Damping Behavior of Wood Filled Polypropylene Composites

Shakti Chauhan,* Ajay Karmarkar, Pankaj Aggarwal

Institute of Wood Science and Technology, Bangalore, India

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ABSTRACT: The damping coefficient (tanδ) of wood flour filled polypropylene composites, having varying filler concentrations were measured using the free vibration decay of disk-shaped specimen, vibrating in flexural vibration mode. The damping coefficients decreased with the increase of filler load in composites. There was no significant difference in damping behavior of composites with and without compatiblizer at low filler level (upto 30%). At higher filler loading (>30%), composites with compatiblizer had lower damping coefficient suggesting improved interfacial adhesion between wood and polypropylene. The damping in composite is attributed to the damping because of the composite constituents and damping at the interface. The damping because of interface was estimated using a model and was found to increase with the increase in filler loading. At higher filler content, damping due to interface in composites with compatiblizer was significantly lower than in composites without compatiblizer suggesting a better interfacial adhesion between the wood filler and polypropylene matrix with compatiblizer. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 2421–2426, 2009

Key words: acoustic; damping; polypropylene; vibration; wood-PP-Composite

INTRODUCTION

Damping is an important acoustic parameter for characterization of materials. It indicates the mechanism by which the elastic energy dissipates in the material once it is set in vibration. It is a critical modal parameter for the design of structures for which, the vibration control and cyclic loading are critical. Damping is also a significant factor for the fatigue life and impact resistance of structures. The phenomenon represents departure of a material from an ideal elastic behavior. In an ideal elastic material, the phase lag between an applied alternating stress and corresponding alternating strain wave would be close to 0°. A phase difference is observed between stress and strain when applied stress contains contributions from either a plastic or viscoelastic nature of the material. For a purely viscous material, the phase difference would be 90°.

It is a well-known fact that a majority of polymers behave like viscoelastic materials and exhibit high damping compared to metallic materials. Most of the mechanical properties of polymers depend on their microstructural characteristics like number of monomeric units,[1] molecular weight, degree of branching and crosslinking, and the main chain stereo configuration. Extensive studies have been carried out on the mechanism of vibration damping in polymers. However in case of composite materials, the presence of fillers or reinforcing agents creates complexities in the internal structure of the material. In such cases, the damping behavior depends not only on properties of individual materials but also on many other factors, like volume fraction of fillers, the quality of interface, loading direction, and plasticization of the polymer matrix.[2] The damping characteristic of a composite under dynamic load is an effective tool for characterizing the morphology of the interface. It has been shown that the damping phenomenon is closely associated with the static creep and stress relaxation characteristics of materials.[3]

Several studies have been carried out to characterize elastic and damping properties of wood-polymer composites using dynamic mechanical analysis. Behzad et al.[4] emphasized the importance of dynamic measurement of elastic and viscous components in characterizing the material performance of composites. Boluk and Schreiber[5] observed differences in the damping characteristics of a polymer and filler without specific interactions (i.e. acid-base). The attributed differences were related to an effective immobilized layer around the filler. The morphology of the plastic is known to change by the addition of

**Present address:* School of Forestry, University of Canterbury, Christchurch, New Zealand
Correspondence to: S. Chauhan (shakti@icfre.org).

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filler in a semicrystalline matrix because of induced nucleation on the filler surface.[6] Harper and Wolcott[7] have thoroughly investigated the influence of maleic anhydride grafted polypropylene (MAPP) compatiblizer on interface between wood and polypropylene. They suggested that MAPP improved interaction between matrix and filler leading to an increased bending stiffness and decreased mechanical damping. Changes in the microstructure of semicrystalline polymer matrix near wood fibers have been reported with the addition of compatiblizer.[8,9] These changes influence the mechanical response of the composite material. Tajvidi et al.[10] observed no significant influence of compatiblizer on loss factor in wood filled HDPE composite at 25% filler content but at 50% filler content, composite with compatiblizer exhibited lower tanδ as compared to that in composite without compatiblizer.

In our earlier paper,[11] we have reported measurement of dynamic elastic constants namely modulus of elasticity, shear modulus, and Poisson's ratio of wood-polypropylene composite using disk-shaped specimens. The use of disk-shaped specimen offers advantages of measuring modulus of elasticity and Poisson's ratio simultaneously from the resonance frequencies of flexure and antiflexure vibration mode. Measurement of damping coefficient by vibration method requires a precise support of the sample at nodes of a specific vibration mode to minimize any damping at the sample-support interface. In this article, we report measurement of damping coefficient, tanδ, of wood-polypropylene composites from the free decaying vibration of a circular disk, vibrating in flexural vibration mode. The damping in composites prepared at varying filler concentration with compatiblizer and without compatiblizer is compared. The relative contribution of the interface region on the damping coefficient of the composites has also been evaluated.

MATERIALS AND METHODS

Polypropylene (Repol H100EY) with melt flow index of 11 g/10 min at 230°C under 2.16 kg load was procured from Reliance Industries, India and was used as the matrix material. Rubber wood flour was used as fillers. Wood chips of rubber wood were dried in an oven at 102°C and then pulverized. The pulverized wood flour was sieved. Wood flour passing through the 30 mesh size sieve was taken for the study. The pulverized flour was oven dried before using for composite preparation to avoid any wood moisture related effects in composites. *m*-Isopropenyl-α, α- dimethylbenzyl-isocyanate (*m*-TMI) grafted polypropylene was used as the compatiblizer. Grafting of *m*-TMI on polypropylene was carried out by melt phase reactive extrusion using a corotating twin screw extruder fitted with a side feeder. Dicumyl peroxide (10% W/W) was used as an initiator. *m*-TMI (10% W/W) was absorbed on porous isotactic polypropylene (Koylene AM120NS) granules and mixed well with 25% of the initiator in a tumble mixer. The *m*-TMI- PP-initiator blend was extruded with the remaining 75% peroxide initiator fed through the side feeder during extrusion. *m*-TMI-*grafted*-PP was collected by air cooling the extrudate on a moving steel belt and granulized. The obtained granules were used as compatiblizer in composite preparation. Composites were prepared by extruding dried wood flour with polypropylene blended with process additives (Zinc stearate, Irganox B-215, Di-octyl phthalate, and paraffin wax) and *m*-TMI-*grafted*-PP (5 wt % of wood) in a 28 mm corotating twin screw extruder equipped with a side feeder and two volumetric feeders. Wood flour was forced fed into molten polymer through the side feeder fitted at a distance of 400 mm from the main inlet, which was used to feed polypropylene blend. The detailed process of synthesis of *m*-TMI-*g*-PP by reactive extrusion and compounding of wood-polypropylene composites has been described in our earlier study.[12]

The composite materials with varying filler concentration ranging from 10 to 50% were prepared in a twin screw extruder with and without compatiblizer. The output of the extruder was 3 mm dia strands of the composite material, which were pelletized and dried in an oven to remove any moisture before molding. The compounded pellets of wood-polymer composites were molded into circular disks (100 mm diameter and 3 mm thickness) using an 80-ton Engle make injection-molding machine. After molding, the specimens were conditioned for at least 3 days, by storing over calcium chloride in a desiccator, before testing. For each composite formulation, three disk samples were taken randomly. Density of the composite was determined by measuring weight and volume of each sample disk. The dynamic modulus of elasticity of composites was measured from the flexural vibration frequencies of disk shaped specimens using the following equation.

$$\mathrm{MoE} = \frac{3\pi^2 f^2 R^4 \rho(1-\upsilon^2)}{K_{\mathrm{na}}^2 t^2} \tag{1}$$

where f is the fundamental frequency of flexural vibration mode, R is the disk radius, ρ is the density, υ is the Poisson's ratio, t is the disk thickness, and K_{na} is a parameter depending in the Poisson's ratio and the disk thickness to radius ratio. The values of the Poisson's ratio were in the range of 0.39–0.43, where the Poisson's ratio decreased with increasing filler content. The value of K_{na} was 9.08 for the Poisson's ratio of 0.43 and the disk dimensions used in the study.[13]

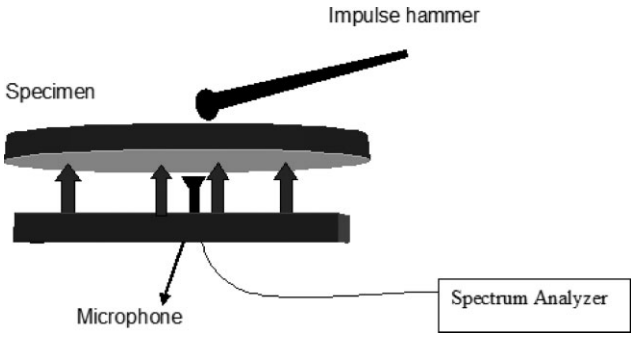

Figure 1 Experimental set-up for capturing vibration spectra.

DAMPING MEASUREMENTS

Techniques used for the measurement of damping depend on the nature of vibration which is categorized either as free vibration (or free decay) or forced vibration. Nowick and Berry[14] summarized the techniques used for measuring vibration damping of materials and structures. In the free vibration method, the decay of the amplitude of vibration generated by an impulse is measured and recorded. The damping is expressed in terms of the logarithmic decrement which is the natural logarithmic value of the ratio of two adjacent peak values of displacement. For a small amplitude plane stress wave, the amplitude of a stress wave of the original amplitude X_o over a number of oscillations in a material is given by:

$$X_n = X_o \exp(-\mathrm{nd})$$

Therefore,

$$\log X_n = \log X_o - \mathrm{nd} \tag{2}$$

where "d" is logarithmic decrement, "n" is the number of cycles, and "X_n" is the amplitude of nth cycle. So the decrement can be obtained from the slope of a plot of $\log X_n$ against n. The damping coefficient is

$$\tan\delta = \frac{d}{\pi} \tag{3}$$

By tracking the stress wave propagation, the spectrum analyzer can capture information about attenuation of the wave over a period of time.

The damping coefficient (tanδ) was measured using free vibration of the circular disk-shaped sample in flexural vibration mode with the nodal circle. Boundary conditions like supporting positions of sample play a critical role in the measurement of damping. Since displacement is minimum at the nodal points, the sample should be supported at the nodal points for a specific vibration mode to minimize the loss of vibration energy at the support points. The nodal circle for flexural vibration of the disk is at 0.68 times the disk radius. The experimental set-up for damping measurement is shown in Figure 1. The disk sample was supported at four points on the nodal circle. The microphone was placed exactly below the specimen center. The sample was gently tapped at the center to set only flexural vibration modes. The amplitude of free vibrations with time was recorded using a FFT analyzer. The damping coefficient, tanδ, was calculated from the decaying-oscillatory damping curve using eq. (2). Measurements were carried out on wood-polypropylene composites with and without compatiblizer at varying filler concentrations of 10, 20, 30, 40, and 50%. For each combination three samples were taken and on each sample three repeated measurements were made.

RESULTS AND DISCUSSION

For small amplitudes, tanδ is independent of vibration amplitude. To ascertain this, tanδ was measured on each disk with different initial amplitudes. Figure 2 indicates variability in measurement of logarithmic decrement, d, for 50% wood powder filled composite. In all cases the value of tanδ was independent of initial amplitude ensuring the correct measurement of damping coefficient.

The vibration damping in composites varies with filler concentration. Typical decay curves observed for virgin polypropylene and wood flour filled composite at 50% filler loading with compatiblizer are shown in Figure 3(A,B), respectively. There was no apparent difference in decay curves for composites with and without compatiblizer. Amplitude peak of every alternate cycle was taken for calculating logarithmic decrement. Vibration amplitude in the disk of pure polypropylene quickly decayed with time as compared to that in wood filled composite. Table I

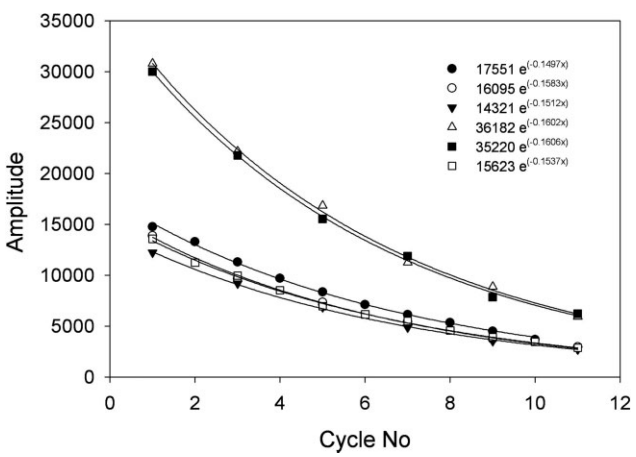

Figure 2 Effect of initial amplitude on decay curve.

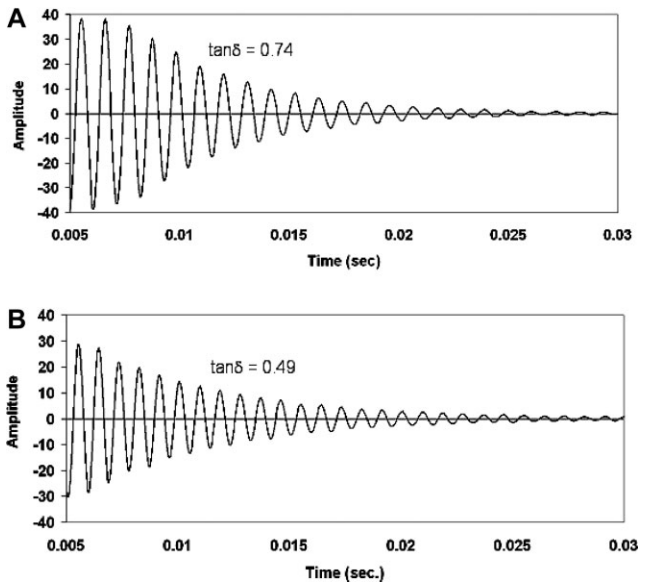

Figure 3 Vibration decay curve (A) Virgin polymer (B) 50% Wood filled composite.

shows the values of density, dynamic modulus of elasticity, and tanδ in wood filled composites with and without compatiblizer. The values shown in parenthesis for tanδ are coefficient of variation (in %) from nine measurements of each formulation. The low coefficient of variation within a formulation signifies the consistency in damping measurement. The damping factor decreased with the increase in filler content in the composites. The value of tanδ at 50% filler loading was 33% lower than that measured at 10% filler loading. A paired *t*-test indicated significant differences in the values of damping factor measured at different filler concentrations. Decrease in damping factor and increase in modulus of elasticity with increasing filler content was observed in composites with compatiblizer as well as without compatiblizer.

As the filler content increased, the material behavior shifted from viscoelastic to elastic and the proportion of dissipated energy reduces, and recoverable strain energy increases. As the energy dissipation in wood is relatively very small (tan δ of the order of 0.01 for dried wood) as compared to that in polypropylene (tan δ of the order of 0.075), the increasing proportion of wood filler contributes in reducing the energy dissipation by the material in the composite. The composites prepared with or without compatiblizer did not exhibit a significant difference in damping coefficients at low filler level (upto 30%). At higher filler loadings of 40% and 50%, tanδ in composite with compatiblizer was lower than in composites without compatiblizer. The difference in damping coefficients at higher filler loading could be attributed to a better interfacial adhesion between wood particles and PP matrix, and improved load transfer from matrix to fibers in the composites with compatiblizer. Similar observations on effect of compatiblization on tanδ in wood-polymer composites have been reported by other researchers. Behzad et al.[4] observed no significant difference in tanδ in HDPE- wood flour composites with and without compatiblizer at 25% wood flour measured by dynamic mechanical analyzer. At 50% wood filler, an influence of compatiblizer on tanδ was reported wherein composites with compatiblizer exhibited lower tanδ. The phenomenon was more prominent above α-transition. Zugenmaier[15] also reported minor differences in damping factor of PP-cellulose microfibres composites with and without MAPP.

Harper[2] reported that with the addition of a coupling agent, a trans-crystalline layer is formed at the interface resulting in higher modulus and strength properties of the composites. An increase in crystallinity of the matrix should decrease the damping by decreasing the amount of polymer that can relax. Karmarkar et al.[12] observed significantly large improvement in strength properties (both tensile and flexural) in the composite with the addition of *m*-TMI-*g*-PP as a coupling agent over uncoupled. With increasing filler content the strength properties of the composite with compatiblizer increased linearly, whereas in composites without compatiblizer the properties either remained same or declined slightly. This improvement in strength properties was attributed to the effective interfacial adhesion

TABLE I
Density, Volume Fraction and Damping Coefficients of Wood Filled Composites

Filler content (%)	Volume fraction (%)	Composite density (kg/m^3)	Composites with compatiblizer		Composites without compatiblizer	
			MoE (MPa)	tan δ	MoE (MPa)	tan δ
0	0	900	2174	0.074 (1.14)	2174	0.074 (1.14)
10	6.9	931	2632	0.073 (2.26)	2725	0.072 (1.47)
20	13.8	970	2868	0.067 (1.47)	2980	0.066 (1.24)
30	21.6	1008	3408	0.060 (0.77)	3408	0.060 (2.50)
40	30.7	1040	3557	0.056 (1.77)	3808	0.058 (1.47)
50	39.7	1085	4152	0.049 (2.61)	4322	0.054 (1.60)

between wood surface and polypropylene. However, such a large difference in damping coefficient was not apparent in composites with and without compatiblizer even at a high fiber loading. Gu[16] suggested that both poor and good interfacial adhesion can lead to increased damping. When the interfacial adhesion is poor, the damping is because of the Coulomb friction, wherein energy gets dissipated at the interface because of the sliding friction mechanism. When the interface is well bonded, as assumed in composites with compatiblizer, increased dislocation density near the fiber matrix interface can lead to increase in damping. As in these composites, the presence of compatiblizer tends to increase the nucleating ability of wood surface resulting in a thick trans-crystalline layer with the increased crystallinity of polymer matrix. The trans-crystalline layer, having a continuous network of gaps could lead to damping in composites with compatiblizer. However, such damping at the interface is expected to be much smaller than the sliding friction.

The total energy dissipation in composites depends on the modulus and damping factors of individual component, their respective fractions in the composite material, and the interfacial adhesion between matrix and filler. Zorowasky and Murayama[17] suggested that, the damping coefficient of the composite materials can be divided into two components, viz. energy dissipation because of the individual components and energy dissipation at interfaces. The damping coefficient of the composite can be written as,

$$\tan \delta_c = \tan \delta_s + \tan \delta_{\text{in}} \tag{4}$$

where $\tan \delta_c$, $\tan \delta_s$, and $\tan \delta_{\text{in}}$ are damping coefficients of composite, total system, and interfaces respectively. The damping coefficient of the total system can be derived from the complex modulus of the fiber and matrix. The complex modulus of the composite obeys the rule of mixtures and can be defined as the volume weighted average modulus of the matrix and fiber i.e.,

$$E_c^* = E_f^* V_f + E_m^* V_m \tag{5}$$

where E is the complex modulus and V is the volume fraction. The subscripts f, m, and c represent filler, matrix and composite respectively. The modulus of the composite can be written in form of an elastic part (E') and a viscous or damping component (E'') as

$$E_c' + iE_c'' = (E_f' + iE_f'')V_f + (E_m' + iE_m'')V_m. \tag{6}$$

The elastic component describes the energy stored in the system, whereas the viscous component describes the energy dissipated during the process.

Thus the eq. (6) can be written as

$$E_c' + iE_c'' = (E_f' V_f + E_m' V_m) + i(E_f'' V_f + E_m'' V_m) \tag{7}$$

According to eq. (7)

$$E_c' = E_f' V_f + E_m' V_m \quad \text{and} \quad E_c'' = E_f'' V_f + E_m'' V_m \tag{8}$$

Mechanical loss factor or damping coefficient of the total system (tanδ) is defined as,

$$\tan \delta_s = \frac{E_c''}{E_c'} = \frac{E_f'' V_f + E_m'' V_m}{E_c'} \tag{9}$$

The eq. (9) can be transformed as,

$$\tan \delta_s = \frac{E_f''}{E_f'}\frac{E_f'}{E_c'} V_f + \frac{E_m''}{E_m'}\frac{E_m'}{E_c'} V_m \tag{10}$$

By definition $\frac{E_f''}{E_f'} = \tan \delta_f$ and $\frac{E_m''}{E_m'} = \tan \delta_m$

$$\tan \delta_s = \frac{\tan \delta_f E_f' V_f + \tan \delta_m E_m' V_m}{E_c'} \tag{11}$$

Thus total system or material damping, $\tan\delta_s$, can be estimated from the elastic modulus, damping coefficient and volume fraction of the individual components, and elastic modulus of the composite. In an ideal interface, the interface thickness is considered as zero as it does not contribute to the damping. In such case, $\tan\delta_c = \tan\delta_s$. In practice, the interface region plays a role in dissipating energies and contributes to the damping. The interface damping can not be quantified directly, but we can estimate it by subtracting the calculated material damping from the total composite damping. The material damping can be calculated using eq. (11).

The total system energy dissipation was estimated for wood flour filled composites assuming damping coefficient of wood to be 0.01. The damping coefficient of polypropylene was 0.074. The dynamic modulus of elasticity of virgin polypropylene and oven dried rubber wood was 2,174 MPa and 10,000 MPa. The variation in damping was because of the material component estimated from the Rule of Mixtures using eq. (11) and correspondingly interface dampings are given in Table II. It is evident that the material damping ($\tan\delta_s$) decreased with the increasing filler content in both types of composites, and compatiblizer does not have any significant influence on the material damping. The relative contribution of the interface in damping in composites was found to increase with the increasing filler content. This increase can be attributed to the increase in total interfacial area resulting in large number of energy dissipation sites. At lower filler level (upto 30%), the interface damping was also nearly the same in both types of composites. At higher filler levels (at 40%

TABLE II
Material and Interface Damping Components in the Composites

Filler content (%)	With compatiblizer		Without compatiblizer	
	tan δ_s	tan δ_{in}	tan δ_s	tan δ_{in}
10	0.060	0.013	0.058	0.014
20	0.053	0.014	0.051	0.015
30	0.043	0.017	0.043	0.017
40	0.040	0.016	0.037	0.021
50	0.033	0.016	0.032	0.022

and 50%), effect of compatiblizer appears to become prominent on interface damping. The interface damping in composites without compatiblizer was about 35% higher than in composites with compatiblizer suggesting the effect of compatiblizer in improving the interfacial linkages between the matrix and wood filler.

CONCLUSIONS

Thermoplastic materials are known to have high damping factor but poor stiffness. This makes them unsuitable for many applications. For structural applications, a combination of high stiffness and high damping is desirable. Filling high damping materials like polypropylene with wood provides a strategy to build newer materials with significantly high stiffness, and still having acceptable damping factors so as to make them suitable for light structure purpose. At 50% filler loading, stiffness of the composite material increased by about 100%, whereas the damping factor decreased only by 33%. Such materials may find applications in areas where reduction of vibration is required like automobile, aircraft interiors, and machinery parts. The benefits of having high damping factor are longer service life of the component, reduction in weight, and reduction in noise.

References

1. Perepechko, I. Acoustic Methods of Investigating Polymers; Mir Publishers: Moscow, 1975.
2. Harper, D. P. Ph.D. Thesis, Washington State University, USA, 2003.
3. Jayne, B. For Prod J 1959, 9, 413.
4. Behzad, M.; Tajvidi, M.; Ehrahimi, G.; Fulk, R. Int J Eng Trans B: App 2004, 17, 95.
5. Boluk, M.; Schreiber, H. Polym Comp 1986, 7, 295.
6. Lipatov, Y. Physical Chemistry of Filled Polymers, Rubber and Plastics Research Association of Great Britain, Shrewsbury, British Library, 1979.
7. Harper, D.; Wolcott, M. Compos A 2004, 35, 385.
8. Stark, N. M.; Rowlands, R. E. Wood Fiber Sci 2003, 35, 167.
9. Yin, S.; Rials, T. G.; Wolcott, M. P. The Fifth International Conference on Wood-Fiber-Plastic Composites; 1999; pp 139–146.
10. Tajvidi, M.; Falk, R. H.; Hermason, J. C.; Felton, C. The Seventh International Conference on Wood-Fiber-Plastic Composites, 2001; p 187.
11. Chauhan, S. S.; Karmarkar, A.; Aggarwal, P. J App Polym Sci 2006, 102, 1706.
12. Karmarkar, A.; Chauhan, S. S.; Modak, J. M.; Chanda, M. Compos A 2007, 38, 227.
13. Martincek, G. J Sound Vib 1965, 2, 116.
14. Nowick, A. S.; Berry, B. S. An Elastic Relaxation in Crystalline Solids; Academic Press: New York, 1972.
15. Zugenmaier, P. 2nd Annual Partnerships for Environmental Improvement and Economic Development Conference; Wood and Cellulose: Building Blocks for Chemicals, Fuels and Advanced Materials; Syracuse, New York, USA, 2000.
16. Gu, W. Ph.D. thesis, Virginia Polytechnic Institute and State University, Blacksburg, Virginia, 1997.
17. Zorowasky, C. F.; Murayama, T. In Proceedings of the 1st International Conference on Mechanical Behaviour of Materials; Society of Material Science: Japan, 1972; p 28.