FATIGUE RESPONSE OF HYBRID MAGNESIUM/CARBON-FIBER/PEEK NANOCOMPOSITE LAMINATES AT ELEVATED TEMPERATURE

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ABSTRACT

High performance Mg based hybrid five-layered composite laminates were fabricated by means of sandwiching the AZ31 Mg foils with the Carbon/PEEK prepregs, in which the nanoparticles SiO₂ were uniformly spreaded at the interfaces, through hot press. The basic mechanical properties of the cross-ply hybrid laminates were obtained by tensile tests, and they were found in good agreement with the prediction by the rule of mixture. Then, the response due to constant stress amplitude tension-tension (T-T) cyclic loading at elevated temperature up to 10⁶ cycles was investigated. The received S-N curves, when normalized by their corresponding ultimate strength, were found very close to each other. That strongly hints the higher resistance to fatigue of cross-ply hybrid laminates at higher temperature is accomplished.

KEY WORDS

Nanocomposite, laminate, mechanical properties, rule of mixture, fatigue.

INTRODUCTION

Magnesium alloys have attracted considerable attention and interest worldwide during the past few years. Because of its low density of 1.7-1.8 Mg/m³, this metal provides the possibility of weight saving in various metallic structures in aircraft, vehicles and other transportation equipment [1-3].

It is well known that the continuous carbon fiber (CF) reinforced PEEK polymer composites possess extraordinary specific strength and stiffness along the longitudinal (or fiber) direction with fiber content up to 61% by volume (vol.). The CF/PEEK composite appears to be a shining star among all PEEK composites, and is being considered as the candidates to replace the conventional epoxy-based composites for aerospace applications.

The inclusion of inorganic fillers into polymers for commercial applications is primarily aimed at the cost reduction, strength, and stiffness improvements [4]. It is worth noting that the inclusion of micrometer sized particulates into polymers, a high filler content (typically greater than 20 vol.%) is generally required to bring the above stated positive effects into play. This would detrimentally affect some important properties of the matrix polymers such as process ability, appearance, density and aging performance. Therefore, composites with improved performance and low particle contents are highly desired. With this concern, the newly developed nanocomposites, i.e., polymers or metals reinforced by nano sized fillers, would come into the competitive candidates.

Until now, it is well-known that fiber-reinforced aluminum laminates (FRALL) have been successfully fabricated and commercialized [5]. The aramid fiber-reinforced aluminum laminates (ARALL) were marketed by the Aluminum Company of America for the applications, such as aircraft lower wing skin, fuselage and tail skins [6]. Moreover, carbon fiber-reinforced aluminum laminates (CARALL) show a superior crack propagation resistance under T-T fatigue [7]. All the above-developed FRALLs contain epoxy-resin polymer, consisting of alternating layers of thin aluminum sheets bonded by layers of high-strength fiber/epoxy prepreg. The service temperature is not expected to exceed 373 K.

In this study, the laminated hybrid nanocomposites were developed for the Mg base alloys, using the widely applied AZ31 sheets, reinforced with the high strength unidirectional CF/PEEK prepregs, in which the optimal nanoparticles of SiO₂ were uniformly spreaded at the interfaces [8]. The PEEK polymer can sustain its mechanical properties up to 423 K or even 473 K [9, 10], thus it is postulated that the current Mg base Mg/Cf/PEEK laminated composites might be utilized at higher temperatures. Then, the mechanical properties at elevated temperature subjected to tensile and cyclic tests were obtained. The failure mechanisms were observed. The improvements of mechanical properties were highlighted.

EXPERIMENTAL

The prepregs of Carbon/PEEK (ICI Fiberite Co., USA) unidirectional plies were cut and stacked into cross-ply [0/90]₉, laminates. The nanoparticles SiO₂ (Wah-Li Co.) possessed the average diameter 15±5 nm, specific surface area 160 ± 20 m²/g, spherical crystallographic and amorphous powder. The optimal amount of SiO₂ was found 1% by wt. of laminate (3% by wt. of PEEK, Vf = 61%) spreaded uniformly on four plies in laminates [8]. The modified diaphragm curing process was adopted as shown in Figure 1.
The AZ31 (Al: 2.5-3%, Zn: 0.6-1.4%, Mn: 0.15-0.7%, Mg: balance) Mg sheet was supplied by Grandmont CO., LTD., Taiwan. The thickness of an alloy sheet is 0.5 mm after rolled, heated and weight flattened with scratch brushing. The density of AZ31 is 1.78 Mg/m³, melting point temperature is 627 °C, ultimate strength is 260 MPa, and stiffness is 2.07 GPa.

Prior to lamination, the slurred AZ31 sheets were subjected to pretreatment by special chemicals in order to create the rough surfaces for better bonding with the APC-2 prepregs. The first try was to mechanically polish the sheet surface using #400 SiC abrasive papers without chemical etching. The following groups were etched by various chemical combinations including HCl, HNO₃, H₂SO₃, (NO₂)₂C₆H₅OH and Cr₂O₃-base etchants, etc. After a series of tests, Cr₂O₃-base etchant was adopted.

Various layers of the APC-2 prepreg were sandwiched with the AZ31 sheets to produce Mg/CF/PEEK laminated nanocomposites. The stacking sequence was AZ31/APC-2/AZ31/APC-2/AZ31 in unidirectional array (with the rolling direction parallel to the continuous fiber direction). Forming of the Mg/APC-2 composite was conducted by means of hot pressing at 400 °C under a pressure of 5.5 kg/cm² for a time duration of 15 min. The hot pressing was conducted under vacuum at 70 cmHg. Actually, the curing process was adopted as shown in Figure 1. The geometry and dimensions of hybrid nanocomposite specimens were shown in Figure 2.

Figure 2. (a) Schematic drawing of the Mg based laminted composite, layers 1, 3 and 5 are Mg and layers 2 and 4 are APC-2 (with 4 foils) . The longitudinal direction is indicated; (b) dimensions of a specimen (unit: mm).

An MTS-810 servohydraulic computer-control dynamic material testing machine was used to conduct the constant stress amplitude T-T cyclic testing with stress ratio = 0.1, frequency = 5Hz, sinusoidal wave form under load-controlled mode at elevated temperatures, such as 25 deg C (RT) [11], 75, 100, 125, 150 deg C (slightly above APC-2 Tg = 143 deg C). An MTS 651 hot chamber was also installed to keep and control the specific temperature of a specimen inside for cyclic testing. A 25.4-mm MTS-634.11F-25 extensometer was used to monitor the strain continuously during the fatigue tests.

RESULTS

The hybrid nanocomposite laminates were fabricated successfully. Statistically, it was found about 15% of total specimens were delaminated by the cutting of panels. However, there was no delamination was observed during the tensile and cyclic tests. The mechanical properties of Mg/APC-2 hybrid five-layered composite laminates of both lay-ups were listed in Table 1.

<table>
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<tr>
<th>Table 1. Mechanical properties of Mg/Cross-ply and Mg/Quasi-isotropic APC-2 composite laminates at RT.</th>
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<td><strong>Properties</strong></td>
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* The experimental data.
** The results predicted by ROM

The predicted results of ultimate strength (σₑₑ) and longitudinal stiffness (Eₑₑ) by rule of mixture (ROM) were also presented in columns 3 and 5 of Table 1 in contrast, respectively. The volume fraction of Mg was 56.23% and that of APC-2 was 43.77%. The mechanical properties of APC-2 were adopted from the previously published data [8]. It was found that only the predicted ultimate strength of hybrid cross-ply laminates was over 10% in comparison with empirical data. Others predicted by ROM were quite close to the obtained data.
The applied stress vs. cycles (S-N) curves of hybrid nanocomposite cross-ply laminates at elevated temperature were presented in Figure 3.

![Figure 3. The S-N curves in hybrid cross-ply nanocomposite laminates at elevated temperature.](image)

The normalized stress vs. cycles curves of hybrid nanocomposite cross-ply laminates were described in Figure 4.

![Figure 4. Normalized S-N curves in hybrid cross-ply nanocomposite laminates at elevated temperature.](image)

It was found that the normalized S-N curves were close to each other.

**DISCUSSION**

According to the published data in [12], the theoretical predication by ROM fits well with the empirical data of Mg/APC-2/Mg/APC-2/Mg composite laminates without adding nanoparticles. However, there existed over 10% error in predicting the strength of hybrid nanocomposite cross-ply laminates in Table 1, that could be mainly attributed to the adding of SiO₂ nanoparticles in APC-2 resulted in the increase of strength.

Compared with experimental databank in [13], Thermal cycling tests were performed on hybrid nanocomposite cross-ply and quasi-isotropic laminate in the temperature range between 25 and 150 deg C until maximum cycles, $10^6$, the applied stress and the fatigue life of the samples were investigated. S-N curves show the stress-life (stress versus logN: stress is the applied stress and N is the number of cycles to fatigue failure or intact until period of $10^6$ cycles) for the samples of hybrid magnesium/carbon-fiber/PEEK nanocomposite laminates at elevated temperature. It is found that at each cycles the applied stress gradually decreases as the temperature increasing for both hybrid nanocomposite cross-ply and quasi-isotropic laminates, respectively. That evidently shows the increase of temperature will reduce the resistance to cyclic loading in thermoplastic matrix laminates. It is caused that the PEEK matrix properties are gradually affected by environmental heating, close to or over Tg, compared to AS-4 fibers. The symbol, $\gamma$, is the fatigue limit, which means that the specimens will be intact after 1 million fatigue cycles. Observations of the samples stress and life relationship at elevated temperature showing the extent of damage indicate that as the temperature increases the PEEK matrix strength at which the brittle to ductile transition occurs to decrease bonding between the fiber and matrix significantly, especially the ambient temperature close to or over Tg. The present researches show that when the environmental temperature less than Tg, hybrid APC-2 nanocomposite laminate is an appropriate system to use in the brittle phase of PEEK matrix, i.e., the degree of crystallinity in hybrid APC-2 nanocomposite system under Tg effectively forced the bonding between the filaments and resin. In order to account the ductile phase of propagation, bonding force in matrix softening that caused the S-N curves in both hybrid nanocomposite lay-ups should be available. On the other hand, from S-N curves in both hybrid nanocomposite lay-ups the general behavior of both lay-ups shows that the most serious degradation of normalized stiffness is at the highest temperature, however, they will possess at least 25-30 % normalized stiffness until $10^6$ cycles. The temperature effects on apparent strength and stiffness are shown previously. So, it is usually caused the degradation in longitudinal stiffness of the lay-up structures by the development of internal PEEK failure. It is noted that the fatigue strength and longitudinal stiffness drop significantly. Since resin system are dominated by the PEEK matrix, which is sensitive to temperature variation, the matrix properties decrease significantly as temperature increases, especially at temperature near the glass transition temperature, while the AS-4 fibers properties is not sensitive to temperature variation. However, the thermosetting matrix composites can not sustain such a long time at elevated temperature, i.e., the Tg of common
thermoset epoxy is almost about 88-125 deg C, such as Hexcel carbon epoxy and Loctite 3564 fast flow epoxy. It is an important reason why the APC-2 composite laminates still have been considered to be one of the best materials in aerospace industry, because of their capabilities of resisting severe environmental conditions. Thermal cycling also resulted in interfacial matrix/fiber reaction that probably strengthened the thermoplastic hybrid APC-2 nanocomposite and caused that the environment resistance of these hybrid APC-2 material at elevated temperature is better than thermosetting composite. Good capabilities bring superior properties and better safety, and I think that’s why thermoplastic APC-2 composite would be popularized so much, no matter they are used in automotive industry or aviation industry.

The picture by energy dispersion spectrum (EDS) as shown in Figure 5 illustrates the uniform spreading of nanoparticles SiO₂ without any aggregation that will cause a defect from microfracture point of view.

Figure 5. Mapping by EDS of a hybrid cross-ply nanocomposite laminate.

Figure 6 shows the surfaces treated by abrasive polishing and etching corrosion, it is also found good adhesive bonding between etched Mg sheet and APC-2 laminate.

Figure 6. Surface of Mg foil (a) polished by 40# SiC abrasive paper, (b) then etched by CrO₃, (c) delaminated fracture surface by cutting, (d) well bonded interface after peeling.

That assures no delamination will occur during the static and dynamic tests.

It is obvious that the capability of resistance to fatigue loading of any material will degrade in the case of rise of temperature. Thus we obtained the S-N curves of hybrid
nanocomposite cross-ply laminates became lowered at elevated temperature. However, the S-N curves were normalized by the ultimate strength of corresponding temperature, they were very close to each other. That strongly hints us nanoparticles not only strengthen the mechanical properties, such as strength and stiffness, of hybrid composite laminates, but improve the fatigue resistance even at higher temperature.

CONCLUSION

The Mg/APC-2/Mg/APC-2/Mg five-layered sandwich nanocomposite cross-ply laminates were fabricated optimally and successfully, and cyclic tests at elevated temperature were conducted systematically.

The concluding remarks were summarized as follows.
1. No delamination was observed in tensile and cyclic tests.
2. Mechanical properties and S-N curves were received.
3. The normalized S-N curves were close to each other.
4. The improvements of mechanical properties and fatigue resistance were achieved.

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REFERENCES