Composites Science and Technology 110 (2015) 145–151

Contents lists available at ScienceDirect

Composites Science and Technology

journal homepage: www.elsevier.com/locate/compscitech

Stochastic simulation of the influence of cure kinetics uncertainty on composites cure

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ARTICLE INFO

Article history:
Received 25 July 2014
Received in revised form 4 February 2015
Accepted 8 February 2015
Available online 13 February 2015

Keywords:
A. Polymer-matrix composites (PMCs)
C. Statistics
B. Curing

ABSTRACT

A stochastic cure simulation methodology is developed and implemented to investigate the influence of cure kinetics uncertainty due to different initial resin state on the process of cure. The simulation addresses heat transfer effects and allows quantification of uncertainty in temperature overshoot during the cure. Differential Scanning Calorimetry was used to characterise cure kinetics variability of a commercial epoxy resin used in aerospace applications. It was found that cure kinetics uncertainty is associated with variations in the initial degree of cure, activation energy and reaction order. A cure simulation model was coupled with conventional Monte Carlo and an implementation of the Probabilistic Collocation Method. Both simulation schemes are capable of capturing variability propagation, with the collocation method presenting benefits in terms of computational cost against the Monte Carlo scheme with comparable accuracy. Simulation of the cure of a carbon fibre–epoxy panel shows that cure kinetics uncertainty can cause considerable variability in the process outcome with a coefficient of variation in temperature overshoot of about 30%.

1. Introduction

Uncertainty in composite manufacture has begun to receive attention in the last decade as a result of the adoption of advanced composites as the main material of choice for large aero-structures. Variability is present in all forms of pre-impregnated and dry textiles and is mainly associated with in-plane and out-of plane fibre misalignment generated during production, handling or storage [1–3]. Experimental results have shown that in-plane fibre misalignment can be represented by a normally distributed variable [2,4] exhibiting strong spatial autocorrelation over the textile [2]. This type of uncertainty can affect the forming process introducing considerable variability in defect generation [2]. In addition, fibre misalignment as well as nesting effects can introduce significant scatter in permeability contributing to the formation of voids or dry patches [5–8]. Similarly to fibre misalignment, macroscopic permeability values can be represented by a normally distributed random variable [5–8]. However, simulation studies have indicated that permeability at the meso-scale (unit cell size) shows higher variations due to local inhomogeneities and cannot be represented by a normally distributed variable [9], a result that is also valid in the case of random mats [10]. In addition to variations in fibre architecture, the cure process can be potentially influenced by environmental/boundary condition uncertainty as well as resin behaviour variability due to variation in handling and storage conditions. Naturally these effects can play a role in cure process defects such as severe temperature overshoots or under-cure and also introduce variability in residual stresses/shape distortion.

The issue of variability during the cure step has received limited attention so far in the literature. The effect of cure temperature variations and cure kinetics uncertainty on cure time has been investigated in a pure simulation study by coupling a cure kinetics model with a Latin Hypercube sampling scheme showing that cure temperature variations tend to dominate cure time variability [11]. Furthermore, taking into account uncertainty in the optimisation of the cure time, has shown that optimal cure time increases with increasing variability [12]. These results, which are based on simulation and hypothesised levels of uncertainty, gain significant practical importance when their conclusions are combined with experimental studies of uncertainty in cure kinetics. In addition to material behaviour and process parameters uncertainty, cure kinetics can show significant variations due to experimental characterisation and data reduction discrepancies [13].

This study aims at the quantification of cure kinetics uncertainty and investigation of its propagation through the manufacturing

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http://dx.doi.org/10.1016/j.compscitech.2015.02.009
0266-3538/

process. The variability can be due to variable resin handling/storage conditions, variations in resin formulation and uncertainty introduced during resin characterisation. A series of experiments was carried out using Differential Scanning Calorimetry (DSC) to characterise cure kinetics uncertainty of a commercial epoxy resin used in aerospace applications. The variability in experimental behaviour was attributed to certain parameters of cure kinetics as expressed by a phenomenological model and the corresponding stochastic object was developed.

The resulting stochastic simulation problem was addressed by coupling a finite element analysis based cure simulation model with conventional Monte Carlo (MC) and the Probabilistic Collocation Method (PCM). The two stochastic simulation approaches were applied to the cure process of a thick carbon fibre–epoxy laminate to investigate the influence of cure kinetics uncertainty on variability in temperature overshoot. Temperature overshoot is defined as the maximum difference between the laminate temperature and mould temperature during the cure cycle. The two schemes were compared in terms of accuracy and efficiency.

2. Deterministic cure simulation model

A simulation of heat transfer effects occurring during the cure was implemented in the finite element analysis solver MSC.Marc. The model was three dimensional and transient. The materials considered were Hexcel G1157 [14] pseudo unidirectional carbon fibre reinforcement and Hexcel RTM6 [15] epoxy resin. The material properties depend on both temperature and degree of cure and the material sub-models of cure kinetics, specific heat capacity and thermal conductivity were implemented in user defined subroutines UCURE, USPCHT and ANKOND [16].

2.1. Cure kinetics model

The cure kinetics model used has been developed for the resin system of this study and is a combination of an *n*th order model and an autocatalytic model [17,18].

The cure reaction rate is expressed as follows:

$$\frac{da}{dt} = k_1(1-a)^{n_1} + k_2(1-a)^{n_2}a^m \tag{1}$$

where a is the current degree of cure, k_1, k_2 the reaction rate constants, and m, n_1, n_2 the reaction orders. The reaction rate constants incorporated diffusion rate limitation terms [19]:

$$\frac{1}{k_i} = \frac{1}{k_{i,c}} + \frac{1}{k_d} i = 1,2 \tag{2}$$

where $k_{i,c}$ are chemical rate constants following an Arrhenius temperature dependence, and k_d is a diffusion rate constant defined as [19]:

$$k_D = A_D e^{\left(\frac{-E_D}{RT}\right)} e^{\left(\frac{-b}{f}\right)} \tag{3}$$

$$k_{iC} = A_i e^{\left(\frac{-E_i}{RT}\right)} i = 1,2 \tag{4}$$

here T is the instantaneous temperature, E_i activation energies, A_i pre-exponential factors, A_D and E_D the pre-exponential factor and activation energy of the diffusion process respectively, R the universal gas constant, b a constant, and f the equilibrium free volume expressed as [19]:

$$f = w(T - T_g) + g \tag{5}$$

where w and g are constants, whilst T_g is the instantaneous glass transition temperature defined as [20]:

$$T_g = T_{go} + \frac{(T_{g\infty} - T_{go})\lambda\alpha}{1-(1-\lambda)\alpha} \tag{6}$$

Table 1
Parameter values for glass transition temperature, specific heat and thermal conductivity sub-models [18,20,21,23].

Parameter	Value
T_{go} (°C)	−11
$T_{g\infty}$ (°C)	206
λ	0.435
A_{fc_p} ($Jg^{-1}C^{-2}$)	0.0023
B_{fc_p} ($Jg^{-1}C^{-2}$)	0.765
A_{rc_p} ($Jg^{-1}C^{-2}$)	0.0025
B_{rc_p} ($Jg^{-1}C^{-2}$)	1.80
Δ_{rc_p} ($Jg^{-1}C^{-2}$)	−0.25
C_{rc_p} (1/°C)	1.10
s (°C)	16.5
A_{lf} ($Wm^{-1}C^{-2}$)	0.0074
B_{lf} ($Wm^{-1}C^{-2}$)	9.7
B_{tf} ($Wm^{-1}C^{-2}$)	0.84

where T_{go} and $T_{g\infty}$ denote the glass transition temperatures for the uncured and fully cured material, respectively and λ is a fitting parameter [18]. The parameters of the glass transition development model are reported in Table 1.

2.2. Specific heat capacity

The composite specific heat capacity is calculated using the rule of mixtures as follows:

$$c_p = w_f c_{pf} + (1 - w_f)c_{pr} \tag{7}$$

where w_f is the weight fraction of the fibre, c_{pf} the fibre specific heat capacity and c_{pr} the specific heat capacity of the resin.

The fibre specific heat capacity is a linear function of temperature and can be expressed as follows [21]:

$$c_{pf} = A_{f_{cp}} T + B_{f_{cp}} \tag{8}$$

where A_{fc_p} is the slope and $B_{f_{cp}}$ the intercept of the linear function [21]. The values of these two coefficients for carbon fibre [21] are reported in Table 1.

The specific heat capacity of the resin depends on both temperature and degree of cure. The dependence on degree of cure is expressed using a dependence on the instantaneous glass transition temperature of the resin (Eq. (6)) as follows:

$$c_{pr} = A_{rc_p} T + B_{rc_p} + \frac{\Delta_{rc_p}}{1 + e^{C_{rc_p}(T - T_g - s)}} \tag{9}$$

here A_{rc_p} and B_{rc_p} are constants expressing the linear dependence of the resin specific heat capacity on temperature for constant material state, while Δ_{rc_p}, C_{rc_p} and s are constants referring to the strength, breadth and temperature shift of the transition around T_g. The values of the coefficients used in Eq. (9) were estimated by fitting to experimental data produced by modulated scanning calorimetry [21] using a Genetic Algorithm and are reported in Table 1.

2.3. Thermal conductivity tensor

Each composite lamina is considered a transversely isotropic material.

The thermal conductivity in the longitudinal direction can be calculated as follows [22]:

$$K_{11} = v_f K_{lf} + (1 - v_f)K_r \tag{10}$$

K_{lf} and K_r are the thermal conductivity of the fibre in the longitudinal direction and of the resin respectively. In the transverse direction the thermal conductivity can be computed as follows [22]: