

The Effects of Physical Aging of PSF/AS4 Laminate on Fatigue

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PSF/AS4 복합재료의 가속노화가 피로강도에 미치는 영향

김형원

Abstract

The effects of aging of PSF/AS4 laminates on fatigue was studied using the new energy release rate analysis. The analysis by the variational mechanics has been useful in providing fracture mechanics interpretation of matrix microcracking in cross-ply laminates. This paper describes the changes of the critical energy release rate (microcracking toughness) according to the aging period under fatigue loading. The master plot by modified Paris-law gives a characterization of a material system's resistance to microcrack formation. PSF [0/90_s]_s laminates were aged at four different temperature based on the glass transition temperature for 0 to 60 days. At all temperatures, the toughness decreased with aging time. The decrease of the toughness at higher temperature was faster than at lower temperature. To assess the effects of aging on fatigue, the unaged laminates were compared with the laminates which had been aged for 60 days at 170°C near 180°C T_g . The slope of dD/dN versus ΔG_m of the aged laminates was lower than that of the unaged laminates. There was a significant shift of the aged data to formation of microcracks at the lower values of ΔG_m .

Key Words: Aging effect, Fracture toughness, Neat resin, Static tensile, Microcracking, Fatigue.

1. INTRODUCTION

When composites are used at the elevated temperatures for long times, the issue of aging becomes important. Aging is defined as time-dependant changes in the composite.

The changes can be due to physical aging or chemical aging. This thesis focuses mostly on physical aging. Physical aging is associated with rearrangement of the polymeric matrix that normally leads to densification. In evaluating composite materials, the importance of aging is the effect on the laminate properties. Some properties that might change are toughness, stiffness, weight, and glass transition temperature. In this thesis, the effect of aging on microcracking toughness was the

main concern. Psf/As4 laminates were chosen as a mode material, because some Psf/As4 prepregs were available in the lab, and the glass transition temperature is relatively low. A low T_g simplifies the aging tests. The most direct way to study aging is to study the neat resin [1]. Three point bending tests were performed for single-edge notched neat resin samples of Psf by a fracture mechanics analysis. The toughness as a function of aging temperature for different periods was obtained. To get the effect on composite toughness, microcracking experiments on [0/90₂]_s laminates of Psf/As4 were performed. The aging process was periodically interrupted and the microcracking data of the laminates were collected. Having microcracking toughness data of Psf/As4 laminates as a function of time and temperature, a master curve was drawn by using time-temperature superposition. An alternative method for assessing the effect of aging on microcracking properties is to observe

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the development of microcracks during fatigue loading. The microcracking fatigue properties were measured for unaged laminates and for laminates that had been aged for 60 days at 170°C. The fatigue results will be presented.

2. EXPERIMENTAL METHODS

2.1 Neat resin Psf 3-point bending tests

2.2.1 Materials and Methods

The neat resin samples that were 3.5 mm thick, 12.6 mm wide and 50.0 mm long was cut to be the single-edge crack with a band-saw first, and sharpened with a fresh razor blade. All cracks were longer than 0.5W. Each sample was then loaded on a 3-point bending fixture in an MTS frame using a cross head rate of 0.01 mm/sec until fracture. By three point bending test, a critical intensity factor K_{IC} was determined by means of Eq. (1).

(1)

where,

$$y = 1.93 - 3.07r + 14.53r^2 - 25.11r^3 + 25.8r^4$$

S ; the length of the sample,

$$S > 4W$$

P ; applied maximum load to fracture,

$$r = \frac{a}{W}$$

Under the plane-strain conditions, can be converted to the G_{IC} ($E=2,480$ Mpa, $\nu=0.3$).

2.2 Results and discussion

The most direct way to study aging of Psf/As4 laminates is to study the neat resin. Polysulfone, an amorphous thermoplastic matrix, is tougher than most thermoset matrix resins. During the aging process, the aging was interrupted and neat resin specimens were tested for fracture toughness. The toughness as a function of aging period at either 175C or 180C is plotted in Fig. 1 (the aging times vary from 0 day to 60 days). The results present the average fracture toughness for two replicate experiments (the error of two samples is about 4.5%). The toughnesses are higher than expected but show a significant toughness loss as aging time increases. The reason that

the toughnesses are higher compared to the results of ref. [1] is probably the sharpness of the original notch formed in the specimen. For brittle materials, it is often possible to propagate a stable crack at the tip of the notch by razor blade method, but for the tougher materials such cracks do not form easily. The toughness decrease was faster and further for aging at 180°C than for aging at 175°C.

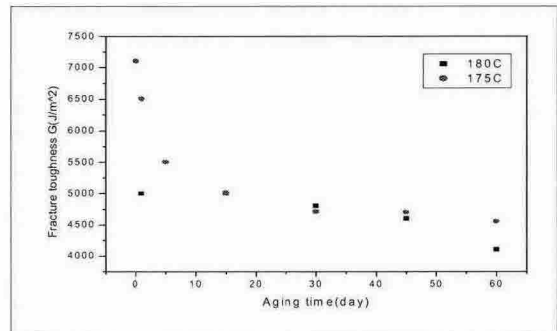


Fig. 1. The toughness as a function of aging time for neat resin Psf specimens. The circle data are from 175°C and the square data are from 180°C. The error of the two samples is about 4.5%

2.2 The Psf/As4 laminates

2.2.1 The unaged Psf/As4 laminates

Neat resin results are significant, but the most important result is the effect on composite toughness. The most likely effect of aging is on the matrix and thus the characterization of composite toughness should concentrate on matrix-dominated failure modes such as microcracking. The first microcracking experiments were to measure the microcrack density in the 90° plies which is the number of microcrack divided by the length of the specimen as a function of applied load for the unaged material. The stacking sequence of the laminates is $[0/90_2]_s$. Once the microcrack density is obtained, a microcracking toughness could be determined by the variational analysis. The variational mechanics analysis determines all components of the stress tensor in the x-z plane. In this paper, we only require the tensile stress in the 90° plies. The result is [2]

$$\sigma_0 = \frac{1}{k_m^{(1)}} \left(\sqrt{\frac{G_{mc}}{C_{3t_1} Y(D)}} - k_{th}^{(1)} T \right) \quad (3)$$

Where, the terms $k_m^{(1)}$ and $k_{th}^{(1)}$ are the effective mechanical and thermal stiffnesses of the 90° plies.

$$k_m^{(1)} = \frac{E_x^{(1)}}{E_c^0} \quad \text{and} \quad k_{th}^{(1)} = -\frac{\Delta \alpha}{C_1} \quad (4)$$

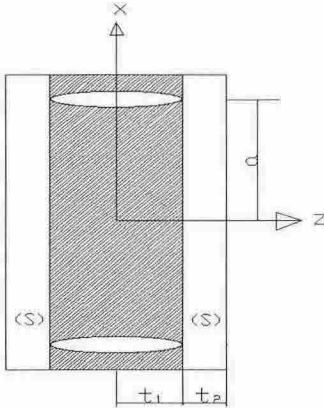


Fig. 2. Edge view of microcracks in the 90° plies of [(S)/90n]s laminates. ((s) is sublaminates)

In eq. (3), there are two unknowns G_{mc} (microcracking fracture toughness), T (the temperature difference that determines the level of residual stresses). T can be measured by various means. Therefore if we have an unknown G_{mc} , we can predict the experimental results (stress (σ_0) vs microcrack density (D)). The unknown G_{mc} can be obtained from the comparison the experimental data with the theoretical line drawn from eq. (3) using a single value of G_{mc} from the mechanical properties in Table 1.

Table 1 The mechanical properties for Psf/As4 unidirectional laminate

E_A (GPa)	E_T (GPa)	G_A (GPa)	G_T (GPa)	ν_A	ν_T	α_A (ppm /°C)	α_T (ppm /°C)	T_{eff}
134	9.8	5.5	3.6	0.3	0.5	-0.09	28.8	-225

The sample dimensions were nominally 12mm wide and 150 mm long and the thicknesses were determined by the stacking sequence.

The result of the microcracking toughness of the unaged Psf/As4 laminates was 450 J/m² and the thermal load term ΔT was -150°C.

2.2.2 The aged Psf/As4 laminates

Psf/As4 [0/90₂]_s laminates were aged at four different temperatures for 0 to 60 days (0 to 1440 hours). Even before any load was applied to the laminate, some microcracks were found in the laminate. These microcracks were assumed to be caused by residual thermal stresses. The observation of microcracks before loading is an indication that G_{mc} is low. The microcracking toughness (G_{ms}) as a function of aging time is plotted in Fig. 2. At all temperatures, the toughness dropped with time (refer to Table 2). The decrease in toughness was faster at higher temperatures than at lower temperatures. At all temperature, there was already a significant drop after only one day of aging. The G_{mc} values were indeed low ranging from a high of 280 J/m² to a low of 90 J/m². The laminate toughness decreased with aging and, on a percentage basis, decreased more than the neat resin fracture toughness. In other words, the ratio of the 60 day aged toughness to the unaged toughness was lower in the composite than in the neat resin. There was also more than an order of magnitude difference between the neat resin toughness and the laminate toughness. The usual explanation for the low laminate toughness is that in the composite the plastic zone size is restricted by the fibers. As a result, the energy dissipation that occurs in the neat resin does not occur in the composite and the toughness is lower.

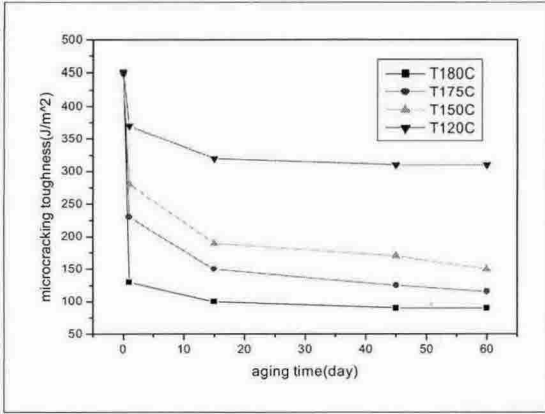


Fig. 3. The microcracking toughness as a function of aging time for $[0/90_2]_s$ Psf/As4 laminates.

Table 2 The microcracking toughness of $[0/90_2]_s$ Psf/As4 laminates as a function of aging time

aging days	$G_{mc}(J/m^2)$ at $T=180^\circ C$	$G_{mc}(J/m^2)$ at $T=175^\circ C$	$G_{mc}(J/m^2)$ at $T=150^\circ C$	$G_{mc}(J/m^2)$ at $T=120^\circ C$
0	450	450	450	450
1	130	230	280	370
15	100	150	190	320
45	90	125	170	310
60	90	115	150	310

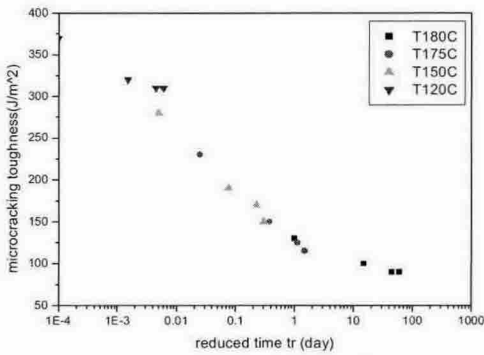


Fig. 4. Microcracking toughness of Psf/As4 laminates as a function of reduced aging time

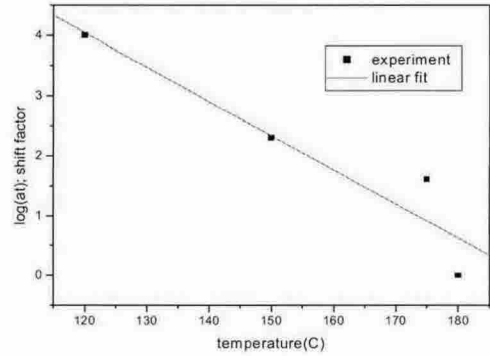


Fig. 5. The shift factor of Psf/As4 laminates as a function of aging temperature

Having mechanical property data of Psf/As4 as a function of time and temperature, a master curve can be drawn by using time-temperature superposition. The microcracking toughness thus was plotted as a function of reduced time, t_r , defined as the following equation.

$$\log t_r = \log \frac{t}{a_T} = \log t - \log a_T \quad (5)$$

where t is actual time and $\log a_T$ is the temperature-dependant shift factor. The test temperature of $180^\circ C$ arbitrarily was selected to be the reference temperature and the data at other temperatures were shifted to achieve the maximum overlap between experimental results on a reduced time plot. The master plot is given in Fig. 4. The master plot defines a smooth curve. In principle, this plot could be used to predict the microcracking toughness after any amount of time at any aging temperature. For some conditions, there was only slight overlap between experiments at two adjacent temperatures. It would be more confident in the master curve if the data at various temperatures had been extended to longer times resulting in more overlap on the reduced time plot.

The shift factor as a function of temperature required to produce the master plot is given in Fig. 5. The qualitative result from Fig. 4 and Fig. 5 is that degradation in microcracking toughness due to physical aging can be observed in reasonably short time scales even well below the glass transition temperature.

2.2.2 Effect of aging on fatigue of Psf/As4 laminates

An alternative method for accessing the effect of aging on microcracking properties is to study the development of microcracks during the fatigue loading. A fracture mechanics interpretation of the propagation of microcracks during fatigue testing is based on a modified Paris-law approach. To apply Eq. (3) to the analysis of fatigue experiments, a modified Paris-law approach was used in which the rate of increase in microcrack density per cycle, dD/dN , during fatigue loading can be predicted using a modified Paris law as the following equation.

$$\frac{dD}{dN} = A \Delta G_m^n \quad (6)$$

where ΔG_m is the range in G_m for the maximum and minimum loads in the fatigue cycle and A and n are material properties that describe the resistance of the material to fatigue-induced microcracks. Eq. (6) used the range in energy release rate, ΔG_m , instead of the range in stress-intensity factor, ΔK . This substitution is made because we have an expression for G_m and the stress intensity factor has no physical meaning for formation of a complete microcrack.

The fatigue tests were run on an MTS 25KN servohydraulic testing frame using load control. The load span was set to cycle between a maximum stress, σ_{max} , and the stress span, $\Delta\sigma_o$. The cycling rate was usually 5 Hz. During the fatigue test, the load cycling was periodically stopped and the microcrack density was measured by examining the sample edges under an optical microscope and counting the cracks. The fatigue experiment is to vary σ_{max} and σ_{min} . For each $\Delta\sigma$, ΔG_m was calculated for low microcrack density and the microcrack density was measured as a function of cycle number. A linear fit to the microcrack density data gives the rate of increase in microcrack density as a function of ΔG_m . Finally, a log-log plot of dD/dN as a function of ΔG_m should be linear with intercept $\log a$ and slope n . To assess the effect of aging on the

microcracking properties of Psf/As4 laminates, the microcracking fatigue properties were measured for unaged laminates and for laminates that had been aged for 60 days at 170°C. From the aging data Fig. 3, the static toughness of the aged laminates for 60 days at 170°C should be about 125 J/m². The fatigue data show that aging had a significant effect on the fatigue properties of the laminates. The slopes of aged ($n=0.816$) and unaged ($n=2.04$) laminates are different, and there is a significant shift of the aged data to formation of microcracks at lower values of ΔG_m . Therefore, as an alternative way to see aging the fatigue test has a good results.

3. SUMMARY

As a simple model system for aging of thermoplastic matrix composites, [0/90₂]_s Psf/As4 laminates were studied. The concern to be researched was the effect of aging on microcracking toughness. A series of aging experiments at four different temperatures from 0 to 60 days have been expressed as a master plot at a single reference temperature by using time-temperature superposition.

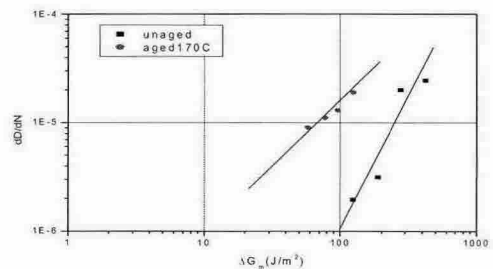


Fig. 6. The microcracking fatigue properties of Psf/As4 [0/90₂]_s laminates

The laminate toughness decreased with aging and, on a percentage basis, decreased more than the neat resin toughness. In other words, the ratio of the 60 day aged toughness to the unaged toughness was lower in the composite than in the neat resin. The energy release rate analysis has been successful in predicting the fatigue experiments using a modified Paris-law approach. The rate of increase in microcrack density per cycle during fatigue loading, dD/dN , could be predicted. The

microcracking fatigue properties were measured for unaged laminates and for laminates that had been aged for 60 days at 170°C. The aging had a significant effect on the fatigue properties of the laminates. There was a significant shift of the aged data to formation of microcracks under fatigue loading.

REFERENCES

1. Highsmith, A. L. and Reifsnider, K. L. (1982), "Stiffness reduction mechanism in composite laminates," ASTM STP 775, 103
2. J. A. Naim (1989), "The strain energy release rate of composite microcracking: A variational approach," *J. Comp. Mat.* 23, 1106
3. S. Liu and J. A. Naim (1990), "Fracture mechanics analysis of composite microcracking: Experimental results in fatigue," *Proc. of the Amer. Soc. of Comp., 5th Tech. Conf.*, 287.
4. Struick L. C. E. (1978), "Physical aging in amorphous polymers and other materials," Elsevier, Amsterdam
5. G. Williams (1984), "Fracture Mechanics of Polymer, Ellis Horwood, Chichester.