Numerical Simulation on the influence of solidification rate on mechanical properties of semi-crystalline thermoplastic composites

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Abstract: - As with the consolidation time, the cooling rate applied during the solidification stage of the composite processing cycle influences the total processing cycle time, as well as the mechanical performance. By controlling the solidification rate, changes in the matrix morphology and crystallinity can be achieved, and hence different mechanical properties may be obtained. Control of the solidification rate is also important in order to control the level and distribution of internal stresses generated within the part during processing. These stresses may cause the composite to warp, resulting in unsatisfactory part quality, and can lead to premature failure of the part, necessitating costly repair or replacement. Therefore, this chapter emphasises the necessity of controlling the solidification rate, and examines its influence on the mechanical properties and the dimensional stability of composites based on CF/PA12 commingled yarns. The influence of solidification rate on crystallinity, morphology, and resulting mechanical properties of thermoplastic composites has become the subject of many research investigations over the last decade. Transverse tensile tests were performed to examine the influence of the solidification rate on the mechanical properties of the CF/PA12 laminates based on commingled yarns. In order to study the influence of solidification rate on interlaminar fracture toughness, mode I interlaminar fracture tests were carried out using the double cantilever beam (DCB) method, Interlaminar fracture toughness.

Key-Words: - Thermoviscoelasticity, Finite element modeling, semi-crystalline, Interlaminar fracture, Crystallinity, Composite processing

1 Introduction

The influence of solidification rate on morphology and mechanical properties of unidirectional composite laminates made from CF/PA12 and GF/PA12 FIT bundles was investigated by Evstatiev *et al.* [1, 2]. The type of reinforcement used was found to play an important role on the crystallisation kinetics and the crystal structure at the fibre/matrix interface. Due to the nucleation ability of carbon fibres, CF/PA12 laminates showed a faster crystallisation rate and a higher degree of crystallinity than GF/PA12 composites. The material used for this investigation was a plain weave fabric made from warp CF/PA12 commingled yarns (588 tex) and weft glass fibres (22 tex). A more detailed description of the material characteristics is given in Section 4.3.1. The fabric was "edge-welded" with a soldering device into 160 × 140 mm specimens, which were then cut and placed into the cavity of a steel matched-die mould of flat geometry. An Acmosan® release agent was previously applied to the inner surfaces of the mould to allow easy removal of the part after moulding. The mould, with inserted fabric plies, was placed in a laboratory hot press, heated to 220°C, held at this temperature for 10 min under an external pressure of 10 bars, and finally cooled down to room temperature while maintaining the pressure. According to the consolidation model validated for the same material in Chapter 4, these processing conditions ensure that the residual porosity of the part is below 0.05%. Microscopic observations confirmed that the chosen processing conditions led to completely consolidated laminates. The thermal histories of the differently processed laminates are schematically represented in Fig. 1. Note that the solidification rates reported above are accurate to within ± 5% between 180°C and 150°C, covering the temperature range associated with crystallisation of the PA12 resin.

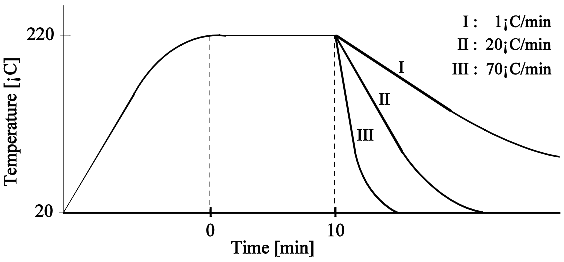

Fig.1. Schematic representation of the different thermal conditions applied during processing of commingled CF/PA12 composites.

2 Numerical Formulation

2.1 Anisotropic thermo- viscoelastic

The viscoelastic relaxation behavior of a polymer can be described by a Prony series expansion, a combination of several Maxwell models in parallel.

$$C_{kl}(t) = C_{kl0} + \sum_{\omega=1}^{N} C_{kl\omega} \exp(-t/\lambda_{kl\omega}) \qquad (1)$$

where C_{kl} are the nine independent relaxation functions of the material, and represent the relaxed and unrelaxed moduli at the relaxation time. The reduced time function given by Schapery in the uniaxial creep case.

$$\xi_{kl\omega}(x,t) = \int_0^t a_{ijkl} \{x, s, T(x,s), \alpha(x,s)\} ds \qquad (2)$$

where $a_{ijkl} = a_T(T(\tau)\ \ )$ is the so-called "temperature shift factor". The time interval $\tau = \xi - \xi'$ is in the absolute temperature *T*.

2.2 FE formulation of anisotropic viscoelastic

For the numerical implementation the anisotropic viscoelastic analysis described by Lin and Hwang based on the work of Christensen. It is assumed that displacements remain small in relation to the specimen thickness and that the dynamic effects of the warpage are insignificant. The resulting expression of the finite element equilibrium equation for an element in a non-isothermal condition is given by

$$\int_{-\infty}^{t} K_{mm}(t,\tau,T,\alpha) \frac{\partial u_m(t)}{\partial \tau} d\tau = F_m^{in}(t) + F_m^{ext}(t) \qquad (3)$$

where K_{mm} F_m^{in} and F_m^{ext}.

The term on the left hand side of Eq. (7) represents the time-dependant element stiffness at time *t* and the right hand side represents the time-dependant element loads at time *t*. $F_m^{in}(t)$ and $F_m^{ext}(t)$ are the components of the load induced by the volumetric shrinkage of the material and by the external boundary constraint. The element force nodal vector $F_m^t(t)$ at time *t* is the sum of $F_m^{in}(t)$ and $F_m^{ext}(t)$ and is given by

$$F^e(t) = \int_{V_e} \int_{\tau=-\infty}^{\tau=t} [B]^T \{C_{ij}(T, t-\tau)\} dV \frac{\partial \varepsilon_j(\tau)}{\partial \tau} d\tau + \int_{\partial V_e} N_{kn}\ \bar{T}_k(t) \partial V \qquad (4)$$

where C_{ij} is the material relaxation function of Eq. (4). For an amorphous thermoplastic C_{ij} is a function of time and temperature only,

$$C_{ij}(t) = \sum_{k=1}^{6} \sum_{l=1}^{6} \eta_{ijkl} C_{kl}(t) \qquad (5)$$

This relaxation function of the material can be expended tensor transformation matrix. For the new formulation following expression for the stiffness matrix at the *p*-th time step, so-call "**LHS**"(left hand side).

$$\text{LHS} = \sum_{s=1}^{p} \sum_{k=1}^{6} \sum_{l=1}^{6} \begin{bmatrix} C_{klo} B_{mnkl} \Delta u_n(t_s) + \\ \sum_{\omega=1}^{N} C_{kl\omega} B_{mnkl} h_{kl\omega}(t_s) \Delta u_n(t_s) \end{bmatrix} \qquad (6)$$

(6)

The recursion formulation scheme is extended to calculate the load vector and stresses as well as the hereditary stiffness terms. The load due to inelastic strain $F_m^{in}(t)$, can be expressed as;

$$F_m^{in}(t_p) = \sum_{s=1}^{p} \sum_{k=1}^{6} \sum_{l=1}^{6} C_{klo} B_{mjkl} \Delta \varepsilon_j^{inel}(t_s) + \sum_{k=1}^{6} \sum_{l=1}^{6} \sum_{\omega=1}^{N} f_{mkl\omega}(t_s) \qquad (7)$$

where $f_{mkl\omega}$ recursion expression.

Finally, the stress at the time step under consideration is expressed as

$$\sigma_i(t_p) = \sum_{s=1}^{p}\sum_{k=1}^{6}\sum_{l=1}^{6} C_{klo}\eta_{ijkl}\Delta\varepsilon_j^{eff}(t_p) + \sum_{k=1}^{6}\sum_{l=1}^{6}\sum_{\omega=1}^{N} s_{ikl\omega}(t_p) \quad (8)$$

using the recursion expression for $s_{ikl\omega}$

2.3 FEM scheme

The latter was performed using the finite element code of "Anisotropic Viscoelastic". The cooling process was modeled as a q uasi-static boundary value problem, assuming the thermal evolution to be decoupled from the build-up of stress and the stresses to have a n egligible effect on the thermal properties of the material. By removing the boundary constraints which represent the mold, the stresses relieved on demolding and the residual stress field in the specimen immediately after demolding can also be derived. The modular structure of this FE code allows for the implementation of additional modules, such as for the description of the crystallization kinetics of a semi-crystalline matrix and its influence on thermo-mechanical response.

3 Thermomechnical Behaviour

3.1 Material Characterization

Predicting internal stresses also requires a description of the composite thermomechanical behavior during cooling. The fiber undulation model used in the analysis, which is similar that developed by Ishikawa and Chou. According to this model, the average in-plane compliance of the matrix, $\overline{C_{ij}}$, can be evaluated as:

$$\overline{C_{ij}} = \frac{1}{a}\int_0^a C_{ij}(\phi)\,\mathrm{d}y \quad (10)$$

where a is half the undulation wave length and ϕ the local off-axis angle. The latter is defined as:

$$\phi(y) = \arctan\left(\frac{\mathrm{d}h(y)}{\mathrm{d}y}\right) \quad (11)$$

where $h(y)$ is a function describing the sectional shape of the weft yarn. In this approach, the relationships obtained from self-consistent field models were used in combination with the elastic properties of the U-shaped composite constituents given in Table 1. (12)

Table 1. Mechanical properties of composite components used in the micromodelling

Material	L (Gpa)	T (Gpa)	LT (Gpa)	TT (Gpa)	υLT	TT
Carbon	40	1	0	.3	.25	.27
Glass fibre	0	0	8.7	8.7	.22	.22
A12	.2	.2	.46	.46	.33	.33

3.2 Results of *Crystallinity*

The evolution of crystallinity of the neat PA12 resin as a function of solidification rate is presented in Figure 6.4. As shown, solidification rates between 1 and 100°C/min lead to an approximately constant degree of crystallinity, whereas at rates below 1°C/min the amount of crystallinity increases with a slowing down of the solidification rate. Figure 6.4 also shows the degree of crystallinity measured for the commingled CF/PA12 laminates cooled at 1, 20 and 70°C/min. A s was observed for the neat polymer, only small variations in crystallinity were obtained for the composite in the range of 1–70°C/min. Within this range of solidification rates, a crystallinity of 41 ± 3% is obtained. This result correlates well to the value of 38% reported by Evstatiev *et al.* [5] for a CF/PA12 composite made of FIT bundles, cooled at 10°C/min during processing. The results presented in Figure 6.4 also indicate that the crystallinity of the PA12 matrix is not significantly affected by the presence of carbon fibres in the laminate. This is probably explained by the two following counterbalancing effects: promotion of crystallisation due to the presence of fibres as nucleation sites, and impediment of crystal growth owing to the very dense packing of fibres ($V_f = 0.56$) in Fig. 2..

3.3 Results of FE simulation

The channels had a length of 120 mm, 44 mm of a height, a thickness of 2.5 mm, a corner internal radius of 2.5 mm, and an enclosed angle of 90° in Figure 1. To manufacture the laminates, the mould containing the fabric plies was heated from room temperature to 220°C, then held at this temperature for 10 min, and finally cooled back to room temperature.

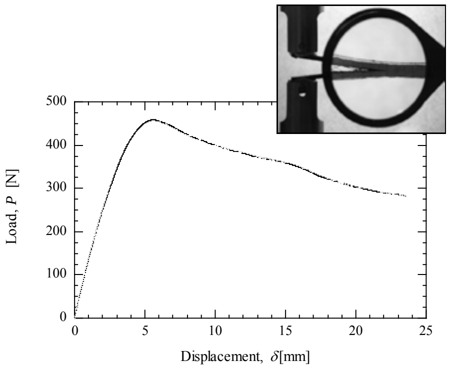

Fig.2 Load–displacement curve in unidirectional commingled CF/PA12 laminates.

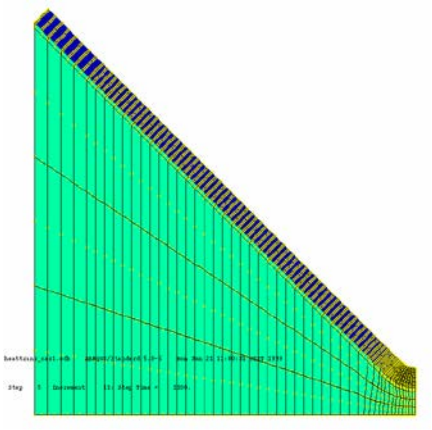

Fig.3 Geometry of the mould used for the manufacture of U-shaped composite laminates FEM model

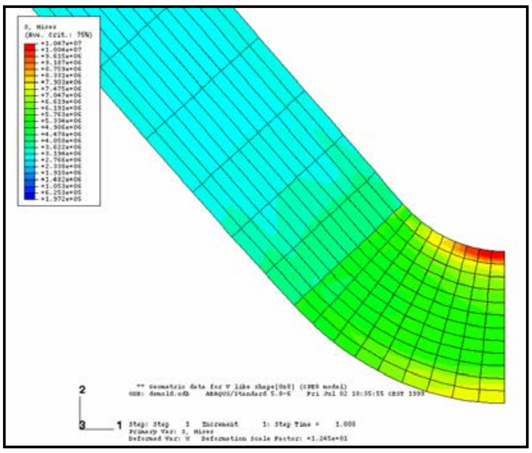

Fig.4 Von-Mises stresses of [90_8], geometry of half a section of a U-shaped laminate after demoulding

4 Conclusion

The build-up of stresses during the cooling of the material was calculated and curvatures on demoulding were predicted by numerical analysis. Precise control of the cooling applied during the solidification stage of the composite processing cycle is of the utmost importance in controlling the manufacturing cost, the matrix morphology, the mechanical performance and the dimensional stability of the final part. Increasing the solidification rate has the effect of reducing the processing cycle time and thus the manufacturing cost.

Thermal gradients within the composite during solidification, due to rapid cooling or unbalanced cooling conditions or complex geometry of the mould, induce internal stresses which may affect the dimensional stability of the part by causing post-processing distortions. The predictive and experimental analyses were conducted on laminated channels of different stacking sequences, which were processed with solidification rates around 10°C/min in the crystallisation temperature range.
The effect of springforward with the viscoelastic model has been investigate the influences of the steel demoulding and the effect of difference in thermal expansion between the mould and the composite component in U-shaped PA12/CF laminates.
Good correlation was obtained between the numerical prediction and the experimental data for the stress profiles and the warpage.

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