Study of the Effect of Regrinding on the Cumulative Damage to the Mechanical Properties of Fiber-Reinforced Nylon 66

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Synopsis

An experimental study of the tensile strength of nylon 66 regrind, with and without short-fiber reinforcement, is reported. For nylon 66 without reinforcement, a uniform increase in tensile strength has been observed with increasing number of moldings. This is due to a further condensation reaction which occurs during molding. For short fiber-reinforced nylon 66 composite, the tensile strength decreases with increasing number of moldings, which is attributed to the cumulative breakage of fiber length through mechanical working. A model based on fiber length analysis is proposed, and the semiempirical equation can be used to predict experimental results and balance cost–end use properties.

INTRODUCTION

Glass fiber–reinforced thermoplastics have been used in the plastic industry for many years mainly because of their excellent combination of mechanical strength, chemical resistance, and moderate cost. By incorporating a small proportion of glass fiber into a thermoplastic resin, mostly in the range 10% to 40%, the tensile strength, modulus, dimensional stability, moisture resistance, corrosion resistance, electrical properties, etc., can be improved dramatically.[1,2] The enhanced properties are closely related to the amount of fibers incorporated as well as the properties of the fiber itself such as, for example, the fiber length, diameter, size, critical length, and fiber strength.[3–5] However, one of the common problems in this phase of industry is the breakage of the fibers due to the mechanical working during the molding process; this always prevents the end product from retaining its maximum enhancement properties. In practical applications, injection molders also often find themselves needing to recycle some reground material from runners, sprues, and rejected parts to maintain a lower cost. A further loss of properties due to excess use of this reground material has been observed.[6] It is thus important for injection molders to search for an optimum molding condition in order to meet the required end product performance.

This report describes the relationship between the composite ultimate tensile strength and variables such as regrind ratio, volume fraction of the fiber, fiber length, etc. Through the simple model we proposed, the injection molders should be able to optimize these variables to meet their cost–performance requirements.

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PROPOSED MODEL

For composite of continuous filament, the tensile strength parallel to the fiber axis can be expressed by a law of mixtures such as

$$\sigma_{uc} = \sigma_f V_f + \bar{\sigma}_m V_m \tag{1}$$

However, in the case of discontinuous fibers, this relationship needs to be modified due to the fact that it is necessary for the resin to bear the load first and then transfer part of it to the fiber through the matrix–fiber interfacial shear stress. Thus, the effective surface area of the fiber, i.e., the diameter and the fiber length, is important in this case. The average tensile stress and thus the strength of the composite is always less than that found for continuous fibers. Using a matrix–fiber stress transfer model, Kelly and Tyson[5] arrived at the following equations:

$$\sigma_{uc} = \sigma_f V_f \left(1 - \frac{L_c}{2L}\right) + \bar{\sigma}_m V_m \qquad L \geqslant L_c \tag{2a}$$

$$\sigma_{uc} = \frac{\tau L}{d} V_f + V_m \bar{\sigma}_m \qquad L < L_c \tag{2b}$$

where $L_c = \sigma_f d / 2\tau$.

Values of critical length L_c have been estimated[7] to be about 8 mils (0.2 mm) for commercial fibers. It is apparent that in order to enhance the composite strength, the average fiber length in the matrix should exceed the critical length by as much as possible without interfering with the processibility. In actuality, fibers are not uniform in length; rather, they are distributed over a mean average length. Here, we assume that the tensile strength of the composite can be treated with the above equations by using the average fiber length instead of summation over contributions from each individual fiber. Our immediate concern is then to understand how this average fiber length would be affected by the mechanical working during injection molding. Filbert[6] has investigated the breakage of average fiber length in glass-reinforced nylon 66 regrind molding. He found that decreasing the real zone temperature or increasing the screw speed and/or back pressure will significantly reduce the average fiber length, thus lowering the tensile strength, Izod impact, and flexual modulus. It has also been found that[6] using 100% regrind material, the average fiber length decreased exponentially to an asymptotic limit, which is near the critical length, with increasing number of remolding.

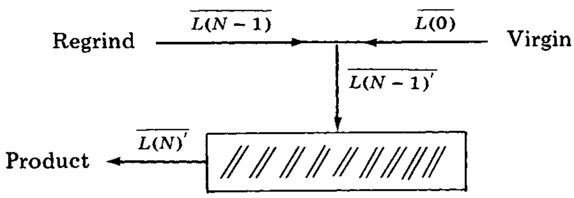
Assume that the average fiber length after the Nth 100% regrind molding, $\overline{L(N)}$, can be expressed as

$$\begin{aligned} \overline{L(N)} &= a \exp(-\alpha N) + b \qquad N = 1, 2, 3, \ldots \\ &= [\overline{L(N-1)} - b] \exp(-\alpha) + b \end{aligned} \tag{3}$$

where $a + b = \overline{L(0)}$ is the initial average fiber length used in the virgin material. The asymptotic limit b and parameters a and α are strongly affected by molding conditions, and their numerical values can be obtained from the intercept and slope of the plot $\ln(\overline{L(N)} - b)$ versus N.

In the case where virgin material is used to mix with the regrind, the average fiber length in each molding could be estimated as follows: Define a regrind ratio

R as the fraction of regrind in the mixed material. Referring to the scheme below,



$$R = \frac{\text{Regrind}}{\text{Regrind + Virgin}}$$

the average fiber length before Nth molding $\overline{L(N-1)'}$ could be expressed as:

$$\overline{L(N-1)'} = \overline{L(N-1)}R + \overline{L(0)}(1-R) \tag{4}$$

where $\overline{L(N-1)}$ is the average fiber length of the reground material. From eqs. (3) and (4), we have

$$\begin{aligned} \overline{L(N)'} &= [\overline{L(N-1)'} - b]e^{-\alpha} + b \\ &= [\overline{L(N-1)}R + \overline{L(0)}(-R) - b]e^{-\alpha} + b \\ &= aR\, e^{-N\alpha} + a(1-R)e^{-\alpha} + b \end{aligned} \tag{5}$$

Thus when $R = 0$, $\overline{L(N)'} = a\, e^{-\alpha} + b$, which is the case when no regrind material is used, and the average fiber length in the product is independent of the number of moldings N. When $R = 1$, corresponding to 100% regrind molding, then the above equation is reduced to $\overline{L(N)'} = a\, e^{-N\alpha} + b$, which is equivalent to eq. (3). Thus, for injection molding with regrind ratio R, the composite tensile strength can be estimated using the following relation:

$$\overline{\sigma_c(N)'} = (\eta\sigma_f V_f + \bar{\sigma}_m V_m) - \eta\sigma_f V_f L_c/(2\overline{L(N)'}) \tag{6}$$

where $\overline{L(N)'}$ is given by eq. (5) and η is introduced into the equation to account for the effect due to the fiber orientation which causes deviation from total alignment.[8]

RESULTS AND DISCUSSION

Two kinds of injection-molded nylon 66 plastics tensile bars as furnished by AMP Corporation were studied in this report: Monsanto M-340, which only contains flame retardent, and Monsanto Vidyne 909, which contains both flame retardant and 20 wt. % (= 11 vol. %) fiber glass. The initial average fiber length, $\overline{L(0)}$, used in Vidyne 909 is 30.2 mils. The tensile test were performed according to the ASTM D-638 method with a strain rate equal to 0.5 cm/min. The average fiber length analysis was performed by using a Quantimate 720 (Cambridge, Imanco), which gives number-average fiber length. Figures 1 and 2 show results of composite tensile strength and average fiber length analysis as a function of the number of moldings. Figure 1 shows that while Vidyne 909 experiences a decrease in tensile strength, Monsanto M-340 shows a slight increase with increasing number of remoldings. The latter phenomenon may be due to further

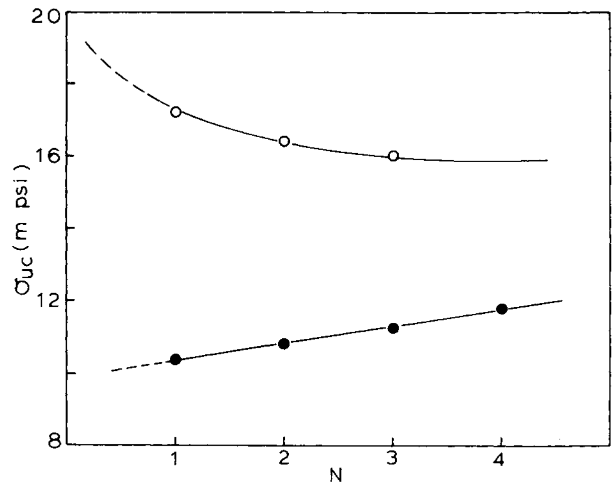


Fig. 1. Tensile strength as function of number of moldings: (O) Vidyne 909 with fiber reinforcement; (●) Monsanto M-340 without reinforcement.

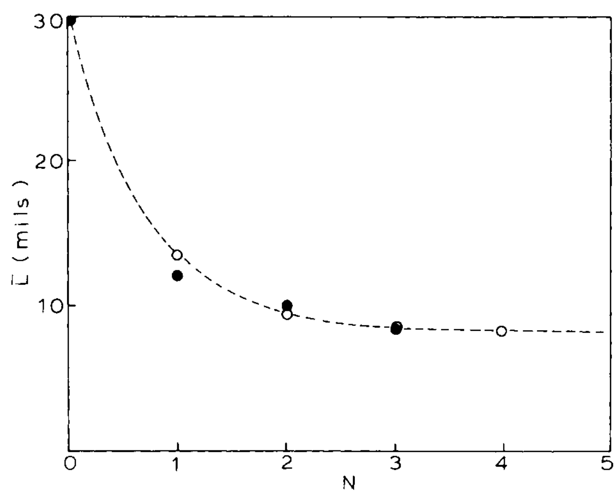


Fig. 2. Average fiber length as function of number of moldings: (●) data from experiment; (O) simulated from eq. (3) with a = 22 mils, b = 8.2 mils, and α = 1.35.

condensation reaction in the polymer which eliminated water or other byproducts from the resin during injection molding. Thus, the real damage to the tensile strength due to fiber breakage should be corrected for this effect, and the real values are shown in Figure 3. From Figure 2 and using eq. (3) to fit the data, we found that a = 22 mils, b = 8.2 mils, α = 1.35 (see dotted curve). Here, we should note that the asymptotic limit length b is close to the critical length L_c shown in the literatures. Thus, we employ eq. (5) together with eq. (6) and let R = 1, to simulate the composite tensile strength. The result agrees well with experimental data as shown in Figure 3 (dotted curve). $\eta\sigma_f V_f$ and $\overline{\sigma}_m V_m$ were found to be 1.15×10^4 and 8.9×10^3 psi, respectively. Ideally,[7] σ_f could be as high as 2.5×10^5 psi; this means the orientation factor η in this case is equal to 0.42. In other words, only 42% of the fibers effectively enhance the tensile strength along the tensile axis. It has been shown from x-ray studies[3] that an injection-molded

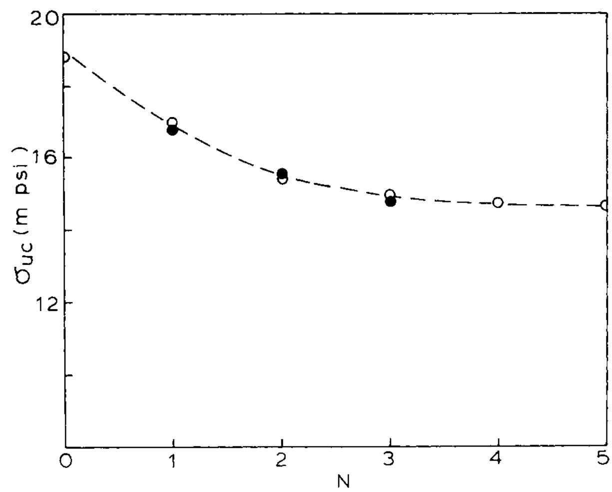


Fig. 3. Tensile strength as function of number of moldings: (●) Vidyne 909 with 20 wt. % fiber glass; (○) simulated using eqs. (5) and (6), $R = 1$, $\eta\sigma_f V_f = 1.15 \times 10^4$ psi, $\bar{\sigma}_m V_m = 8.9 \times 10^3$ psi; a, b, and α are the same as in Fig. 2.

tensile bar may have between 33% and 90% of the fibers oriented along the tensile axis, while the rest of the fibers are oriented along the flow line fronts of the interior. Using extrusion molding with the smallest die through which the melt would flow without excessive plugging will give samples which provide maximum orientation along the tensile axis.[3]

The good agreement between the predicted and the experimental result demonstrates the usefulness of the idea of employing the average fiber length to estimate the composite tensile strength.

Using variables obtained from the above experiments, we can demonstrate the effect of regrind ratio R on the tensile strength of the composite. This is shown in Figure 4. When $R = 0$, no regrind material is used in the feed, as one would expect, and the composite tensile strength is independent of the number

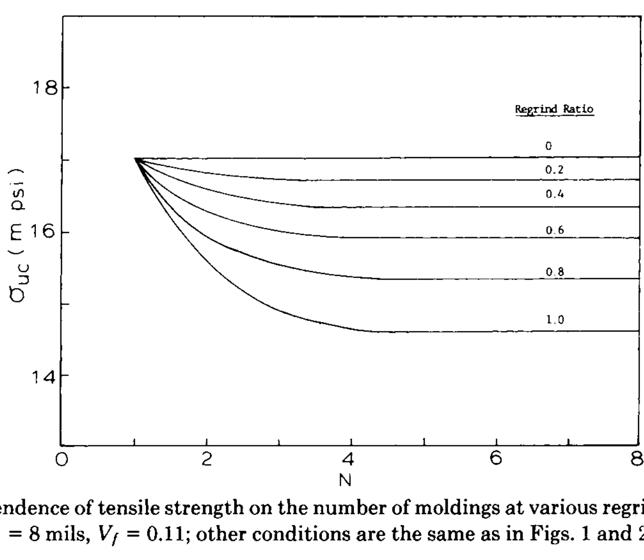


Fig. 4. Dependence of tensile strength on the number of moldings at various regrind ratios; $L(0) = 30.2$ mils, $L_c = 8$ mils, $V_f = 0.11$; other conditions are the same as in Figs. 1 and 2.

of molding. However, as R increases, the dependence become more evident, and the final property which corresponding to N equal to infinity decreases sharply with increasing amount of regrind material used.

The amount of fibers used and their average fiber length also play an important role in the composite tensile strength. Assuming $R = 0.5$, Figure 5 displays the effect of volume fraction V_f and the initial fiber length $\overline{L(0)}$ on the composite tensile strength. As one realizes, the composite tensile strength increases dramatically with increasing amount of fibers incorporated into the resin. It should also be noted that for a lower fraction of fibers in the resin, varying the initial average fiber length does not change the composite tensile strength appreciably, unlike the case of a composite with high fiber content, which has a stronger dependence on $\overline{L(0)}$.

The regrind ratio R is also important in determining the final properties.

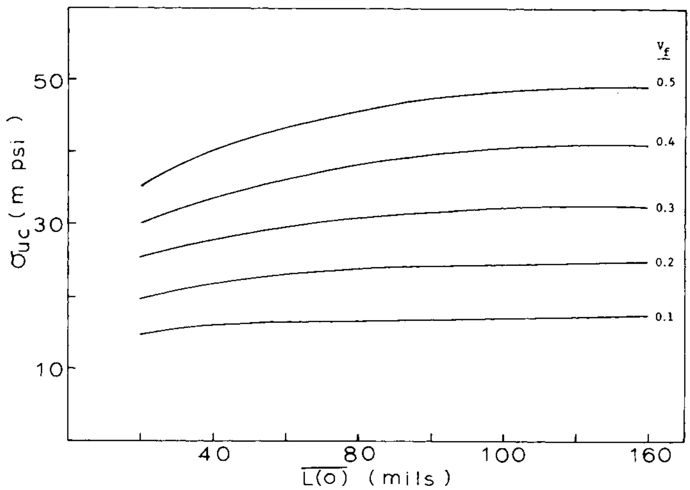


Fig. 5. Dependence of tensile strength on the average initial fiber length at various fiber fractions; $R = 0.5$, $N = \infty$, other conditions are the same as in Figs. 1 to 3.

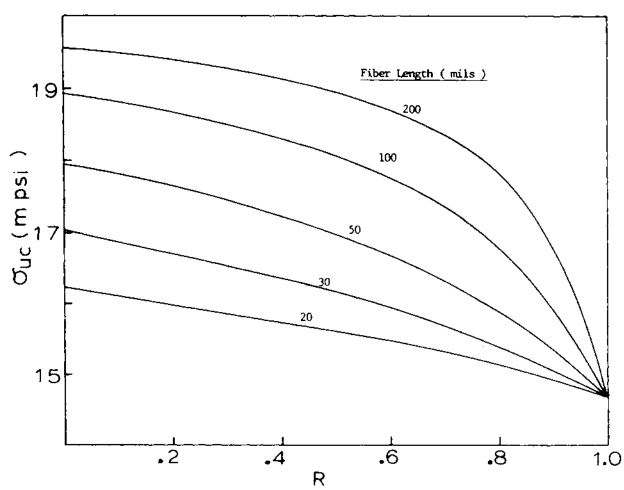


Fig. 6. Dependence of tensile strength on the regrind ratio at various average initial fiber lengths. All conditions are the same as in previous figures.

Figure 6 shows this effect. For any chosen fiber, the composite tensile strength decreases with increasing regrind ratio, and the effect is even more prominent if more fiber is used in the resin, as can be predicted from Figure 5. Figure 6 together with Figure 5 could be used as references to choose the kind of fibers and the amount needed in order to meet the required product performance.

Figure 7 displays the dependence of tensile strength on the parameters α and R, with b equal to 8.2 mils. Both b and α would be affected by molding conditions. The whole spectrum will shift to a higher tensile strength without changing very much the shape itself as the minimum length b increases. It is thus important for a molder to establish functional relationships between parameters α and b and molding conditions such as screw speed, back pressure, real zone temperature, and other adjustable conditions.[9] The most ideal condition is, of course, to yield the smallest α and largest b possible.

All of the above predictions are based upon the assumption that the critical length L_c is equal to 8 mils. Theoretically, L_c could also be varied with different coupling agents applied to the surface of the fibers. The effect of varying the critical length on the tensile strength is displayed in Figure 8, where weak dependence is observed for low fiber content. When the fiber content is high, the tensile strength decreases appreciably with increasing critical fiber length, which results from weak bonding between the fiber and the resin.

Finally, we should point out that this report only demonstrates the usefulness of the proposed model in predicting the composite tensile strength from various parameters. Serious injection molders should study regrind materials to at least six remoldings. From average fiber length analysis together with the tensile measurement for each remolded sample, accurate values of parameters such as η, σ_f, $\overline{\sigma}_m$, α, and b could then be obtained. A further study on this model will be to measure the tensile strengths for samples with various regrand ratios. From the comparison between the measured properties and the theoretical predictions, a rigorous test of the present model can then be obtained.

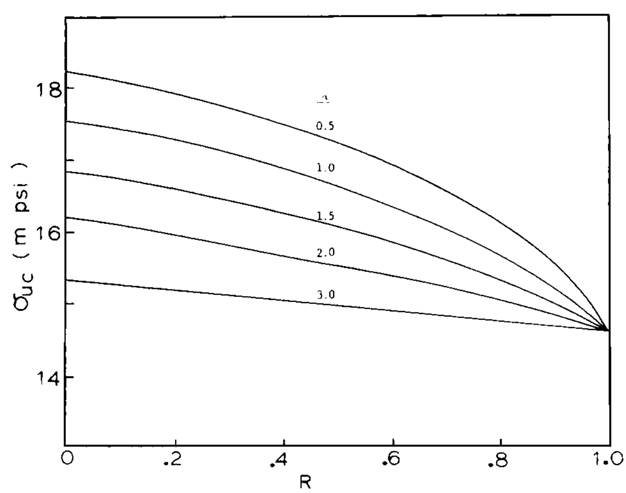


Fig. 7. Dependence of tensile strength on the regrind ratio at various parameters α. Conditions are the same as in previous figures.

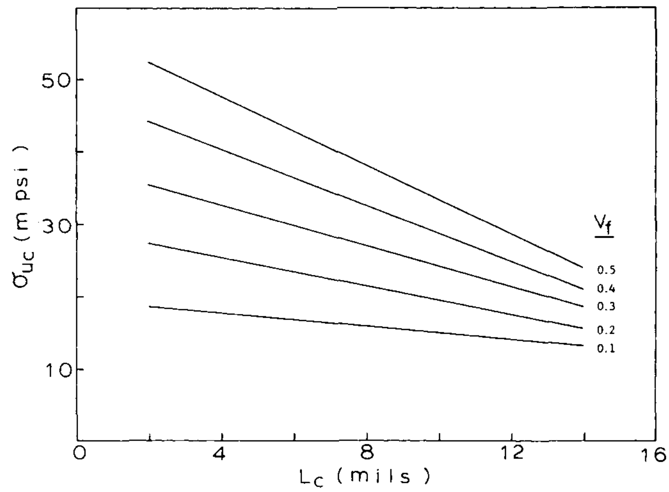


Fig. 8. Dependence of tensile strength on the critical length at various fiber fractions. Other conditions are the same as in previous figures.

Notation

σ_{uc} ultimate tensile strength of the composite
σ_f tensile strength of glass fibers
$\bar{\sigma}_m$ tensile strength of matrix at strain corresponding to composite failure
V_f volume fraction of glass fibers
V_m volume fraction of matrix
L length of glass fibers
L_c critical length of glass fibers
d diameter of fiber
τ strength of adhesive bond between fiber and matrix
η orientation factor
R regrind ratio
$\bar{L}$ average fiber length
b minimum fiber length
α molding parameter

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