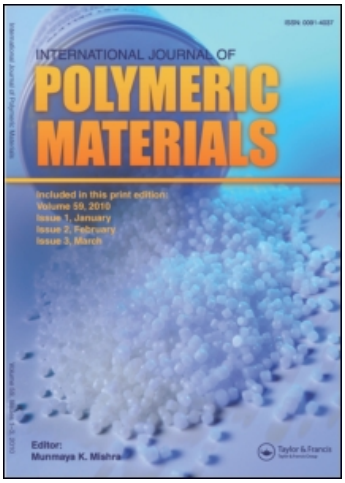

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Improvements of Fibreboard Properties through Fibre Activation with Silane

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Thermomechanical pulp (MDF fibre) was reacted with γ-aminopropyltriethoxysilane. The adducts formed was mixed with urea formaldehyde (UF) resin followed by hot press. Well-conformed boards were produced by the process. Modified boards showed significant improvement in modulus of elasticity, modulus of rupture and water absorption. The total swelling performance of the modified boards was attributed to predominantly by irreversible swelling.

KEY WORDS Fibreboard-chemical modification-γ aminopropyltriethoxysilane-rubberwood-plasticization.

INTRODUCTION

Various works have been done on the chemical modification of solid wood, and it has been reviewed extensively by Rowell[1] and Banks.[2] The success of the modification on solid wood, especially in improving the dimensional stability, enables the technique to be applied on wood composites. More recently, interest has developed in the use of a di- or polyfunctional reagents so that some of the functional groups react with cell wall components and remain attached to the cell wall surface, while, unreacted groups are available for subsequent secondary reactions.[3,4] Rozman *et al.*,[5] showed that mechanical strength and dimensional stability of medium density fibreboard (MDF) could be enhanced through fibre activation and subsequent copolymerization with vinyl monomer. It is well known that chemical modification of wood improves dimensional stability as well as mechanical properties. These involve reacting wood with reactive chemicals at elevated temperatures in solution form. Rozman *et al.*[6] showed that solid rubberwood sample treated with γ-methacryloxypropyltrimethoxysilane displayed better mechanical strength and dimensional stability than the untreated samples. The method which employed very mild condition involved introduction of a coupling agent which has dual functionality which would establish a bridge between wood component on the one hand and polymeric bulking agents on the other. The purpose of this work was to chemically modify wood fibre

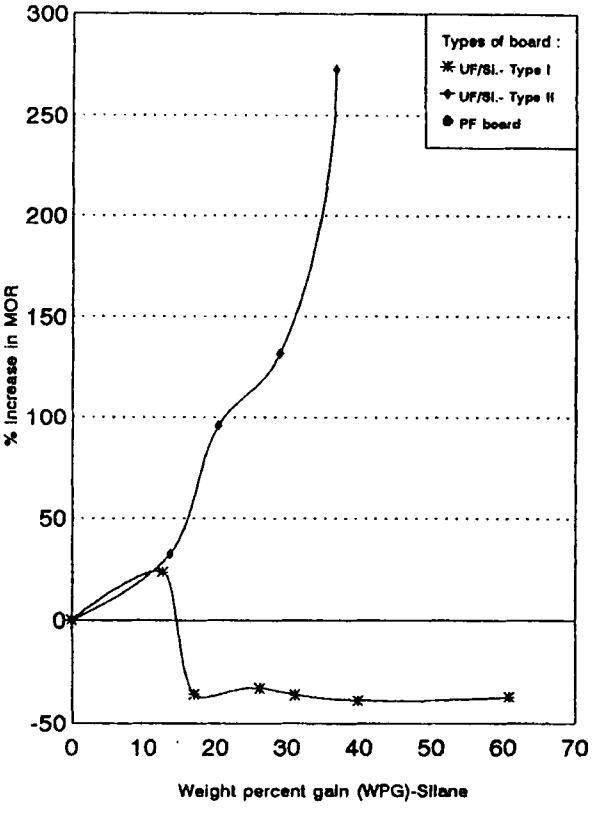

FIGURE 1 % Increase in MOR.

with γ-aminopropyltriethoxysilane and followed by blending with urea formaldehyde (UF) resin.

Reaction of silane represented by Y—R—SiX_3, with the hydroxyl groups of wood could be accomplished by the —SiX_3 portion of the molecule through its hydrolysis product, $Si(OH)_3$ (see Figure 9). The Y unit is an amino group as in the case of γ-aminopropyltriethoxysilane, which could undergo reaction with methylol groups of UF resin. The hypothetical chemical structure of wood-silane coupling agent in the interfacial area can be presented as in Figure 10. Through this mechanism, it was to be hoped that this system would impart some enhanced properties to the composite.

EXPERIMENTAL PROCEDURE

Thermomechanical pulp fibre produced from rubberwood and refined at high temperature and pressure suitable for medium-density fibreboard (MDF) production was acquired from the Hume Fibreboard Sdn. Bhd., Nilai, Malaysia. The fibre was used as delivered with the moisture content about 10%. The fibres were soaked with silane/methanol solution for 3 hours at room temperature. The silane used was γ-aminopropyltriethoxysilane. The solution was drained from the fibres and the fibres

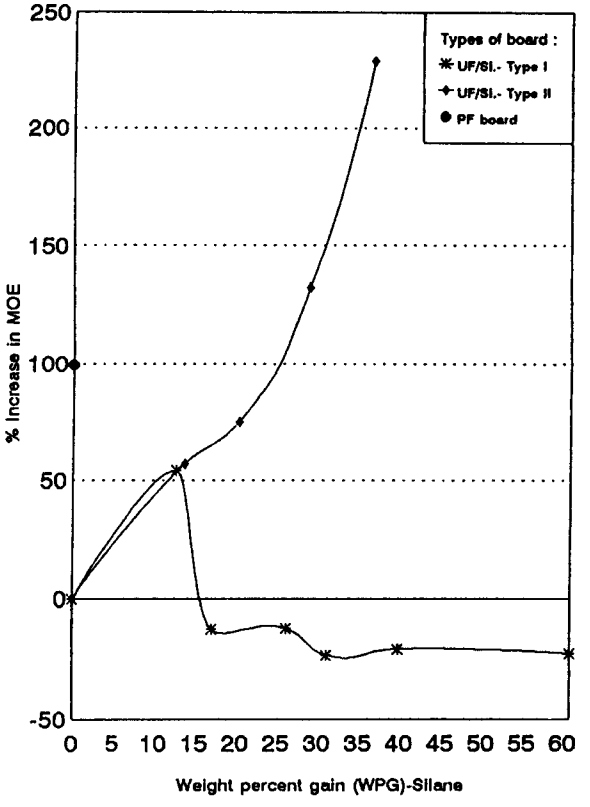

FIGURE 2 % Increase in MOE.

were placed in an oven for curing process at 110°C for 5 hours. The fibres were then cooled in a desiccator and the weight percent gain (WPG) was measured (based on ovendry weight). The treated fibres were mixed thoroughly with 25% solid content UF solution (12% based on dry weight of untreated fibre) and 2% catalyst (based on UF weight). The mixture was laid as mat in a round stainless steel mould and heated under pressure at 150°C for 15 minutes. Two types of board were produced, Type I, boards with similar density, 750–780 kg/m^3 and type II, boards with similar fibre content (the density varies accordingly with the level of modification). Control boards were produced by hand-mixing 25% solid content UF solution (12% based on dry weight of untreated fibres) and 2% catalyst (based on UF weight). For the purpose of comparison, phenol-formaldehyde (PF) boards were also produced with the same manner as UF boards.

Six rectangular samples of dimensions approximately 9.0 × 1.5 × 0.50 cm (length × width × thickness) were cut from each board to be used for the static bending test. Internal bonding (IB) test was carried out using 9 samples of dimensions approximately 3.0 × 3.0 × 0.5 cm. Eight rectangular samples of dimensions approximately 3 × 1.5 × 0.5 cm (length × width × thickness) were used for the dimensional stability test. All test samples were conditioned at 23 ± 2°C and 55 ± 5% relative humidity until constant mass was achieved, before being tested.

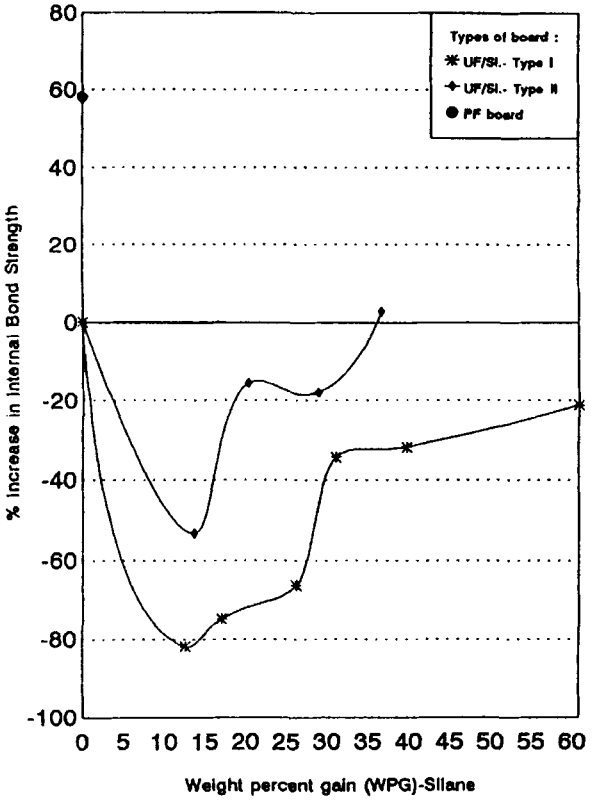

FIGURE 3 % Increase in internal bond strength.

For the dimensional stability test, the conditioned samples were weighed and dimensions were measured before being immersed in cold water at 25°C for 24 h. On removal, the samples were allowed to drain for a few minutes, then surface-dried with absorbent paper, and the mass and dimensions of each sample were measured. The samples were then dried in an oven at 60°C for 5 hours and reconditioned (23 ± 2°C and 55 ± 5% relative humidity). After the samples were conditioned, the weight and dimensions were measured.

The modulus of rupture (MOR) was measured as follows:

$$\text{MOR} = \frac{3 \times W \times L}{2 \times b \times d^2}$$

where W is the ultimate failure load; L, the span between centres of support; b, the mean width of the sample; and d, the mean thickness of the sample.

The modulus of elasticity (MOE) was measured as follows:

$$\text{MOE} = \frac{L^3 \times \Delta W}{4 \times b \times d^3 \times \Delta S}$$

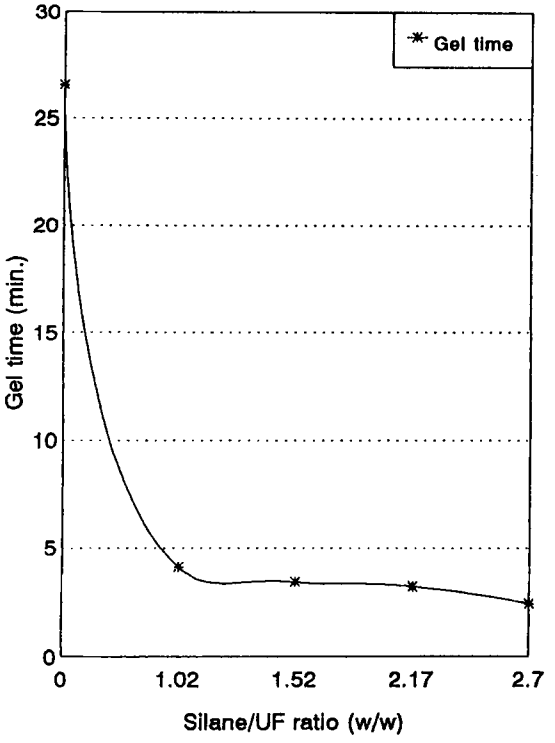

FIGURE 4 Gel time vs. silane/UF ratio.

where L is the span between the centres of supports; ΔW, the increment in load; b, the mean width of the sample; d, the mean thickness; and ΔS, the increment in deflection corresponding to W.

Water absorption (WA) was measured as follows:

$$\text{WA} = \frac{M_2 - M_1}{M_1} \times 100$$

M_2 is the mass of the sample after immersion (g), M_1 is the mass of the same before immersion (g).

Thickness swelling was measured as follows:

$$\text{Reversible swelling} = \frac{t_w - t_e}{t_0}$$

$$\text{Irreversible swelling} = \frac{t_e - t_0}{t_0}$$

t_0 = initial thickness of the sample (conditioned), t_w = thickness of wetted sample, t_e = thickness of the sample after soaking and reconditioned at 55% RH and 23°C.

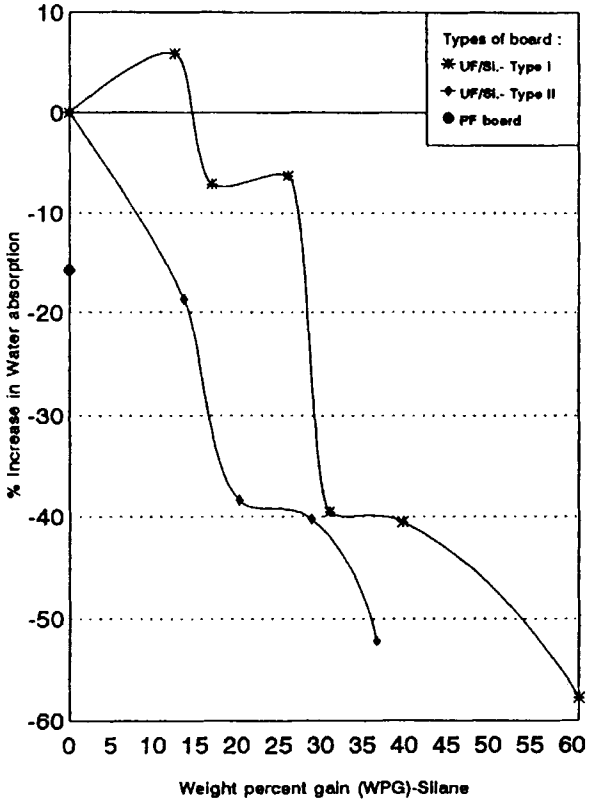

FIGURE 5 % Increase in water absorption.

$$\% \text{ Increase in properties (\%) over control sample} = \frac{P - Po}{Po} \times 100$$

P = properties of modified sample, Po = properties of control sample.

Gel time measurements were carried out by physical observation. Silane/UF solution were prepared with different ratio of silane and UF (weight/weight) and in the presence of catalyst, similar to the ones present in the modified boards.

RESULTS AND DISCUSSION

The results on the improvements in MOR as a result of treatment over control are presented in Figure 1. The results showed that the % increase in MOR for type II boards increased significantly. The improvement increased accordingly with the increase of the level of modification. In fact, type II samples with degree of modification of about 40 WPG showed significantly higher improvement in MOR than PF boards. There is a possibility that silane which present in the board could have resulted in greater plasticization of the fibres and in turn resulted in greater densification of the board. Thus, the results show that as the level of modification increases

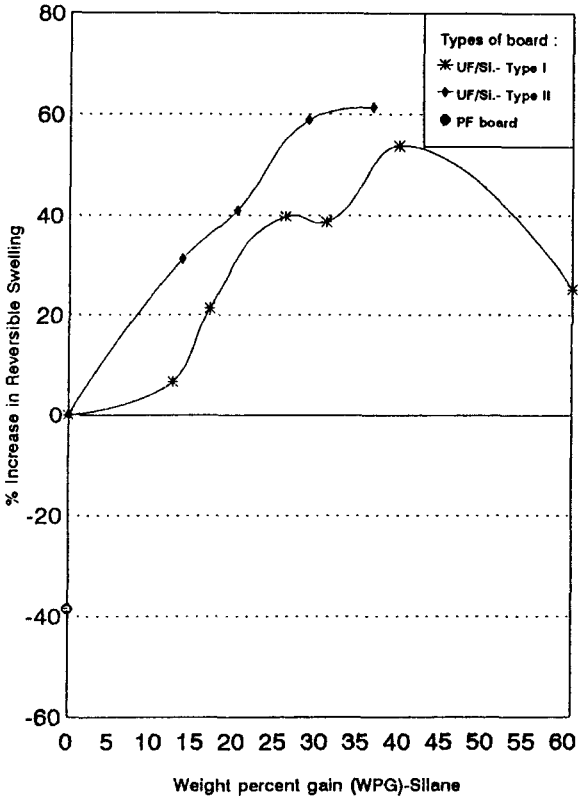

FIGURE 6 % Increase in reversible swelling.

with concomitant increase in plasticization and densification, there is corresponding increase in the % increase in MOR for type II boards. However, there was little improvement in MOR for type I boards with modification to about 13 WPG. Further modification resulted in the decrease of the modulus to the level below that displayed by the control boards. Though the fibres were modified to the same degree for these two types of board, only type II boards show a corresponding increase in MOR. This may due to the fact that, although the WPG is increased, corresponding increase in MOR could not be achieved due to lesser fibre content in the type I as compared to type II boards. This shows that the modification per se is not sufficient, the fibre content also plays a predominant role in determining the improvement in MOR of the product.

The percentage increase in MOE results which is shown in Figure 2 also display trend similar to that shown by MOR results. For type II boards, all modified boards showed significantly higher stiffness than the control boards. Boards with 40 WPG again as in the case of MOR showed greater stiffness than the PF boards. Type I boards showed greater stiffness than the control boards at lower modification level, however, upon further modification there was no improvement in MOE compared to the control boards. These results again confirm the importance of fibre content vis-a-vis modification as discussed earlier in the case of MOR.

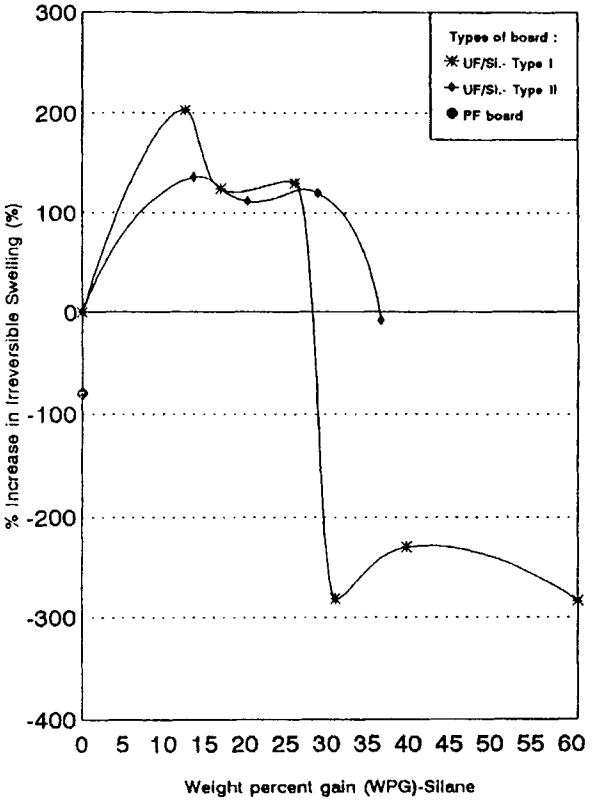

FIGURE 7 % Increase in irreversible swelling.

Results of % increase in internal bonding over controls (Figure 3) display different trend from those shown by MOR and MOE results. For type II boards at lower modification, i.e., at about 13 WPG, the internal bonding strength was significantly lower than the ones shown by control boards. Similar trend was also shown by type I boards which showed significant decrease in internal bond strength for modification from 10 to 30 WPG. However, as the modification was further increased, the internal bonding was accordingly increased to the extent similar (no difference statistically) to the controls. Thus, these results implied that at lower modification, silane somehow interfered with the cure of UF in the board. This might be due to accelerated cure of UF in the presence of silane and the consequent effect of precure. Experiments were conducted to examine whether the silane reduced the gel time of UF and it was from that it had in fact done so, as can be seen in Figure 4, where gel time was significantly reduced compared to UF with normal catalyst alone. At lower modification, a dominant role was played by the silane in inducing the precure of UF adversely affecting the IB. Under these conditions the beneficial effect of plasticization to achieve quick and efficient consolidation before reaching the cure temperature and thereby averting the risk of precure was not achieved. As the modification increased, on the other hand, it is probable that increased plasticization could have

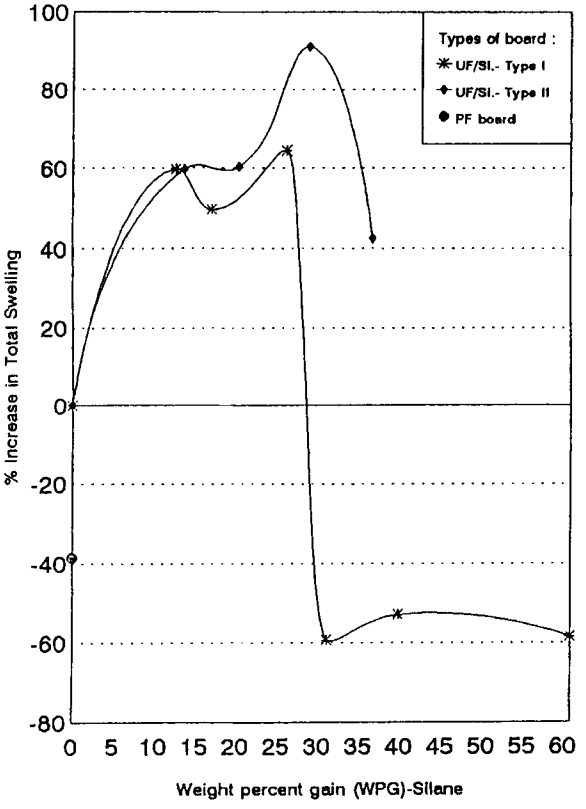

FIGURE 8 % Increase in total swelling.

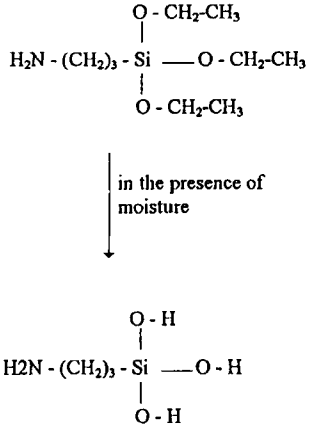

FIGURE 9 Hydrolysis of silane.

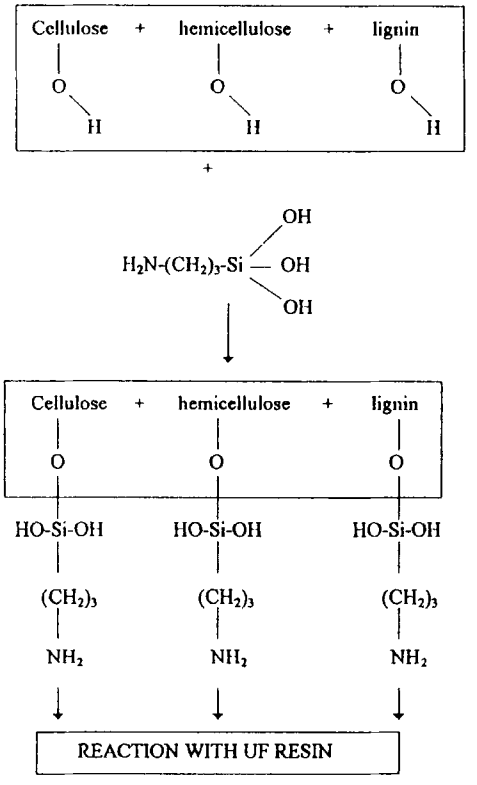

FIGURE 10 Hypothetical reaction of wood and silane.

facilitated quicker consolidation to the facial density thereby eliminating the risk of precure caused by radiant heat from the platens during the hot press operation.

The percentage increase in water absorption results are presented in Figure 5. The results showed that as the WPG was increased, water absorption showed a decreasing trend both for type I and II. Type II boards showed a higher reduction in water absorption compared to type I boards. Almost all type II boards modified at different level of modification showed significant reduction compared to the untreated UF boards. Type I boards also showed significant reduction except for the ones modified to modification level lower than 30 WPG.

Results of thickness swelling are presented in Figure 6. Generally, both types of boards showed no improvement in reversible swelling. Since the reversible swelling relates to the hygroscopicity of the sample,[7] it showed that the modification did not instill additional hydrophobicity in the composites to the extent lower than the controls. Type II boards showed greater increase in reversible swelling than type I and this might due to the fact that there was more fibre in the former which resulted in more water entered the cell wall and caused greater swelling.

Type II boards showed no improvement in the results of irreversible swelling as compared with the controls (Figure 7). For type I boards only those with higher modification degree of 31, 40 and 60 WPG showed significant reduction in irreversible swelling to the extent that the thickness shrank lower than the initial thick-

ness. This might be due to the fact that, at higher modification and in the presence of water more unreacted silanol groups would be available for the formation of homopolymer and caused shrinkage. This effect was also shown for the ones modified to 37 WPG in type II, though the effect was not to the same extent as shown by the type I samples. Since type II boards contained more fibre and more compact, the degree of shrinkage was lower compared to type I boards where the fibre content was lower and more porous. The results of total swelling (Figure 8), which was the summation of reversible and irreversible swelling showed that irreversible swelling contributed more than reversible swelling towards total swelling of the samples. Thus, these results indicated that polymerization of silane occurred mainly outside the cell wall, i.e., in the lumen and in pores. Although, the matrix formed was able to retard water absorption significantly better than the controls, it seemed that the absorbed water after a period of time was able to enter into the cell wall and caused swelling (reversible swelling) to the same or greater extent than controls. Instead of reacting with the cell wall and act as coupling agent with UF, the presence of silane caused premature curing of UF (as shown by gel time results) thus, interrupted the normal cure reaction of UF with wood. Thus, this interruption which was to be believed to cause reduction of internal bonding, might also be the cause for the greater irreversible swelling which occurred at about the same range of modification level.

CONCLUSIONS

The work demonstrates that by chemically modifying the fibre with silane, boards with enhanced properties of MOR, MOE and water absorption could be produced. The results indicate that the silane system behaves like wax where the retardation of water absorption was enhanced. The total swelling of the modified samples was contributed mainly by the irreversible swelling.

Acknowledgment

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