OPTIMISATION OF THE PROCESSING VARIABLES FOR HIGH POLYMER LOADING IN COMPRESSED WOOD USING RESPONSE SURFACE METHODOLOGY

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ZAIDON A, KIM GH, PARIDAH MT, BAKAR ES & RUSHDAN I. 2012. Optimisation of the processing variables for high polymer loading in compressed wood using response surface methodology. Response surface methodology (RSM) was applied to optimise processing variables in achieving high polymer loading in compressed sesenduk (*Endospermum diadenum*) wood. Central composite design (CCD) using RSM with three process variables, namely, phenol formaldehyde concentration (PC), pre-curing time (PCT) and compression ratio (CR) was used in this study to optimise effects of these variables on polymer loading. A quadratic model was obtained for polymer loading through this design. The experimental values were in good agreement with the predicted ones and the model was highly significant with correlation coefficient of 0.915. Within the pre-curing time applied in this study, the independent variable did not show any significant effect on polymer loading. Interactions between PC and PCT, PC and CR, and PCT and CR were negligible. Maximum polymer loading was achieved with 36% phenol formaldehyde solution and 74% compression ratio.

Keywords: Response surface method, central composite design, *compreg*

ZAIDON A, KIM GH, PARIDAH MT, BAKAR ES & RUSHDAN I. 2012. Pongoptimuman pemboleh ubah pemprosesan untuk memperoleh ketahanan polimer yang tinggi dalam kayu termampat menggunakan keadah permukaan respons. Kaedah permukaan respons (RSM) digunakan untuk mengoptimumkan pemboleh ubah pemprosesan bagi memperoleh ketahanan polimer yang tinggi dalam kayu sesenduk (*Endospermum diadenum*) termampat. Reka bentuk komposit pusat (CCD) menggunakan RSM dengan tiga pemboleh ubah proses iaitu kepekatan fenol formaldehid (PC), masa pra-pematangan (PCT) dan nisbah mampatan (CR) digunakan dalam kajian ini untuk mengoptimumkan kesan pemboleh ubah terhadap ketahanan polimer. Model kuadratik untuk ketahanan polimer telah diperoleh menggunakan reka bentuk ini. Nilai yang diperoleh daripada eksperimen adalah sebaik nilai yang diramalkan dan model yang diperoleh sangat signifikan dengan pekali korelasi iaitu 0.915. Dalam had masa pra-pematangan yang digunakan dalam kajian ini, pemboleh ubah bebas tidak menunjukkan sebarang kesan ketara terhadap ketahanan polimer. Interaksi antara PC dengan PCT, PC dengan CR dan PCT dengan CR sangat kecil. Ketahanan polimer maksimum boleh dicapai dengan larutan 36% fenol formaldehid dan nisbah mampatan 74% .

INTRODUCTION

Lesser-known timber species in Malaysia are still not fully utilised due to their poor inherent properties. These timbers are usually low in density but have potential to be utilised for high-value-added products if their properties can be improved. Chemical modification of wood either by bulking, internal coating or cross-linking is attractive for the improvement of strength properties, dimensional stability and durability of wood against decay (Hill 2006). An effective method to improve properties of low density wood involves phenol formaldehyde (PF) resin impregnation and compression at considerably high hot-pressing pressure (Shams & Yano 2004). This product is known as compressed wood (*compreg*). It has also been found that PF-impregnated wood strips followed by laminating in a hot press can produce high quality *compreg*

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laminates which have potential applications in parquet flooring, panelling and furniture components (Zaidon et al. 2010).

One of the dominant factors in improving properties and dimensional stability of treated wood is the level of polymer loading. Density and mechanical properties of compressed wood increase with increasing polymer loading coupled with the densification process (Yano et al. 2001). On the other hand, dimensional stabilisation of compressed wood is dependent on polymer loading and polymer distribution in the treated wood (Rowell & Konkol 1987, Inoue et al. 1991). Another factor worth considering is the molecular weight of the PF resin. PF resin with molecular weight (Mw) of 290–480 is able to penetrate into the cell wall and significantly reduces swelling (Rowel 2005). However, PF resin with Mw of 820 remains in the cell lumen without resulting in any significant stability (Furuno et al. 2004). High-polymer loading in compressed wood can be achieved by optimising the processing variables. These include Mw of the PF resin, concentration of the PF resin, pre-curing time and compression ratio (ratio of final thickness to initial thickness of wood).

Response surface methodology (RSM) is a mathematical and statistical technique used for analysing effects of several independent variables (Myers & Montgomery 2002). In many cases, the relationship between the response and independent variables is not known. Therefore, in the first step of RSM, it is crucial to approximate the response in terms of analysing independent variables. This process usually employs a low-order polynomial equation in a predetermined region of the independent variables, which will later be analysed to locate the optimum values of independent variables for the optimum response (Ceylan et al. 2008). In this study, a model for high polymer loading in compressed wood of sesenduk *(Endospermum diadenum)* was developed using RSM. Independent variables studied were PF concentration (PC), pre-curing time (PCT) and compression ratio (CR).

MATERIALS AND METHODS

Methodology

The material used in this study were wood strips of sesenduk (*E. diadenum)*. Treating solution used was low molecular weight phenol formaldehyde (approximately 600 Mw, LmwPF). Pre-weighed, air dry (15% moisture content) wood strips of 25 × 150 × 5 mm thick were treated with LmwPF solution using vacuum-pressure process. The process involved 30 min vacuum followed by filling the set up with solution and soaking under pressure of 690 kPa for 30 min. After treatment they were pre-cured in an oven at 65 °C prior to compression in a hot press at 150 ± 2 °C for 20 min (Rowell & Konkol 1987). The *compreg* was then conditioned in a conditioning room at 25 ± 2 °C and 65 ± 2% relative humidity until constant weight. The weight was recorded. The polymer loading was determined as follows:

$$\text{PL}\ \% = 100\left(\frac{W_f - W_i}{W_i}\right) \qquad (1)$$

where W_f is the constant weight in a conditioning room after treatment and W_i is the constant weight in a conditioning room before treatment.

Response surface methodology and central composite design

To explore the effect of variables on the response in the region of investigation, a central composite design (CCD) with three factors and three levels was performed. CCD with RSM has previously been used to evaluate the effect of multiple parameters on properties of panel product (Wang & Lam 1999, Li et al. 2009). It is considered as a robust mathematical and statistical technique for models that alleviated tribulation where multiple parameters may influence responses.

Previous work (Zaidon et al. 2010) showed that treating solution concentration, pre-curing time and compression ratio were considered the most effective independent variables. The percentage of polymer loading was taken as the response. The CCD was applied using Design Expert Software (State Ease, Design Expert 8). The range and the level of variables under investigation are given in Table 1.

In the regression equation, the test variables were coded according to equation 2:

$$xi = \frac{(x_i - x_{i0})}{\Delta x_i} \qquad (2)$$

where *xi* is the independent variable coded value, x_i is the independent variable real value, x_{i0} is the independent real value on the centre point

Table 1 The range and level of the variables

Factor	Variable	Unit	Range and level of actual and coded values				
			-α	-1	0	1	α
x_1	PC	%	20	24.05	30	35.95	40
x_2	PCT	Hour	3	4.21	6	7.79	9
x_3	CR	%	70	74.05	80	85.95	90

PC = phenol formaldehyde concentration, PCT = pre-curing time, CR = compression ratio

(Elibol 2004, Roriz et al. 2009) and Δx_i is the interval, i = 1, 2, 3.

A quadratic model equation was used to express the response as a function of independent variables as shown in the following equation.

$$Y = \beta_0 \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} x_i x_j + \varepsilon \quad (3)$$

where Y is the predicted response, β_0 is the constant coefficient, β_i is the linear coefficient, β_{ii} is the quadratic coefficient, β_{ij} is the interaction of the coefficient, and x_i and x_j are the coded factors (Aghaie et al. 2009).

Three important parameters, i.e. concentration of phenol formaldehyde (PC = x_1), pre-curing time (PCT = x_2) and compression ratio (CR = x_3) were selected as the independent variables and the polymer loading (PL = Y) was the dependent response variable. Each of the independent variable was studied at five different levels containing two star points (α = 1.68) and seven replications of the central point with a total of 20 experiments. The effect on polymer loading corresponding to combined effects of the three variables was studied in their specific ranges: PC from 24.05–40.00%, PCT 3–9 hours and CR (per cent change of thickness after compressing) 70–90%. The plan of CCD in coded and actual levels of the three independent variables are shown in Table 2. The results of the experimental design were analysed and interpreted using Design Expert Software (Stat Ease version 8) and the response 3D plots were built using the same software.

RESULTS AND DISCUSSION

The actual and predicted percentages of polymer obtained through RSM analysis are presented in Table 3. Actual values corresponded to the measured response data for a particular run and predicted values were evaluated from the model. We used quadratic model to explain the mathematical relationship between independent variables and the dependent response. In the first stage of analysis, the results showed that the coefficients, x_2, x_2^3, x_1, x_2, $x_1.x_3$ and $x_2.x_3$ were statistically insignificant. Therefore, they were deleted from the equation and added to the lack of fit. As a result, new regression models were obtained for both coded and actual factors (equations 4 and 5 respectively):

$$Y = 57.9027 + 14.60537x_1 - 2.1196x_3 + 2.358194\, x_1^2 + 3.316324\, x_2^2 \quad (4)$$

$$PL, \% = 673.08 - 1.546\, PC - 15.36\, CR + 0.067\, PC^2 + 0.094\, CR^2 \quad (5)$$

The results of analysis of variance (ANOVA) are shown in Table 4. The model of the equation and each term were significant at the 1% level. The equation was highly reliable as reflected by the high coefficient of determination value (r^2), 0.915. The model also revealed a statistically insignificant lack of fit (p = 0.224) at 5% level. Since this value was more than 0.05, the model was statistically appropriate for further analysis.

The effects of variables on polymer loading are shown in Figures 1–3. A graph was constructed by plotting the central values of the variables that affected polymer loading, i.e. 30% PC, 6 hours PCT and 80% CR. The effect of PC and PCT on the response at a fixed CR of 80% is shown in Figure 1. It could be seen that polymer loading increased with increasing resin concentration regardless of pre-curing time. The response value reached its highest value (74%) at 36% resin concentration. This result was comparable with that of Zaidon (2009). Using similar treatment process, the author found that sesenduk treated with 30–40% LmwPF had polymer loading of 68.61–85.52%. It is anticipated that when the treating concentration increases to a certain high value, the loading reaches a plateau and remains

Table 2 Experimental conditions of central composite design (CCD)

Run	Coded value			Actual value		
	x_1	x_2	x_3	PC (%)	PCT (hours)	CR (%)
1	-1	-1	-1	24.05	4.21	74.05
2	1	-1	-1	35.95	4.21	74.05
3	-1	1	-1	24.05	7.79	74.05
4	1	1	-1	35.95	7.79	74.05
5	-1	-1	1	24.05	4.21	85.95
6	1	-1	1	35.95	4.21	85.95
7	-1	1	1	24.05	7.79	85.95
8	1	1	1	35.95	7.79	85.95
9	-1.68	0	0	20	6	80
10	1.68	0	0	40	6	80
11	0	-1.68	0	30	3	80
12	0	1.68	0	30	9	80
13	0	0	-1.68	30	6	70
14	0	0	1.68	30	6	90
15	0	0	0	30	6	80
16	0	0	0	30	6	80
17	0	0	0	30	6	80
18	0	0	0	30	6	80
19	0	0	0	30	6	80
20	0	0	0	30	6	80

PC = phenol formaldehyde concentration, PCT = pre-curing time, CR = compression ratio

constant even if more concentration is used. Rowell (2005) stated that higher solute content in PF solution might increase the viscosity, thus, limiting the penetration through the minute structure of the wood. The results also revealed that the range of PCT used did not significantly affect polymer loading. According to Shams and Yano (2004), the purpose of pre-curing is to plasticise the cell wall before initiating collapse at lower pressure. PF resin is effective since it polymerises before curing and fixes the deformation condition permanently after curing (Shams & Yano 2011).

The effect of PC and CR on polymer loading at fixed PCT of six hours is presented in Figure 2. Increasing the PC and decreasing the CR increased the polymer loading in the wood. Maximum loading of 80% was attained at 36% PC and 74% CR. Figure 3 shows the effect of PCT and CR on the response at a fixed PC of 30%. Polymer loading increased with decreased CR at any PCT. PCT gave no significant effect on PL in the wood within the range of the tested period. The maximum loading was 63.5%. The relationship between CR and PL is not clear. However, it is believed that the lower the CR, the more PF resin fixes the cell deformation. As a consequence, more polymer is retained in the wood.

Adequacy of the model

The fitted model needs to be assessed to ensure that it gives sufficient approximation of the results obtained in the experimental conditions. The coefficient of multiple regressions, r^2, is a global statistic parameter to assess the fit of a model (Myers & Montgomery 2002). In this model, r^2 was 0.915, which indicated the fitness of the model (Table 4). For further validation of the model, adjusted r^2 was used for confirming the model adequacy. The adjusted r^2 was calculated to be 0.958 which indicated a good model for use in field conditions. A residual

Table 3 The actual and predicted polymer loading percentages

Run	Response		R-studentised residual
	Observed	Predicted	
1	52.33	51.09	0.517
2	83.44	80.30	1.311
3	54.18	51.09	1.290
4	79.44	80.30	-0.360
5	45.44	46.85	-0.590
6	78.79	76.06	1.139
7	45.7	46.85	-0.481
8	80.57	76.06	1.883
9	40.32	40.01	0.179
10	84.84	89.14	-2.480
11	56.2	57.90	-0.673
12	59.61	57.90	0.675
13	68.28	70.85	-1.482
14	62.3	63.72	-0.819
15	57.32	57.90	-0.230
16	54.44	57.90	-1.369
17	59.91	57.90	0.794
18	56.21	57.90	-0.669
19	56.7	57.90	-0.476
20	59.53	57.90	0.643

Table 4 ANOVA for response surface reduced quadratic model

Source	Sum of square	Df	Mean square	F-value	Prob > F
Model	799.21	4	799.21	109.96	< 0.0001
x_1	2913.24	1	2913.24	400.83	< 0.0001
x_3	61.36	1	61.36	8.44	0.0109
x_1^2	80.94	1	80.94	11.14	0.00450
x_2^2	160.07	1	160.07	22.02	0.000289
Residual	7.27	15	7.27		
Lack of fit	8.75	10	8.75	2.04	0.224
Pure error	4.30	5	4.30		
Total	799.21	19			
r^2	0.9148		Adjusted r^2	0.9582	

analysis was also carried out for validating the model accuracy (Myers & Montgomery 2002). Identification of the outliers was performed by examining the internally studentised residuals. The residual should be approximately normal with mean zero and unit variance. Figure 4 shows that none of the studentised residuals had value higher than 2. To validate the model further and check the outliers, the r-studentised values were calculated (Table 3). All the values were within -2.5 to +2.5 (values between –3 and +3 were the acceptable limit), thereby validating the model.

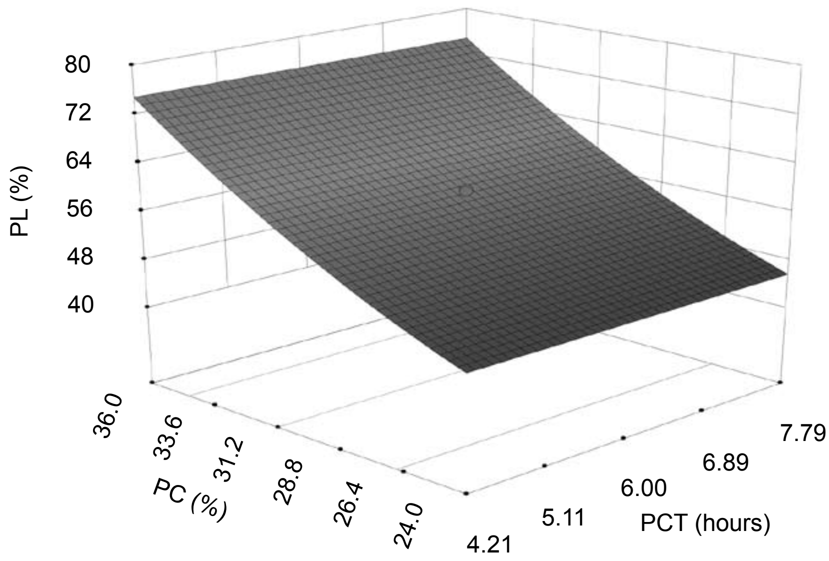

Figure 1 3D-surface plot of polymer loading (PL) in *compreg* wood as a function of phenol formaldehyde concentration (PC) and pre-curing time (PCT) at a fixed value of 80% compression ratio

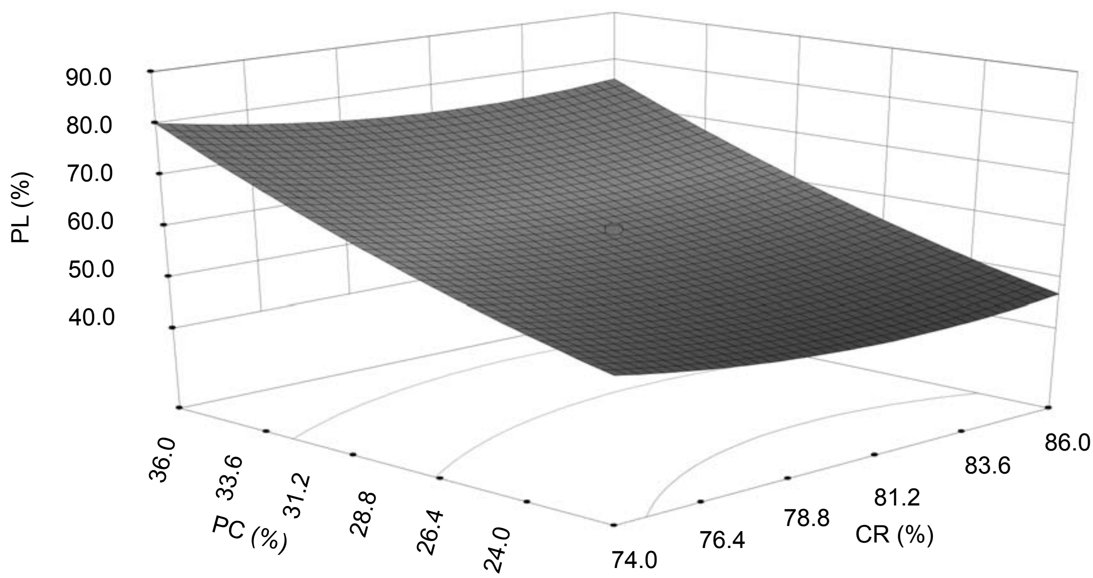

Figure 2 3D-surface plot of polymer loading (PL) in *compreg* wood as a function of phenol formaldehyde concentration (PC) and compression ratio (CR) at a fixed value of six hours pre-curing time

CONCLUSIONS

Response surface methodology was successfully applied to determine the optimal operational conditions for maximum PL in *compreg E. diadenum* wood. A quadratic model in terms of PC, PCT and CR was developed. The experimental values were in good agreement with predicted ones and the model was highly significant with correlation coefficient of 0.915. The independent variable did not show any significant effect on polymer loading. Interactions between PC and PCT, PC and CR, and PCT and CR were negligible. Maximum polymer loading was achieved when treated with 36% phenol formaldehyde solution and 74% compression ratio. The minimum polymer loading in the wood was found to be treatment with 20% PF concentration and 85% CR, with a value of 40.3%. At each centre point of the

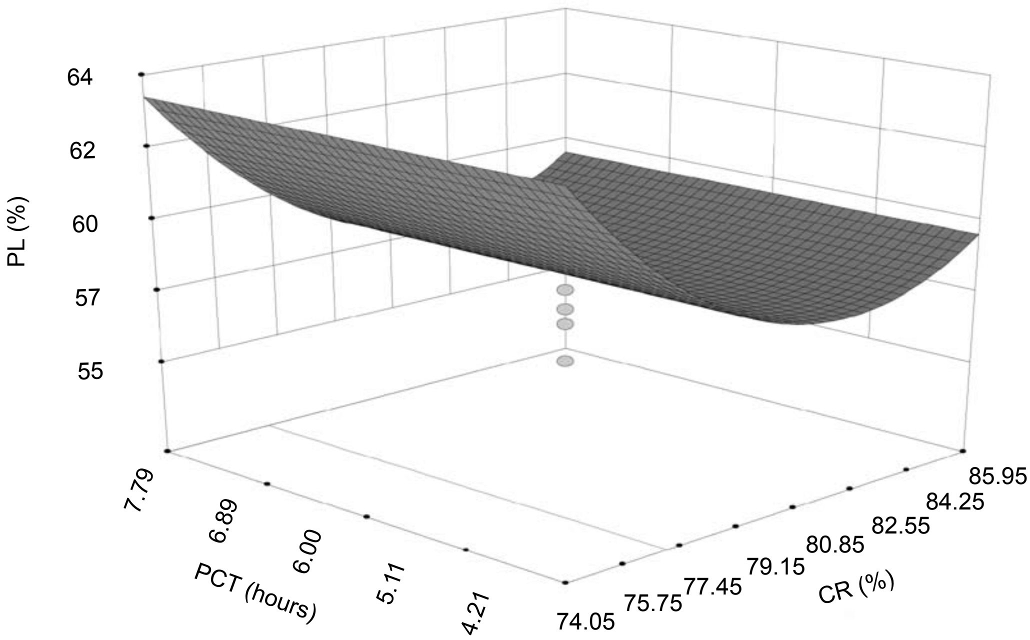

Figure 3 3D-surface plot of polymer loading (PL) in *compreg* wood as a function of pre-curing time (PCT) and compression ratio (CR) at a fixed value of 30% phenol formaldehyde concentration

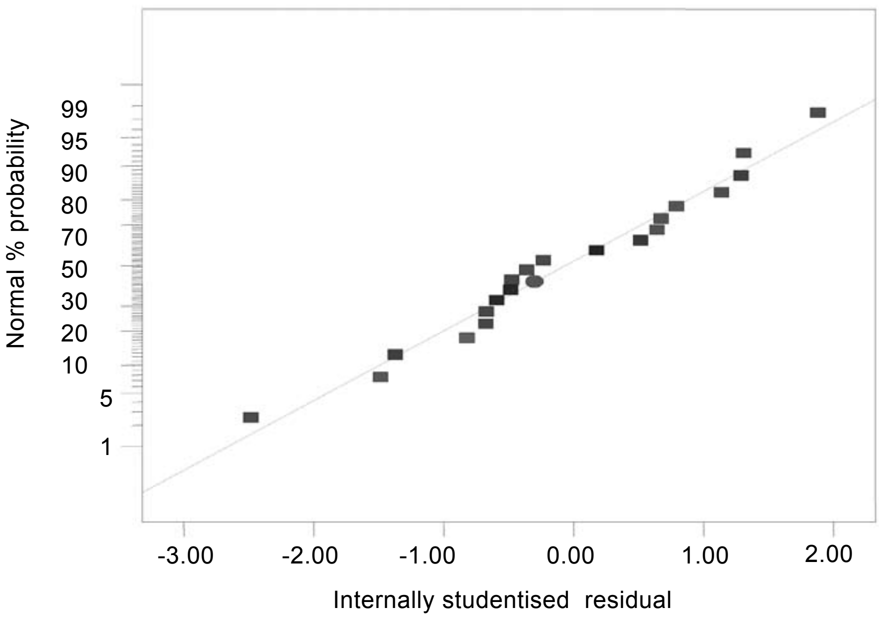

Figure 4 Studentised plot showing normal % probability versus internally studentised residuals

independent variables, which were PC 30%, PCT 6 hours and CR 80%, the optimal polymer loading was in the range of 57–60%.

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