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Effect of Jatropha Seed Oil Meal and Rubber Seed Oil Meal as Melamine Urea Formaldehyde Adhesive Extender on the Bonding Strength of Plywood

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Abstract: Extenders are added to adhesive formulations to reduce resin utilization leading to cost savings. In this work, Melamine Urea Formaldehyde (MUF) resin was synthesized in the laboratory and Jatropha seed oil meal (JM) with Rubber seed oil meal (RM) were added to the Melamine Urea Formaldehyde (MUF) resin in adhesive mix formulations for interior plywood manufacturing using a laboratory press. These formulations were compared with commercialize industry wheat flour (IF). Response Surface Methodology (RSM) was used for identification of the optimum temperature and pressing time for wood adhesive performance. The experiments were conducted with temperature range from 100 to 150°C and pressing time from 50 to 250 sec. The result indicates that the effect of the extender type on plywood bonding strength was significant. The extender with high protein content has greater bonding strength performance.

Key words: Jatropha seed oil cake, rubber seed oil cake, melamine urea formaldehyde, RSM, extender

INTRODUCTION

The continuing increasing global demand for energy, the impending depletion of fossil fuels and concern over global climate change has led to resurgence in the development of alternative energy sources. *Jatropha curcas* L. and *Hevea brasiliensis* have acquired significant economic importance for its seed oil which can be converted to biodiesel is emerging as an alternative to petro-diesel. Besides this, the by-product of jatropha and rubber mill such as JM and RM is also increased directly. Moreover, Malaysia remain as top ten wood suppliers of wood-based products in the world, especially to export in Europe, Japan, Taiwan, Singapore and Middle East. Total exports of wood-based products from Malaysia was RM 20.5 billion in 2010 (http://www.mtib.gov.my) and this industry based exports in Malaysia rises nearly 40% compared to the past ten years. However, the production cost of the commercial adhesive for wood composite is increasing day by day because most of them are petrochemical based. In wood industry, the common uses synthetic resin is thermosetting and thermoplastic. The examples of thermosetting resin are MUF, Urea Formaldehyde (UF), Melamine Formaldehyde (MF), Phenol Formaldehyde (PF) and etc. Whereas, Polyvinyl acetate (PVAc), Polyvinyl alcohol (PVA) and Ethylene Vinyl Acetate (EVA) are the common uses thermoplastic resin in wood industry (Sellers, 1998). Resin and extender is the main material for an adhesive for plywood production. Adhesives often compounded with extender to increase viscosity and reduce production cost. Wood surface consists of small holes and the extender in the adhesive is used to fill up all the small holes at the surface of wood for strengthening the boding interactions among the components. Apart from this, extender also used to reduce the penetration of resin into the small holes of the wood (Pizzi, 1994). There are several types of extender which are used by the industries, such as, wheat flour, plastic, corn starch flour, tapioca flour and others. One of the Malaysian largest wood products producers, Shin Yang Chemical Sdn, Bhd, uses wheat flour (industrial flour) as the extender. The protein content of wheat flour is around 8% (Perry, 1944). Recently, our laboratory has been evaluated oil palm by-products, Palm Kernel Meal (PKM) as alternative extender in MUF based plywood adhesive. The result shows that, the plywood performance PKM as an extender is comparable with the industry's standard extenders. The significant high

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protein content inside PKM enhances the plywood performance (Ong *et al*., 2011). The goal of the present study was to investigate suitability of JM and RM in enhancing the plywood performance.

MATERIALS AND METHODS

Jatropha seed oil meal (JM) with Rubber seed oil meal (RM) preparation: Jatropha seed (*Jatropha curcas* L.) and Rubber seed (*Hevea brasiliensis*) was purchase from the local supplier. After being washed and dried at 60°C in oven for 16 h, Jatropha seed and Rubber seed was undergone oil extraction process to obtain oil free JM and RM. The JM and RM was pulverized in order obtain a desired size of 50 μm fine powder.

Industrial flour preparation: The industrial flour was provided by Shin Yang Chemical Sdn. Bhd. The raw industrial flour was to sieve by using 50 μm sieve to obtain uniform fine powder and stored.

Melamine-urea-formaldehyde (MUF) resin preparation: The method of MUF resin preparation was adopted from Bono *et al*. (2003, 2006, 2007, 2008). The main materials were used to produce MUF resin which contained formaldehyde, melamine and urea. In brief, 100 mL of 37% formalin is poured in a 500 mL three-necked flask and required amount of melamine and urea is added under vigorous stirring. The pH of the mixture was adjusted to 8.5-9.0. The temperature of the mixture was maintained at 80°C for 2 h so, that resin formed and resin mixture was cooled down to 60°C, excess amount of urea is added to the resin solution. The stirring was continued until the temperature was reach to the room temperature and finally transferred to plastic container for further testing.

Wood adhesive production: For the adhesive (glue) preparation, MUF resin was weighted and transfer into a beaker, of urea was added into the MUF resin and mixed well with a mixer (KHIND Model SM 210) for 5 min. The extender (IF, JM, RM) was added to the above mixture and mixed well for 5 min. The hardener (ammonium chloride) was added into the mixture and mixed for 5 min (the amount of the hardener is subjected to change).

Type II plywood production: The production of type II plywood was conducted using red-meranti 300×300× 3.3 mm veneer. In order to get consistence result veneer was maintain at 10% moisture content and the equal amount of wood adhesive were used at every plywood produced. The adhesive was applied onto two sides of a core veneer using a glue spreader. The unfinished plywood was left at room temperature for 5 min., before it was cold pressed 9 kg cm^{-2} for 20 min.

Table 1: Constraints for hot press

Parameter	Lower limit	Higher limit
Temperature (°C)	100	150
Pressing time (sec)	50	250
Types of extender	JM, RM, IF	

JM: Jatropha seed oil meal, RM: Rubber seed oil meal, IF: Industry wheat flour

The unfinished plywood was removed after 20 min from the cold press device and left free 5 min before it was transferred to hot press device. During hot pressed, pressure at 9 kg cm^{-2} and temperatures of 100-150°C were applied. The process was conducted for 50-250 seconds. After the hot press completed, the plywood product was released from the device and stored at room environment for further testing.

Wood performance test (shear strength test): The shear strength of type II plywood was determined by bonding test according to the Japanese Agriculture Standard (JAS) for structural plywood (Japanese Agricultural Standard for Plywood, 2003). Total of nine plywood test pieces (25×80 mm) were tested for every trial plywood panel produced. Prior to the test, the test pieces were soaked in a hot water bath at 60°C for 3 h and followed by soaking it at cold water bath at room temperature. Once the test pieces reached cold state, then it was used for shear strength test. Test was conducted using universal testing machine while the plywood test pieces were wet.

Experimental design: The experimental design was conducted using Response Surface Methodology (RSM), Design Expert software (version 6.10, Stat Easy Inc., Minneapolis, USA). For the hot press plywood production, the effect of three independent variables of temperature, pressing time and types of extender were investigated. The constraint of component proportion is shown in Table 1.

RESULTS AND DISCUSSION

The experimental result obtained for temperature and pressing time of RM, JM and IF as extender used the proposed experimental design is given in Table 2.

The studies of the effects of different type of extender for MUF resin towards shear strength performances were conducted at various parameters as well as temperature and pressing time. In addition, the experiments were carried out in find out the optimum and maximum shear strength of MUF resin in the particular range of temperature and pressing time.

Table 2: Average shear strength performance of various types of extenders

No. of run	Temperature, x_1 (°C)	Pressing time, x_2 (sec)	Average shear strength of 9 specimens of plywood, Y_1 (MPa) IF, x_3	JM, x_3	RM, x_3
1	100	50	0.90	1.03	1.04
2	125	250	1.11	1.22	1.22
3	150	50	1.08	1.13	1.12
4	100	150	0.96	1.25	1.25
5	125	150	1.36	1.47	1.45
6	125	50	1.03	1.09	1.08
7	100	250	0.93	1.12	1.12
8	125	150	1.37	1.49	1.40
9	150	250	1.06	1.10	1.11
10	150	150	1.10	1.22	1.27
11	125	150	1.33	1.43	1.48
12	125	150	1.30	1.37	1.38
13	125	150	1.28	1.39	1.50

Initial range of temperature between 100 to 150°C and pressing time between 50 to 250 sec were selected for the experiment. However, the experimentation at the lower ranges of temperature and pressing time were impossible to evaluate the results. This is due to the veneers failed stick with core face. Therefore, the shear strength test could not be conducted. This phenomena occur due to the failed of boding formation to hold the veneers and core face together.

In addition, the experiments for the temperature above 150°C and pressing time 250 sec was also found to be failed. This due to the plywood failed to be cut into pieces for bonding test. Therefore, the bonding test couldn't be conducted. However, this phenomenon occurs due to the large air bubbles occur between the veneers and core face. Besides that, the wood cracks were found at the surface of wood during the hot press. As concluded that, the suitable range of temperature 100 to 150°C and the pressing time is 50 to 250 sec for the experimental study.

Development of regression model equation: A polynomial regression equation was developed by using Three level factorial design to analyze the factor interactions by identifying the significant factors contributing to the regression model. The complete design matrix together with the response values obtained from the experimental works are given in Table 2. The shear strength of extender was found from 0.90 to 1.50 MPa.

According to the sequential model sum of squares, the models were selected based on the highest order polynomials where the additional terms were significant and the models were not aliased. For shear strength performance of plywood, quadratic models was suggested by the software and selected due to higher order polynomial. The final empirical models in term of coded factors for shear strength performance (Y_1) is shown in Eq. 1:

Table 3: Analysis of variance (ANOVA) for response surface quadratic model for shear strength of plywood

Source	Sum of squares	df	Mean square	F-value	Prob>F	Comment
Model	0.98	11	0.089	22.17	<0.0001	Significant
x_1	0.019	1	0.019	4.82	0.0370	
x_2	0.013	1	0.013	3.32	0.0794	
x_3	0.12	2	0.062	15.50	<0.0001	
x_1^2	0.18	1	0.18	44.57	<0.0001	
x_2^2	0.32	1	0.32	80.05	<0.0001	
x_1, x_2	5.633E-03	1	5.633E-03	1.40	0.2465	
x_1, x_3	0.016	2	8.089E-03	2.01	0.1529	
x_2, x_3	1.378E-03	2	6.889E-04	0.17	0.8432	
Residual	0.11	27	4.015E-03			
Lack of fit	0.082	15	5.443E-03	2.44	0.0630	Not significant
Pure error	0.027	12	2.230E-03			

$$Y_1 = 1.38 + 0.033x_1 + 0.027x_2 - 0.080x_3[1] + 0.036x_3[2] - 0.15x_1^2 - 0.20x_2^2 - 0.022x_1x_2 + 0.042x_1x_3[1] - 0.024x_1x_3[2] - 0.012x_2x_3[1] + 0.004444x_2x_3[2] \quad (1)$$

Positive sign in front of the terms indicates synergistic effect, whereas negative sign indicates antagonistic effect. The quality of the model developed was evaluated based on the correlation coefficient value. The R^2 value for the equation was 0.9003. This indicated that 90.03% of the total variation in the shear strength performance of the plywood. The closer the R^2 value to unity, the better the model will give predicted values which are closer to the actual values for the response. the R^2 of 0.9003 for Eq. 1 was considered relatively high, indicating that there was good agreement between the experimental and the predicted shear strength performance of plywood from this model.

Statistical analysis: The result of the surface quadratic model in the form of analysis of variance (ANOVA) are given in Table 3 for shear strength of plywood. ANOVA is required to justify the significance and adequacy of the models. The mean squares were obtained by dividing the sum of the squares of each of the variation sources, the model and the error variance, by the respective degrees of freedom. If the value of Prob>F less than 0.05, the model term are considered as significant. From the Table 3, the model F-value is 22.17 and Prob<0.0001 implied that this model was significant. In this case, x_1, x_3, x_1^2 and x_2^2 factors was significant model term whereas x_2, x_1, x_2, x_1, x_3 and x_2, x_3 were insignificant to the response. From the statistical results obtained, it was shown that the above models were adequate to predict the shear strength performance within the range of variables studied. Figure 1 shows the predicted values versus the experimental values for shear strength performance. As can be seen, the predicted values obtained were quite close to the experimental values, indicating that the models developed were successful in capturing the correlation between operating parameter to the response.

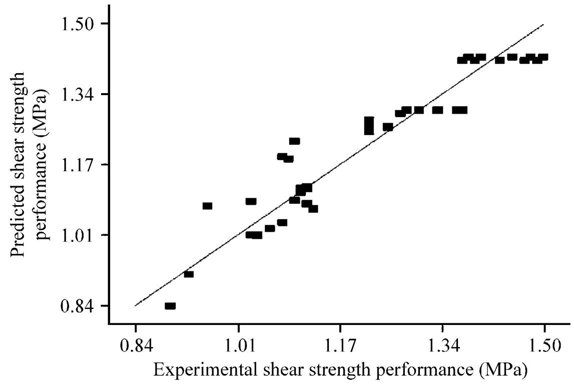

Fig. 1: Predicted vs. experimental shear strength performance (MPa)

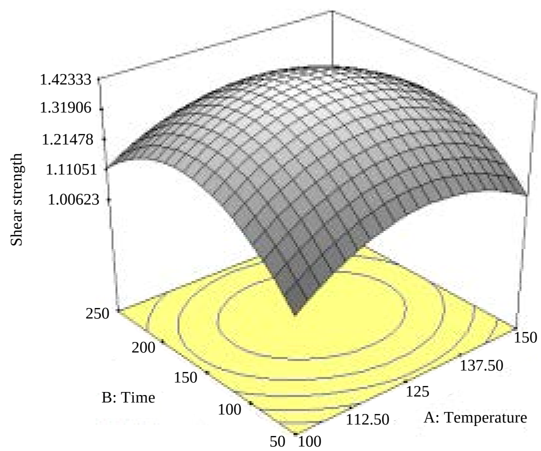

Fig. 2: Design expert plot for RM as extender

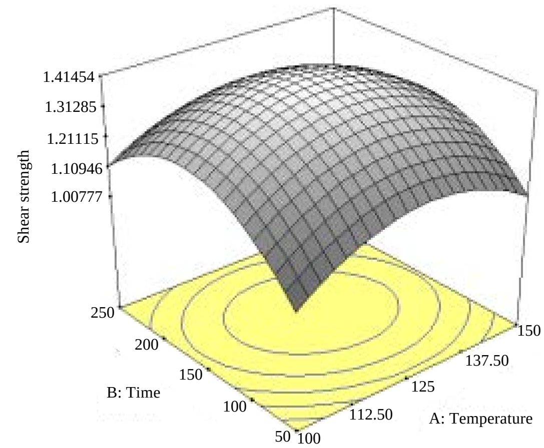

Fig. 3: Design expert plot for JM as extender

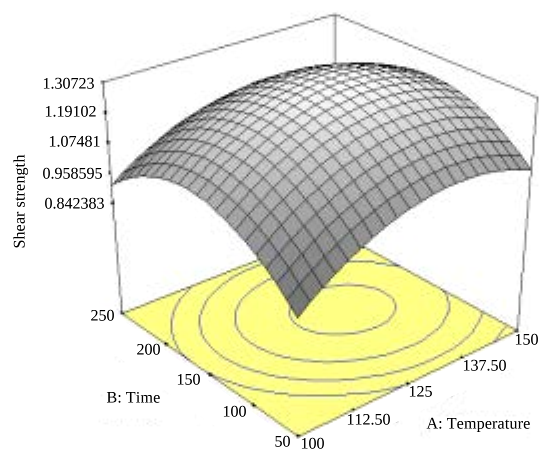

Fig. 4: Design expert plot for IF as extender

Shear strength performance: Referring to Table 3, different types of extenders showed the largest F-value 15.50 among the factors, indicating that this variable imposed the significant effect on the shear strength performance of plywood. The effect of temperature was significant as well. However, the effect of pressing time on the response was relatively insignificant. The interaction effects between the variables were insignificant. Figure 2-4 shows the three dimensional response surface which was constructed to show the effects of the pressing temperature, time and different types of extenders on the plywood shear strength performance. The effects of pressing temperature and different types of extenders were studied as they were found have significant effects on the response. As can be seen from Fig. 2-4, shear strength performance increases with increase in pressing time and temperature. However, shear strength performance decrease with increase in pressing time and temperature after the response met the highest value. The response surface in Fig. 2-4 shows curvature. The highest shear strength performance value was obtained when both the variables (pressing time and temperature) were at the middle within the range studied. Besides that, RM has given highest shear strength performance value among the extenders. However, in this work, all the variables studied were found to have synergistic on the shear strength performance in plywood application. This was expected as the progressive temperature rise and longer pressing time would increase the bonding strength of the plywood until reach maximum and then it would decrease as the pressing temperature and time continuing rise. Besides that, effect of different types of extenders was expected as higher protein content extender provided better response. As shown by the analysis, RM and JM shown greater result than IF due to protein content of RM and JM are higher than IF. The

Table 4: Model validation

Pressing temperature, x_1 (°C)	Pressing time, x_2 (sec)	Different types of extenders, x_3	Shear strength performance (MPa) Predicted	Experimental	Error (%)
126	159	RM	1.42	1.38	2.82
125	158	JM	1.41	1.36	3.55
131	152	IF	1.31	1.27	3.15

higher amount of protein will increase the shear strength of the wood adhesive. This due to the amino group $-NH_2$ inside the extenders enhanced the bonding formation between the wood adhesive and surface of veneer (Ong *et al*., 2011). This result also agreement with the work done by Ong *et al.* (2011). The protein content of RM, JM and IF are 28, 24 and 8%, respectively (Perry, 1944; Babatunde *et al*., 1990; Lestari *et al*., 2011). Therefore, RM and JM shear strength value very close this due insignificant different of protein content.

Process optimization: Three level factorial has been used to optimize the parameters affecting the shear strength performance response. In this optimization analysis, the target criteria was set as maximum values while the values for variables were set in the ranges being studied. The predicted and experimental results of shear strength obtained at optimum conditions are shown in Table 4. The optimum shear strength performance of RM was obtained by using pressing temperature 126°C and time 159 sec was 1.38 MPa. The optimum shear strength performance of JM was obtained by using pressing temperature 125°C and time 158 sec was 1.36 MPa. The optimum shear strength performance of IF was obtained by using pressing temperature 131°C and time 152 sec was 1.27 MPa. It was observed that the experimental values obtained were in good agreement with the value calculated from the models, with relatively small errors which only 2.82, 3.55 and 3.15%, respectively for RM, JM and IF.

CONCLUSION

The study on the effect of plywood preparation variables on the shear strength performance of plywood have been conducted by using three level factorial design. Through analysis of the response surface, pressing temperature and different types of extenders imposed the greater effect on the shear strength performance. The optimum shear strength performance of RM was obtained by using pressing temperature 126°C and time 159 sec was 1.38 MPa. The optimum shear strength performance of JM was obtained by using pressing temperature 125°C and time 158 sec was 1.36 MPa. The optimum shear strength performance of IF was obtained by using pressing temperature 131°C and time 152 sec was 1.27 MPa. It was observed that the experimental values obtained were in good agreement with the value calculated from the models, with relatively small errors. Besides that, this work presented that extender is important in enhancing the wood performance for wood adhesive. The higher amount of protein will increase the shear strength of the wood adhesive. This due to the amino group $-NH_2$ inside the extender enhanced the bonding formation between the wood adhesive and surface of veneer, core face and plywood (Ong *et al*., 2011).

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