

Stress Relaxation of Woodfiber–Thermoplastic Composites

Debes Bhattacharyya, James Manikath, Krishnan Jayaraman

Centre for Composites Research, Department of Mechanical Engineering, University of Auckland, Auckland, New Zealand

Received 19 August 2003; accepted 4 January 2006

DOI 10.1002/app.24018

Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Woodfiber–polypropylene and woodfiber–waste polyethylene composites have been produced by injection molding and by hot pressing the thermoplastic between woodfiber mats. The stress relaxation under constant strain in these composites has been studied at 25, 50, and 80°C. The results have been compared with similar experiments performed on neat thermoplastics. It is interesting to note that the presence of woodfibers as reinforcement in the composites restricts the stress relaxation, but their effectiveness decrease with the increase in ambient temperature.

Composites made by hot pressing the woodfiber mat and the thermoplastic are found to exhibit a lesser amount of relaxation than those made by injection molding the same combination. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 401–407, 2006

Key words: relaxation; thermoplastics; woodfiber–thermoplastic composites; viscoelastic properties; natural fiber composites; Maxwell model

INTRODUCTION

Extensive studies have been carried out over the past few years to investigate the mechanical properties of woodfiber–thermoplastic composites.^{1–7} Woodfibers are biodegradable, inexpensive, easily renewable, and possess lower density than mineral fillers. Woodfiber–thermoplastic composites are attracting increased interest for industrial applications such as construction materials and storage pallets. These composites are cheaper, environment friendly, and have higher stiffness compared to virgin thermoplastics. However, the long-term behavior of the composites under load might restrict their usefulness. So, it is imperative that the time-dependent characteristics of the thermoplastic composites reinforced with woodfibers are studied.

A review of the literature reveals that the majority of woodfiber–thermoplastic composites research around the world has focused on the methods of manufacturing and fiber–plastic interface modification,^{8,9} but very little has been reported on their time-dependent properties. Bhagawan et al.¹⁰ have studied the stress-relaxation behavior of short jute fiber–nitrile rubber composites and reported a two stage relaxation pattern in these composites. Varughese et al.¹¹ have found that the stress-relaxation pattern in sisal fiber–natural rubber composites is influenced by the fiber–rubber interface. George et al.¹² have conducted stress-relaxation

tests on short pineapple fiber–polyethylene composites to study the effects of fiber loading, fiber length, chemical treatment, and fiber orientation on relaxation behavior and observed that the incorporation of fibers results in a decrease in the rate of relaxation.

In the present work, two processing methods, hot pressing with woodfiber mats made on a dry mat forming machine and the widely used injection molding, have been used. Woodfiber–polypropylene and woodfiber–waste polyethylene composites have been made by both these methods. The fiber mass content has been varied between 20 and 30% in the injection-molded composites while it is close to 20% in hot-pressed composites. The stress relaxation of these composites under constant strain has been studied at temperatures of 25, 50, and 80°C and compared with that of the neat thermoplastics.

EXPERIMENTAL

Materials

The pulp and paper industry separates wood into its component fibers to make paper products. These woodfibers themselves are composites of cellulose microfibrils held together by a lignin and hemicellulose matrix.¹³ High temperature mechanical pulp Pinus Radiata fibers, because of their lignin covered surfaces,¹⁴ have advantages as reinforcement in comparison to the highly hydroxylated kraft fibers used for paper-making, and have been used in this study. These Pinus Radiata fibers, often loosely categorized as medium density fibers (MDF), are slightly weaker than kraft fibers, are cheaper to produce, and more amenable to

Correspondence to: K. Jayaraman (medept@auckland.ac.nz).

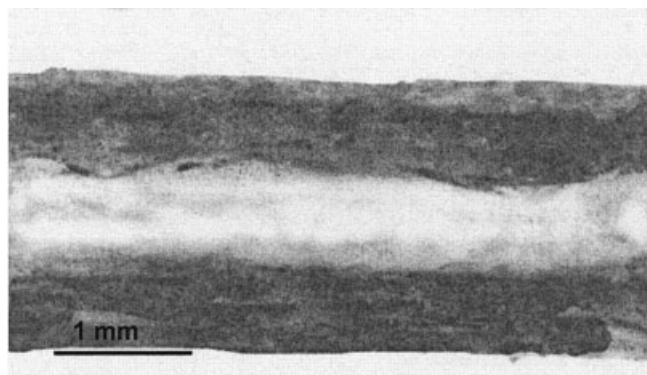


Figure 1 Cross section of a hot-pressed woodfiber-polypropylene prepreg showing the fiber-reinforced outer layers and the polypropylene-rich central region.

surface modification.^{15,16} The MDF used in this study were light brown in color with width varying from 15 to 40 μm and length from 1.5 to 5 mm.

Cotene PP 9800 polypropylene granules and a mix of waste plastics were used as the matrix materials. Waste plastics, recovered through kerbside recycling in Auckland, New Zealand, consist of a variety of thermoplastic and thermosetting materials. Sorting the waste into individual materials is not economically viable, except for the separation of easily identifiable materials. Hence, soft drink containers (PET) were separated out of the waste plastics and the remainder was used in this study. This consisted of a mixture of milk bottles (HDPE) and cleaning product bottles made of different types of polyolefins (mainly LDPE and some polypropylene) in the ratio 1:3.

Specimen preparation

Specimens of neat polypropylene, waste polyethylene, woodfiber-polypropylene composites and woodfiber-waste polyethylene composites were made by injection molding in a Boy 50 injection molder. In the case of composites, fibers were premixed with the polymer in a laboratory scale corotating twin screw extruder, model tsa EMP26-30. The precompounded extrudate was ground into pellets, which were then fed into the injection molding machine. Test specimens were

molded from the composite materials with the barrel temperature set at 200°C and the mold set at 30°C.

Woodfiber mats were produced using a novel, proprietary dry mat former,^{7,17} as a precursor to composite sheet manufacturing. Dry fiber suspended in air was forced by an air-turbine through a fiber delivery head on to a perforated drum rotating about a partially perforated suction pipe. Air was sucked into the rotating drum through its surface and out through the suction pipe that also served as the axle of the drum. The fiber was filtered from the air stream by the surface of the drum and then flattened out by an external roller. Successive layers of fibers were built up on the surface of the drum and eventually formed a dry fiber mat.

Dry fiber mats were interleaved with layers of thermoplastic powder and the material was consolidated by the application of heat and pressure. Unconsolidated, interleaved material was placed between aluminum plates with an aluminum spacer frame inserted to control final sheet thickness and then enclosed by silicone rubber diaphragms. A vacuum of 50 kPa was drawn from the diaphragms to remove air and any water vapor emitted during consolidation. The assembly was then placed between the platens of a hot press and the material was consolidated for a period of 5 min at 180°C and 1.3 MPa. The prepregs formed were sandwich-type composites in which the fiber-reinforced layers form the outer layers and the central region consisted of thermoplastic as shown in Figure 1. The tensile strengths and moduli of the materials under consideration are listed in Table I.

Testing

The stress-relaxation tests were performed at 25, 50, and 80°C inside an Instron environmental chamber (model 3119-006) mounted on an Instron 5567 universal testing machine. Injection-molded samples were dog bone shaped (with nominal test area dimensions of $80 \times 10 \times 4 \text{ mm}^3$), whereas rectangular strips (with dimensions of $120 \times 15 \times 2 \text{ mm}^3$) were cut from the prepregs for testing. Ease of sample preparation was the criterion for the choice of sample shape and tests done indicated that the sample shape did not

TABLE I
Tensile Strengths and Moduli of the Materials Under Consideration

Material	Fiber mass content (%)	Tensile strength at break (MPa)	Tensile modulus at 0.2% elongation (GPa)
Polypropylene	0	23.0	1.0
Woodfiber-polypropylene composite	20	26.4	2.4
Waste HDPE	0	24.3	2.8
Waste LDPE	0	21.7	2.5
Woodfiber-waste polyethylene composite	20	19.9	2.1

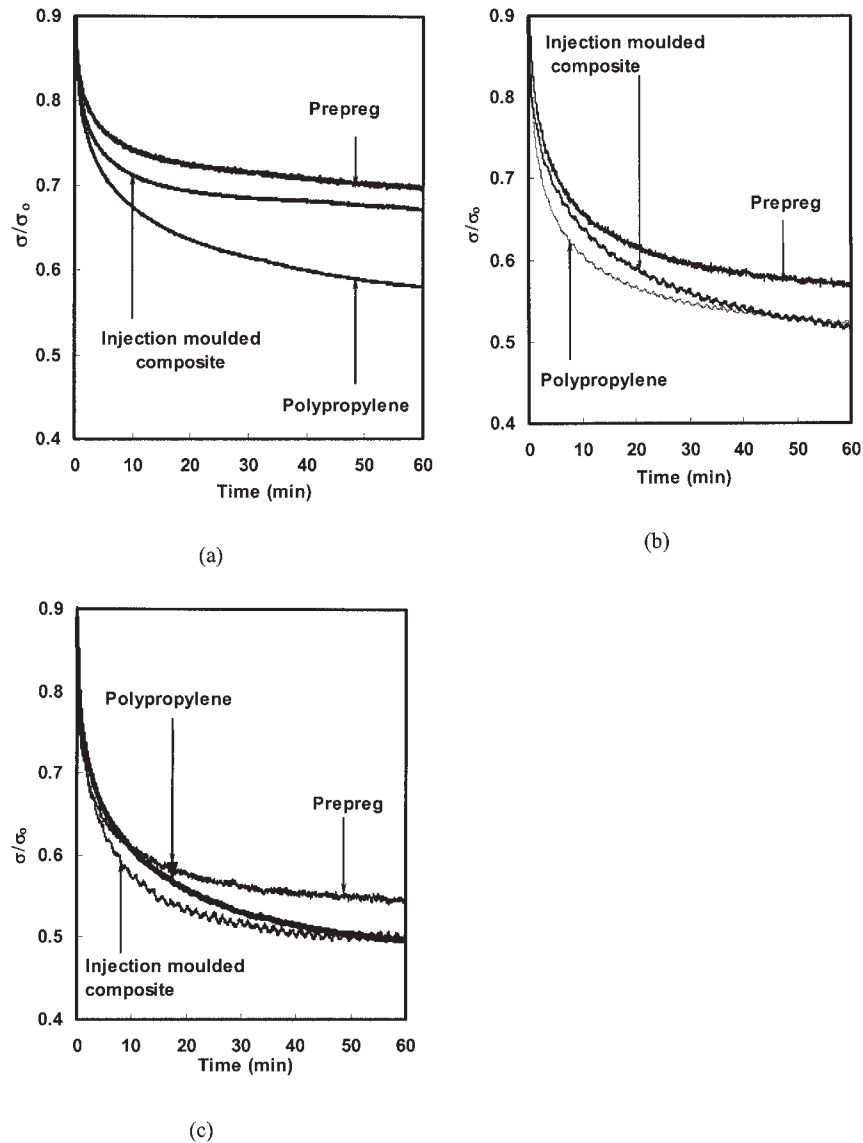


Figure 2 Stress-relaxation behavior of polypropylene and woodfiber-polypropylene composites of 20% fiber mass content at (a) 25°C, (b) 50°C, and (c) 80°C.

have much effect on the results of the relaxation studies. The samples were preheated for 30 min at the testing temperature before the start of the test. A stress, which was 30% of the maximum stress for the different types of samples at each of the different temperatures, was applied to the samples and the strain was held constant. The decay in stress with time was studied and the normalized stresses (σ/σ_0 ; i.e., instantaneous stress/initial stress) under constant strain were plotted against time.

RESULTS AND DISCUSSION

Woodfiber-polypropylene composites

The stress-relaxation behavior of neat polypropylene and woodfiber-polypropylene composites with a fiber

mass content of 20% is shown in Figure 2. Figure 2(a) clearly shows that the amount and rate of stress relaxation are less for the woodfiber composites than those for polypropylene at temperatures close to the room temperature. Interestingly at increased temperatures of 50 and 80°C, Figures 2(b) and 2(c), respectively, polypropylene- and the injection-molded composites exhibit greater relaxation than the prepreg and their relaxation curves come much closer to each other. This may be explained by the fact that a preimpregnated specimen possesses essentially a sandwich structure with two outer layers containing woodfiber-rich layers impregnated with the matrix polymer, whereas injection-molded specimens contain more uniformly distributed mixture of woodfibers and polymer. It is also to be noted that most of the fibers in the outer

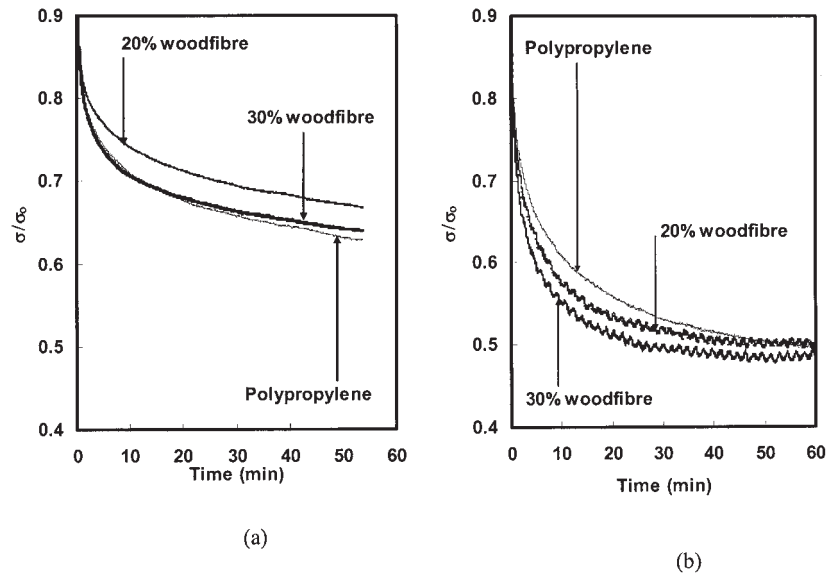


Figure 3 Stress-relaxation behavior of injection-molded polypropylene and woodfiber-polypropylene composites of 20 and 30% fiber mass contents at (a) 25°C and (b) 80°C.

layers retain some of the interlocked structure generated during the mat formation,^{7,17} thus resulting in stiffer skins (see Table I for further evidence of this notion of stiffness increase). Therefore, the overall tensile behavior of a prepreg specimen is largely controlled by the deformation characteristics of these fiber-rich outer layers, which have to undergo the same amount of deformation as that of the core material (connected in parallel) and as a result these specimens relax less than the injection-molded and neat polypropylene specimens. At the softened state of the polymer, the bonding between the fibers and the matrix is expected to be weaker and the short fibers, coupled with weak interfacial bonding, start behaving like fillers in the polymer-rich areas and fail to share the imposed load.¹⁸ This explains the greater relaxation in general, demonstrated by the composite specimens at higher temperatures, irrespective of their manufacturing method. Another contributing factor may be an increase in the amount and rate of stress relaxation of woodfibers at elevated temperatures.¹⁹

The stress-relaxation characteristics of injection-molded neat polypropylene and woodfiber-polypropylene composites of 20 and 30% fiber mass contents are shown in Figure 3 for two different temperatures. At 25°C, the composite with a fiber mass content of 30% more or less follows a relaxation pattern very similar to that of neat polypropylene, and it might be interesting to note that the specimens with 25% woodfiber mass content follow a similar trend (not shown). However, more interestingly at 80°C, the composite specimen with fiber mass contents of 20 and 30%, unlike their compression-molded counterparts, relax faster than the polypropylene specimen, Figure 3(b). This can be explained by

considering the simple linear viscoelastic model of Maxwell with a spring and a dashpot in series.²⁰ The rate and amount of the relaxation are controlled by the deformation characteristics of the viscous element and the stiffness of the spring element. A larger viscous deformation or a stiffer elastic part should amount to a larger relaxation response with the rate varying in a similar manner. With the addition of woodfibers, the stiffness, as shown earlier, increased and in conjunction the polymer became softer at the elevated temperature of 80°C. These two effects, coupled together, generally produced larger relaxations for composites with 30% fiber content, showing more pronounced effects. This also explains the behavior of the injection-molded specimen in Figure 2(c).

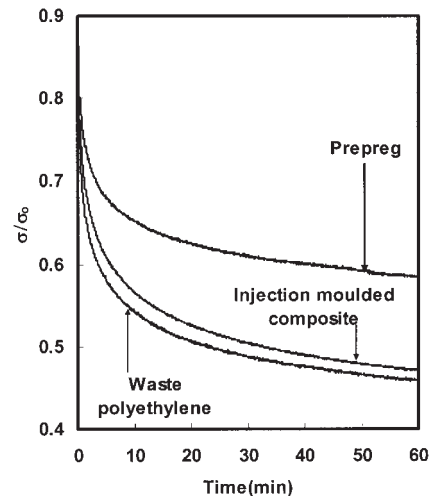


Figure 4 Stress-relaxation behavior of waste polyethylene and woodfiber-waste polyethylene composites of 20% fiber mass content at 25°C.

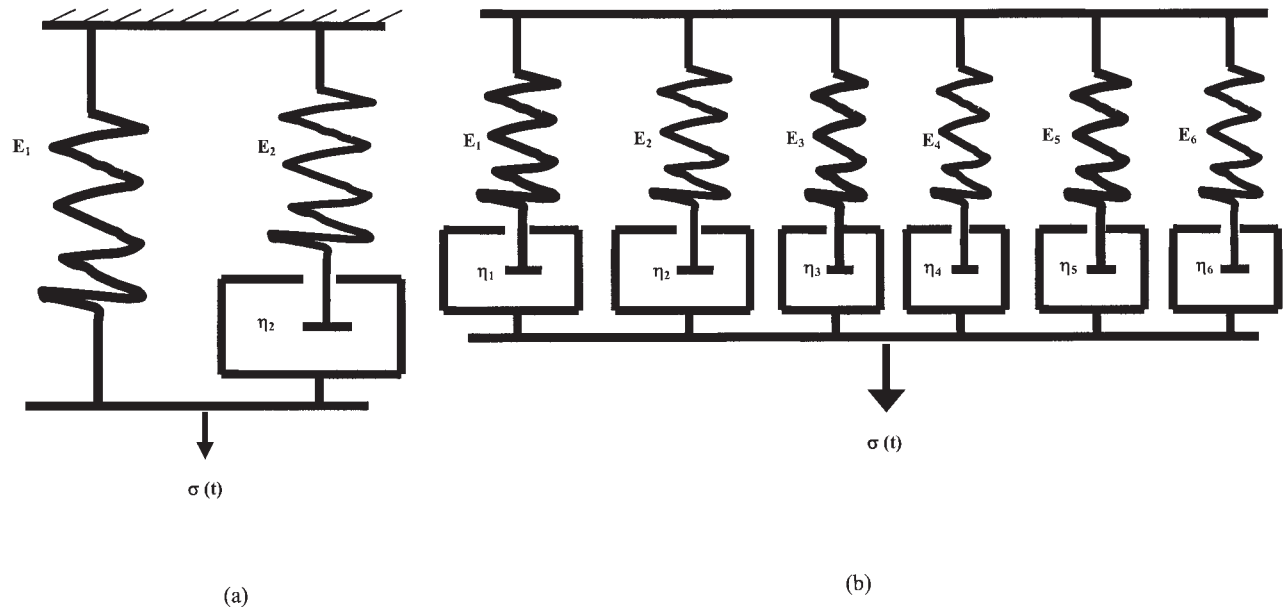


Figure 5 Schematic diagram showing the components of (a) a three-element Maxwell relaxation model and (b) a six-element Maxwell relaxation model.

Woodfiber-waste plastic composites

The stress-relaxation curves of waste polyethylene and woodfiber-waste polyethylene composites made by injection molding and hot pressing are shown in Figure 4. The woodfiber contents in the injection-molded specimens and the prepregs were about 20% by mass. Stress-relaxation tests were performed at only 25°C and 50°C, as higher temperatures gave very low maximum stresses for these composites. The stress-relaxation results indicate that the preimpregnated polyethylene specimens, as their polypropylene counterparts, show lesser amount of stress relaxation

compared to those of the injection-molded composites. The difference between the two types of polyethylene composites is more pronounced than in the case of polypropylene composites.

Comparison of the experimental results with Maxwell models

The relaxation behavior of polypropylene and woodfiber-polypropylene composites can be represented by the Maxwell body, a combination of a spring and a dashpot in series. In stress-relaxation experiments, the

TABLE II
Constants for the Maxwell Relaxation Models for Polypropylene and Woodfiber-Polypropylene Composites of 20% Fibre Mass Content at 25, 50, and 80°C

Material	Fibre mass content (%) / testing temperature (°C)	σ_1	σ_2	σ_3	τ_1 (s)	τ_2 (s)	τ_3 (s)
Prepeg	20/80	0.601	0.35	—	—	108	—
Injection-moulded composite	20/80	0.53	0.39	—	—	249	—
Polypropylene	0/80	0.55	0.22	0.16	12,797	157	88
Prepeg	20/50	0.68	0.29	—	—	105	—
Injection-moulded composite	20/50	0.58	0.36	—	—	227	—
Polypropylene	0/50	0.63	0.28	0.10	14,782	125	27
Prepeg	20/25	0.716	0.217	—	—	155	—
Injection-moulded composite	20/25	0.69	0.287	—	—	185	—
Polypropylene	0/25	0.68	0.264	0.14	21,197	121	38

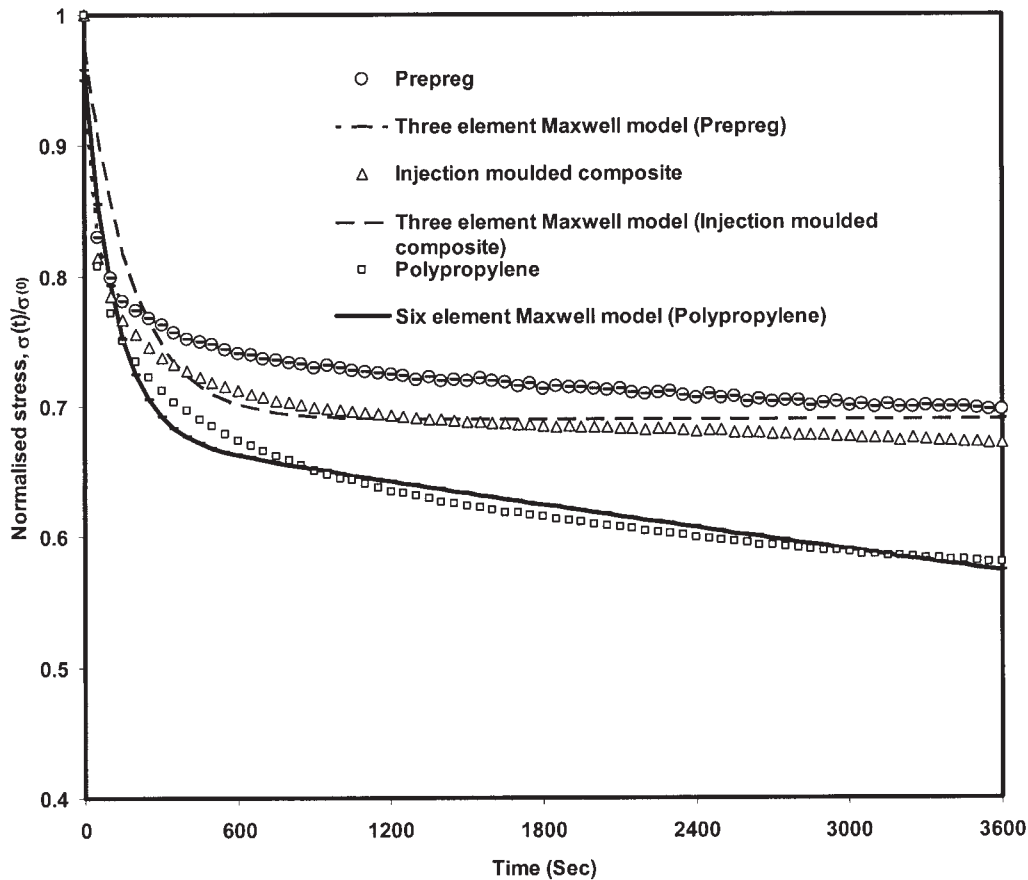


Figure 6 Predicted and experimental relaxation data for polypropylene and woodfiber-polypropylene composites of 20% fiber mass content at 25°C.

instantaneous strain is realized in the spring element and the dashpot describes the viscous behavior. To accurately fit the experimental data, a three-element relaxation model consisting of a single spring in parallel with a Maxwell body was used for the composites and a six-element relaxation model consisting of three Maxwell bodies in parallel was used for polypropylene, Figure 5. The governing equation for the three-element model during relaxation can be written as $\sigma(t)/\sigma_0 = \sigma_1 + \sigma_2 \exp(-t/\tau_2)$; where " τ_2 " is the relaxation time constant²¹; a similar governing equation can be written for the six-element relaxation model. A code was written in Matlab to calculate the equation constants using the experimental results for woodfiber-polypropylene composites of 20% fiber mass content and polypropylene at 25, 50, and 80°C. The magnitudes of these equation constants could be determined by iteratively varying the time constant, thereby providing the best fit of the Maxwell model to the experimental stress-relaxation curve; the constants thus calculated are shown in Table II. These best-fit curves, plotted in Figure 6, display trends similar to the experimental results.

CONCLUSIONS

Woodfiber composites restrict stress relaxation more than plastics at temperatures about 25°C. As temperature increases, the effectiveness of woodfibers in restricting stress relaxation decreases. But even at 80°C, prepregs made using woodfiber mats show lesser amount of stress relaxation than neat polypropylene. It is interesting to note that the sheet prepregs exhibit less stress relaxation compared to the samples made by injection molding. This is due to the fact that the overall tensile behavior of a prepreg specimen is largely controlled by the deformation characteristics of the fiber-rich outer layers, which have to undergo the same amount of deformation as that of the core material. The prepregs of waste polyethylene and woodfiber mats exhibit similar behavior, which suggests that such prepregs can lead to products with good time-dependent properties.

The authors thankfully acknowledge the support provided by the Plastics Institute of New Zealand and the Foundation for Research, Science and Technology, New Zealand while conducting this research. The authors also thank Anjaneya Prasad Penneru for his help with the relaxation models.

References

1. Raj, R. G.; Kokta, B. V.; Maldas, D.; Daneault, C. *J Appl Polym Sci* 1989, 37, 1089.
2. Maldas, D.; Kokta, B. V. *J Appl Polym Sci* 1990, 40, 917.
3. Maldas, D.; Kokta, B. V. *J Comp Mater* 1991, 25, 375.
4. Felix, J. M.; Gatenholm, P. *J Appl Polym Sci* 1991, 42, 609.
5. Balatinecz, J. J.; Park, B.-D. *J Mater Sci* 1999, 34, 24.
6. Coutinho, F. M. B.; Costa, T. H. S. *Polym Test* 1999, 18.
7. Bowis, M. E. Ph.D. Thesis, Department of Mechanical Engineering, University of Auckland, 1997.
8. Bledzki, A. K.; Gassan, J. *Prog Polym Sci* 1999, 24, 221.
9. Lu, J. Z.; Wu, Q.; McNabb, H. S. *Wood Fiber Sci* 2000, 32, 88.
10. Bhagawan, S. S.; Tripathy, D. K.; De, S. K. *J Appl Polym Sci* 1987, 33, 1623.
11. Varughese, S.; Kuriakose, B.; Thomas, S. *J Appl Polym Sci* 1994, 53, 1051.
12. George, J.; Sreekala, M. S.; Thomas, S.; Bhagawan, S. S.; Neelakantan, N. R. *J Reinforc Plast Compos* 1998, 17, 651.
13. Walker, J. C. F. In *Primary Wood Processing*; Walker, J. C. F., et al, Eds.; Chapman and Hall: London, UK, 1993.
14. Kibblewhite, R. P.; Brookes, D.; Allison, R. W. *TAPPI* 1980, 63, 133.
15. Beshay, A. D.; Kokta, B. V.; Daneault, C. *Polym Compos* 1985, 6, 261.
16. Zadorecki, P.; Mitchell, A. J. *Polym Compos* 1989, 10, 69.
17. Bhattacharyya, D.; Bowis, M.; Jayaraman, K. *Compos Sci Technol* 2003, 63, 353.
18. Mackenzie, C. I.; Scanlan, J. *Polymer* 1984, 25, 559.
19. van Houts, J.; Bhattacharyya, D.; Jayaraman, K. *Holzforschung* 2003, 57, 391.
20. Caddell, R. M. *Deformation and Fracture of Solids*; Prentice-Hall: Englewood Cliffs, NJ, 1980; p 124.
21. Bodig, J.; Jayne, B. A. *Mechanics of Wood and Wood Composites*; Van Nostrand Reinhold: New York, 1982.