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SCALE-UP AND MODELING OF FORCED CHEMICAL VAPOR INFILTRATION

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ABSTRACT

The forced flow-thermal gradient chemical vapor infiltration (FCVI) process has been scaled-up from a maximum of 7.6 cm dia. disk to fabrication of 24 cm dia. disks, 1.6 cm in thickness. The components are turbine rotor subelements produced from polar weave Tyranno fibers preforms. The disks were subjected to non-destructive testing and spin-tested to high rpm. The processing conditions were modeled with the GTCVI code to aid in optimization.

INTRODUCTION

The past decade has seen the development of the forced flow-thermal gradient chemical vapor infiltration (FCVI) process as an alternative to isothermal-isobaric chemical vapor infiltration (ICVI) to meet differing structural composite requirements. Whereas ICVI can efficiently produce thin-wall, complex-shape parts, it has significant difficulty producing thicker-wall materials (> 3 mm). Using ICVI for thicker components requires many weeks to months of infiltration time, interrupted by periodic machining of the surfaces to open gas pathways that have become sealed due to overcoating. The FCVI of thicker sections can be effected in a single step, however, and at sufficiently high rates to require tens of hours or less.

In ICVI the reactant gases are allowed to flow around the preform and diffuse into the inner volume. In FCVI the reactant stream is forced to flow through the preform, thus only depending on diffusion to reach the finest porosity within, for example, fiber bundles. The imposition of the thermal gradient prevents

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premature sealing of the gas entrance surface, thus obviating the need for periodic machining. Details of the processes have been extensively described elsewhere.¹⁻⁶

The objective of the reported work was to scale-up the Oak Ridge National Laboratory (ORNL) FCVI technique to demonstrate fabrication of components approaching 30 cm in width and 2.5 cm in thickness. Previously, systems were available with capability to produce materials with a diameter of only 7.6 cm. The effort was designed as a demonstration of the ease of scaling the process and to provide for a comparison of the produced components with those prepared by other means.

This paper is an account of the scale-up FCVI unit specifications and the resultant infiltration system that was manufactured. The preform selected for the demonstration, a turbine rotor subelement, is described along with the results of infiltration, including non-destructive evaluation of the components. The fibers used were the SiC-based Tyranno™ (Ube Industries, Tokyo, Japan) and the matrix was SiC. A computer model for the process, GTCVI,⁷ was used to predict densification progress. Finally, the infiltrated preforms were subjected to spin-testing to determine their strength.

SCALED-UP FCVI FACILITY

Specifications

The specifications for the scale-up FCVI unit included requirements such as hot and cool surface temperatures and their control, minimal radial gradients, instrumentation, and exhaust gas cleaning. These are indicated below:

Heating: Hot surface minimum temperature of 1500°C
Radial gradients < 10°C
Variable cooling of cool surface

Fixturing: Hold preform shape
Capacity to 30 cm diameter x 2.4 cm thickness
Exclude reactants from heating elements

Instrumentation: Measure diametral temperature profile
 Meter reactant flow
 Measure reactant inlet pressure
 Read power to heating elements
 Provide for automated ramp to temperature and shut-down
 Computer interface for data acquisition and control

Exhaust gas: Recirculating caustic scrubber
 Explosive gas dilution

FCVI System

The furnace and power supply components of the FCVI system were designed and constructed by Advanced Vacuum Systems of Ayer, MA. Figure 1 is a schematic of the furnace with the important features indicated. The furnace shell and lid are water-cooled. Access is gained through the top lid which rises on pneumatic lifters. The inside of the lid contains two concentric heating elements which are independently controlled, and therefore allow control of the radial temperature gradient. Also in the lid are two pneumatic hold-down pins, which press down on the preform fixture to compress the preform and seal the fixture against reactant gas leakage into the furnace cavity. The reactant gas inlet fits into the bottom of the fixture and the exhaust exits the fixture through a graphite tube that mates with a metal exhaust line. Graphite foil is used as gasket material in the fixture and to help seal the gas entrance and exhaust tubes.

Figure 1. FCVI furnace schematic for large-scale component fabrication

The bottom surface can be cooled with either a water-cooled plate or with forced air blown through annuli below the furnace. A baffle is fixed such that the flow of air can be directed to varying degrees into the inner or outer annulus, thus allowing control over the lower surface radial gradient. Temperature measurement is via thermocouples that are emplaced above and below the preform fixture (six each). Temperatures are maintained utilizing control thermocouples on the hot side (controlling power to the heating elements) and on the cool side (controlling positioning of the air flow baffle).

The remaining system components were assembled on site. Reactant flow control is via a Tylan Source V vapor source for methyltrichlorosilane (MTS) and MKS mass flow meters for hydrogen. Inlet reactant stream pressure is measured with a MKS Baratron. A caustic wet scrubber supplied by Vanair was used for neutralizing the exhaust gas, which was previously diluted with air below the lower explosive limit for hydrogen. Figure 2 is a photograph of the system together with two infiltrated disks.

Figure 2. Photograph of the FCVI furnace with two infiltrated disks.

MODELING

GTCVI was used to simulate densification of the spiral weave disk using the saced-up FCVI facility. Figure 3 shows the model grid consisting of 15x25 elements where the preform itself consists of a 5x12 element grid. In this configuration the center hole of the preform is open except that the top and bottom plates are solid preventing direct flow into or out of this region. The overall gas flow pattern in the preform is shown also. Gas flow near the outer perimeter is significantly less than near the center.

Figure 3. The arrow size and direction indicate the magnitude and direction of the gas flow. This image is for the unfiltrated preform.

Figure 4 shows the overall progress of densification using three sets of infiltration conditions. For a preform with 40% fiber volume fraction, increasing the MTS flow rate from 3 g/min to 6 g/min reduces the residual porosity after 30 hours from 20% to 10%. Actual conditions used for the experimental runs at ORNL more closely match the model run for a preform with 30% fiber and a 5 g/min MTS flow rate. These model results indicate that modification of the reagent injection system to allow higher MTS flow rates is needed to achieve higher densification rates.

Figure 4. The CVI model shows densification for different MTS flow rates and 30% (open squares) and 40% (filled squares) fiber loading.

The model also shows the densification profile at intermediate times during processing. As seen in Figure 5, density increases more rapidly near the center of the disk, but after 40 hours is relatively uniform, ranging from 80 to 90% of full density.

PREFORMS

The preforms used for demonstrating scale-up were constructed of a modified polar weave fabric developed for the U.S. Air Force Wright Laboratory "High Temperature, Non-Metallic Turbine Engine Evaluation" program under Allied-Signal Corp. (Phoenix, AZ) by Textile Technologies Inc. (Hatboro, PA). The component is the JETC Phase III CMC compressor subelement, which is 24.6 cm in diameter and 1.27 cm thick, with a 1.27 cm diameter central hole. Since the original program preforms had glass matrix prepreg added to the fibers before weaving, the satin weave that was used in that study provided adequate fiber density. However, the FCVI preforms required no prepreg, and thus the resulting satin weave did not have adequate fiber loading. The weave was modified to a plain weave, which resulted in satisfactory preforms. The resultant fiber loading was ~33%.

Figure 5. Two plots show the density profile after 18 hours (top) and after 40 hours of infiltration (bottom).

After removing the sizing by heating in the furnace, ~ 200 nm of pyrolytic carbon was applied to the fibers of the preform. This was accomplished by

placing the preform in the FCVI furnace and holding the top surface temperature at 1100°C, the pressure at 6.6 Pa, and flowing propylene diluted in argon through the preform. After two hours the furnace was shut-down and the preform turned over. The process was then repeated. This ensured that the coating was applied uniformly throughout the preform. Weight gain measurements confirmed the coating thickness.

DENSIFICATION

The preforms were infiltrated utilizing constant flows of 5 g/min MTS and 5 l/min H₂, and a hot surface temperature of 1200°C. The results agree well with those predicted by the GTCVI model (Table I).

Table I. Results of the infiltration runs.

Spec. No.	No. Turns	Thickness (cm)	Run Time (h)	Void Fraction
794	50	1.59	40	16.5%
795	50	1.59	40	16.4%
806	40	1.27	40	13.6%

NON-DESTRUCTIVE EVALUATION

The densified preforms were examined utilizing digital radiography and x-ray computerized tomography. Figure 6 is a digital radiograph of specimen no. 806 which shows relatively uniform densification. The radial striations correspond to channels in the bottom surface of the holder which are passages for the reactant gas. In addition, the radial density variation appears to correspond to variations in fiber loading. This is a natural result of the polar weave, which requires the initiation of new radial fiber tows with increasing distance from the center.

Figure 7 is a x-ray computerized tomographic image of a diametral section of specimen no. 806. Some nonuniformity in density can be seen, which is attributable to the internal puckering of the weave. The edges of the disk also appeared to be slightly folded upwards, due to somewhat too tight a fit in the holder. Results similar to those for no. 806 were observed for nos. 794 and 795.

Figure 6. Digital radiograph of specimen number 806.

Figure 7. X-ray computed tomographic image of a diametral section of specimen number 806.

SPIN TESTING

The results of spin testing performed at Williams International, Inc., Walled Lake, Michigan were inconclusive due to eccentricities in the densified preforms. Specimen no. 795 was spun up to 21,080 rpm and became unstable, causing shearing of the mounting quill. The equivalent stress was 193 MPa. The disk struck the wall of the spin test pit, yet remained intact. A second test was performed with the same disk which again sheared the quill at 20,850 rpm, indicating a destructive harmonic at that speed. This time upon striking the wall the disk delaminated (it struck the edge of the groove left in the lead wall by the first test).

Specimen no. 806 was designed to overcome the test problems encountered with no. 795. The disk was made thinner to allow the test rig to overcome any imbalance. In addition, the holes in the fixture top plate were made smaller to reduce puckering in the cloth. This specimen, however, proved to have an eccentric placement of the center hole which caused the disk to encounter a critical point at 14,700 rpm, stalling the apparatus. The disk was intact, but only tested to 70.2 MPa.

CONCLUSIONS

It has been demonstrated that the FCVI process can be relatively easily scaled by a factor of at least three. Polar weave disks were successfully infiltrated to significant densities within ~40 h. The process, however, has yet to be optimized in the new apparatus, or even for the polar weave preforms, therefore it is likely that significant improvements can still be obtained in densification times and ultimate densities. The 20% residual porosity after 40 hours predicted by the GTCVI model is in good agreement with experimental results. The GTCVI model also proved useful in indicating appropriate flows and temperatures for these initial runs. Nondestructive evaluations indicated some nonuniformities in densification, largely resulting from puckering and other nonuniformities in the woven preform. The spin testing results are encouraging, with a maximum stress of 193 MPa measured without failure of the component. The results of this effort indicate further scale-up of FCVI and process commercialization have a good chance of success.

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REFERENCES

1. E. Fitzner and R. Gadow, "Fiber-Reinforced SiC," *Ceram. Bull.* **65** [2] 326-35 (1986).
2. R. Naslain, "Two-Dimensional SiC/SiC Composites Processed According to the Isobaric-Isothermal Chemical Vapor Deposition Gas Phase Route," *J. Alloys and Compounds* **188** 42-48 (1992).
3. J.Y. Rossignol, F. Langlais, and R. Naslain, "Tentative Modelization of Titanium Carbide C.V.I. Within the Pore Network of Two-Dimensional Carbon-Carbon Composite Preforms," pp. 596-614 in *Proc. 9th Int'l. Conf. CVD*, eds. McD. Robinson et al., The Electrochemical Society, Pennington, New Jersey (1984).
4. D.P. Stinton, A. J. Caputo, and R.A. Lowden, "Synthesis of Fiber-Reinforced SiC Composites by Chemical Vapor Infiltration," *Ceram. Bull.* **65** [2] 347-50 (1986).
5. T.M. Besmann, B.W. Sheldon, R.A. Lowden, and D.P. Stinton, "Chemical Vapor Infiltration," *Science* **253** 1104-1109 (1991).
6. T.D. Gulden, J.L. Kaae, K.P. Norton, and L.D. Thompson, "Forced-Flow Thermal-Gradient Chemical Vapor Infiltration (CVI) of Ceramic Matrix Composites," pp. 546-552 in *Proc. 11th Int'l. Conf. CVD*, eds. G.W. Cullen and K.E. Spear, The Electrochemical Society, Pennington, New Jersey (1990).
7. T. Starr, A. Smith, T. Besmann, J. McLaughlin, and B. Sheldon, "Modeling of Chemical Vapor Deposition for Composite Fabrication," pp. 231-240, in *Proceedings of the Conference on High Temperature Ceramic Matrix Composites*, eds. R. Naslain, J. Lamon, and D. Doumeingts, Woodhead Pub. Ltd., Cambridge (1993).

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