-modified starch binder, the solid content was 24.50, 22.44, and 23.06% for HW10, HW20, and HW30, respectively**.** This showed that NaOH gelatinised, glutardialdehyde-modified starch binder has a relatively higher solid content than hot water gelatinised, glutardialdehyde-modified starch binder. According to Gavrilović-Grmuša et al. [8], the binder particle size would increase as the solids content increased, and their viscosities were also affected by the percentage of solids content. This finding was supported by the observation during the binder-making process, where the NaOH gelatinised glutardialdehyde-modified starch binder was noticeably more viscous than the control binder.

3.2. Physical Properties Analysis

3.2.1. Density and Moisture Content

According to Frihart and Hunt [7], density is the most important physical property of wood, where it is the weight or mass divided by the specimen's volume at given moisture content. Table 2 shows the mean density and moisture content values for both plywoods using NaOH gelatinised glutardialdehyde modified starch binder and hot water gelatinised glutardialdehyde modified starch binder at three different percentages of glutardialdehyde used, respectively.

The density of plywood spread with NaOH gelatinised glutardialdehyde modified starch binder is higher than plywood spread with hot water gelatinized glutardialdehyde modified starch binder with 0.38 g/cm^3 the highest and 0.34 g/cm^3 as the lowest. The raw material density may affect the final plywood density, besides the compression during production and the ability of the binder to hold the veneer together. This could relate to the solid content results mentioned previously, where NaOH gelatinised glutardialdehyde modified starch binder has a higher solid content percentage than hot water gelatinised glutardialdehyde modified starch binder. ANOVA analysis showed no significant difference between different types of plywood, which may indicate that the density of the raw material played a more important role in determining the final plywood density than the binder type. The moisture content percentage ranges from 19.34 to 23.67%. The differences between the samples are not statistically different to each other.

Density and moisture content of plywood Table 2

Sample	Density [/gcm^{-3}]			Moisture content [%]		
	Value	Standard deviation	ANOVA analysis	Value	Standard deviation	ANOVA analysis
Na10	0.37	0.02	a	21.84	1.70a	a
Na20	0.38	0.03	a	21.60	2.12a	a
Na30	0.38	0.02	a	19.34	1.34a	a
HW10	0.34	0.01	a	19.63	1.21a	a
HW20	0.36	0.01	a	21.59	0.73ab	ab
HW30	0.36	0.02	a	23.67	0.61b	b

*different letters in the same column show significant differences at α value of 0.05

3.2.2. Water Absorption and Thickness Swelling

Table 3 shows the water absorption rate for both plywoods manufactured using NaOH and hot water gelatinised, glutardialdehyde-modified starch after immersion in water. Plywood using NaOH-gelatinized, glutardialdehyde-modified starch binder showed lower water absorption rate after being immersed in water at 1 hr, 2 hrs, and 4 hrs, compared to the plywood using hot water gelatinised glutardialdehyde-modified starch binder.

The water absorption rate of the manufactured plywood compared between different types of binder and percentages of glutardialdehyde used Table 3

Water absorption [%]		Sample					
		Na10	Na20	Na30	HW10	HW20	HW30
1hr	V	24.56	23.56	21.13	29.27	27.17	24.99
	SD	0.75	1.56	0.52	2.49	3.47	0.95
	A	b	ab	a	a	a	a
2hrs	V	31.52	29.59	27.35	32.03	31.26	30.16
	SD	0.52	1.00	1.45	2.46	3.00	1.20
	A	b	ab	a	a	a	a
4hrs	V	37.49	36.57	35.60	37.17	37.34	34.52
	SD	1.20	0.61	0.13	2.56	3.87	0.50
	A	a	a	a	a	a	b
12hrs	V	56.64	54.85	52.85	48.67	49.56	45.05
	SD	0.57	2.16	2.02	3.48	3.66	0.90
	A	a	a	a	a	a	a
24hrs	V	71.60	71.09	67.85	60.30	59.45	52.65
	SD	1.48	3.12	3.16	3.45	3.14	1.74
	A	a	a	a	b	ab	a

*V is value, SD is standard deviation, and A is ANOVA analysis

**different letters in the same column and same gelatinisation type show significant differences at α value of 0.05

However, after being submerged in water for 12 and 24 hrs, plywood using NaOH-gelatinised, glutardialdehyde-modified starch binder showed a higher water absorption rate than plywood using hot water gelatinised glutardialdehyde modified starch binder.

Thickness swelling indicates the increment of the plywood thickness after being immersed in water for a certain period. Table 4 shows the thickness swelling rate for all manufactured plywood. Plywood with hot water-gelatinised binder showed higher thickness swelling overall compared to plywood with NaOH-gelatinised glutardialdehyde-modified starch binder. Despite having a higher water absorption rate for plywood using NaOH-gelatinised glutardialdehyde-modified starch binder, it has a lower thickness swelling rate due to stronger bonds inside the wood structural panels that slow down the water penetration rate. Generally, the thickness swelling for all plywood showed an increasing trend since the particles in the plywood had expanded over the immersion period. The geometry of wood particles, their structure, and the presence of many voids in the plywood panel allow internal swelling to influence the thickness swelling rate [9].

Table 4

Thickness swelling of the manufactured plywood compared between different types of binder and percentages of glutardialdehyde used

Thickness swelling [%]		Sample					
		Na10	Na20	Na30	HW10	HW20	HW30
1hr	V	1.33	1.83	1.18	3.00	5.22	4.45
	SD	0.55	0.26	0.42	0.47	0.46	0.24
	A	a	a	a	a	b	b
2hrs	V	2.67	2.49	1.80	3.49	5.98	5.02
	SD	0.09	0.23	0.36	0.27	0.65	0.22
	A	b	b	a	a	b	b
4hrs	V	3.10	3.06	2.54	3.85	6.46	5.37
	SD	0.28	0.42	0.42	0.21	0.47	0.22
	A	a	a	a	a	b	b
12hrs	V	4.19	3.68	3.22	4.79	6.75	5.91
	SD	0.40	0.84	0.43	0.12	0.46	0.70
	A	a	a	a	a	b	b
24hrs	V	4.48	4.22	3.77	5.56	7.27	6.31
	SD	0.40	0.74	0.26	0.33	0.54	0.19
	A	a	a	a	a	b	ab

*V is value, SD is standard deviation, and A is ANOVA analysis

**different letters in the same column and same gelatinisation type show significant differences at α value of 0.05

3.3. Mechanical Properties Analysis

3.3.1. Bending Strength

The results for modulus of rupture, or MOR, for plywood bending strength are shown in Figure 1. MOR reflects the strength properties of wood where the maximum load-carrying of a member in bending is proportional to the maximum moment endured by the specimen. The results showed the MOR for plywood with all types of binder was on an increasing trend where it was 152.89, 174.85, and 179.32 N/mm^2 for Na10, Na20, and Na30 plywood, respectively, while 124.88, 156.94, and 175.26 N/mm^2 for HW10, HW20, and HW30 plywood, respectively. All the manufactured plywood passed the Japanese Industrial Standard, which required a minimum of 20.0 N/mm^2 of MOR for plywood with thickness ranging from 12 to 21 mm.

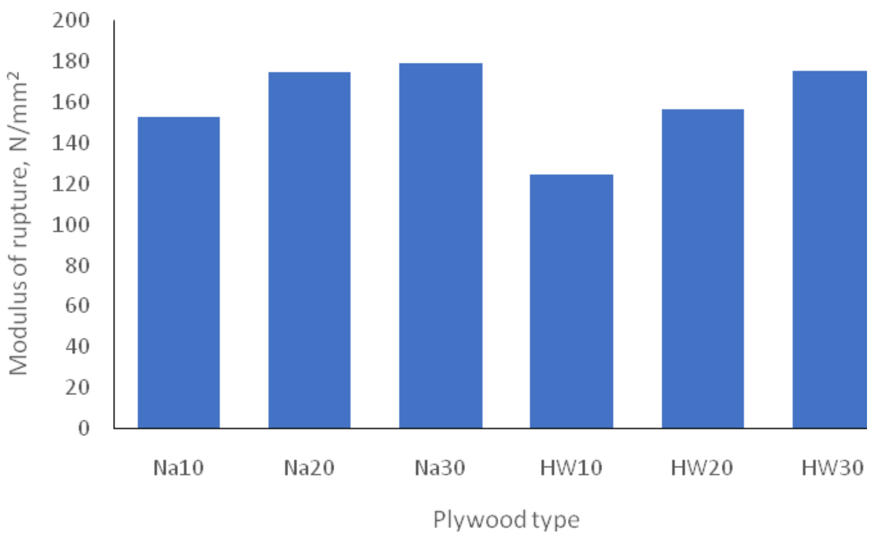

Fig. 1. *Modulus of rupture for bending strength of all samples*

The modulus of elasticity, or MOE, indicates the measure of the resistance to bending deflection relative to the stiffness. Elasticity expresses that distortions produced by low stress under the proportional limit are completely recoverable after loads are removed. However, when more stress is loaded beyond the proportional limit, distortion will occur, also called a failure or plastic deformation [7]. Figure 2 shows the results for the MOE for plywood bending strength. With a higher percentage of glutaraldehyde, the trend increased for both plywoods with NaOH and hot water-gelatinised glutardialdehyde-modified starch binder. The MOE for plywood bending strength with NaOH-gelatinised glutardialdehyde modified starch binder were 8027.50, 9689.76, and 10603.90 N/mm^2 for Na10, Na20, and Na30, respectively. Meanwhile, for HW10, HW20, and HW30 plywood, the MOE for plywood bending strength was 10214.07, 10471.45, and 10540.36 N/mm^2, respectively.

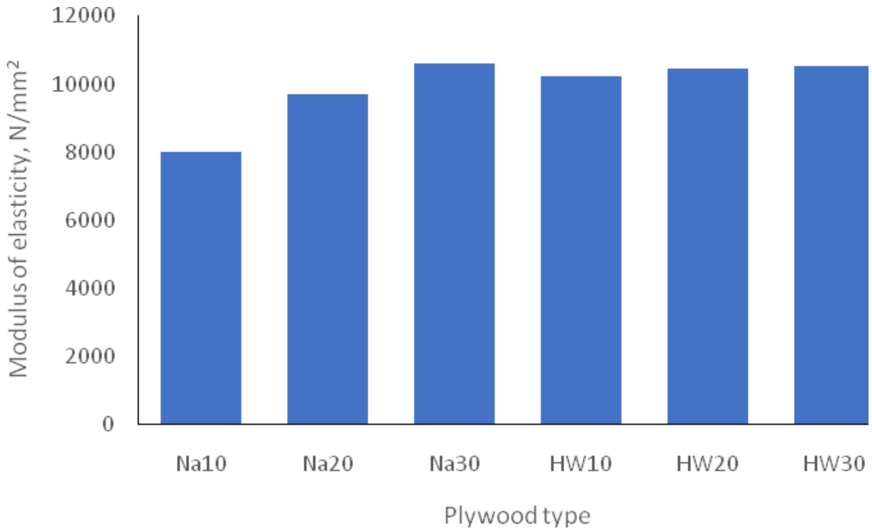

Fig. 2. *Modulus of elasticity for bending strength of all samples*

3.3.2. Tensile Shear Strength

Figure 3 shows tensile shear strength for all manufactured plywood. The higher percentage of glutaraldehyde in the formulation leads to higher tensile shear strength of plywood using NaOH-gelatinised starch. However, this trend was the opposite for plywood made using hot water-gelatinised starch. The mean values of Young's modulus for plywood with NaOH-gelatinised glutardialdehyde-modified starch binder were 709.92, 772.11, and 1385.16 N/mm^2 for Na10, Na20, and Na30, respectively. Meanwhile, the recorded values of plywood with hot water-gelatinised glutardialdehyde-modified starch binder were 997.46, 809.74, and 651.64 N/mm^2 for HW10, HW20, and HW30, respectively. The difference in results could be due to the higher moisture content of plywood with hot water-gelatinised glutardialdehyde-modified starch binder compared to plywood with NaOH-gelatinised glutardialdehyde-modified starch binder.

3.3.3. Comparison

Table 5 shows the comparison between various types of previously researched natural substance-based adhesive plywood. It was found out that the glutaraldehyde modified starch plywood, taken from the Na30 sample, showed comparable mechanical strength. The MOR of Na30 was slightly higher than the plywood made using partially delignified wood while the MOE was slightly lower at 10603.90, compared to 18000 by the closest competitor.

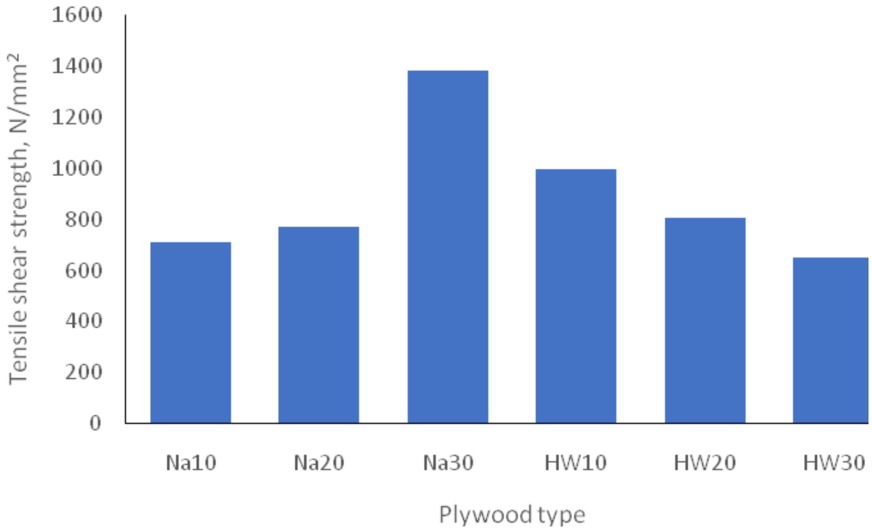

Fig. 3. *Tensile shear strength of all samples*

Plywood types made by previous researchers Table 5

Plywood type	MOR (bending) [N/mm^2]	MOE (bending) [N/mm^2]	Reference
Glutaraldehyde modified starch	179.32	10603.90	This work
Plywood from Partially Delignified Densified Wood	155.05	18000	[11]
Beech Bark in Adhesive Mixtures	100	9000	[19]
Urea formaldehyde (UF) resin with bark flour	78.45	4373.20	[4]
Corn starch and tannin mixture	70	4271	[15]
Dimethylol dihydroxy ethylene urea crosslinked rice starch- natural rubber latex-based adhesive	63	6840	[21]
Soya based adhesive	45.93	-	[17]

4. Conclusions

From the refreshing results of this research, NaOH and hot water-gelatinised glutardialdehyde-modified starch are suitable for plywood binders. The strength produced by plywood using modified starch binder passed the Japanese Agricultural Standards (JAS) No. 233:2003 [12] requirements. Plywood using NaOH-gelatinised glutardialdehyde-modified starch binder showed better results in most tests conducted, whereas in the tensile shear strength test, the results were comparable to the plywood using a hot water-gelatinised glutardialdehyde-modified starch binder with the different trend of results obtained. It was also proven that moisture content significantly affects the mechanical properties of manufactured plywood. Plywood using NaOH gelatinised glutardialdehyde-modified starch binder with lower moisture content showed better performance in mechanical testing. In addition, most of the preferable results were obtained from plywood using NaOH-gelatinised 30% glutardialdehyde-modified starch binder (Na30), indicating that different glutardialdehyde: starch ratios affect plywood properties.

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