Characterization of Fatigue Strength and Damage Behavior in Glass-Particle-Reinforced Nylon 66 Composites*

Keiichiro TOHGO**, Yasuo ITOH** and Taewoo KIM**
**Department of Mechanical Engineering, Shizuoka University,
3-5-1, Johoku, Hamamatsu 432-8561, Japan
E-mail: tmktoug@ipc.shizuoka.ac.jp

Abstract
This paper deals with fatigue strength and damage behavior of glass-particle-reinforced Nylon 66 composites. Fatigue tests are carried out on seven kinds of glass-particle-reinforced Nylon 66 composites in which a volume fraction of glass particles and interface treatment between particles and matrix are changed. The fatigue strength is low in the interface-untreated composite and is high in the interface-treated composite as compared with that of the Nylon 66. Variation of stiffness during fatigue is determined from the equilibrium stress-strain relations obtained by multi-step relaxation tests after given stress cycles, which eliminate the influence of viscous deformation of the Nylon 66 matrix. The stiffness reduces significantly at the early stage of stress cycles and then becomes constant in the Nylon 66 and both composites. In the fatigue tests under the applied stress for the same fatigue life, the stiffness reduction is more remarkable in both composites than in Nylon 66 because of the debonding damage. From the variation of the equilibrium stress-strain relations during fatigue, it is found that the viscous component of deformation gradually disappears with increasing stress cycles.

Key words: Particle-Reinforced Composites, Polymer Matrix Composites, Fatigue, Viscous Deformation Behavior, Equilibrium Stress-Strain Relation, Debonding Damage

1. Introduction
The technique of improving mechanical performance of polymeric materials by dispersing inorganic particles or rubber particles has been widely applied to cost effective polymer-matrix composites. It is generally supposed that in the case of perfect bonded interface between particles and matrix the stiffness and yield stress of the composites are enhanced by hard particles and are reduced by soft particles. In the composites, however, a variety of damage modes such as fracture of particles, interfacial debonding between particles and matrix, and shear yielding and cracking in matrix develop from early stage of deformation under monotonic and cyclic loads. As these damage modes strongly affect the deformation and fracture process of the composites, mechanisms underlying in achieved mechanical performance are more complex. The dominant damage mode depends on the relative strength of the particle, matrix and interface, as well as on the external loading. Therefore, in order to extent the application of the composites to a variety of engineering components and to develop the optimized high performance composites, it is important to make clear systematically the roles of particles and damage playing during deformation and fracture processes (1)-(21).
From the viewpoint mentioned above, many investigations have been carried out on the mechanical performance of particle-reinforced polymeric composites. Especially, much attention has been paid in toughening of epoxy-matrix composites because epoxies exhibit relatively high tensile strength and low fracture resistance. For rubber-particle-reinforced epoxies, it is well accepted as common knowledge that the toughening is achieved by enhanced shear yielding in the epoxy matrix after void formation due to damage of rubber particles \(^{(5)}\), \(^{(6)}\). On the other hand, for inorganic-hard-particle-reinforced polymeric composites, the toughening effect of particles and its mechanism still remain as controversial topics to be discussed. Several toughening mechanisms of the composites have been proposed \(^{(7)-(13)}\), which are microcracking, crack pinning, shear yielding, debonding of particles, crack bridging, crack deflection, crack front bowing, and so on. The achieved mechanical performance and major toughening mechanism depend on the intrinsic properties of the matrix and particles and the interfacial strength. Based on the observation of micro-mechanical deformation and damage on the fracture surface in glass-particle-reinforced epoxies, Lee et al. \(^{(10)}, \,(11)\) concluded that the matrix shear yielding is one of the major energy absorbing mechanisms and the fracture toughness does not depend on the surface treatment of glass particles. Kawaguchi et al. \(^{(12),(13)}\) reported for glass-particle-reinforced epoxies that the moisture absorption decreases the yield stress of the epoxy matrix and the interfacial strength, and the toughness is improved by the debonding damage and increased matrix ductility on the moisture exposed composites.

The investigations on inorganic-particle-reinforced thermoplastic-matrix composites are relatively limited \(^{(14)-(18)}\). Tohgo et al. \(^{(16)-(18)}\) investigated the roles of intact particles and debonding damage on the tensile strength and fracture toughness in glass-particle-reinforced Nylon 66 composites, and derived the conclusions as follows: The tensile strength is enhanced by the intact hard particles and is reduced by the debonding damage, while the fracture toughness is reduced by the intact hard particles due to high stress concentration at a crack-tip and is enhanced by the debonding damage due to the energy dissipation and stress release around a crack-tip. Furthermore, according to the detailed numerical analysis of damage around a crack-tip in elastic-matrix and elastic-plastic-matrix composites based on a damage theory \(^{(19),(20)}\), Tohgo et al. \(^{(21)}\) suggested that the toughening due to debonding damage is questionable in the elastic-matrix composites while it is likely in the elastic-plastic-matrix composites. From these results, it is imagined that the toughening due to debonding damage and successive matrix yielding is more effective in the more ductile matrix composites such as thermoplastic composites and moisture exposed epoxy composites.

The strength and damage behavior in particle-reinforced polymeric composites under cyclic loading are also important in the application to engineering components. Investigations on fatigue behavior of the composites are very limited comparing to those on the mechanical performance under static loading \(^{(22)-(28)}\). Most of them were concerned about the fatigue crack growth behavior on the same context as toughening under static loading \(^{(22)-(28)}\). Deformation and damage behavior before main crack formation in the composites under cyclic loading are also major interests for researchers, engineers and material developers \(^{(17),(29)}\). Stiffness reduction during cyclic loading is occasionally used to identify the fatigue degradation of materials. However, because polymeric materials exhibit viscous deformation, the stress-strain relation and apparent stiffness depend on the strain rate or frequency of cyclic loading. In order to obtain the stiffness reflecting the damage during fatigue, some consideration is necessary in measuring stiffness of polymeric materials.

In the present report, fatigue strength and damage behavior of the glass-particle-reinforced Nylon 66 composites have been investigated. Fatigue tests are carried out on seven kinds of glass-particle-reinforced Nylon 66 composites, in which a volume fraction of glass particles and interface treatment are changed. Stiffness reduction is
obtained by measuring the viscous deformation behavior during fatigue tests. The influence of the particle volume fraction and debonding damage on the fatigue strength is discussed from a viewpoint of the stiffness reduction and viscous deformation behavior.

2. Materials and Experimental Procedure

The raw materials were a Nylon 66 (Leona 1302S, Asahi Kasei Corp.) and two types of spherical glass particles (GB210, Toshiba Ballotini Corp.) with and without surface treatment. An average diameter of the spherical glass particles was 17\( \mu \)m, and the surface treatment of glass particles was given with the silane-coupling agent (Sila-Ace S330, Chisso Corp.) to improve the interfacial strength between the particles and matrix. A matrix material (Nylon 66) and six kinds of glass-particle-reinforced Nylon 66 composites were fabricated by injection molding technique. In the composites, a glass particle volume fraction was varied in 10%, 20% and 30%, and treated or untreated glass particles were used. The composites containing untreated particles are referred to as interface-untreated composite, while the composites containing treated particles are referred to as interface-treated composite. Furthermore, these materials are represented by GN-0 for the Nylon 66, and by GN-10, GN-20 and GN-30 for the interface-untreated composites, and by GNIT-10, GNIT-20 and GNIT-30 for the interface-treated composites, using a number of the particle volume fraction. As the Nylon 66 is highly hygroscopic, the materials were dried in a thermo chamber at 100\( ^\circ \)C for 8 hours before tests.

As polymeric materials generally exhibit the viscous deformation, their stiffness depends on strain rate under monotonic loading and on frequency under cyclic loading. To describe the viscous deformation of polymeric materials, an over stress viscoplastic model which was extended from a viscoelastic model by Krempl et al.\(^{(30)}\),\(^{(31)}\) has been well used. In the over stress viscoplastic model, a total strain rate is given as the sum of elastic strain rate and viscoplastic strain rate:

\[
\dot{\epsilon} = \dot{\epsilon}_e + \dot{\epsilon}_p = \frac{\sigma}{E^*} + f(\sigma - \sigma^*)
\]  

where \(E^*\), \(\sigma\) and \(\sigma^*\) are the Young’s modulus, applied stress and equilibrium stress, respectively. The function \(f(\sigma - \sigma^*)\) is the viscoplastic strain rate given as a function of over stress \(\sigma - \sigma^*\). Sun et al.\(^{(32)}\)-(\(^{(34)}\) developed the constitutive model to describe the time-dependent nonlinear deformation of uniaxial-fiber-reinforced polymer-matrix composites based on the over stress viscoplastic model. The purpose of the present investigation is not to determine the constitutive model of the present materials, but is to discuss the material degradation during fatigue by variation of the equilibrium stress-strain relation which is obtained by using the concept of the over stress viscoplastic model\(^{(34)}\). As the viscoplastic strain rate is given as a function of the difference between the applied stress and equilibrium stress, the equilibrium stress is obtained by stress relaxation test to \(\sigma = \sigma^*\).

Figure 1 shows the configurations of two kinds of dog-bone shape smooth specimens. Multi-step relaxation tests during tensile loading-unloading were carried out on the specimen as shown in Fig. 1 (a) at 1mm/min of crosshead speed to obtain the equilibrium stress-strain relations for three kinds of materials (GN-0, GN-10 and GNIT-10). The stress-strain relations of the composites were obtained from the load-elongation curves, where the elongation of gauge length 25mm was measured by an extensometer. Figure 2 shows an example of the stress-strain curves obtained by the multi-step relaxation tests. When the tensile stress was applied to the composites at 1mm/min crosshead speed up to a certain stress level and then the crosshead was fixed, the applied stress gradually decreased down to the equilibrium stress by stress relaxation. In the present study, 20 minutes was adopted as the relaxation time enough to reach the equilibrium state. By several steps of the relaxation tests during tensile loading-unloading, the equilibrium stress-strain relation eliminating the influence of viscous deformation was obtained. The multi-step relaxation
tests were carried out for two or three cycles of tensile loading-unloading to the final failure.

Fatigue tests were carried out on the specimen as shown in Fig. 1 (b) by a sinusoidal cyclic load with stress ratio of 0.15 and frequency of 0.05 to 3Hz to obtain the S-N curves for the seven kinds of materials. Furthermore, for the three kinds of materials (GN-0, GN-10 and GNIT-10), fatigue tests were carried out again on the specimen as shown in Fig. 1 (a) under the applied stress for the fatigue life of $10^3$ cycles, and then during fatigue the multi-step relaxation tests were conducted on the given load cycles such as the first, 10th,

---

![Diagram](image1)

(a) Specimen for multi-step relaxation tests

![Diagram](image2)

(b) Specimen for Fatigue tests

Fig. 1 Specimen configurations.

![Diagram](image3)

Fig. 2 Stress-strain relation by multi-step relaxation test during tensile loading-unloading

![Diagram](image4)

Fig. 3 Multi-step relaxation test during fatigue test.
50th, 100th cycle and so on, as shown in Fig. 3. The multi-step relaxation tests were performed after the relaxation for 20 minutes under stress free to remove the influence of viscous deformation due to cyclic loading so far. A crosshead speed was 6mm/min, which corresponds to 0.05-1 Hz, for the fatigue tests and 1mm/min for the multi-step relaxation tests.

3. Results and Discussions

3.1. Equilibrium stress-strain relations under tensile loading-unloading

Tensile strength and damage behavior of the present materials were previously investigated in the references (16),(18). Although the main damage mode is debonding damage between the glass-particles and Nylon 66 matrix in both interface-treated and untreated composites, in the interface-treated composite the debonding damage is reduced because of high interfacial strength. The tensile strength of the present materials is shown as a function of the particle volume fraction in Fig. 4. The tensile strength in Fig.4 is slightly high as compared with that in the previous reports (16),(18). This is supposed to be the influence of moisture absorbed in the composites. The present materials were dried in a thermo chamber at 100\(^\circ\)C for 8 hours before tests while the materials in the previous reports were tested without special treatment for drying. However, the influence of the particle volume fraction and interface treatment on the tensile strength is almost the same in the present and previous materials; namely with an increase in the particle volume fraction, the tensile strength increases up to 10% of particle volume fraction and then slightly decreases in the interface-treated composites, and decreases in the interface-untreated composites.

![Fig. 4  Tensile strength as a function of particle volume fraction.](image)

Figure 5 shows the equilibrium stress-strain relations for GN-0, GN-10 and GNIT-10 obtained by the multi-step relaxation tests. The equilibrium stress-strain relations correspond to the stress-strain relations under the loading-unloading at the infinitesimal strain rate, and exhibit the deformation properties containing the influence of the particles and damage but eliminating the influence of viscous deformation of the matrix. The equilibrium stress-strain relation is high in GNIT-10 by the glass particles, and low in GN-10 by the debonding damage as compared with that of GN-0. Two kinds of stiffness, namely the Young’s moduli, \(E\) and \(E^*\) are defined from the conventional stress-strain relation and equilibrium stress-strain relation under tensile loading as in Fig. 2, and are shown as functions of the strain before unloading in Fig. 6. As \(E\) is affected by a strain rate, \(E\) is higher than \(E^*\). At the initial state before straining, \(E\) and \(E^*\) of GN-10 and GNIT-10 are higher than those of GN-0 because of the stiffening effect due to the glass particles. The variation of \(E^*\) with the strain reflects the influence of the damage developed in the materials. The gradual decrease in \(E^*\) of GN-0 and GNIT-10 seems to reflect the
development of microscopic-level damage in Nylon 66 matrix, and the rapid decrease in $E^*$ of GN-10 and the difference in $E^*$ between GN-10 and GNIT-10 correspond to the evolution of the debonding damage between the particles and matrix in GN-10.

3.2. Fatigue strength

Figure 7 shows the S-N curves of the composites obtained by fatigue tests. The data of tensile strength are also plotted against $1/4$ stress cycle in Fig. 7. From this figure, the fatigue strength for the fatigue life of $N=10^2$ and $10^5$ cycles is obtained as a function of the particle volume fraction as shown in Fig. 8. The fatigue strength in the interface-treated composites is higher than that in the interface-untreated composites, and this difference increases with an increase in the particle volume fraction. The influence of the particle volume fraction and interface treatment on the fatigue strength in Fig. 8 is similar to that on the tensile strength in Fig. 4.

3.3. Variation of equilibrium stress-strain relation during fatigue

For GN-0, GN-10 and GNIT-10, the fatigue tests were carried out again under the applied stress for fatigue life of $10^3$ cycles, and the equilibrium stress-strain relations obtained by the multi-step relaxation tests for the several fatigue stages are shown in Fig. 9. The applied stress and number of cycles to failure were $\Delta \sigma=53.0$ MPa and $N_f=999$ cycles for GN-0, $\Delta \sigma=51.3$ MPa and $N_f=1371$ cycles for GN-10, and $\Delta \sigma=67.0$ MPa and $N_f=586$ cycles for GNIT-10, respectively. It should be noted that under this fatigue condition the applied stress is much higher in GNIT-10 than in GN-0 and GN-10. On three materials, the equilibrium stress-strain relation gradually changes with an increase in stress cycles. To
characterize the variation of equilibrium stress-strain relation during fatigue, the Young’s modulus $E^*_N$, maximum equilibrium stress $\sigma^*_{\text{Nmax}}$ and accumulated strain by fatigue ratcheting $\varepsilon^*_{\text{N}}$ are determined from the equilibrium stress-strain relation for the Nth stress cycle as shown in Fig. 10. Then normalized Young's modulus $E^*_N/E^*_1$, normalized maximum equilibrium stress $\sigma^*_{\text{Nmax}}/\sigma^*_{\text{max}}$ and accumulated strain $\varepsilon^*_N$ are shown as functions of stress cycles in Figs. 11, 12 and 13, respectively. The stiffness reduces significantly at the early 10 stress cycles and then becomes constant on the Nylon 66 and both composites as shown in Fig. 11. The stiffness reduction is more remarkable in both composites than in the Nylon 66, namely 50% of the initial stiffness in GN-10 and GNIT-10 and 80% in GN-0. The stiffness reduction in both composites seems to be attributed to the debonding damage between the particles and matrix. On the other hand, the stiffness reduction in GN-0 seems to be caused by the microscopic-level damage in Nylon 66. The similar behavior of stiffness reduction in GN-10 and GNIT-10 indicates that the debonding damage even in the GNIT-10 becomes easy to occur under cyclic loading. However, it is speculated from the results of tensile loading-unloading tests in Fig. 6 that the stiffness reduction in GN-10 is more rapid than in GNIT-10 during the early 10 stress cycles.

As shown in Fig. 9, the equilibrium stress-strain relation for the first stress cycle exhibits the significant nonlinear deformation and low maximum equilibrium stress, and then this nonlinearity of deformation decreases and the maximum equilibrium stress increases with an increase in stress cycles. This means that the viscoplastic component of deformation gradually disappears and the hardening occurs during fatigue. Since these are probably matrix dominant behavior, the gradual increase in the normalized maximum...
equilibrium stress is almost the same in three materials as shown in Fig. 12. The accumulated strain during fatigue is higher in both composites than in the Nylon 66 as shown in Fig. 13. This is also attributed to the debonding damage of glass particles, because the composites behave as a deformable porous material after debonding damage.

In Figs. 11 and 13, no difference between GN-10 and GNIT-10 is observed because of the fatigue tests under the applied stress for the fatigue life of $10^3$ cycles. Under this fatigue condition, as the applied stress is much higher in GNIT-10 than in GN-10, the damage evolution is supposed to be the same level in both composites. In order to generalize the present results on the fatigue degradation of the composites, the same kind of fatigue tests under different stress levels is necessary.

Fig. 9 Variation of equilibrium stress-strain relation with Fatigue.
Fig. 10  Definition of Young's modulus, maximum equilibrium stress and accumulated strain by fatigue ratcheting for the Nth stress cycles.

Fig. 11  Variation of Young's modulus with fatigue.

Fig. 12  Variation of maximum equilibrium stress with fatigue.

Fig. 13  Variation of accumulated strain with fatigue.
3.4. Fractography

Figure 14 shows the micrographs taken at the fatigue crack growth region in the fracture surfaces of GN-0, GN-10 and GNIT-10. The fracture surface of GN-0 is covered with the fatigue striation. GN-10 exhibits the appearance characterized by the smooth glass surface, traces of debonded particles and striation-like pattern. On the other hand, the fracture surface of GNIT-10 consists of the glass particles, shallow dimples and thin ridges. The difference in fracture surface between both composites suggests the difference in the microscopic process of fatigue crack growth. In GN-10, fatigue crack propagates by striation-formation mechanism in the porous matrix material after full-debonding of glass-particles. On the other hand, in GNIT-10, void nucleation by partial debonding of glass-particles and void growth occurs under relatively high stress, and the crack propagates by coalescence between the crack-tip and voids. However, as the fatigue crack growth occurs in the final stage of fatigue, the macroscopic degradation behavior mentioned in the previous section is not affected by the difference in the micro-mechanism of fatigue crack growth between both composites.

![Micrographs of fatigue fracture surface.](image)

4. Conclusions

In order to characterize the fatigue strength and damage behavior in glass-particle-reinforced Nylon 66 composites, fatigue tests and multi-step relaxation tests during fatigue were carried out on seven kinds of composites with different particle volume fraction and different interface treatment. The obtained conclusions are summarized as follows.

(1) With an increase in the particle volume fraction, the fatigue strength decreases in the interface-untreated composite, and increases up to 10% of particle volume fraction and then slightly decreases in the interface-treated composite.

(2) On the fatigue tests for the fatigue life of $10^3$ cycles, the stiffness reduces significantly at the early stage of stress cycles and then becomes constant in the Nylon 66 and both composites. The stiffness reduction is more remarkable in both composites than in the Nylon 66 because of the debonding damage between particles and matrix.

(3) From the variation of the equilibrium stress-strain relations with stress cycles, it is found that the viscous component of deformation gradually disappears and the hardening occurs with increasing stress cycles.
References


