Carbon Science

Vol. 1, No. 2 September 2000 pp. 60-63

Studies on ILSS and Acoustic Emission Properties of Carbon-Carbon Composites

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(Received June 24, 2000; accepted August 17, 2000)

Abstract

In this work, the carbon fibers-reinforced carbon matrix composites made with different carbon char yields of phenolic resin matrix have been characterized by mechanical flexural tests for acoustic emission properties. The composites had been fabricated in the form of two-dimensional polyacrylonitrile based carbon fibers during the carbonization process. It was found that the composites made with the carbon char yield-rich of resin matrix result in better mechanical interfacial properties, i.e., the interlaminar shear strength (ILSS) of the composites. The data obtained from the acoustic emission monitored appeared to show that the composites made with carbon char yield-rich were also more ductile. From the acoustic emission results, the primary composite failure was largely depended on the debonding at interfaces between fibers and matrix. The interlaminar shear strengths of the composites were correlated with the acoustic emission results.

Keywords : *Carbon Fibers, Acoustic Emission, Phenolic Resin, Carbon Char Yield, Interlaminar Shear Strength.*

1. Introduction

It is well known that the carbon fibers-reinforced carbon matrix composites, or simply, carbon-carbon (C-C) composites consist of a fibrous carbon substrate in a carbonaceous matrix. They are developed due to the demands of the aircraft and aerospace industries. Their use as rocket propulsion components, reentry tips, exhaust cones and friction pads, leads to the evaluation of their ablation performance [1, 2]. Phenolic resins are systematically being incorporated into carbon and graphite technology. Today they are commonly used as starting materials, in the resin-based process, for C-C composite preparation by a procedure employing pyrolysis and carbonization of resin matrix and subsequent densifications. Since they are easy to handle and give considerable carbon char yields (50~70% at 800 in a nitrogen atmosphere) [3-6]. Meanwhile, it is known that acoustic emission (AE) is a transient elastic wave generated by the rapid release of energy within materials as they undergo deformation or fracture. In fibers-reinforced composites, the easily recognizable sources of AE have been studied in the generation, propagation, and detection of composite cracking for the glass fibers- [7], carbon fibers-reinforced epoxy matrix [7-9], and alumina fibers-reinforced aluminum composites [10]. However, the AE studies on extremely heat-treated C-C composites have been studied in a very few [11].

The objective of the present work is to extend the investigations on the acoustic emission properties, resulting from the matrix cracking, debonding at fiber-matrix interfaces, fiber fracture, and delamination, of the C-C composites made with different carbon char yields of phenolic resins. During the mechanical flexural tests on the C-C composites, the AE is monitored as a function of load. The data recorded are also discussed with the results obtained from the interlaminar shear strength (ILSS) of composites.

2. Experimental

Polyacrylonitrile (PAN) based high-strength carbon fibers (Toray, T-300, 6000 monofilaments), with a weight of 317 $g \cdot m^{-2}$, were used in the form of two-dimensional plain weave made from commercially standard oxidation surface treatment of fibers. The different types of carbon char yield of phenol-formaldehyde resins (supplied by Kangnam Chem. Co., Korea) were used as matrix precursors of the composites to be studied, i.e., carbon yields: $45 \pm 1\%$ (Type I), $55 \pm 1\%$ (Type II), and $65 \pm 1\%$ (Type III) measured at 800°C in an inert atmosphere.

The green composites from the eight plies of resin impregnated carbon fabrics were prepared in autoclave at 1.0 MPa. and 490 K for 60 min. The composites carried out by the liquid phase impregnation and carbonization [1] of up to 1370 K. The heating rate to the carbonization temperature was fixed to 2 $K \cdot min^{-1}$ in an inert atmosphere. The carbonization retention time of the specimens at 1370 K was 120 min. The thermo-mechanical properties were characterized from thermal thickness changes or thermal expansion of the green composites using a du Pont Co. model 9900 thermo-mechanical analyzer (TMA) up to 970 K. The heating rate to the TMA temperature was fixed to 10 $K \cdot min^{-1}$ in an inert atmosphere. The mechanical interfacial properties of the

composites were the short-beam flexural test to obtain the ILSS, according to ASTM D2344 (the distance between supports divided by the thickness of specimen, L/d=4; cross-head speed, 0.5 mm · min^{-1}), as follows:

$$ILSS=\frac{3}{4}\cdot\frac{F}{bd} \quad (1)$$

where, F is the load at the moment of break (N), b the width of the specimen (m), and d the thickness of the specimen (m).

The bulk fiber volume fraction of the green composites was about 60% (± 2%) for all composites, and more than seven specimens were tested for each of the composites studied.

The schematic apparatus of AE detection during the mechanical flexural tests (L/d=16; cross-head speed, 1.0 mm · min^{-1}) is shown in Fig. 1. The acoustic signal was detected by a piezoelectric transducer which converts it into an electrical signal. The AET model MAC300L transducer with a resonant frequency of 300 kHz was located in the test section. Silicon grease was used as a couplant and the sensor was held in place with adhesive tape. This signal was amplified in order to obtain reliable AE data as a function of time. The transducer was connected to a 60 dB, 250-500 kHz AET model 160B bandpass filter and preamplifier which fed the signal to an AET model 5000 AE analyzer. The total amplification of the recording system was 86 dB and the threshold was set at 0.5 V. Three types of data were collected such as event counts, cumulative event counts and relative signal energy distribution derived from the peak amplitude of the AE responses.

3. Results and Discussion

Fig. 2 exhibits the result of TMA measured as a function of heat treatment temperature (HTT) for all composites studied. As expected, it is shown that the behaviors of all composites studied are similar to those of the purely cured phenolic resins [12]. The thermal thickness changes of the specimens studied show in rapidly decreasing in the range of 670~870 K. These observations may be explained by high thermal isotropic shrinkage of the resin matrix, resulting from the outgassing of very-low-weight aromatic species. In this HTT range, it gives an information for the high performance C-C composites, which is related to more detailed thermal behaviors against sudden thickness changes of the composites. Since a sudden isotropic shrinkage of the matrix can influence matrix cracking and, in worst, delamination of composites. In this work, it is not clarified that the thermomechanical behavior of all specimens studied is greatly influenced by different carbon char yields of resin matrix. However, a slightly stable thermal decomposition is observed in the composites made with carbon char yields-rich, as shown in Fig. 2.

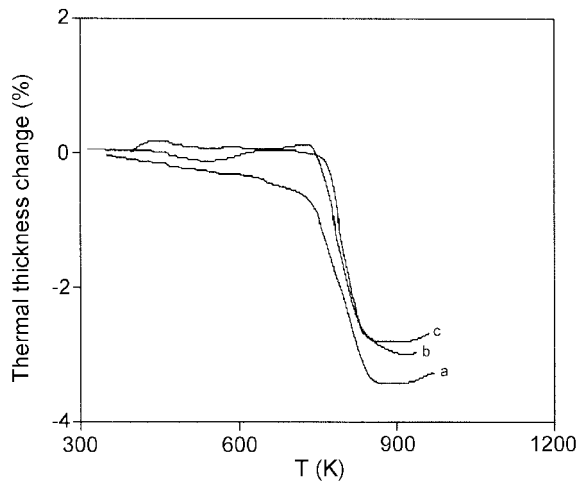

Fig. 2. TMA diagram of the C-C composites studied. Curve a (Type I); b (Type II), and c (Type III).

In Fig. 3, the evolutions of bulk density of the composites characterized by the water displacement technique, according to the ASTM D796-66 as functions of number of densifications are shown. As expected, these results indicate that the bulk density of C-C composites increases as the densification increases. Also, the composites made with higher car-

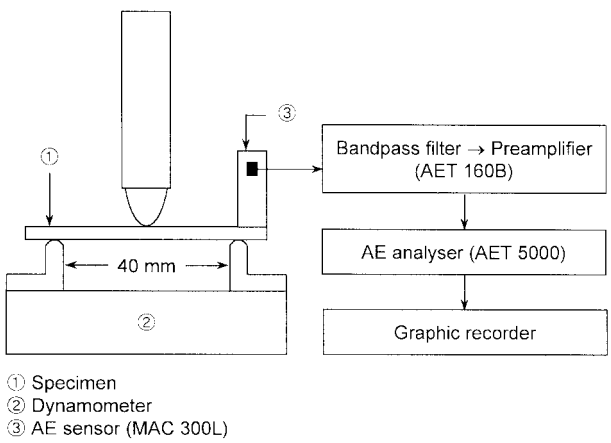

Fig. 1. Schematic diagram of AE detection during the mechanical test.

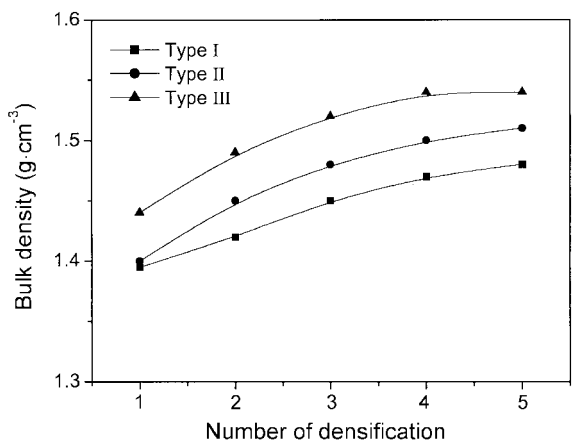

Fig. 3. Evolutions of bulk density of the composites as functions of number of densifications.

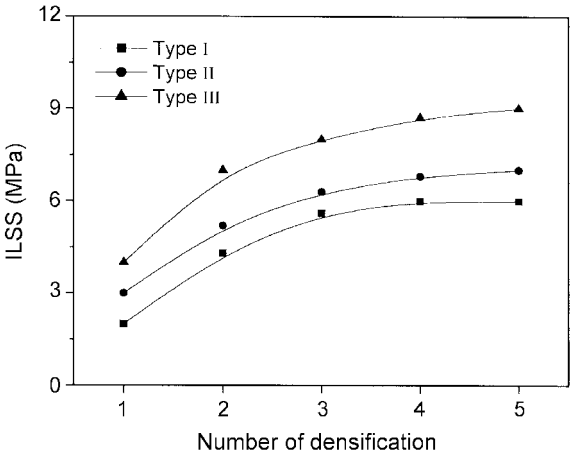

Fig. 4. Results of ILSS of the composites as functions of number of densification.

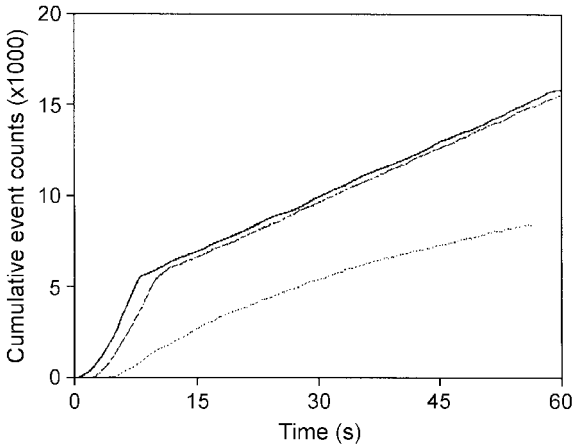

Fig. 6. AE event counts of the C-C composites as a function of elapsed time. — (Type I), – · – (Type II), and ····· (Type III).

bon char yield have relatively higher bulk densities than do the composites made with lower carbon char yield of the resin matrix.

Meanwhile, ILSS is recognized as one of the critical failure modes in fibers-reinforced composites, and depends greatly on the fiber-matrix interfacial adhesion and resin matrix properties [13]. In this work, the ILSS is only influenced by the different types of carbon char yields of the resin matrix. The results of the ILSS of composites studied as a function of the number of densification is shown in Fig. 4. It becomes possible to observe that, as the number of densification increases or the bulk density of C-C composites increases, as shown in Fig. 3, the ILSS of the composites systematically enhance with increasing the carbon char yields of resin matrix. This seems to be a consequence of the resulting in reducing the sudden thermal matrix degrading or in improving the interfacial adhesion areas between fibers and matrix in a composite system during pyrolysis.

Fig. 5 shows the typical examples of AE responses for all composites studied after first densification cycle, versus elapsed time of up to 60 sec. As an experimental result, it is clearly found that the maximum event counts of AE of all composites studied are built up between zero and 15 sec, and retard the elapsed time as increasing the carbon char yields of resin matrix, together with decreasing the maximum AE event counts. This observation can be explained by improving the physical or intermolecular forces between the fibers and the resin matrix of a composite [14], as indicated in above ILSS results of Fig. 4.

In Fig. 6, another identifiable behavior exhibits that the cumulative acoustic emission events versus the elapsed time of the C-C composites decreases with increasing the carbon char yields of the resin matrix. The AE responses for the composites made with higher carbon char yield (Type III) may possibly be explained as arising because the residual carbon char in a composite causes to have intrinsic ductile properties after pyrolysis.

The relative energy distribution of AE events derived from the peak amplitude of AE responses during elapsed time (here, 60 sec) is presented in Fig. 7. In this Figure, three bundle sections in Gaussian-like models are observed in each case of all composites studied. It is generally accepted

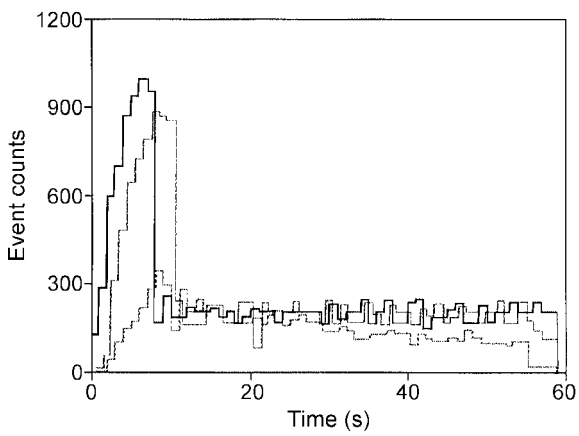

Fig. 5. AE event counts of the C-C composites as a function of elapsed time. — (Type I), – · – (Type II), and ····· (Type III).

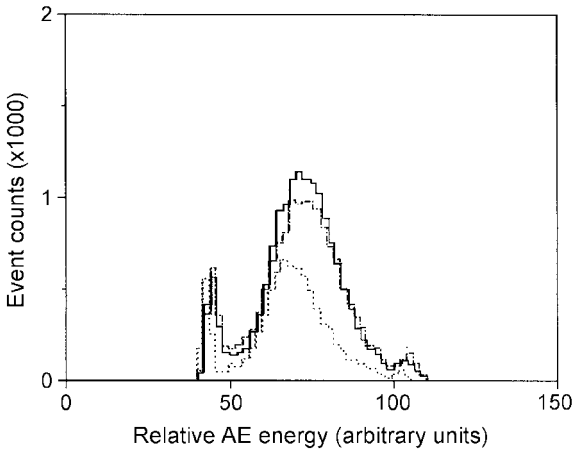

Fig. 7. AE event counts of the C-C composites as a function of relative signal energy during the elapsed time of 60 s. — (Type I), – · – (Type II), and ····· (Type III).

that the increased AE event counts are due to composite damage and that the event counts with low, middle and high range responses are generated from matrix cracking, debonding at the fiber-matrix interfaces, and fiber relaxation or fracture during composite failure, respectively [8-10]. Therefore, it is found that the damage of C-C composites would be largely dominated by debonding at fiber-matrix interfaces, as demonstrated in Fig. 7. This investigation is reasonable, considering that excellent mechanical properties and long durability of the composites necessitate a good fiber-matrix interfacial adhesion, which transfers the load stress from one matrix to the other via the fiber [14, 15]. As mentioned above the ILSS results, this is due to the increase in the ductile interface properties between fibers and matrix in a case of the composites made with the carbon char yield-rich of the resin matrix. It is thus noted that an effective and optimized interfacial property between carbon fibers and phenolic resin matrix may be necessary to improve the intrinsic ductile behaviors for high-performance two-dimensional C-C composites.

4. Conclusions

From the results of short-beam bending tests, the increasing of carbon char yields of the matrix precursors leads to an improvement of the mechanical interfacial properties of the C-C composites. In particular, the results obtained from AE studies show that the major damage to the composites is dominated by debonding at the fiber-matrix interfaces. This seems to be a consequence of the resulting in growing the fiber-matrix physical or intermolecular adhesion of the C-C composites, which could be proved in ILSS behaviors. This resemblance suggests that the suitable of high carbon char yields may be expected either to attain useful levels of thermo-mechanical properties or to achieve good interfacial adhesion of C-C composites.

Acknowledgement

This work is partially supported by grant No. 2000-2-30100-011-3 from the Basic Research Program of the Korea Science & Engineering Foundation.

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