SPATIAL-TEMPORAL SEMI-EMPIRICAL DYNAMIC MODELLING OF THERMAL GRADIENT CVI PROCESSES

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Abstract: Thermal gradient chemical vapour infiltration (CVI) appears to have much promise as a process to produce carbon/carbon composites and has several advantages over conventional isothermal CVI. Using a graphite heater in the centre of porous disk preforms, Zhao *et al.* (2006) reported excellent densification and no surface preform plugging. The aforementioned work produced a large amount of spatial and dynamic temperature data but falls short of providing a model of temperature over space (r) and time (t). Hence, using the data reported by Zhao *et al.* (2006), this article develops and presents two dynamic models for deposition temperature as a continuous function of t and distance from the heater surface, r. One model uses only two adjustable parameters and the other five and both capture the phenomenological structural behavior quite well. Hence, the approach presented in this work provides a methodology to model real processes using a limited amount of data and produces important continuous dynamic and spatial behaviour that can prove valuable in controlling and optimizing thermal gradient CVI processes.

Keywords: carbon/carbon composites; chemical vapour infiltration; modelling.

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INTRODUCTION

Chemical vapour infiltration is a relatively new process for forming carbon reinforced carbon composites. This is an important process because carbon/carbon composites have been proven to have excellent high-temperature mechanical properties and high thermal stabilities (Li *et al.*, 2005; Zhang and Hüttinger, 2003; Vignoles *et al.*, 2004; Delhaes, 2002; Birakayala and Edward, 2002). This material exhibits high strength, stiffness, toughness, thermal shock, wear-resistance, ablation performance, friction resistance and is also lightweight (Zhang and Hüttinger, 2003). These composites are already being used where high temperatures and friction are an issue such as in brake pads for planes and on space crafts (Birakayala and Edward, 2002); unfortunately though because of high processing times this material is extremely expensive, limiting its possible usage.

The conventional CVI process consists of infiltration of fibre preforms with a carrier gas such as helium and a low molecular weight hydrocarbon such as methane or ethane at constant temperature and pressure. The temperature is usually around 1000°C and the hydrocarbons react rapidly to form larger molecules that deposit into the carbon matrix at a high rate typically plugging the entrance of pores and inhibiting infiltration and thus, densification. A promising alternative to overcome the limitations of the isothermal CVI process is thermal gradient CVI (Tang *et al.*, 2003; Golecki *et al.*, 1995; Probst *et al.*, 1999; Jiang *et al.*, 2002). As the name suggest, thermal gradient CVI seeks to control carbon deposition by controlling the temperature profile in the preform. This is done by creating a higher temperature where deposition is to occur and maintaining a lower temperature where deposition is not to occur. Filling of the pores is controlled by temperature in the zones of the preform. Thermal gradient CVI was demonstrated in Zhao *et al.* (2006) using an electrical furnace consisting of a heated rod between two electrodes with cylindrical donut shaped preforms slid over the rod. Thus, the preforms were heated in the radial direction from the center out. Natural gas was introduced into the reactor from the cold outer surface at a controlled rate and reached the hot deposition zone by diffusion. The deposition zone is controlled by temperature and a thermocouple inserted into the preforms that moves in the radial direction outward as the temperature rises and densification is completed.

This work develops two types of dynamic semi-empirical models of the temperature field of the preform continuously over time and space for the process in Zhao *et al.* (2006) using only the data they reported. Moreover, this article will demonstrate a new model building process for this application and provide theorists and practitioners with a means to obtain accurate spatial and dynamic response surface behaviour. The models give the temperature (T) as a function of time (t) and radial distance from the heater surface (r). The difference between the models is the dynamic form they use. Model 1 uses a third order, critically damped transfer function and depends *only on two* adjustable coefficients. Model 2 uses a sigmoidal transfer function and depends *only on five* adjustable coefficients. Model 2 fits the data better but Model 1 has fewer adjustable coefficients. By 'adjustable coefficients', we mean model parameters that are estimated under least squares estimation.

Our procedure consists of basically three steps. The first step uses temperature versus time data, at fixed values of r, to obtain the dynamic models for temperature. Next we determine models for the initial temperature and model parameters as a function of r. In the final step we incorporate these models for the parameters, which are functions of r, into $T(t,r)$.

We present this work using the following outline: in the following section we use the data in Zhao *et al.* (2006) and develop the dynamic models. The next section describes the model building procedures to obtain initial temperature and the dynamic parameters as a function of r. We present and compare both our final models with the three dimensional surface plot in Zhao *et al.* (2006) in the penultimate section 4 and give concluding remarks in the final section.

DYNAMIC MODELLING

Collecting the Modelling Data

In this section we develop dynamic fits for 11 fixed values of r. The data we used come from figures 5 and 6 in Zhao *et al.* (2006) and are given in Figure 1. The plot on the left gives temperature versus time (t) data at constant values of r and the other one gives temperature versus r data at constant values of t. We used the nine curves in the left plot to obtain T_0 for the values of r shown. We estimated these values by mildly extrapolating to $t = 0$. We added two values of r (80 and 100 mm) using information from the right plot. Their T_0 values were obtained from the $t = 3$ h curve and their dynamic data were obtained from all the curves at their values of r. Note that the initial temperatures across r represent the temperature profile in the radial direction at $t = 0$, where we are assuming that $t = 0$ is the time that the flow of gas was started, i.e., deposition began. Based on the surface temperature plot in Zhao *et al.* (shown later in Figure 7) we used a maximum temperature of 1000°C. Next, we use this information to dynamically model temperature at the 11 values of r for Model 1 first and then Model 2.

Model 1

From the data in Figure 1, for each of the eleven values of r, we obtained a 'best fit' dynamic transfer function for Model 1 using a third order, critically damped transfer function, with a step input. This model form was accepted after evaluating a few low order functions. The Laplace domain form of this transfer function is given by equation (1) below:

$$\hat{T}_r'(s) = \frac{(1000 - T_{r0})}{s(\hat{\tau}_r s + 1)^3} \tag{1}$$

where '^' is used to represent 'estimate'. In the time domain,

$$\hat{T}_r'(t) = \hat{T}_r(t) - T_{r0} \tag{2}$$

where T_{r0} is the initial temperature at a distance r from the heater surface and $\hat{\tau}_r$ is the estimated time constant for the transfer function at a distance r from the heater surface. By applying the method of partial fraction expansion and then transforming to the time domain (see Seborg *et al.*, 2004)

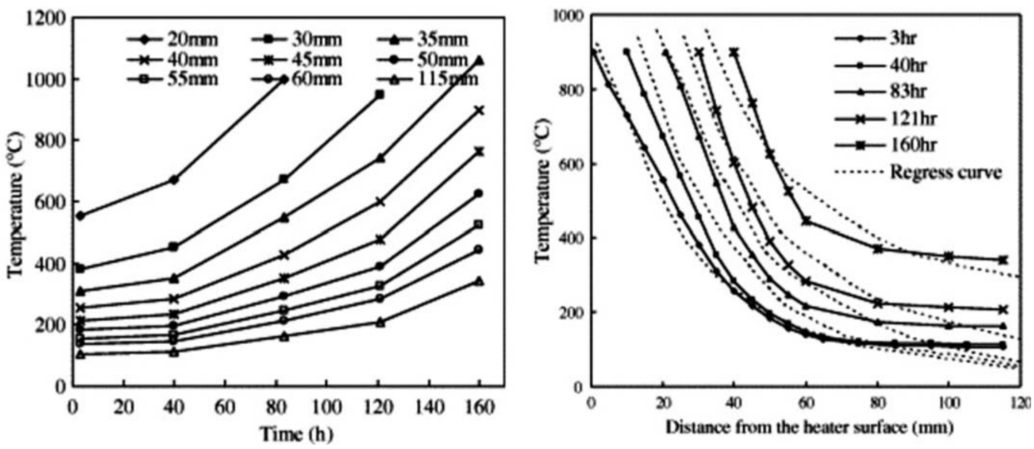

Figure 1. The plots taken from Zhao *et al.* (2006) used to provide the modelling data. The plot on the left gives temperature versus t curves for different values of r and the one on the right give temperature versus r curves for different values of t. The plot on the left was used to obtain T_0 data at each value of r shown except $r = 80$ mm and $r = 100$ mm. These two were estimated using the 3 h curve from the plot on the right.