Factors affecting the properties of composites made by 4D printing (moldless composites manufacturing)

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Abstract 3D printing is the process where layers of materials are deposited to make structures of complex geometries. 4D printing is a process that combines 3D printing with the application of some activating agent in order to change the shape of the manufactured part after the process, e.g. the flat structure will change its shape to take up the desired complicated shape such as “curved” or “S” shaped. As such the 3D printing process does not have to spend time to print the intricate parts, and the process can be faster. The requirement for 4D printing is that materials with special characteristics that are responsive to an activating agent need to be used. 4D printing of composites utilizes the same concept of 4D printing, except that the materials used are long fiber composite materials. 4D printing of composites utilizes the shrinkage of the matrix resin, and the difference in coefficients of thermal contraction of layers with different fiber orientations to activate the change in shape upon curing and cooling. This behavior can be used to make parts with curved geometries without the need for a complex mold. As such manufacturing of pieces of curved shapes can be fast and economical. However, the degree of shape changing depends on the material properties, the fiber orientation, the lay up sequence and the manufacturing process. This paper presents the results obtained from a study on the effects of these aspects on the shape of the curved parts.

Keywords 3D printing, 4D printing, Composites, Complex structures, Automated composites manufacturing

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Introduction

3D printing (also known as additive manufacturing) is a powerful process that enables the efficient manufacturing of parts with complex shapes. While 3D printing offers an immense range of complexities, the process is relatively slow and it may not be most suitable for mass production. In efforts to take the technology further, the concept of 4D printing was introduced. The process can produce a self folding structure that can change its configuration under certain stimulating conditions, such as the absorption of some liquid like water, or the application of heat, or light. The process can also be some form of origami self folding mechanism. The material used in the 4D printing process is usually some soft plastics with special properties.

The concept of 4D printing using these types of mechanism is extended to the concept of 4D printing of composites (or moldless composite manufacturing) in this paper. The method of automated composite manufacturing using automated fiber placement (AFP) can be considered as a form of 3D printing using long and continuous fiber composites. On top of that, if the composite lay up sequence can be designed such that the composite structure changes its configuration upon curing (a form of activation process), then composite structures with curved configuration can be made without the need for a curved mold (basis for the term moldless composite manufacturing).

Consider for example the manufacturing of a spring having the shape of an S as shown in Fig. 1. Using the conventional method of manufacturing, one would need to make a curved mold out of some solid material such as metals, then laying up the prepregs on the mold to make the composite piece. The mold making is expensive and time consuming. However, if the lay up sequence is done right, only flat laminates need to be made.
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temperature (or both), the deformations of the different layers along different directions will make the flat laminate to reconfigure, which can be useful for the design of the flat laminates, a systematic analysis needs to be done. The calculated results should be validated using experimental values. This paper presents the theoretical formulation, along with the experimental results for validation.

For thermoset matrix composites, the reconfiguration of the laminate upon curing and cooling can be thought to be due to two mechanisms (with the assumption that the stiffness of the material is sufficiently high): One is the resin shrinkage due to chemical reactions between the molecules in the resin, and the other is due to mismatch in coefficients of thermal contraction along different directions in different layers in the laminate upon cooling from the cure temperature down to room temperature. Each of these two mechanisms is described below.

**Contribution from resin shrinkage**

For curing of thermoset resin, the liquid resin transforms into a solid via chemical reaction between the molecules. This transformation can take place at different temperatures depending on the type of resin. The transformation can take place at room temperature for resins such as polyesters. For most aircraft types of composite where the resin is epoxy, the transformation usually takes place at higher temperature (about 180 °C). The temperature at which the majority of the transformation takes place is called the cure temperature. There is significant shrinkage of thermoset resin due to this transformation.

A very rigorous modeling work on the effect of cure shrinkage and thermal contraction coefficients for composite laminates was performed by Bogetti and Gillespie. In this work, the process of resin cure and modulus development was considered to consist of three phases. Phase 1 is when the resin is a liquid and the modulus of the material is almost zero. When the resin gels, the degree of cure is denoted as $\alpha_{gel}^\text{mol}$. During phase 2, the resin transforms from the gel state to the solid state, from $\alpha_{gel}^\text{mol}$ to $\alpha_{solid}^\text{mol}$ and the modulus increases to its saturated value. Most of the shrinkage occurs within this phase. Phase 3 is the saturation state when there is no further shrinkage or modulus development (due to shrinkage). Note that the modulus is very low for a good part of the of the shrinkage development. As such even though shrinkage occurs in the early stage, this shrinkage may not contribute to the re-configuration of the part. The model proposed by Bogetti and Gillespie consists of many sub-models. The variation of temperature in the part and the development of cure of the resin were determined using energy balance and kinetic equations. Micromechanics equations were then used to determine the lamina properties. Laminate equations were then used to determine the residual stresses and curvatures. Comparison between calculated results and experimental results performed by Hahn and Kim for an unsymmetric laminate (90/0) showed good agreement for the curvature.

The work done by Bogetti and Gillespie was to understand the behavior of the materials that gives rise to the development of the residual stresses in the laminates. Certain assumptions on the values of different properties had to be made in order to proceed with the analysis. For example, the range of the degree of cure for the development of the modulus (from $\alpha_{gel}^\text{mol}$ to $\alpha_{solid}^\text{mol}$) is different from the total amount of shrinkage from the liquid state to the solid state and this needs to be assumed. Also the transverse modulus of carbon fiber composite needs to be assumed. The development of the resin...
modulus is assumed to take the form of the rule of mixture based on the development of cure. These and other assumptions give rise to some degree of approximations to the result obtained. Instead of going through the tedious calculations while getting approximate results, here a more simple method is proposed to get the approximate results. The validity of the method is determined by the end experimental result on the curvature of the reconfigured laminate.

In an earlier study on the shrinkage and modulus development of an epoxy resin, the method of ultrasonic was used to determine the development of these quantities. Figs. 2 and 3 show the results. In these figures, the amount of volume shrinkage and the values of the moduli depend on the amount of curing agents. In Fig. 3, the modulus at the beginning of the curing process is about 3 GPa. As the process time increases up to about 40,000 s, there are discontinuities in the modulus curves. This is due to the loss of ultrasonic signals when they are trying to traverse the rubbery state of the resin. At 40,000 s, the modulus is about 5 GPa. After that time, the modulus shows continuous increase up to a final value of about 6 GPa. Assuming that the significant reconfiguration takes place only after the modulus has reached its final saturation state, the corresponding period of time is from about 80,000 s to 100,000 s. The volume shrinkage varies from about 3.4 to 3.6% (taking the case of 40 phr). The resin shrinkage range is taken to be the difference between 3.6 and 3.4% (0.2%). The relation between volumetric shrinkage and linear shrinkage for an isotropic material can be as follows:

Relation between change $\Delta V$ and linear change $\Delta x$:

$$\frac{\Delta V}{V} = \left(\frac{x + \Delta x}{x}\right)^{3} - \frac{x}{x} \approx 3 \frac{\Delta x}{x}$$

(1)

A volumetric shrinkage of 0.2% would correspond to a linear shrinkage of 0.066%.

### Contribution from cooling from cure temperature to room temperature

Based on a normal manufacturing procedure for long fiber composites, after the sample is cured, it is cooled from the cure temperature to room temperature. The difference in the

| Table 1 Properties of the constituents$^a$ |
|------------------------------------------|
| $E_1$ (GPa) | 155.0 |
| $E_2$ (GPa) | 12.10 |
| $G_{12}$ (GPa) | 4.40 |
| $v_{12}$ | 0.248 |
| $a_1$ ($10^{-6}$/°C) | -0.018 |
| $a_2$ ($10^{-6}$/°C) | 24.3 |
| $\alpha_1$ (%) | 0 |
| $\alpha_2$ (%) | 0.066 |

Note: $\frac{\Delta V}{V} = \frac{\Delta x}{x}$

contractions of layers of different orientations will contribute to the reconfiguration of the laminates.

### Laminate theory to determine the radius of curvature

Due to the relative thinness of the structure, the reconfiguration can be analyzed using laminate theory. The determination of the strains and curvatures due to shrinkage and thermal effects can be obtained using the procedure outlined below:

The laminar properties of carbon/epoxy composites are given in Table 1. Values for the shrinkage coefficients are obtained using the above discussion.

Laminate matrix:

$$A_i = \int \bar{Q}_{ij} dz \quad B_i = \int \bar{Q}_{ij} dz \quad D_i = \int \bar{Q}_{ij} dz$$

(2)

where $i, j = 1, 2, 6$.

and

$$\bar{Q}_{11} = \frac{E_1}{1 - \nu_{12}^2} \frac{E_2}{1 - \nu_{21}^2} \frac{E_3}{1 - \nu_{31}^2}$$

(3)

and $E_i$, $E_j$, $G_{ij}$, $\nu_{ij}$, and $\nu_{ij}$ are moduli and Poisson ratios of a layer, respectively, and were given in Table 1.

Also $m = \cos \theta$ and $n = \sin \theta$ where $\theta$ is the angle between the fiber direction in a layer relative to the $x$ coordinate axis of the laminate.

For the case of a laminate subjected to a temperature change $\Delta T$, the relations for the strains and curvatures are given as:

$$\begin{bmatrix}
\varepsilon_x^o \\
\varepsilon_y^o \\
\gamma_{xy}^o \\
\kappa_x^o \\
\kappa_y^o \\
\kappa_{xy}^o
\end{bmatrix} =
\begin{bmatrix}
a_{11} & a_{12} & a_{16} & a_{17} & b_{11} & b_{12} & b_{16} & b_{17} & N_{11}^T \\
a_{12} & a_{22} & a_{26} & a_{27} & b_{22} & b_{26} & b_{27} & n_{2} & N_{22}^T \\
a_{16} & a_{26} & a_{66} & a_{67} & b_{66} & b_{67} & b_{67} & n_{6} & N_{66}^T \\
b_{11} & b_{12} & b_{16} & b_{17} & d_{11} & d_{12} & d_{16} & d_{17} & M_{11}^T \\
b_{12} & b_{22} & b_{26} & b_{27} & d_{22} & d_{26} & d_{27} & n_{2} & M_{22}^T \\
b_{16} & b_{26} & b_{66} & b_{67} & d_{66} & d_{67} & d_{67} & n_{6} & M_{66}^T \\
\end{bmatrix}$$

(5)
Similarly for a laminate subjected to shrinkage, one has the relation:

\[
\begin{bmatrix}
\varepsilon_x^o \\
\varepsilon_y^o \\
\gamma_{xy}^o \\
\kappa_x^o \\
\kappa_y^o \\
\kappa_{xy}^o
\end{bmatrix} = 
\begin{bmatrix}
a_{11} & a_{12} & a_{16} & b_{11} & b_{12} & b_{16} \\
b_{11} & b_{12} & b_{16} & a_{21} & a_{22} & a_{26} \\
a_{16} & a_{26} & a_{36} & b_{16} & b_{26} & b_{36} \\
b_{16} & b_{26} & b_{36} & d_{16} & d_{26} & d_{36}
\end{bmatrix} \begin{bmatrix}
N_x^o \\
N_y^o \\
N_{xy}^o \\
N_{xx}^o \\
N_{yy}^o \\
N_{xy}^o
\end{bmatrix}
\]

(5a)

where the column on the left hand side represents the in-plane strains and curvatures at the mid plane of the laminate, the square matrix represents components of the compliance, and the column on the right hand side represents the thermal stress resultants (or shrinkage stress resultants), and thermal moment resultants (or shrinkage moment resultants), respectively.

For laminates made of thermoset matrix composites, the shrinkage effect takes place during the curing of the resin which occurs at relatively high temperature. Subsequently, upon cooling from the cure temperature to room temperature, the difference in thermal contraction of layers at different fiber orientations will also give rise to residual stresses. These stresses in turn contribute to the reconfiguration.

The inverse of the relations (5) are:

\[
\begin{bmatrix}
N_x^o \\
N_y^o \\
N_{xy}^o \\
M_x^o \\
M_y^o \\
M_{xy}^o
\end{bmatrix} = 
\begin{bmatrix}
A_{11} & A_{12} & A_{16} & B_{11} & B_{12} & B_{16} \\
B_{11} & B_{12} & B_{16} & A_{21} & A_{22} & A_{26} \\
A_{16} & A_{26} & A_{36} & B_{16} & B_{26} & B_{36} \\
B_{16} & B_{26} & B_{36} & D_{16} & D_{26} & D_{36}
\end{bmatrix} \begin{bmatrix}
\varepsilon_x^o \\
\varepsilon_y^o \\
\gamma_{xy}^o \\
\kappa_x^o \\
\kappa_y^o \\
\kappa_{xy}^o
\end{bmatrix}
\]

(6)

and

\[
\begin{bmatrix}
N_x^o \\
N_y^o \\
N_{xy}^o \\
M_x^o \\
M_y^o \\
M_{xy}^o
\end{bmatrix} = 
\begin{bmatrix}
A_{11} & A_{12} & A_{16} & B_{11} & B_{12} & B_{16} \\
B_{11} & B_{12} & B_{16} & A_{21} & A_{22} & A_{26} \\
A_{16} & A_{26} & A_{36} & B_{16} & B_{26} & B_{36} \\
B_{16} & B_{26} & B_{36} & D_{16} & D_{26} & D_{36}
\end{bmatrix} \begin{bmatrix}
\varepsilon_x^o \\
\varepsilon_y^o \\
\gamma_{xy}^o \\
\kappa_x^o \\
\kappa_y^o \\
\kappa_{xy}^o
\end{bmatrix}
\]

(6a)

where the \(A, B, D\) are given by Equation (2).

Equation (5) can be abbreviated as:

\[
(\varepsilon) = [C] (R)
\]

(7)

And Equation (6) can be abbreviated as:

\[
(R) = [S] (\varepsilon)
\]

(8)

where \([C]\) is the compliance matrix and \([S]\) is the stiffness matrix, and we have:

\[
[C] = [S]^{-1}
\]

(9)

Also, the thermal stress resultants and thermal moment resultants are given as:

\[
N_x^o = \int \left( \bar{Q}_{11} \alpha_x^o + \bar{Q}_{12} \alpha_y^o + \bar{Q}_{16} \alpha_{xy}^o \right) \Delta T \, dz
\]

(10a)

\[
N_y^o = \int \left( \bar{Q}_{16} \alpha_x^o + \bar{Q}_{21} \alpha_y^o + \bar{Q}_{12} \alpha_{xy}^o \right) \Delta T \, dz
\]

(10b)

\[
N_{xy}^o = \int \left( \bar{Q}_{21} \alpha_x^o + \bar{Q}_{22} \alpha_y^o + \bar{Q}_{26} \alpha_{xy}^o \right) \Delta T \, dz
\]

(10c)

\[
M_x^o = \int \left( \bar{Q}_{11} \alpha_x^o + \bar{Q}_{12} \alpha_y^o + \bar{Q}_{16} \alpha_{xy}^o \right) \Delta T \, dz
\]

(10d)

\[
M_y^o = \int \left( \bar{Q}_{16} \alpha_x^o + \bar{Q}_{21} \alpha_y^o + \bar{Q}_{12} \alpha_{xy}^o \right) \Delta T \, dz
\]

(10e)

\[
M_{xy}^o = \int \left( \bar{Q}_{21} \alpha_x^o + \bar{Q}_{22} \alpha_y^o + \bar{Q}_{26} \alpha_{xy}^o \right) \Delta T \, dz
\]

(10f)

Where \(\alpha^o\) are the off-axis coefficients of thermal expansion and are given as:

\[
\alpha_x = \alpha_x m^2 + \alpha_y n^2 \quad \alpha_y = \alpha_y m^2 + \alpha_z n^2 \quad \alpha_{xy} = 2(\alpha_x - \alpha_y)mn
\]

(11)

and \(\sigma_x\) and \(\sigma_y\) are on-axis coefficients of thermal expansion of a particular layer, and were given in Table 1.

Similarly for shrinkage, the equations are:

\[
N_x^o = \int \left( \bar{Q}_{11} \varepsilon_x^o + \bar{Q}_{12} \varepsilon_y^o + \bar{Q}_{16} \varepsilon_{xy}^o \right) \, dz
\]

(12a)

\[
N_y^o = \int \left( \bar{Q}_{16} \varepsilon_x^o + \bar{Q}_{21} \varepsilon_y^o + \bar{Q}_{12} \varepsilon_{xy}^o \right) \, dz
\]

(12b)

\[
N_{xy}^o = \int \left( \bar{Q}_{21} \varepsilon_x^o + \bar{Q}_{22} \varepsilon_y^o + \bar{Q}_{26} \varepsilon_{xy}^o \right) \, dz
\]

(12c)

\[
M_x^o = \int \left( \bar{Q}_{11} \varepsilon_x^o + \bar{Q}_{12} \varepsilon_y^o + \bar{Q}_{16} \varepsilon_{xy}^o \right) \, dz
\]

(12d)
\[ M'_x = \int_0^1 (Q_{11} \varepsilon'_x + Q_{12} \varepsilon'_y + Q_{16} \varepsilon'_z) z \, dz \]  
\[ M'_y = \int_0^1 (Q_{12} \varepsilon'_x + Q_{22} \varepsilon'_y + Q_{26} \varepsilon'_z) z \, dz \]  

where the off-axis shrinkage strains are obtained by:
\[ \varepsilon'_x = \beta_1 m^2 + \beta_2 n^2 \quad \varepsilon'_y = \beta_1 n^2 + \beta_2 m^2 \quad \varepsilon'_{xy} = 2(\beta_1 - \beta_2)mn \]  

where \( \beta_1 \) and \( \beta_2 \) are the coefficient of shrinkage of a unidirectional layer, along the fiber direction, and transverse to the fiber direction, respectively, and were given in Table 1.

By obtaining the strains and curvatures as shown in Equation (5), the deformation of the laminate due to shrinkage during curing, or cooling down from either the curing temperature (or processing temperature), can be obtained.

Example

A simple illustration of the above concept would be to determine the curvature of a laminate made using [0/90] stacking sequence. This is an un-symmetric laminate and it may curl up upon curing and cooling from the curing temperature. It is of interest to determine the curvature of the final part.

As such,
\[ \varepsilon'_x = 0 \quad \text{and} \quad \varepsilon'_y = -0.00066 \]

For 0° layer:
\[ \varepsilon'_x = 0, \quad \varepsilon'_y = -0.00066, \quad \varepsilon'_{xy} = 0 \]

For 90° layer:
\[ \varepsilon'_x = -0.00066, \quad \varepsilon'_y = 0, \quad \varepsilon'_{xy} = 0 \]

Contribution from difference in coefficients of thermal contraction upon cooling

The curing temperature is 177 °C. The thickness of each layer is 0.125 mm. The curvature can be calculated using the formulation as follows:

From Equation (4):
\[ Q_{11} = 155.7 \text{ GPa} \quad Q_{12} = 3.02 \text{ GPa} \quad Q_{22} = 12.16 \text{ GPa} \quad Q_{46} = 4.40 \text{ GPa} \]  
\[ (14) \]

From Equation (3), for 0° layer:
\[ \bar{Q}_{11} = 155.7 \text{ GPa} \quad \bar{Q}_{12} = 3.02 \text{ GPa} \quad \bar{Q}_{22} = 12.16 \text{ GPa} \quad \bar{Q}_{46} = 4.40 \text{ GPa} \]  
\[ (15) \]

And for the 90° layer:
\[ \bar{Q}_{11} = 12.16 \text{ GPa} \quad \bar{Q}_{12} = 3.02 \text{ GPa} \quad \bar{Q}_{22} = 155.7 \text{ GPa} \quad \bar{Q}_{46} = 4.40 \text{ GPa} \]  

Solving Equation (23) yields:
\[ N_y^T + N_y^r = A_{66} \kappa_y \]  
\[ (24) \]

From Equation (11), for 0° layer:
\[ \alpha_x = -0.018 \times 10^{-6}/\text{C} \quad \alpha_y = 24.3 \times 10^{-6}/\text{C} \quad \alpha_{xy} = 0 \]  

And for 90° layer:
\[ \alpha_x = 24.3 \times 10^{-6}/\text{C} \quad \alpha_y = -0.018 \times 10^{-6}/\text{C} \quad \alpha_{xy} = 0 \]  

Combined effect

From Equation (2):
\[ A_{11} = h (155.7 + 12.16) = 167.86 \text{ h (GPa – m)} \]  
\[ A_{12} = h (3.02 + 3.02) = 6.04 \text{ h (GPa – m)} \]  
\[ A_{22} = h (12.16 + 155.7) = 167.86 \text{ h (GPa – m)} \]  
\[ A_{46} = h (4.40 + 4.40) = 8.80 \text{ h (GPa – m)} \]  
\[ (19) \]

and
\[ B_{11} = -\frac{\nu}{2} (155.7) + \frac{\nu}{2} (12.16) = -71.77 h^2 \]  
\[ B_{12} = -\frac{\nu}{2} (12.16) + \frac{\nu}{2} (155.7) = 71.77 h^2 \]  
\[ B_{22} = 0 \]  
\[ B_{46} = 0 \]  
\[ B_{66} = 0 \]  
\[ (20) \]

\[ D_{11} = \frac{\nu}{2} (155.7 + 12.16) = 55.95 h^3 \]  
\[ D_{12} = 20.1 h^3 \]  
\[ D_{22} = 2.93 h^3 \]  
\[ D_{46} = 0 \]  
\[ D_{66} = 0 \]  
\[ (21) \]

The stiffness matrix can be written as:
\[ [S]^T = \begin{bmatrix} 167.86 h & 6.04 h & 0 & -71.77 h^2 & 0 & 0 \\ 6.04 h & 167.86 h & 0 & 0 & 71.77 h^2 & 0 \\ 0 & 0 & 8.80 h & 0 & 0 & 0 \\ -71.77 h^2 & 0 & 0 & 55.95 h^3 & 2.01 h^3 & 0 \\ 0 & 71.77 h^2 & 0 & 2.01 h^3 & 55.95 h^3 & 0 \\ 0 & 0 & 0 & 0 & 2.93 h^3 & 0 \end{bmatrix} \]  
\[ (22) \]  

Due to the presence of the zeros, the stiffness relation can be broken down into three separate equations as:
\[ \begin{bmatrix} N_y^T + N_y^r \\ M_y^T + M_y^r \\ \alpha_y \end{bmatrix} = \begin{bmatrix} A_{11} & A_{12} & B_{11} & 0 & 0 \\ A_{12} & A_{22} & 0 & B_{22} & 0 \\ B_{11} & 0 & D_{11} & D_{12} & \kappa_y \\ 0 & B_{22} & D_{12} & D_{22} & \kappa_y \end{bmatrix} \begin{bmatrix} \varepsilon_x \\ \varepsilon_y \\ \kappa_x \\ \kappa_y \end{bmatrix} \]  
\[ (23) \]  

\[ N_y^T + N_y^r = A_{66} \kappa_y \]  
\[ (24) \]  

\[ M_y^T M_y^r = D_{66} \kappa_y \]  
\[ (25) \]
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Using the known values to evaluate the strains and curvatures in Equations (26)–(29), one has:

\[ \varepsilon_x = \varepsilon_y = -0.00147 \]  

\[ \kappa_x = \frac{1}{B_{11}} \left[ N_x^T + N_y^T - A_{11} \varepsilon_x - A_{12} \varepsilon_y \right] \]  

\[ \kappa_y = \frac{1}{B_{22}} \left[ N_y^T + N_y^T - A_{13} \varepsilon_x - A_{22} \varepsilon_y \right] \]  

\[ \varepsilon_x = \frac{Y_x(M_x^T + M_y^T - X_x) - X_x(M_x^T + M_y^T - Y_y)}{Y_x Y_x - Y_y Y_y} \]  

\[ \varepsilon_y = \frac{Y_y(M_x^T + M_y^T - X_y) - X_y(M_x^T + M_y^T - Y_y)}{Y_x Y_x - Y_y Y_y} \]

where

\[ X_1 = B_{11} - \frac{\alpha_{12}}{\alpha_{11}} A_{11} - \frac{\alpha_{12}}{\alpha_{12}} A_{12} \]  

\[ X_2 = -\frac{\alpha_{12}}{\alpha_{11}} A_{11} - \frac{\alpha_{12}}{\alpha_{12}} A_{12} \]  

\[ X_3 = \frac{\alpha_{12}}{\alpha_{11}} (N_x^T + N_y^T) + \frac{\alpha_{12}}{\alpha_{12}} (N_y^T + N_y^T) \]  

Using Equation (10), the thermal resultants are evaluated as:

\[ N_x^T = N_y^T = 3.71 \times 10^{-3} h \Delta T \]  

\[ M_x^T = -M_y^T = -1.12 \times 10^{-4} h^2 \Delta T \]  

With \( \Delta T = 20-177 = -157 \) °C, we have:

\[ N_x^T = N_y^T = -0.0582 h \]  

\[ M_x^T = -M_y^T = -0.0176 h^2 \]  

Using Equation (12), the shrinkage resultants are evaluated as:

\[ N_x^T = N_y^T = -0.0238 h \]  

\[ M_x^T = -M_y^T = -0.0075 h^2 \]  

Substituting values into Equations (30) and (31) gives:

\[ X_1 = 58.92 h^2 \]  

\[ X_2 = 0 \]  

\[ X_3 = 0.0616 h^2 \]  

\[ Y_1 = 0 \]  

\[ Y_2 = -58.92 h^2 \]  

\[ Y_3 = -0.0616 h^2 \]

Using the known values to evaluate the strains and curvatures in Equations (26)–(29), one has:

\[ \varepsilon_x = \varepsilon_y = -0.00147 \]  

\[ \kappa_x = -\kappa_y = \frac{0.00242}{h} \]  

Examination of Equation (40) shows that the curvatures depend on \( 1/h \) where \( h \) is the thickness of the layer group. This means that a laminate with stacking sequence of [0/90] has curvatures that are two times smaller than a laminate with a stacking sequence of [0/90].

Using the value of \( h = 0.130 \) mm (1.30 \( \times \) 10\(^{-4} \) m), one has:

\[ \kappa_x = \kappa_y = -18.62 m \]  

Giving

\[ R_x = -0.054 m = -5.4 cm \]  

\[ R_y = 0.054 m = 5.4 cm \]  

Experimental work

To verify the above calculations, a sample of carbon/epoxy composites with lay up sequence [0/90] was made using AFP machine and autoclave curing. The material was CYTEC 977-2 carbon epoxy prepregs. The curing temperature was 177 °C. Initially a flat stack of prepregs was made. The curing cycle is shown in Fig. 4. After curing, the laminate becomes curved, as shown in Fig. 5. The radius of curvature of the sample is measured to be 5.9 cm.

Discussion

(1) The difference between the calculated curvature and the experimental curvature is due to the many assumptions made for the values used in the calculation (5.9 cm vs. 5.4 cm). First is the amount of shrinkage that contributes to the reconfiguration. The results shown in Figs. 3 and 4 are for one particular type of resin and even these depend on the amount of the curing agents. These can have effect on the final result. Secondly the values shown in Table 1 also depend on the particular type of fiber, resin, and volume fraction. Thirdly, the modulus of the resin also changes as the temperature decreases.
The model shows that there are two curvatures, $R_x$ and $R_y$. However from experiment, only one curvature can exist at any one time. Fig. 5 shows the case. Whichever curvature is obtained from the mold depends on the constraint of the mold which favors one curvature over the other. This configuration can be snapped to take up the other curvature.

### Effect of different stacking sequences

The above example shows the situation for the unsymmetric laminate with the lay up sequence of [0/90]. It would be of interest to see the effect of varying the number of the respective layers 0 and 90 on the developed curvature. Table 2 shows the radius of curvatures for different laminates.

Note that the calculated radii of curvatures are different from the experimental results due to the assumptions discussed above. The experimental values have a range due to the variation of the radii of curvature from sample to sample, with the same lay up sequence. For the series of laminates [0/90], when $n$ increases from 1, the radius becomes smaller when $n = 2$ but increases for values of $n$ more than 2. This is because

![Figure 5 Curved piece after curing](image)

![Figure 6 Edge view of the prepreg stacks](image)

| Laminate   | Radius of curvature-calculated-cm | Radius of curvature-experimental-cm |
|------------|----------------------------------|-------------------------------------|
| 0/90       | 5.4                              | 5.6–7.2                             |
| 0/90,      | 10.8                             | 13.3                                |
| 0/90,      | 16.2                             | 15.9                                |
| 0/90,      | 4.7                              | 5.6–6.7                             |
| 0/90       | 5.7                              | 6.3–6.7                             |

Figure 6 Edge view of the prepreg stacks
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apart from the different amounts of contractions along different directions, there is also the effect of the change in the location of the neutral axis as the number of layers changes.

S shape pieces

In order to demonstrate the features of the 4D printing process, flat stacks of prepregs that reconfigure into S shaped laminates were examined. The lay ups for the S shape laminates are shown in Fig. 6. The steps are as follows:

Step 1: At the first elevation, initially a layer of 0° orientation is placed on the tool up to a certain length (this is called layer 1).

Step 2: At the same elevation, a layer at 90° orientation is then placed overlapping with a portion of the length of the first 0° layer (this is called layer 2).

Step 3: At the second elevation, a layer at 90° orientation is laid on top of the 0° layer (this is called layer 3).

Step 4: At the same second elevation, a layer at 0° orientation is placed overlapping with the third layer (this is called layer 4).

Three plates were made. The difference between them is the relative length between the left segment and the right segment of the laminate, as shown in Fig. 6.

Three flat configurations of the stack of prepregs are shown in Fig. 7. Upon curing and cooling, the flat stacks of prepregs reconfigured to have the S shape piece as shown in Fig. 8.

The curved configurations of the three laminates are traced and are shown in Fig. 9. Segment AB corresponds to the unsymmetric laminate [0/90] while segment DE corresponds to the unsymmetric laminate [90/0]. This gives the S shape configuration. The overlapped region BCD is straighter because the lay up sequence in this segment is [0/90/0].

The dimensions for the three plates are shown in Table 3. The table shows the radii of curvature at two ends of the S piece. There is variation of the radii of curvature from sample to sample, even though the lay up sequence is the same [0/90] or [90/0]. This variability is subject of a further investigation.

The edge tracings for the three plates are placed together as shown in Fig. 10.

Conclusion and further work

It is possible to lay up prepregs of composites in a flat stack, then upon curing, curved laminates can be obtained. This depends on the proper lay up sequence in the laminate. The
phenomenon depends on the asymmetric nature of composite laminates. By capitalizing on the self reconfiguration nature of the composite laminates, parts with complex geometries may be made without having to make molds of complex shapes. It is planned to develop engineering parts with complex geometries using this concept.

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