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## RANKING THE INFLUENCE OF KEY UNCERTAINTIES IN THE CURING OF THERMOSET LAMINATES

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**Abstract:** *The different characteristics of high and low thickness laminates means it is often optimal to process them under different conditions. Processing parts with tapered thicknesses can therefore be an issue as the entirety of the part must be processed in the same environment. To better understand how a processing environment can be optimised to minimise inhomogeneity across a tapered part, sensitivity analyses and Monte-Carlo simulations were performed. The sensitivity analyses showed how changes in key processing parameters affect the sensitivity of cure time to part thickness under different conditions. The results gave strong evidence to suggest that environments with a high heat transfer coefficient and a cure cycle designed for thick laminates were the most conducive to producing tapered parts with the greatest spatial uniformity. Results from the Monte-Carlo simulations showed the cure cycle recommended for thick Hexply M21 laminates was suited to most tapered parts but was highly conservative.*

**Keywords:** Thermoset; Curing; Modelling

### 1. Introduction

In many applications it is desirable for a composite part to be tapered, such as with an aircraft wing skin or wind turbine blade, in order to account for the decreasing lift generated towards the tips. Given the entirety of a part must be cured in the same environment, it is of great practical importance to know the characteristics of the curing environment that will result in the most desirable properties throughout the finished part. For example, to minimise the inhomogeneity in the degree of cure across the final part the aim will be to create a processing environment that minimises the cure time disparity between the thickest and thinnest sections of the part.

The curing process of thermoset parts is driven by temperature, as part thickness increases the reduced ability to dissipate exothermic heat causes greater transverse temperature inhomogeneities, leading to greater through thickness cure time variability. To mitigate this, it is common practice to use cure cycles with lower ramp rates and pre-dwells as parts become thicker. However, when a part is tapered the need for a gentler cure cycle will vary across the part, thus introducing a trade-off between part quality and processing efficiency. The question to be answered being, is it optimal to tailor the cure cycle for the thickest section thereby maximising quality throughout or does the optimal cycle lie somewhere between the two extremes? The answer depends on the desired outcome and the material being considered.

This study considers the processing of a tapered composite laminate made with Hexply M21, a high-performance epoxy system that is representative of the class of particle interleaf material systems. The desired outcome considered here was maximising manufacturing efficiency

through minimising cure time, while keeping the temperature overshoot below 10°C to avoid melting the thermoplastic particles. For superior fidelity, analysis considers uncertainty in material properties and processing conditions, therefore the aim is framed accordingly, achieving a cure time with minimum mean while avoiding temperature overshoots of 8°C (lower to account for thermocouple uncertainty) or more in 95% of cases.

The datasheet for Hexply M21 contains two cure cycle recommendations depending on the thickness of the part, the scenario is explored where the dimensions of a tapered part cover the thickness domains of the two cycles, and when each of the cure cycles should be used to achieve the desired outcome. In addition, a more general analysis is conducted to identify how different combinations of processing conditions affect the sensitivity of cure time to part thickness with the aim of establishing processing environments that are most conducive to minimising degree of cure variability in a tapered part.

## Method

### Finite Element Model

The curing process in the Hexply M21 carbon fibre laminates was modelled using a finite elements model that coupled transient heat transfer with cure kinetics. The assumption of negligible thickness change during each simulation was used to simplify the heat transfer analysis to a 1D problem [1].

The geometry of the 1D model is represented in Figure 1, it consisted of a homogenised M21/IMA carbon fibre reinforced epoxy laminate in ideal contact with a tool. Simulations were performed with tool properties representative of invar and a carbon fibre reinforced epoxy composite. Results were reported at the five equally spaced points across the laminate domain shown in Figure 1, this enabled the variations of cure time and temperature overshoot across the thickness to be computed.

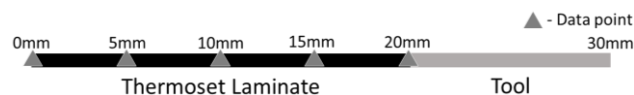


Figure 1. The geometry used in the finite element model (case of 20mm part thickness and 10mm tool thickness)

### Cure Kinetics Model

The cure kinetics model for Hexply M21 (Eq. (1)) was based on a variant of the Kamal and Sourour equation [2] and was successfully formulated and validated by Mesogitis et al [3].

$$\frac{d\alpha}{dt} = k_1(1 - \alpha)^{n_1} + k_2\alpha^m(1 - \alpha)^{n_2} \quad (1)$$

Where  $\alpha$  is the degree of cure,  $n_1$ ,  $n_2$  and  $m$  are reaction orders, and  $k_1$  and  $k_2$  are reaction constants which are composed of a diffusion term in addition to a chemical term to capture the effect of diffusion rate limitation phenomena [3], both have Arrhenius temperature dependence. The diffusion term was defined as in Eq. (2).

$$k_D = A_D \exp\left(-\frac{E_D}{RT}\right) \exp\left(-\frac{b}{w(T - T_g) + g}\right) \quad (2)$$

Where  $A_D$  is the pre-exponential constant,  $E_D$  is the activation energy of the diffusion process,  $R$  is the universal gas constant,  $T$  is temperature,  $T_g$  is glass transition temperature and,  $b$ ,  $w$  and  $g$  are constants. There is a chemical term for each reaction constant, they are defined in the same manner as Eq. (2) but without the second exponential. An initial degree of cure of 1% was assumed.

### Heat Transfer Model

The heat equation used for the composite domain is shown in Eq. (3).

$$\rho C_p \frac{dT}{dt} = k \frac{d^2T}{dx^2} + h(T - T_\infty) + L \frac{d\alpha}{dt}$$

Where  $\rho$  is the effective density,  $C_p$  is the specific heat capacity and  $k$  is the transverse thermal conductivity. The values of these parameters for M21 were obtained in the material characterisation conducted by Mesogetis et al [3]. The second term on the right-hand side represents the convective boundary condition,  $h$  is the convective heat transfer coefficient and  $T_\infty$  is the cure cycle temperature at time  $t$ . The final term, containing volumetric latent heat  $L$  and degree of cure  $\alpha$  represents the exothermic heat generated during the curing reaction and is responsible for the coupling between the heat transfer model and the cure kinetics model. Volumetric latent heat is defined in Eq. (4).

$$L = \rho_m(1 - v_f)H_T \quad (4)$$

Where  $\rho_m$  is the density of the matrix,  $v_f$  is the fibre volume fraction and  $H_T$  is the total heat reaction obtained through differential scanning calorimetry measurements [3]. Eq. (3) without the last term on the right-hand side was used for the tool domain. The solution of the model involved convective boundary conditions and an initial temperature of 293K through the thickness.

### Sensitivity Analysis

Sensitivity analyses were used to identify the set of processing parameters that minimised the influence of part thickness on the cure time. Cure time was defined as the time required to reach a degree of cure of 90% across the thickness. For different sets of processing parameters, cure time was predicted with part thicknesses of 5mm and 25mm, in each case the sensitivity was predicted using these values.

The independent variables considered were convective heat transfer coefficient, tool thickness and tool material, each with two levels. The cure cycle was also varied, the two cure cycles specified in the Hexply M21 datasheet were used, one for laminates with thicknesses less than 15mm, the other for laminates with thicknesses between 15 and 48mm. The cure cycle for thinner laminates consisted of a 2°C per minute ramp to 180°C, followed by a 120-minute dwell at 180°C and finishing with a 5°C per minute cooldown. The cure cycle for thicker laminates consisted of a 1°C per minute ramp to 150°C, an initial 180-minute dwell at 150°C, a 1°C per minute ramp to 180°C, a 120-minute dwell at 180°C and a 5°C per minute cooldown.

A central difference approximation of the sensitivity of cure time to part thickness was used for the analysis, the metric is defined in Eq. (5).

$$\frac{d\theta}{d\phi} = \frac{\theta(\phi + \Delta\phi) - \theta(\phi - \Delta\phi)}{2\Delta\phi} = \frac{\theta(\phi = 0.025) - \theta(\phi = 0.005)}{0.02} \quad (5)$$

Where  $\theta(\phi)$  is cure time when part thickness is  $\phi$  and  $\Delta\phi$  is the amount by which the part thickness was shifted. Central difference was used to capture the sensitivity of cure time to part thickness in the thickness domains of both cure cycles using a single metric.

To ensure the parameter space was thoroughly explored, analysis was conducted following the full-factorial experiment design shown in Table 1. The two levels assigned to each parameter are shown in Table 2. The lower heat transfer coefficient value was informed by industrial oven data and the higher by industrial autoclave data to provide values that were relevant to each environment. The two sets of properties given to the tool were obtained from the literature [4] and were intended to represent invar and a cured carbon fibre reinforced epoxy laminate, these properties are presented in Table 3.

To isolate the influence of the independent variables during the sensitivity analyses the remaining parameters were fixed, ensuring the chosen parameter was the only source of variability.

*Table 1. A full factorial experiment to determine the processing environment cure time and temperature overshoot are least sensitive to part thickness*

Experiment No.	Cure Cycle	HTC	Tool Thickness	Tool Material
1	-	-	-	-
2	-	-	-	+
3	-	-	+	-
4	-	-	+	+
5	-	+	-	-
6	-	+	-	+
7	-	+	+	-
8	-	+	+	+
9	+	-	-	-
10	+	-	-	+
11	+	-	+	-
12	+	-	+	+
13	+	+	-	-
14	+	+	-	+
15	+	+	+	-
16	+	+	+	+

Table 2. The key defining the two levels of the independent variables in the full factorial experiment (Table 1)

	-	+
<b>Cure Cycle</b>	t < 15 mm	15 < t < 48 mm
<b>HTC</b>	35 Wm <sup>-2</sup> K <sup>-1</sup>	150 Wm <sup>-2</sup> K <sup>-1</sup>
<b>Tool Thickness</b>	5 mm	15 mm
<b>Tool Material</b>	Invar	Composite

Table 3. Properties assigned to the tool materials in the heat transfer sub-model

Tool Material	$\rho$ [kgm <sup>-3</sup> ]	$C_p$ [Jkg <sup>-1</sup> K <sup>-1</sup> ]	$k$ [Wm <sup>-1</sup> K <sup>-1</sup> ]
Invar	8100	505	12
Carbon fibre reinforced epoxy composite	1550	800	0.7

## Stochastic Simulations

The model described above was incorporated into a stochastic framework, COMSOL Livelink for MATLAB enabled the model to be called repeatedly via a MATLAB script, allowing stochastic data to be generated using the Monte-Carlo method. Monte-Carlo trials were performed with cure kinetics and processing parameters being treated as normal random variables with means and variances derived from experimental data, this is in accordance with a number of previous studies [5, 6]. The means and variances for the cure kinetics parameters were obtained from differential scanning calorimetry data for M21 [3], those for the processing conditions were obtained from measurements within industrial ovens and autoclaves. Simulations were performed with part thicknesses between 5 and 45mm in 10mm increments for both recommended cure cycles. A convergence analysis was used to show 500 Monte-Carlo trials were sufficient to ensure the output distributions produced were repeatable.

The objective was to determine which cure cycle would minimise the cure time while producing temperature overshoots exceeding 8°C less than 5% of the time. Temperature overshoot was defined to be the maximum temperature difference above the dwell temperature through the thickness. The value of cure time and temperature overshoot were captured for each trial. The results for the two outputs were analysed to ascertain the statistical parameters of interest.

## 2. Results and Discussion

### Sensitivity Analysis

The sensitivity of cure time to part thickness for each experiment in Table 1 is shown in Figure 2. The clearest feature in Figure 2 is that the sensitivity in the first half of experiments is significantly higher than in the second, that is with all other dependent variables the same, the sensitivity of cure time to part thickness was consistently lower with the cure cycle recommended for thicker laminates.

With the thicker laminate cure cycle, it is evident that the increase in heat transfer coefficient leads to a reduction in sensitivity. This trend is also observed with the thin laminate cure cycle apart from with the thicker tool, as in experiments 3 and 7 where the tool was invar and, 4 and 8 where the tool was composite.

The influence of tool thickness appears clear with the thick laminate cure cycle, a thicker tool causing an increase in sensitivity. This same trend is observed for the experiments with the thin laminate cure cycle with higher heat transfer coefficient, but the opposite is observed with the lower heat transfer coefficient.

Finally, the effect of going from an invar tool to a composite tool was generally to increase sensitivity. The only exception to this in the data presented was when going from experiment 5 to 6, where the heat transfer coefficient was representative of an autoclave and the thinner tool was used.

Overall, the results from the sensitivity analysis indicated that of the processing environments considered, those in experiment 13 produced the lowest sensitivity of cure time to part thickness. Suggesting that homogeneity in a tapered thermoset composite part is maximised when processing it on a thin, low conductivity tool in an environment with a high heat transfer coefficient using a cure cycle with gentle heating ramps and pre-dwells. However, the closeness of experiments 14, 15 and 16 indicate that heat transfer coefficient and the selection of cure cycle were the most influential parameters. These results point to the suitability of autoclaves for producing tapered parts with uniform quality, due to the ability to augment the heat transfer coefficient with applied pressure. Despite the general superiority of the thick laminate cure cycle the conditions during experiments 3 and 6, that is low heat transfer coefficient and a thick invar tool and, high heat transfer coefficient and a thin composite tool respectively, presented a promising compromise between part uniformity and processing efficiency.

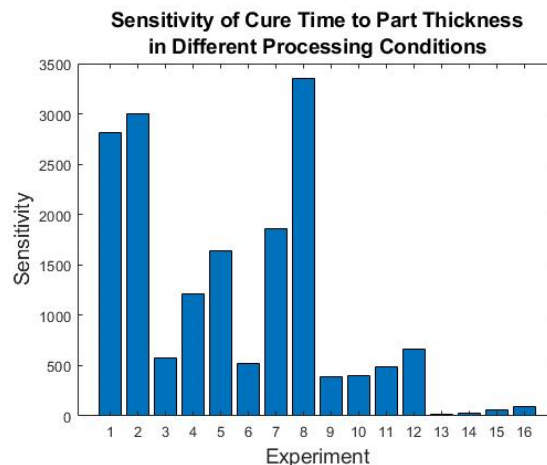


Figure 2. The sensitivity of cure time to part thickness in each processing environment of the full factorial design

### Stochastic Analysis

The mean cure times of the 500 Monte-Carlo trials performed using the two cure cycles with part thicknesses from 5 to 45mm are shown in Figure 3. The higher ramp rates and lack of a pre-dwell in the thin laminate cure cycle resulted in mean cure times that were significantly shorter.

Cure times were generally similar with the two tool materials, but slightly faster with the composite tool, the lower conductivity made temperature overshoots more likely.

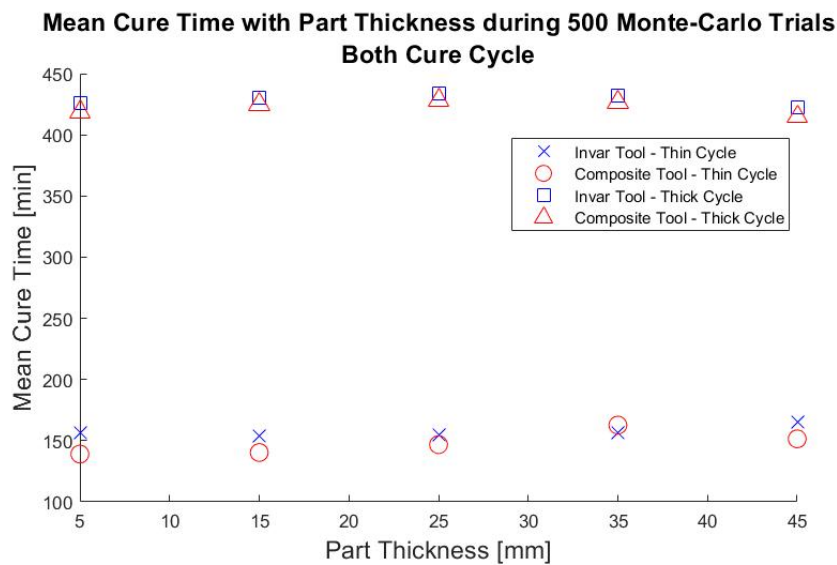


Figure 3. Mean cure time of 500 Monte-Carlo trials with the thin and thick laminate cure cycles

Given the dominance of the thin laminate cure cycle for minimising cure time, it was necessary to use the temperature overshoot criteria to select the appropriate cure cycle in each case. The thin laminate cure cycle was prone to cause much larger temperature overshoots, in most cases 8°C was exceeded significantly more than the 5% limit. The only cases that did not violate the limit were part thicknesses of 5 and 15mm with an invar tool. When the tool was composite, the thick laminate cure cycle was required in all cases. This difference was due to the lower conductivity of the composite tool stifling the dissipation of exothermic heat. On the other hand, with the thick laminate cure cycle the temperature overshoot criteria was satisfied in all cases.

Therefore, despite the significantly lower processing efficiency depicted in Figure 3, to produce acceptable quality tapered composite parts that have thicknesses outside the 5 and 15mm range or when a low conductivity tool is used, the cure cycle designed for thick laminates must be followed. The large difference of the two cure cycles and the universal applicability of the thick laminate cycle, suggests that for the majority of the tapered parts in the specified 15-48mm thickness range this cycle is too conservative and much greater efficiency could be achieved without violating the temperature overshoot criterion.

### 3. Conclusion

To determine which combination of the chosen processing parameters resulted in an environment where cure time was least sensitive to part thickness, a campaign of sensitivity analyses was conducted. Clear trends for two of the individual parameters were apparent, these were that the higher heat transfer coefficient and the cure cycle recommended for high thickness laminates consistently resulted in a lower sensitivity and were therefore more conducive to part uniformity. These results gave evidence to suggest that with the parameters considered, processing environments with both a heat transfer coefficient representative of an



autoclave with high pressure (i.e., around 7bar) and the Hexply M21 cure cycle recommended for thicker laminates would produce the most uniform tapered parts. Weaker influences were observed for tool material and tool thickness. The lowest sensitivity was predicted when the thick laminate cure cycle and high heat transfer coefficient were combined with a thin invar tool, however, the lower influence of the tool properties suggested these values were less critical.

Stochastic simulations showed that the thinner laminate cure cycle resulted in significantly lower cure times. However, the unacceptably large temperature overshoots in the majority of cases meant that the thick laminate cure cycle was almost always required. The thick laminate cure cycle produced acceptable temperature overshoots for all part thicknesses considered with both the invar and composite tools. The universal applicability of this cycle suggested it was overly conservative in most cases. Consequently, future work could investigate a systematic approach of tailoring cure cycles to tapered parts with thicknesses in the considered thickness range.

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