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Foamed phenol formaldehyde (PF) with naturel foaming agent and characterization adhesive properties

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Abstract – In this study, the phenol formaldehyde resin volume was increased (foamed) using natural foaming agents. Increasing production costs force manufacturers to produce using fewer raw materials. In recent years, in the wood sector, studies have been carried out on glue efficiency by foaming the glue. In this study, egg white (EW), sodium bicarbonate (SB), cattle gelatin (GL), wheat starch (WS) and wheat flour (WF) were used as foaming agent. 5-10 % foaming agent was added to the PF and foamed by mixing at 3000 rpm with a mechanical mixer. Foamed PF has been used to bond black pine wood (BP). Internal bond (IB), density, water absorption (WA) and jell time analyses were performed to characterize foamed PF. The foamed sample made with EW was determined to be the best by the study's findings. In terms of increasing IB strength, the foaming process with GL has also been successful. Other foaming agents have also been successful in the foaming process, but their IB strength has been reduced. In this regard, it is advantageous to use EW and GL agents for foaming PF glue. More research should be done on this topic to determine how different foaming agents, in different proportions, affect the efficiency of glues. Glue savings can thus be realized in industries that use glue.

Keywords – Cattle gelatin, foaming, foamed adhesive, internal bond, phenol formaldehyde

I. INTRODUCTION

Foaming the adhesives is an old issue which was studied researchers because the find an effective gluing process. The first studies on this issue were conducted in the United States. Menger [1], described the preparation of a glue mixture for plywood production. The mixture consists of a urea formaldehyde condensation product, casein, urea, ammonia solution, water, ammonium chloride, butylated naphthalene sulfonic acid, and wood meal. Air is incorporated into the mixture using a stirring mechanism until its volume expands to three to five times the original volume. The resulting glue mixture is applied to wood veneers using roller machines at a rate of 200-250 cubic centimeters per square meter of surface. The plywood is assembled by coating both sides of a 2 mm thick camwood veneer with the glue and placing untreated veneers on either side with perpendicular grain alignment. The assembly is then pressed at 90-100 °C and 10 atmospheres of pressure for 10 minutes, ensuring adhesion and destroying the foamy structure of the glue. The writer claims three different processes for adhesion using a liquid foam adhesive. The first process involves coating wood pieces with a foam consisting of urea formaldehyde condensation product, casein, ammonia, and ammonium chloride. The pieces are then pressed together at 90-100 °C and a pressure of about 10 atmospheres. The second process uses a similar foam but substitutes bone glue for casein. The third process replaces casein with blood albumen in the foam. In all three processes, the wood pieces are cemented together through pressing under specific temperature and pressure conditions.

Gillern and Freeman [2] introduced a technique for bonding materials, specifically wood, through the utilization of foamed adhesives with gap-filling properties. These adhesives were formulated by incorporating foaming agents into amine-modified aldehyde condensation polymers. The resulting resins exhibited rapid curing at room temperature and demonstrate exceptional performance as construction adhesives.

Cone and Steinberg [3] studied about foamed adhesive for plywood process. The process of manufacturing plywood involved the continuous propulsion of liquid plywood glue, maintaining a predetermined flow rate. As the glue was propelled, it was simultaneously foamed. The foamed glue was then continuously extruded or applied onto multiple wood veneers. The flow rates of both the unfoamed and foamed glue were carefully controlled to ensure an equal unit weight of liquid glue per unit time. Once the veneers were arranged into a plywood assembly, they were pressed together, causing the foam to break and the veneers to unite. This method can be applied more broadly to produce laminar products by applying foamed liquid onto solid surfaces.

In another study, Cone and Steinberg [4], reported that a rapidly-foamable phenolic resin liquid plywood glue designed for use in a plywood production line, specifically in conjunction with an in-line continuous foaming unit. The glue formulation consists of an alkaline aqueous solution containing a highly advanced phenol aldehyde resin that possesses fast setting properties. Additionally, a dried animal blood foaming agent was incorporated into the glue. To control the viscosity of the glue, glyoxal or another aldehyde compound was included. When the glue was agitated in the presence of air or another gas, it uniformly forms foam with a density around 0.2 gm/cc within approximately two seconds.

Wawzonek [5], described a method and particle board composition involve the use of urea formaldehyde resin adhesive while effectively suppressing formaldehyde emissions. This was achieved by adding a small amount of a weak acid and calcium metal silicate to the formaldehyde resin. The weak acid and calcium metal silicate acted as suppression agents, reducing the release of formaldehyde.

Detlefsen et al. [6] reported a highly advanced phenolic resin with improved foamability. This was achieved by incorporating lime (calcium hydroxide) into the early stages of the reaction mix. The solution was formed through the reaction of phenol, aldehyde, water, and alkali. The invention also encompassed a plywood glue composition that included the lime-containing highly advanced phenolic resin solution, along with dried animal blood and an optional aldehyde glue viscosity-lowering agent.

In another study, Watters and Wellons [7], foamed the UF resin by freon 12 gas and obtained improved internal bond and modulus of rapture in particle board due to foaming. They reported that production cost can be decreased by this foaming process. But in this process freon 12 gas was too expensive. Foaming process was effective, but it was not used by manufacturers because of its cost.

Also, the furniture industry is increasingly relying on lightweight particleboard to facilitate easier transportation and assembly for customers. A novel method for producing foaming particleboard is proposed, involving the addition of azodicarbonamide (AC) foaming agent to a formulation consisting of oven-dry poplar particles and phenol formaldehyde resin (PF resin). The study explores the impact of the AC foaming agent and adhesive contents on the mechanical, physical, and chemical properties of the particleboards. The results indicate that the addition of the AC foaming agent significantly influences the particleboard properties, with optimal performance achieved at a density of 0.6 g/cm^3, a PF resin amount of 12 %, and an AC foaming agent amount of 1 %. Furthermore, the appearance of pores on the particle surface is attributed to the radical pyrolysis of the foaming agent, as confirmed by FTIR results, and these pores also affect the particleboard properties [8].

Not much work has been done on the foaming of glues. The work done is briefly summarized. Other studies are Kelleci et al. [9] about Eco-friendly particleboards with low formaldehyde emission and enhanced mechanical properties produced with foamed urea-formaldehyde resins, Dorsey et al. [10] about studies of foamed epoxies, Sellers [11] about Foamed adhesives for particleboard, Hojilla-evangelista [12] about Adhesive qualities of soybean protein-based foamed plywood glues.

In this study, we aimed to produce foamed PF by natural foaming agent. Thus, eco-friendly foamed glue could be produced. We used the egg white, cattle gelatin, sodium bicarbonate, wheat starch and wheat flour to foam the PF resin.

II. MATERIALS AND METHOD

A. *Materials*

Natural foaming agents (EW, SB, GL, WS and WF) were purchased local market. PF and Ammonium sulphate (AS) was purchase by online marketing. Materials ingredients and ratios in the samples were given in Table 1.

Table 1. Materials ratios in samples

Sample Code	**PF^1 (gr)**	**AS^2 (gr)**	**EW^3 (gr)**	**GL^4 (gr)**	**SB^5 (gr)**	**WS^6 (gr)**	**WF^7 (gr)**
PF	200	15	-	-	-	-	-
EW	200	15	10	-	-	-	-
GL	200	15	-	10	-	-	-
SB	200	15	-	-	10	-	-
WS	200	15	-	-	-	10	-
WF	200	15	-	-	-	-	10

1: 48% Phenol formaldehyde solution, 2: 30% Ammonium sulfate, 3: 10% protein %90 water, 4: 100% solid, 5: 100% solid, 6: 100% solid, 7: 100% solid.

B. *Preparation of samples*

PF resins were mixed by naturel foaming agent according to given ratios in Table 1. 5 foamed sample and 1 control sample (PF) were prepared by mechanic blender in a 600 ml glass container at 3000 rpm (rotary per minute) (Fig.1). IB analysis sample was prepared from 2 pieces of with 3 mm x 50 mm x 50 mm BP woods which were poured their surfaces 1 g foamed PF. Two pieces of BP woods were sticked together and pressed with a hand clamp by manually. Pressed samples were placed in oven at 150 °C for 20 minutes.

Fig. 1 Foamed PF samples (a), and initial level (b)

C. *Characterization of samples*

Characterization of samples were carried out mechanically (by internal bond) and physically (density and water absorption). Internal bond (IB) analysis was carried out according to TS EN 319. Water absorption (WA) analysis was carried out according to TS EN 317. Density of samples were determined according to TS EN 323.

D. *Statistical analysis*

Results were presented statistically. One-way Anova statistical analysis method was done to obtained data by SPSS 20 software program. Duncan analyses were applied to obtained data and determined same samples which were same group.

III. RESULTS

Results show that jell-time were similar to each other, but Duncan analyze result show that there are four group in jell-time (Table 2). Foaming agents affect the IB strength and carried out five group according to Duncan analysis. Samples densities also very close to each other but nevertheless samples carried out five group in Duncan analysis. The WA performances of the samples were also very close to each other.

Table 2. Analysis (Duncan) results

Samples	Jell-Time (second)	IB (N/mm^2)	DN (g/cm^3)	WA (%)
PF	220 b	1,02 c	1,36 e	2,2 a
EW	230 d	1,21 e	1,32 c	2,3 d
GL	224 c	1,12 d	1,30 a	2,2 b
SB	225 c	0,98 b	1,31 b	2,3 c
WS	220 b	0,95 ab	1,32 cd	2,3 de
WF	215 a	0,91 a	1,32 d	2,3 e

According to the results of the One-way Anova statistical analysis, in all results, the P value (significant degree) was < 0.05. This shows that there are significant differences in jell-time, internal bond, density and water absorption results between samples (Table 3). The descriptive results of the samples with a 95 % confidence interval are given in Table 4.

Table 3. One-way Anova results

		Sum of Squares	df	Mean Square	F	Sig.
Jell-time	Between Groups	427	5	85,4	21,6	,00
	Within Groups	47	12	3,9		
	Total	474	17			
Internal bond	Between Groups	,208	5	,042	122	,00
	Within Groups	,004	12	,00		
	Total	,212	17			
Density	Between Groups	,006	5	,001	285	,00
	Within Groups	,000	12	,000		
	Total	,006	17			
Water absorption	Between Groups	,065	5	,013	146	,00
	Within Groups	,001	12	,000		
	Total	,066	17			

Table 4. Descriptives results

		Mean	Std. Deviate	Std. Error	95%	
					Lower	Upper
Jell-time	PF	220	1,50	,88	216	224
	EW	229	1,52	,88	225	233
	GL	224	2,00	1,15	219	228
	SB	225	1,00	,57	222	227
	WS	220	1,00	,57	217	222
	WF	214	3,60	2,08	205	222
	Total	222	5,28	1,24	219	224
Internal bond	PF	1,02	,02	,01	,97	1,07
	EW	1,21	,02	,00	1,17	1,25
	GL	1,12	,03	,01	1,06	1,18
	SB	,96	,02	,00	,92	1,00
	WS	,94	,01	,01	,91	,96
	WF	,91	,02	,01	,86	,95
	Total	1,02	,11	,03	,97	1,08
Density	PF	1,36	,00	,00	1,35	1,37
	EW	1,31	,00	,00	1,31	1,32
	GL	1,30	,00	,00	1,30	1,31
	SB	1,31	,00	,00	1,30	1,31
	WS	1,32	,00	,00	1,31	1,30
	WF	1,32	,00	,00	1,32	1,32
	Total	1,32	,02	,00	1,31	1,33
Water absorption	PF	2,21	,01	,01	2,18	2,23
	EW	2,35	,01	,01	2,32	2,37
	GL	2,25	,01	,01	2,22	2,27
	SB	2,27	,01	,01	2,24	2,29
	WS	2,35	,01	,00	2,33	2,36
	WF	2,37	,01	,01	2,34	2,39
	Total	2,30	,06	,01	2,26	2,33

According to the results of the statistical analysis, it was concluded that the foaming process with egg white, sodium bicarbonate, cattle gelatine, wheat starch and wheat flour revealed significant differences in PF glue. In particular, the use of egg whites (EW) has led to an increase in the IB strength of PF (Fig. 2). Similarly, GL increased IB strength, but SB, WF and WS foaming agents caused the IB value to decrease. There was some variation in the densities of the samples with the use of natural foaming agent. The foaming agents reduced the densities of the samples. When Fig. 3 was examined, it was seen that the GL agent reduced the density the most.

IV. DISCUSSION

The naturel foaming agents were successful to increase the PF resin volume. The internal IB strength in the mechanical properties had increased with the addition of GL and EW. The reason for this is thought to be that there is ovalbumin in EW and collagen in GL. Previous studies have also reported that the use of EW increases the strength of IB [9], [13]. The negative effect of EW use on PF is that it prolongs the jell time. When Fig. 2 was examined, it was determined that EW extended the jell time more than the others. This is thought to be caused by the amount of water in ovalbumin. When Fig. 3 was examined, it was seen that there was a change in the physical properties of the samples as a result of the foaming process. The foaming process reduced the densities of the samples. This is a desirable feature for the manufactured product. Especially in particle board construction, a lot of research was done on low density board production [4], [6], [11], [14]–[17]. In these studies, similar to our study, it used the process of foaming the glue. When Fig. 3 was examined, an increase in the WA of the samples was seen. This increase is negligible because foaming glue can save glue.

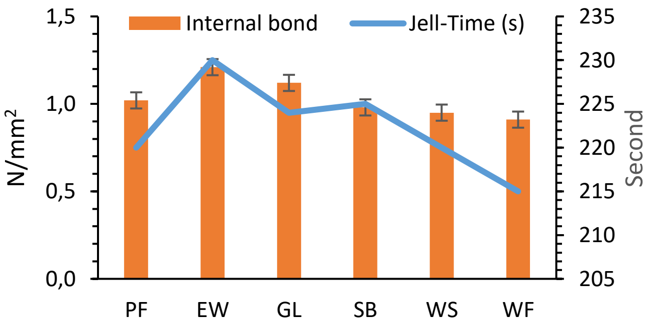

Fig. 2 IB strength of samples rely on jell-time

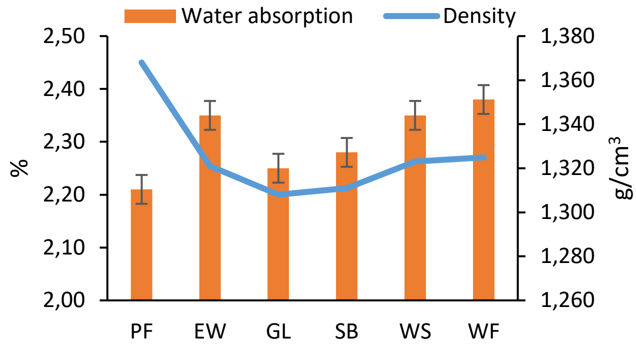

Fig. 3 Physical properties of foamed PF sample

V. CONCLUSION

In this study, PF glue was foamed using natural foaming agents. Foaming of glues is an important issue in terms of increasing gluing efficiency. According to the results of the study, it was determined that the foamed sample made with EW was the best. The foaming process with GL has also been successful in terms of increasing the IB strength. Other foaming agents have also been successful in the foaming process but have reduced the IB strength. In this respect, it is useful to prefer EW and GL agents for foaming PF glue. Further research on this topic should be carried out to determine how different foaming agents, in different proportions, affect the efficiency of glues. In this way, glue savings can be achieved in industries that use glue.

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