FORCED FLOW-THERMAL GRADIENT CHEMICAL VAPOR INFILTRATION FOR CARBON-CARBON COMPOSITES: MODELING AND EXPERIMENT

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INTRODUCTION

Current commercial chemical vapor infiltration (CVI) for carbon-carbon composites uses the isothermal process. This approach requires long processing times and is restricted to thin components. These limitations can be removed by using the forced flow-thermal gradient (FCVI) process. This approach has been successful for the related technology of ceramic matrix composites (CMC's) where it has produced thick composite components with uniform density in less than ten hours[1]. Recent work at Georgia Tech has demonstrated that similar gains are possible for carbon CVI[2].

The development of FCVI has benefitted from parallel development of a process model[3]. This model not only has enhanced understanding of the process and guided experimental investigations during the process development program, but also provides a design tool for future applications of the process to large, complex components.

EXPERIMENTAL

The FCVI process was used to fabricate a carbon-carbon composite in the shape of a right circular disk. A reagent gas mixture was forced to flow through a 0.85 cm thick, 4.8 cm diameter fiber preform in a resistively heated, hot wall furnace. The carbon matrix was deposited from a gas mixture of propylene and hydrogen. The preform consisted of layers of square weave cloth using T-300 3K tow, with alternating 0/30/60° orientation between the layers and 57 vol% fiber in the final preform. Cooling of the gas inlet produced a temperature gradient through the thickness of the disk of 300-350°C prior to densification.

Eight infiltration runs were completed representing a two level-three factor designed experiment. Process factors included the temperature on the cool side of the preform, the propylene concentration and the overall gas flow rate. For each run the process was terminated when the inlet pressure reached 40 kPa above ambient.

Infiltration time and final density (Table I) ranged from 28.5 to 2.8 hrs and 1.62 to 1.71 g/cm^2 respectively. Additional experimental details and characterization results are reported elsewhere[2].

Table I. Results of carbon FCVI designed experiment.

run ID	reagent conc. (v%)	bottom temp. (C)	gas flow (sccm)	run time (hrs)	bulk density (g/cm^2)
(1)	25	850	200	28.5	1.68
A	25	950	200	11.5	1.71
B	50	850	200	9.0	1.67
AB	50	950	200	6.0	1.62
C	25	850	400	21.5	1.68
AC	25	950	400	7.0	1.68
BC	50	850	400	7.8	1.66
ABC	50	950	400	2.8	1.64

MODEL DEVELOPMENT

The experimental runs were modeled using GTCVI, a "finite volume" model developed at Georgia Tech to assist optimization of the CVI process for ceramic matrix composites[3]. In this model the infiltration system, including the preform, furnace and associated fixturing, is dividing into a mesh of orthogonal, 3-D volume elements. Setting the boundary conditions to match process parameters, a steady-state solution to the coupled equations for mass and heat transfer is obtained, yielding a matrix deposition rate for each volume element. The resulting increase in density is calculated with successive time steps.

Successful modeling of the FCVI process depends on reasonable estimation of the carbon deposition kinetics and of the transport and microstructure properties of the preform and partially densified composite. For carbon deposition we assume a first-order, Arrhenius rate expression, consistent with other studies of carbon deposition from propylene[4]. Values for the rate constant and activation energy were obtained from an infiltration run using periodic deposition of SiC layers to "mark" the deposit thickness at various times and positions[5].

The gas permeability and the internal pore surface area of the preform and partially densified composite were estimated from the filament diameter, strand filament count and weave and stacking dimensions using a percolation model[6]. In the absence of experimental data for this composite, we made a rather arbitrary estimate for the thermal conductivity assuming values of 0.01 and 0.08 W/cmK at 50% and 10% porosity and interpolating between these values with an inverse linear function.

Eight model runs were performed approximating the conditions in Table I. In the model it is more convenient to set the furnace temperature, and values of 1250 and 1350°C were found to give approximately 850 and 950°C at the bottom (inlet side) of the preform. The run was complete at an inlet pressure of 100 kPa above ambient.

RESULTS AND DISCUSSION

The results of the model runs are shown in Table II. Overall they match experimental results well, as seen in Figure 1. Further, the model offers insight into the infiltration process by showing the progress of densification within the preform (Figure 2). Initially, the density in a particular area shows a rapid increase but slows after reaching approximately 85% full density. At this density the porosity within the tows is filled and subsequent densification involves deposition in the larger pores between tows and between cloth layers. The densification rate near the hot face is higher than that near the cool face but the difference between these narrows near the end of the run.

With this simple geometry and within this narrow range of process conditions, a complex, 3-D finite volume model may not be needed to identify optimum process conditions. However, validation of model parameters with these experiments will allow application of the model to more complex geometries where choices concerning gas inlet and outlet placement and imposed thermal gradients greatly increase.

Table II. Results of carbon FCVI model runs.

run ID	reagent conc. (v%)	furnace temp. (C)	gas flow (sccm)	run time (hrs)	fraction dense (%)
(1)	25	1250	200	25.0	88.8
A	25	1350	200	13.6	88.9
B	50	1250	200	12.0	88.8
AB	50	1350	200	6.7	88.7
C	25	1250	400	20.6	88.3
AC	25	1350	400	11.1	88.5
BC	50	1250	400	10.1	88.6
ABC	50	1350	400	5.3	88.6

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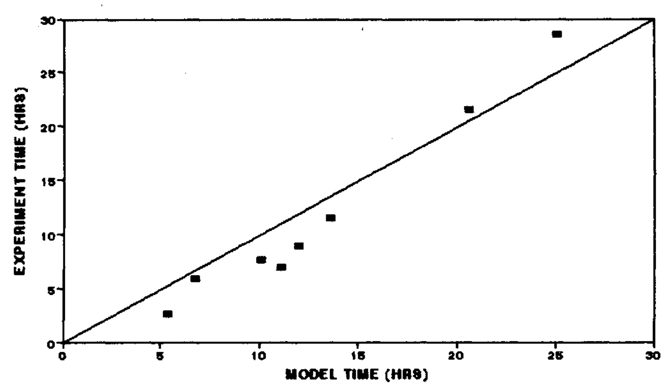

Figure 1. Model run time matches experiment.

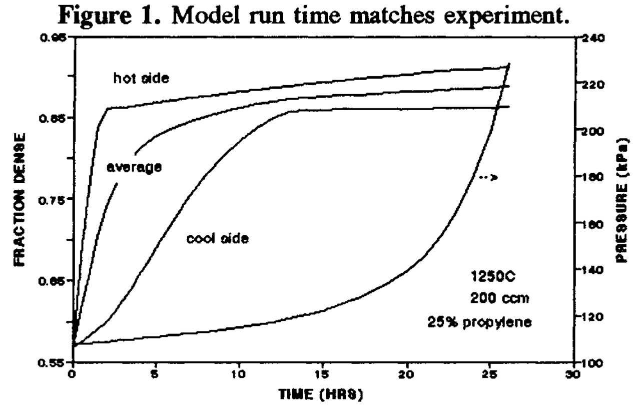

Figure 2. Density and inlet pressure increase during run.