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The Performance of Melamine Urea Formaldehyde (MUF) Based Particleboard with Wheat Flour as Filler

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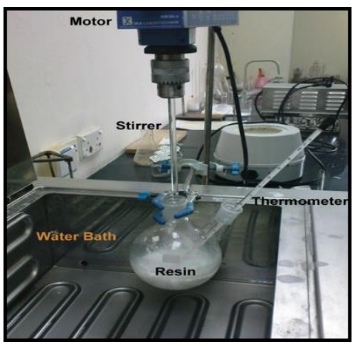
Graphical abstract



Abstract

In this study, melamine urea formaldehyde (MUF) resin was used as wood adhesive. The MUF was synthesized in three stages. The MUF resin based particleboard was produced using wheat flour as filler. The parameters that have been used to evaluate the performance of MUF resin are: water absorption (WA), thickness swelling (TS), modulus of rupture (MOR) and modulus of elasticity (MOE). The data limits designed was analyzed by using response surface methodology (RSM). The models were developed for four response variables, i.e. WA, TS, MOR, and MOE. The range of temperature, pressing time and wheat flour filler content were 110–150ºC, 80 to 250 sec and 10-20% (w/w) respectively. From the analysis of variance (ANOVA), the optimal conditions were established at 149.8ºC of temperature, 250.0 sec of pressing time, and 10.0% (w/w) of wheat flour filler.

Keywords: Melamine urea formaldehyde; water absorption; thickness swelling; modulus of rupture; modulus of elasticity

Abstrak

Dalam kajian ini, Melamin Urea Formaldehyde (MUF) telah digunakan sebagai perekat kayu. Penghasilan resin MUF adalah berdasarkan tiga peringkat. Selepas itu, resin MUF akan diuji melalui penghasilan papan partikel dengan penambahan tepung gandum. Parameter yang telah digunakan untuk menilai prestasi resin adalah: penyerapan air (WA), ketebalan (TS), modulus keretakan (MOR) dan modulus kekenyalan (MOE). Had data direka dianalisis dengan menggunakan tindak balas metodologi permukaan (RSM). Model telah dibangunkan selama empat pemboleh ubah tindak balas, iaitu WA, TS, MOR, dan MOE. Pemboleh ubah yang digunakan adalah suhu tekanan panas untuk panel dalam julat 110ºC–150ºC, masa tekanan panas dalam julat 80 hingga 250 saat dilakukan dan penggunaan kuantiti tepung dalam panel adalah dalam julat 10-20% (w/w). Daripada analisis varians (ANOVA), keadaan paling optimum yang dicapai adalah pada suhu 149.8ºC, 250.0 saat bagi masa untuk tekanan panas, dan 10.0% (w/w) kandungan tepung gandum.

Kata kunci: Melamin urea formaldehyde; penyerapan air; ketebalan; modulus keretakan; modulus kekenyalan

1.0 INTRODUCTION

Adhesives systems that are used for the production of wood panel products are heterogeneous mixture. This type of mixture is consisting primarily of resin with extenders, fillers and catalysts. The adhesive mixture or blending of a resin with other ingredients frequently results in reducing overall glue costs.

Starch-based and protein-based adhesives were the early wood adhesives for bonding wood products [1]. Nowadays, the formaldehyde based resin is commonly used such as urea formaldehyde (UF), melamine formaldehyde (MF), phenol formaldehyde (PF) and melamine urea formaldehyde (MUF). UF resin is used as binder or adhesive in particleboard and medium density fibreboard for composition panels. Meanwhile MF is used in the building and construction industries for the laminates and surface coatings. PF also is used in construction industry and building for insulation binder, wood production and laminates. Beside, MUF resin is widely used in wood industries, coating technology, paper industries and kitchenware production.

Adhesives are mainly used in the processing of wood products especially engineered woods such as particleboard, woods panels, fiber board and plywood. Since adhesives are used in many different applications with wood, a wide variety of types are used [2]. Basically, wood adhesives can be classified into natural adhesives and synthetic adhesives. However, natural adhesives further can be classified into plant based adhesives and animal based adhesives. These types of adhesives are synthesized from natural sources such as animal protein, blood protein or even soy-bean protein. Besides that, synthetic adhesives consist of two types: thermoplastic resins and thermosetting resins. All these types of wood adhesive are utilized based on the suitability with the requirement of wood products. However, adhesives systems that are used for the production of wood panel products such as veneer, particleboard and plywood usually are heterogeneous mixture. This type of mixture consists of primarily of resin with extenders, fillers and catalysts. The adhesive mixture or blending of a resin with other ingredients frequently results in reducing overall cost. Moreover, it was reported that the benefit of using extenders and fillers is to manipulate the hygroscopity of the adhesive mixture [3]. Due to the reduction of solid wood supply, the world demand for wood products is growing and this trend is expected to continue in the years to come [4, 5]. In that case, quality of wood product should be sustained so that it can compete with the world economic trend of wood based industry. In order to produce a good quality of wood product, wood adhesive become the major thing that need to be put into consideration.

Particleboard industries are still relying on UF resin as binding agents. The utilization of UF resin is most preferable due to low cost and desired particleboard properties [6]. There are efforts done by researchers in lowering the amount of formaldehyde in UF, however this brought another problem where it has severely impaired the already vulnerable properties of the boards, in particular their water resistance [7]. In order to improve the moisture resistance, fortification of UF resins with melamine was investigated [8, 9]. The content of melamine in MUF resin requires as low as possible as MUF resin is expensive. However, reduction of melamine in MUF creates another problem where the MUF exterior grade performance is noticeably worse. However when the MUF resin content is low, there will be a limitation in the increment of resin consumption. This condition then results in the higher production cost of wood product. Thus, filler is desirable so as to increase the solid content in MUF resin. Filler helps to enhance resins performance by filling the void or gap within the board surface as well as avoids weak bond. Wheat flour is one of the natural sources that can be used as filler for the MUF resin. Wheat flour possesses high content of protein which enhances the bonding formation between the wood products.

Therefore the aim of this work was to produce particleboard from wood particles by using wheat flour as filler with MUF resin and to investigate the performance of the wood particleboard. The performance test includes the studying of water absorption (WA), thickness swelling (TS), modulus of rupture (MOR) and modulus of elasticity (MOE).The MUF based particleboard process with wheat flour as filler was optimized by using response surface methodology (RSM) [10].

■2.0 MATERIAL AND METHOD

2.1 MUF Resin Preparation

The MUF resin was prepared using analytical grade melamine, industrial grade urea and 37% (w/w) formaldehyde as the raw materials. The method of synthesis of MUF resin was adopted from reflux process [11,12,13]. MUF resin was prepared by three stages. In the first stage, formalin was placed into the three neck flask as shows in Figure 1. Then, melamine was poured into the flask followed by first urea. Urea was poured later in order to avoid a faster polymerization process. After that, the mixture was blended homogenously by using a stirrer which is connected to a motor as shown in Figure 1. In this stage, the mixture forms a white-colored solution. The water bath temperature was set at 90^{o}C and pH of the solution was checked. The mixture was acidic, thus it was required to adjust the pH of solution by adding few drops of 10 % of caustic soda (NaOH). The pH range was in 8.8 to 9.0 or in alkaline range. Initial temperature and initial pH of solution was then recorded. Then, temperature and pH of the solution was recorded for every 5 minutes until the end of production process.

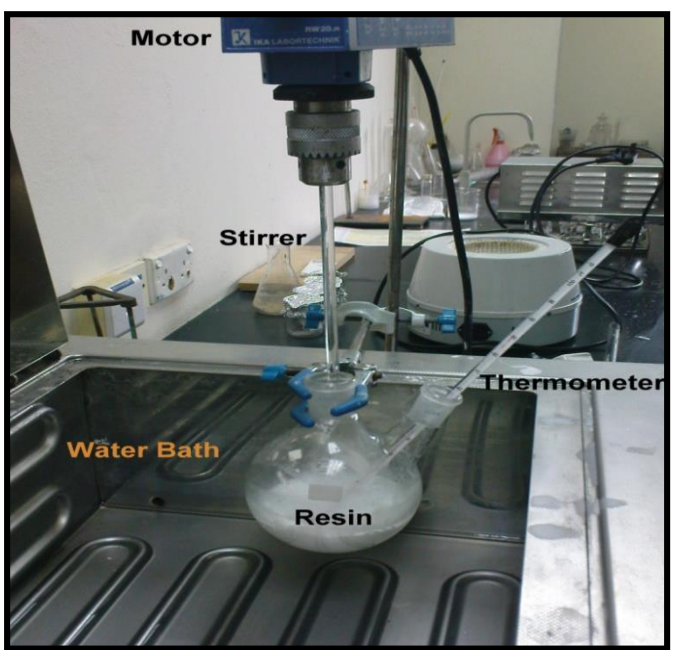


Figure 1 Equipment setup for MUF resin production

The heating process was continued until it reaches 80oC. The white solution was turned to a clear solution at this stage. Meanwhile, end point test can be conducted at this stage. The end point test was determined by dropping the mixture of solution into a beaker of water at 30oC temperature for every 5 minutes to 10 minutes. When the end point was reached, the heating process was stopped while stirring. Caustic soda was added slowly in order to increase the pH in the range of 8.8 to 9.5. This was done to stop the polymerization of the resin. The solution was then cooled to ambient temperature by immersing the flask into water bath. Second urea was added when the temperature dropped to 60oC. The cooled resin was transferred to a plastic bottle or container for further testing and particleboard production.

2.2 Filler Preparation

Wheat flour was used as filler in this study. It was purchased from 1Borneo, Kota Kinabalu, Sabah, Malaysia. The flour was dried in order to use as filler for the particleboard production for a period of 6 hours in an oven at 60oC.

2.3 Production of Particleboard with Wheat Flour as Filler and Characterization

Wood particles provided by School of Forestry and Tropical Science of University Malaysia Sabah (UMS) were used for producing a particleboard. The wood consists of Acacia mangium. Particles were dried in an oven for a period of 12 hours before used and the moitutre content was found to be <5%.

The target density of particleboard was 600 kg/m3 and the final target board equivalent moisture content (EMC) was 12%. The 12% EMC is actually the total moisture of the finished particleboard that consists of moisture of Acacia mangium particles and moisture of the resin. Based on the calculations in the preparation of particleboard, required values such as particle oven dry weight, raw particles weight, weight of resin, actual weight of resin and amount of distilled water were obtained. Then, the resin was mixed with wood particles by using rotating drum-type blender. After mixing, wood particles were molded. Then, particles were hot-pressed in the hot-press machine for 80 to 250 sec with the temperature between 110°C to 150°C. Once hot pressed was done, the board was cooled down. Cooling was done by cold press machine. After that, board was cut into pieces for the testing purpose. Test samples were prepared and conditioned. The performance test for particleboard was conducted.

WA and TS tests were carried out based on method B of ASTM D1037-93 which is 24 hours soaking test. The 50mm x 50mm piece of particleboard was soaked in water at room temperature for 24 hours. Initial thickness and weight of particleboard before being soaked were measured. After soaking, immediately, the thickness and weight of particleboard were measured. The thickness was measured by using vernier caliper. This was done to calculate the thickness swelling and water absorption for the board respectively. MOR and MOE were estimated according to the Japanese Industrial Standard (JIS 5908-1994) by using GOTECH testing machine (Model AI-7000 L10). The crosshead speed was set at 10 mm/min. The test specimen dimension was 150 x 50 mm.

2.4 Experimental Design and Optimization

The experimental settings were designed by using RSM. The experimental design was conducted by using Design Expert Software (Version 7.0.0, Stat Easy Inc, and Minneapolis, USA). In this study, the parameters were involved are the temperature, pressing time and wheat flour filler content. The responses of the parameters were the performance of particleboard in terms of WA, TS, MOR and MOE. Constraints of experimental design were presented in Table 1.

Table 1 Constraint for experimental design

Component/Parameter	Unit	Low Limit	High Limit
Temperature	°C	110	150
Pressing time	Seconds	80	250
Wheat flour content	% (w/w)	10	20

The effect of wheat flour as filler for MUF resin for bonding strength in terms of MOR and MOE was studied at various temperature, pressing time and wheat flour filler content. In addition, the experiments were carried out to find out the optimum condition of WA, TS, MOR and MOE in the particular range of temperature, pressing time and wheat filler content. Initially, a single particleboard was produced by using only MUF resin alone with the temperature of 150oC and pressing time 300 sec and adopted as a control element in order to estimate the difference in the performance of MUF resin in the presence of wheat flour and without wheat flour filler. The values of WA, TS, MOR and MOE were found to be 32.60%, 4.71%, 2.091 N/mm2, and 499.820 N/mm2 respectively.

The range of temperature was 110oC and 150oC and pressing time range in between 80 to 250 sec. The MUF resin content was fixed at 8% and the amount of wheat flour used in the range of 10% to 20% particleboard solid content. The experiment was conducted until 150oC only as the maximum temperature for a particleboard pressing temperature is 160oC.

Higher temperature with longer pressing time can cause the bond quality deteriorated as the melamine content in the resin decreases. The range of wheat flour filler content was in between 10-20%, the pressing time range was in 80 to 250 sec and the range of pressing temperature was 110oC – 150oC. The responses that need to be analyzed in this experiment are WA, TS, MOR and MOE.

■3.0 RESULTS AND DISCUSSIONS

The experimental results of this study are summarized in Table 2. This Table shows the data obtained from experiments conducted for various percentages of wheat flour filler, different temperature of pressing and different values of pressing time. This data was then evaluated by using D-Optimal Design Expert software (version 7.0.0, Stat Easy Inc., Minneapolis, USA) [14,15].

In order to analyze the experimental data in Table 2, the individual graph of WA, TS, MOR and MOE are plotted for various values of temperature, wheat flour filler amount and curing period. Each of the result was then analyzed on three different percentages of wheat flour filler content that is 10%, 15% and 20%. Based on the analysis of variance (ANOVA) from the Design Expert Software, all the equations were derived for each of the response based on the data as shown in Table 2. The model of 2F1 was found to fit the analysis of WA and TS response. On the other hand, MOR and MOE responses were based on the quadratic analysis.

Table 2 Experimental vs predicted results of particleboard testing

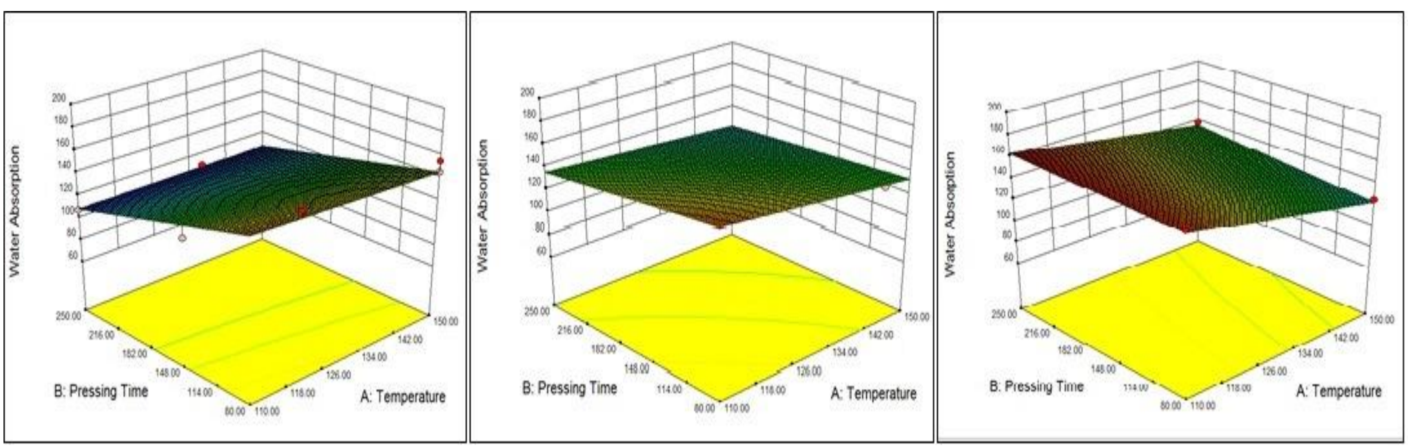
Run	Experiment parameter			Responses							
				Experimental				Predicted			
	Temperature (°C)	Pressing time (sec)	Wheat flour content, % (w/w)	WA (%)	TS (%)	MOR (N/mm^2)	MOE (N/mm^2)	WA (%)	TS (%)	MOR (N/mm^2)	MOE (N/mm^2)
1	136.01	250.00	10.00	109.58	8.40	2.41	468.07	108.47	7.13	2.30	475.33
2	110.00	80.00	20.00	161.86	21.30	0.98	358.08	160.33	23.19	0.99	352.87
3	136.01	250.00	10.00	109.58	8.40	2.41	468.07	108.47	7.13	2.30	475.33
4	120.00	80.00	10.00	159.72	23.40	1.09	88.10	153.54	21.29	1.24	132.33
5	126.07	80.00	14.01	164.56	25.40	1.10	89.05	149.25	21.26	1.10	65.51
6	110.00	250.00	10.00	107.82	9.10	1.53	438.36	108.57	13.34	1.75	447.54
7	126.15	181.51	20.00	122.42	26.70	1.13	412.23	149.35	24.77	1.18	454.33
8	150.00	80.00	20.00	124.64	19.30	1.03	90.05	122.68	16.41	1.00	88.81
9	110.00	250.00	16.44	150.84	30.00	1.64	458.18	143.77	27.67	1.52	428.58
10	150.00	80.00	10.00	155.39	22.50	1.02	91.05	145.71	23.73	1.01	106.12
11	150.00	80.00	20.00	124.64	19.30	1.03	90.05	122.68	16.41	1.00	88.81
12	150.00	250.00	20.00	139.00	14.90	2.43	520.07	135.89	16.00	2.40	508.17
13	110.00	140.89	10.00	128.00	14.20	1.25	422.09	139.11	17.92	0.98	417.08
14	150.00	180.82	14.08	125.73	11.20	1.13	403.05	126.42	13.64	1.32	431.91
15	147.50	91.27	15.00	126.45	10.40	1.04	103.05	134.86	19.56	0.93	85.72
16	150.00	80.00	10.00	145.39	22.50	1.02	91.05	145.71	23.73	1.01	106.12
17	110.00	80.00	20.00	161.86	21.30	0.98	358.08	160.33	23.19	0.99	352.87
18	125.29	185.50	13.75	162.32	20.20	1.24	339.51	134.49	18.38	1.21	378.36
19	150.00	250.00	20.00	139.00	14.90	2.43	520.07	135.89	16.00	2.40	508.17
20	135.00	141.18	10.00	107.78	24.40	1.14	443.07	134.82	17.06	1.16	335.82

3.1 Effect of Wheat Flour Content on WA

Twenty experiments were conducted with four replicates. From the ANOVA analyses, the experimental results can be fitted into a quadratic model, as shown in the equation 1 in terms of actual factors temperature (A), pressing time (B) and amount of wheat flour content(C):

$$WA,\% = 165.3851 + 0.2977A - 0.7435B + 5.5214C + 1.5145 \times 10^{-3}AB - 0.0680AC + 0.0297BC \quad (1)$$

Figure 2 (a, b and c) shows the results of WA at 10%, 15% and 20% of wheat flour filler content. From the Figure 2 (a, b and c), it can be concluded that WA increasing significantly with the increased of wheat flour content.



(a) (b) (c)

Figure 2 WA at 10%, 15% and 20% of wheat flour content

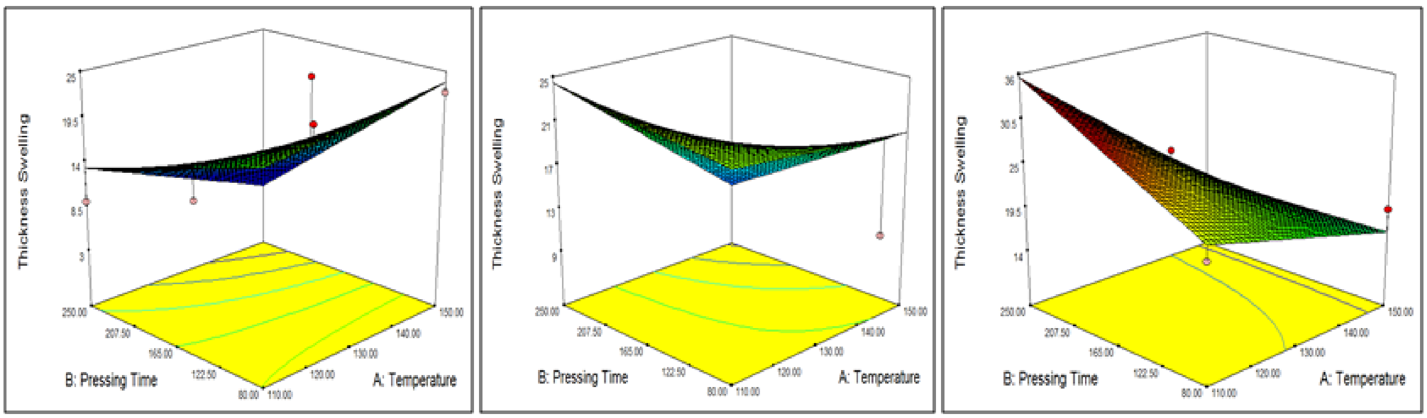
MUF resin and wheat flour contributes to hydrophobic condition in melamine in which result in reduced water absorption. Wheat flour exhibits higher protein content that consist of hydroxyl group which can enhance bonding strength between adhesive and wood. However, over addition of wheat flour in the resin may worsen the water resistance of resin as adhesive cannot be easily applied to the substrate. Thus, from the Figure 2 (a, b and c), it can be concluded that 20% wheat flour filler content promotes higher WA compared to panels made with 15% and 10% wheat flour content. Based on Figure 2 (a, b and c) it is can also be concluded that the WA decreases with the increases of temperature. The significant difference of WA can be seen between 110ºC and 150ºC. The higher the temperature, the lower the WA performed by the particleboard. Higher temperature generates more heat transfer surface area of substrates in the panel thus more bonding can built up which results in better water resistance. Besides that, the hot pressing time of a particleboard also affects the mechanical and physical properties of it, this is due to the insufficient or over curing of the resin resulted in indecent internal bonding between the particles and binder [16].

3.2 Effect of Wheat Flour Content on TS

The results of Table 2 can be fitted into a quadratic model, as shown in the equation 2 in terms of actual factor temperature (A), pressing time (B) and amount of wheat flour content (C):

$$TS,\% = -22.7786 + 0.4827A + 0.05022B + 2.1109C - 1.8829\times10^{-3}AB - 0.02508AC + 0.01149BC \quad (2)$$

Figure 3 (a, b and c) shows the results of TS at 10%, 15% and 20% of wheat flour filler content. Figure 3 (a, b and c) shows that TS of particleboard is increasing with the increase of wheat flour filler content from 10% to 20%. Figure 3 (a, b and c) also exhibits that TS is decreasing when higher temperature is applied during production of particle board. However, over addition of the wheat flour as filler brought the drawback of lack resistance towards water. This is because of wheat flour which contains water-soluble carbohydrates would reduce the water resistance of adhesive bonds, thus increases the TS of the particleboard. However, the hydroxyl groups that exist in wheat flour also another reason for the reduction in water resistance of a particleboard.



(a) (b) (c)

Figure 3 TS at 10%, 15% and 20% of wheat flour content

From Figure 3 (a, b and c), it is obvious that temperature of 110ºC shows higher TS compared with 150ºC. Similarly with WA result, higher pressing temperature promotes higher heat transfer surface areas in the panel which results in more bonding built up and hence gives better water resistance. Other than that, longer and shorter pressing time of a particleboard would affect the mechanical and physical properties, since insufficient or over curing of the resin which resulted in change in internal bonding between the particles and binder [17].

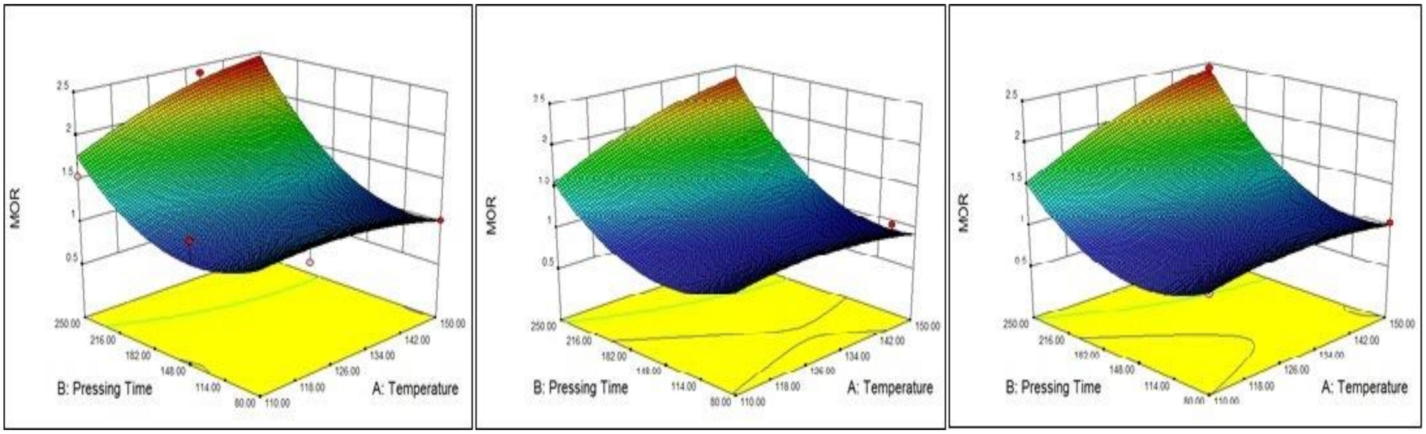
3.3 Effect of Wheat Flour Content on Static Bending Test (MOR and MOE)

The dependence of temperature (A), pressing time (B) and amount of wheat flour content(C) on the modulus of rupture (MOR) and modulus of elasticity (MOE) was correlated from the ANOVA analysis as shown in the equations 3 and 4:

$$MOR\ (N/mm^2) = 0.7580 + 0.0521A - 0.0321B - -0.1779C + 1.2926\times 10^{-4}AB + 5.2802\times 10^{-4}AC - 1.2439\times 10^{-5}BC - 2.7947\times 10^{-4}A^2 + 6.4224\times 10^{-5}B^2 + 3.2948\times 10^{-3}C^2 \quad (3)$$

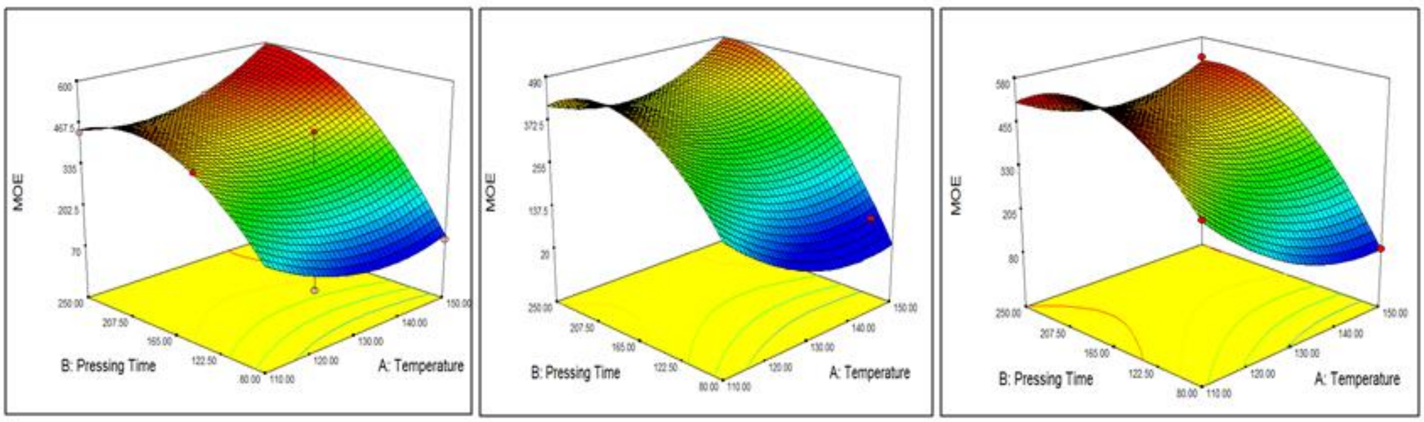
$$MOE\ (N/mm^2) = 3461.9763 - 51.2590A + 3.4162B - 20.6218C + 0.038167AB - 0.3833AC - 0.04167BC + 0.1895A^2 - 0.0177B^2 + 2.6573C^2 \quad (4)$$

Figure 4 (a, b and C) and Figure 5 (a, b and C) show the results of modulus of MOR and MOE at 10%, 15% and 20% of wheat flour filler content. From these figures, it can be concluded that MOR and MOE increase significantly with longer curing period and increase in temperature. This indicates that better polymer curing at higher temperature. However, MOR and MOE decrease with shorter pressing time and increase in temperature. MUF resin viscosity increases with wheat flour filler content. Derkyi *et al*. [17] found that increasing the solid content results in the increase of resin viscosity. This physical property may help in enhancing the bonding strength between the substrate. From the interaction Figure 4 (a, b and C) and Figure 5 (a, b and C), 20% wheat flour content performed better compared to 10% and 15% of wheat flour content as filler. It is obvious that curing period affects the physical and mechanical properties of a particle board due to insufficient or over curing of the resin [13].



(a) (b) (c)

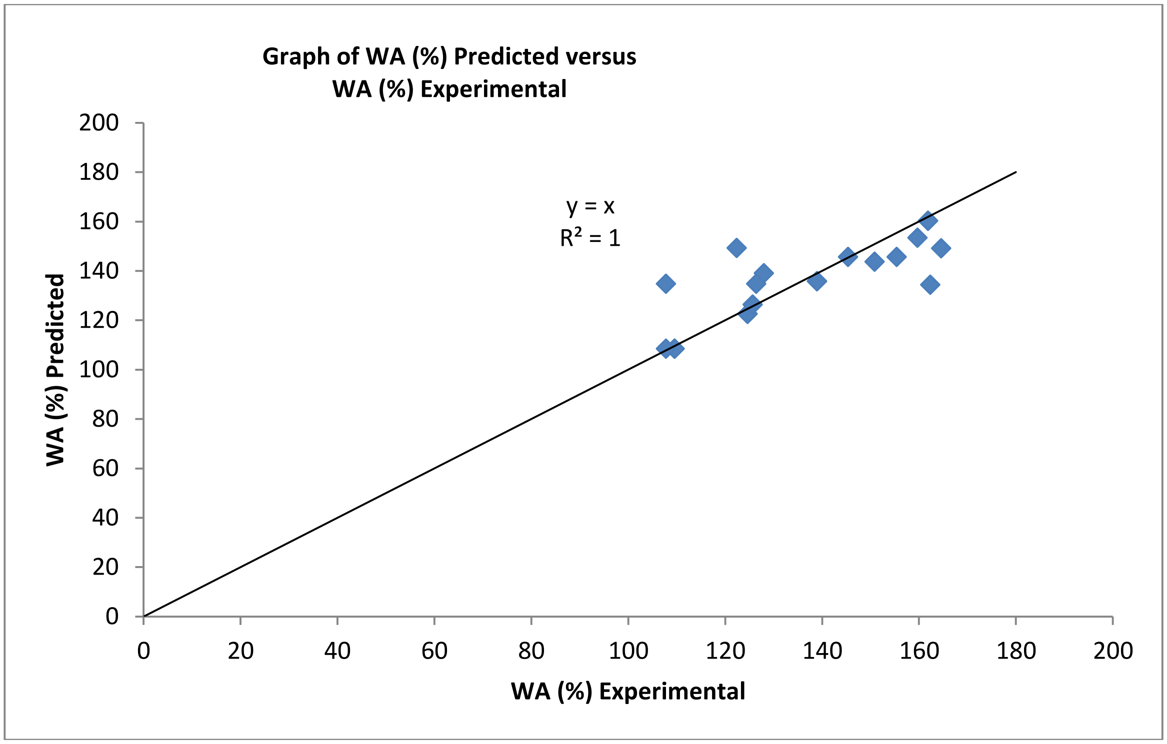
Figure 4 MOR at 10%, 15% and 20% of wheat flour content



(a) (b) (c)

Figure 5 MOE at 10%, 15% and 20% of wheat flour content

The adequacy of the empirical equations can be assessed based on the data points close to the diagonal line. All the Equations (1-4), were predicted with the data as shown in Table 2. The predicted and experimental values are shown in Figure 6. The Figure 6 indicate the good correlation between the experimental and predicted values of WA, TS, MOR, MOE as the points are adjacent to the straight line. This confirms the adequacy of the models predicted.


Graph of WA (%) Predicted versus
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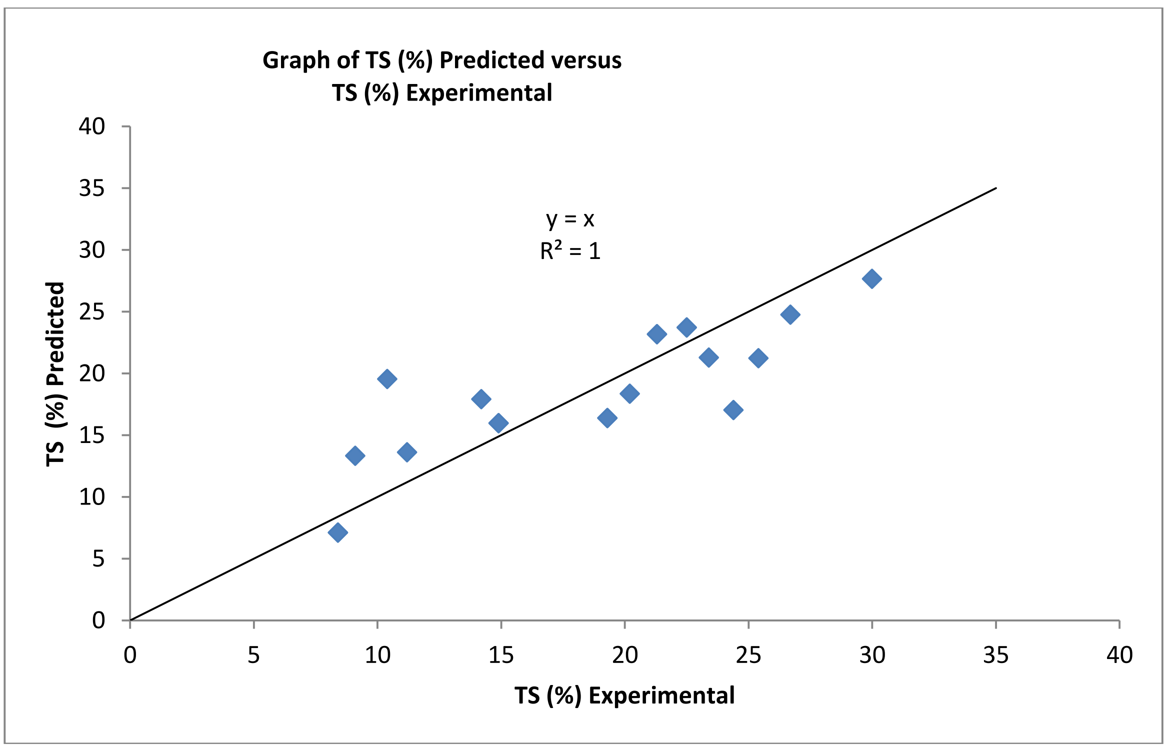
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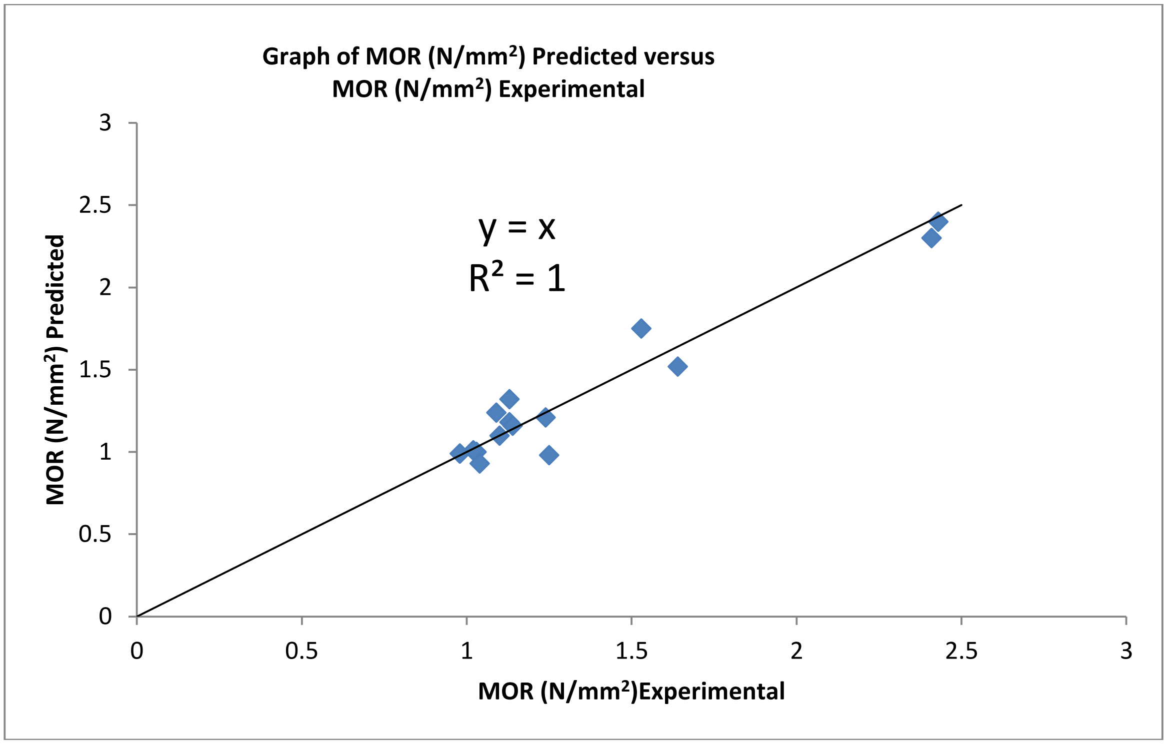

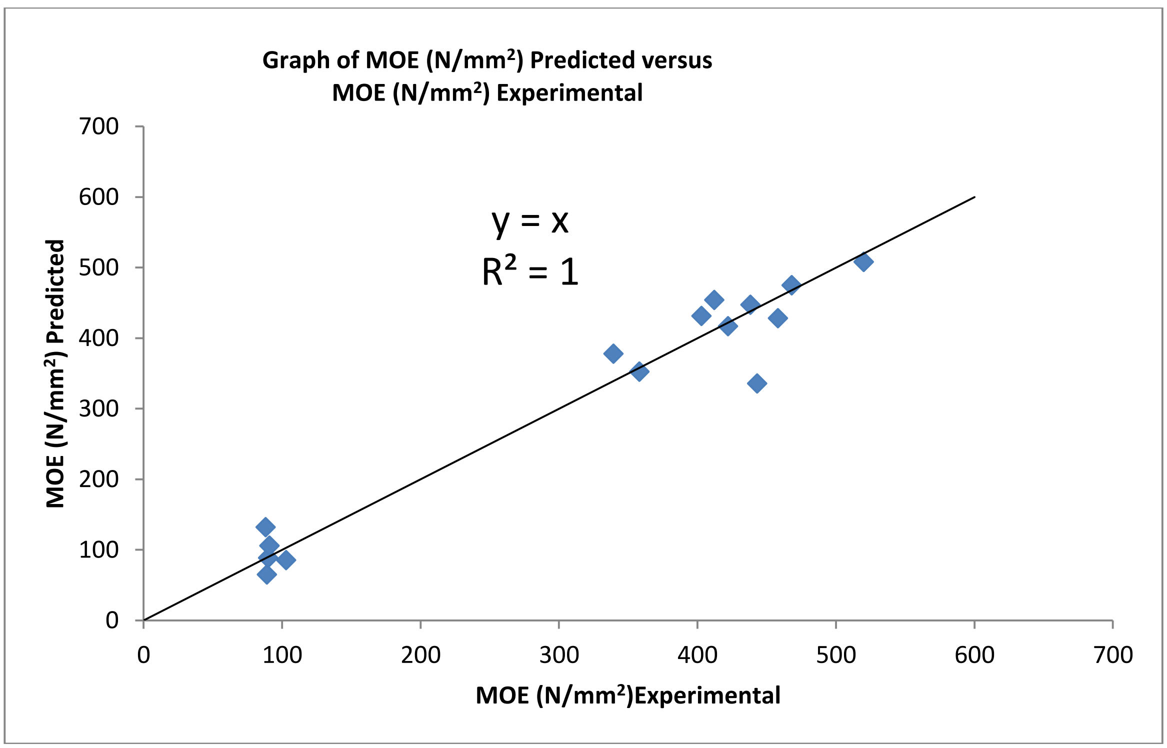

Figure 6 Predicted vs. experimental values of WA, TS, MOR, MOE

3.4 Optimization of MUF Resin with Wheat Flour Filler

The optimization of this study is based on the target values for each response as shown in Table 3. In the optimization parameter WA, TS, MOR and MOE were analysed. Minimum target of water absorption and thickness swelling were desired so as to obtain an optimum condition for MUF resin to perform better water resistance properties. Besides that, for MOR and MOE, maximum values were desired so that an optimum condition can be achieved for a better performance of MUF resin with wheat flour as filler.

Table 3 The criteria for optimization of MUF Resin with wheat flour as filler

Responses	Target	Range	
		Low	High
Water absorption (WA)	Minimum	107.78	162.32
Thickness swelling (TS)	Minimum	8.40	30
Modulus of rupture (MOR)	Maximum	0.98	2.43
Modulus of elasticity (MOE)	Maximum	88.10	520.07

The criteria above can be achieved by the suggested conditions as shown in Table 4. Table 4 shows the optimal condition of each experimental parameter to meet the desired response. It shows that the suggested condition of MUF resin with wheat flour as filler is 149.88^{o}C of pressing temperature, 250 sec of curing period and 10% of wheat flour filler. Figure 7 shows the desirability of optimization conditions for MUF resin with wheat flour filler and it was found to be 0.997.

Table 4 Suggested condition for MUF Resin with wheat flour as filler

Experimental parameter	Suggested condition
Temperature (^{o}C)	149.8
Pressing time (sec)	250.0
Wheat flour content, % (w/w)	10.0

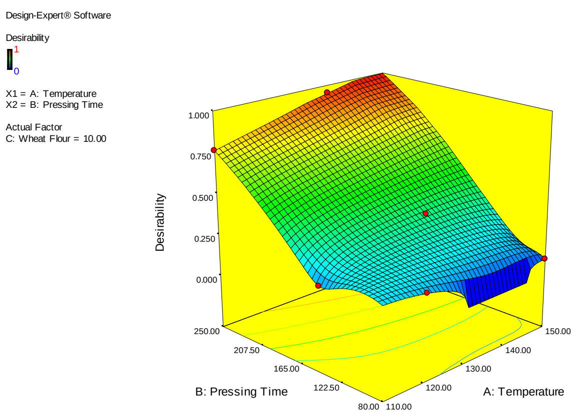

Figure 7 Graph of desirability of optimization condition for MUF resin with wheat flour

■4.0 CONCLUSIONS

In this study, the analysis of the experimental responses shows that wheat flour gives better performance based on the percentage of it used as filler. The optimal conditions for MUF resin with wheat flour filler established at 149.88^{o}C of temperature, 250.00 sec of pressing time, and 10.00% (w/w) of wheat flour filler. It was found that most of their performance gives a satisfactorily result but lower performances as compared to the experimental control value. Thus, it can be concluded MUF resin can be utilized with wheat flour as filler but the pressing time should be done longer so as to obtain a complete polymerization between adhesive, filler and wood particles.

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