1 Introduction

Carbon fibers exhibit excellent engineering properties as a reinforcement in composite materials for the aerospace, automotive, infrastructure and sporting applications [1–2]. 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 polyacrylonitrile (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 [3], high thermal and electrical properties with highly anisotropic microstructure [4]. 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 residual stress or strain due to the microstructure is very complicated. The measurement method of the thermal expansion of CFRPs and carbon fibers has been shown in the literatures using several experimental techniques [5-8]. For example, M. Trinquecostet et al. [8] reported that the longitudinal and transverse thermal expansion coefficients associated with PAN-based carbon fiber was evaluated using an in-situ experimental technique in a wide temperature range. Longitudinal thermal expansion was measured by the sag method [14] applied to single carbon fiber. The transverse thermal expansion was evaluated by

![Figure 1 Microstructure of carbon fiber and CFRP: (a) fracture surface observation of K13D pitch-based carbon fiber, (b) carbon fiber reinforced epoxy matrix composite.](image-url)
measuring the temperature dependence of its apparent diameter in the SEM camber with high magnification. This method had a lower accuracy and a smaller highest temperature due to the thermoelectric effect. R. Kulkarn and O. Ochoa [7] reported that the longitudinal and transverse thermal expansion coefficients associated with PAN-based and pitch-based carbon fibers were evaluated using an in-situ Transmission Electron Microscope (TEM) in the temperature of 300 – 1300K. The images of carbon fibers due to the temperature dependence were directly taken by TEM at the same magnification. The transverse and longitudinal CTE was calculated by dimensional changes of the carbon fiber. However, Thermal strain distribution in the carbon fiber and local strain mismatch at the fiber/matrix interface have not been investigated during temperature.

In the present study, we have focused on measurement method of nano scale deformation and strain distribution around the fiber/matrix interface and the laminate interface in CFRP during temperature via in situ field emission scanning electron microscope (FE-SEM) observations.

2 Experimental Procedure

2.1 Materials

Carbon fiber used in this study was an ultrahigh modulus pitch-based (K13D) carbon fiber. The K13D pitch-based carbon fiber was supplied from Mitsubishi Chemical Functional Products, Inc. The as-received fibers had been subjected to commercial surface treatments and sizing (epoxy compatible sizing). Figure 1 (a) shows a typical example of the transverse section microstructure of the K13D carbon fiber after observing the tensile fracture surface [3] and the transverse section of the CFRP manufactured by conventional prepreg technology. Inhomogeneous alignment of the carbon fibers can be seen in the figure 1 (b). The CFRP is used as a component for the hybrid CFRP material [9]. The fiber volume fraction is approximately 0.6.

2.2 Measurement of thermal deformation

In order to measure the thermal deformation at different scales, a small rectangular block sample with 1 mm thick parallel to the fiber axis was cut from the unidirectional CFRPs, and also cut from the hybrid-CFRP. To allow direct observation, one side
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The sample was polished with diamond paste up to 0.25 µm.

The method on thermal deformation in materials during temperature can be measured using several experimental techniques, such as various full-field non-contact optical methods including interferometric, scanning electron microscope grating and digital image correlation (DIC) methods. The DIC method has been widely accepted and used as a powerful and flexible tool for the surface deformation measurement in the field of experimental solid mechanics. The DIC method also has some advantages, compare with the interferometric optical method, such as simple experimental set up and specimen preparation and wide range of spatial resolution. However, the object specimen surface must have a random pattern with high quality image to realize nano-scale deformation measurement. The spattering technique was used to introduce nano-random pattern onto the sample surface [10].

In order to measure the micro deformation, an electron moiré method has developed using a micro grid pattern fabricated onto the specimen surface [11]. A grid pattern was fabricated by using electron beam lithography [12]. An electron-sensitive resist (Nippon Zeon ZEP-520-22) was coated onto the specimen surface using a spin coater at a speed of 2,000 rpm for 120 s and then baked in an oven for 4 hour at 50 °C. We then mounted the specimen on the specimen stage in an FE-SEM (Quanta 200 FEG, FEI Corp.) for electron beam exposure using the beam blanking device. After the electron beam exposure, the specimen was developed in a solution of ZED-N50 (Nippon Zeon) for 10 s and then immediately rinsed in ZMD-B (Nippon Zeon) for 20 s. The specimen was coated with a very thin layer (10–20 nm) of gold by sputtering. After removing the resist by using an organic solvent, a model grid formed on the specimen surface. The real-time scanning lines in the FE-SEM acted as a reference grating, which was superimposed onto the model grid to produce a moiré pattern. A very thin polymer resist (EBR9, Toray Corp.) was coated onto the specimen surface after finishing the grid preparation, and gold was then coated onto the entire region of the surface using an ion coater (IB-5, EIKO Engineering Co., Ltd.). This gold coating was necessary to provide a conductive surface that prevented the accumulation of electron charge while viewing the specimen in the FE-SEM in the high vacuum mode. Figure 2 shows a typical example of a pattern consisting of a grid pattern with 500 nm spacing and a nano-cluster pattern ranging from 20 to 100 nm. 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.

A heating/cooling stage of Joule-Thomson refrigerator using nitrogen [13] gas was installed into the FE-SEM chamber, as shown in Figure 3. In
order to measure multiscale 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 from macro to nanometer scales.

The temperature of the sample was given at a rate of 10K/min. The sample was cooled down to the initial 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 macroscale thermal deformation of the specimen 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 macroscale 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 localized thermal deformation and strain distribution at different temperatures using the initial and after thermal loading.

3 Results and discussions

Figure 4 shows the macroscopic deformation observed by the electron moiré method under the different temperature of 170, 250, and 330K for the transverse of CFRP. The moiré fringe patterns were clearly generated in the region of the grid pattern at a magnification of 249. The spacing of the moiré fringe lines was decreased with increasing temperature. Based on moire theory, the average thermal strain, \( \varepsilon \), can be calculated using fringe spacing at initial and after thermal loading, as following.

\[
\frac{\varepsilon}{1 + \varepsilon} = \frac{N(1/\delta_2 - 1/\delta_1)}{\rho_f}
\]
Where, \( N \) and \( P_f \) are the number of electron beam lines and the number of grid lines in the measured area, respectively. The \( \delta_0 \) and the \( \delta_1 \) are the moiré fringe spacing at initial and after temperatures, respectively. The average CTE of the transverse CFRP was determined to be \( 5.18 \times 10^{-5} \).

Local thermal deformation at nanometer scale around the fiber/matrix interface in the direction of longitudinal and transverse can be measured by the DIC method using a nano-cluster pattern.

Figure 5 shows the relative thermal deformation and strain distribution in the perpendicular to the fiber direction, \( y \), at different temperature range of 40K, 120K and 200K analyzed by Digital Image Correlation method, observed at magnification of 30,000. The white dotted line indicates the fiber/matrix interface. In the \( y \) component, even the temperature range of 40 the localized deformation is clearly observed in the epoxy matrix near the interface, and it is increased with increasing the temperature range. It is clear that the relative deformation value in the \( x \) (103 nm) is approximately 2 times larger than that at the value of \( y \) (191 nm). The inhomogeneous deformation in the fiber has a tendency to be larger with distance from the interface with increasing temperature range. This anisotropic and inhomogeneous deformation behavior is caused by the differences of the coefficient of thermal expansion in the transverse and longitudinal directions due to texture microstructure of pitch-based carbon fiber [7]. The thermal strain distribution shows that the localized negative strain is clearly observed and increased in the epoxy matrix near the interface with increasing temperature range. The localized positive strain is observed in the carbon fiber, it is increased with increasing temperature range. The strain value near the fiber surface seems to be constant with increasing temperature range.

Figure 6 shows the average thermal strain distribution in the fiber direction, \( x \), and perpendicular to the fiber direction, \( y \), at different temperature ranges. The average strain in the \( x \) component of carbon fiber is decreased with increasing the temperature range, it has the negative
CTE to the fiber direction. On the other hand, the average strain in y component of the fiber is increased with increasing the temperature range, it has positive CTE. The localized compressive strain is formed in the matrix near the fiber with increasing temperature range. So, the localized thermal strain behavior is significant for determining the interface delamination. The compressive strain of approximately 0.08, for example, is locally formed in the epoxy matrix near the interface despite the positive CTE of the epoxy. In the operating temperature, the transverse strain is linearly increased, while the longitudinal strain is slightly decreased with increasing temperature.

Figure 7 shows FE-SEM photographs of the transverse cross-sectional fracture surface in the mesophase pitch-based carbon fiber K13D. The fiber showed a radial texture in the overall transverse surface at low magnification (Figure 7 (a)). The core area about 2 µm from the fiber center showed a random texture (Figure 7(c)). Most domains were of looped and bent shape shown independent orientation from that of the neighboring domain. In the outer area, the constituent domains showed a well developed linear shape, being aligned in the radial orientation. No looped or bent shaped domains were found in the outer area. The length of domain in the outer area was from 600 nm to 1000 nm (Figure 7(b)).

Figure 8 shows the relative thermal deformation in the radial direction at different temperature range of 40K and 120K analyzed by Digital Image Correlation method, observed at magnification of 15,000. The white dotted line also indicates the fiber/matrix interface. Inhomogeneous thermal deformation is found at the interface. The inhomogeneous deformation arises from the radial CTE of the fiber and the matrix. That is strongly affected by the fiber distribution in the matrix.
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The radial and longitudinal strains in the carbon fiber versus loading temperature calculated from the deformation data are shown in figure 9. Each value is averaged in the fiber direction at each temperature. In the operating temperature, the radial strain is almost linearly increased, while the longitudinal strain is slightly decreased with increasing temperature. The slope of strain versus temperature is considered to be an approximate estimate of CTE of the carbon fiber. The radial and longitudinal CTEs are determined to be $4.16 \times 10^{-5}$ and $-2.35 \times 10^{-6}$, respectively. The radial CTE of carbon fiber is higher than that CTE of pitch-based carbon fiber compared with literature [7]. On the other hand, the radial CTE of the fiber is slightly small compare with macroscopic CTE obtained from electron moiré method.

This suggests that the nano scale deformation and strain distribution in the small area will affect the CTE due to the anisotropy and inhomogeneous microstructure in the carbon fiber. It is considered the transverse CTE after polishing is different with as-received carbon fiber. The present study can be used to provide higher-order of thermal expansion behaviors, such as thermal expansion inhomogeneity and anisotropy, interface delamination, deformation gradients needed for developing the thermal damage mechanism understanding of the CFRP.

4 Conclusions

The developed nano-sclae pattern is applied to measure in-plane thermal deformation and strain distribution in a carbon fiber-reinforced composite 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


