Advanced Composite Materials
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/tacm20

Influence of thermal history on transverse cracking in a carbon fiber reinforced epoxy composite

Keiji Ogi

Version of record first published: 02 Apr 2012.

To cite this article: Keiji Ogi (2002): Influence of thermal history on transverse cracking in a carbon fiber reinforced epoxy composite , Advanced Composite Materials, 11:3, 265-275

To link to this article: http://dx.doi.org/10.1163/156855102762506308

Adv. Composite Mater., Vol. 11, No. 3, pp. 265–275 (2003)

Also available online - www.vsppub.com

Influence of thermal history on transverse cracking in a carbon fiber reinforced epoxy composite

KEIJI OGI*

Department of Materials Science and Engineering, Faculty of Engineering, Ehime University, 3 Bunkyocho, Matsuyama, Ehime 790-8577, Japan

Received 2 March 2002; accepted 21 October 2002

Abstract—This paper presents the influence of thermal history on transverse cracking in carbon fiber reinforced epoxy (carbon/epoxy) composites. At first, measurement of micro-Vickers hardness and Fourier transform-infrared spectrum (FT-IR) analysis of resin are conducted to investigate the effect of thermal history on resin. Secondly, shrinkage strain and transverse strength are measured for carbon/epoxy unidirectional laminates. Thirdly, the effect of temperature and duration of thermal history on transverse cracking in cross-ply laminates is experimentally investigated. The change in the residual stress due to post-cure is also estimated for the cross-ply laminates. Transverse cracking in the cross-ply specimens after thermal history initiates at lower stresses than the critical stresses in virgin specimens. It is found that thermal history increases the hardness of resin; however, it does not change the chemical structures in resin. The decrease in the strength is mainly caused by degradation of the fiber/matrix interface.

Keywords: Carbon/epoxy; thermal history; post-cure; mechanical properties; transverse cracking.

1. INTRODUCTION

As composite materials have been widely used in various industrial fields, it is important to accumulate data on strength and lifetime under practical thermal conditions to ensure the reliability of those materials. It is essential to quantitatively evaluate the change in the mechanical properties and geometry when the material is subjected to thermal history during the fabrication process.

While the damage evolution [1, 2] and creep behavior [3, 4] of carbon/epoxy laminates under various temperatures have been investigated, effect of thermal history before mechanical loading on the deformation and damage has not been clarified. In general, appropriate post-cure processes increase the modulus and

*E-mail: kogi@eng.ehime-u.ac.jp

strength of fiber reinforced thermosetting composites due to relaxation of thermal stress and shrinkage of resin [5, 6]. However, strength can be decreased by degradation at the fiber/matrix interface after thermal history with severe thermal conditions [7]. It is confirmed that the unidirectional carbon/epoxy composites shrink in the transverse direction during a curing process [8] and post-cure process. The author has investigated the effect of post-cure for 4 h on mechanical properties and transverse cracking and has found that the shrinkage due to post-cure increases the tensile residual stress in the transverse ply of cross-ply laminates and decreases the critical stress for transverse cracking [9].

In the present study, we conducted thermal treatment with duration of 4 or 100 h at 150°C or 200°C using the same carbon/epoxy composite as that in the previous work [9]. First, measurement of micro-Vickers hardness and FT-IR analysis of resin in the composite were performed before and after thermal history. Secondly, shrinkage strain and transverse strength of unidirectional laminates were measured after the thermal history. Thirdly, transverse cracking behavior in the cross-ply laminates was studied taking transverse strength and residual stress into account. Finally, fracture surfaces were observed by scanning electron microscopy (SEM) to reveal the fracture behavior.

2. EXPERIMENTAL

2.1. Material and thermal history

The carbon/epoxy unidirectional ([90°]) and cross-ply ($[0^\circ/90^\circ_3]_S$) laminates were fabricated by means of an autoclave system at a curing temperature of 180°C. The cured thickness was about 1.1 mm and fiber volume fraction was 0.63 for both laminates. In some unidirectional laminates, thermocouples and heat-resistant strain gages with the gage length of 10 mm were embedded between prepregs and co-cured.

At first, the specimens were cut out of unidirectional and cross-ply laminate plates and heated for 4 or 100 h at temperatures 150 or 200°C in air under atmospheric pressure. Hereafter, we denote five thermal history conditions by Virgin, 150°C/4 h, 200°C/4 h, 150°C/100 h and 200°C/100 h. Secondly, the longitudinal and transverse strains in the unidirectional laminates were measured with the strain gages embedded in both directions during thermal history. Table 1 shows material properties of a virgin unidirectional laminate [9].

Table 1.
Material properties of a carbon/epoxy composite

E_1 GPa	E_2 GPa	G_{23} GPa	α_1 K^{-1}	α_2 K^{-1}
154.6	8.28	2.85	-0.5×10^{-6}	35.5×10^{-6}

2.2. Vickers hardness and FT-IR analysis

After polishing the cross-section of virgin and thermally-treated [90°] specimens, micro-Vickers hardness H_V of resin between fibers were measured for applied load of 1 gf. Resin rich regions at the interlayers were selected for measurement of hardness.

In addition, FT-IR spectrum of resin near the specimen surface was analyzed to reveal the effect of thermal history on chemical structures in resin for five thermal conditions.

2.3. Tensile tests

The dumbbell-shaped [90°] specimens for tensile tests were fabricated to measure transverse strength S_2 [9]. The strain gages and glass/epoxy tabs were glued at the center and on both ends of the specimens, respectively. Tensile tests were performed at a loading speed of 4.9 N/s at room temperature to obtain the transverse stress – strain curves.

Coupon-shaped cross-ply specimens with end tabs were also used for tensile tests [9]. The two strain gages with the gage length of 30 mm were glued on the specimen surface. Thus, the total gage length was 60 mm. Tensile tests of cross-ply specimens were conducted in the same way as [90°] specimens. The number of transverse cracks taking place within the gage length of 60 mm was estimated from change in the longitudinal strain measured with the two strain gages.

The surfaces of transverse cracks in the cross-ply specimens with and without thermal history were observed by means of SEM focusing on fiber/matrix interfaces.

3. RESULTS

3.1. Hardness and FT-IR analysis of resin

Micro-Vickers hardness of resin for each thermal history is shown in Fig. 1. The hardness tends to increase with increasing thermal treatment temperature for both 4 h and 100 h, although some scatter due to measurement error is observed. Table 2

Table 2.
Summary of experimental results for various conditions of thermal history. The symbols H_V, $-e_2$, S_2, $\Delta\sigma_2$ and σ_f denote the hardness of resin, transverse shrinkage strain in the [90°] specimens, transverse strength of the [90°] specimens, tensile residual stress increment in the transverse layer of the cross-ply specimens and failure strength of the cross-ply specimens, respectively

	H_V	$-e_2$ ($\mu\varepsilon$)	S_2 (MPa)	$\Delta\sigma_2$ (MPa)	σ_f (MPa)
Virgin	22.2	0	74.1	0	596
150°C/4 h	25.2	700	76.5	5	573
200°C/4 h	23.9	2500	63.4	18	562
150°C/100 h	24.0	700	59.2	5	504
200°C/100 h	25.1	3000	46.6	21	251

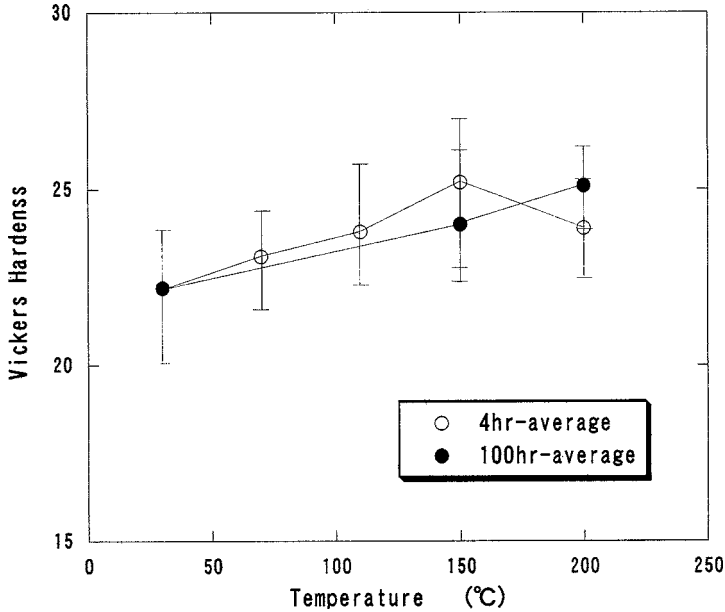


Figure 1. Micro-Vickers hardness of resin for various conditions of thermal history.

Table 3.
Wave numbers and corresponding chemical structures at the peaks in Fig. 2. The symbol B denotes a benzene ring

Wave numbers (cm^{-1})	~3400	~2900	1591 1298 715	1506	1236 817	1182	1140	1101
Chemical structures	−OH or −NH_2	−CH_2−	B−NH_2	B	−COC−	−SO_3H	B−SO_2−B	$-SO_4^{2-}$

B: Benzene ring.

shows the hardness of resin H_V, transverse shrinkage strain in the [90°] specimens $-e_2$, transverse strength of the [90°] specimens S_2, tensile residual stress increment in the transverse layer of the $[0^\circ/90_3^\circ]_S$ specimens $\Delta\sigma_2$ and failure strength of the $[0^\circ/90_3^\circ]_S$ specimens σ_f.

Figure 2 shows the FT-IR spectra of resin for five thermal history conditions. Table 3 summarizes the wave numbers and chemical structures that correspond to the peaks in Fig. 2. Aromatic sulfon, aromatic amine and sulfoxide included in the epoxy resin and hardener are detected as peaks. Difference among five conditions is not observed.

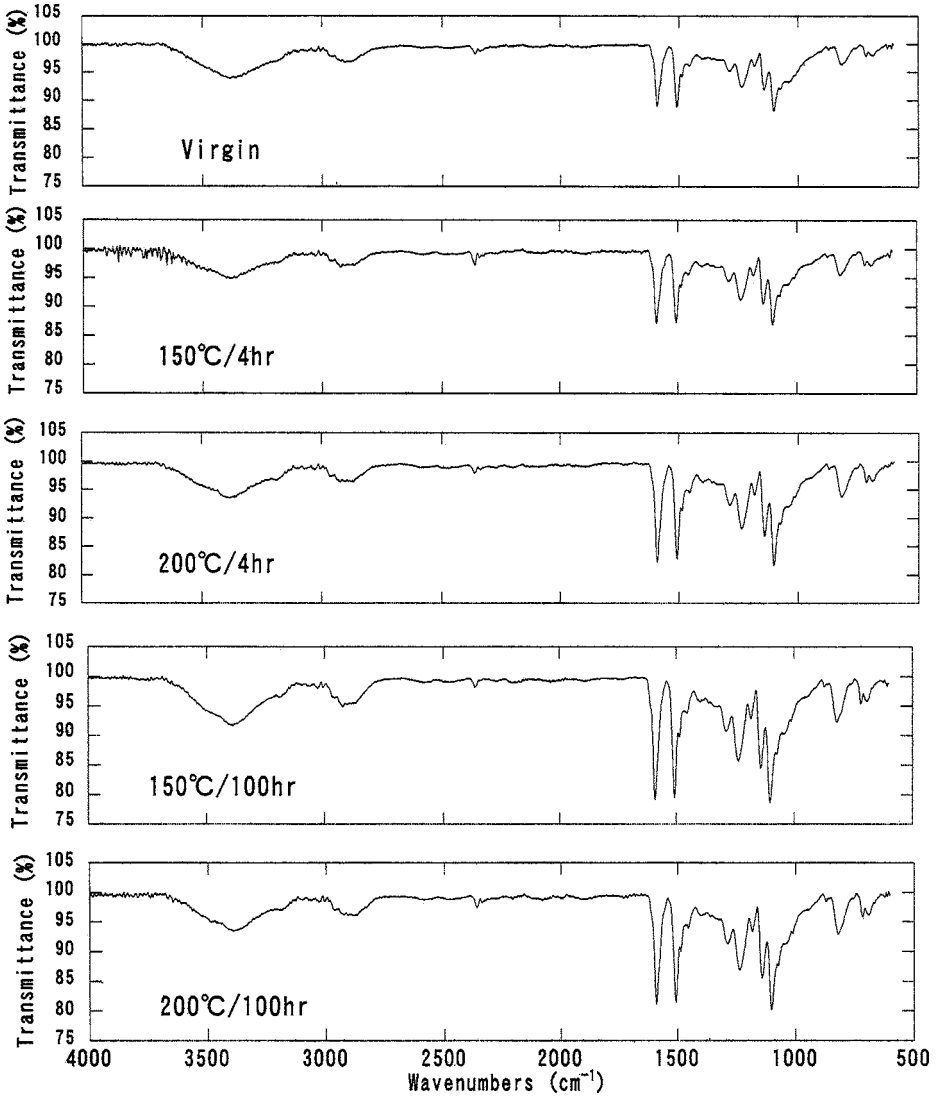


Figure 2. FT-IR spectra of resin before and after thermal history.

3.2. Shrinkage strain

Figure 3 shows the permanent transverse shrinkage strain $-e_2$ in the [90°] specimens after thermal history. The shrinkage strain is not proportional to temperature but remarkably increases above 180°C. This shrinkage strain is of the same magnitude as the decrease in the strain during keeping temperature constant (150 or 200°C). On the other hand, the longitudinal strain decreases with temperature rise and returns to approximately zero at room temperature [9].

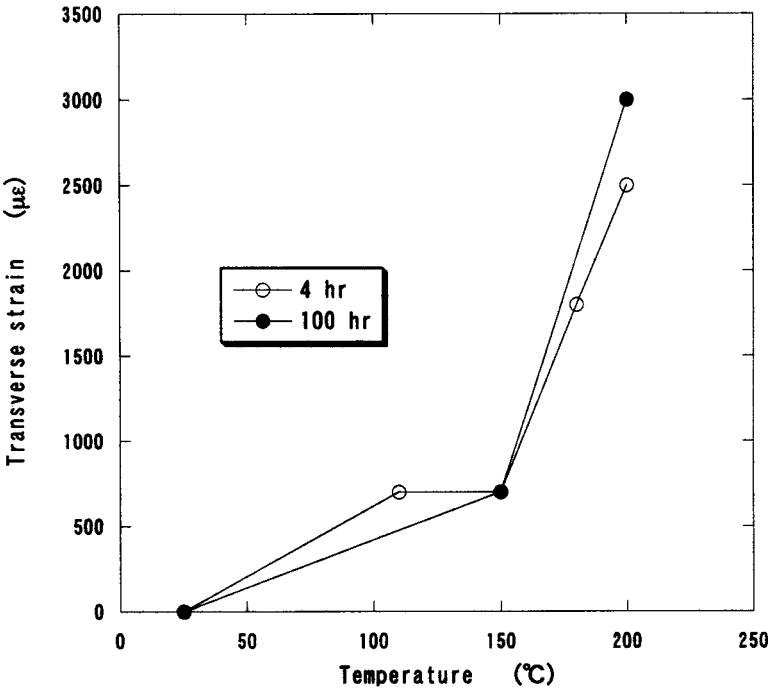


Figure 3. Permanent shrinkage strain in the transverse direction of unidirectional laminates.

3.3. Transverse strength

The average transverse strength measured using [90°] specimens for five conditions is plotted in Fig. 4 and summarized in Table 2. Transverse strength slightly increases for 150°C/4 h while it decreases for 200°C/4 h. On the other hand, transverse strength for 150°C/100 h and 200°C/100 h is lower than that for Virgin, 150°C/4 h and 200°C/4 h.

Figure 5 shows transverse crack density against applied stress in the cross-ply specimens for each thermal history. The critical stress for transverse cracking decreases with increasing thermal treatment temperature and the crack density is larger at higher temperature. In the case of 200°C/100 h, the specimens fracture with little transverse cracking. Final failure stress σ_f of the cross-ply specimens is presented in Table 2. The failure strength decreases in order of Virgin, 150 °C/4 h, 200°C/4 h, 150°C/100 h and 200°C/100 h. Especially, the failure strength for 200°C/100 h is approximately reduced to half of that for Virgin.

3.4. Fracture behavior

Since effect of temperature has already been discussed in the previous work [9], effect of duration of thermal history is compared in the present paper. Figure 6 shows SEM photographs of transverse cracks in the cross-ply specimens for Virgin, 200°C/4 h and 200°C/100 h. In the thermally-treated specimen, amount of resin

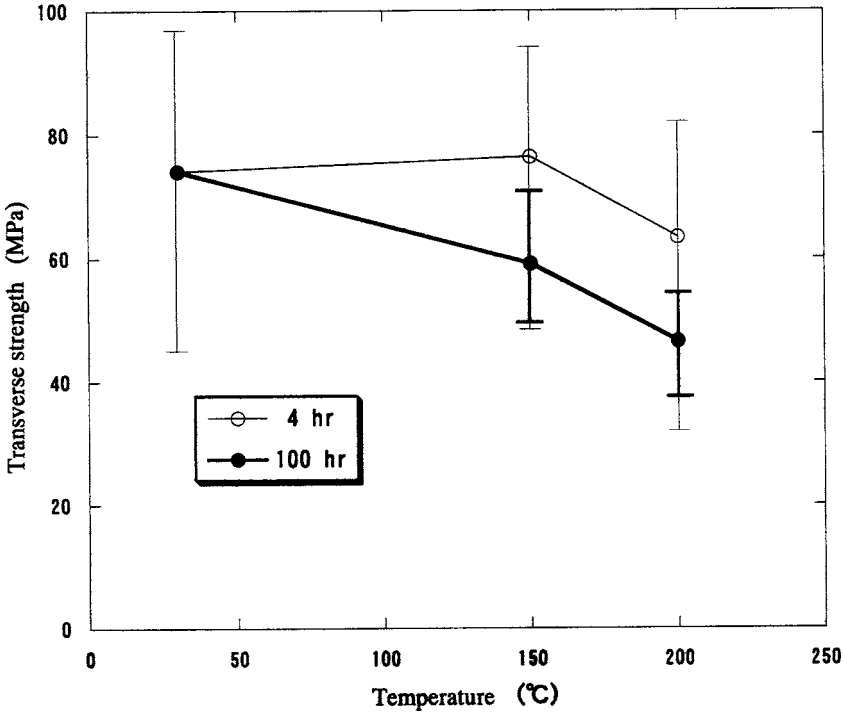


Figure 4. Effect of temperature and duration of thermal history on transverse strength of unidirectional laminates.

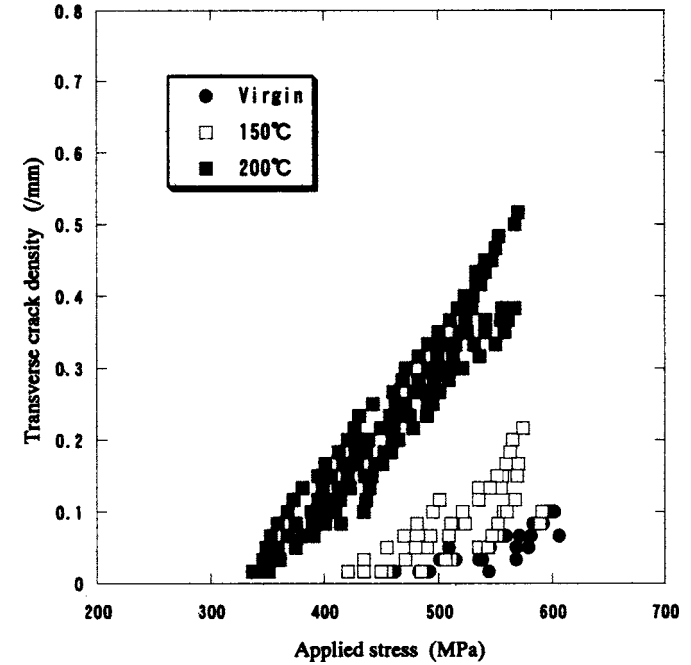
attached to fiber/resin interfaces is of small magnitude compared with that in Virgin. Fracture morphology for 150°C/4 h and 150°C/100 h is similar to that for 200°C/4 h and 200°C/100 h.

4. DISCUSSION

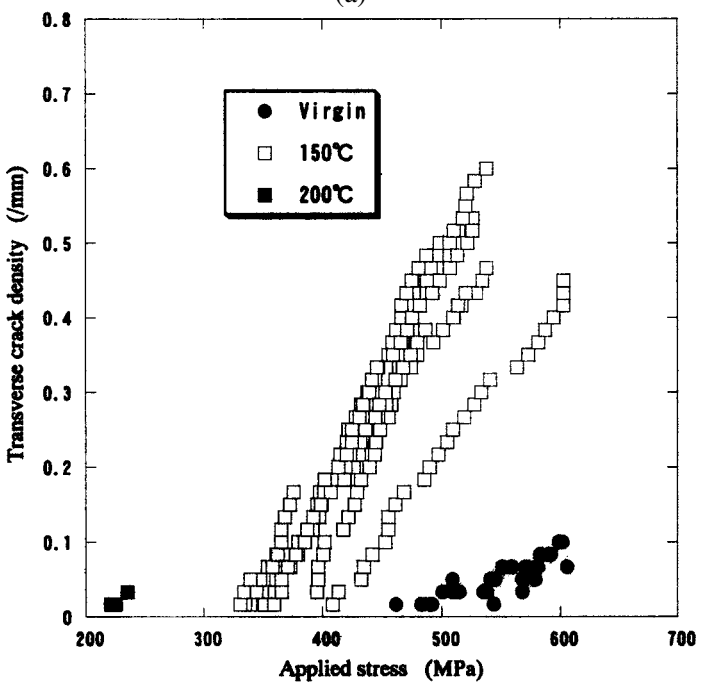
4.1. Effect of thermal history on resin

As shown in Fig. 1, hardness of resin is increased by thermal history. In general, hardness of resin is ascribed to the yield strength of resin. On the other hand, change in chemical structures of resin observed in acid environment [10] is not caused by the present thermal history (Fig. 2). It should be noted that the present FT-IR was conducted only for resin and that the results did not reflect the change of bonding at the fiber/resin interface. Accordingly, it is presumed from the above results that resin is hardened and not degraded by thermal history.

Shrinkage in the transverse direction (Fig. 3) is caused by post-cure of resin, namely, reduction in volume due to cross-linking of uncured resin [6]. Remarkable increase in the shrinkage strain above 150°C may be ascribed to the fact that the glass transition temperature T_g is about 165°C. At temperatures higher than T_g, the above shrinkage easily occurs because motion of polymer chains becomes free.

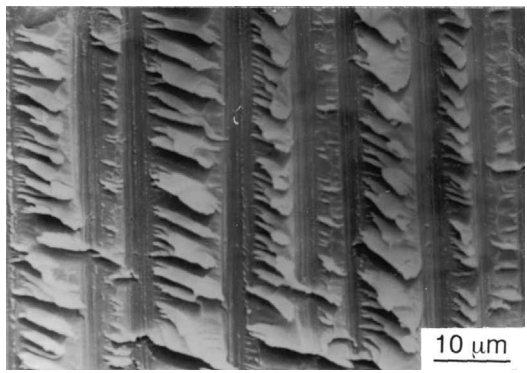


(a)

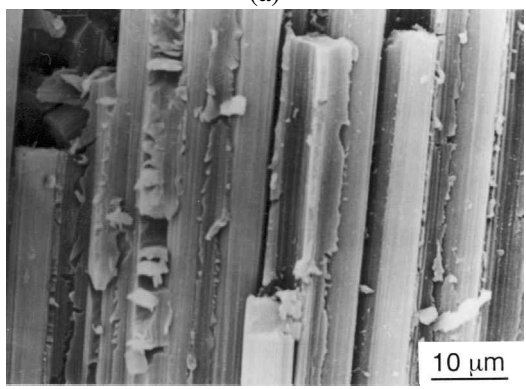


(b)

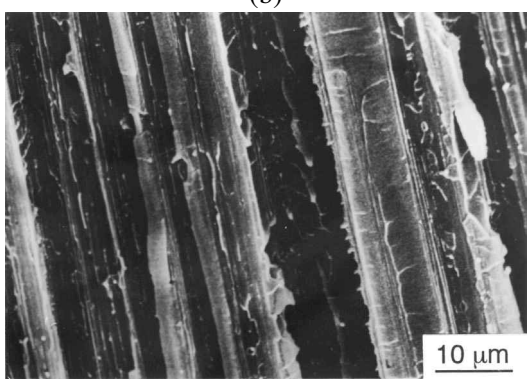
Figure 5. Effect of temperature of thermal history with duration of (a) 4 h and (b) 100 h on transverse cracking in cross-ply laminates.



(a)



(b)



(c)

Figure 6. SEM photos showing surfaces of transverse cracks in cross-ply specimens for (a) Virgin, (b) 200 °C/4 h and (c) 200°C/100 h.

4.2. Transverse strength and transverse cracking

Since deterioration of resin was not observed after thermal history as is stated previously, the main reason for decrease in transverse strength (Fig. 4) is concluded to be degradation of fiber/matrix interface. Degradation of interface is also

supported by the fracture morphology (Fig. 6). In addition to this degradation, increase in residual stress in the cross-ply specimens results in decrease in the onset stress of transverse cracking (Fig. 5). Residual stress increment is estimated using expressions similar to thermal residual stress based on the lamination theory [9]. The shrinkage strain $-e_2$ in the [90°] specimens and the corresponding tensile residual stress increment $\Delta\sigma_2$ in the transverse ply of the cross-ply specimens are presented in Table 2. The large residual stress increment for 200°C/4 h and 200°C/100 h leads to reduction of transverse strength and the critical strength for transverse cracking.

Final failure strength as well as the onset stress of transverse cracking decreases with increasing temperature and duration of thermal history (Table 2). In particular, the cross-ply specimens for 200°C/100 h fracture at low stress after very few transverse cracks are generated. One of the presumable reasons for this is the delamination at relatively low stress which propagates through the width and then along the length to final failure of the cross-ply specimens. For further discussion on failure strength of the cross-ply laminate, longitudinal strength of the unidirectional laminate and interlaminar fracture toughness between the longitudinal and transverse layers should be measured for each condition of thermal history

5. CONCLUDING REMARK

Effect of thermal history on transverse cracking in a carbon/epoxy composite was studied. As temperature and duration of thermal history increase, transverse strength in the unidirectional laminates and the onset stress of transverse cracking in the cross-ply laminates decrease. Degradation at the fiber/matrix interface and increase in residual stress due to shrinkage are reasons for decrease in strength after thermal history while there was no evidence to show degradation of resin.

REFERENCES

1. H. S. Kim, K. Ogi, T. Matsubara, W. X. Wang and Y. Takao, Microscopic damages of quasi-isotropic carbon/epoxy laminates under various temperatures, *J. Soc. Mater. Sci. Japan* **47**, 599–605 (1998) (in Japanese).
2. K. Ogi, H. S. Kim, T. Maruyama and Y. Takao, Influence of hygrothermal conditions on the damage processes in quasi-isotropic carbon/epoxy laminates, *Compos. Sci. Technol.* **59**, 2375–2382 (2000).
3. K. Ogi and Y. Takao, Evolution of transverse cracking in CF/epoxy cross-ply laminates under creep loading, *J. Reinf. Plast. Compos.* **13**, 220–230 (1999).
4. K. Ogi and Y. Takao, Time-dependent behavior of deformation and transverse cracking in cross-ply laminates, *Adv. Composite Mater.* **10**, 39–62 (2001).
5. K. A. Lindsey and C. D. Rudd, Effects of post-cure on the interfacial properties of glass fiber-urethane methacrylate composites, *J. Mater. Sci. Lett.* **12**, 894–897 (1993).

6. R. Tucker, P. Compston and P.-Y. B. Jar, The effect of post-cure duration on the mode I interlaminar fracture toughness of glass-fibre reinforced vinylester, *Composites Part A* **32**, 129–134 (2001).
7. J.-R. Hwang and A.-S. Lee, Thermal damage effects on mode II interlaminar fatigue crack propagation of graphite/epoxy composites, *Compos. Struct.* **27**, 389–393 (1994).
8. A. S. Crasto and R. Y. Kim, On the estimation of shrinkage and thermal strains during composite cure, in: *Proc. 8th Japan–US Conf. Compos. Mater.*, pp. 813–822 (1998).
9. K. Ogi, Effect of post-cure on mechanical properties of CF/epoxy composites, *J. Soc. Mater. Sci. Japan* **51** (5), 518–523 (2002) (in Japanese).
10. Y. Tomita, K. Morioka, T. Iseki and S. Nakahori, Environmental effect on strength and fracture of advanced CFRP composite, *J. Soc. Mater. Sci. Japan* **50**, 1196–1200 (2001) (in Japanese).