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Effect of thermal strain inhomogeneity on fiber/matrix interface debonding for carbon fiber-reinforced polymer matrix composite

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Abstract

The thermal strain distribution in CFRP around interfaces on the temperature dependence ranging from 170K to 370K was investigated by in-situ FE-SEM observation induced the heating and cooling stage into the FE-SEM chamber. CFRP laminated with 0 and 90 degree orientation fibers was fabricated using PAN-based, pitch-based carbon fibers with epoxy matrix. The electron moiré method was applied to measure the laminate residual strain and digital images correlation method was applied to measure the localized deformation and strain distribution around the interfaces at nm scale acquired before and after thermal loading. The non-uniformity residual strain at different length scales, then debonding and damage evolutions at the interfaces such as fiber/matrix interface and the laminated interface are discussed.

1. Introduction

Carbon fibers exhibit excellent engineering properties as a reinforcement in composite materials for the aerospace, automotive, infrastructure and sporting applications (Fitzer1989). Carbon fiber-reinforced polymer matrix composites (CFRP) are candidate for space structures due to their high specific stiffness and low coefficient of thermal expansion (CTE). CFRPs are typically fabricated by stacking sequence with multi-plies of different fiber directions, and curing at elevated temperatures under pressure and/or in vacuum. When a CFRP is cooled down to room temperature from the fabrication temperature, the residual stress and/or strain arise from the differential CTE of the fiber and the matrix, and the laminate residual stress also arise from the difference between the ply CTEs in the longitudinal and the transverse directions due to the mismatch in the thermomechanical properties of the fiber and the matrix. Carbon fibers are fabricated from three organic precursor materials of polyacylonitrile (PAN), pitch and rayon followed by a heat treatment. PAN-based carbon fibers generally have high strength, high modulus and low density, and pitch-based carbon fibers tend to have high modulus (Naito 2008), high thermal properties with highly anisotropic microstructure (Kumar 1997). The anisotropic microstructure of the carbon fiber plays an important role in determining the thermal expansion of CFRP and the delamination at the fiber/matrix interface or the laminate interface during temperature. The effect of the thermal expansion due to anisotropic microstructure on the interface strain inhomogeneity is not fully understood because of the difficulty of multi-scale strain measurement. The measurement method on the thermal expansion of CFRPs and carbon fibers has reported in the literatures (Pradere 2008, Kulkarni 2006). However, Thermal strain distribution in the carbon fiber and local strain mismatch at the fiber/matrix interface has not been investigated during temperature. In the present study, we have focused on measurement method on strain distribution inhomogeneity at different scales in CFRP due to thermal loading via in situ field emission scanning electron microscope (FE-SEM) observations.

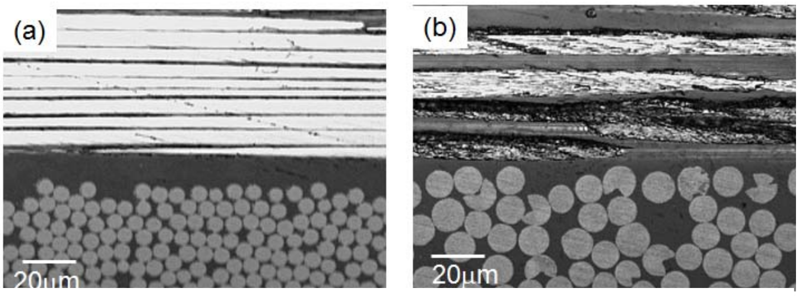

Figure 1 Typical example of microstructure for CFRP, (a) the vicinity of interface between 0 and 90 degree orientation IM600 fibers, (b) the vicinity of interface between 0 and 90 orientation K13D fibers.

2.Experimental procedure

The material used in this study was carbon fiber reinforced polymer composite consist of an ultrahigh strength PAN based IM600 carbon fiber and an ultrahigh modulus pitch based K13D carbon fiber, and epoxy matrix because of their hierarchical internal structures over a wide range of length scales. The laminates were made of seven plies with the 0 and 90 degree orientations by stacking sequence (Figure 1). The fiber volume fraction is approximately 0.6. Inhomogeneous alignment of the both carbon fibers can be seen in the figure 1. The fiber volume fraction is approximately 0.6.

In order to measure the thermal deformation inhomogeneity at different scales, a small rectangular sample with 1 mm thick parallel was cut from the CFRP laminate. To allow direct observation, one side of the sample was polished finally with diamond paste up to 0.25 μm. Before measuring thermal strain, the developed multi-scale pattern consisting of different shape and size (Tanaka 2011) was dawn onto the polished surface of the sample. In the present study, a grid pattern fabricated by using electron

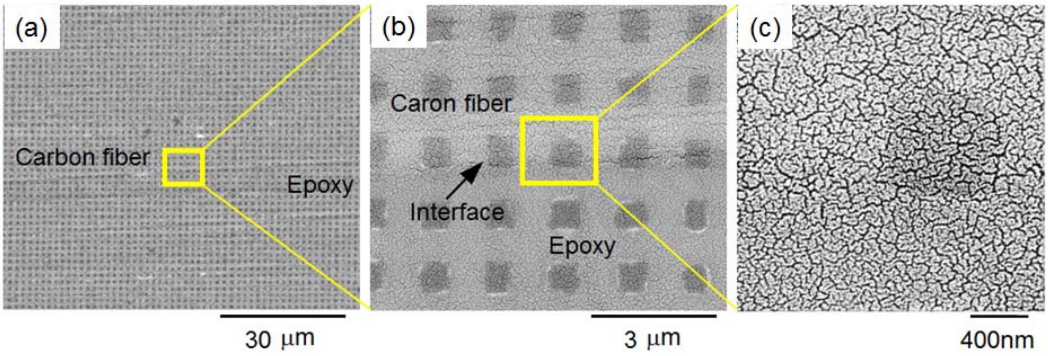

Figure 2 A typical example of multi-scale pattern consisting of (a) grid, (b) the vicinity of fiber/matrix interface at the magnification of the boxed region in (a), and (c) nano random pattern at the magnification of the boxed region in (b).

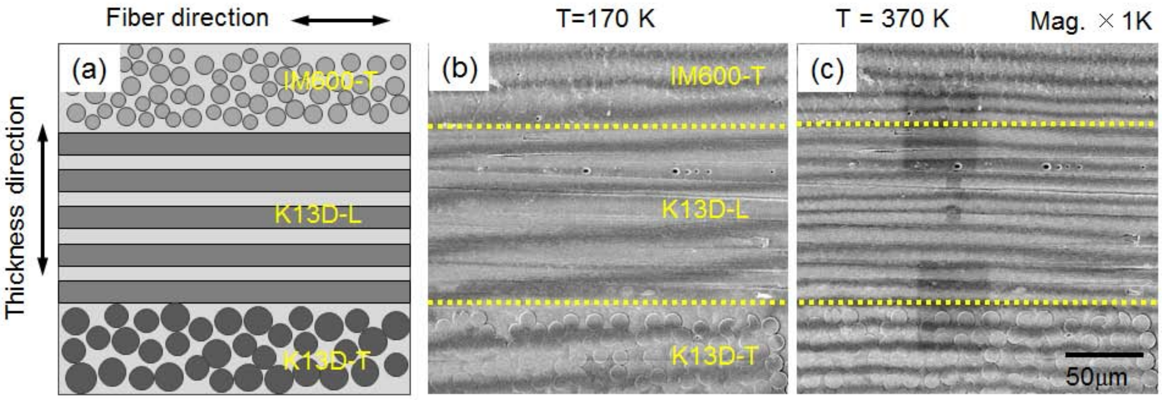

Figure 3 (a) The illustration of the measured area with microstructure, (b) and (c) microscopic thermal deformation behaviors observed by the electron moiré method under different temperatures of 170K and 330K, respectively.

beam lithography (Kishimoto 1991) and random pattern fabricated by spattering were used as multi-scale pattern, as shown in Figure 2. These patterns could be clearly distinguished by backscattered electron imaging (BSEI). The fiber/matrix interface is also distinguished by BSE mode due to reflectance of backscattered electron. An electron moiré method was used to measure the micro deformation in the laminates with different orientation. Digital image correlation method (DIC) was used to measure the nano deformation in carbon fiber and fiber/matrix interface by using random pattern (Figure 2(c)).

A heating/cooling stage using Joule-Thomson effect with high pressure nitrogen gas was installed into the FE-SEM chamber. In order to measure multi-scale deformation and strain inhomogeneity, digital images of 1024 × 884 with 16-bit value intensity were obtained at various step of temperature by using an in-situ FE-SEM observation from macro to nanometer scales. The temperature of the sample was given at a rate of 10K/min. The sample was initially cooled down to the temperature value of 170K and images were taken at 170, 210, 250, 290, 330 and 370K with a hold time 20 min to stabilize the thermal expansion at each temperature. The micro-scale thermal deformation and strain of the laminate was measured using the grid pattern produced in an FE-SEM through the interference between the electron beams (reference grid) and the grid (master grid) by the electron moiré method. It allowed micro-scale thermal deformation measurement by selecting the appropriate real-time scanning lines in the FE-SEM. The two-dimensional digital image correlation using the commercial software VIC-2D was used to analyze the nano-scale thermal deformation and strain distribution such as fiber/matrix interface at different temperatures using the initial and after thermal loading.

3. Results and discussions

Figure 3 shows the microscopic deformation behaviors observed by the electron moiré method under the different temperature of 170 and 330K in the thickness direction. The moiré fringe patterns were clearly generated in the region of the grid pattern at a magnification of 1000. The dotted line indicates the laminate interface between different fiber orientations. The spacing of the moiré fringe lines were decreased with increasing temperature. In particular, the fringe lines in the transverse direction of K13D fibers are locally distorted with increasing the temperature due to inhomogeneous deformation by inhomogeneous fiber distribution. Based on mire theory, the average thermal strain in each laminate was calculated by using fringe spacing at initial and after thermal loading. Figure 4 shows the averaged thermal strain as a function of temperature in each laminate for thickness and fiber directions. The averaged thermal strain is quite different between thickness and fiber directions. The thermal strains in the thickness direction are increased with increasing temperature while the thermal strains in the fiber direction are decreased. The CTE of pitch-based K13D carbon fiber is higher than that at PAN-based IM600 carbon fiber. This behavior is strongly affected by the longitudinal thermal expansion of carbon fiber. The average CTE for K13D-L, K13D-T, IM600-T and fiber direction were determined to be 66.4×10^{-6}, 51.0×10^{-6}, 26.5×10^{-6} and -0.34×10^{-6}, respectively. The results show that the laminate microstructure with different direction affects strongly CTE values.

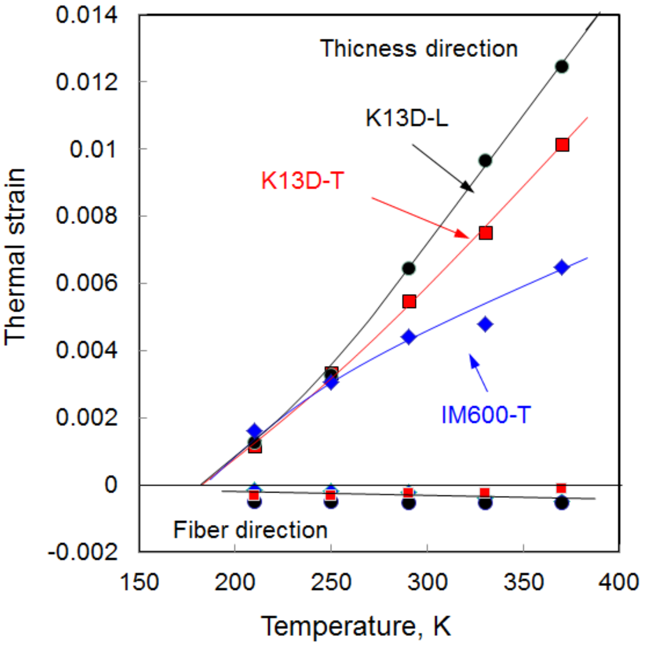

Figure 4 The averaged thermal strain as a function of temperature in each laminates with thickness and fiber directions analyzed by moire theory.

On the other hand, local thermal deformation at nanometer scale around the fiber/matrix interface in the transverse direction of carbon fiber can be measured by the Digital Image Correlation method (DIC) using a nano-random pattern. Figure 5 (a) - (c) show the thermal deformation and shear strain distribution in the cross section of pitch based K13D fiber around fiber/matrix interface at different temperature of 210K and 370K, observed at magnification of 15,000. The white dotted circle indicates the fiber/matrix interface. Local deformation observed in the epoxy matrix near the interface increases with increasing temperature. It is considered that the inhomogeneous deformation arises from the radial CTE of the pitch-based fiber due to the microstructure of the graphite crystals. In particular, the debonding between fiber and matrix is clearly observed at temperature of 370K (Figure 5(c)). The shear strain distribution around the fiber is also shown in figure 5 (a') – (c'). The localized shear strain located at 45 degree appears in carbon fiber near the interface and it increases with increasing temperature. In contrast, the shear strain in the opposite direction from the carbon fiber surface

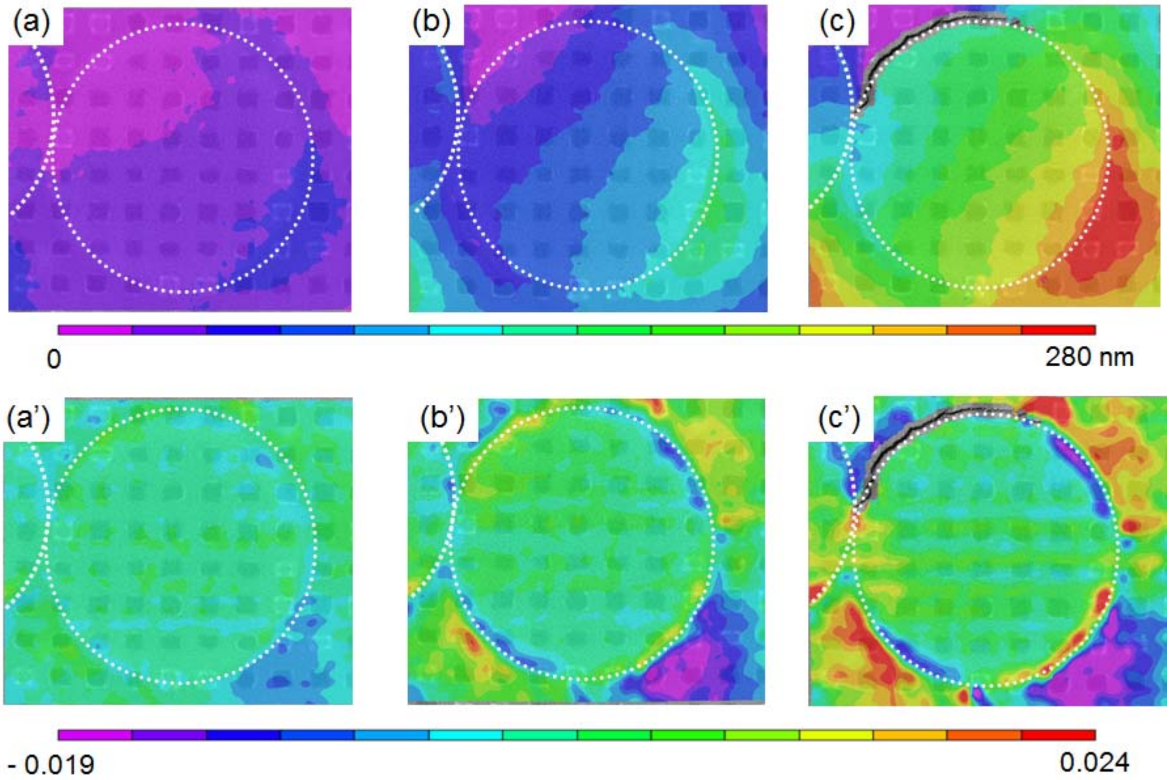

Figure 5 The thermal deformation and shear strain distribution in the cross section of pitch-based K13D fiber around fiber/matrix interface, (a) and (a') T=210K, (b) and (b) T=290K and (c) and (c') T=370K.

appears in the matrix near the interface. This result show that the maximum shear strain occurs at the interface between the carbon fiber and the matrix. It is considered that the debonding initiates from the fiber/matrix interface caused by shear strain. This information is an important role in determining the damage initiation and evolution and the effect of local damage on macro scale thermal deformation behavior.

4. Conclusion

The developed multi-scale pattern is applied to measure thermal deformation and strain distribution in a CFRP by using in situ FE-SEM observations. The present study provides deformation behaviors at different length scales and their related boundary conditions such as interface damage initiation and evolution, and localized thermal deformation gradients needed for developing gradient continuum plasticity at the interface for understanding hybrid CFRP composite materials.

References

Fitzer E. (1989), "PAN-based carbon fibers – present state and trend of the technology from the viewpoint of possibilities and limits to influence and to control the fiber properties by the process parameters", *Carbon,*Vol. 27(5), 621-641.

Kishimoto S., Egashira M., Shinya N. and Carolan R.A. (1991), “Local micro deformation analysis by means of microgrids and electron moire fringe method”, *Proc. 6th Int. Conf. on Mech. Behavior of materials*, Kyoto.

Kulkarni R. and Ochoa O. (2006), “Transverse and longitudinal CTE measurements of carbon fibers and their impact on interfacial residual stress in composites”,*J. Comp. Mat.*, Vol. 40(8), 733-754.

Kumar S., Wang Y. (1997), “Fibers, fabrics, and fillers”, Composites engineering handbook, New York: Dekker.

Naito K., Tanaka Y., Yang J.-M. and Kagawa Y. (2008), “Tensile properties of ultrahigh strength PAN-based ultrahigh modulus pitch-based and high ductility pitch-based carbon fibers”, *Carbon*, Vol. 46(2), 189-195.

Pradere C. and Sauder C. (2008), “Transverse and longitudinal coefficient of thermal expansion of carbon fibers at high temperatures (300-2500K)”, *Carbon*, Vol. 46(14), 1874-1884.

Tanaka Y., Naito K., Kishimoto S. and Kagawa K. (2011), “Development of a pattern to measure multiscale deformation and strain distribution via in situ FE-SEM observations”, *Nanotechnology*, vol. 22, 115704.