UNCERTAINTY QUANTIFICATION OF MATERIAL MODELS FOR PROCESS SIMULATION

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ABSTRACT

In composite process simulation, the ability to accurately capture changes in material behavior, such as degree of cure/crystallinity, viscosity, modulus and thermal expansion during the process is of great importance for making accurate predictions about the outcome of the process. Material behavior is commonly described by a mathematical equation whose parameters have been determined by fitting the equation to experimental data. This paper demonstrates the use of contemporary statistical methods to fit both parametric and nonparametric material models, how to quantify and reduce prediction error and uncertainty and how to avoid overfitting. Methods highlighted include: least-squares regression, Gaussian process regression, empirical Bayes and k-fold cross-validation. By quantifying prediction error and uncertainty, the value of data quantity and experimental design can be quantified, which is very useful when designing test matrices for material characterization.

Keywords: process modelling, cure kinetics, statistics, uncertainty
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1. INTRODUCTION

Composites processing is a complex multi-physics process where heat and pressure are applied to an uncured composite material on a tool, with the goal of producing a composite part that meets specific performance requirements [1]. For thermoset polymer composites, these requirements typically include a min/max thermal history, full cure, high glass transition temperature, low porosity, good fiber alignment, and good dimensional fidelity. The material suppliers provide general guidance on recommended process conditions to achieve high quality parts but when making large and geometrically complex parts, such as in the aerospace industry, the process often requires a significant amount of fine tuning until high quality parts are produced repeatedly and consistently [2].

1.1 Process Simulation

Development and fine tuning of composites processes can be very challenging, time consuming and costly if done by trial-and-error. This has led to a widespread use of process simulation, where a digital replica of the real process is created on a computer. Using process simulation, the outcome of a process can be simulated for different process conditions so that much of the trial-and-error can be done virtually rather than experimentally.

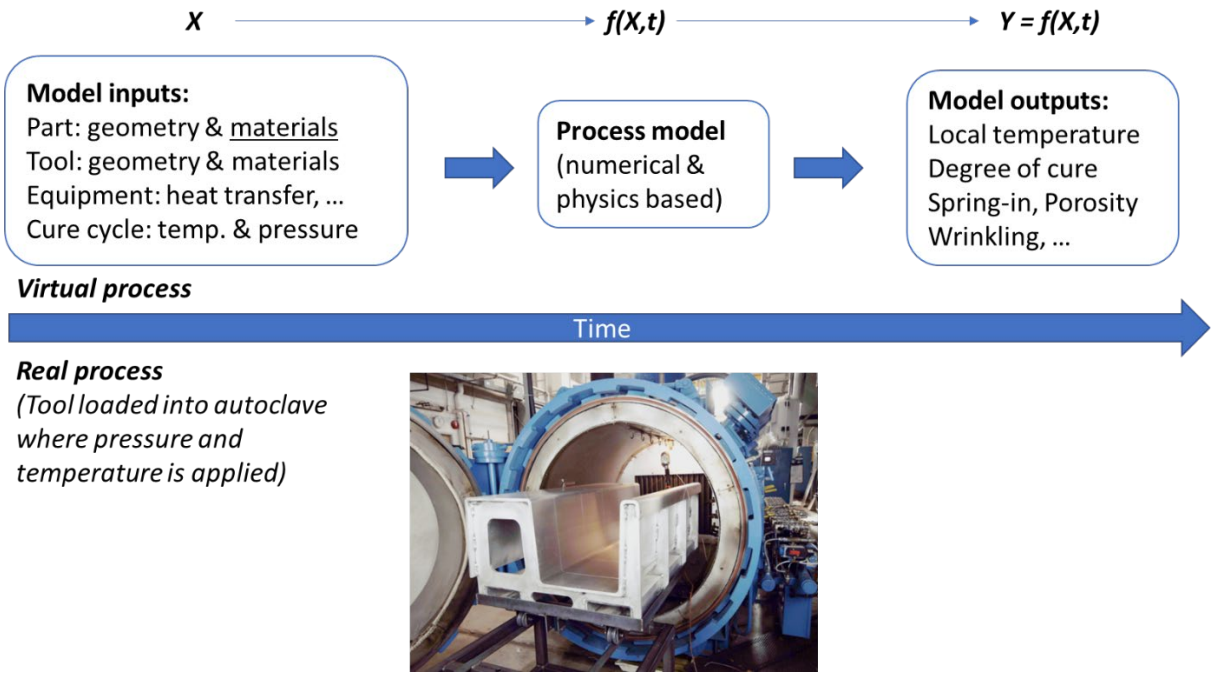

Figure 1. Virtual (top) and real autoclave process (bottom).

A process model can be thought of as a function f, that maps an input vector X to an output vector Y, Figure 1.

$$Y = f(X, t) \tag{1}$$

Where t represents time. A process model is typically a numerical implementation of a complex set of differential equations and the input vector X includes all the initial conditions and boundary conditions required to solve for the output variables of interest, Y. In many cases, these initial conditions and boundary conditions are not fixed values but complex and evolving material properties which are described by constitutive models themselves. For a process model to be useful, it must be able to give accurate predictions with sufficiently low uncertainty. There are some differences in terminology used in the scientific computing community [8] and the statistics community with respect to uncertainty. In the scientific computing community, we often talk about uncertainty from a modeler's point of view. Uncertainty in the model output Y is a combined effect of the uncertainty in model input X, the model form and numerical implementation thereof. The uncertainty is not just due to statistical randomness but everything that may result in the input or model not representing the physical reality correctly. We will use a broad definition of uncertainty in this paper.

The basis of most process models is a heat transfer calculation where the spatial and temporal temperature distribution within the part and process tools are calculated based on the prescribed equipment temperature and pressure. Error and uncertainty in process model predictions are due to uncertainty in the model inputs, X and bias in the process model itself, f. Convective and conductive heat transfer are well understood and our mathematical description of these phenomena

accurately model reality, but there can be considerable uncertainty in their boundary conditions. Much of the prediction error and uncertainty in the output of a process model is due to bias in material constitutive models such as degree of cure, viscosity and resin modulus. There is an abundance of different phenomenological equation forms for material constitutive models in the literature that aim to describe the same material property, such as cure kinetics. Most commercially available resins for high performance composites have complex and proprietary chemistries, which makes it very difficult to select an appropriate functional form from the literature that adequately describes the behaviour of the resin to be modeled with little bias.

The foundation for how material properties evolve during the process is the material state, which for thermosets is typically defined by the degree of cure x and temperature T. Note that x (degree of cure) is one of the components of the input vector X to the process model f. The degree of cure is typically calculated from a rate equation g of the form:

$$\dot{x} = g(x, T) \tag{2}$$

where $\dot{x}$ represents the derivative of x with respect to time. Prediction error in $\dot{x}$ will propagate through the larger process model f and cause prediction errors in the model output Y.

Uncertainty in process model inputs and outputs are typically not addressed explicitly; best practices dictate than some form of small-scale validation test is performed where model predictions and experimental results are compared to build confidence in model predictions. There has been very little work done in explicitly quantifying uncertainty in model inputs X and calculating how uncertainties are propagated though the process model f to the outputs Y of interest in composites process modelling. Without quantifying uncertainty and prediction error more precisely and objectively, it is challenging to assess the quality of the cure kinetics model and its fitness for use in different regions of the x-T space.

1.2 Objectives

This paper demonstrates how contemporary statistical methods and modelling techniques can be used to quantify and reduce error and uncertainty in process model inputs that are derived from experimental data. The focus is on cure kinetics, eq. (2), which is one of the most important inputs to a process model as accurate modelling of other material properties such as viscosity and resin modulus is contingent on having a high-quality cure kinetics model. By explicitly quantifying uncertainty and prediction error we can assess the value of data quantity with regards to model quality and design more efficient test matrices. Ultimately, the use of statistical methods can greatly reduce the time and cost of fitting robust material models and give objective measures of model accuracy at the same time.

2. BACKGROUND

When fitting a material constitutive model, there are two main categories of models to choose between: parametric and nonparametric models.

2.1 Parametric Modelling

Parametric models are models with a finite number of explicit parameters and have a fixed structure or functional form. An example of a simple parametric cure kinetics model based on chemical reaction dynamics [5] is:

$$K_i = A_i e^{-\Delta E_i/RT}$$
$$\dot{x} = \begin{cases} (K_1 + K_2 x)(1-x)(B-x) & x \leq x_c \\ K_3(1-x) & x > x_c \end{cases} \tag{3}$$

Fitting a parametric model to experimental data involves determining the model parameters such that the model best fits the data. This is typically done by imposing a distribution on the measured data, then using frequentist statistical procedures such as least-squares to estimate the parameters, and finally parameter uncertainty is estimated using asymptotic methods or bootstrapping. The primary advantages of parametric models are that they are generally interpretable and can incorporate the underlying physics of a problem to apply known bounds and constraints on the solution [6]. The potential disadvantage of parametric models is that if the selected functional form doesn't describe the process well, the model bias will prevent a good fit from being achieved.

2.2 Nonparametric Modelling

Conversely, nonparametric models impose very few assumptions and are nearly completely driven by data. The primary advantage of a nonparametric model is that by avoiding the assumption of a specific functional form, the model has less constraints and a wider range of model shapes can be explored. The disadvantages of nonparametric models are that they aren't very interpretable and usually require more data to train effectively than a parametric model [6].

A powerful nonparametric model is the Gaussian process (GP). A GP is a form of stochastic process such that any finite subset $\{x_1, x_2, \ldots x_n\}$ of the domain X is a multivariate Gaussian random variable [7]. GP regression involves taking a weighted average of datapoints where weights are determined by a smooth covariance function (or kernel function). GP regression is also a form of Bayesian inference, and the result is a multivariate Gaussian posterior distribution with a mean vector and covariance matrix. As such, GP regression not only results in smooth model curves, but also inherently produces information about the uncertainty in its predictions.

The most widely used covariance function is the Gaussian radial basis function (RBF) kernel.

$$k(x_i, x_j) = exp\left(-\frac{1}{2\sigma^2}\left\|x_i - x_j\right\|^2\right) \tag{4}$$

Where σ^2 is referred to as the kernel bandwidth and determines the width of the kernel. Small bandwidths assign greater weights to a smaller window of neighbouring datapoints, whereas large bandwidths assign lesser weights to a larger window of neighbouring datapoints. Kernel bandwidth is the primary parameter to determine when fitting a Gaussian process regression model and determines the smoothness of the model.

If we want to make n_2 predictions $y_2 = f(x_2)$ based on n_1 previous observations (x_1, y_1), given a prior mean function $m(x)$ and covariance function $k(x, x)$ we can write:

$$\begin{bmatrix} y_1 \\ y_2 \end{bmatrix} \sim \mathcal{N}\left(\begin{bmatrix} \mu_1 \\ \mu_2 \end{bmatrix}, \begin{bmatrix} \Sigma_{11} & \Sigma_{12} \\ \Sigma_{21} & \Sigma_{22} \end{bmatrix}\right) \tag{5}$$

Where:

$$\begin{aligned} \mu_1 &= m(x_1) \\ \mu_2 &= m(x_2) \\ \Sigma_{11} &= k(x_1, x_1) \\ \Sigma_{22} &= k(x_2, x_2) \\ \Sigma_{12} &= k(x_1, x_2) = \Sigma_{21}^T \end{aligned} \tag{6}$$

Combining multivariate Gaussian theorem and Bayes' theorem, we can then get the posterior mean and covariance matrix [7]:

$$\begin{aligned} \mu_{2|1} &= \mu_2 + \Sigma_{21}\Sigma_{11}^{-1}(y_1 - \mu_1) \\ \Sigma_{2|1} &= \Sigma_{22} - \Sigma_{21}\Sigma_{11}^{-1}\Sigma_{12} \end{aligned} \tag{7}$$

2.3 Empirical Bayes

One way to incorporate physical knowledge into a Gaussian process is to use empirical Bayes methods. Contrary to standard Bayesian methods where the prior mean and distribution are fixed before data is observed, empirical Bayes methods estimate the prior mean and distribution from the data. More specifically, a parametric, physics-based prior mean function is fit to the training data to use as a "starting point" for the GP regression model. This helps constrain the GP posterior and can guide the model in regions where training data is sparse, giving the benefits of both physical knowledge of the underlying process and flexible modelling.

2.4 Model Fitting Best Practices

One of the biggest perils in model fitting is overfitting the data. The first step in fitting a model, which is often overlooked in composite materials modelling, is to partition the experimental data into a training set and a test set. The model is then fit to the training set and the test set is withheld to be used for validation purposes only. The procedure of withholding a portion of the data allows for proper assessment of how well a model generalizes to new data and helps avoid overfitting.

In statistics and machine learning, predictor variables are often referred to as features. When features have considerably different scales, it can be beneficial to transform them to a common scale. For parametric models, this generally isn't necessary as there is often a closed-form objective function to minimize or maximize. However, many nonparametric models are sensitive to differences in feature scale. In the case of a Gaussian process, the kernel function computes the Euclidean distance between points and thus it is important that the features have similar scales so as not to bias the covariance. A common method of feature scaling is standardization, where features are scaled to zero mean and unit variance by subtracting the unscaled mean and dividing by the standard deviation, eq. (8):

$$x_{scaled} = \frac{x - \mu}{\sigma} \tag{8}$$

2.5 Performance Metrics

A common metric for assessing model quality is the root-mean-squared-error (RMSE), which represents the average prediction error of the model, eq. (9):

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(y_i - \hat{f}(x_i)\right)^2} \tag{9}$$

where, n is the number of data points, y_i is the observed data point at $x = x_i$ and $\hat{f}(x_i)$ is the model prediction at that point. A low RMSE value indicates that the model predicts the known data points well and that there is good agreement between model and data. RMSE is often normalized by a value such as the data mean or interquartile range so that it may be expressed as a percentage. In this paper, RMSE is normalized by the mean of the training response data.

In order to determine how well the model generalizes to new data before performing the final validation on the test set, a k-fold cross-validation is often performed. K-fold cross-validation involves partitioning the training dataset into k subsets, training the model using $k - 1$ of the subsets and validating it on the remaining subset. This procedure is then repeated $k - 1$ times.

3. EXPERIMENTATION

3.1 Materials Characterization

To create a cure kinetics model, eq. (2), experimental data for the resin of interest is needed. Cure kinetics data is generated using a Dynamic Scanning Calorimeter (DSC), which measures the heat flow during the cure reaction. The curing reaction is typically characterized by running a series of dynamic ramp tests and isothermal tests. In a dynamic ramp test, the sample is equilibrated at a low temperature and then heated at a predefined rate. In an isothermal test, the sample is equilibrated at a low temperature and then heated to a predefined hold temperature at a very high rate. Together, these tests are designed to bracket any ramp rates and hold temperatures anticipated in the manufacturing process. Below are some details about the characterization process used for this paper:

- Material characterized: HexPly M18/1.
- Material form: prepreg with Hexcel AS4C fibres
- Fibre volume fraction: 55%.
- Sample mass: 20 mg
- Pan type: T-zero aluminum hermetic
- DSC used: TA Q2000
- Sampling frequency: 1 Hz
- Dynamic ramp tests conducted: 1, 2, 3, 4, 5 ˚C/min

- Isothermal tests conducted: 100, 140, 155, 180 ˚C

In total, 5 dynamic ramp tests and 4 isothermal tests were conducted. The resulting full dataset (large) was divided into two smaller sub-sets (medium and small) so that the effect of dataset size on resulting model quality could be assessed. Note that the same test data is used in all data sets, only the amount of data differs. The test matrix for three data sets is shown in Table 1.

Table 1. Cure kinetics test for large, medium and small datasets.

Dataset	Dynamics	Isothermals
Small	1, 3, 5 ˚C/min	N/A
Medium	1, 3, 5 ˚C/min	140, 155, 180 ˚C
Large	1, 2, 3, 4, 5 ˚C/min	100, 140, 155, 180 ˚C

3.2 Model Fitting and Assessment

All data processing and analysis shown in this paper was conducted using custom python scripts based on well-established modules and libraries such as numpy, scipy, pandas and scikit-learn. Both parametric and nonparametric cure kinetics models were fit to compare their performances and observe their strengths and weaknesses. For all models, 70% of the data was used for training and 30% was withheld for validation.

Parametric models were generated by determining the optimal parameters for eq. (3) using least-squares with scipy's curve fitting function, scipy.curve_fit(). Parameter covariance was determined automatically by scipy.curve_fit() using asymptotic methods. A 10-fold cross-validation was then conducted before determining the test set RMSE.

Nonparametric models were generated using Gaussian process regression with empirical Bayes. First, a simple parametric prior mean function derived from the natural log of the Arrhenius terms in eq. (3): $ln\,\dot{x} = A - (B/T)$ was fit to the training data. Next, features were standardized using eq. (8) before training the GP. For cure kinetics, the features are degree of cure x and temperature T, and the response is the cure rate $\dot{x}$. A 10-fold cross-validation was then performed on a series of GP regression models trained using various kernel bandwidths. The kernel bandwidth was selected so that the lowest cross-validated RMSE was achieved while still producing sufficiently smooth model curves. GP regression model uncertainty was calculated by taking the square root of the diagonal of the posterior covariance matrix to be the standard deviations in the model predictions.

Note that eq. (3) is far more rudimentary than what Convergent would typically use for a cure kinetics characterization. It was selected for this study due to its use in the literature and to demonstrate the procedure of fitting a parametric model compared to a non-parametric model.

4. RESULTS

4.1 Raw Data

The raw test data for all the dynamics and isothermals was reduced to the form of eq. (2) and plotted in Figure 3.

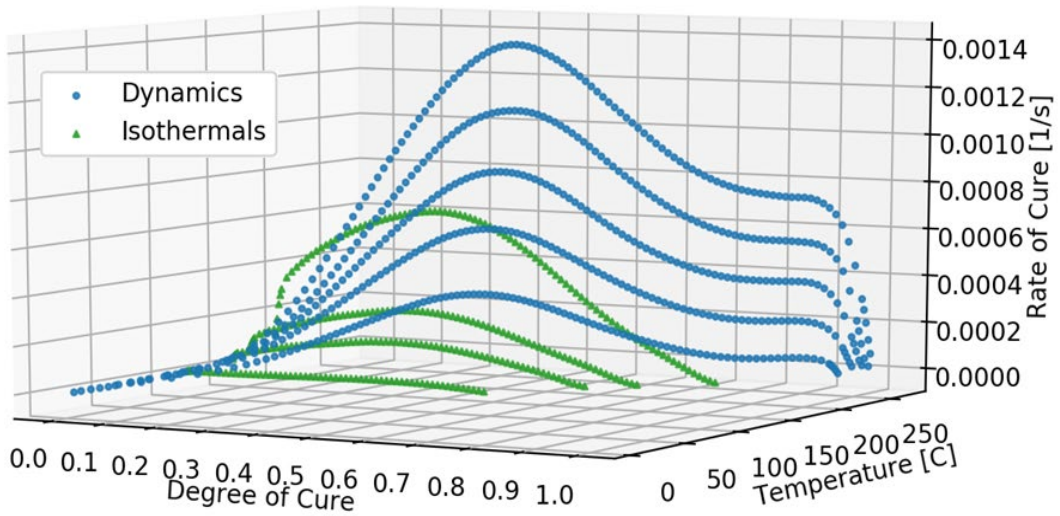

Figure 2. Cure kinetics test data.

4.2 Parametric Model

The parametric model, eq. (3), was fit to the large dataset and the resulting cure rate predictions for the isothermal and dynamic runs are shown in Figures 3 and 4. Solid lines represent model predictions and scatter points represent experimental data. As seen from the Figures, the agreement between model predictions and experimental data is poor, which is a reflection that the form of the parametric model used does not represent the underlying curing reaction well.

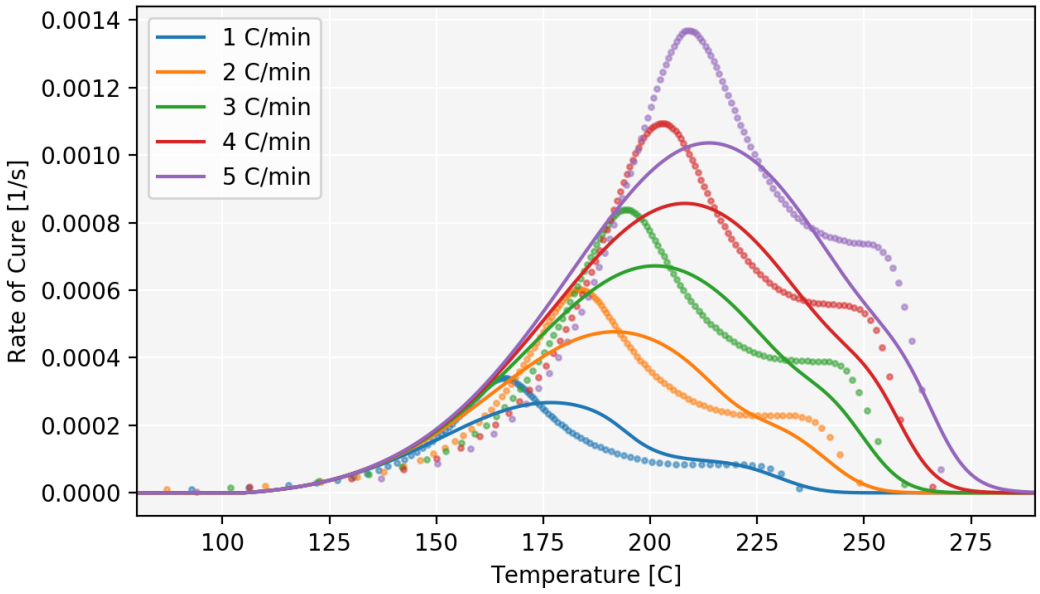

Figure 3. Comparison of parametric model predictions and experimental data for dynamic runs.

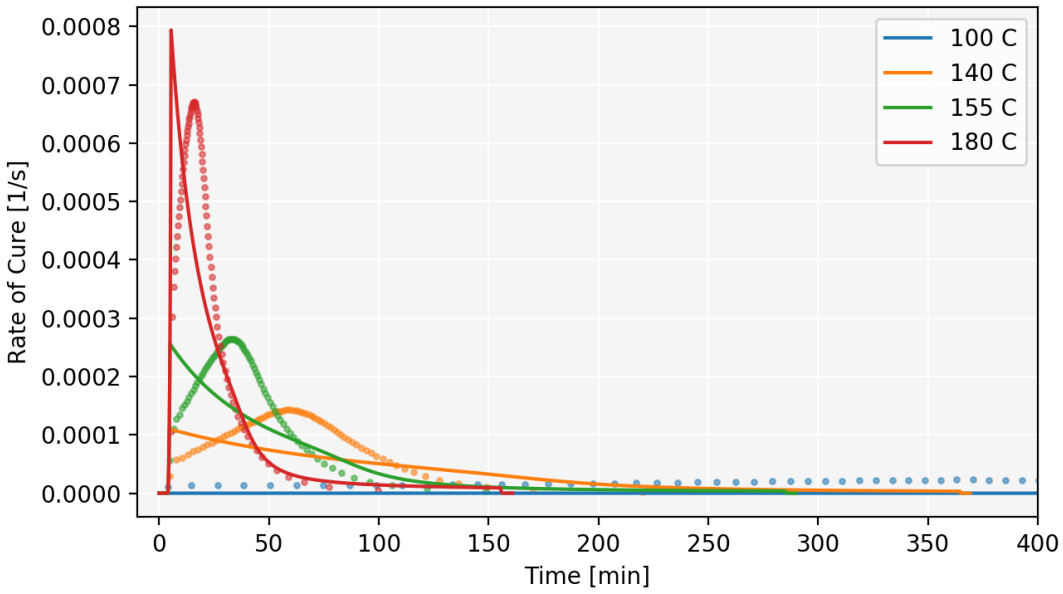

Figure 4. Comparison of parametric model predictions and experimental data for isothermal runs.

4.3 Nonparametric Model

To determine the optimal kernel bandwidth for the GP regression model, which is the only user defined parameter in the model, the average cross-validated RMSE was calculated as a function of the kernel bandwidth, Figure 5.

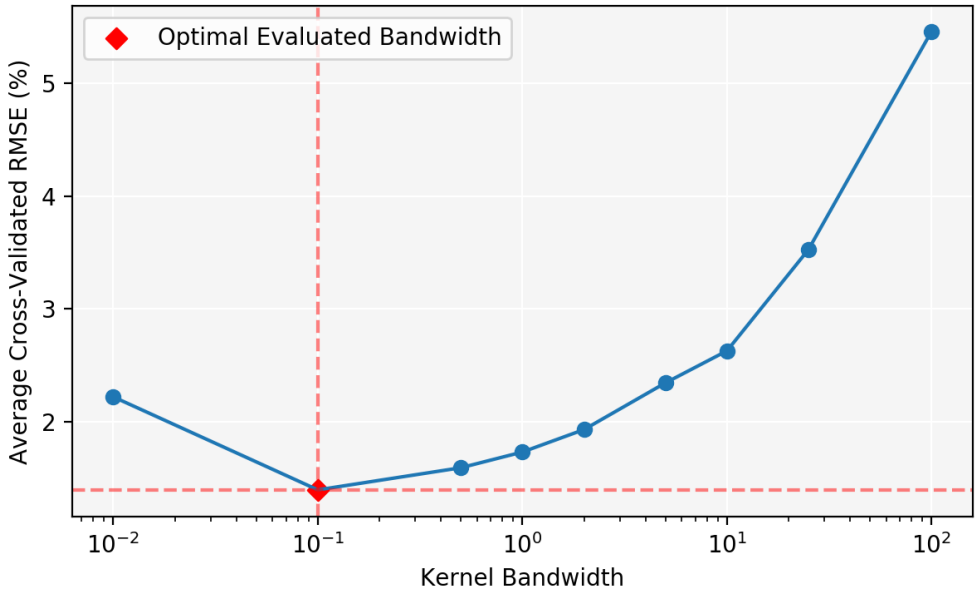

Figure 5. Average cross-validated RMSE as a function of kernel bandwidth.

Note that typically, the "type II maximum likelihood" procedure would have been used to select the kernel bandwidth [7], but cross-validation was used for the sake of simplicity in this paper. The selected bandwidth was slightly greater than the optimal evaluated bandwidth from the k-fold analysis in order to increase model smoothness.

With the optimal bandwidth determined, cure rate predictions using the GP model were performed. Figures 6 and 7 show the predicted cure rate for the dynamic and isothermal runs as well as the 95% credible bands on the mean predictions (shaded regions). Once again, solid lines represent model predictions and scatter points represent experimental data.

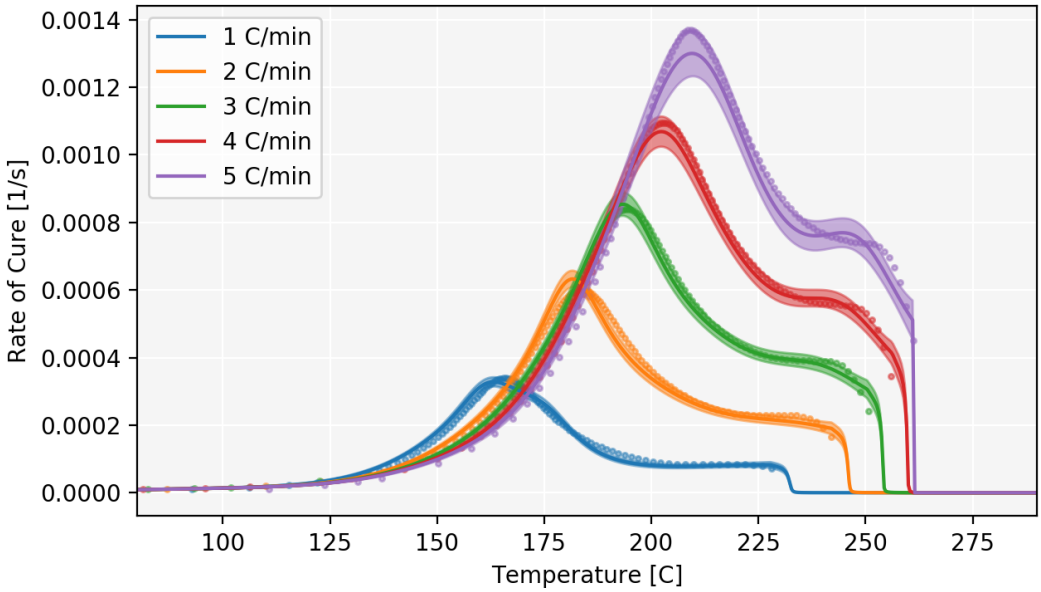

Figure 6. Comparison of GP model predictions and experimental data for dynamic runs.

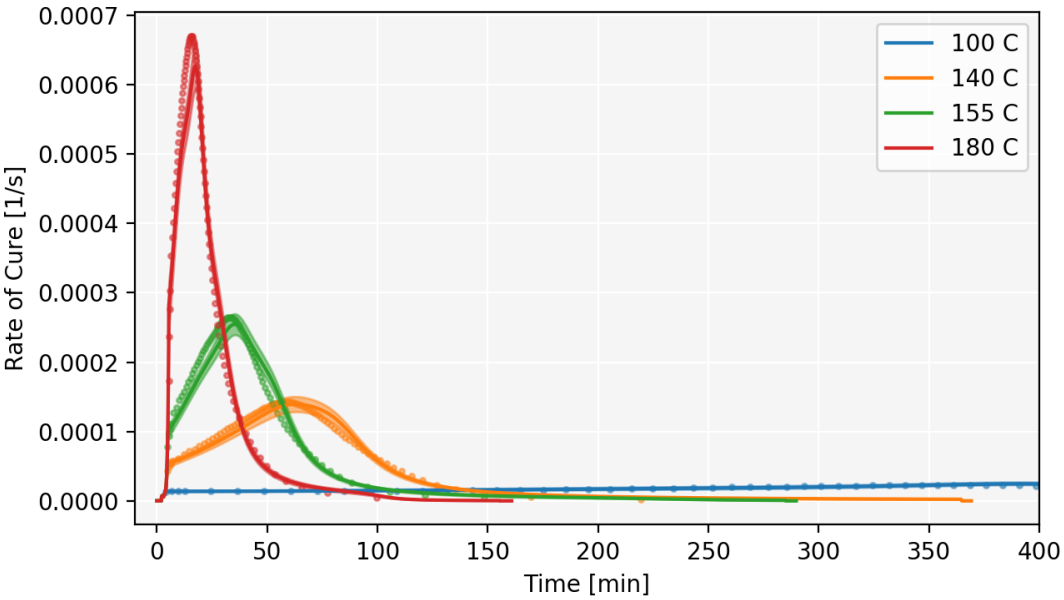

Figure 7. Comparison of GP model predictions and experimental data for isothermal runs.

A quick visual comparison of figures 3 and 4 with figures 6 and 7 shows that the GP model fits the experimental data much better than the parametric model and that the GP model gives realistic measures of confidence in the model predictions with respect to the experimental data. We can also visualize the confidence in the GP model predictions as a function of degree of cure x and temperature T by plotting the 95% credible intervals as contours in the form of a process map [3].

This gives a good understanding of the model uncertainty in different regions of the feature space (x, T). Figures 8, 9 and 10 show the credible intervals for the GP model predictions when trained on to the small, medium and large data sets, respectively. Credible intervals were normalized by their respective model predictions in order to be expressed as a percentage.

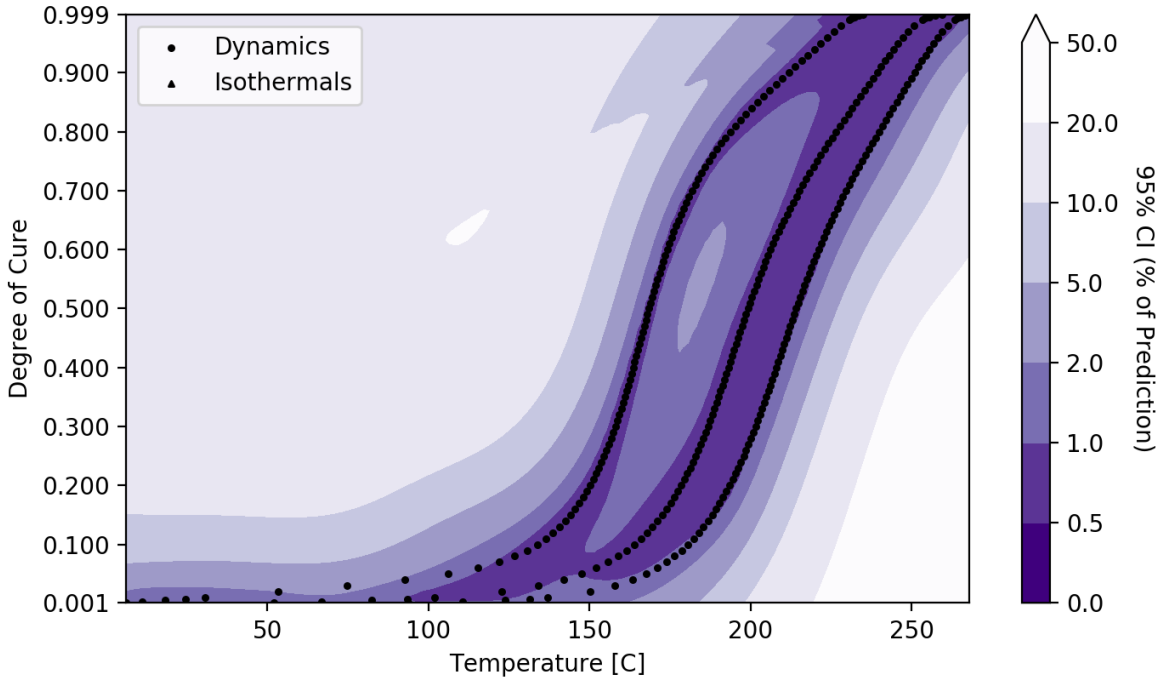

Figure 8. Uncertainty contours for the GP model trained on the small data set.

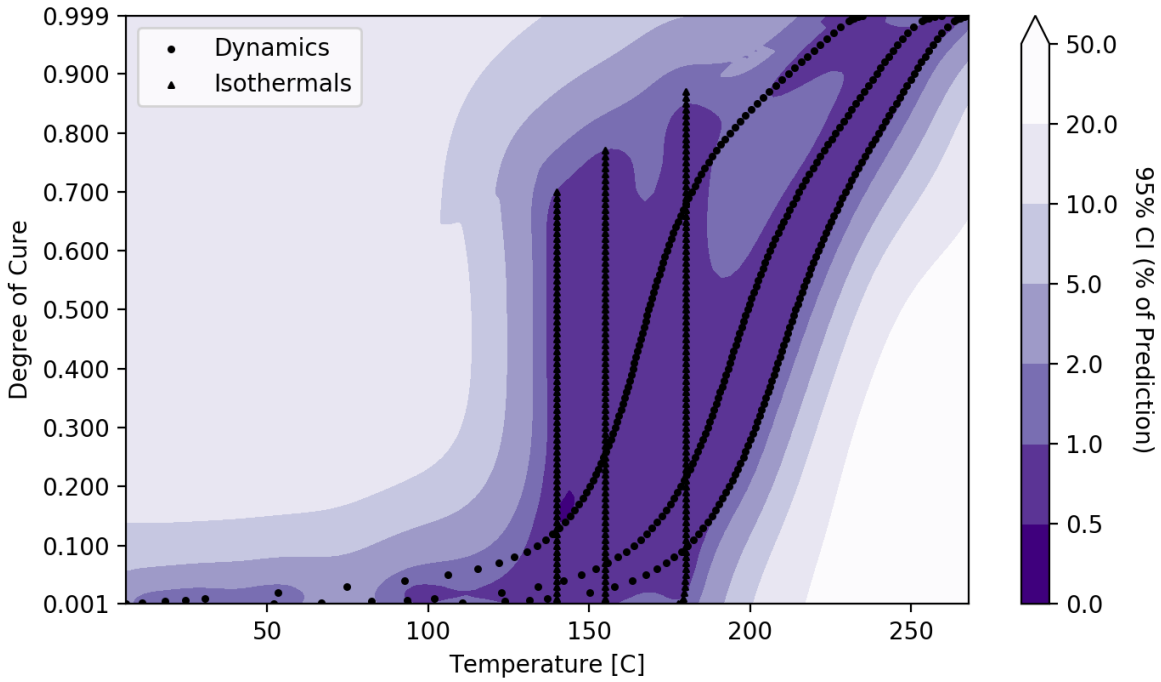

Figure 9. Uncertainty contours for the GP model trained on the medium data set.

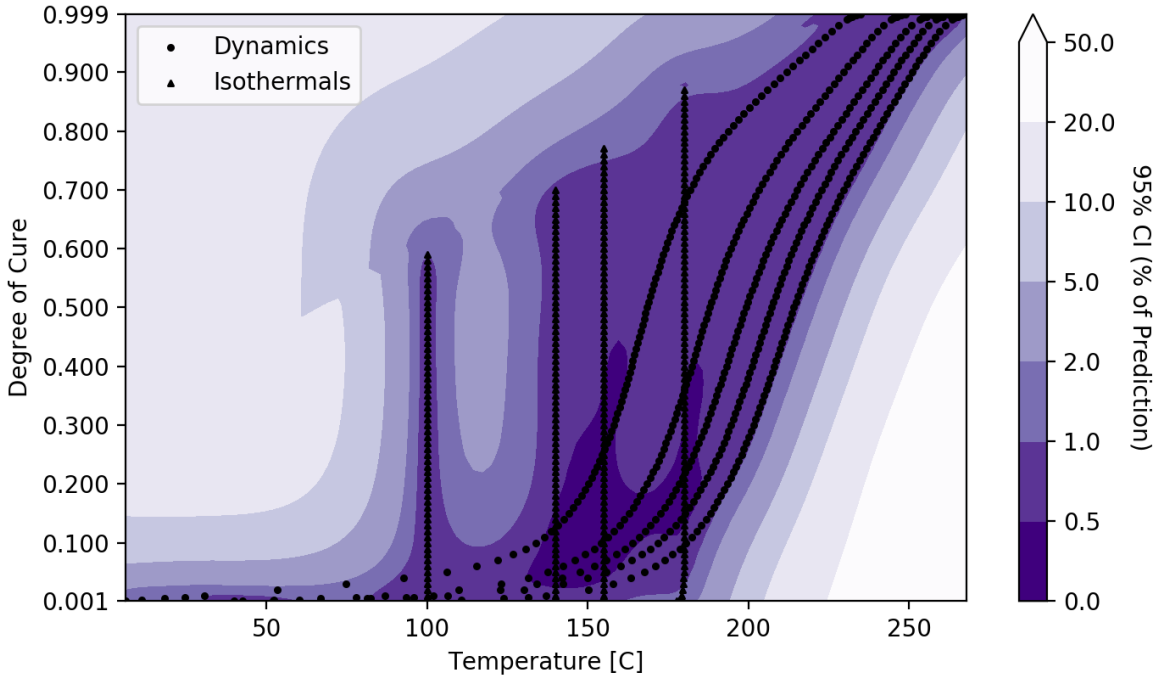

Figure 10. Uncertainty contours for the GP model trained on the large data set.

As seen in Figures 8-10, the more data the model is trained on, the larger the region with low prediction uncertainty. The figures also give a very good indication of which regions more tests should be performed in to reduce prediction uncertainty.

4.4 Comparison of Parametric and Nonparametric Models

To compare the predictive accuracy for the parametric and nonparametric models for the three data sets, the test set RMSE's were calculated and are presented in Figure 11.

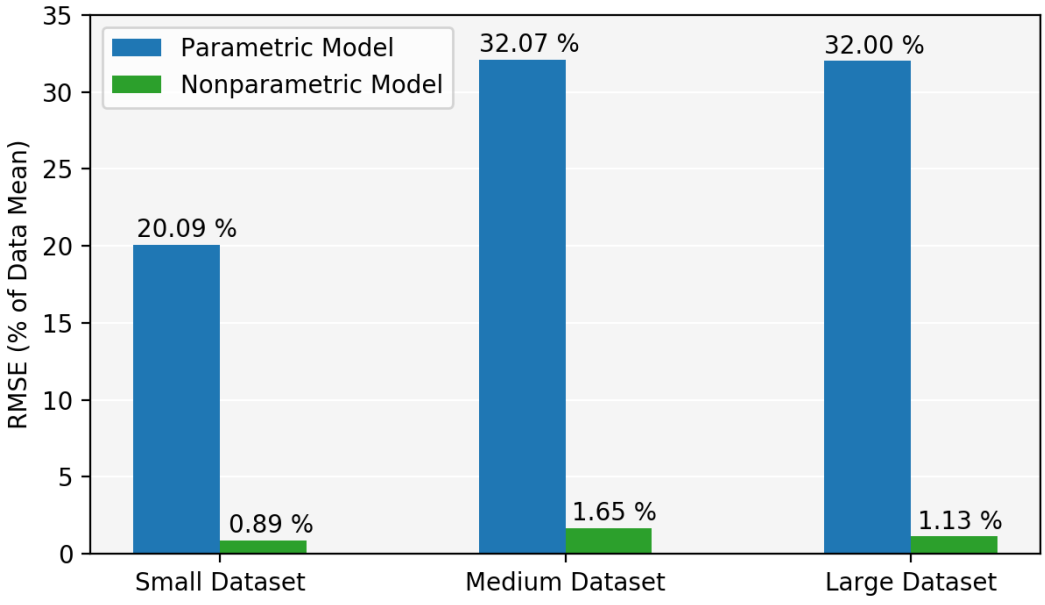

Figure 11. Test set RMSE for parametric and nonparametric models for the three data sets.

Figure 11 shows that the predictive accuracy of the nonparametric model is vastly superior to that of the parametric model that was used in this study. The parametric model has a much lower test RMSE for the small data set compared to the medium and large data sets that included isothermal holds. This is an indication that the functional form use in the parametric model is not well suited to representing the isothermal data, which enters the diffusion regime of the cure reaction. Conversely, the nonparametric model RMSE is relatively insensitive to the type and amount of test data.

5. DISCUSSION

The Gaussian process model greatly outperforms the traditional parametric cure kinetics model used in this work. In addition to having a significantly lower RMSE, the GP model also automatically gives the uncertainty in its model predictions, which is very important for understanding the feature space (x, T) in which the model is reliable. This is not to say that nonparametric cure kinetics models are unequivocally superior to parametric models, and it should be noted that Convergent currently has 34 different parametric models that are used to describe cure kinetics behaviour most of which have far greater flexibility to capture complex behaviour than eq. (3). However, this demonstrates that proper functional form selection is imperative when attempting to fit a parametric model to experimental cure kinetics data. Finding the proper functional form can be difficult for complex resin chemistries, and equations are often highly nonlinear and contain many parameters, making it difficult to find an optimal fit using frequentist methods such as least-squares. Nevertheless, a nonparametric model will only ever be as good as the data it is trained on, and many regions of the process space (x, T) are difficult or impossible to access through experimentation. Physical understanding and intuition are extremely valuable as they can help guide nonparametric models in the absence of quality data and prevent them from making predictions that disobey our best physical understanding in regions with little or no data. A very simple GP prior mean function was used in this paper, but even better results could potentially be achieved by fitting a more detailed and representative parametric prior function which includes information about diffusion and other effects which are difficult or costly to measure experimentally.

There is great value in incorporating statistics into material characterization and process modelling in general. Without using any statistical metrics to evaluate the predictive accuracy and uncertainty of a model, it is impossible to determine whether a model is fit for use. Basic practices like partitioning training and test datasets, computing RMSE and performing k-fold cross-validation makes it possible to understand the strengths and limitations of a model and identify how to improve it through more efficient experimental design. Once uncertainty has been quantified in the inputs to a process model, eq. (1), there are a variety of methods for forward propagation of uncertainty to the outputs, such as Monte Carlo methods. This way, the maximum benefit can be achieved from process simulation and the physical processing of composite parts can be better informed. Figure 12 shows an example of forward propagation of uncertainty through a RAVEN process model to give a 95% credible interval on an exothermic temperature prediction using Monte Carlo methods.

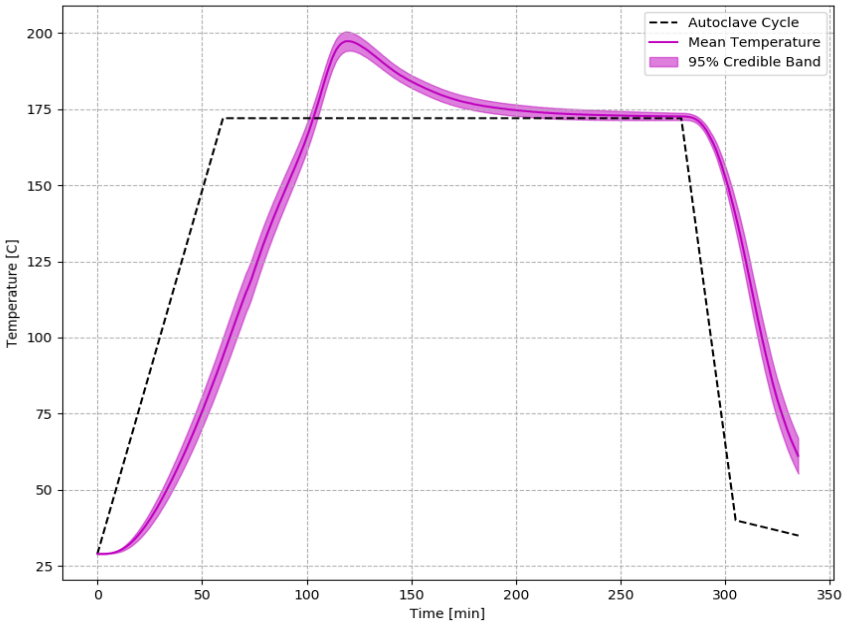

Figure 12. Virtual thermocouple thermal history simulation in RAVEN V4 using forward propagated uncertainties from process model inputs.

6. CONCLUSION

This study showed how to quantify predictive error and uncertainty in composite material models, highlighted some of the problems with current standard approaches to composite materials constitutive modelling and presented some alternative approaches to remedy these problems. If attempting to fit a parametric model to complex material data, it is imperative to select a functional form which sufficiently describes the underlying physics. Nonparametric methods can provide extra flexibility in fitting complex data sets but suffer from low interpretability and don't reflect any physical knowledge or intuition. It is possible to combine the benefits of physical understanding and flexible modelling by fitting a physics-based parametric prior mean function for a Gaussian process to help guide it in regions of the feature space where good quality data is sparse or nonexistent. By incorporating statistical practices into materials characterization and process modelling procedures, it is possible to extract far greater information and benefit than by attempting to do everything deterministically. Uncertainty that is quantified in process simulation inputs can ultimately be propagated to the final output of interest using Monte Carlo methods, benefitting the physical part manufacturing process by providing the best possible virtual support.

7. REFERENCES

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