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Citric acid, an environmentally friendly adhesive and wood impregnation material-review of research

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Abstract. Citric acid's potency to be adhesive is proven with an increasing quality of product bonding resulted from wood and other lignosellulosic materials. Wood waste, bagasse, sorghum and corn stalks, and bamboo are quality particleboard materials after addition of 20% citric acid. Moulding made of a mixture of tree bark and citric acid with the same concentration is also able to pass repeated boiling test without having any damage. Ester bond between wood's hydroxyl group and citric acid's hydroxyl group is formed at 180°C and gets more optimal as the pressing temperature increases during production process. This results in an increase of board's physical-mechanical properties pursuant to the standard. Some researches remain presenting non-standard test parameters and this is a challenge to be answered with further research. The density of Samama wood impregnated with 10% citric acid has its density increased between 17.11% and 20.13%. Samama wood does not experience a recovery of set after it has been pressed for 50 minutes at 180°C.

1. Introduction

Environmental issue is one important point in future technology development including adhesive technology for lignosellulosic materials. Alternatives to various petroleum resin based adhesives like urea formaldehyde (UF), phenol formaldehyde (PF) and melamine formaldehyde (MF) start to be searched for since they are evidently less environmentally friendly. Formaldehyde emission generated by these adhesives is hazardous to health. To reduce the emission may be made by changing adhesive formula[1], adding surface layer[2] and changing adhesive[3, 4].

Many environmentally friendly adhesive alternatives have been studied to substitute or even potentially replace formaldehyde containing materials. One of these alternatives is a compound generated from plant of genus citrus, which is citric acid. Citric acid may well bind hydroxyl groups of wood[5], thus may well be distributed and form *cross-link*. Many researches of citric acid have been presented with various methods and are continuously developed to generate the best product. In addition, citric acid has also been used as a fixation material for Samama wood densification. Find this more in Tabel 1.



Table 1. Citric acid as adhesive for lignocellulosic materials.

Num.	Application	Support
1	Adhesive for Molding with tree bark material	[5-7]
2	Adhesive for particleboard with material:	
	a. Nypa frond and oil palm midrib	[8-10]
	b. Wood waste	[11-13]
	c. Bagasse	[14, 15]
	d. Sorghum and corn stalks	[16, 17]
	e. Bamboo	[18-21]
3	Samama wood impregnation	[22]

2. Citric Acid as adhesive

2.1. Tree bark

Acacia (*Acacia mangium*) tree bark powder is mixed with citric acid and poured onto iron molding at 180°C, 4 MPa for 10 minutes. The Modulus of Rupture (MOR) and Modulus of Elasticity (MOR) moldings employ adhesive of 20 wt% citric acid, each at 18.1 MPa and 4.9 GPa. The molding is not damaged during the repeated boiling treatment. Based on the result of Fourier transform infrared spectra, ester bond formation is seen between citric acid's carboxyl group and tree bark's hydroxyl group, particularly at high temperature. These chemical bonds confirm that citric acid is worthy to be used as natural adhesive[5-7].

2.2. Particleboard

2.2.1. *Nypa frond* (*Nypa fruticans* Wurm) and oil palm midrib. An addition of 10% citric acid increases the physical and mechanical properties of particleboard made of nipa palm midrib[8-10]. The optimal temperature for pressing process is 180°C with a duration of 15 minutes. In other research employing oil palm midrib, the optimal temperature for the best quality is 200°C with a duration of 10 minutes[9].

2.2.2. *Wood waste*. *Albizia chinensis* wood waste based particleboard is made using a mixed adhesive of citric acid and sucrose. The sucrose addition serves to provide hydroxyl groups and increase the number of ester groups. Sucrose mixture in adhesive formula evidently increases particleboard's physical properties. On the contrary, sucrose without citric acid does not show a good binding property at 180°C[11]. The optimal temperature for particleboard making from wood waste is 200°C with specific pressure of 3.6 MPa and duration of 10 minutes. The best quality wood is shown with a mixed adhesive of citric acid and sucrose 50:50 wt%, with solid content 15 wt%[12]. Other research with a ratio of adhesive to solid content 25:70 and 30 wt% shows the internal bonding (IB), MOR and thickness swelling (TS) values pursuant to JIS A5908 (2003) for board type 18[23]. The wood waste used is that of softwood with making process at 200°C and pressing duration for 10 minutes.

2.2.3. *Bagasse*. *Saccharum sp.* processing at sugar mill leaves bagasse. This material may be made particleboard using citric acid adhesive[14]. The best quality particleboard in the research uses 10% citric acid at press temperature 200°C. The specific pressure of the production process is 2 MPa with duration of 10 minutes. The particleboard produced has its density of 0.66 g/cm³ with indicators of quality including 4.88% moisture content, 1.98% thickness swelling, 29.29% water absorption, 1121 N/mm² modulus of elasticity, 5.04 N/mm² modulus of rupture and 0.17 N/mm² internal bonding. Furthermore, other research presents a combination of 20 wt% citric acid, press temperature 200°C for 10 minutes which produces the best quality sweet sorghum (*Sorghum bicolor*) bagasse particleboard[15]. The board's Dry bending (DB), Internal bonding (IB) and Thickness swelling (TS)

are pursuant to JIS A 5908 (2003). The results of infrared spectra analysis show that ester's degree of bonding increases in line with an increase of temperature and press time.

2.2.4. Sorghum Stalk and Corn Stalk. Particleboard produced from a mixture of sorghum stalk and *Albizia chinensis* wood waste has also been reported using citric acid adhesive. The best quality board is produced from a combination of 100% sorghum stalk with press temperature 200°C, press time 10 minutes and pressure 25 kgf/cm². Usage of 20% adhesive results in the best quality board in comparison with 15% adhesive[16]. Meanwhile, for particleboard made of corn stalk, the use of 20% citric acid adhesive also shows board with Japanese quality standard, JIS A5908 (2003)[17]. Figure 1 shows display comparison between corn stalk particleboard with citric acid and without citric acid adhesives.

2.2.5. Bamboo. Betung bamboo (*Dendrocalamus asper*) particle may be made good quality board using 20 wt% citric acid[18]. This board making process is performed at pressure temperature 200°C for 10 minutes, resulting in optimal IB, MOE and MOR values of 0.44 MPa, 15.1 MPa and 4.6 GPa, respectively. The board's IB and MOR values are pursuant to JIS A5908 (2003), particularly particleboard type 8. Its MOE value is pursuant to a higher grade, which is type 13. In comparison with bamboo composite board using formaldehyde based adhesive, either particleboard products, composite beam[20] or OSB[19], the results of this research further confirm the advantage of citric acid as adhesive material. Besides mechanical properties, the dimension stability of particleboard using betung bamboo, tali bamboo (*Gigantochloa apus*) and hitam bamboo (*Gigantochloa atroviolacea*) particles also increases and is pursuant to JIS A5908(2013) after citric acid addition[21]. However, the type of bamboo factor does not present significant difference in physical and mechanical properties.

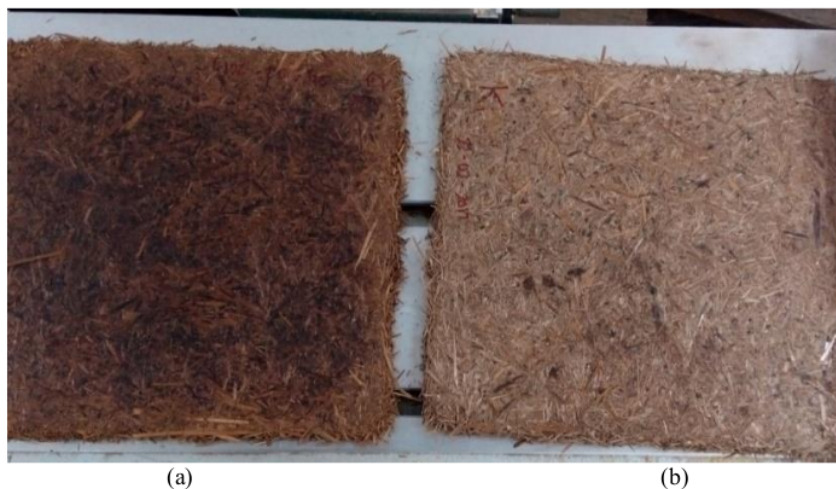


Figure 1. Particleboard using citric acid (a) and without citric acid (b). Photo: [17].

3. Citric Acid improves Samama wood density

3.1. Adhesive impregnation for wood density improvement

This paper tries to elaborate citric acid usage as adhesive, including presentation of adhesive impregnated into wood. An impregnant generally used is formaldehyde based resin [24-28]. The purpose of such quality modification is to improve wood's density, dimension stability and strength [29, 30]. Wood's durability from deterioration caused by physical, chemical and biological factors are bonus for each treatment.

Various adhesive impregnation methods have been performed, from simple to complicated ones. One of the methods is pre-compression, particularly applied to wood with low density. This method is

able to improve water absorption into wood[31]. The pre-compression method is certainly not applied independently, but combined with other methods for adhesive maximum retention.

3.2. Characteristics of Samama impregnated with citric acid

Researches of citric acid impregnation using pre-compression method have been conducted on Samama wood[22]. Pre-compression is performed at 100°C for 1 hour until reaching drying set condition. The second phase is to put wood into *autoclave* under pressure between 5 – 7.5 kg/cm² (at room temperature) for 4 hours, submerged in citric acid solution. The solutions used are 5% and 10%. The third phase is repressing at 180°C with press time variable of 10, 20, 30, 40, 50 and 60 minutes. This series of method has successfully improved Samama's oven-dry density at 0.44 - 0.45 g/cm³, an increase between 17.11 - 20.13%. This density increase is relatively low compared to the density increase of wood impregnated using phenol formaldehyde adhesive. The reason of this is that phenolformaldehyde adhesive generally fills in lumen, while citric acid is expected to get into cell wall and form ester bond like what occurs in particleboard.

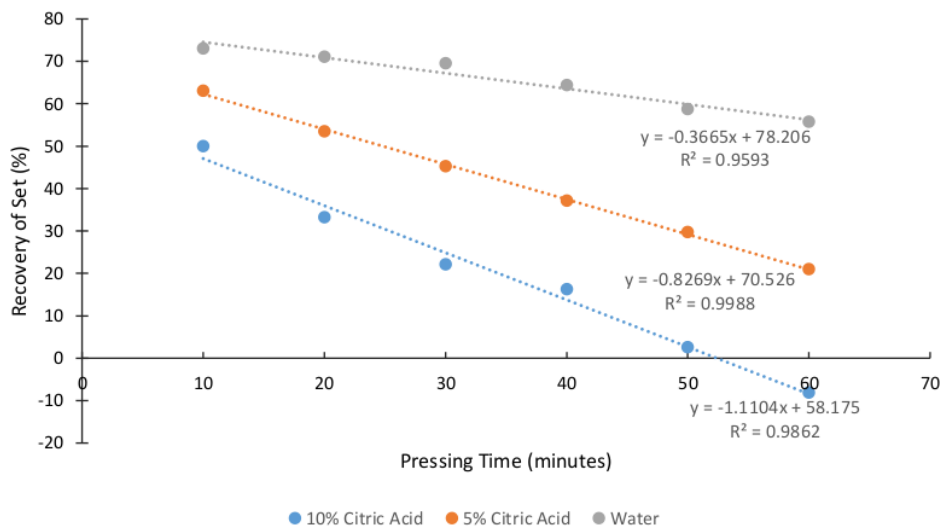


Figure 2. The recovery of set of wood test sample impregnated with citric acid[22].


Figure 2 shows that the recovery of set (RS) of Samama without citric acid is still high (>55%). An addition of 5% citric acid seems to be able to reduce RS, but the value is still 21% with wood impregnation at 180°C for 1 hour. Samama wood fixation occurs at a concentration of 10% citric acid with press time of 50 minutes. If desired, the temperature and citric acid concentration may be increased to reduce press time.

4. Conclusion

Citric acid is an environmentally friendly material evidently having advantages in its usage as adhesive for wood and materials with lignocellulose. Citric acid is able to bind OH group in wood to form ester bond, especially when the production process is performed at above 180°C. The application of 20% citric acid produces quality molding and particleboard, pursuant to the standards. Some parameters which are not pursuant to the standards are continuously anticipated and some solutions have been presented for them. Furthermore, as an impregnation material, the use of 10% citric acid is able to improve Samama wood density up to 20%. A recovery of set (RS) = 0 occurs after 50 minutes of pressing at 180°C.

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