ESTIMATING MAXIMUM WATER ABSORPTION OF WOOD FIBER/ POLYMER FLUFF COMPOSITES

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ABSTRACT

The objective of this study was to develop a model to estimate the maximum water absorption (*MWA*) of wood fiber/polymer fluff composites as a function of polymer fluff content and board density. Polymeric diphenylmethane diisocyanate (PMDI) resin bonded dry-process wood fiber/polymer fluff composites were used in this study. Six polymer fluff contents (0, 15, 30, 45, 60, and 100%) and four target oven-dry board densities in the range of 0.50–1.00 g/cm^3 were studied. A water immersion test was conducted on these boards. The effect of irreversible thickness swelling after water immersion (TS_i) on the estimation of the maximum water absorption was evaluated. It was shown that the irreversible thickness swelling had a quadratic relationship with polymer fluff content and a linear relationship with oven-dry board density. The TS_i of the composites used in this study was in the range of only 0.04–4.20%, which was negligible in the estimation of maximum water absorption. The prediction of maximum water absorption from the *MWA* model developed in this study was over 95% accuracy for most of the specimens. The maximum water absorption had a linear relationship with the polymer fluff content and a reciprocal relationship with board density.

Keywords: wood composites, wood fiber/polymer fluff composites, water absorption, maximum water absorption.

INTRODUCTION

Total water uptake is an index to evaluate the water resistance of wood composites. To avoid a long duration test for obtaining maximum water absorption (*MWA*) of composites, a 24-h water immersion test is usually used to evaluate the water resistance (ASTM 1987). However, if water absorption rates are different for different composites, it is hard to make a useful comparison on the water resistance just using water absorption (*WA*) values after a 24-h water immersion (Shi et al. 1999). For the new generation of wood/plastic lumber, different wood-plastic combinations may have different water absorption rates. Therefore, ASTM D1037 may not be appropriate for determining water absorption for these composites. The maximum water absorption values of

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the composites should be measured and compared. Wood fiber/polymer fluff composites are processed from wood fibers and polymer mixtures contained in automobile fluff, also called automobile shredder residue (ASR). The components in polymer fluff are mainly plastics such as polyurethane, polypropylene, poly(vinyl chloride) (PVC), acrylonitrile, butadiene styrene (ABS), and etc. Other components such as rubber, paper, and wood chips are also contained in polymer fluff (Jody et al. 1994). Wood and polymer fluff exhibit different water absorption behavior. Wood is a hygroscopic and hydrophilic material that can adsorb moisture from surroundings over a wide range of relative humidities. Most of the components in polymer fluff, such as plastics and rubbers, are hydrophobic materials. Incorporation of the less hygroscopic polymer fluff materials in wood furnish will potentially improve the water resistance of the composites. To determine the water uptake of the hybrid composite material systems, it is desirable to formulate a model to predict their maximum water absorption using the known processing parameters.

Two factors are important for modeling the maximum water absorption of wood fiber/polymer fluff composites, the wood to polymer fluff ratio (or polymer fluff content), and board density. Theoretically, the higher the board density, the fewer voids in the composites, and the greater difficulty for water penetrating into the composites. Also, the higher the polymer fluff content, the less the hydrophilic character of the composites, and the more water resistance of the composites. It is the objective of this study to theoretically develop a model to estimate the maximum water absorption of wood/polymer fluff composites as a function of polymer fluff content and board density.

DEVELOPMENT OF THE *MWA* MODEL

The water absorption of wood fiber/polymer fluff composites will account for the water absorption of both wood and polymer fluff materials. The addition of an adhesive also needs to be considered. The estimation of the *MWA* of the composites can be based on the total voids available in the composites. The number of extra voids created by the irreversible thickness swelling and the bound water contained in wood elements comprising the composites also need to be determined.

Wood is a cellular material that is composed mostly of hollow, elongate, spindle-shaped cells. Water in wood has two basic forms: bound or hygroscopic water and free or capillary water (Siau 1984). Bound water is hydrogen bonded to the hydroxyl groups and intra-cellulose spaces of wood. Free water is found in the lumens or voids of the wood and is held only by weak capillary forces without hydrogen bonding. Fiber saturation point (FSP) is the equilibrium moisture content (EMC) at which the cell walls are saturated (amount of bound water) while the lumens and voids are empty (Siau 1984). The cell-wall saturation occurs when the relative humidity approaches 100%.

As was mentioned previously, polymer fluff is mainly composed of different plastics, rubber, paper, and wood chips. Among these materials, most plastics absorb little moisture (usually below 0.5% when saturated) because water molecules cannot penetrate into either the crystals or the amorphous polymers of the plastics. There are no voids in the plastics except for polyurethane foam. However, polyurethane foams are processed from polyurethane. If the density of polyurethane is used during the calculation, it is not necessary to consider the voids. The water absorption for polymer fluff is due to mainly the hydrogen bonding occurring in the plastic materials. Very few plastics exhibit high water absorption. For example, nylon 6 has a 9% water absorption when saturated because of the great proportion of hydrophilic amide groups (Elias 1993). Other materials mixed in the polymer fluff such as wood chips and paper probably contribute the major part of water absorption in polymer fluff materials.

Equation to calculate the total voids in wood fiber/polymer fluff composites

After the specimen is saturated with water, all the voids will be occupied by water molecules. If the voids in polymer fluff material are neglected and compensated for by using the determined water absorption data of the polymer fluff material, the total volume (cm^3) of void space in the oven-dried composites (V_v^{TOT}) can be expressed as:

$$V_v^{TOT} = V^b - V^f - (V^{wd} - V_v^{wd}) - V^r \quad (1)$$

in which V_v^{wd} is total void volume in the wood (cm^3), V^b is the volume of the board (cm^3), V^f is the volume of the fluff in the composites (cm^3), V^{wd} is the volume of the wood (cm^3), V^r is the volume of the resin (cm^3).

In the manufacture of wood composites, the wood mat is consolidated under high pressure. A certain compression ratio, characterized by the ratio of the board density to material density, is required for the composites to ensure a good interfacial bonding. Density is defined as the weight (g) over volume (cm^3). Wood has a much lower density (i.e. 0.40 g/cm^3) than that of the polymer fluff materials (i.e. 1.10 g/cm^3). Therefore, for wood fiber/polymer fluff composites, it can be assumed that only the wood material is densified during the pressing procedure and the solid volume of polymer fluff in the composites should remain the same. Also, because the density of the dry wood cell wall (1.53 g/cm^3) is much higher than that of the composites (for example, 0.90 g/cm^3), it can also be reasonably assumed that the compression occurs only in the volume of voids in the wood. Therefore, the density of the wood cell wall will not change when the wood is compressed into composites. Based on the above assumptions, the following equations can be used to estimate the volumes of the materials and the voids in the material.

The volume of polymer fluff:

$$V^f = \frac{R \cdot W}{\rho_f} \quad (2)$$

The volume of wood:

$$V^{wd} = \frac{(1 - R)W}{\rho_{wd}} \cdot \frac{1}{\alpha} \quad (3)$$

The volume of voids in wood:

$$V_v^{wd} = \frac{(1 - R)W}{\rho_{wd}} \frac{1}{\alpha} - \frac{(1 - R)W}{\rho_{cw}} \quad (4)$$

The volume of resin:

$$V^r = \frac{R_r \cdot W}{\rho_r} \quad (5)$$

in which ρ_f is density of polymer fluff (g/cm^3), ρ_{wd} is oven-dry density of wood (g/cm^3), ρ_r is density of the resin (g/cm^3), W is oven-dry weight of composite board ($= \rho_b \cdot V_b$), ρ_b is oven-dry board density (g/cm^3), R is fractional polymer fluff content, R_r is fractional resin level, α is compression ratio of the composites (*board density/material density*), and ρ_{cw} is density of wood cell wall (g/cm^3).

Combining Eqs. (1)–(5) after reorganization, the total voids in wood fiber/polymer fluff composites can be expressed as:

$$V_v^{TOT} = \left(\frac{1}{\rho_b} - \frac{R}{\rho_f} - \frac{R_r}{\rho_r} - \frac{(1 - R)}{\rho_{cw}}\right) \cdot W \quad (6)$$

The total voids calculated using Eq. (6) are based on the oven-dry condition. During the water absorption process, thickness swelling and linear expansion of the specimens also occur. Thickness swelling of platen-compressed composites is composed of reversible and irreversible thickness swelling. Reversible swelling is due to the hygroscopic nature of wood, which is related to bound water in the wood cell wall. Irreversible swelling is due to springback of the compressed wood and the breakage of adhesive bonds between wood elements. A major part of the irreversible thickness swelling occurs after the board is removed from the hot press (stress release). During water immersion tests, due to moisture penetration into the board, some irreversible thickness swelling also occurs. This part of the irreversible thickness swelling may create extra voids in the composites. The irreversible thickness swelling (TS_i) can be expressed as:

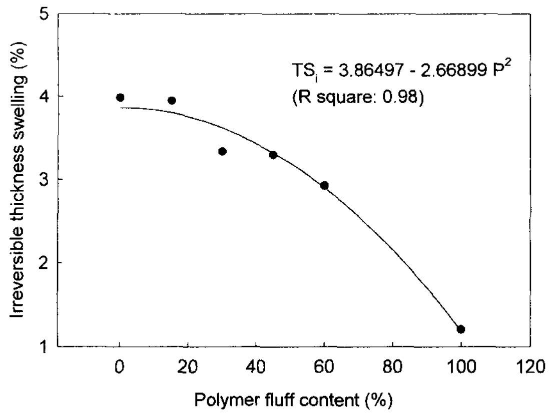

FIG. 1. Relationship between irreversible thickness swelling (TS_i) and polymer fluff content (P).

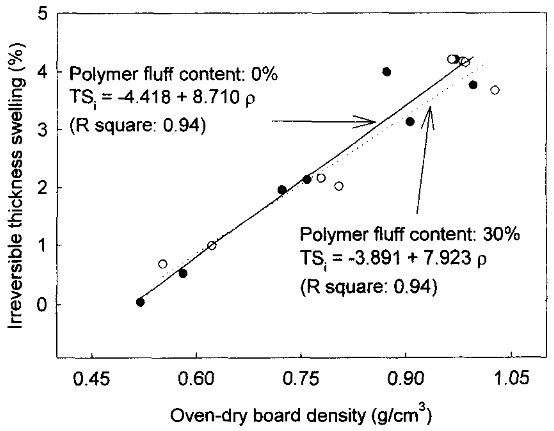

FIG. 2. Relationship between irreversible thickness swelling (TS_i) and oven-dry board density (ρ).

$$TS_i = \frac{T_o' - T_o}{T_o} \qquad (7)$$

in which T_o is initial oven-dry thickness of the specimen, T_o' is oven-dry thickness of the specimen after immersion.

Equation (7) can be reorganized as:

$$T_o' = (1 + TS_i)\cdot T_o \qquad (8)$$

In this experiment, because the specimen dimensions used in the immersion test were small (76.2 × 76.2 mm), the linear expansion that was below 1% (Shi and Wang 1997) could be neglected. Therefore, the total voids after immersion ($V_v^{TOT'}$) can be calculated as:

$$V_v^{TOT'} = (1 + TS_i)\cdot V_v^{TOT} \qquad (9)$$

Equation to calculate the maximum water absorption of wood fiber/polymer fluff composites

The total weight of water in the composites is equal to the summation of the free water in the voids of the composites, bound water in the wood cell wall, and water in polymer fluff materials. Therefore, the maximum water absorption of wood fiber/polymer fluff composites can be expressed as:

$$\begin{aligned} MWA &= \frac{W_{wt}}{W}\cdot 100 \\ &= [100\cdot \rho_{wt}\cdot V_v^{TOT'} + M_{bw}^{wd}\cdot(1 - R)\cdot W \\ &\quad + M^f\cdot R\cdot W]/W \qquad (10) \end{aligned}$$

in which W_{wt} is weight of the water in the composites, W is weight of the composite board, M^f is maximum water absorption of polymer fluff (%), and M_{bw}^{wd} is maximum bound water content of wood fiber (%).

Combining Eqs. (6), (9), and (10), the maximum water absorption of wood fiber/polymer fluff composites can be obtained as:

$$\begin{aligned} MWA &= 100\cdot\rho_{wt}(1 + TS_i) \\ &\quad \times \left(\frac{1}{\rho_b} - \frac{1}{\rho_{cw}} - \frac{1}{\rho_r}R_r - \left(\frac{1}{\rho_f} - \frac{1}{\rho_{cw}}\right)R\right) \\ &\quad + M_{bw}^{wd} - (M_{bw}^{wd} - M^f)R \qquad (11) \end{aligned}$$

In Eq. (11), irreversible thickness swelling (TS_i) in the water immersion test, maximum bound water content in wood fiber (M_{bw}^{wd}), and saturated water absorption for immersion of polymer fluff (M^f) need to be determined for calculating the MWA of the composites. Density of the board (ρ_b), fractional polymer fluff content (R), and fractional resin level (R_f) are the known experimental parameters.

EXPERIMENTAL

All the wood fiber/polymer fluff composites were processed using hardwood fibers (75% aspen, 25% other hardwood) and automobile polymer fluff particles of size of 35 mesh (0.5 mm) bonded by polymeric diphenylmethane diisocyanate (PMDI) resin. Wood fiber/polymer fluff composites at a target oven-dry board density of 0.90 and six different polymer fluff contents (0, 15, 30, 45, 60, and 100%) were used in this experiment. Composites with four different target oven-dry board densities (0.50, 0.73, 0.90, and 1.00 g/cm^3) were also processed at polymer fluff contents of 0 and 30%. All the boards were processed at a press temperature of 150°C, press time of 4 min, and resin solids level of 4%.

Water immersion tests

Water immersion tests were conducted to measure the maximum water absorption of the composites. The specimen dimensions used for the water immersion test were selected as 76.2 × 76.2 × 3.18 mm. To prevent the cycling effect on the specimens' absorbing moisture from the surroundings, the specimens were cut after hot pressing and immediately placed in a desiccator with desiccant at the bottom (relative humidity was measured as 35% by using an Omega RH 70 Digital Hygro-thermometer). All the specimens were conditioned in the above-described dried condition for over one month. To ensure the same moisture content for all the specimens, before the water immersion test, all the specimens were oven-dried and cooled in a desiccator. In the immersion test, specimens were immersed in distilled water 25 mm below the water surface. The weight and thickness of each specimen were measured before the immersion tests and after the specimens were saturated (after 100 h). Before each measurement, the water on the specimen surface was removed using a lint-free paper towel. After the water immersion tests, all the specimens were oven-dried. The irreversible thickness swelling of the wood fiber/polymer fluff composites for all the specimens used in the immersion test was calculated by measuring the initial oven-dry thickness (T_o), maximum thickness after water immersion (T_∞), and the oven-dry thickness after immersion (T'_o). Equation (7) was used to calculate the irreversible thickness swelling.

Measurement of the maximum bound water content of wood fibers ($M_{bw}{}^{wd}$)

The maximum bound water content of wood, also referred to as fiber saturation point (FSP), is defined as the moisture content at which the cell wall is saturated while the voids are empty (Siau 1995; Babiak and Kudela 1995). Many methods have been proposed to measure the FSP, for example, extrapolation of adsorption isotherms to unit relative humidity, a method based on shrinkage and swelling of wood, nonsolvent water technique, porous plate method, and nonfreezing water technique. The analysis of the methods presented shows that the FSP value can be strongly influenced by the method used. The values of FSP obtained by different methods vary in the range 13–70% (Babiak and Kudela 1995).

In this experiment, the water vapor method was used to determine the maximum bound water content. Wood fibers were placed in a desiccator 25 mm above the surface of the distilled water on the bottom until it reached equilibrium and was subsequently weighed. The relative humidity is assumed to be near 100%. Four replicates were used in the measurements. The FSP of the wood fibers used in the experiment was measured as $M_{bw}{}^{wd} = 33.52 \pm 0.92\%$.

Measurement of the saturated water absorption for immersion of polymer fluff (M^f)

Saturated water absorption of polymer fluff represents the greatest capacity of polymer fluff materials to absorb water. The following procedures were used:

1. Oven-dry the polymer fluff material and weigh the samples.
2. Soak the polymer fluff material in distilled water for over three days.
3. Drain the water in the material and place the material in a desiccator with the distilled water on the bottom (water vapor condition) for 15 days (equilibrium).
4. Weigh the samples again and calculate the water absorption of the material.

TABLE 1. *Parameters used in the model to predict the maximum water absorption from immersion experiments.*

Parameters	Value	Sources
ρ_{wt}	1.00 g/cm^3	
ρ_{wd}	0.40 g/cm^3	Note: the specific gravity of aspen is 0.36 (Bodig and Jayne 1993) at 12% moisture content. Oven-dry density was found by using the relation chart of specific gravity and moisture content (FPL 1987) which is: 0.40 g/cm^3 for aspen.
ρ_{cw}	1.53 g/cm^3	Siau 1984
ρ_r	1.24 g/cm^3	Adhesive supplier
ρ_f	1.1 g/cm^3	Estimated value from the composition of the polymer fluff
R_r	0.04	Experimental parameter
M_{bw}^{wd}	33.52 ± 0.92%	Determined by experiment
M^f	21.00 ± 5.53%	Determined by experiment

ρ_{wt}: density of water;
ρ_{wd}: density of wood;
ρ_{cw}: density of wood cell wall;
ρ_r: density of resin;
ρ_f: density of polymer fluff material;
R_r: fractional resin content;
M_{bw}^{wd}: maximum bound water content of wood;
M^f: water absorption of polymer fluff.

Four replicates were used for the measurements. The saturated water absorption of the polymer fluff was measured as: $M^f = 21.00 \pm 5.53\%$. The high measured water absorption value may be because of the hydrophilic materials contained in the polymer fluff, such as paper and wood fibers.

All the known and measured parameters for wood fiber/polymer fluff composites are summarized in Table 1.

RESULTS AND DISCUSSION

Evaluation of the effect of the irreversible thickness swelling on estimation of MWA *model*

Figure 1 shows the irreversible thickness swelling (TS_i) vs. polymer fluff content at an oven-dry board density of 0.90 g/cm^3. A quadratic relationship was found between the polymer fluff content and irreversible thickness swelling. The relationships between the irreversible thickness swelling and oven-dry board density at two polymer fluff contents (0 and 30%) are shown in Fig. 2. It is seen from Fig. 2 that the irreversible thickness swelling is also proportional to the board density. The irreversible thickness swelling is due primarily to the release of the internal stress of the compressed composites and the breakage of adhesive bonding between the elements in the composites. The higher the density of the composites, the greater the internal stress in the composites, and the higher the irreversible thickness swelling. Figure 3 shows the relationship for irreversible thickness swelling vs. polymer fluff content and oven-dry board density. It is seen from Fig. 3, there is an interaction between polymer fluff content and board density on the irreversible thickness swelling. At a higher board density ($\rho > 0.7$ g/cm^3), as the polymer fluff content increases, irreversible thickness swelling decreases because polymer fluff materials have a higher density than wood fibers. The compression ra-

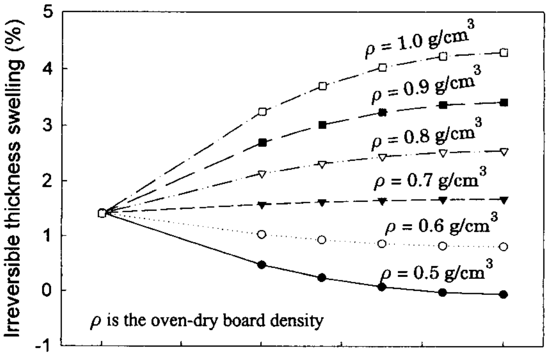

FIG. 3. Irreversible thickness swelling vs. polymer fluff content and oven-dry board density.

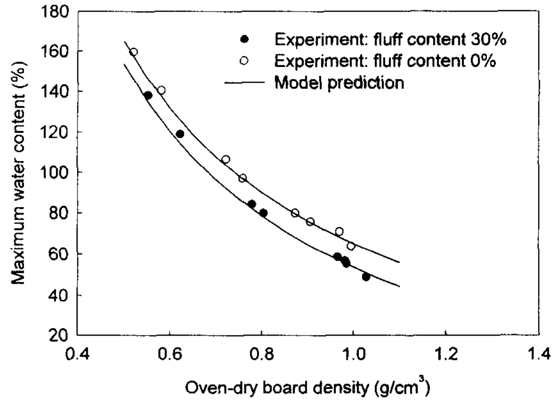

FIG. 4. Maximum water absorption of composites manufactured with different oven-dry board density.

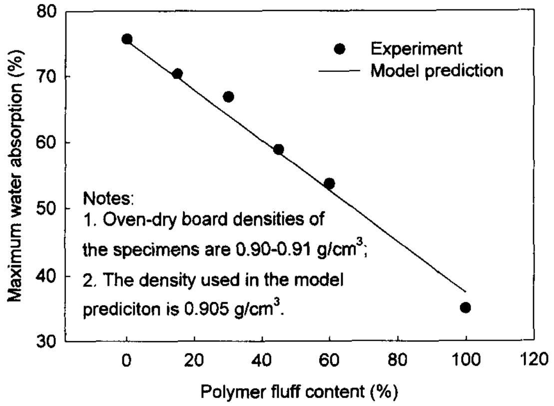

FIG. 5. Maximum water absorption of composites manufactured with different polymer fluff contents.

tio of the higher fluff content composites is lower. Therefore, the internal stress in higher polymer fluff content composites is lower, which therefore lowers the irreversible thickness swelling. However, when the board density is too low ($\rho < 0.7$ g/cm^3), incorporation of the higher density polymer fluff will have higher irreversible thickness swelling. This is because the compression ratio is too low to form enough intimate contacts between the elements. A lower interfacial bonding between the elements will lead to more bonding breakages of the composites during the water immersion tests, and therefore induce a higher irreversible thickness swelling.

Based on this relationships shown in Fig. 3, an equation is established showing the irreversible thickness swelling as a function of both fractional polymer fluff content (R) and oven-dry board density (ρ_b):

$$TS_i = (-4.418 + 5.856R^2) + (8.710 - 8.744R^2)\rho_b \quad (12)$$

In this experiment, it is shown that the irreversible thickness swelling of the specimens after water immersion was only in the range of 0.04–4.2%. Because of the small portion of irreversible thickness swelling, the contribution of the irreversible thickness swelling portion to the total voids in the composites is very small.

Evaluation of the MWA *models*

By applying the values of all the parameters in Table 1 to Eq. (11), the expression of the *MWA* model is:

$$MWA = 100\cdot(1 + TS_i) \times \left(\frac{1}{\rho_b} - 0.6859 - 0.2555R\right) + 0.3352 - 0.1252R \quad (13)$$

If the TS_i is neglected for the maximum water absorption estimation, the *MWA* model can be simplified by as:

$$MWA = \left(\frac{1}{\rho_b} - 0.3807R - 0.3507\right) \times 100 \quad (14)$$

As is shown in Eq. (14), the maximum water absorption of wood fiber/polymer fluff composites has a reciprocal relationship with the oven-dry board density and a linear relationship with the polymer fluff content.

Table 2 shows the maximum thickness swelling values of wood fiber/polymer fluff composites with different polymer fluff contents and board specific gravities in the water immersion test both from experimental measurements and the estimation of the developed *MWA* model. The comparison of the model considering the irreversible thickness swelling (Eq. (13)) and the simplified model (Eq. (14))

TABLE 2. *Results of the maximum water absorption of wood fiber/polymer fluff composites measured from immersion tests and estimated by the* MWA *model.*

		Maximum water absorption (%)				
			Model considering irreversible thickness swelling (Eq. (13))		Simplified model (Eq. (14))	
Polymer fluff content (%)	Oven-dry board density (g/cm^3)	Measured value in experiment	Estimation value	Error (%)	Estimation value	Error (%)
0	0.520	159.64	157.37	1.42	157.23	1.51
	0.580	140.35	137.89	1.75	137.23	2.22
	0.722	106.59	104.70	1.77	103.39	3.00
	0.758	97.04	98.21	1.21	96.83	0.22
	0.873	79.89	80.97	1.35	79.51	0.48
	0.906	75.72	76.77	1.39	75.32	0.52
	0.970	70.75	69.43	1.87	68.03	3.84
	0.996	63.75	66.74	4.70	65.39	−2.58
15	0.904	70.34	71.15	1.16	69.85	0.69
30	0.552	138.21	135.31	2.10	134.80	2.47
	0.622	118.91	115.09	3.21	114.21	3.95
	0.778	84.17	83.18	1.17	81.99	2.59
	0.804	79.82	79.12	0.88	77.92	2.38
	0.965	58.51	58.19	0.54	57.16	2.30
	0.981	56.62	56.45	0.29	55.46	2.05
	0.984	55.33	56.11	1.40	55.12	0.39
	1.027	49.05	51.81	5.63	50.92	−3.80
45	0.911	58.75	58.54	0.36	57.62	1.92
60	0.900	53.54	53.92	0.73	53.20	0.64
100	0.907	34.91	37.30	6.84	37.08	−6.21

is also shown in Table 2. It can be seen in Table 2 that the prediction accuracies from Eq. (13) and (14) are similar. Therefore, the simplified *MWA* model neglecting the irreversible thickness swelling can be used directly to predict the maximum water absorption of wood/polymer fluff composites. The developed *MWA* model provides a very good prediction with over 95% accuracy for most of the specimens.

Figure 4 shows the maximum water absorption as a function of oven-dry board density of both neat wood fiberboard and wood fiber/polymer fluff composites at a polymer fluff content of 30%. It can be seen from Fig. 4 that as the board density increases, the maximum water absorption decreases, but the relationship is not linear. The *MWA* model has a good fit with the experimental data. The maximum water absorption as a function of the polymer fluff content is also depicted in Fig. 5; it can be seen that as the polymer fluff content increases, the maximum water absorption decreases linearly. Therefore, the higher polymer fluff content and board density will give a better water resistance for wood fiber/polymer fluff composites.

CONCLUSIONS

A *MWA* model was developed to estimate the maximum water absorption of wood fiber/polymer fluff composites based on the calculation of the total voids contained in the composites. Composites with six polymer fluff contents and four target board densities were used to conduct water immersion tests to evaluate the model. The effects of polymer fluff content and board density were studied.

Results obtained in this study indicate that maximum water absorption of wood fiber/polymer fluff composites decreases as the