Use of Almond Shell Powder in Modification of the Physical and Mechanical Properties of Medium Density Fiberboard

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This study evaluated the effects of almond shell powder content as an extender, as well as the effects of varying paraffin contents, temperature, and press time on the properties of medium-density fiberboard. Response surface methodology (RSM) based on a five-level, four-variable central composite rotatable design was applied to evaluate the effects of independent variables on the modulus of rupture (MOR), internal bonding (IB), and thickness swelling (TS) of medium-density fiberboard. Mathematical model equations were derived from computer simulation programming to optimize the properties of the panels. These equations, which are second-order response functions representing MOR, IB, and TS, were expressed as functions of four operating parameters of panel properties. Predicted values were found to be in agreement with experimental values (R^2 values of 0.93, 0.90, and 0.90 for MOR, IB, and TS, respectively). The study showed that RSM can be efficiently applied in modeling fiberboard properties. It was found that almond shell powder maintained the MOR, IB, and TS at desirable levels up to 10.6% as an extender in the resin. Using 1.7% paraffin as a sizing agent, a press temperature of 158 °C, and a duration of 6.43 min had the highest impact on the improvement of the studied properties of medium-density fiberboard.

Keywords: Medium-density fiberboard; Response surface methodology; Properties; Paraffin; Extender; Press time and temperature

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INTRODUCTION

The physical and mechanical properties of fiberboard are strongly affected by manufacturing variables, such as binder content, board density, duration and temperature of heat treatment, type of additive, and sizing agents. Many researchers have investigated the correlation between the properties of fiberboards with adhesive application (Roffael *et al.* 2003; Roffael *et al.* 2005), matt forming and heat transfer (Cao *et al.* 2007), and adhesive curing (Heinemann 2004). In this way, different factors such as sizing agents, extenders, temperature, and press duration could be introduced during manufacturing to improve certain properties.

Dimensional stability is one of the most important characteristics of wood-based composites. This characteristic is affected by the hydroxyl groups of cell wall polymers. Dimensional stability can be improved by replacing or covering the hydroxyl groups with a less hygroscopic material, such as a sizing agent. However, this material negatively affects the mechanical properties of wood composites (Chow *et al.* 1996). Moreover, the hydrophobic properties of paraffin waxes have a short-term effect. In contrast to these results, Akrami *et al.* (2011) mentioned that neither adding paraffin nor press time and temperature had a significant influence on modulus of rupture (MOR) and modulus of elasticity (MOE) in medium-density fiberboard (MDF) panels. Chow *et al.* (1996) investigated the effect of fiber treatment, resin content, and wax content on the mechanical and physical properties of dry-process hardboard made from hemlock. It was concluded that increasing resin content and paraffin content significantly altered the mechanical and physical properties of the board.

One of the biggest advantages of using extenders in urea formaldehyde (UF) resin is the use of less base resin, which leads to cost savings. Resin extenders applied in the production of MDF usually improve the efficiency of standard binders while simultaneously maximizing the usefulness of the base resin by maintaining or improving its properties. Hojilla-Evangelista and Bean (2004) evaluated the use of sorghum flour as an extender in plywood production and concluded that UF resin containing sorghum flour as a protein extender had mixed properties and appeared to be superior to those of the standard wheat flour-based plywood glue. The viscosity and adhesion strength of the sorghum-based plywood glue were substantially improved to acceptable levels by increasing the amount of sorghum flour in the glue mixture. By using the organic extender, it seems that the viscosity of the adhesive increases with increasing protein content. It was observed that the presence of more protein can provide additional reactive groups for cross-linking and polymerization reactions with resin and wood (Frihart and Wescott 2004).

Press temperature and duration offer the potential to increase resistance to moisture absorption, as well as the dimensional stability of wood fiber composites, by modifying the hygroscopicity of wood fiber *via* the degradation of hemicelluloses, lignin, and cellulose (Winandy and Krzysik 2007). The effects of press temperature and time on the properties of medium-density fiberboard were studied by Kargarfard *et al.* (2009). They showed that the MOR of boards was increased with increasing press temperature. Also, the effect of press temperature on MOE was significant, and the MOE of boards increased with increasing press temperature. The results revealed that increasing steaming time and press time had negative effects on internal bonding (IB). On the other hand, press time had a significant effect on dimensional stability; the minimum of thickness swelling was obtained after three minutes of press duration.

The objectives of this study were to determine the effects of content of powdered almond shell as a resin extender with regard to various levels of paraffin content on matt, temperature, as well as duration of press time on the manufacture of MDF panels with equal densities. This study investigated the interrelated effect of the aforementioned parameters on the physical and mechanical properties of MDF.

EXPERIMENTAL

Materials

Preparation of boards

A total of 30 groups of medium-density fiberboard with nominal dimensions of 350 mm ×350 mm ×10 mm were involved in this study. Bagasse fiber was collected from a local pulp and paper mill (Pars Paper Co.; Haft Tapeh, Iran). To avoid felting of the fibers caused by wet conditions, they were dried in a laboratory oven. After taking samples from the oven-cooled fibers, they were preserved in polyethylene bags. The fibers were dried to a 3% moisture content.

Methods

In the production of experimental panels, a mixture of urea formaldehyde and powdered almond shell (as an extender) was used as a binder with ratios of 100/0, 95/5, 90/10, 85/15, and 80/20 (by weight). Liquid resins (55% solid) were sprayed on the fibers as they were tumbled (for 3 min) in a rotating drum. After adding resin, solid powdered paraffin was added to the fibers at five levels: 0%, 0.5%, 1%, 1.5%, and 2% based on oven-dry fiber weight, according to the experimental design. The mixture of fibers, resin, and paraffin was then tumbled for 2 min.

The resonated fibers were laid up by hand into a 350 mm×350-mm mat on a metal plate, and then manually pre-pressed. The initial mat thickness was about 15 cm, and the target thickness of the panel was 1 cm. The mats containing 13% moisture content were exposed to hot pressing. The hot press was applied at a pressure of 35 kg/cm^2 at five levels of temperatures, *i.e*., 145, 155, 165, 175, and 185 °C. Depending on the treatment number, five pressing time durations (5, 6, 7, 8, and 9 min) were applied, as shown in Table 1. The produced panels were conditioned at 65±5% relative humidity and 20 °C to reach a moisture content of about 12% before being cut to final dimensions of 350 mm × 350 mm × 10 mm.

Table 1. Coded and Actual Levels of Variables Considered for the Design

Variable	Unit	Coded Level of Variable				
		−2	−1	0	1	2
X1: paraffin content	%	0	0.5	1.0	1.5	2.0
X2: extender content	%	0	5	10	15	20
X3: press temperature	°C	145	155	165	175	185
X4: press time	min	5	6	7	8	9

Analysis of MDF properties

Response surface methodology (RSM) was used to develop a mathematical model in the form of multiple regression equations for responses (modulus of rupture (MOR), internal bonding (IB), and thickness swelling (TS)). The independent variables (weight ratio of UF/melamine formaldehyde (MF) resins, paraffin content as a sizing agent, press temperature, and press duration) were viewed as a surface on which the mathematical model was fitted. The second order polynomial (regression) equation was used to represent the response surface Y, which is given by Balasubramanian *et al*. (2008) in Eq. (1):

$$Y=\beta_0+\sum_{i=1}^{4}\beta_i X_i+\sum_{i=1}^{4}\beta_{ii}X_i^2+\sum_{i=1}^{4}\sum_{j=i+1}^{4}\beta_{ij}X_iX_j \qquad (1)$$

A five-level, four-factor (CCRD) was employed. The fractional factorial design consisted of 16 factorial points, eight axial points (two axial points on the axis of each design variable at a distance of 2 (α=±2) from the design center), and six center points. The variables and their levels selected for the study are represented in Table 1. Table 2 shows the actual experiments that were carried out to develop the model. Hence, the total number of tests required for the four independent variables is 4^2 + (4×2) + 6 = 30 (Cochran Cox 1962; Manonmani *et al*. 2007). Subsequent regression analysis, analysis of variance (ANOVA), and response surfaces were generated using Design Expert Software (Version 6.0.6) from Stat-Ease, Inc. (Minneapolis, MN, USA). The optimization of technological parameters for producing ideal mechanical and physical properties was achieved through software numerical optimization.

RESULTS AND DISCUSSION

The modulus of rupture, internal bonding, and thickness swelling of the panels were functions of paraffin content (x_1), extender content (x_2), press temperature (x_3), and press time (x_4) (Table 2).

Table 2. Experimental Design Matrix and Results

Run	X_1	X_2	X_3	X_4	MOR (MPa)	IB (MPa)	TS (%)
1	1.0	10	140	7	18.6	0.61	9.4
2	1.0	10	180	7	19.0	0.70	12.8
3	0.5	5	150	8	25.2	0.77	14.0
4	1.0	10	160	9	22.0	0.82	13.8
5	1.5	5	170	6	21.5	0.75	10.5
6	0.5	15	150	8	20.0	0.77	12.6
7	1.0	10	160	7	23.7	0.78	12.4
8	1.5	15	150	6	21.0	0.60	9.0
9	1.0	10	160	7	23.0	0.79	13.0
10	0.5	5	170	6	21.8	0.87	14.3
11	1.0	20	160	7	23.0	0.72	13.8
12	0.5	15	170	6	22.8	0.75	14.4
13	1.0	10	160	7	23.4	0.79	12.4
14	1.0	0	160	7	26.0	0.83	13.1
15	0.5	15	170	8	23.0	0.90	14.8
16	1.5	15	150	8	21.0	0.70	9.2
17	1.5	15	170	6	23.0	0.68	10.4
18	1.0	10	160	7	23.5	0.80	12.2
19	2.0	10	160	7	22.0	0.77	8.8
20	1.0	10	160	5	20.5	0.73	11.5
21	0.0	10	160	7	24.0	0.99	15.2
22	1.5	5	170	8	20.4	0.80	10.7
23	1.0	10	160	7	23.3	0.83	12.0
24	0.5	15	150	6	20.0	0.70	12.1
25	0.5	5	170	8	22.0	0.82	13.4
26	0.5	5	150	6	22.0	0.74	11.3
27	1.5	15	170	8	19.8	0.78	12.2
28	1.0	10	160	7	24.0	0.79	12.7
29	1.5	5	150	6	22.0	0.69	10.1
30	1.5	5	150	8	24.0	0.74	10.0

MOR, modulus of rupture; IB, internal bonding; TS, thickness swelling

For the four factors, the selected polynomial (regression) could be expressed as follows (Eq. 2),

$$Y=B_0+b_1x_1+b_2x_2+b_3x_3+b_4x_4+b_{11}x_1^2+b_{22}x_2^2+b_{33}x_3^2+b_{44}x_4^2+b_{12}x_1x_2 +b_{13}x_1x_3+b_{14}x_1x_4+b_{23}x_2x_3+b_{24}x_2x_4+b_{34}x_3x_4 \quad (2)$$

where b_0 is the term of the regression equation, the coefficients b_1, b_2, b_3, and b_4 are linear terms, the coefficients b_{11}, b_{22}, b_{33}, and b_{44} are quadratic terms, and the coefficients of b_{12}, b_{13}, b_{14}, b_{23}, b_{24}, and b_{34} are interaction terms. The developed final mathematical model equations in coded form are given below in Eqs. (3-5):

$$MOR(MPa)=23.34-0.34x_1-0.60x_2-4.2x_3+0.18x_4+0.27x_2^2-1.16x_3^2 -0.55x_4^2-0.36x_1x_3-0.37x_1x_4+0.88x_2x_3-0.46x_2x_4-0.57x_3x_4 \quad (3)$$

$$IB\ (MPa)=0.78-0.04x_1-0.02x_2+0.034x_3+0.028x_4+0.02x_1^2-0.03x_3^2+0.021x_2x_4 \quad (4)$$

$$TS\ (\%)=12.4-1.57x_1+0.075x_2+0.80x_3+0.39x_4-0.41x_3^2+0.34x_2x_3 \quad (5)$$

Experiments were conducted to verify the regression equations (Eqs. 2 through 5). Six manufacturing runs are made of applying different values of paraffin, extender content, press temperature, and time other than what were used in the design matrix. The results obtained were found to be quite satisfactory, and the details are presented in Table 3.

Table 3. Results of Confirmation Experiment

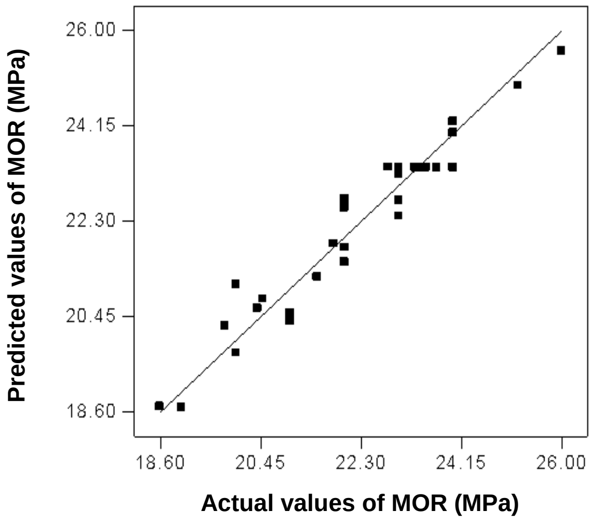
Independent Variables				MOR(MPa)		IB(MPa)		TS(%)	
x_1	x_2	x_3	x_4	Estimate	Predict	Est.	Pred.	Estimate	Predict
1.7	10.61	158.62	6.43	22	22.83	0.77	0.734	8.8	9.88
1.7	10.60	158.58	6.44	22	22.83	0.77	0.734	8.8	9.87
1.7	10.58	158.58	6.44	22	22.83	0.77	0.734	8.8	9.87
1.7	10.66	158.63	6.44	22	22.82	0.77	0.733	8.8	9.88
1.7	10.58	158.60	6.44	22	22.83	0.77	0.734	8.8	9.87
1.7	10.56	158.57	6.44	22	22.83	0.77	0.734	8.8	9.87

According to the analysis of variance (ANOVA), calculated F-ratios were larger than the tabulated values at a 95% confidence level; hence, the models are considered to be adequate. Other criteria commonly used to illustrate the adequacy of a fitted regression model are the coefficient of determination (R^2), the calculated R^2 values, and adjusted R^2 values above 90% and 70%, respectively. These values indicate that the regression models were adequate. The validity of the regression models developed was further tested by drawing scatter diagrams. Moreover, Table 4 shows that there were significant differences between the physical and mechanical properties of the fiberboard.

Table 4. ANOVA Test Results

	S. of Squares		M. Squares		D. Freedom		F	R^2	Ad. R^2	Remarks
	Regression	Residual	Regression	Residual	Regression	Residual				
MOR	85.30	5.77	7.12	0.34	12	17	21	0.93	0.89	Adequate
IB	0.16	0.02	0.02	0.01	7	22	29	0.90	0.87	Adequate
TS	84.80	9.42	14.13	0.41	6	23	34	0.9	0.87	Adequate

Typical scatter diagrams for all the models are presented in Figs. 1 through 3. The observed values and predicted values of the responses are scattered close to the 45° line, indicating an almost perfect fit with the developed empirical models (Kim *et al.* 2003).



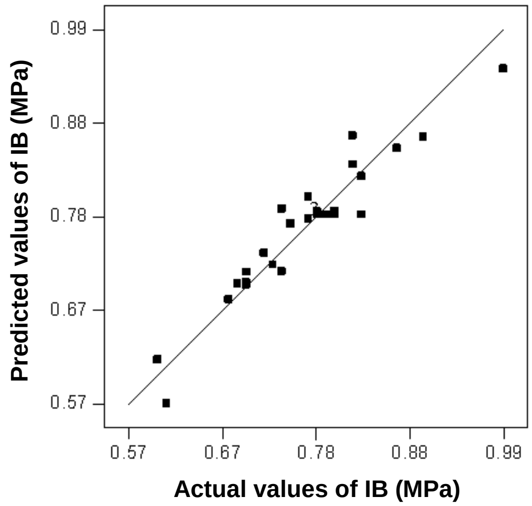


Fig. 1. Scatter diagram of the MOR

Fig. 2. Scatter diagram of the IB

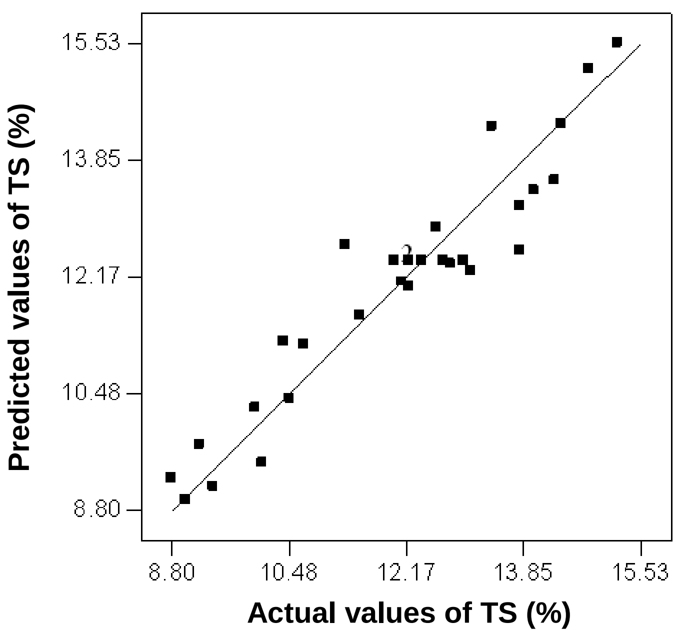


Fig. 3. Scatter diagram of the TS

The effect of different process parameters on the modulus of rupture (MOR), internal bonding (IB), and thickness swelling (TS) of MDF predicted from the mathematical models using the experimental observations are presented in Figs. 4 through 7, showing the general trends between cause and effect.

Figure 4 shows the direct effect of paraffin content on MOR, IB, and TS. It seems that increasing paraffin content decreases the IB and TS of panels, as is well-known from previous studies on the impact of the addition of paraffin emulsion on panel quality.

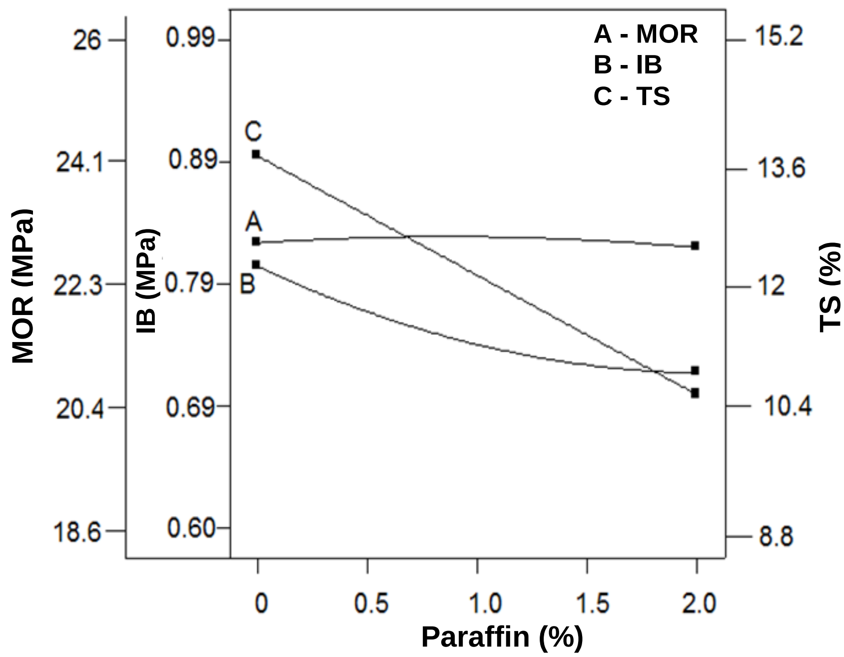


Fig. 4. Direct effect of paraffin content on MOR, IB, and TS

Increasing the amount of added paraffin emulsion makes the particle surface too inert, which means that during adhesive application, fiber coatings are reduced (Antonovi *et al.* 2010). This lowers adhesion efficiency with fibers and consequently reduces mechanical values. Another factor that influences the properties of wood composite panels is the surface wettability of fibers. Because of the waxy layer on the surface of fibers and the resulting decreased wettability, it is difficult for glue to penetrate into the sides of fibers coated with paraffin, which significantly reduces its ability to chemically react with hydroxyl groups; consequently, the formation of gel nails during the bonding process becomes difficult (Shi and Tang 2010). Also, the higher values of contact angles that result from higher amounts of paraffin cause more discontinuity in the resin film layer for increasing diameters of adhesive drops, concentration of stress on unglued regions, and failure of the glue bonds at the inter-phases between resin and wood; consequently, they decrease the internal bonding strength of the specimens. Moreover, the waxy layer (paraffin) encircling the fibers inhibits sufficient contact between the functional groups of resin and the OH groups of fibers, so that water-soluble UF resin is chemically incompatible with the fibers. This is probably one of the primary factors responsible for the reduction in bond quality.

According to Hundhausen *et al.* (2009), the wettability of wood veneer treated with paraffin was significantly lower than that of untreated veneer. It was found that the untreated samples exhibited the most hydrophilic surface, reflected by a decreasing angle, whereas the paraffin-treated samples showed steady angles at approximately 120°. Additionally, moisture is an important factor for increasing thermo-plasticity (Kelly 1977). Decreasing the thermoplastic properties of fibers treated with paraffin caused a lower efficiency of moisture in this situation; on the other hand, decreasing wettability and thermo-plasticity could lead to a decrease in inter-fiber contact, which is confirmed by the IB shown in Fig. 4. The speed of heat transfer is affected by the permeability of the fibers. Using paraffin decreases the permeability of fiber surfaces and consequently decreases fiber thermo-plasticity. In this case, moisture cannot behave as a plasticizer, so the mechanical interlocking necessary for forming good contact between the fibers could not be achieved.

Figure 5 shows the direct effects of extender content on MOR, IB, and TS. It is evident that as extender content was increased from 0% to 20%, the MOR and IB of the panels decreased, while TS increased steadily. At the lowest and highest extender contents, higher and lower MOR and IB were observed, respectively.

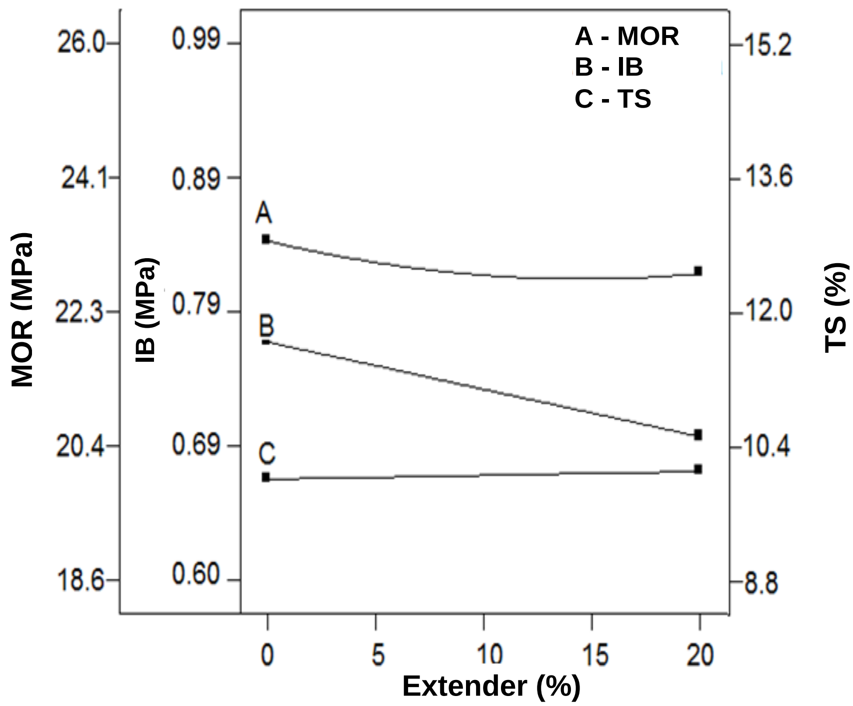


Fig. 5. Direct effect of extender content on MOR, IB, and TS

The results of this study are in contrast to those of other researchers. Doosthoseini and Zarea-Hosseinabadi (2010) reported a modifying effect of extender on urea formaldehyde resin used in plywood manufacturing by decreasing resin viscosity and controlling its penetration into the wood tissue, as well as improving bonding. However, this result is in agreement with those of several previous studies (Ajiwe *et al.* 1998; Lee *et al.* 2003). According to Batalla *et al.* (2005), the strength of wood panels decreases as filler content increases, because the void content increases as well. In fact, the increased porosity reduces the load-bearing volume of the sample and introduces stress concentrations, which make the material less stiff and resistant.

According to Fig. 5, thickness swelling caused by water absorption increased with extender content, probably because the porosity content increased as well. As porosity increases, water finds less resistance to penetration into the sample (Nemli *et al.* 2003). With increasing extender content, the absorption of the resin water extender increases as well, because of its high specific surface area and the quantity of hydrophilic groups (OH) on this surface. Consequently, the moisture coating surface of fibers decreases. This decrease causes slower heat transmission into the core and lower overall plasticization of the fibers, thus slowing the curing of resin and the setting of the boards. This decreases the formation of cross-linkage of the resin polymer and bonding strength between fibers.

The direct effect of press temperature on MOR, IB, and TS is shown in Fig. 6. This indicates that with increasing press temperature, MOR initially rises and reaches its maximum at a nearly average value of the manufacturing value, then starts decreasing. This suggests that the maximum MOR was in the middle value range of the press temperature.

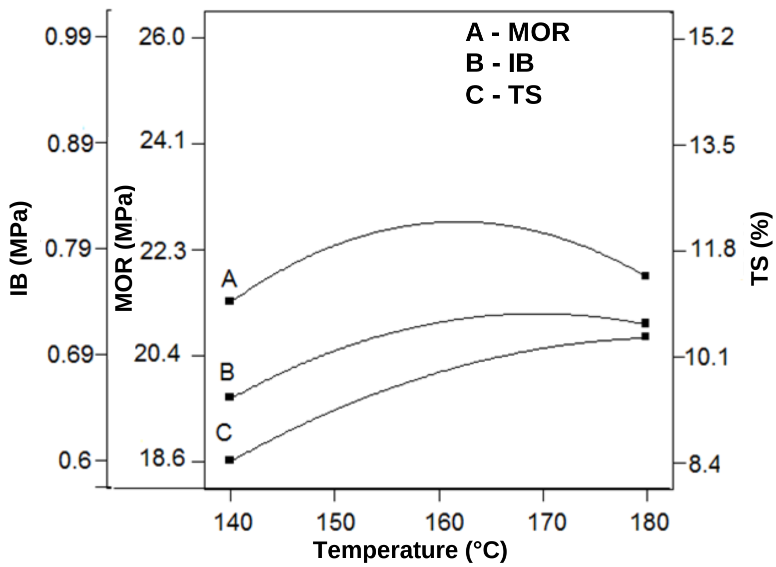


Fig. 6. Direct effect of press temperature on IB, MOR, and TS

With increasing temperature, the largest temperature gradient exists between the surface and core layer of the matt during compression. Higher temperature at the surface layers softens the wood fibers, providing a lower compression modulus and higher compaction. The slowly increasing temperature at the center is not high enough during this time period to plasticize the material, resulting in the spring-back of the fibers to counteract the densification of the surface layers (Zombori 2001). Higher temperatures produce more pronounced density profiles, and therefore larger differences exist among the surface and core layers of the panels. With increasing pick density and decreasing core density, shear stresses can be induced by the contrast between pick and core densities in the panels as reflected in the T- or Y-shaped line of failure formed from the core to the bottom surface (Wong *et al.* 2000).

It is well known that the outermost layers of the panels are the most important factor for increasing or decreasing MOR. With increasing plate temperature, the cross-linkage of resin at the upper and bottom surface layers will be broken at the end of the hot-pressing period, and could form weak layers on the outer-most surfaces. Under low temperatures and limited press time, the fibers of the outermost layers could be easily softened and reach the plastic point for the formation of good inter-fiber contact. However, this causes a reduced consolidation rate, slower heat transition into the core, and lower overall plasticization of the fibers, resulting in less inter-fiber contact; simultaneously, a delay in the curing of resin and the setting of the middle layer of the board occurs. A high compaction ratio may improve contact between the fibers. With increasing press plate temperature, the compaction ratio of panels increases because of improvement in the thermoplastic behavior of wood fibers. Fibers are sufficiently compressed for effective contact between fibers because of a high compaction ratio. Although this increases internal bonding, it cannot improve resistance to thickness swelling. It is obvious that thickness swelling is generally related to internal bonding strength. However, thickness swelling increased because of breaking cross-linkages of resin polymer and a lack of bonding strength on the outermost layers, which were exposed directly to the hot pressing plate.

Figure 7 shows the direct effect of press time on MOR, IB, and TS. It indicates that when press time was increased from 5 to 7 min, the MOR of panels did not increase, but decreased.

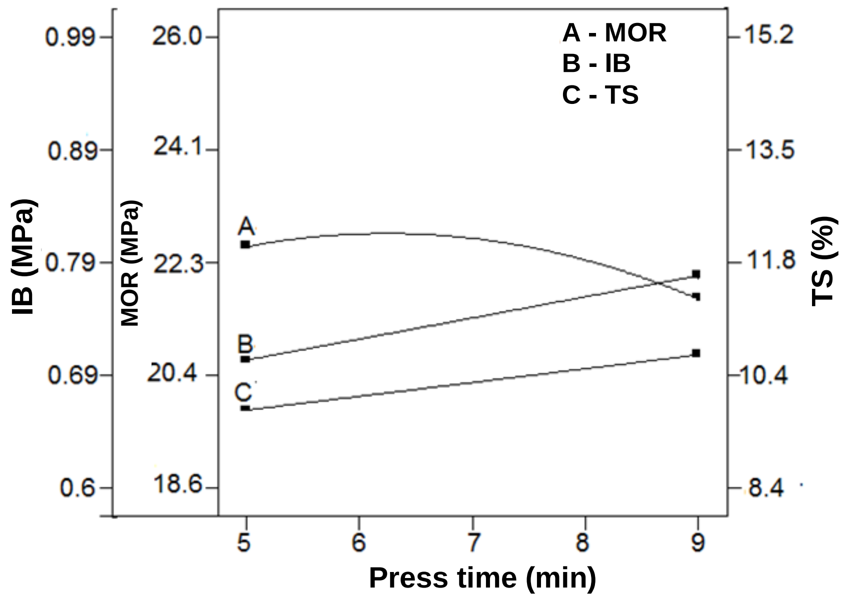


Fig. 7. Direct effect of press time on IB, MOR, and TS

According to the analysis of variance (ANOVA, Table 4), there were significant differences between the physical and mechanical properties of fiberboards. This may be because a shorter time is required to transmit heat from the face to the core. Prolonged pressing is expected to result in a higher degree of resin degradation, especially in the outermost layers; thus, a reduction will result in bending strength of the panels. Conversely, with increased pressing time, internal bonding increases due to an increased rate of consolidation, faster heat transmission into the core, and higher plasticization of the fiber for obtaining a well-connected area between fibers. However, increased pressing time increases thickness swelling because of chemical bonding degradation created by resin in the surface layers, although the internal bonding increases. In the hot-pressing process, the content of water migration from the center of the panels to the external atmosphere increases and at the same time, press time soars. This causes the saturated vapor pressure to become less than the bonding strength of the glue line created among the fibers, and resinated fibers can become more overlapped. It is well known that one of the important factors that can affect TS is the poly-condensation level of resin in the core layer of panels. With increasing press duration, the poly-condensation process in the core increases, but this increase accelerates the breakage of the chemical linkages in surface layers. This causes TS to increase because of an increase in TS resin cross-linkage values in the surface layers.

CONCLUSIONS

1. The model predicted the general effects of production variables on the panel properties. This research revealed that the performance of bagasse fiberboard is strongly dependent upon paraffin content, extender type and content, and press temperature and time. With increasing amounts of paraffin in the mixture, IB and TS decreased drastically, whereas MOR decreased steadily. This may be the result of the inert characteristics of the fiber surface, low wettability of fibers, low speed of heat transfer, and higher values of resin drop contact angles on surfaces.
2. It was determined that as almond shell powder content was increased from 0% to 20%, the MOR and IB of panels decreased, while TS increased steadily. At the lowest and highest extender contents, higher and lower MOR and IB values were observed, respectively. This decrease is related to porosity, which makes the panels less stiff and less resistant to water penetration. It also causes an increase in specific surface area and the quantity of hydrophilic groups.
3. Increases in press time and temperature initially led to increased MOR, which reached a maximum level at a nearly average value of the manufacturing value, then started decreasing. With increasing plate temperature and press time, internal bonding and thickness swelling increased. With increasing temperature or press time, the cross-linkage of resin in the outermost layers could be broken at the end of the hot-pressing period. Under low temperature or press time, the fibers of the outermost layers could be easily softened. However, this caused a decrease in the rate of heat transmission into the core and lower overall fiber plasticization, as well as a delay in curing the resin and setting the middle layer of the board.

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