Distribution function of fiber length in thermoplastic composites

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Abstract

Due to the strong correlation with material properties, fiber length distribution in polymer composites has been investigated. The distribution exhibits steep increase until peak followed by long tail decay, which has been analyzed in terms of the Weibull distribution. However, interpretation of the stochastic process behind the Weibull distribution is not trivial, particularly for the shape parameter. In this study, a simple distribution function is proposed based on the two Poisson processes at the breakage of loaded fibers; one of them is a series of memoryless events for the fiber breakage with a characteristic length λ_a, and the other one is another series of memoyless events blocking the adjacent breaks with an interval length of λ_b. The proposed function was examined against literature data for nylon-6 composites containing glass fibers and carbon fibers. For the glass fiber composites for which the processing conditions were varied at the same fiber content, the proposed function captures the experimental data as well as the Weibull distribution. The obtained λ_a shows a good correlation with the mechanical properties of the composites to hint the relation to the processing conditions. For the carbon fiber composites for which the fiber content was varied, the proposed distribution better captures the long tail decay than the Weibull distribution whereas the

Weibull distribution better captures the data around the sharp peak for high fiber contents than the proposed distribution. It so appears that the proposed function provides a straightforward analysis of the fiber length distribution in polymer composites, in particular for the long tail decay that is of importance for the material properties.

Keywords

Thermoplastic composite; fiber length distribution; Weibull distribution; Poisson process

Introduction

For fiber reinforced thermoplastics (FRTP), the fiber length and its distribution are of importance due to their strong influence to the material properties. Bowyer and Bader [1] have theoretically derived the fiber contribution in the modulus of materials that consist a group of fibers with various lengths. They showed the consistency of their theory with experimental data for glass fiber composites of nylon 66 and polypropylene. Fukuda and Chou [2] constructed the theory to derive the mechanical strength of composites from the distribution in fiber length and orientation. Fu and Lauke [3] further improved the theory to take account of the dependences of the fiber strength and the critical fiber length on the inclination angle. In these theories, the fiber length and its distribution are given a priori. But for the case of thermoplastic composites with discontinuous fibers, the fiber length and its distribution are the resultant of fiber breakage during the process, as shown by Lunt and Shortall [4], for example, and investigations of fiber breakage have been performed extensively. Czarnecki and White [5] examined the effect of compounding on the damage of glass, aramid and cellulose fibers for polystyrene composites to propose a mechanism of fiber breakage induced by the Euler buckling in shear flow. von Turkovich and Erwin [6] investigated the breakage of glass fiber in polyethylene composites with various fiber contents to suggest that the effects of inter-fiber collisions are not significant for fiber breakage. On the other hand, Fisa [7] measured the glass fiber degradation during the compounding with polypropylene to report that the inter-fiber collisions are of importance as well as the fiber-polymer interactions. He also reported that the fiber length is dependent on the work of mixing. Gupta et al. [8] have shown for polypropylene composites containing glass fiber that the fiber length distribution strongly depends on the initial fiber length before the compounding. On the contrary, Karsli et al[9] examined the effect of initial fiber length for carbon-fiber reinforced polypropylene prepared by extrusion compounding followed by injection molding to conclude that the initial fiber length has no significant influence on the fiber length distribution in the final product. Accumulation of experimental studies has induced the development on experimental methods for the fiber length measurement. For example, Fu et al [10] discussed the accuracy of image analysis to propose a correction on the measured fiber length. Miettinen et al [11] have developed the non-destructive method using X-ray microtomography. Apart from these experimental studies, theoretical attempts have been made to predict the fiber length distribution from the processing conditions. For fibrous materials in general, Meyer et al. [12] have derived the fundamental equation that describes the evolution of fiber length distribution through the breakage. Salinas and Pittman [13] investigated the breakage of isolated glass fibers under shear to show that the critical shear stress of the buckling [14] coincides with the fiber breakage condition, as earlier reported by Czarnecki and White [5] for the polystyrene composites. On the basis of the buckling-induced breakage, Shon et al. [15] constructed a kinetic model for the development of fiber length during the compounding to describe the rate constant as a function of the shear stress applied to the fiber. Inceoglu et al. [16] modified the model by Shon et al. [15] to assume that the kinetic constant is a function of the mechanical energy rather than the shear stress. Durin et al. [17] and Phelps et al [18] have taken into account of the distribution of the breakage probability along the fiber. Vas et al [19] theoretically discussed the effect of fiber length distribution on the breakage of the composite.

In spite of the attempts mentioned above, functional foam describing the fiber length distribution has not been thoroughly discussed. In some studies, number average and weight average lengths are often used to characterize the distribution. However, such measures are insufficient to describe the actual distribution that typically has a sharp peak followed by long tail decay (See Phelps et al [18] for example). In particular, the long tail portion is of importance in the framework of the critical length theory [20]. The long tail decay can be explained by the simplest assumption for the fiber breakage process in which the breakage events occur independently with the same probability (as Poission process)[19]. If the initial fiber length before the breakage is infinite, the distribution function is derived as

$$P(l;\lambda) = \frac{1}{\lambda}\exp\left\{-\frac{l}{\lambda}\right\} \tag{1}$$

Here, λ is the average fiber length. Monotonic decay distributions being similar to this exponential decay function have been reported in some cases [5,6,19]. However, most of the cases the fiber length distribution does not exhibit monotonic decay but it shows a peak at a certain short length, if the number of fibers are counted with a sufficient fine interval. To capture such a distribution, instead of eq 1, the Weibull distribution [21] (or equivalently referred to as Tung's distribution [22]) is used, for which the frequency of breakage is assumed to be dependent on the fiber length with a power-law manner;

$$P(l;\lambda,k) = \frac{k}{\lambda}\left(\frac{l}{\lambda}\right)^{k-1}\exp\left\{-\left(\frac{l}{\lambda}\right)^{k}\right\} \tag{2}$$

Here, k is the shape parameter (so-called Weibull modulus) and λ is the scale parameter. If $k = 1$, eq 2 is identical to eq 1 and the breakage occurs as Poisson process.

On the other hand, for $k > 1$, the frequency of breakage increases with increasing fiber length and the distribution has a peak being consistent with experiments, as shown by Fu et al [10,23] for example. However, the shape parameter is difficult for discussion. Jayatilaka and Trustrum [24] have theoretically shown that the shape parameter is directly related to the distribution of crack size on the fiber. They assumed that the crack size distribution decays with a power-law manner. This assumption is rationalized when the fiber strength is dominated by relatively large cracks so that the effects of crack size distribution for small cracks are negligible. However, the power-law decay assumption may not be realistic for the fibers with a rather small diameter that limits the crack size. Another difficulty is the determination of the value of k for which the Weibull plot [25] is used on the basis of the assumption of linear relationship between $\ln(\ln(1/(1 - F(l)))$ and $\ln l$ with respect to the cumulative distribution function $F(l)$. In some cases, the linear relation is not strictly achieved, and the fitting to determine the value of k is a matter of arbitrariness.

In this study, we consider a simple stochastic process for the fiber breakage in the processing of thermoplastic composites to propose a distribution function of the fiber length. The proposed functional form contains two characteristic lengths. One of them reflects the probability of breakage, which may be of characteristic for the processing, and the other stands for the blocking probability of adjacent breakage, which may be related to the fiber toughness. The proposed distribution function is examined to the fiber distribution reported in literature for nylon-6 thermoplastic composites containing glass fibers [26] and carbon fibers [27] , in comparison to the Weibull distribution. Details are shown below.

Distribution Function

Hereafter, we consider a process for thermoplastic composites in which long fibers are supplied to an extruder to be mixed with molten thermoplastic. The initial fiber length is assumed to be infinite. During the process, the fiber is broken to exhibit a length distribution in the final product. In the simplest assumption, the fiber breakage is regarded as a memoryless process (Poisson process) in which each breakage occurs independently. In this assumption, the distribution function of the fiber length l after the processing is given as eq 1. Such a breakage process is, however, unrealistic because it predicts amount of fibers with infinitesimally short lengths. In reality, the toughness of fiber prohibits the successive breakage from adjacent breaks with an infinitesimal interval, as suggested by the buckling theories[5,13]. To consider such an effect, we introduce a stochastic process in which the adjacent breakages are separated with another Poisson process. Then the distribution function becomes

$$P(l;\lambda_a,\lambda_b) = \frac{1}{\lambda_a-\lambda_b}\left[\exp\left(-\frac{l}{\lambda_a}\right) - \exp\left(-\frac{l}{\lambda_b}\right)\right] \quad (3)$$

Here, λ_a and λ_b are the characteristic lengths for the breakage and for the separation between adjacent breakages, respectively. (It is fair to note that Tzoumanekas and Theodorou [28] have introduced the distribution function given in eq 3 for the distribution of entanglement molecular weight of polymers.) According to the corresponding stochastic processes, λ_a may be characteristic of the processing (or shear stress in relation to the buckling theories) whereas λ_b may be of fiber toughness (related to the Young's modulus and the fiber diameter). The cumulative distribution for eq 3 is given as

$$F(l;\lambda_a,\lambda_b) = \int_0^l P(l';\lambda_a,\lambda_b)dl' = \frac{1}{\lambda_a-\lambda_b}\left[-\lambda_a\exp\left(-\frac{l}{\lambda_a}\right) + \lambda_b\exp\left(-\frac{l}{\lambda_b}\right)\right] \quad (4)$$

The number average and the length-weighted average are given as

$$\langle l\rangle = \int_0^\infty lP(l;\lambda_a,\lambda_b)dl = \lambda_a + \lambda_b \quad (5)$$

$$\langle l\rangle_w = \int_0^\infty l^2P(l;\lambda_a,\lambda_b)dl \Big/ \int_0^\infty lP(l;\lambda_a,\lambda_b)dl = \frac{2\left({\lambda_a}^2+\lambda_a\lambda_b+{\lambda_b}^2\right)}{\lambda_a+\lambda_b} \quad (6)$$

It is noteworthy that in the limit of $\lambda_a \to \lambda_b$ eq 3 becomes Erlang-2 distribution

(Erlang distribution with phase 2);

$$P(l;\lambda_a) = \frac{1}{\lambda_a{}^2} l \exp\left(-\frac{l}{\lambda_a}\right) \quad (7)$$

Results and discussion

In this section we examine the proposed distribution function (eq 3) for a few experimental data sets reported for nylon-6 thermoplastic composites with discontinuous fibers.

Figure 1 shows the fitting to the data for the length distribution of glass fiber processed in a polyamide 6, reported by Ularych et al [26]. The fiber content, fiber diameter and fiber length before processing were 30wt%, 13 μm and 4.5mm, respectively. Five samples were processed with different processing conditions for which the details are not available. In the figure, in addition to the linear scale plots (left panels), semi-logarithmic plots (right panels) are shown to magnify the distribution in the long tail region. The fitting parameters are summarized in Table I.

The agreement with the data for eq 3 (solid curve) is similar to Weibull distribution (broken curve) if the distribution is seen in linear scale (left panels). However, for the long tail region, the two distribution functions behave differently; the decay rate for the long fiber is larger for the Weibull distribution than that for the proposed distribution. Because the decay in the Weibull distribution reflects the shape parameter being larger than unity, it diminishes always faster than the proposed distribution. In the long tail region, the proposed distribution well describes the data for the materials I and IV whereas the Weibull distribution is consistent with the data for the material II and III.

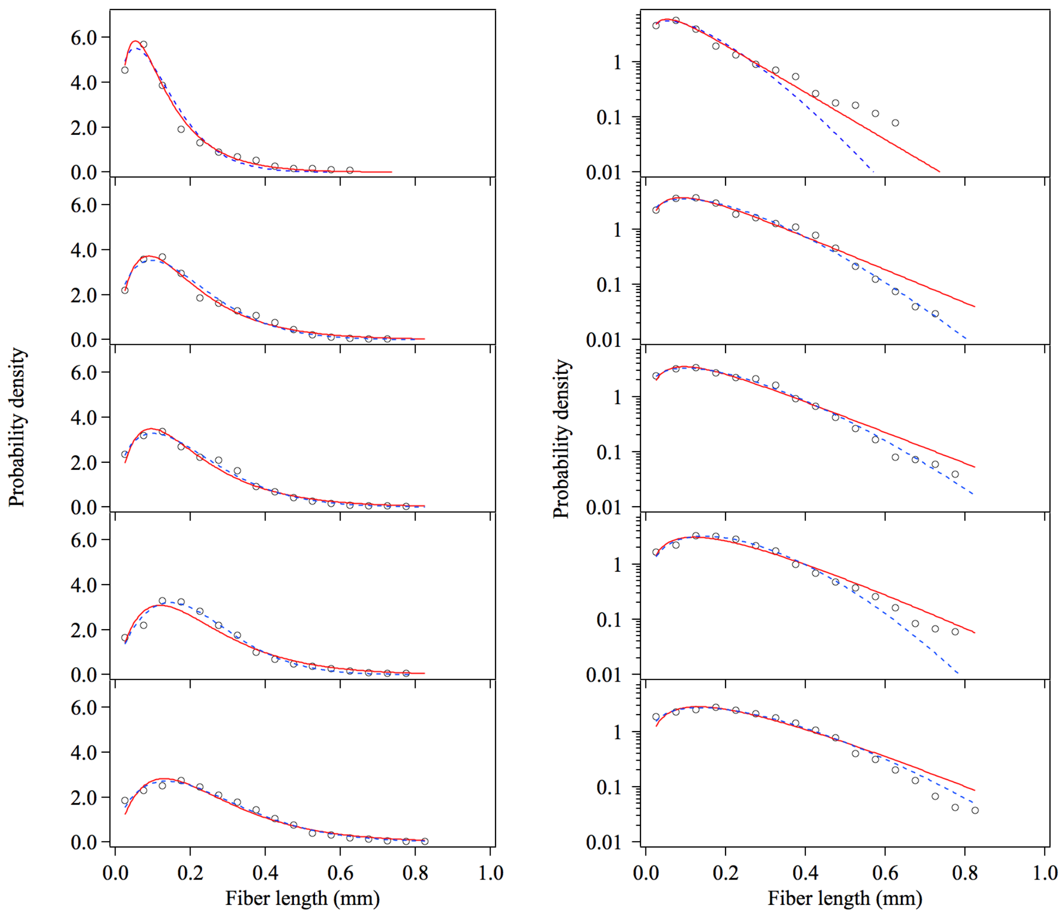

Figure 1 Glass fiber length distribution for nylon-6 composites. 5 panels from top to bottom show the results for the material I to V processed with different conditions. Circle is the experimental result extracted from the literature [26]. Solid and broken curves are eqs 3 and 2, respectively. The parameters are summarized in Table I. Right

panels show the data in semi-logarithmic plot to magnify the long tail decay.

Table I Fitting parameters for the nylon-6 glass fiber composites

Sample	(Parameters for Weibull distribution)		(Parameters for eq 3)	
	k	λ(mm)	λ_a(mm)	λ_b(mm)
I	1.39	0.06	0.03	0.10
II	1.47	0.10	0.06	0.15
III	1.47	0.12	0.06	0.16
IV	1.71	0.90	0.12	0.12
V	1.52	0.14	0.14	0.12

Figure 2 demonstrates the correlation between the parameters in the distribution functions, λ, λ_a and λ_b and the mechanical properties. Among the three parameters, λ_a shows the strongest correlation with the mechanical properties with power-law dependence. For the other parameters (λ and λ_b) the correlation with mechanical properties is relatively weak. This result is in harmony with the idea underlying the proposed distribution; λ_a is varied via the processing conditions whereas λ_b stands for the fiber properties. For the Weibull distribution the variation of the shape parameter k is rather small, as suggested from the origin of this parameter in relation to the crack distribution along the fiber. (Consequently, the correlation with the mechanical properties is rather weak and not shown here.) On the other hand, the scale parameter λ behaves similarly to λ_b, and the effect of processing condition is not visible in comparison to that in λ_a.

According to the critical length theory [20], the mechanical properties are dominated by the amount of fibers that have super-critical fiber lengths. The amount of such fibers can be quantified via the ratio of super-critical fibers, $P(L_C) = 1 - F(L_c)$, where L_c is the critical fiber length and $F(l)$ is the cumulative distribution. The calculation of $P(L_C)$ is thus straightforward for given L_c as long as the parameters of the distribution functions are given. As reported by Ularych et al [26], for the examined composite materials $P(L_C)$ shows a excellent correlation with the mechanical properties if L_c is chosen at 0.2mm for the Weibull distribution. Because the proposed distribution function is not significantly discrepant from the Weibull distribution around $l = 0.2$ mm (see Figure 1), similar correlation with the mechanical properties is observed for the proposed distribution function (not shown). Note that the result may differ for larger L_c values owing to the difference between the two distribution functions in the long tail region.

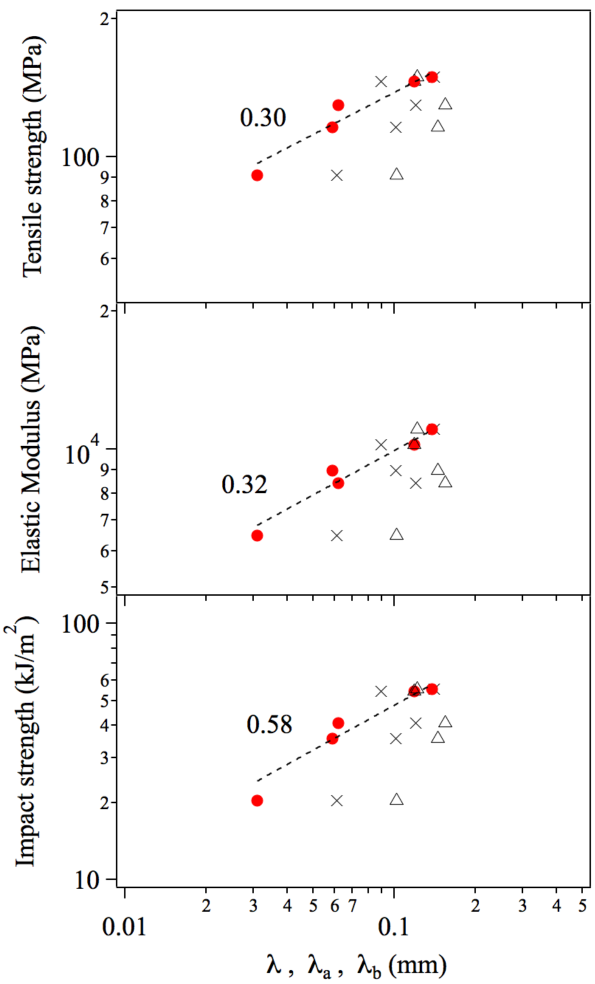

Figure 2 Correlation between the parameters for the distribution functions and the mechanical properties for the glass fiber composites[26]. Cross, circle and triangle are λ, λ_a and λ_b, respectively. Dotted line shows the power-law fitting of the relation between λ_a and the mechanical properties. The power-law exponents are indicated in the panels.

We turn our attention to a carbon fiber system [27] composed by nylon-6 and carbon fiber with the diameter of 7.2 μm. Because the material was processed via LFT-D (long fiber thermoplastic direct process), the initial fiber length was virtually infinite. The carbon fiber fraction was varied from 30wt% to 45wt%. For further details, see the literature [27].

Figure 3 shows the fiber length distribution in a similar manner to Figure 1. In the linear plot (left panels) in which the fitting around the peak is magnified, eq 2 (Weibull distribution) provides better agreement than eq 3, specifically for the materials with high fiber content (see the two panels from the bottom). Conversely, as shown in the semi-logarithmic plot (right panels), the distribution in the long tail region is better captured by eq 3 rather than eq 2. It is fair to note that the Weibull distribution may attain better fitting if we neglect the single point at the peak. But even in such a case the long-tail decay in the Weibull distribution is steeper than that in the proposed distribution owing to the shape parameter $k > 1$, as mentioned for Figure 1. The discrepancy between data and the Weibull distribution in the long tail region may be of critical importance for the evaluation of fiber length distribution with respect to the critical length theory[20]. However, it is fair to note that the data reliability in the long tail region may be not sufficient due to rather small number of fibers.

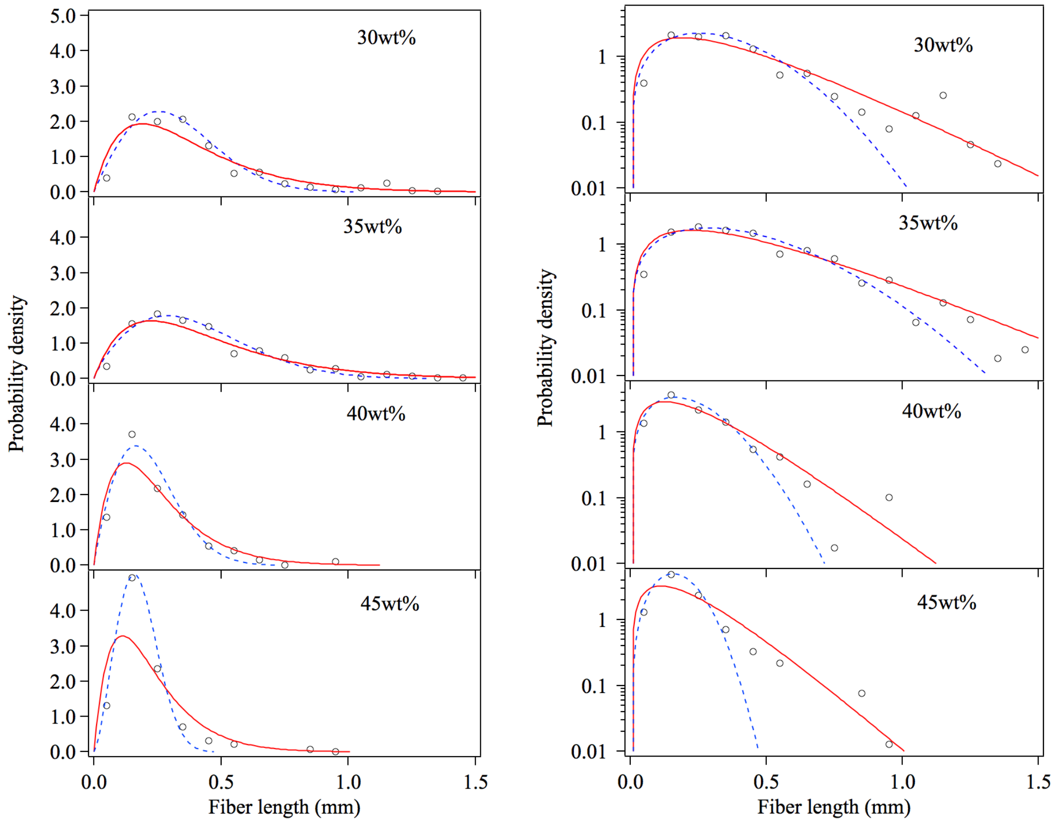

Figure 3 Carbon fiber length distribution for nylon-6 composites processed via LFT-D with various fiber contents from 30 to 45wt% from top to bottom. Circle is the experimental result extracted from the literature [27]. Solid and broken curves are eqs 3 and 2, respectively. The parameters are summarized in Table II. Right panels show the data in semi-logarithmic plot to magnify the long tail decay.

Table II Fitting parameters for the nylon-6 carbon fiber composites

Fiber content (wt%)	(Parameters for Weibull distribution) k	λ(mm)	(Parameters for eq 3) λ_a(mm)	λ_b(mm)
30	1.94	0.14	0.19	0.19
35	1.80	0.24	0.22	0.22
40	1.89	0.07	0.13	0.13
45	2.42	0.02	0.11	0.12

The parameters obtained from the fitting demonstrated in Figure 3 are summarized in Table II. Interestingly, for this specific case, λ_a and λ_b are very close to each other so that the distribution function virtually becomes the Erlang-2 distribution (eq 7), regardless of the fiber content. The coincidence between λ_a and λ_b suggests a strong correlation between the two stochastic events, which are breakage of the fiber and blockade of adjacent breaks. Owing to the strong effects from the fiber content, the correlation between the parameters and the material properties is not visible (not shown).

Conclusion

The functional form for fiber length distribution in thermoplastic fiber composites was proposed on the basis of the two Poisson processes for the fiber breakage and the blockade of adjacent breaks. The proposed function reasonably captures the experimental data for nylon-6 composite materials reinforced by glass fibers and carbon fibers. For the case of glass fiber composites for which the processing conditions were varied and the fiber content was common, the characteristic length for the fiber breakage, λ_a, has a strong correlation with the mechanical properties of the material. This observation suggests a correlation between λ_a and the processing. For the case of carbon fiber composites, the characteristic lengths for the two stochastic processes are very similar to each other, which suggest a strong correlation between the fiber breakage and the blockade of adjacent breaks. In comparison to the Weibull distribution, the proposed distribution tends to better capture the long tail decay whereas the Weibull distribution better captures the data around the sharp peak. The data reproducibility of the proposed function in the long tail decay may be of advantage in the framework of the critical length theory in which the longer fibers dominate the material properties. Nevertheless, further comparison to the other experimental data is apparently required for evaluation on the applicability of the proposed function. In particular, experimental data with sufficient quality in the long tail region are necessary. The limitation of the proposed function is the assumption for the infinite fiber length as the initial condition. The functional form for the initial conditions with finite fiber length is to be derived. The relation to the kinetic theories based on the fiber buckling and the fiber properties are the other direction to be investigated. The studies toward such a direction are in progress and the results will be published elsewhere.

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