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## The significance of spatial temperature variations in large volume oven curing

Adam Fisher<sup>1,2</sup>, Arthur Levy<sup>2</sup>, James Kratz<sup>1</sup>

<sup>1</sup>The Bristol Composites Institute, University of Bristol

<sup>2</sup>Laboratoire de Thermique et Energie de Nantes, université de Nantes

The production of thermoset composite laminates in an oven or an autoclave involves considerable uncertainty, the majority of which comes from two main sources, the processing conditions, and the material itself. The complexity inherent in thermoset composite materials accommodates various unknown factors such as: initial degree of cure, resin composition, volume fractions and fibre alignment, all of which are liable to vary spatially within a part and from part-to-part [1]. Similarly, the processing conditions contain uncertainties, for example in temperature, pressure, tool thickness and localised heat transfer coefficients, as such the prescribed values are rarely those seen by the material. However, despite this, it is common practice to use deterministic cure kinetics models to predict the extent of a cure reaction [2]. These models assume values assigned to both the material properties and the processing conditions conform to a set of predetermined values.

In this study the validity of assuming a deterministic cure kinetics model was considered, addressing uncertainty in the cure kinetics parameters and the applied temperature values in the chosen model. The material considered during this study was HexPly® M21 particle interleaf toughened epoxy resin system. It has been found that a cure kinetics model that combines an nth order model and an autocatalytic model is capable of representing the cure reaction of this material [2]. The reaction rate constants have a diffusion term in addition to the conventional chemical component to capture the diffusion rate limitation phenomena.

The effect of the uncertainty of the inputs was assessed by considering its effect on the final degree of cure of the chosen material. To this end the deterministic cure kinetics model described above was used to propagate the uncertainty of the inputs to the final degree of cure, the Monte-Carlo method was used to produce a distribution of the output. The Monte-Carlo method required that the stochastic parameters were represented by distributions so a value could be sampled for each trial. To capture uncertainty in the cure kinetics parameters that was representative of the M21 material, the available 17 sets of dynamic DSC data [2] were fitted to the cure kinetics model, producing 17 values of each cure kinetics parameter. The relevant statistics of these values were then used to generate the distributions for each parameter. Sampled values from these distributions were then used as inputs to the cure kinetics model for each Monte-Carlo trial. It is noted that the DSC data was collected (i) by a single operator, (ii) using the same machine, (iii) on neat resin samples, and (iv) the material was from a single batch, thus eliminating four potential sources of uncertainty.

To determine which of the cure kinetics parameters had the most influence on the degree of cure, a sensitivity analysis was conducted. The sensitivity analysis showed that four of the cure kinetics parameters had a significant influence on the final degree of cure, the most influential of these were the two activation energies in the chemical component. The influence of two of the reaction orders was also above the remaining parameters but was less significant than the activation energies. In comparison with these four parameters the influence of the remaining parameters was negligible, thus in accordance with previous work, for the sake of efficiency they were treated as deterministic [3].

Following on from Mesogitis et al [3] who found that an activation energy and a reaction order followed a normal distribution, analysis was performed under the null hypothesis that the chosen parameters were normally distributed. A combination of visual tools and hypothesis testing was used to increase the reliability of the conclusion. Quantile-Quantile plots showed that in all cases the majority of the data points were close to the line representing a normal distribution. These observations were supported by results from Shapiro-Wilk normality tests, where all values were well within the 95% confidence interval that is standard for the test. It is noted that this result was achieved for the order of reaction  $m$  after an extreme

value was removed. The inability to reject the null hypothesis was consistent with the literature thus it was decided to assume the parameters were normally distributed.

A total of 10,000 Monte-Carlo trials were performed to generate an accurate representation of the distribution of the degree of cure values caused by the stochastic cure kinetics parameters after 1 and 2 hours at 180°C. These values were chosen as the recommended cure cycle for the M21 material consists of a 2-hour 180°C dwell. The histogram in Figure 1 representing the degree of cure after an hour indicates the distribution is symmetric about the mean and looks to be normally distributed. The long tails indicate the potential for large variability in the degree of cure during the curing process. The physical consequences of this variability could be the initiation of residual stresses, and dimensional instabilities that could cause issues in the assembly process. After 2 hours the histogram depicts a significant reduction in variability with most of the degrees of cure exceeding 95%. In both cases the deterministic degree of cure is a good approximation of the stochastic averages, suggesting that as the degree of cure increases and the variability decrease the need for stochastic methods decreases.

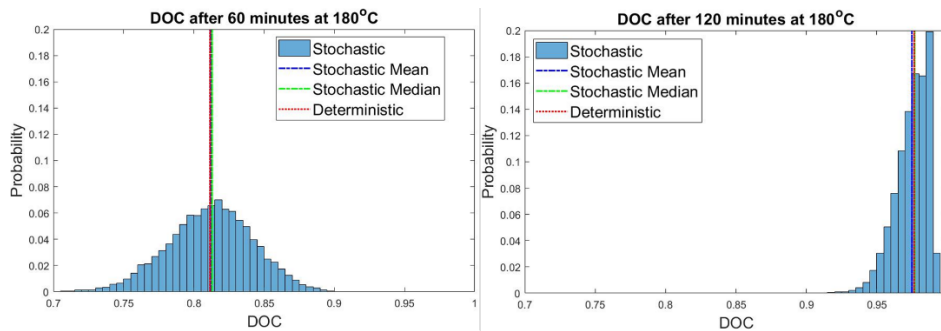


Figure 1. Histograms of final degree of after 60 (left) and 120 (right) minutes at 180°C

Analysis of thermocouple data from inside a range of ovens has shown that the amount of deviation from the programmed temperature (the temperature typically assumed in a deterministic model) is highly variable and can be large. The nature of the temperature variation depends on oven specific features, hence the oven-to-oven variability. In additions to temperature, there was also significant variability in the heat transfer coefficients calculated in each oven. Heat transfer coefficient is largely dependent on airflow, again this is influenced by oven specific features such as: the location of fans and exhausts, size and shape. All the ovens considered here were empty, however, under operating conditions the positioning of parts will obstruct the airflow, leading to variability between operations [4].

In this study attention was focused on a case study involving a 5m pre-production oven that was empty apart from thermocouples and the rig used to hold them. The spatial temperature variation was mapped using a 3x3 grid, at each node of the grid there was a lumped-mass calorimeter consisting of two thermocouples, one to measure air temperature, the other embedded in a stainless-steel rod allowing the heat transfer coefficient to be calculated. The temperature measurements were taken during an oven program that consisted of an initial 5°Cpm ramp from ambient temperature (25°C) to 120°C, a 30-minute hold at 120°C, a second ramp of 1°Cpm to 180°C and a 60-minute hold at 180°C.

The measured temperature distribution in this oven was characterised by a significant temperature gradient from front to back, the left-right temperature was more uniform, but variation was still present. The hottest region was at the front (closest to the door) and was the only region where the temperature closely followed the temperature program. In contrast, at the back of the oven where it was coldest, from the end of the first ramp the temperature was consistently below the prescribed temperature, with a peak difference of above 20°C in air temperature and above 50°C in the lumped mass. The largest temperature differences occurred in the lumped masses towards the end of the aggressive first ramp, where the lag caused by the higher heat capacity was most pronounced.

To put into perspective the significance of the measured temperature variation, the cure kinetics model was run with the air temperature measured at each calorimeter as the applied temperature. The results followed a similar pattern to the temperature, with the front measurements producing values closest to the expected degree of cure, the middle values do not deviate massively, but the rear values do. The peak discrepancy of nearly 20% represents a part with significant degree of cure gradients that would likely cause many issues including those mentioned above.

The temperature in the lump masses is dependent on the local heat transfer coefficient. Unlike the temperature measurements, the calculated heat transfer coefficients at any time varied little throughout the oven suggesting uniform airflow. The exception being at the central rear node where heat transfer coefficient was consistently greater, this was attributed to its proximity to the exhaust of the oven, demonstrating the potential effect of an oven specific feature.

To compare the influence of the oven temperature variability with the uncertainty in the cure kinetics parameters on the degree of cure, a sensitivity analysis was conducted. After a brief period at the beginning of the cycle where the temperature is low, the influence of the temperature exceeds any of the cure kinetics parameters and its peak reduced sensitivity value of 40% is more than double that of any of the cure kinetics parameters. This analysis clearly shows that the variability of the temperature in this oven has significantly more influence on the cure process than the uncertainty in the cure kinetics parameters.

A more general approach was taken to see what deviation from the programmed temperature cycle would produce an equivalent effect on the degree of cure as the uncertainty in the cure kinetics parameters and oven temperature. Treating the programmed temperature as the mean, the temperature was shifted downwards by a standard deviation assuming a range of coefficients of variation (COV) from 2% to 10%. The degree of cure under each of these cycles was predicted using the cure kinetics model and compared with those computed for the uncertain cure kinetics parameters and the lowest air temperature measurement from the oven. The 95% confidence interval ( $\pm 1.96sd$  assuming normality) for the uncertain cure kinetics parameters just covers the 2% COV curve, whereas the maximum deviation from the set temperature in the 5m oven is equivalent to a notional oven with a temperature COV of 8%.

The above analysis has shown that the measured variability in the oven temperature had a much greater influence on the degree of cure than the uncertainties in the cure kinetics parameters. Furthermore, due to the well documented [5] sensitivity of the cure process to temperature and the much greater influence exhibited, it has been shown that far less variability would be necessary before an equivalent influence on the degree of cure is realised. This suggests in cases where oven temperature variability is likely to be high, such as within large ovens, efforts to reduce the uncertainty in the final degree of cure of thermoset composite parts would be more effectively spent reducing oven temperature variability than in attempting to measure and account for the many sources of uncertainty in the material.

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