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Transverse crack initiation under combined thermal and mechanical loading of Fibre Metal Laminates and Glass Fibre Reinforced Polymers

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Abstract. The paper describes a temperature-dependent extension of the classical laminate theory (CLT) that may be used to predict the mechanical behaviour of Fibre Metal Laminates (FML) at cryogenic conditions, including crack initiation. FML are considered as a possible alternative class of structural materials for the transport and storage of liquified gases such as LNG. Combining different constituents in a laminate opens up the possibility to enhance its functionality, e.g. offering lower specific weight and increased damage tolerance. To explore this possibility, a test programme is underway at the University of Twente to study transverse crack initiation in different material combinations under combined thermal and mechanical loading. Specifically, the samples are tested in a three-point bending experiment at temperatures ranging from 77 to 293 K. These tests will serve as a validation of the model presented in this paper which, by incorporating temperature-dependent mechanical properties and differential thermal expansion, will allow to select optimal material combinations and laminate layouts. By combining the temperature-dependent mechanical properties and the differential thermal contraction explicitly, the model allows for a more accurate estimate of the resulting thermal stresses which can then be compared to the strength of the constituent materials.

1. Introduction
The search for Fiber Metal Laminates (FML) is inspired by the aeronautic industry, where concepts such as GLARE⁴ combine aluminium sheets with plies of unidirectional glass fibre reinforced epoxy. FML can have better damage tolerance, corrosion resistance, lower specific weight and higher yield strength compared to their constituents [1, 2]. As with any composite material, combining several constituents in a laminated structure allows to optimally exploit the strong points of each material, while its weaker properties are compensated by a different ply in the laminate. This way optimal structural material may be designed for different cryogenic applications.

Double walled steel tanks are presently used for the road transport of LNG. In this research a new class of cryogenic structural materials will be explored to replace the inner wall of such
Crack formation and delamination in a FML. The black circles schematically indicates the glass fibres, the red lines cracks in the polymer matrix and delamination fronts in between the plies.

The new materials should have a competitive advantage over the conventional solution, which can be achieved by combining specific properties, such as specific weight, thermal conductivity or impact strength, of different materials in a laminated structure. One concept for the inner wall consists of a thin walled metal tank around which continues glass fibres are wound. This method allows a large part of the steel to be replaced by glass fibre reinforced epoxy, thereby reducing the weight. The construction of a thin-walled vessel of steel followed by the winding of glass fibres around it can be achieved using conventional techniques.

The glass fibres are wound around the metal layer under two angles of continuous unidirectional fibres. Such a glass epoxy ply is strong in the direction of the fibre, but weak transverse to the fibres. Due to the combined thermal and mechanically loading, transverse cracks may initiate, leading to local delamination as schematically shown in figure 1. These cracks initiate and propagate between the fibres. The different orientations of the plies may cause the transverse cracks of one layer to cross the transverse crack of the next one, resulting in possible leak paths. One of the functionalities of the steel layer is to prevent such a leakage path through the wall.

Although laminates allow to combine different constituents, the mismatch in thermal expansion between the different materials in general results in the build-up of thermal stress during cool-down from the production - to the application temperature, which will influence the performance of a material. A first-order model is required to predict the thermal and mechanical response of the laminate and thus to select the most suitable candidate combinations for further testing. Many classical laminate theory (CLT) models are available, but they do not take into account the change in stiffness in calculating the thermal stress and therefore give less accurate stress and deformation estimates at cryogenic temperatures. In this work, a model based on CLT was constructed which takes the interplay between residual stresses at room temperature and changing stiffness explicitly into account. This treatment leads to an additional component of the thermal stress which allows to predict the mechanical, thermal and residual stresses in the laminate as a function of temperature more accurately.

2. Classical Laminate Theory for cryogenic temperatures

Classical laminate theory (CLT) is a straightforward but powerful modeling tool that can be used to estimate the stress, strain and curvature of laminated structures under thermal and mechanical loading [3]. However, the material properties such as the Young’s modulus $E$, the coefficient of thermal expansion $\alpha$ and the yield- and ultimate strength ($\sigma_y$ and $\sigma_u$) of some materials may change significantly between room temperature and cryogenic temperatures [4, 5]. Therefore, to provide reliable estimates the material properties in the CLT for cryogenic temperatures cannot be used temperature independent. Indeed, the stiffness of a laminate will in
Figure 2. Schematic representation of a laminate and the different coordinate systems. The indices 1 and 2 refer to the fibre direction (1) and transverse to it (2) of a ply. The global coordinate system, denoted by *, is under an angle \( \theta \) to the ply. In a CLT model, the material is assumed to be infinitely extended in the \( x \)- and \( y \)-direction, while the \( z \)-direction consists of \( n \) plies. \( h \) is the overall thickness, \( z_i \) the position of the interface between plies \( i \) and \( i+1 \) [6].

general change significantly with temperature and therefore also its stress-strain relation.

The semi-1D classical laminate theory models the laminate as an infinitely extended flat plate and considers only in-plane deformations. Therefore, plane stress conditions can be assumed (\( \sigma_{33} = \sigma_{13} = \sigma_{23} = 0 \)). Moreover, all materials are assumed to remain in the linear elastic regime. A schematic of such a laminate is shown in figure 2. Under these assumptions, the relation between the non-zero components of the stress \( \sigma \) and strain \( \epsilon \) of the \( k \)th ply at a given temperature \( T \) can be written as

\[
\begin{pmatrix}
\sigma_1 \\
\sigma_2 \\
\sigma_{12}
\end{pmatrix}_k =
\begin{bmatrix}
\frac{E_1}{1-\nu_{12}\nu_{21}} & \frac{\nu_{12}E_1}{1-\nu_{12}\nu_{21}} & 0 \\
\frac{\nu_{12}E_1}{1-\nu_{12}\nu_{21}} & \frac{E_2}{1-\nu_{12}\nu_{21}} & 0 \\
0 & 0 & G_{12}
\end{bmatrix}
\cdot
\begin{pmatrix}
\epsilon_1 \\
\epsilon_2 \\
\epsilon_{12}
\end{pmatrix}_k
\] or \( \{\sigma\}_k = [C]_{k,T} \cdot \{\epsilon\}_k \),

(1)

where the indices 1 and 2 refer to the fibre direction (1) and transverse to it (2) in the case of an unidirectional fibre reinforced polymer, \([C]\) is the stiffness matrix, \(G_{12}\) the shear modulus and \(\nu\) the poisson ratio. All the material properties in the compliance matrix are temperature dependent. For anisotropic materials such as FML or glass fibre reinforced polymers (GFRP), the material properties depend on the relative angle \( \theta \) of the ply compared to the global coordinate system of the laminate. The stress in a ply in the global laminate coordinate system can be derived as

\[
\begin{pmatrix}
\sigma_1^* \\
\sigma_2^* \\
\sigma_{12}^*
\end{pmatrix}_k
= 
\begin{bmatrix}
\cos^2 \theta & \sin^2 \theta & -2 \cos \theta \sin \theta \\
\sin^2 \theta & \cos^2 \theta & 2 \cos \theta \sin \theta \\
\cos \theta \sin \theta & -\cos \theta \sin \theta & \cos^2 \theta - \sin^2 \theta
\end{bmatrix}_k
\cdot
\begin{pmatrix}
\sigma_1 \\
\sigma_2 \\
\sigma_{12}
\end{pmatrix}_k,
\]

(2)

or \( \{\sigma^*\}_k = [T]^{-1}_k \cdot \{\sigma\}_k \)

where \([T]\) is the transformation matrix. The stress and strain of the \( k \)th ply in the global laminate coordinate system ply can be calculated as

\[
\{\sigma^*\}_k = [T]^{-1}_k \cdot [C]_{k,T} \cdot [R] \cdot \{\sigma\}_k.
\]

(3)

where \([C^*]_{k,T}\) is the temperature dependent stiffness matrix of the \( k \)th ply in the global coordinate system and \([R]\) the Reuter matrix.
The strain in a ply depends on the mechanical applied stresses $\sigma$, on the thermal expansion over a temperature difference from the glass temperature $T_g$ to the temperature $T$ and on the strain component $\epsilon_{cs}$ due to the chemical shrinkage of the epoxy during the curing process [7]. The strain in the $k$th ply (assuming an unstrained state at $T_g$) can therefore be written as

$$\{\epsilon^*\}_k = [S^*]_{k,T} \cdot \{\sigma\}_k + \int_{T_g}^{T} \{\alpha^*\}_{k,T} dT + \{\epsilon^*_{cs}\}_k,$$  \hspace{1cm} (4)

where $[S^*]_{k,T} = [C^*]_{k,T}^{-1}$ is the compliance matrix. The strain of the laminate can be expressed as $\{\epsilon^*\}_k = \{\epsilon^0\} + z_k\{\kappa\}$, where $\{\epsilon^0\}$ is the mid-plane strain and $\{\kappa\}$ the curvature. Inserting this in equation 4 and inverting it gives the stress in the local coordinate system as

$$\{\sigma^*\}_k = [C^*]_{k,T} \cdot (\{\epsilon^0\} + z_k\{\kappa\} - \int_{T_g}^{T} \{\alpha^*\}_{k,T} dT - \{\epsilon^*_{cs}\}_k).$$  \hspace{1cm} (5)

The stress on a ply can be divided in a mechanical, a thermal and a residual component. The residual stress component accounts for all stresses due to the curing process. This includes the chemical shrinkage of the epoxy, the moisture content and the cooling from the curing temperature to room temperature. The residual component is the stress level at which a specimen initially starts and is therefore independent of temperature. The thermal stresses are defined as the stresses due to a temperature change from room to the temperature $T$. The main component of the thermal stress simply originates in the different thermal expansion of each ply. A second part of the thermal stress comes from the changing of the stiffness of the ply. However, as the material properties change, so does the stiffness matrix $[C]_k$. When $[C]_k$ changes with temperature, the strain from the residual components will result in different stresses, which need to be included in the thermal stress. Hence, the second part of the thermal stress is the difference in stiffness ($[C^*]_{k,T} - [C^*]_{k,293}$) between room- and actual temperature $T$ multiplied by the residual strain. The stresses from the residual stress can therefore be separated in equation 5 in a thermal ($[C^*]_{k,T} - [C^*]_{k,293}$) and a residual ($[C^*]_{k,293}$) term. The three components of the stress can therefore be written as

$$\{\sigma^*\}_k = [C^*]_{k,T} \{\epsilon^0\} + z_k[C^*]_{k,T}\{\kappa\} - [C^*]_{k,T} \int_{T_g}^{T} \{\alpha^*\}_{k,T} dT$$

$$- ([C^*]_{k,T} - [C^*]_{k,293}) \int_{293}^{T} \{\alpha^*\}_{k,T} dT + \{\epsilon^*_{cs}\}_k,$$  \hspace{1cm} (6)

The applied force resultant $N^{Ap}$ on the laminate can be calculated by adding up the forces in each ply and can be written as

$$\{N^{Ap}\} = \sum_{k=1}^{N} \left( \int_{z_{k-1}}^{z_k} \{\sigma^*\}_k \cdot dz \right) = \{A\} \cdot \{\epsilon^0\} + \{B\} \cdot \{\kappa\} - \{N^T\} - \{N^{Res}\},$$  \hspace{1cm} (7)

with $N^T$ the thermal force resultant and $N^{Res}$ the residual force resultant. A similar expression can be derived for the moment resultant. These resultants can then together be written as

$$\begin{pmatrix} N^{Ap} \\ M^{Ap} \end{pmatrix} = \begin{bmatrix} A & B \\ B & D \end{bmatrix} \begin{bmatrix} \epsilon^0 \\ \kappa \end{bmatrix} - \begin{bmatrix} N^T \\ M^T \end{bmatrix} - \begin{bmatrix} N^{Res} \\ M^{Res} \end{bmatrix},$$  \hspace{1cm} (8)

where the A-, B- and D-matrix are the laminate extensional-, coupling- and bending stiffness matrix, defined as
\[ A_{ij}; B_{ij}; D_{ij} = \sum_{k=1}^{N} (C_{ij}^e)_{k,T} \int_{z_{k-1}}^{z_k} 1; z; z^2 dz \]  

The thermal and residual resultant are 'fictive' components, representing the force and moment required to compensate for the shrinkage of the laminate. The stiffness matrix can be used to calculate the strain and curvature of the laminate under the combined action of the thermal and residual components. By inverting equation 8, the mid-plane stress and curvature can directly be derived from the applied force and momentum and the residual and thermal components as

\[
\{ \epsilon^0 \} = \begin{bmatrix} a & bT \\ b & d \end{bmatrix} \begin{bmatrix} N_{\text{Apl}}^T \\
M_{\text{Apl}}^T \end{bmatrix} + \begin{bmatrix} N_{\text{Th}}^T \\
M_{\text{Th}}^T \end{bmatrix} + \begin{bmatrix} N_{\text{Res}}^T \\
M_{\text{Res}}^T \end{bmatrix}.
\]

where the residual force and -moment resultants are given by

\[
N_{i}^{\text{Res}} = \sum_{k=1}^{N} (C_{ij}^e)_{k,293}(z_k - z_{k-1})(\int_{293}^{T_g} (\alpha_j^*(T))_k dT + \{ \epsilon_{cs} \}_k)
\]  

and

\[
M_{i}^{\text{Res}} = \frac{1}{2} \sum_{k=1}^{N} (C_{ij}^e)_{k,293}(z_k^2 - z_{k-1}^2)(\int_{293}^{T_g} (\alpha_j^*(T))_k dT + \{ \epsilon_{cs} \}_k).
\]

The thermal force and -moment resultants are given by

\[
N_{i}^{\text{Th}} = \sum_{k=1}^{N} (z_k - z_{k-1}) \times 
\left( (C_{ij}^e)_{k,T} \int_{T}^{293} (\alpha_j^*(T))_k dT + (C_{ij}^e)_{k,T} - (C_{ij}^e)_{k,293} \left( \int_{293}^{T_g} (\alpha_j^*(T))_k dT + \{ \epsilon_{cs} \}_k \right) \right)
\]

and

\[
M_{i}^{\text{Th}} = \frac{1}{2} \sum_{k=1}^{N} (z_k^2 - z_{k-1}^2) \times 
\left( (C_{ij}^e)_{k,T} \int_{T}^{293} (\alpha_j^*(T))_k dT + (C_{ij}^e)_{k,T} - (C_{ij}^e)_{k,293} \left( \int_{293}^{T_g} (\alpha_j^*(T))_k dT + \{ \epsilon_{cs} \}_k \right) \right).
\]

The first term between the brackets of equation 13 and 14 is the usual thermal stress due to the thermal expansion of the plies, which can be found as \( \alpha \Delta T \) in most textbook derivations of the CLT [3], which use a temperature-independent stiffness matrix. In this derivation both \( \alpha \) and \([C]_k\) are temperature dependent. The second term, however, is an additional contribution which follows explicitly from taking this temperature dependence into account. Depending on the material choices and on the extent of the cool-down, this new component may contribute significantly to the thermal stress and hence the overall properties of the material.

### 3. Model

The analytical model described above can be readily evaluated (using numerical integration over the temperature history) to predict the cryogenic properties of laminated structures starting from the CLT theory with explicit inclusion of the temperature dependent material properties.
Figure 3. Estimation of a glass-fibre epoxy cross-ply, with in diamonds: the residual stress, in squares: the thermal stress, in circles: the transverse strength and in triangles: the resembling maximum mechanical stress before transverse crack initiation. Applied stress lower the maximum applied stress are expected safe, whereas higher stress will result in failure by a transverse crack or yielding.

This CLT formulation allows to estimate the stress and strain in each ply due to the combined action of applied mechanical loading and the stress and strain from the thermal and residual component. By performing a parametric sweep over temperature and force and by comparing it with the ultimate strength of the material, the maximum applied stresses before failure can be estimated. It should be noted that when the temperature is reduced, some material combinations become stronger (so that a higher mechanical load can be applied before failure), whereas others may weaken.

The model is thus able to predict the maximum applied mechanical stress before failure, the residual stress and the thermal stress, which together sum up to the transverse strength of the unidirectional ply $S_{2t}$. An illustration of such a calculation of 'safe' stress levels, as function of temperature for a typical cross-ply glass-fibre reinforced epoxy is shown in figure 3. Note how the maximum applied stress becomes a non-monotonic function of temperature, reflecting the relatively steep temperature dependence of the thermal stress (compared to the ultimate strength) at lower temperatures. This model will be validated with a series of three-point bending experiments at 77 K in a liquid N2 bath cryostat and from 173 to 293 K in a temperature-controlled N2 gas environment. The most important material properties as input for the model, $E_2$ and $S_{2t}$ will be measured on [90] UD glass specimens with three-point bending tests and $\alpha_2$ by strain gauges for low temperatures and a thermal mechanical analyser (TMA) up to the glass temperature. Transverse crack initiation in [90/0]$_S$ GFRP and both [90/Aluminium]$_S$ and [90/Steel]$_S$ FML will be measured over the temperature range and compared to the model.

So far, the model is static with a constant temperature over the laminate. In the envisaged FML application, cryogenic tanks will warm up and be cooled again many times and therefore
the model needs to be dynamic. During cooling, thermal gradients will occur over the laminate, especially at high cooling rates encountered e.g. during filling with the cryogenic liquid. This will result in different thermal stresses on either side of the ply. As discussed above, the stiffness matrix \( [C_{ij}] \) changes with temperature. Temperature profiles across the thickness of the laminate are readily taken into account by subdividing each ply in a number of subplies and assuming the temperature in each of these to be constant. The relative simplicity of the CLT formalism allows for a sufficiently fine subdivision without excessive increase in computational cost. For the calculation of the components of the ABD’-matrix with equation 9, the subdivision of the ply ensures that the actual plies stiffness is used and not the stiffness calculated from material properties at the average temperature of a ply. The thermal stress in a ply depends strongly on temperature. For the calculation of the thermal resultants, a significant error may occur if one temperature is used for the whole ply. By subdividing the ply into subplies result in a better estimation of the thermal stress and strain can be derived.

4. Conclusion & Prospects

The design of cryogenic transport or storage vessels constructed from light-weight, high-strength laminates requires a better understanding of thermal stresses throughout the laminate at cryogenic temperatures and their effect to the crack initiation in such a laminate. A model has been formulated based on the classical laminate theory which estimates the mechanical, thermal and residual stresses in the laminate as function of temperature. The model includes temperature dependent material properties explicitly and specifically takes into account the change in laminate stiffness. It was shown how, in conjunction with residual stresses at room temperature, the temperature dependence of the stiffness leads to an additional thermal strain term which may constitute a significant correction on the ’standard’ term that stems from differential thermal contraction.

Guided by this model, several FML types, as well as their constituent materials, will be tested for transverse crack initiation under combined thermal and mechanical loading in a three-point bending experiment at temperatures ranging from 77 to 293 K. The CLT model is used to predict the mechanical behaviour and the crack initiation of the laminate. The three-point bending tests will be performed to validate this model and to get a better understanding in the behaviour of FMLs at cryogenic temperatures. In a LN2 cryostat and in a temperature chamber from 173 to 293 K, the transverse flexural strength and modulus on [90] GFRP specimen and the transverse crack initiation on [90/0]_S, [90/Aluminium]_S and [90/Steel]_S specimen will be measured. The coefficient of thermal expansion will be separately measured with strain gauges and with a TMA from room to the glass temperature. Additionally, transverse crack initiation will be independently monitored with a cryogen-compatible acoustic emission method during the 77 and 293 K experiments.

References

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