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EFFECT OF GEOMETRY ON THERMAL AGING BEHAVIOR OF CELION/LARC-160 COMPOSITES

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EFFECT OF GEOMETRY ON THERMAL AGING BEHAVIOR OF CELION/LARC-160 COMPOSITES

BY

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SUMMARY

Laminates of Celion®/LARC-160, fabricated in thicknesses from 4 to 16 ply and in unidirectional, x-ply and fabric ply configurations, were isothermally aged at temperatures of 204, 260 and 316°C for periods up to 15,000 hours. Weight-loss of the test panels was measured at selected intervals during aging. At the lower aging temperatures, it was observed that panel thickness and ply arrangement influenced the apparent stability: i.e., thicker panels degraded less than thin panels and unidirectional panels degraded less than x-ply or fabric reinforced panels. At higher aging temperatures, all panel configurations and thicknesses converged toward the same behavior.

INTRODUCTION

The National Aeronautics and Space Administration is currently conducting research directed at exploiting the full weight-saving potential

*Celion is a registered trademark of BASF Structural Material, Inc.

of composite materials for a wide range of aerospace structural applications. Carbon fiber reinforced epoxy composites have been studied for decades and are currently being used in applications requiring long-term thermal exposures at temperatures below 177°C. Potential applications for composite materials, however, include spacecraft requiring use at temperatures up to 316°C for lifetimes up to 100 hours and subsonic and future supersonic aircraft applications requiring use at temperatures of 170°C to 260°C for periods of 50,000 to 70,000 hours.

Currently, the only readily available composite matrix materials suitable for this temperature range is the class of addition polyimides represented by the NASA-developed PMR-15 and LARC-160 resins. A number of studies have been made of the long-term thermal stability of these and similar materials (refs. 1 to 6). One difficulty in predicting long-term stability from accelerated and/or small scale tests is the effect of sample geometry (ref. 2). In ref. 2, a distinct difference between the measured stability of small SBS test specimens and larger 6 inch square panels, as well as an effect of sample thickness was reported. It was asserted that these effects could be observed by monitoring weight loss during thermal aging.

This paper reports on a study to further define this effect of geometry on the apparent thermal stability of Celion/LARC-160 composites. Three different geometric configurations of several thicknesses of unidirectional, x-ply and fabric reinforced panels of LARC-160/Celion were thermally aged at temperatures of 204°C, 260°C and 316°C for times up to 15,000 hours. Results are presented showing the weight-loss of each panel as function aging time and temperature.

EXPERIMENTAL PROCEDURE

Materials and Fabrication

The polyimide matrix resin used in all the laminates in this study was LARC-160. LARC-160 is a solventless resin system based on the ethyl esters of 3,3',4,4'-benzophenone tetracarboxylic dianhydride (BTDA) and 5-norbornene-2,3-dicarboxylic acid (NA), and Jefferson Jeffamine AP-22 aromatic amine mixture (ref. 7). The carbon fiber used for the unidirectional and x-plied laminates, $(0.90)_s$, was Celion 6000 continuous filament yarn, sized with a DuPont NR-150B2 polyimide precursor solution. The fabric reinforcement material was Celion 3000 continuous filament yarn, sized with an epoxy compatible resin, woven into a 8 harness satin weave. Layups of these prepregs, approximately 30 cm x 30 cm, were vacuum bag autoclave processed to a maximum temperature of 330°C according to the cure temperature profile shown in figure 1. After fabrication, the laminates were subjected to ultrasonic C-scan inspection. Fiber volume, resin content, void content and thickness were determined for each acceptable laminate. These are shown in Table I. The panels had low to moderate void content and generally a fiber volume of 50 to 60 percent. The glass transition temperature (Tg) was measured as 349°C, indicating adequate cure. Each of the acceptable 30 cm square laminates were cut into the test panel configurations shown in figure 2.

Isothermal Aging

Forced-convection horizontal airflow ovens were used for isothermal aging at 204°C, 260°C and 316°C. The average air velocity was approximately

0.75 m/sec. The panels were supported vertically on their edges, 12 mm apart, with air flowing freely between each panel. The panels were subjected to exposure times up to 15,000 hours. At predetermined intervals, all panels were removed for weight loss determinations and then replaced. Each time, the oven was cooled to room temperature over a period of two hours to lessen the thermal shock when the panels were removed. When the aging of the panels was completed, they were each examined closely with a low power microscope and photographed.

Since the resin content of the panels varied, the weight-loss data were normalized to a uniform 30 percent resin content.

RESULTS AND DISCUSSION

One objective of this study was to examine the effect of the exposed panel edge on the measured weight loss. The results reported in reference 2, indicated that weight loss occurred preferentially from the surface of the panel edge perpendicular to the direction of the fiber (0° edge) in unidirectional laminates. The three panel configurations aged in this study (fig. 2) provided a variation of this parameter. Configuration I has the largest 0° edge surface area relative to both total edge surface or panel volume, and II the smallest. The effect of these three panel configurations on long-term thermal aging behavior of unidirectional panels aged at 204°C is shown in figures 3 to 5. The weight-loss behavior of the 4-ply panels in figure 3 shows a variation with panel configuration but not that which was expected. The configuration II panel (least 0° edge area) degraded the most and the panel I (most 0° edge area) the least. This appears to indicate that the 90° edge of these panels is the preferential site for weight-loss. However in figure 4, the weight-loss curves for the 8-ply panels, there is little difference between the three configurations. This is also true for the 16-ply panels shown in figure 5. In figures 6 and 7 the 4-ply unidirectional weight-loss curves for aging at 260°C and 316°C are shown. Neither of these show any significant effect of panel configuration. In fact, none of the unidirectional panels aged at 260°C and 316°C showed any effect of panel configuration. It can be seen in figures 8 and 9 that the x-ply and fabric reinforced aged at 204°C, as expected, also show no effect of panel configuration. The aging data at 260°C and 316°C were similar.

The effect of panel thickness on weight-loss behavior is illustrated in figures 10 to 12. The weight-loss curves of the unidirectional square panels aged at 204°C exhibit a systematic variation, with the 16-ply having the least weight-loss and the 4-ply having the most. This same behavior was shown by the x-ply panels but the fabric reinforced panels show little effect of panel thickness. These same general trends were repeated when the panels were aged at 260°C and 316°C (figs. 13 to 18). The thinner (4 and 8 ply) unidirectional panels, however, show an increasing tendency to degrade at the same rate. When aged at 316°C these two are essentially the same, whereas the 16-ply panel remains distinctly more stable.

The variation in weight-loss with ply configuration (unidirectional, x-ply and fabric) is typified by the 8-ply aging curves shown in figures 19 to 21. The other thicknesses show the same trends. When aged at 204°C the unidirectional panels are significantly more stable than the x-ply or fabric panels. There is little or no difference between fabric reinforced and x-plied panels. As the aging temperature is increased to 260°C and 316°C the difference between the three ply configurations disappears. At 316°C all three degrade essentially at the same rate.

CONCLUSIONS

A general conclusion of this study is that for moderate sized panels the geometric shape does not significantly influence weight-loss during thermal aging. The panel thickness did significantly effect the weight-loss of unidirectional and x-ply specimens aged at 204°C and 260°C. The thicker panels degraded the least, the thinnest degraded the most. This effect was greatest at 204°C, less at 260°C and essentially absent at 316°C. There was no significant effect of panel thickness observed in the fabric reinforced panels aged at any temperature. The ply configuration (unidirectional, x-ply or fabric) was an important factor in panels aged at 204°C. The unidirectional panels were significantly more stable than the fabric and x-ply panels. On aging at 260°C and 316°C there was no real difference in the three forms.

At low aging temperatures, variables such as panel thickness and ply arrangement effect the observed stability. At higher aging temperatures all panel configurations converge toward the same behavior.

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TABLE I--INITIAL PROPERTIES OF CELION/LARC-160 LAMINATES

PANEL TYPE	THICKNESS in.	FIBER VOL. %	RESIN WT. %	VOIDS %
4-Ply Uni	.023	53.2	36.8	4.4
8-Ply Uni	.047	67.2	25.2	2.2
16-Ply Uni	.092	54.0	38.3	0.5
4-Ply X-Ply	.022	53.8	36.9	3.1
6-Ply X-Ply	.033	48.5	42.8	1.9
8-Ply X-Ply	.043	50.5	40.4	3.2
16-Ply X-Ply	.080	52.1	40.1	0.4
4-Ply Fabric	.060	48.6	42.7	1.9
8-Ply Fabric	.126	54.6	37.3	0.9

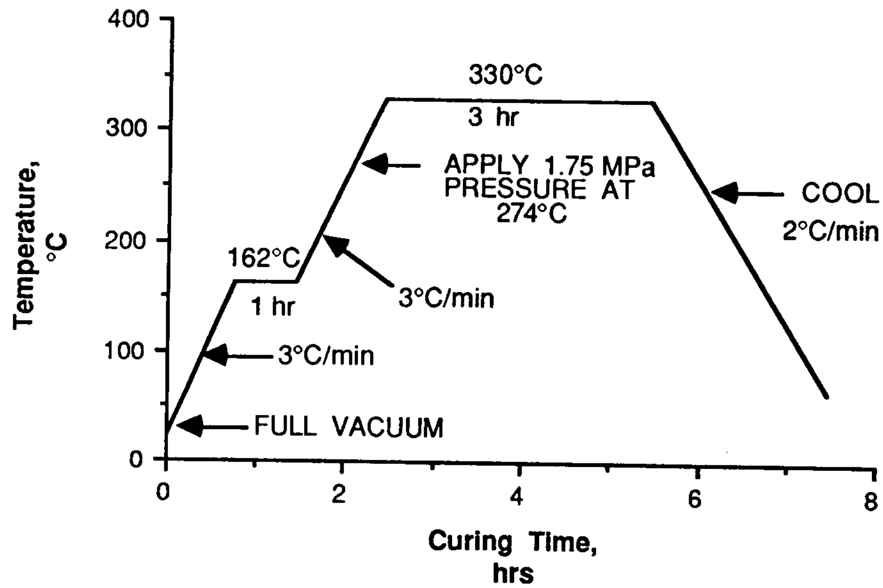

Figure 1. Temperature profile for Celion/LARC-160 autoclave curing process.

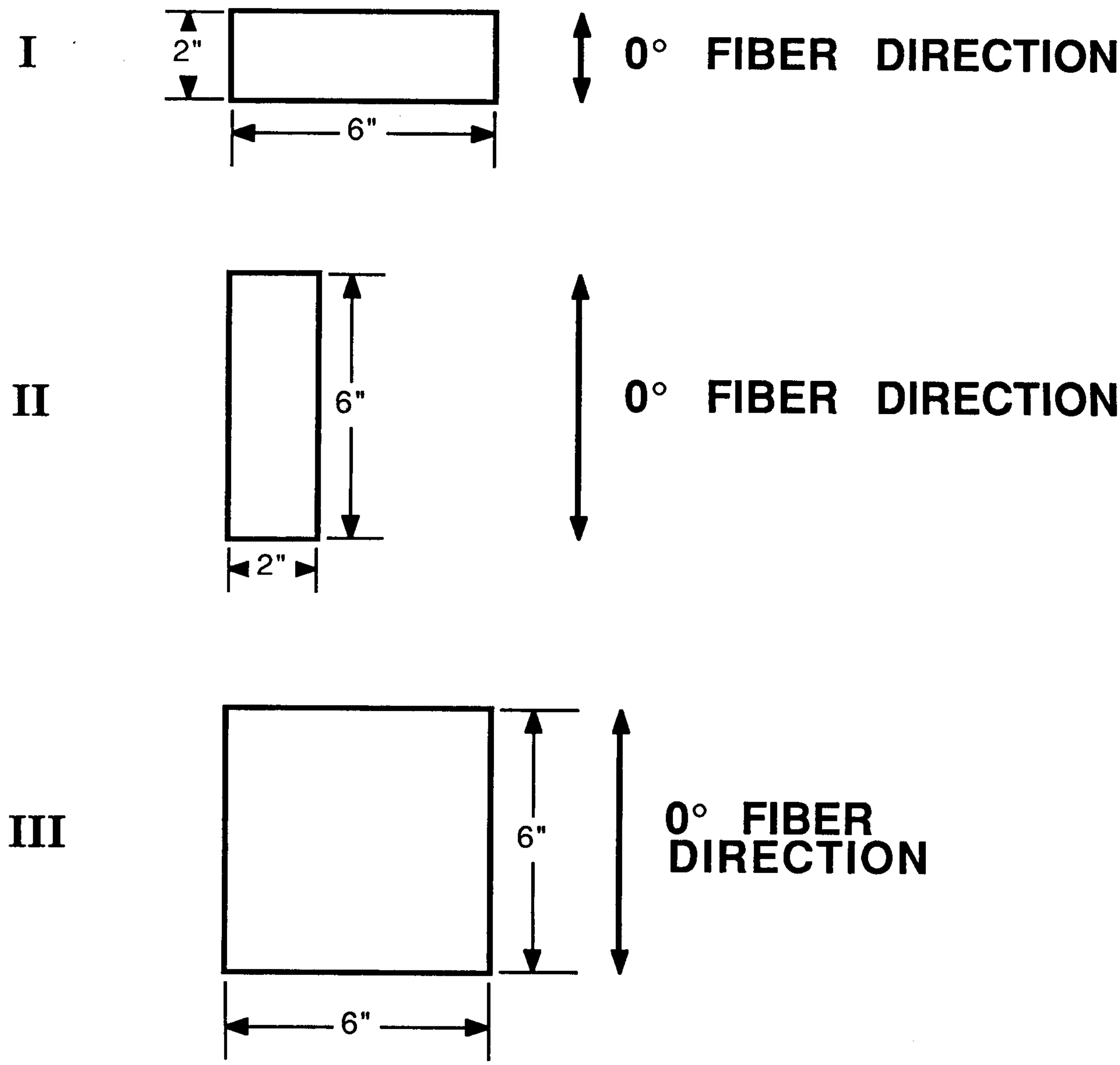

Figure 2. Test panel configurations.

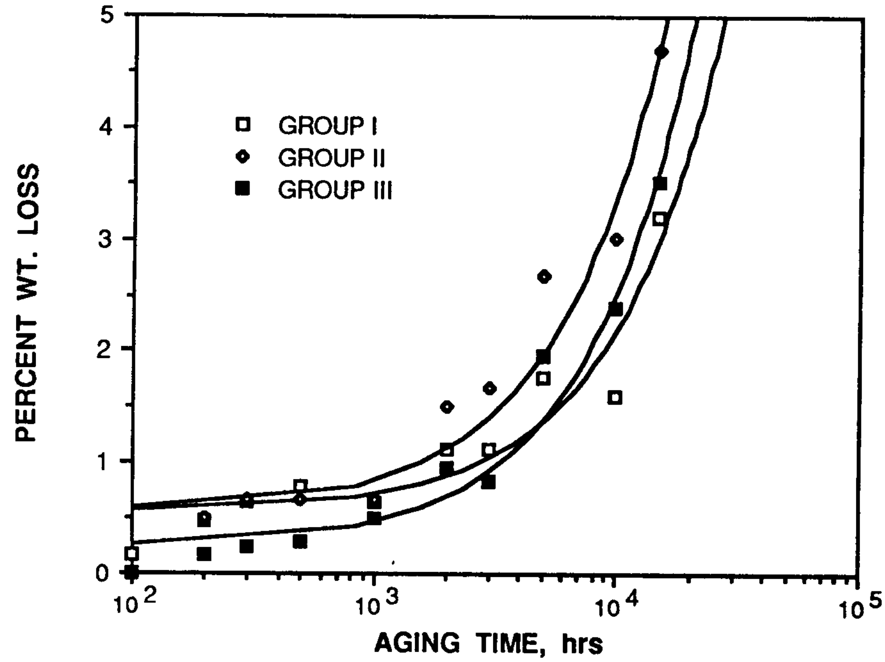

Figure 3. Weight loss of 4-ply unidirectional panels of Celion/LARC-160 aged at 204°C.

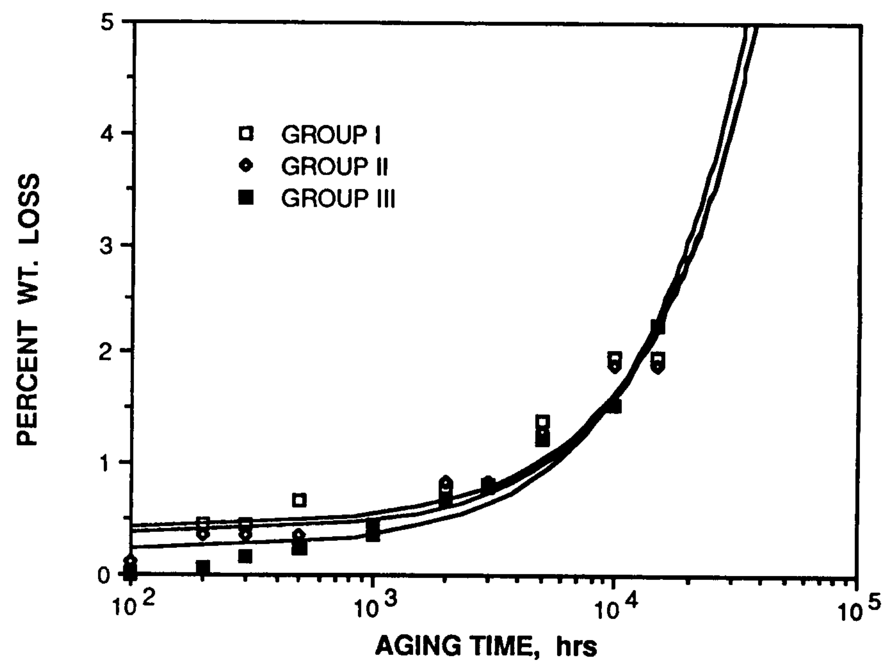

Figure 4. Weight loss of 8-ply unidirectional panels of Celion/LARC-160 aged at 204°C.

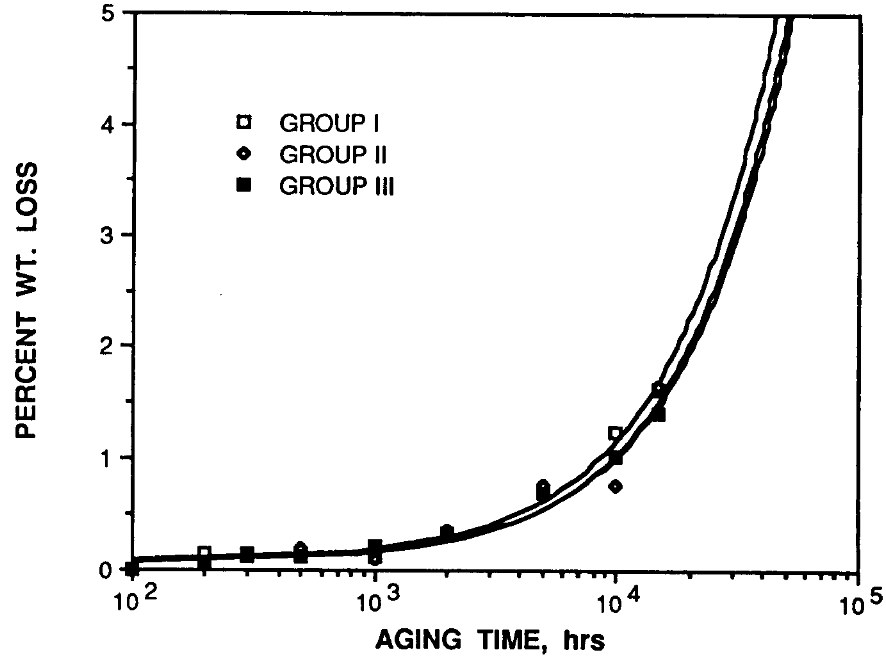

Figure 5. Weight loss of 16-ply unidirectional panels of Celion/LARC-160 aged at 204°C.

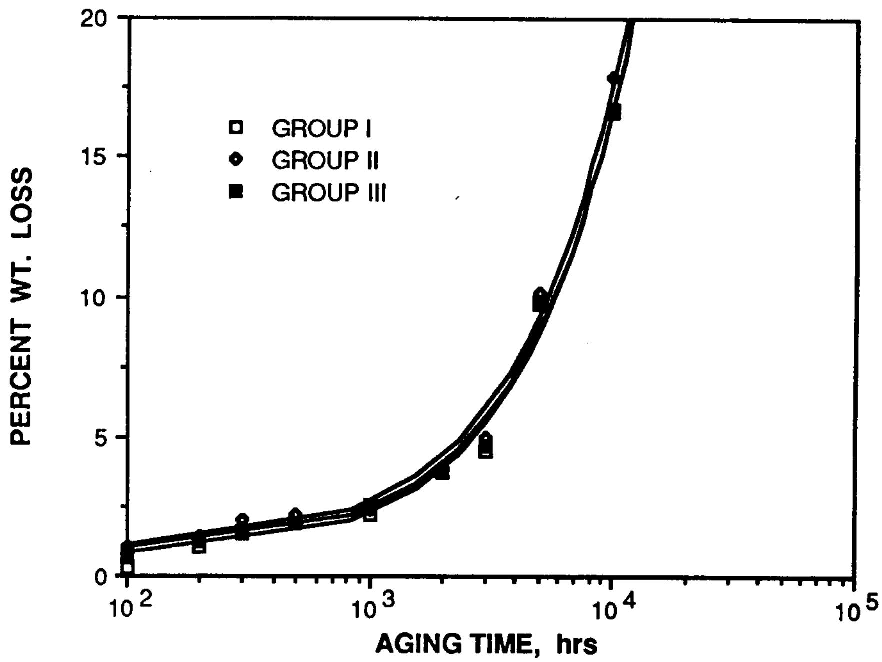

Figure 6. Weight loss of 4-ply unidirectional panels of Celion/LARC-160 aged at 260°C.

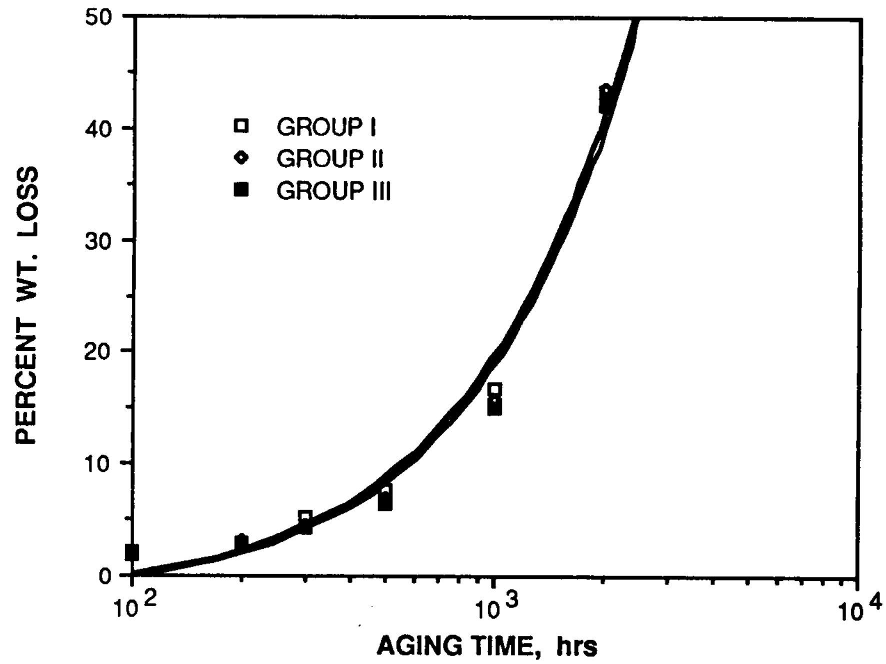

Figure 7. Weight loss of 4-ply unidirectional panels of Celion/LARC-160 aged at 316°C.

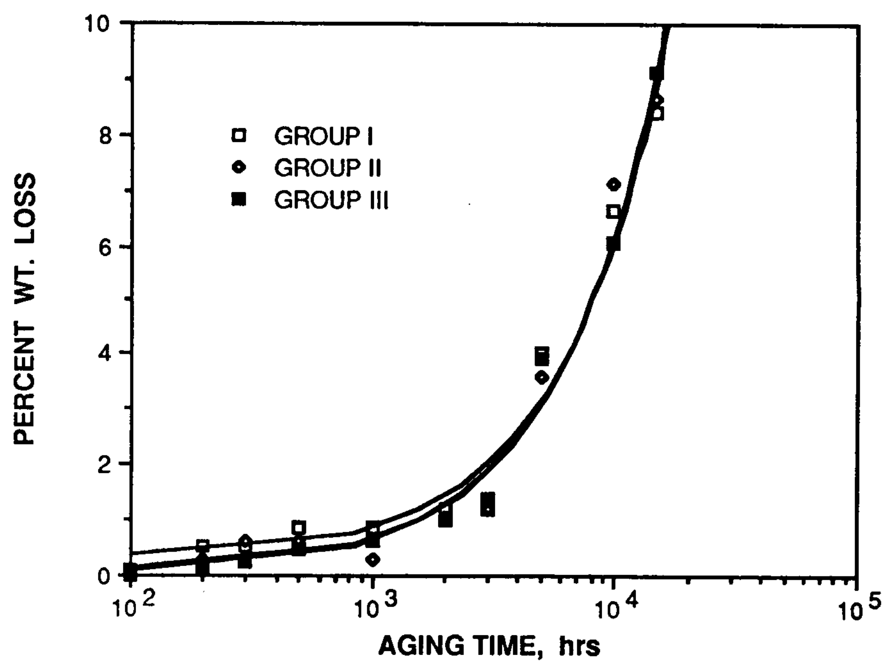

Figure 8. Weight loss of 4-ply X-plied panels of Celion/LARC-160 aged at 204°C.

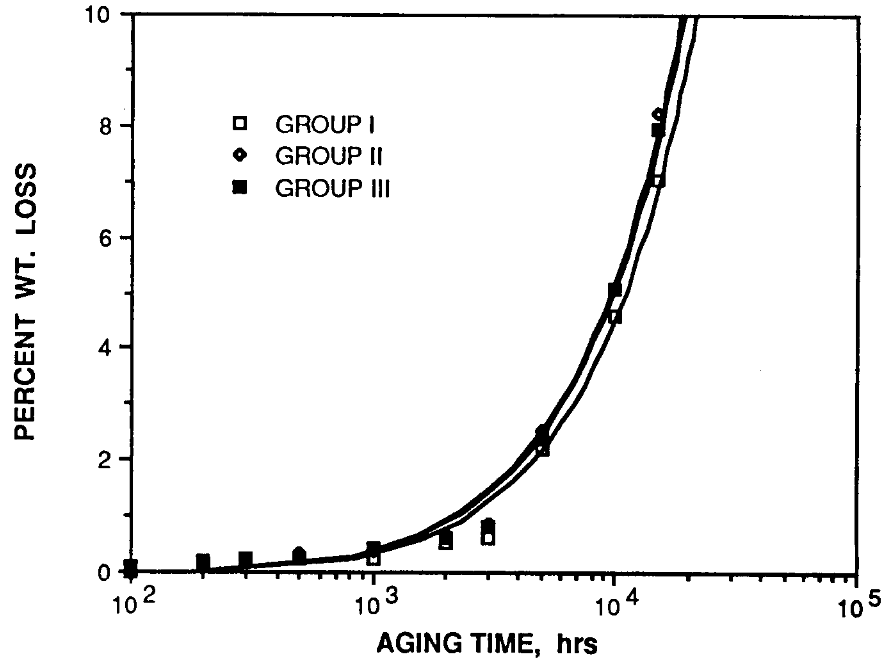

Figure 9. Weight loss of 4-ply fabric reinforced panels of Celion/LARC-160 aged at 204°C.

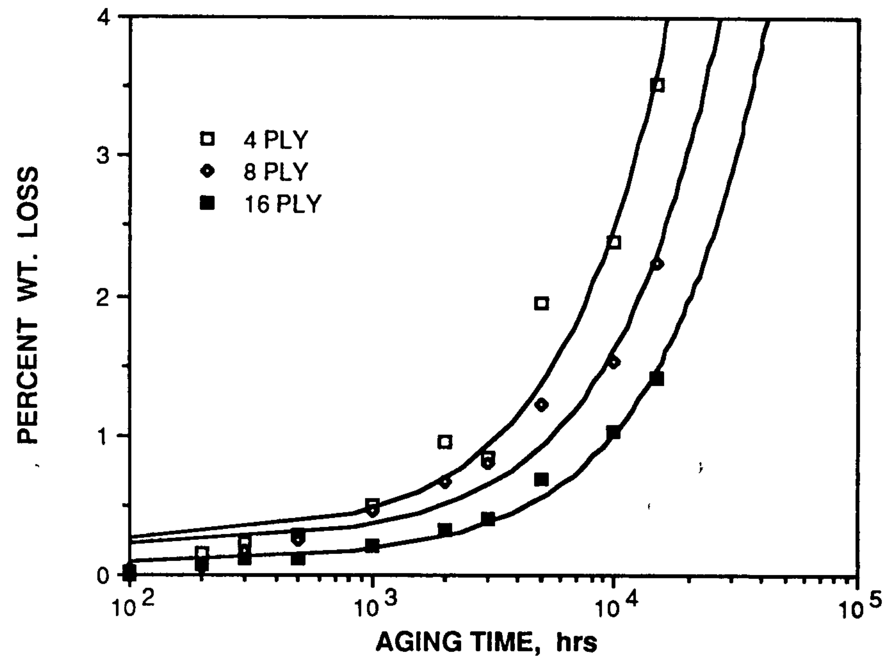

Figure 10. Weight loss variation with panel thickness of unidirectional Celion/LARC-160 square panels aged at 204°C.

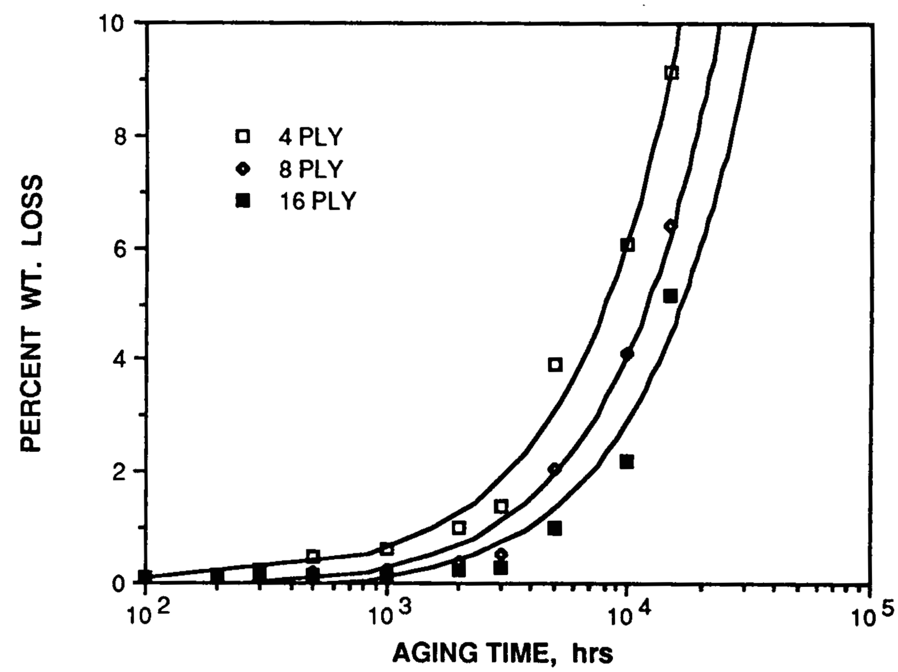

Figure 11. Weight loss variation with panel thickness of X-plied square panels of Celion/LARC-160 aged at 204°C.

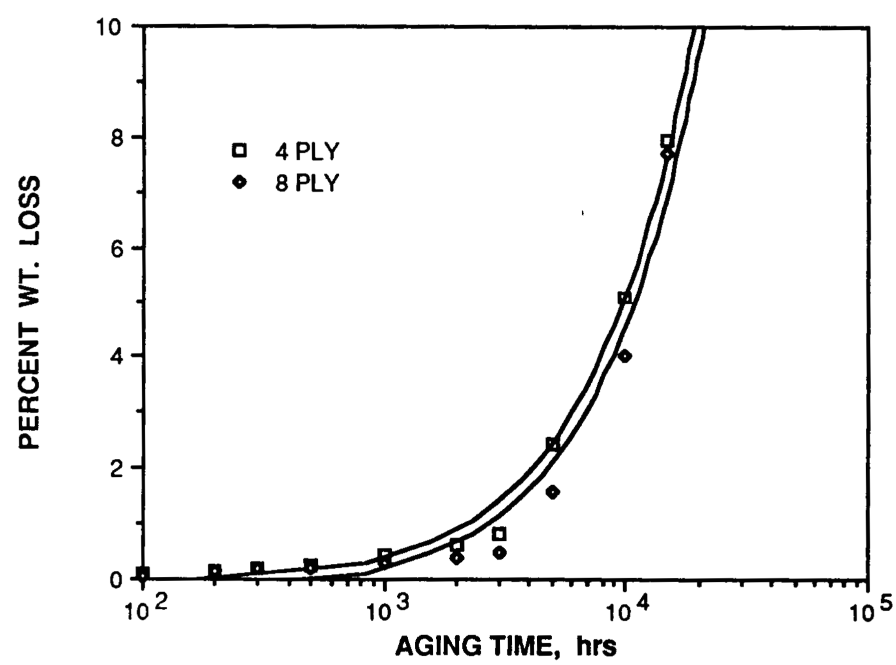

Figure 12. Weight loss variation with panel thickness of fabric reinforced square panels of Celion/LARC-160 aged at 204°C.

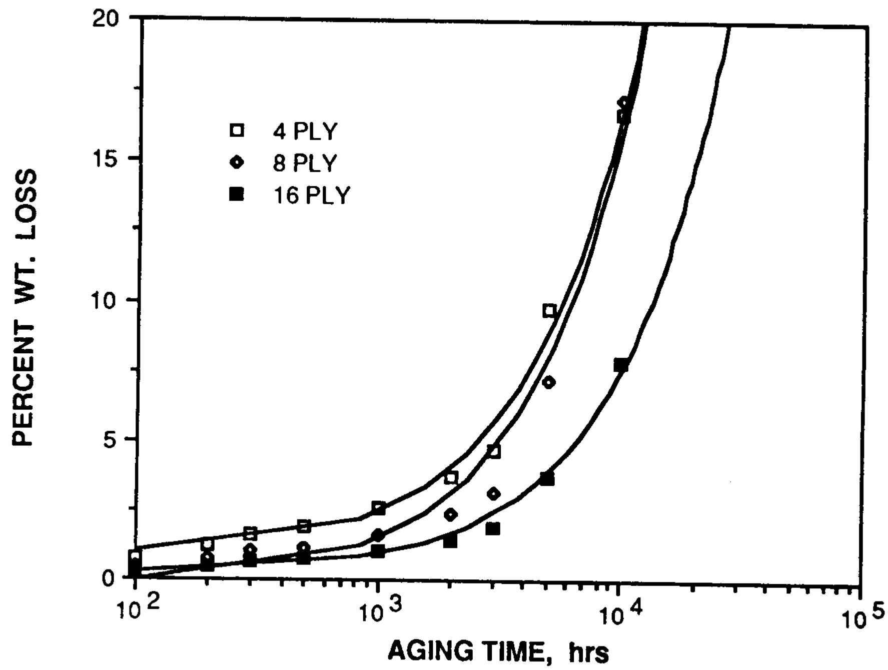

Figure 13. Weight loss variation with panel thickness of unidirectional square panels of Celion/LARC-160 aged at 260°C.

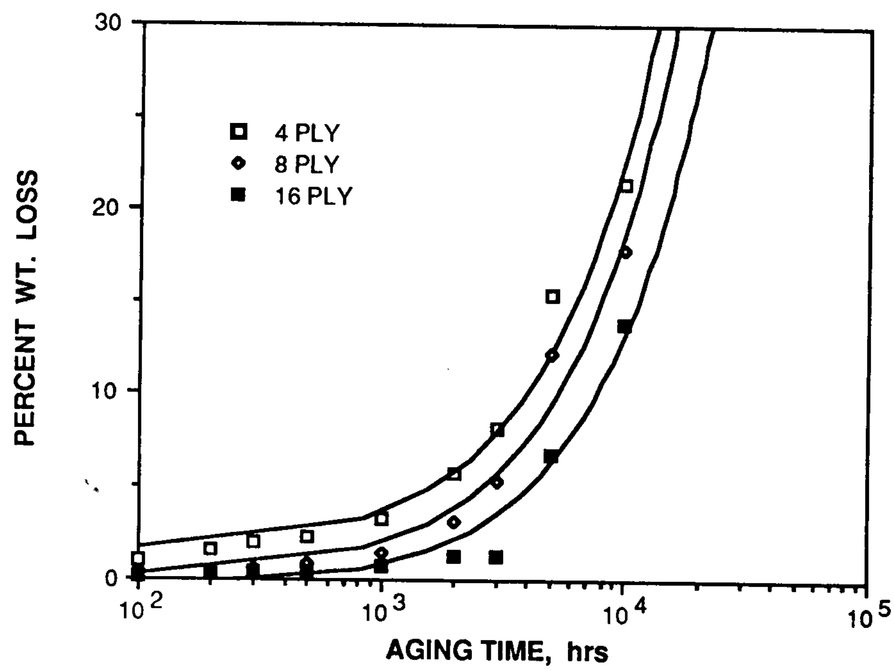

Figure 14. Weight loss variation with panel thickness of X-plied square panels of Celion/LARC-160 aged at 260°C.

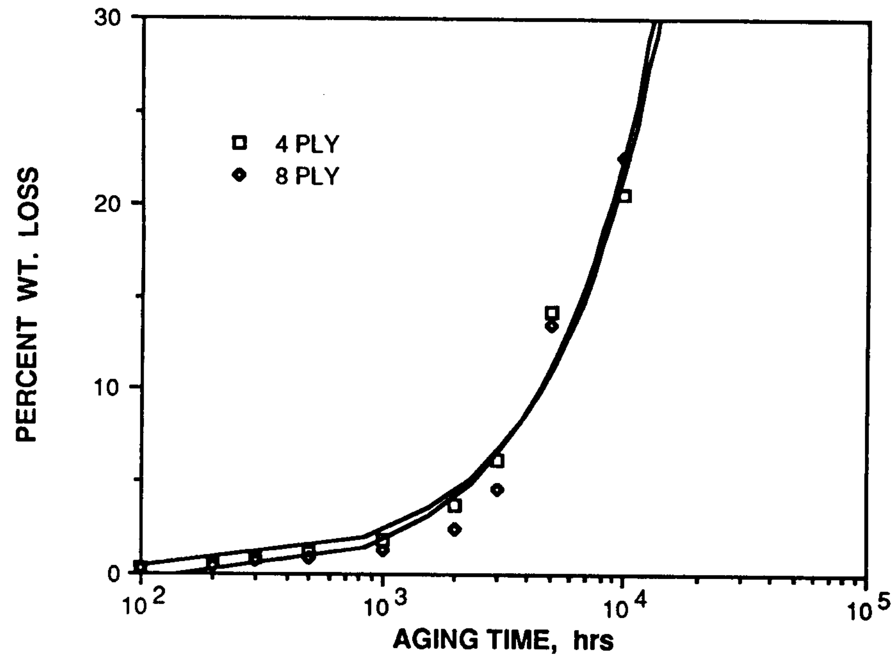

Figure 15. Weight loss variation with panel thickness of fabric reinforced square panels of Celion/LARC-160 aged at 260°C.

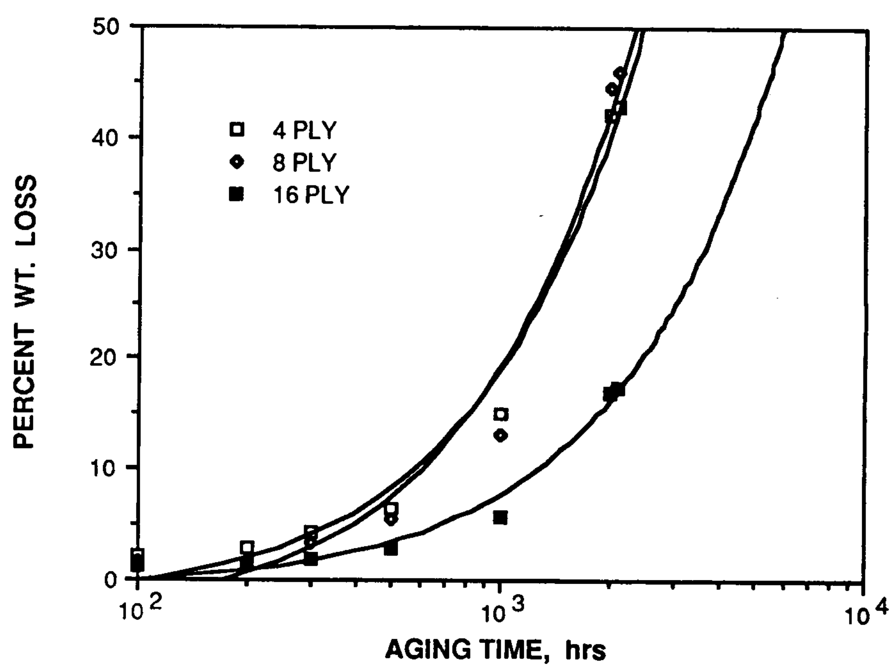

Figure 16. Weight loss variation with panel thickness of unidirectional square panels of Celion/LARC-160 aged at 316°C.

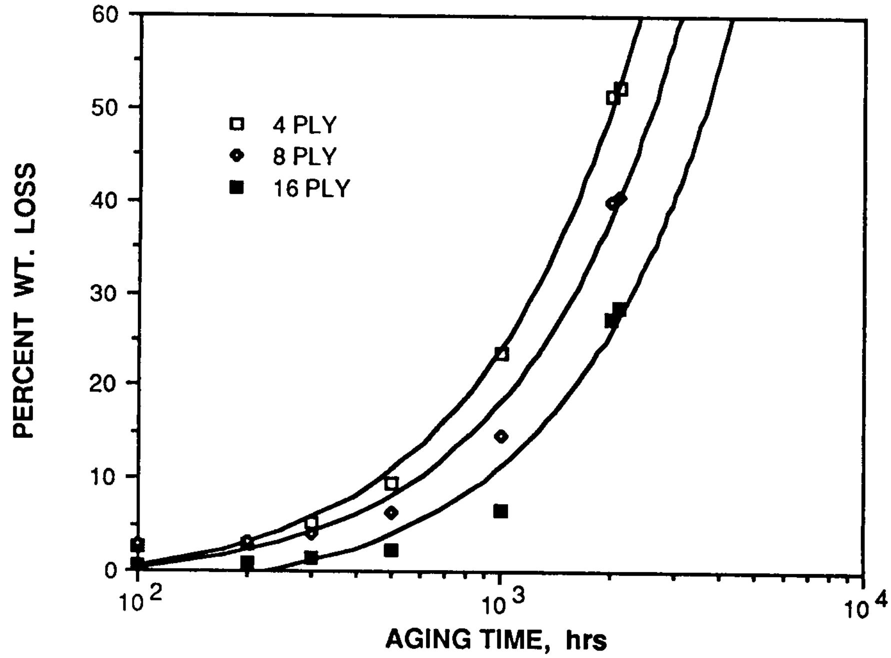

Figure 17. Weight loss variation with panel thickness of X-plied square panels of Celion/LARC-160 aged at 316°C.

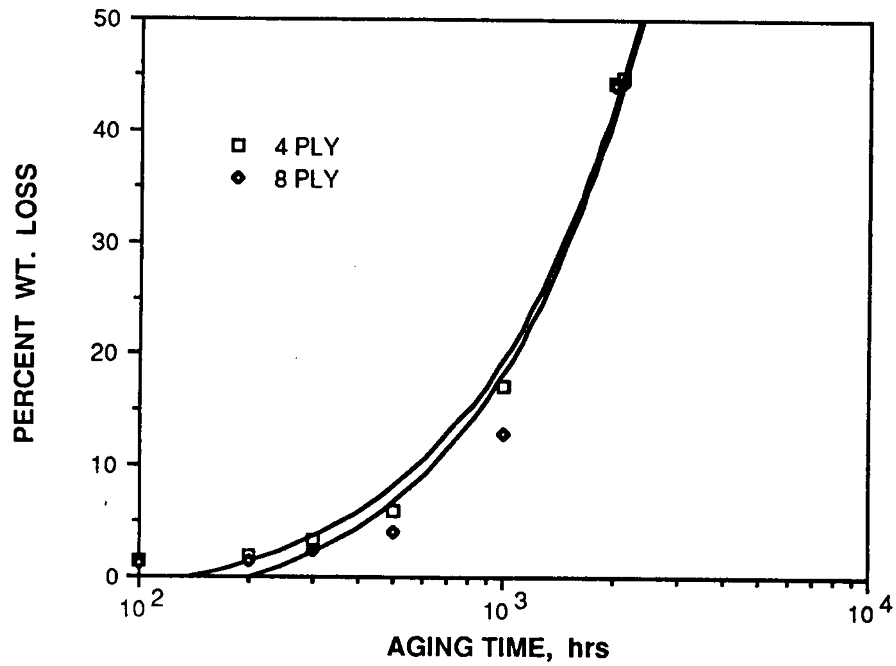

Figure 18. Weight loss variation with panel thickness of fabric reinforced square panels of Celion/LARC-160 aged at 316°C.

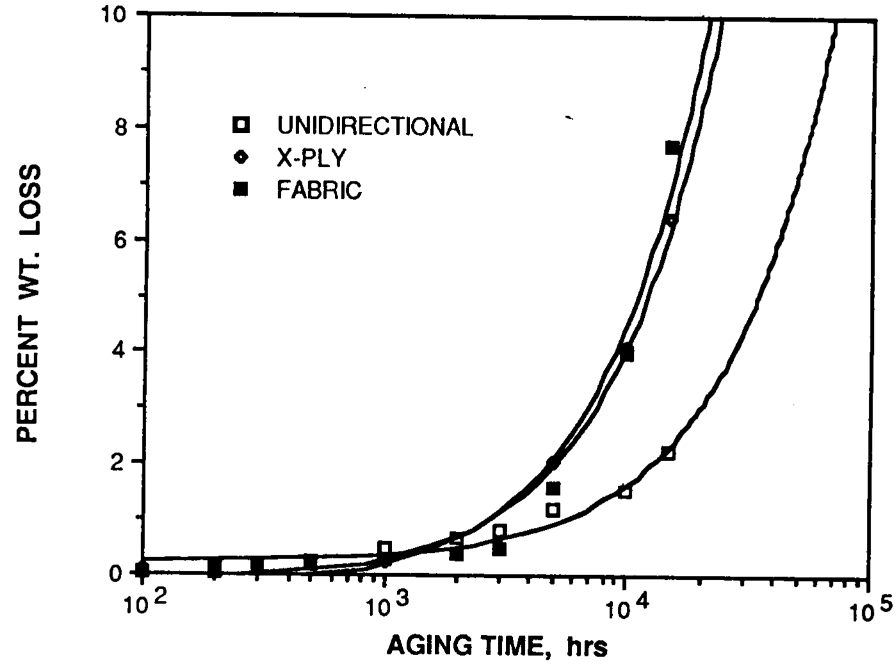

Figure 19. Weight loss of 8-ply square panels of Celion/LARC-160 aged at 204°C.

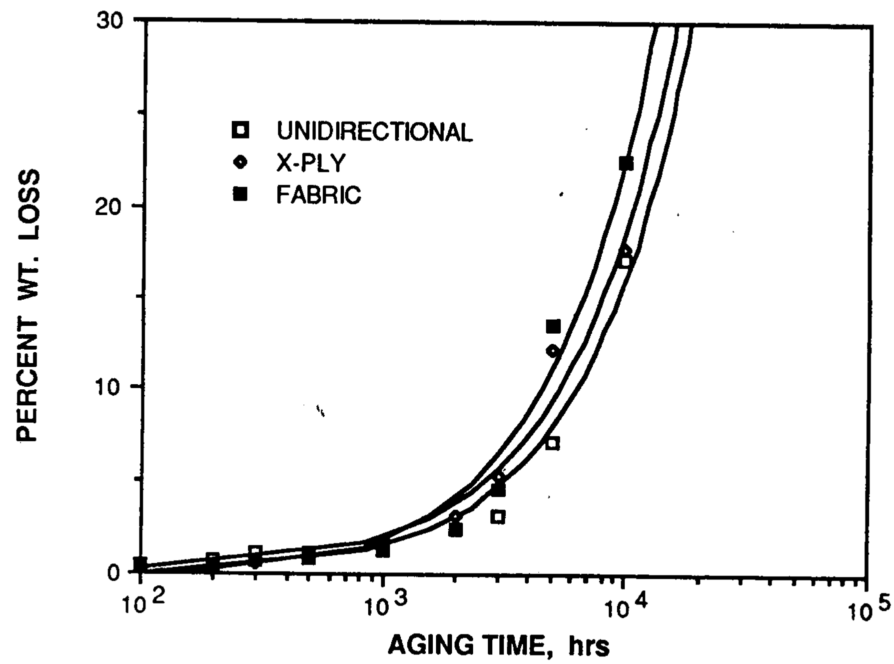

Figure 20. Weight loss of 8-ply square panels of Celion/LARC-160 aged at 260°C.

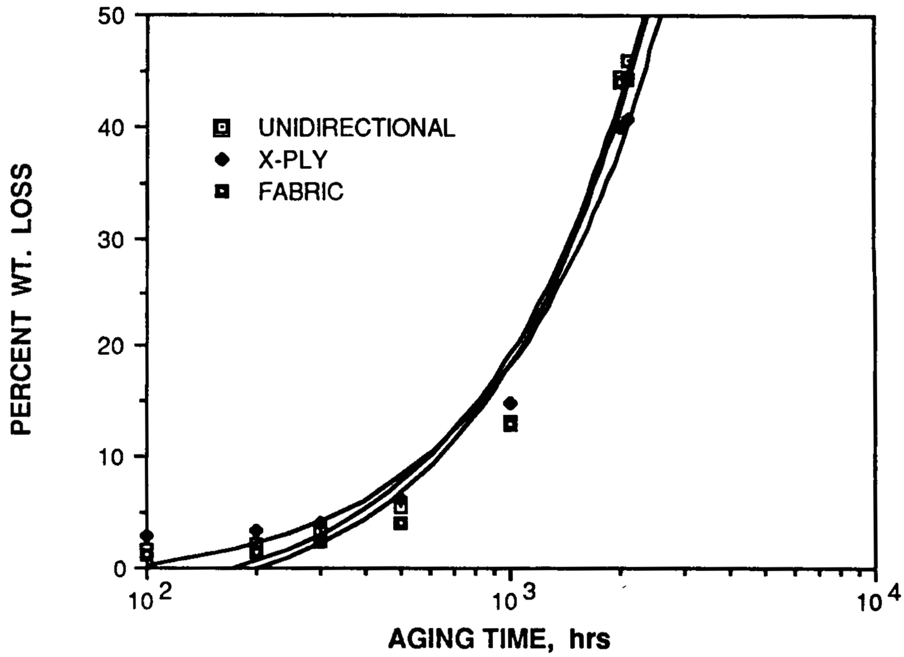

Figure 21. Weight loss of 8-ply square panels of Celion/LARC-160 aged at 316°C.

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16. Abstract

Laminates of Celion®/LARC-160, fabricated in thicknesses from 4 to 16 ply and in unidirectional, x-ply and fabric ply configurations, were isothermally aged at temperatures of 204, 260 and 316°C for periods up to 15,000 hours. Weight-loss of the test panels was measured at selected intervals during aging. At the lower aging temperatures, it was observed that panel thickness and ply arrangement influenced the apparent stability: i.e., thicker panels degraded less than thin panels and unidirectional panels degraded less than x-ply or fabric reinforced panels. At higher aging temperatures, all panel configurations and thicknesses converged toward the same behavior.

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