Coextruded Polyethylene and Wood-Flour Composite: Effect of Shell Thickness, Wood Loading, and Core Quality

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ABSTRACT: Coextruded recycled polyethylene and wood-flour composites with core–shell structure were manufactured using a pilot-scale coextrusion line. The influence of wood loadings and thickness of the shell layer and core quality on mechanical and water absorption properties of the composites were investigated. Core–shell structured profile can significantly improve flexural and impact strengths of composites especially when a relatively weak core was used. However, the coextruded profile with unreinforced shell may have a reduced modulus when a strong core was used. The shell layer also protected coextruded composites from long-term moisture uptaking, leading to improved dimensional stability compared with the corresponding un-coextruded controls. When the shell thickness was fixed, less wood loading in the shell layer did not cause obvious flexural modulus and dimension change but improved impact strength and water resistance of the coextruded composites. When wood loading in the shell layer was fixed, increased shell thickness improved impact strength but affected modulus negatively. Thickened shell layer helped reduce water uptaking but did not change dimensional stability of coextruded composites remarkably. Overall enhancement of composite strength was more pronounced for the weaker core system. Thus, the coextrusion technology can be used to achieve acceptable composite properties even with a relatively weak core system—offering an approach to use recycled, low quality plastic-fiber blends in the core layer. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 118: 3594–3601, 2010

Key words: coextrusion; mechanical properties; recycling

INTRODUCTION

Coextrusion is a process in which two or more polymer materials are extruded and converged upon a single feedblock or die to form a single multilayer structure (e.g., flat, annular, or core–shell profiles).[1–3] It has become one of the most advanced plastic processing technologies in creating multilayer composites with different complementary layer characteristics and in making properties of final products highly "tunable." For example, target composite properties such as oxygen and moisture barrier, shading and insulation, and mechanical properties can be adhibited by incorporating one or more layers with target properties.[4–7] In addition, coextrusion can significantly reduce material and production costs, and help recycle the used material.[1–3]

Because of those advantages, coextrusion technology is widely used in plastic composite films and sheets,[8,9] and coated tubes and pipes.[10–12] However, the potential of applying coextrusion technology to wood plastic composites (WPCs) is just being recognized,[13] and the work focused on this aspect is very limited. Few published studies include virgin polyvinyl chloride (PVC)[14] and high density polyethylene (HDPE)[15,16] based WPC with a core–shell structure. It was reported that coextruded PVC-wood composite had reduced moisture uptake rate, increased flexural strength, and decreased flexural modulus compared with regular PVC-wood control samples.[14] Coextruded HDPE-wood composites also had improved moisture resistance and color stability. However, mechanical performance of the composite system was less understood from the published studies.[15,16] Several commercial coextruded PVC-wood composites are currently available (e.g., Crystal White™ Railing from Louisiana-Pacific Corporation and Celucor™ from Royal Group Technologies). However, there is still no report yet on commercialized coextruded PE-wood composite products. As PE-based WPC has the largest market share among various WPC products,[17] the

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TABLE I
Formulations and Material List for Core and Shell Layers

Layer	Material and contents	Provider
Core A	Recycled WPC A (HDPE:LDPE:wood:additives = 20 : 20 : 55 : 5)	Commercial WPC blend
Core B	Recycled WPC B (HDPE:LDPE:wood:additives = 30 : 10 : 55 : 5)	Commercial WPC blend
Shell A	Virgin HDPE (HGB 0760): 100%	ExxonMobil Chemical Co., TX
Shell B	Pine wood flour (60 mesh): 5–35wt %	American Wood Fibers
	PE-*g*-MA (Epolene^{TM} G2608): 6 wt % of wood	Eastman Chemical Co., TN
	Virgin HDPE (HGB 0760): 62.9–94.7 wt %	ExxonMobil Chemical Co., TX

performance of its coextruded composites needs to be investigated.

In recent years, most commercial WPCs use recycled, commingled plastic blends as their base resin system. It is an environmentally beneficial and economically competitive approach for the WPC industry. However, the utilization of multicomponent commingled, recycled plastics was a significant challenge in maintaining target composite properties. It was due to the incompatibility among different plastics which limits their recyclability and requires additional treatment technologies.[18,19] In addition, recycling used WPC material poses a significant industrial problem. Recycled WPC material often suffers large strength loss due to degradation from moisture and weathering[20,21] which limits its direct use in making new WPC. Coextrusion with its ability to create a layered structure may offer a practical solution to the use of recycled WPC material.

In this study, we simulated industrial manufacturing process and produced coextruded WPC using recycled PE-based WPC blends as raw material. The objectives of the study were (1) to develop a pilot-scale coextrusion system for manufacturing coextruded WPC, and (2) to investigate the influences of wood loading and thickness of the shell layer and core quality on mechanical and water absorption (WA) properties of the resultant composites.

EXPERIMENTAL

Raw material preparation

Raw materials and the formulation used in this study are listed in Table I. Material for Core A and Core B were commercial PE-based blends with pine fibers supplied by a local WPC manufacturer. Core A contained more low-density polyethylene (LDPE) than Core B. Thus, Core A was significantly weaker than Core B. The 5% additives include coupling agent (polyethylene-grafted maleic anhydride, PE-*g*-MA), lubricant, and colorant. Even though the exact content of each component is not known, the blend represented typical commercial WPC formulation. The virgin HDPE for Shell A was used as received. Materials used for Shell B were compounded before coextrusion by a Leistritz Micro-27 co-rotating parallel twin-screw extruder (Leistritz Corporation, Allendale, NJ) with temperature controlled at 170°C in all zones. The extrudates were air-cooled and then pelletized. The pine fiber loading in Shell B were 5, 15, 25, and 35% based on the total shell weight.

Coextrusion system and composite manufacture

A pilot-scale coextrusion system (Figure 1) was developed for this work. In this system, the Leistritz extruder (length-to-diameter ratio of 40 : 1) was used to produce the core blends. A Brabender 32 mm conical twin-screw extruder (Brabender Instruments Inc., South Hackensack, NJ) with a length-to-diameter ratio of 13 : 1 was used for the shell layer extrusion. The Leistritz machine was equipped with two weight-in-loss feeders, whereas the Brabender machine had two gravimetric feeders. Each extruder was controlled by an independent computer with commercial software. A specially-designed coextrusion die was used to produce a solid profile with a target cross-section area of 13 mm × 50 mm and

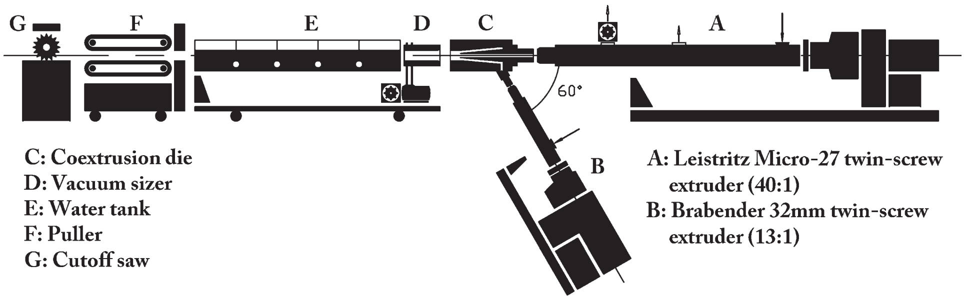

Figure 1 Schematic diagram of the pilot-scale coextrusion system.

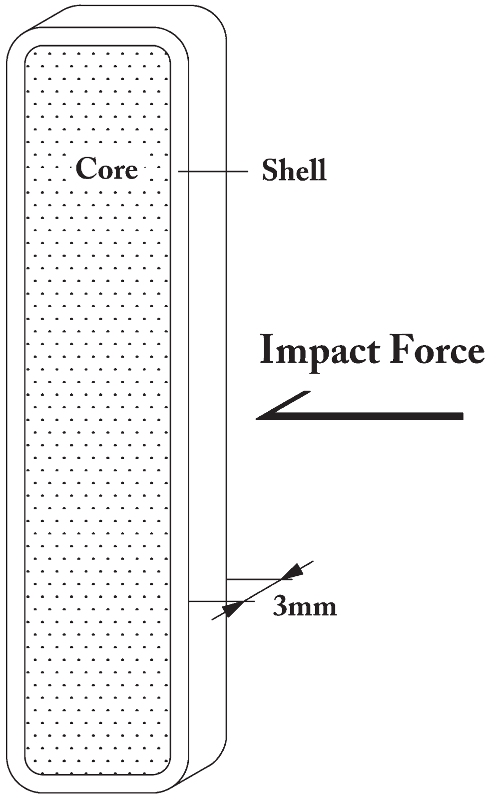

Figure 2 Impact strength testing sample.

shell thickness up to 1.5 mm. A specially-designed water-cooled vacuum sizer was used to maintain the target dimension of the coextruded composites. The coextruded composites were cooled through a 2 m water tank with controlled water spray. The extrusion speed was maintained by a speed-controlled puller (Al-Be Industries, Fullerton, CA).

During coextrusion process, the screw rotational speed of the Leistritz extruder for the core layer was kept constant. The thickness change of the shell layer was done by adjusting screw rotational speed of the Brabender extruder. Processing temperatures in the main processing zones of the Leistritz extruder were controlled between 165 and 175°C and were kept the same for both Core A and Core B formulations. Processing temperatures of the Brabender extruder varied with different shell materials (i.e., 150 and 165°C for Shell A and Shell B, respectively). The un-coextruded control samples (core only) were also extruded using the same die.

Characterization

All specimens were conditioned for 72 h at a temperature of 23 ± 2°C and a relative humidity of 50 ± 5% before sampling and characterization. A high-resolution digital camera was used to take photographs of the cross sections of coextruded samples. The thickness of both shell and core layers in each sample was then measured from the digital photographs with the aid of Image-Pro Plus 6 (Media Cybernetics, Inc.) software. A sample from each composite system was randomly selected and impact-fractured. A SEM (Hitachi S-3600N, Japan) was used to analyze the morphology of the fractured surfaces coated with gold before observation. The acceleration voltage used was 15 KV.

Three-point flexural test was carried out using an Instron 5582 testing machine (Instron Co, Norwood, MA). A crosshead speed of 6mm/min was used according to the ASTM D7031-2004. Izod impact strength without notching was tested using a Tinius Olsen Mode 1892 impact tester (Tinius Olsen Inc., Horsham, PA) following the ASTM D256. The samples with 3 mm along the extrusion direction were obtained by cross-cutting the extruded profiles. The impact force was perpendicular to the extrusion direction (Figure 2). At least five replicates were used for each test.

WA and thickness swelling (TS) tests were conducted under water soaking condition specified in the ASTM D7031-2004. Nominal length, width, and thickness of samples were 25.4 mm, 50 mm, and 12 mm, respectively. All sample edges were unsealed. Thickness measurements were done at the center point of each sample. The WA and TS were calculated using the following equations:

$$\mathrm{WA} = \left(\frac{\mathrm{M_t} - \mathrm{M_0}}{\mathrm{M_0}}\right) \times 100\% \tag{1}$$

$$\mathrm{TS} = \left(\frac{\mathrm{H_t} - \mathrm{H_0}}{\mathrm{H_0}}\right) \times 100\% \tag{2}$$

where M_t and M_0 were sample mass at time t and initial time; H_t and H_0 were sample thickness at time t and initial time. Three replicates were used to obtain average value for each test.

RESULTS AND DISCUSSION

Morphology of coextruded WPC

Typical cross-section images of coextruded composites are shown in Figure 3. The shell layer had uniform thickness on all four sides and the core was fully encapsulated by the shell layer. Vacuum sizing played a significant role in maintaining surface quality and target core–shell thickness ratio. The higher wood fiber loading in the core layer can be clearly seen from the SEM micrographs (Figure 4). Because the same resin system (i.e., PE) was used for both core and shell layers, there was good interfacial bonding as observed from the SEM micrographs (Figure 4).

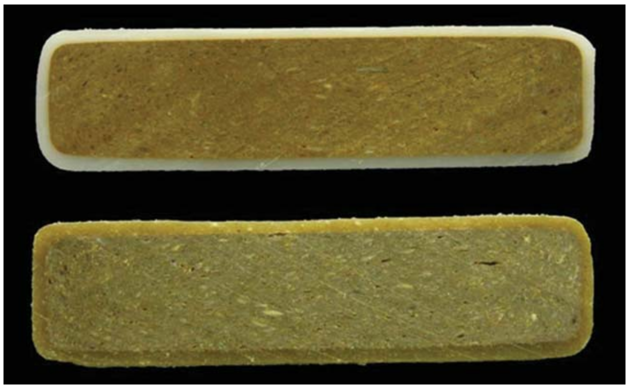

Figure 3 Cross sections of coextruded core–shell WPC. Top: pure HDPE shell; bottom: wood-filled HDPE shell. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

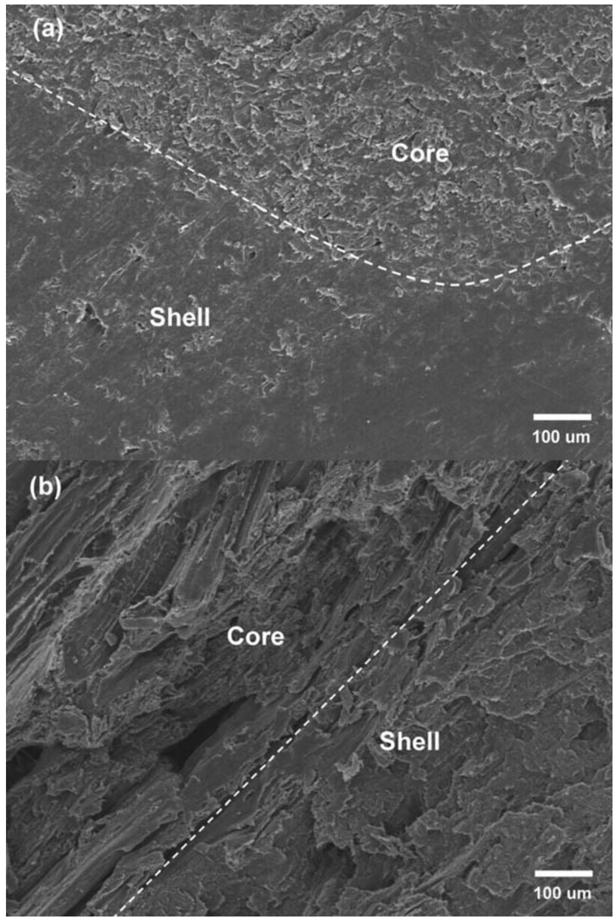

Figure 4 SEM micrographs of coextruded WPC: (a) sawn surface and (b) impact fractured surface. Wood loading in core and shell were 55% and 15%, respectively.

Effect of wood loading in shell layer

The effect of four different wood loadings (i.e., 5, 15, 25, and 35%) in the shell layer on mechanical properties of coextruded composites is summarized in Table II. The shell thickness was fixed at 1 mm in all cases. Un-coextruded (core only) composites were used as control group. It was observed that coextruded core–shell structure significantly improved flexural strength of composites. There were up to 94% and 28% increases, respectively, for Core A and Core B composite systems, compared with the corresponding control samples. The shell layer restricted the deformation of the core layer and consequent generation of cracks during a three-point bending test. Further, the shell layer had higher tensile strength than the core layer due to smaller wood content in the shell layer.[22] The presence of the shell layer effectively prevented cracks from propagating towards the center of the specimen. Therefore, flexural strengths of the final coextruded composites were improved. For the same reason, coextruded composites with comparatively

TABLE II
Summary of Mechanical Properties Effected by Wood Loading in Shell Layer [a]

Wood loading in shell layer (wt %)	Flexural strength (MPa)	Flexural modulus (GPa)	Impact strength (KJ/m^2)
Core A (Recycled WPC A)			
No shell	14.1(0.8)D	1.75(0.17)A	1.98(0.25)D
Shell A	21.1(0.8)B	1.31(0.05)B	5.74(0.68)A
5	26.0(0.5)A	1.74(0.06)A	5.24(0.74)A
15	27.4(0.1)A	1.89(0.11)A	4.02(0.59)B
25	22.1(0.3)B	1.69(0.04)A	2.91(0.48)C
35	16.5(1.6)C	1.70(0.21)A	2.85(0.38)C
Core B (Recycled WPC B)			
No shell	26.0(1.5)B	2.13(0.17)A	2.56(0.16)C
Shell A	–	–	–
5	32.2(1.9)A	1.77(0.17)B	5.41(0.60)A
15	33.4(0.4)A	1.89(0.09)B	4.88(0.38)A
25	32.1(1.5)A	1.82(0.08)B	3.51(0.34)B
35	28.0(1.3)B	1.78(0.14)B	3.56(0.50)B

[a] Mean values with the same capital letter for each category are not significantly different at the 5% significance level; numbers in the parenthesis are standard deviation based on five specimens.

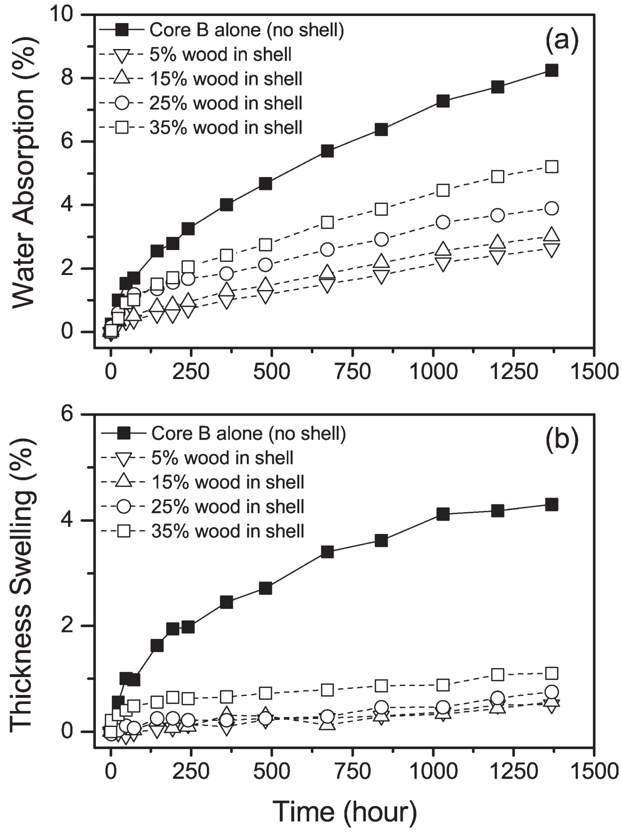

Figure 5 Effect of wood loading in shell layer on water absorption and thickness swelling properties of coextruded composites.

less wood content in the shell layer (5 or 15%) showed better flexural strength than those with more wood (35%). This trend can be seen in both core systems. When wood loading in the shell layer was fixed, the coextruded composites based on the weak core (Core A) gained more improvement in flexural strength than those based on the strong core (Core B) did. Moreover, the strength values of the former seemed commercially acceptable when wood loading in shell layer was not greater than 25%, which implied that a strong composite can be produced even with a relatively weak core layer using coextrusion technology. Thus, coextrusion technology makes it possible to use recycled, low-quality material in the core layer and a relatively strong shell for achieving targeted overall composite properties.

Wood loading in the shell layer seemed not significantly affect flexural modulus of coextruded composites based on the statistical analysis. This trend occurred in both core systems. This is possibly because that shell layer was only minor portion of the entire system. Flexural modulus of the composite usually reflects intrinsic properties of the whole material. Another observation was that, compared with un-coextruded specimen (core only), the addition of shell layer decreased modulus of those coextruded composite containing stronger core (Core B). It implied the negative influence of coextruded shell layer on stiffness of composite. The reason might be associated with relatively lower wood content and consequent less stiffness in shell layer with respect to the core layer. This observation was in agreement with that obtained by Matuana and Jin[14] in PVC-based coextruded WPC research. Therefore, maintaining stiffness of coextruded composites needs special attention in future studies.

For impact strength of coextruded composites, coextruded shell layer showed obvious protection effect for the core layer. Those results were expected because shell layer contained more PE which tended to be more flexible and tougher. It also helped absorb impact energy and prevent crack propagation during impact fracturing. Therefore, the protection effect by shell layer was more significant as wood loading decreased from 35 to 5%. This trend was seen in both core systems.

The long-term WA and TS properties of coextruded composites with varying wood loadings in the shell layer are shown in Figure 5 using the Core B composite group as an example. It was obvious that the shell layer protected coextruded composites from moisture uptaking. As shown in Figure 5, the un-coextruded samples (core only) absorbed much more water than coextruded composites after $\sim$ 1368 h (57days) water soaking. This could be attributed to better moisture barrier ability of PE. For the same reason, it was not surprised that WA of composites obviously decreased as wood loading in shell layer decreased. The TS of coextruded composites was also much lower than that of un-coextruded core layer, which indicated much better dimensional stability of coextruded composites. This characteristic is very important for future outdoor use of those coextruded composite. Moreover, we did not observe much thickness difference between those composites containing from 5–25% wood in the shell layer. It was possibly because of the restriction effect by core–shell structure. It indicated that moderate wood loading in the shell layer might not affect dimension stability of resultant composites significantly.

Effect of shell layer thickness

Pure HDPE as shell layer was first investigated using the Core A composite system. The relationships between shell thickness and various mechanical properties are shown in Figure 6. The P-values were obtained based on the null hypothesis of nonlinear correlation between the two variables (i.e., property and shell thickness). We observed somewhat positive linear correlation between either flexural or impact strength and shell thickness [Figure 6(a,c)]. These correlations were not perfect according

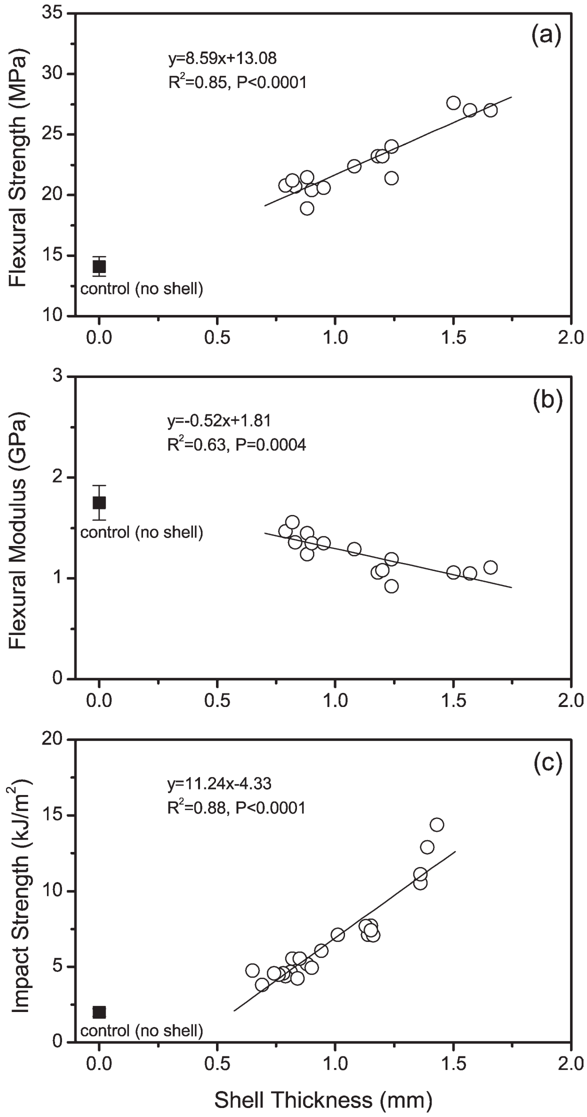

Figure 6 Effect of thickness of pure HDPE shell on mechanical properties of coextruded composites.

to correlation coefficient, R^2. The increased strengths were possibly attributed to thicker shell layer and better tensile strength and toughness of thickened pure PE shell layer. The flexural modulus of composites decreased as shell thickness increased and a weak linear correlation still existed [Figure 6(b)]. Thus, a thick shell layer should be avoided for coextruded PE-based WPC. This conclusion is similar to that for coextruded PVC-based WPC research.[14] Composites coextruded with pure HDPE shell layers had close WA and TS properties as shown in Figure 7. It can be clearly seen that all coextruded composites had better moisture absorption and dimensional stability properties compared with un-coextruded ones (core only). Also, composites with pure PE shells were more stable dimensionally than those with wood-filled ones (Figure 7 versus Figure 5).

Mechanical properties of coextruded composites with the Core B system and wood-filled shell layers with varying thickness are summarized in Figure 8. The wood loadings in the shell layer were fixed at 5% and 25%. As expected, flexural strengths of the coextruded composites were all higher than those of controls (un-coextruded core). However, it seemed no significant linear correlation between flexural strength and shell thickness at the 1% significance level [Figure 8(a)]. This result was different with that in pure HDPE encapsulated composites. In this case, composites possibly reached the ultimate reinforcing capability of the shell layer due to the use of a stronger core (Core B). Flexural modulus had a slightly negative linear correlation with shell thickness according to the P-value and R^2 [Figure 8(b)]. It was because that relative volume and weight portion of the shell layer increased quickly as thickness increased, while stiffness of the shell layer was lower than core layer because of less wood content in the shell layer. Thus, a comparatively thinner shell layer was preferable in order for composites to avoid significant modulus loss. Impact strength of composites was positively linearly correlated with shell thickness [Figure 8(c)] as expected.

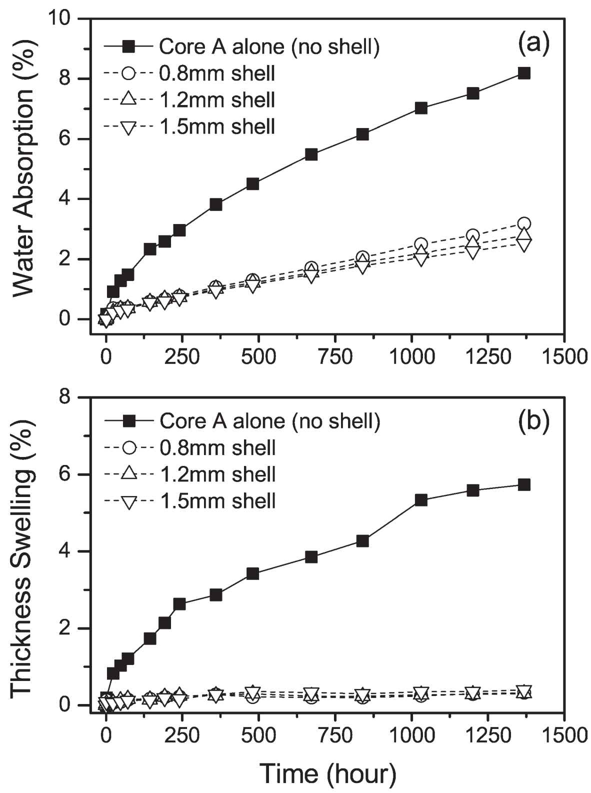

Figure 7 Effect of thickness of pure HDPE shell on water absorption and thickness swelling properties of coextruded composites.

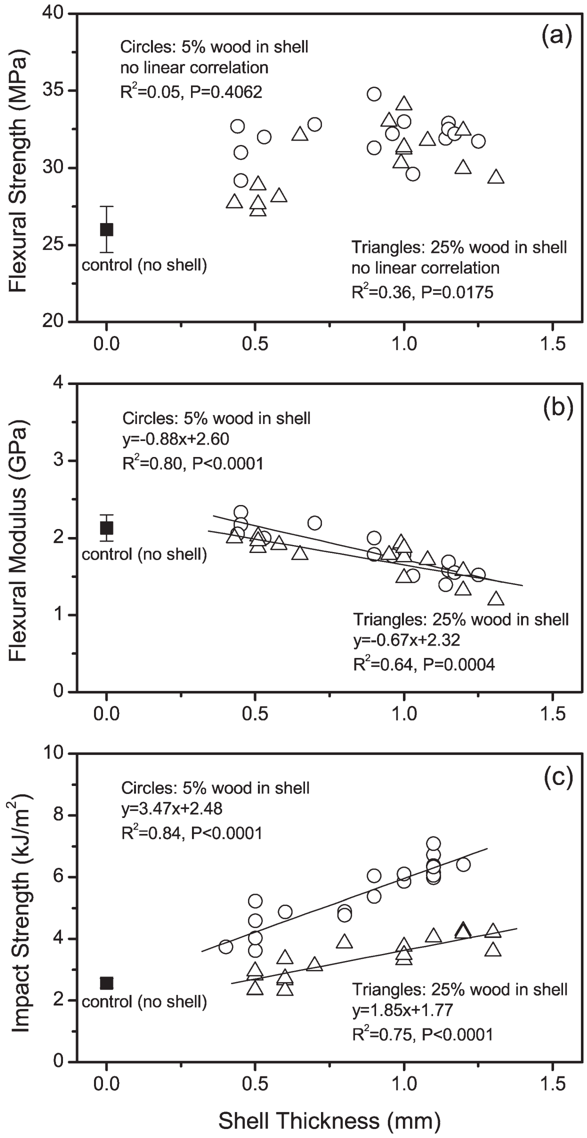

Figure 8 Effect of thickness of wood-filled HDPE shell on mechanical properties of coextruded composites.

The WA properties of composites containing 25% wood in the shell layer are shown in Figure 9. The trend shown in Figure 9 is typical for the composites with moderate wood loading in the shell layer. Obviously, increased shell thickness led to lower WA of composites but did not cause significant TS change. The difference between Figures 7 and 9 was that the WA properties of composites coextruded with wood-filled HDPE shell were somewhat more sensitive to shell thickness change than those having pure HDPE shell were.

CONCLUSIONS

Core–shell structured profile can significantly improve flexural and impact strengths of composites especially when a comparatively weaker core was used. However, it may reduce the modulus of composites when higher modulus core was used. The coextruded composites had significantly better long-term moisture resistance and dimensional stability compared with the corresponding un-coextruded controls. At a fixed shell thickness, less wood loading in the shell layer did not affect flexural modulus, but significantly increased impact strength of coextruded composites. Decreased wood loading in the shell layer did not cause obvious dimensional change but improved WA of coextruded composites. Thus, the work suggested that high wood loading (>25%) in the shell layer should be avoided. At a fixed wood loading in the shell layer, thickening the shell layer improved impact strength but reduced modulus of composites. Thus, a comparatively thinner shell layer was preferable to avoid significant modulus loss of composites. Increased shell thickness helped reduce water uptaking of coextruded composites but did not change dimensional stability remarkably. The work successfully demonstrated that coextrusion technology makes it possible to use recycled, low quality plastic-fiber blend in the core layer and a relatively good shell layer for achieving acceptable overall composite properties.

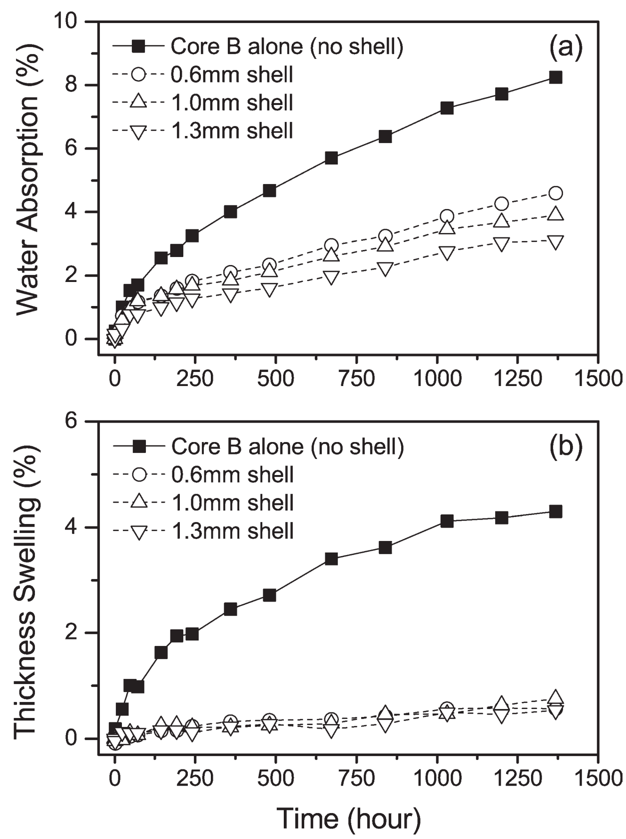

Figure 9 Effect of thickness of wood-filled HDPE shell (containing 25% wood) on water absorption and thickness swelling properties of coextruded composites.

References

1. Bauser, M.; Sauer, G.; Siegert, K. Extrusion; ASM International: Materials Park, Ohio, USA, 2006.
2. Djordjevic, D. Coextrusion; iSmithers Rapra Publishing: Shawbury, United Kingdom, 1992.
3. Rauwendaal, C. Polymer Extrusion; Hanser Verlag: Munchen, Germany, 2001.
4. Averous, L. J Macromol Sci Polym Rev 2004, 44, 231.
5. Jarus, D.; Hiltner, A.; Baer, E. Polymer 2002, 43, 2401.
6. Mavridis, H.; Shroff, R. N. Polym Eng Sci 1994, 34, 559.
7. Perkins, W. G. Polym Eng Sci 1999, 39, 2445.
8. Mitsoulis, E. J Polym Eng 2005, 25, 393.
9. Wang, L.; Shogren, R. L.; Carriere, C. Polym Eng Sci 2000, 40, 499.
10. Sun, J. J.; Koh, Y. H.; Choi, W. Y.; Kim, H. E. J Am Ceram Soc 2006, 89, 1713.
11. El-Salmawy, A.; Kimura, Y. Text Res J 2001, 71, 145.
12. Doshi, S. R.; Charrier, J. M.; Dealy, J. M. Polym Eng Sci 1988, 28, 964.
13. Rowell, R. M. J Polym Environ 2007, 15, 229.
14. Jin, S.; Matuana, L. M. J Vinyl Addit Technol 2008, 14, 197.
15. Stark, N. M.; Matuana, L. M. Proceedings: Coating Wood and Wood Composites: Designing for Durability, Seattle, WA., July 23–25, 2007; p 1.
16. Stark, N. M.; Matuana, L. M. Proceedings: 4th Wood Fibre Polymer Composites International Symposium, Bordeaux, France, March 30–31, 2009; p 1.
17. Klyosov, A. A. Wood-Plastic Composites; Wiley-Interscience: New York, USA, 2007.
18. Tall, S.; Karlsson, S.; Albertsson, A. C. Polym Polym Compos 1998, 6, 261.
19. Vilaplana, F.; Karlsson, S. Macromol Mater Eng 2008, 293, 274.
20. Stark, N. M. J Appl Polym Sci 2006, 100, 3131.
21. Stark, N. M.; Matuana, L. M. J Appl Polym Sci 2003, 90, 2609.
22. Yao, F.; Wu, Q. L.; Lei, Y.; Xu, Y. J. Ind Crop Prod 2008, 28, 63.