Thermal and volumetric property analysis of polymer networks and composites using elevated temperature digital image correlation

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Abstract:

Digital Image Correlation (DIC), which exploits non-contact advantages and full-field analysis, provides more data in-situ that are not possible with traditional techniques. In this work, elevated temperature digital image correlation techniques were applied to a glassy polymer network via thermal expansion and contraction experiments to study volumetric behavior during the curing process. A glassy epoxy network was tested in both cured and under-cured states and heated to the ultimate cure temperature. Matrix volume changes due to both thermal expansion and cure shrinkage were quantified. Concurrently, the thermal expansion of aerospace-grade composite laminates was also observed in matrix and fiber-dominant directions. Additionally, the strain-free temperature of a non-symmetric composite laminate was identified through thermal compensation of process-induced curvatures. Finally, laminate dimension changes were related to the strain-free temperature as means to probe process-induced strains within composite laminates. Thermal properties of the neat matrix and composite laminates were compared to traditional techniques, validating the benefit of elevated temperature digital image correlation for composite matrix qualification.

Keywords:

Digital image correlation

Polymer-matrix composites (PMCs)

Residual strain

Thermal expansion

Elevated temperature testing

1. Introduction

The rising exploitation of polymer composites for use in primary and secondary structures of aerospace, automotive and marine applications is significant and requires further exploration into manufacturing and design parameters. For the proliferation of composite materials, property simulation of new chemistries has become vital to the aerospace community. In this, understanding property development of composites during the manufacturing stage is essential for simulation. The development of thermal, mechanical and rheological properties of thermoset matrices through the multiple stages of manufacturing is complex, and a holistic understanding of key properties is critical for model development and accuracy. While significant effort has been put into the testing, modeling and prediction of property development, there is a continuous demand for test improvement for enhanced material performance analysis. As simulation of composite materials continues to multiply across all size scales, it becomes critical to understand, at a fundamental level, the developing properties and physical changes that take place within the matrix during cure.

Molecular-level strains are introduced within the matrix due to volume changes during the manufacturing of continuous fiber composite laminates, and lead to a number of failure modes including geometry deformation, matrix micro-cracking and ply delamination, among others.[1–3] The sources of these strains are complex, ranging from material properties to processing factors such as tooling material or autoclave thermal gradients; however, a clear consensus exists that dominant contributing factors include a differential in the coefficients of thermal expansion (CTE) between the matrix and reinforcement and, to a lesser extent, matrix chemical shrinkage. Understanding the magnitude of these contributing factors, as well as any potential areas for control, is critical for material and process qualification.

The basics of strain creation with a composite material involve constraint placed on volume changes of the matrix by the fiber environment. The community agrees that a strain cannot build until the matrix has developed an appreciable mechanical strength; a number of authors have shown that, in the processing of thermoset networks, strain creation begins on gelation.[1,4] In this rubbery state, however, strain relaxation is significant, and molecular segments are able to rearrange to dissipate energy.[5] As molecular weight continues to build throughout cure, the polymer network's ability to realize these cooperative motions decreases until the point of vitrification, at which point continued reaction slows drastically and a number of network properties significantly change. For example, vitrification marks measurable increases in Young's, shear and bulk moduli, and increases in moduli correspond with increases in relaxation times.[6] For this reason, it is assumed that strain relaxation ceases upon vitrification and this corresponding temperature is often denoted as the strain-free temperature.[7–9] Due to high values of moduli upon vitrification, additional reaction and bond formation imparts strain as the matrix continues to cure, and may also increase the strain-free temperature.[4] From this glassy state, the material still must undergo cooling from the cure temperature to room temperature. This corresponds with a significant change in matrix volume inducing sizeable thermal residual strains, which are considered to be the largest contributors to process-induced strain within composite materials.[10]

Due to the multifaceted mechanisms of strain creation, the measurement of residual strains is complex. While tools such as classical laminate theory allow for the calculation of residual strains based on constituent material properties, techniques which directly measure the strains are not common. Unlike more established polymer analysis techniques, there is a lack of consensus within the community to a preferred method to measure or represent the magnitude of

strains. Strain measurement devices such as Fiber Bragg grating sensors and strain electric gauges raise question as to at which point the measurement becomes valid during the cure due to the developing interface between the part and the sensor.[10] Conversely, techniques which measure laminate dimension changes or developing mechanical strength of uncured, impregnated fibers, such as dynamic mechanical or thermomechanical analysis, do not represent the environment present in the manufacturing of high performance composite laminates.[4,11] Because of the implications of in-situ techniques such as these, deformation of laminates from initial, pre-cured to final cured states continues to be a common method for researchers. In this, process-induced strains are examined through either curvature reduction of an angled part or by the introduction of curvature upon curing a flat laminate of a non-symmetric laminate sequence. In each scenario, it is understood that the magnitude of residual strain relates to the deviation of mold geometry. In the case of an angled tool, a decrease in interior angle, also called “spring-in,” corresponds to an increase in residual strain within the laminate. Conversely, the creation of curvature upon the curing of a flat, non-symmetric laminate is also related to the magnitude of residual strain, wherein increased curvature corresponds to increased residual strain. These techniques are based on measurements made before and after laminate curing. This limits the knowledge of process-induced strain development to an initial (prior to curing) and final (after curing) conditions. We believe heating cured laminates to elevated temperatures provides additional insight into the residual strain state, and offers an additional response to laminate deformation. Further, we show that a single experimental setup can be used for accurate measurement of critical matrix properties which serve as the contributing factors to process-induced strains within continuous fiber composite laminates.

2. Experimental

2.1 Materials

CYCOM 977-3 prepolymer matrix and prepreg was supplied by Cytec Aerospace Materials and used as received.

2.2 Glassy Polymer Network Preparation

The prepolymer matrix was heated and degassed at 125 °C (10^{-3} Torr) and cast into silicone molds (internal cavity 130 x 12 x 6 mm). It was then cured by the manufacturer's recommended cure cycle (MRCC) (temperature ramp at 1.8 °C/min to 177 °C, 6h isothermal hold). For chemical shrinkage studies, samples were removed from the oven after a 30 min isothermal hold at 177 °C and quickly cooled to room temperature. Samples approximately 15 x 12 x 6 mm were manually prepared from the cast 130 x 12 x 6mm bars.

2.3 Composite Laminate Preparation

Prepreg materials were stored in vacuum-sealed containers at sub-ambient temperatures, and equilibrated at room temperature prior to use. Flat composite panels (30.48 x 30.48 cm) were fabricated into unidirectional (UD) $[0_8]$, off-axis (OA) $[0_4/45_4]$, and non-symmetric (NS) $[0_8/90_8]$ laminates on flat Invar tools with steel caul plates. Panels were cured by the manufacturer's recommended cure cycle (1.8 °C/min to 177 °C, 6h hold) in an American Autoclave under vacuum bag and pressure ($\sim 10^{-3}$ Torr and 0.689 MPa, respectively) to fully compact plies and assure void-free laminates. Panels were machined into 2.54 x 7.62 cm coupons using a diamond saw fitted with cutting fluid. Due to non-traditional lay-up sequences, some coupons developed cracks during machining and were not used. Non-symmetric $[0_8/90_8]$ and off-axis $[0_4/45_4]$

laminates exhibited significant concave curvature with respect to the 90° and 45° faces, respectively.

2.3 Thermal Characterization

Parallel plate rheological experiments were conducted to determine the gelation point of the prepolymer matrix through the MRCC on a Thermal Analysis (TA) ARES-G2 rotational rheometer using 8mm aluminum plates with a 0.3 mm gap and frequency of 1 Hz. The development of the shear storage (G') and loss (G'') moduli were observed, with the $G'=G''$ crossover used for the identification of gelation, which occurred at 30min into the isotherm at 177 °C.

The volumetric properties of the epoxy networks were also determined from thermomechanical analysis (TMA) in comparison to results from elevated temperature DIC. Cured matrix cubes approximately 10 x 10 x 10 mm were manually cut and polished. The experiments were conducted using a Thermal Analysis (TA) Q400[®] instrument fitted with a quartz stage and macro expansion probe. Specimens were heated at 1 °C/min from 25 °C to 177 °C using a 0.05 N force. Linear coefficients of thermal expansion were determined by the slope of a linear fit to sample length vs. temperature curves from 25-50 °C. Laminate measurements were taken parallel and perpendicular to the fiber direction (0° and 90°, respectively) using 1 x 1 cm UD [0₈] specimens.

2.4 Elevated Temperature Digital Image Correlation

Digital image correlation (DIC) measures specimen deformation by tracking random surface patterns in gray-scale image sets from the un-deformed to deformed states, allowing for non-contact full-field analysis of surface strain.[12–14] DIC strain analysis was conducted using

a GOM Optical Measuring Techniques ARAMIS 3D Deformation Analysis System (Trilion Quality Systems). Specimens were speckle coated to create a non-uniform surface pattern using high-temperature white and black aerosol paints with a black solid basecoat and white speckles in the appropriate size range of 2-5 pixels.[15] In the case of composite laminates, a basecoat was not necessary. Digital gray scale images were captured via two 2M digital CCD cameras equipped with 50mm lenses. In this setup, the DIC cameras were placed squarely and level in front of a Memmert UF55 Plus laboratory oven fitted with a glass window. Placement of the cameras was chosen such that the camera span, camera angle, measuring distance, and viewing angle were appropriate for elevated temperature experiments. A camera angle of 11.8° was chosen for optimized lighting and minimal glare through the laboratory oven window. Camera calibration was performed using a 25mm x 20mm standard placed within the oven (at ambient temperature) to obtain a resolution deviation of less than 0.03 pixels. Images were taken through the glass door throughout the calibration procedure. To improve lighting conditions, the lens aperture was opened to the maximum, and shutter speed was increased to approximately 120 ms. A black backdrop was placed behind the cameras to reduce glare.

During the heating event, images were taken at a frame rate of 1 image per minute to align with the oven temperature ramp rate of $1^\circ\text{C}/\text{min}$. To increase accuracy of surface strain maps between ambient and elevated temperature, several steps were taken. These included the definition of analysis areas, assignments of start points, and masking of improperly focused/illuminated areas. For more details regarding DIC techniques please refer to our previous works on strain development and recovery in epoxy networks.[16,17]

Elevated temperature digital image correlation has found application in a number of research topics including high temperature deformations,[18] thermally activated shape memory

polymers,[19] calculations of Poisson's ratio for soft materials,[20] and the coefficient of thermal expansion for thin films.[21] Using elevated temperature digital image correlation, the expansion behavior of cured matrix and laminates can be accurately measured for key insights into their volumetric properties. Further, as laminates are heated to elevated temperature, process-induced residual strains are relieved, and this response can be captured via DIC. The expansion behavior, as well as reduction of laminate deformation, each relate to thermal residual strains and are critical for material and process qualification. The temperature at which all residual strains have been relieved is known as the strain-free temperature, which directly relates to the magnitude of process-induced strains and is also used in composite failure models.[22] In this work, we propose differences in thermal expansion and deformation between various lay-up sequences as means to probe the internal strain state of composite laminates.

3. Results and Discussion

3.1 Chemical Shrinkage

A unique benefit to DIC techniques is the ability of the software to calculate numerous responses from a single series of images. While DIC is typically used as a full-field strain measurement technique, the ARAMIS software also provides analytical reference tools, known as “primitives” related to the computed surface maps, including points, lines, planes, best-fit shapes, etc. These tools can be used to track dimension changes from image to image. Line lengths calculated along the surface map can be used for analysis of sample expansion or contraction throughout the image series. In this work, five lines whose lengths were nearly equal to the sample's height (approximately 13.6 mm) were created. Maximum line lengths were found to reduce the noise associated with the measurement. Length changes were then tracked from the

first image prior to heating (L_0) through the MRCC. In this case, a maximum heating rate under the MRCC (2.8 °C/min) was chosen to limit the convolution of chemical shrinkage with thermal expansion. The average change in line length is plotted in Figure 1. Initially the partially-cured sample undergoes thermal expansion. As the cure temperature is approached, the curve deviates from linearity and begins to decrease. Once the sample has equilibrated near the cure temperature (sample temperature approximately 175.5 °C), all changes in line length are assumed to be associated with continued crosslinking, or chemical shrinkage. The $\Delta L/L_0$ curve shows a negative slope until nearly the end of the 6h isothermal hold, at which point it approaches a plateau as the cure approaches completion. From this study, we measured chemical shrinkage beyond gelation to be 0.8%.

[INSERT FIGURE 1]

3.2 Thermal Expansion of Matrix and UD Fiber-Reinforced Composites

Cooling the matrix within the glassy state, from the cure temperature to room temperature, is often cited as the largest contributing factor in strain creation in continuous fiber composite laminates.[4,10] Measurement of the glassy state CTE relevant within the processing temperature window is desirable to simulate the cooling process. Through line length measurements on the surface of glassy polymer network during heating, elevated temperature DIC lends itself to a direct comparison to the linear coefficient of thermal expansion as measured by TMA. Figure 2 shows the average dimension change ($\Delta L/L_0$) of five vertical lines in heating to the cure temperature, 177 °C. The length of each line spans the height of the strain map on a cured sample (approximately 13.6mm in length). As with chemical shrinkage measurements, increasing the length of the analysis tool increases the signal to noise ratio. Calculated linear

coefficients of thermal expansion by DIC and TMA from 25-50 °C are shown in Table 1. The relationship between change in line length and temperature throughout this temperature window shows a clear increase, with a “knee” at 118.5 °C by bilinear fit, suggesting a glassy-state expansion event.

[INSERT FIGURE 2]

[INSERT TABLE 1]

Similar procedures for thermal expansion of neat matrix were applied to a UD composite laminate to probe dimension changes due to the presence of the fiber. A UD laminate coupon was clamped and horizontally suspended, in this case above a NS laminate (method discussed in Section 3.3). Five lines were placed parallel or orthogonal to the fiber orientation of the observed laminate surface, each spanning a length nearly equal to the width of the coupon (approximately 25 mm). Laminate orientation and reference line placement (parallel and orthogonal) are shown in Figure 3. Dimension changes parallel and orthogonal to the fiber orientation allowed for calculation of the effective glassy state CTE in these two directions. CTE values taken from 50-100 °C are reported in Table 1 with comparison to measurements obtained via TMA. Along the fiber orientation, volumetric properties are fiber-dominant and lead to thermal contraction, as denoted by a negative CTE observed in both techniques. Conversely, thermal expansion perpendicular to the fiber orientation is matrix dominant and the glassy state CTE is significantly larger. The presence of the fibers, however, significantly decreases the observed CTE as compared to the neat matrix. Decreasing the macroscopic CTE of a polymer through incorporation of low CTE reinforcements is well established.[23–25]

[INSERT FIGURE 3]

3.3 Laminate strain-free temperature

Of the laminate sequences fabricated, NS laminates have the most significant difference in fiber orientation, leading to significant interlaminar strain. Some of this strain is relieved through geometry deformation, and large aspect ratio coupons have significant concave curvature towards the 90° surface. The residual strains causing curvature may be relieved upon heating. Elevated temperature DIC can be used to accurately track curvature compensation during heating to identify the strain-free temperature. The NS laminate was constructed with 16 plies (as opposed to eight), allowing for a thicker edge and a larger surface map. The coupon was anchored at one end with a clamp, fitted with a thermocouple, and the oven was heated at 1 °C/min to 250 °C (see Figure 3, bottom). In addition to primitives, the ARAMIS software also allows users to create reference planes and monitor point-to-plane distances. Point-to-plane distances can be tracked through the image series using stage points to quantify laminate curvature changes with respect to the vertical axis during the heating event. A representation of stage point placement is shown in Figure 3, while distance change vs. temperature for the stage points is shown in Figure 4. A clear plateau is observed for all stage points regardless of proximity to the anchoring clamp. From these curves, the laminate reached maximum curvature compensation at a laminate surface temperature of 199 °C. On cooling, the laminate redevelops curvature as thermal strains build, and at room temperature displays increased curvature due to exposure to temperatures above the ultimate cure temperature.

[INSERT FIGURE 4]

3.4 Laminate Curvature Development

As the temperature deviates from the strain-free temperature, the matrix and fiber undergo significantly different volume changes, as previously observed in UD laminates, leading to non-uniform contraction. This non-uniform dimension change, which leads to curvature development in laminates which lack symmetry about the mid-plane, can be captured using elevated temperature DIC. If we consider the strain-free temperature to be the temperature at which all process-induced strains within the composite laminate have been relaxed, it is logical to regard dimensions at this temperature as the reference lengths, or L_0 . In this case, we re-heat the laminate to the strain-free temperature to compensate the residual strains induced during the cooling stage, with the assumption that thermal expansion upon re-heating produces a similar response to thermal contraction upon cooling. This allows for a simulation of the cooling stage in laminate processing, during which a significant proportion of thermal residual strains are introduced.

The effective laminate thermal properties such as CTE are combinatorial between the matrix and fiber properties, and determined by laminate ply sequence. Traditional contact-based techniques such as TMA are capable of one-dimensional measurements. Therefore, these measurements are an average response from all plies within the laminate. Non-contact techniques, however, are capable of simultaneous multidirectional measurements on a surface. In the case of a composite laminate, these measurements probe the local environment of the topmost ply, thus more accurately representing the interactions between the matrix and fibers. Elevated temperature DIC is uniquely positioned to quantify differences of the free (matrix-dominant) and hindered (fiber-dominant) thermal expansion/contraction directions on the surface of a composite laminate. Expansion/contraction anisotropy of different laminate sequences

represents the variability of a composite due to the matrix and fiber constituent properties and deformation from process-induced strains. Composite anisotropy is represented in Figure 5, which shows the average dimension change parallel and orthogonal to the fiber orientation on the observed laminate surface. The fiber orientation on the observed surface of all laminates was 0° . Dimension changes along the surface map with respect to dimensions at the strain-free temperature were chosen to simulate the glassy state cooling process. As the temperature differential from the strain-free temperature increases, dimension changes parallel and orthogonal to the fiber axis are measurably different for the laminate sequences tested. This relationship is unique and demonstrates the benefits of elevated temperature DIC. Listed in Table 2 are the glassy-state CTE measurements as taken by DIC, in with comparison to TMA measurements. In both the OA and NS laminates, the parallel and orthogonal CTE values are decreased in comparison to the UD laminate values in Table 1. Within the TMA, the OA laminate was oriented such that the measurement was taken in the direction of the 0° fibers. Measurements by TMA are significantly different from either directional measurement taken by DIC due to being an average dimension change of all plies in the laminate.

[INSERT FIGURE 5]

[INSERT TABLE 2]

The UD laminate does not contain off-axis lamina, and thus the expansion/contraction properties of the observed surface are highly anisotropic with respect to the fiber orientation. Length changes parallel to the fiber orientation are restricted throughout the temperature window tested. Orthogonal to the fiber orientation, however, exhibits significant increase/decrease in line length with increasing/decreasing temperature, indicating matrix-dominant thermal

expansion/contraction. The OA and NS laminates exhibit a similar difference in parallel vs. orthogonal directions; however, the expansion/contraction behavior of their observed surfaces is also affected by the nearby off-axis plies. The angular difference in fiber orientation of the NS laminate is 90° , thus the relative matrix and fiber-dominant directions are orthogonal about the mid-plane of the laminate. Therefore, expansion/contraction of the observed surface in the matrix-dominant direction is resisted by nearby layers whose expansion/contraction properties are fiber-dominant in this direction. This is apparent by the smaller slope of the NS laminate orthogonal to the fiber direction compared to the UD laminate. Due to a smaller angular difference in the OA laminate, this constraint is less prominent, which corresponds with increased dimension change in the orthogonal direction as compared to the NS laminate. However, both the OA and NS laminates have plies which resist free expansion of the observed surface, decreasing the effective CTE in the matrix-dominant direction in comparison to the UD laminate.

In Figure 5, it should be noted that the OA and NS laminates both show non-zero slopes parallel to the fiber orientation, while the curve of the UD laminate has effectively zero slope. On cooling below the strain-free temperature, residual strains build and cause curvature to develop within laminates which do not have symmetry about the mid-plane. By this method, changes in line length parallel to the fiber axis represent curvature development. The UD laminate has symmetry about its mid-plane, thus no curvature changes contribute to the $\Delta L/L_0$ calculations. This corresponds with large thermal expansion/contraction anisotropy relative to the fiber direction. To show this, we compare differences in $\Delta L/L_0$ values for the parallel and orthogonal directions in Figure 6. Differences in slopes represent the magnitude of curvature development during glassy-state cooling below the strain-free temperature. This demonstrates how varied fiber

orientations affect laminate expansion/contraction properties upon heating/cooling. Direct measurement of laminate thermal properties is a unique capability of elevated temperature DIC, and can complement thermal strain calculations based on classical laminate theory.

[INSERT FIGURE 6]

4. Conclusions

The objective of this research was to develop elevated temperature digital image correlation techniques to study the volumetric behavior of glassy polymer networks and composite laminates during thermal expansion/contraction events. A holistic understanding of a composite material's constituent properties is necessary for the proliferation of composites manufacturing and use. Process-induced strains are a challenge to the composites community, yet there persists a lack of standard testing methods. Traditional laminate deformation techniques, which only examine laminate geometry before and after cure, often exhibit unexpected or noisy data. We propose using dimension changes throughout a heating event, such as expansion differences and curvature compensation, as means to probe the residual strain state of composite laminates. Judicious selection of laminate sequences can be used to exhibit differences in matrix and fiber volumetric properties, thus giving insight to their compatibility with respect to process-induced strains. Through the use of software reference tools, we demonstrate elevated temperature DIC can be used for accurate identification of the strain-free temperature, which is a critical property for simulation of composite processing and performance. Dimensional measurements can be taken for multiple directions simultaneously and, in the case of a composite laminate, are representative of a local environment on the laminate surface rather than a combined laminate average, as is the case in traditional techniques. This gives the researcher increased insight into the compatibility of the volumetric properties of

the constituent components. Ultimately we found this technique to be equally advantageous and simple to use as traditional DIC methods, requiring only extra consideration for system calibration and lighting.

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References

- [1] M. Merzlyakov, G.B. McKenna, S.L. Simon, Cure-induced and thermal stresses in a constrained epoxy resin, *Compos. Part A Appl. Sci. Manuf.* 37 (2006) 585–591.
- [2] J.F. Timmerman, M.S. Tillman, B.S. Hayes, J.C. Seferis, Matrix and Fiber influences on the cryogenic microcracking of carbon fiber/epoxy composites, *Compos. Part A Appl. Sci. Manuf.* 33 (2002) 323–329.
- [3] T. Garstka, N. Ersoy, K.D. Potter, M.R. Wisnom, In situ measurements of through-the-thickness strains during processing of AS4/8552 composite, *Compos. Part A Appl. Sci. Manuf.* 38 (2007) 2517–2526.
- [4] P.A. Olivier, A note upon the development of residual curing strains in carbon/epoxy laminates. Study by thermomechanical analysis, *Compos. Part A Appl. Sci. Manuf.* 37 (2006) 602–616.
- [5] J. Lange, S. Toll, J.-A.E. Månson, Residual stress build-up in thermoset films cured above their ultimate glass transition temperature, *Polymer (Guildf)*. 36 (1995) 3135–3141.
- [6] J.K. Gillham, Formation and Properties of Thermosetting and High Tg Polymeric Materials, *Polym. Eng. Sci.* 26 (1986) 1429–1433.
- [7] P. Prasatya, G.B. McKenna, S.L. Simon, A Viscoelastic Model for Predicting Isotropic Residual Stresses in Thermosetting Materials: Effects of Processing Parameters, *J. Compos. Mater.* 35 (2001) 826–848.
- [8] N. Ersoy, T. Garstka, K. Potter, M.R. Wisnom, D. Porter, M. Clegg, G. Stringer, Development of the properties of a carbon fibre reinforced thermosetting composite through cure, *Compos. Part A Appl. Sci. Manuf.* 41 (2010) 401–409.
- [9] P.P. Parlevliet, H.E.N. Bersee, A. Beukers, Measurement of (post-)curing strain development with fibre Bragg gratings, *Polym. Test.* 29 (2010) 291–301.
- [10] M. Mulle, F. Collombet, P. Olivier, Y.-H. Grunevald, Assessment of cure residual strains through the thickness of carbon–epoxy laminates using FBGs, Part I: Elementary specimen, *Compos. Part A Appl. Sci. Manuf.* 40 (2009) 94–104.
- [11] H. Yu, S.G. Mhaisalkar, E.H. Wong, Direct Measurement of Cure-Induced Stress in Thermosetting Materials by Means of a Dynamic Mechanical Analyzer, *Macromol. Rapid Commun.* 27 (2006) 1393–1397.
- [12] T.C. Chu, W.F. Ranson, M.A. Sutton, Applications of digital-image-correlation techniques to experimental mechanics, *Exp. Mech.* 25 (1985) 232–244.
- [13] H.A. Bruck, S.R. McNeill, M.A. Sutton, W.H. Peters, Digital image correlation using Newton-Raphson method of partial differential correction, *Exp. Mech.* 29 (1989) 261–267.
- [14] Peters, WH, W. Ranson, Digital imaging techniques in experimental stress analysis, *Opt. Eng.* 21 (1982) 427–431.
- [15] GOM, ARAMIS User Manual, (2007).
- [16] S.R. Heinz, J.S. Wiggins, Uniaxial compression analysis of glassy polymer networks using digital image correlation, *Polym. Test.* 29 (2010) 925–932.

- [17] S. Heinz, J. Tu, M. Jackson, J. Wiggins, Digital image correlation analysis of strain recovery in glassy polymer network isomers, *Polymer (Guildf)*. 82 (2016) 87–92.
- [18] J.S. Lyons, J. Liu, M.A. Sutton, High-temperature deformation measurements using digital-image correlation, *Exp. Mech.* 36 (1996) 64–70.
- [19] A.J.W. McClung, G.P. Tandon, K.E. Goecke, J.W. Baur, Non-contact technique for characterizing full-field surface deformation of shape memory polymers at elevated and room temperatures, *Polym. Test.* 30 (2011) 140–149.
- [20] M. Jerabek, Z. Major, R.W. Lang, Strain determination of polymeric materials using digital image correlation, *Polym. Test.* 29 (2010) 407–416.
- [21] P. Bing, X. Hui-min, H. Tao, A. Asundi, Measurement of coefficient of thermal expansion of films using digital image correlation method, *Polym. Test.* 28 (2009) 75–83.
- [22] J. Gosse, S. Christensen, Strain Invariant Failure Criteria for Polymers in Composite Materials, in: 42nd AIAA/ASME/ASCE/AHSIASC Structures, Struct. Dyn. Mater. Conf. Exhib., American Institute of Aeronautics and Astronautics, Seattle, WA, 2001.
- [23] S. Sulaiman, C.M. Brick, C.M. De Sana, J.M. Katzenstein, R.M. Laine, R.A. Basheer, Tailoring the Global Properties of Nanocomposites. Epoxy Resins with Very Low Coefficients of Thermal Expansion, *Macromol. Rapid Commun.* 39 (2006) 5167–5169.
- [24] S. Wang, Z. Liang, P. Gonnet, Y.H. Liao, B. Wang, C. Zhang, Effect of nanotube functionalization on the coefficient of thermal expansion of nanocomposites, *Adv. Funct. Mater.* 17 (2007) 87–92.
- [25] M.M. Shokrieh, A. Daneshvar, S. Akbari, M. Chitsazzadeh, The use of carbon nanofibers for thermal residual stress reduction in carbon fiber/epoxy laminated composites, *Carbon N. Y.* 59 (2013) 255–263.

Figure Captions:

Figure 1: Average change in linear dimension due to heating and continued curing.

Figure 2: Average dimension change due to thermal expansion of cured matrix.

Figure 3: Laminate placement for thermal expansion and strain-free temperature studies, with representative reference lines and stage points.

Figure 1: Distance changes for several points along the laminate length upon heating.

Figure 2: Changes in line length parallel and perpendicular to the fiber orientation due to heating.

Figure 3: Differences in line length changes with temperature for each laminate sequence.

Table 1: CTE values of the epoxy network and UD laminate obtained via TMA and DIC.

	Matrix CTE ($\mu\epsilon/^\circ\text{C}$)	UD Laminate CTE – Parallel to Fiber ($\mu\epsilon/^\circ\text{C}$)	UD Laminate CTE – Orthogonal to Fiber ($\mu\epsilon/^\circ\text{C}$)
TMA	62.7	-1.1	31.6
DIC	60.0	-0.65	31.4

Table 2: Glassy-state CTE values for variable fiber orientation laminates.

Laminate Sequence	DIC CTE – Parallel to Fiber ($\mu\epsilon/^\circ\text{C}$)	DIC CTE – Orthogonal to Fiber ($\mu\epsilon/^\circ\text{C}$)	TMA CTE – Parallel to 0° ($\mu\epsilon/^\circ\text{C}$)
$0_8/90_8$	-5.3	23.8	13.7
$0_4/+45_4$	-1.5	27.8	4.4

Figure 1: Average change in linear dimension due to heating and continued curing.

Figure 2: Average dimension change due to thermal expansion of cured matrix.

Figure 3: Laminate placement for thermal expansion and strain-free temperature studies, with representative reference lines and stage points.

Figure 4: Distance changes for several points along the laminate length upon heating.

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