INTERIM REPORT

FINITE-VOLUME MODEL FOR CHEMICAL VAPOR INFILTRATION
INCORPORATING RADIANT HEAT TRANSFER

A.W. Smith and T.L. Starr

May 1995

Report prepared by
School of Materials Science and Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0245
under
Subcontract 19X-55901
for
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831
managed by
Martin Marietta Energy Systems
for the
U.S. Department of Energy
under Contract No. DE-AC05-84R21400

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER
DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assume any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
A FINITE-VOLUME MODEL FOR CHEMICAL VAPOR INFILTRATION
INCORPORATING RADIANT HEAT TRANSFER

A.W. Smith and T.L. Starr

May 1995

Research sponsored by the U.S. Department of Energy,
Fossil Energy
Advanced Research and Technology Development Materials Program
Report prepared by
School of Materials Science and Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0245
under
Subcontract 19X-55901
for
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831
managed by
Martin Marietta Energy Systems
for the
U.S. Department of Energy
under Contract No. DE-AC05-84R21400
# TABLE OF CONTENTS

ABSTRACT .......................................................... 1

INTRODUCTION ...................................................... 1

MODEL DEVELOPMENT ............................................ 3

CVI MODELING .................................................... 8

CONCLUSIONS ...................................................... 13
LIST OF FIGURES

Figure 1. Infiltration system at Oak Ridge National Laboratory is designed for tube shapes. .................................................. 4
Figure 2. Simulation domain for finite volume model includes gas spaces and solid materials. .............................................. 4
Figure 3. Temperature profile with no radiative heat transfer. .................... 10
Figure 4. Temperature profile with radiative heat transfer. ......................... 10
Figure 5. Temperature profile matches experiment only when radiative heat transfer is included. ........................................... 11
Figure 6. ID and OD temperatures change with densification. Using an "effective" conductivity, the temperatures diverge from the correct radiation-included result. ........................................... 12
Figure 7. ID and OD densities increase at different rates. Density increase with "effective" thermal conductivity does not match that with radiation included. . . . 12
ABSTRACT

Most finite-volume thermal models account for the diffusion and convection of heat and may include volume heating. However, for certain simulation geometries, a large percentage of heat flux is due to thermal radiation. In this paper a finite-volume computational procedure for the simulation of heat transfer by conduction, convection and radiation in three dimensional complex enclosures is developed. The radiant heat transfer is included as a source term in each volume element which is derived by Monte Carlo ray tracing from all possible radiating and absorbing faces.

The importance of radiative heat transfer is illustrated in the modeling of chemical vapor infiltration (CVI) of tubes. The temperature profile through the tube preform matches experimental measurements only when radiation is included. An alternative, empirical approach using an "effective" thermal conductivity for the gas space can match the initial temperature profile but does not match temperature changes that occur during preform densification.

INTRODUCTION

The chemical vapor infiltration (CVI) process is used to produce high quality composites with ceramic and carbon matrices. In some cases a thermal gradient is maintained through the composite preform in order to control the densification rate and to avoid premature closing of gas transport to the preform interior. This forced flow/thermal gradient method (FCVI) offers dramatic improvement in processing times.

In certain system configurations the preform is not in direct physical contact with the heating surfaces of the furnace and heat is transferred primarily by thermal radiation. Previous models for CVI adequately account for only the conduction and convection components, although radiation can be approximated using a high, "effective" conductivity for the gas space. Since this approach is highly empirical and will not work for non-simple geometries, a need exists for a model which provides a general and consistent formulation of the radiant heat transfer between the radiating surface of the furnace and the absorbing surfaces of the preform.

Research sponsored by the U.S. Department of Energy, Fossil Energy Advanced Research and Technology Development Materials Program, DOE/FE AA 15 10 10 0, Work Breakdown Structure Element GT-1(A)
Radiation heat transfer is one of the dominant heat transfer mechanisms in a variety of practical engineering problems. The physical rules and mathematical formulas of radiative exchange between "gray" surfaces separated by non-participating media are well established. However, modeling thermal radiation by discretization methods such as finite-volume, finite-difference, or finite-element is difficult. The interaction of the radiating surfaces separated by non-participating media destroys the tri-diagonal or banded nature of the matrices created by the normal discretization methods. The temperature at one node within the simulation domain does not depend solely on the nearest neighboring nodes. Nodal temperatures at radiating surfaces will depend upon the amount of energy absorbed from the other radiating surfaces, as well as the energy emitted from the node itself. This effectively couples the nodal temperature to all of the other radiating and absorbing nodes.

Another problem with modeling thermal radiation is the large number of radiating surfaces created by the discretization of the simulation domain. The calculation of the visual interaction (view factors) between these surfaces is time consuming. Fortunately, once the view factors have been calculated for a particular geometry, they remain invariant as long as the geometry remains unchanged.

Several approaches have been proposed to circumvent the problems mentioned. An "effective" thermal conductivity may be assigned to the non-participating medium. Heat is then transferred as a diffusion process. The advantage of this approach is that it maintains the matrix characteristics of the original discretization technique. It also eliminates the need to calculate the view factors. This approach can be successful for simple geometries where the "effective" thermal conductivity can be calculated from the problem dimensions, conductances of other regions of the domain and the boundary conditions, or can be established empirically. However, it would not be adequate for more complex geometries where there are different path lengths between different pairs of surfaces and in which shading of radiating/absorbing surfaces occurs. This technique may also fail in cases where the temperature of the non-participating medium does not depend upon the temperature of the radiating/absorbing surfaces. An example of this would be two radiating/absorbing surfaces separated by a flowing gas whose temperature is dominated by convection.

Another approach is to separate the simulation domain into sub-regions in which analytic solutions of the radiant heat transfer may be calculated. These analytic solutions are then added as source terms to the heat transfer equation. This method also maintains the matrix
characteristics of the original discretization technique. However, it requires that the user supply an analytic calculation of the radiant energy exchange among the participating surfaces. For non-simple geometry analytic solutions may not exist.

The characteristics of a successful simulation program for modeling thermal radiation include maintaining the matrix characteristics of the original discretization technique, eliminating the need to define a non-physical, "effective" thermal conductivity, no need for prior knowledge of the radiant energy exchange within the simulation domain. Finally, it should not add a large computational cost to the solution of the overall problem.

The objective of this paper is twofold: First, introduce a method of including the radiant energy in the modeling of the heat transfer equation. The second objective is to investigate the effect of using the radiant heat transfer in the modeling of the chemical vapor infiltration process.

MODEL DEVELOPMENT

An example of a FCVI system to be modeled is shown in Figure 1. This system is designed to fabricate tube shapes by densifying a woven cloth preform wrapped around a perforated mandrel. Reactant gas is forced from the center of the tube outward. A thermal gradient is maintained by cooling the center of the tube while heating the outside of the preform in a cylindrical heating element.

The modeling of CVI involves mathematical description of transport and reaction phenomena within a simulation domain. The simulation domain for the tube CVI system, shown in Figure 2, includes the fibrous preform, graphite fixturing, a perforated graphite mandrel, and open gas space. The open spaces include the gas injection system and the area between the preform and the heating element. The processes to be modeled include heat transfer due to conduction, convection and radiation, transport and reaction of gaseous reactant species, and pressure-driven flow of the gas. Differential equations representing these phenomena can be written in the following steady-state form:

\[ \nabla (\rho \phi u) = \nabla \cdot (\Gamma \nabla \phi) + S \]  

where \( \phi \) is temperature, pressure or concentration, \( u \) is the gas velocity, \( \rho \) and \( \Gamma \) are constants and \( S \) is a source term. Using the finite volume method of Patankar\(^4\), the discretized version of this equation is solved over the simulation domain which is divided into control volumes.
Figure 1. Infiltration system at Oak Ridge National Laboratory is designed for tube shapes.

Figure 2. Simulation domain for finite volume model includes gas spaces and solid materials.

Heat Transfer

The heat transfer equation contains both diffusion and convection components, and a source term:

\[ \nabla (C_p u T) = \nabla \cdot (K \nabla T) + S \]

(2)

where \( C_p \) is the heat capacity of the flowing gas and \( K \) is the thermal conductivity of the material. The source term \( S \) contains any heat generated or absorbed by the volume element, such as heat from chemical reaction or from absorption of microwave energy. This source term also will be used to account for the thermal radiation.
Calculation of the diffusive and convective contributions to the heat balance for each volume element is straightforward given the flow rate and heat capacity of the gas, the thermal conductivities of the materials, and the thermal boundary conditions. The heat flux terms for each volume element depend only on these quantities and on the temperatures of the adjoining volume elements. Since the radiation contribution may depend on the temperatures of more distant volume elements, these cannot be included as flux terms in the same manner. Instead, these are calculated and included as a source term.

To attain this goal a ray tracing program is coupled to the main finite-volume program as a pre-processing step. Based on the discretization of the simulation domain the ray tracing program calculates the view factors of the control volume surfaces. The view factors are then stored for later use. During the solution of the heat transfer equation the view factors are used to calculate the energy exchange between the radiating surfaces by standard formulas based on the nodal temperatures. The radiant energy is then added as a source term to the heat flow equation, this maintains the original matrix structure. To obtain a self-consistent solution several iterations of the heat flow equation are required to incorporate the non-linear behavior of the radiant energy exchange.

Ray Tracing

The first step in the calculation of heat transfer by thermal radiation is to identify all of the control volumes which represent a solid and have a face bordering on the gas phase. Since more than one face of a control volume may be adjacent to the gas, the list of radiating surfaces may contain the same element more than once. The list also includes boundary elements which represent the gas phase. Cycling through the list of surfaces, the view factors between the radiating planes are calculated. The area \( A_i \) of one radiating face is subdivided into 25 blocks of equal area. From the center of each of these areas several hundred rays of random orientation are traced. Note that the faces of the adjoining volume element describe a constrained volume for the ray; i.e. the ray must hit one of the faces of the volume before it can move to an adjoining element. Given that the originating face is not concave, the ray may not re-hit the surface from which it originated. Therefore, simultaneously solving the three dimensional equation of the ray (in cartesian coordinates),

5
where \((X_0, Y_0, Z_0)\) and \((X_1, Y_1, Z_1)\) are the coordinates of the starting and ending points and \(\cos \alpha, \cos \beta\) and \(\cos \gamma\) are the direction cosines, and three dimensional equations of each of the planes representing the faces of the control volume,

\[
\frac{X_1}{a} + \frac{Y_1}{b} + \frac{Z_1}{c} = d,
\]

(4)
gives the points of intersection between the ray and the planes. Subject to the size constraints of the faces dictated by the dimensions of the control volume, only one plane can satisfy all of the physical requirements. This is the next plane which the ray hits as it is traced through the simulation domain.

If the control volume immediately adjacent to the intersected plane contains the gas phase, the ray is transferred to that control volume and the tracing process is repeated. For control volumes adjacent to the plane of intersection which are on the boundary two cases may occur. The first represents a reflecting boundary. In this case, the surface normal of the plane is determined and the ray of light is reflected based on the incoming angles. Then the tracing process continues as described above. In the second case, an absorbing boundary, a counter representing the point of origin and point of termination is incremented by one. This terminates the tracing of this particular ray. A new ray trace is then started from the area \((A_i)\). If the control volume adjacent to the face struck represents a solid the same treatment as an absorbing boundary is followed.

Once all of the rays from one face \((A_i)\) are traced, all of the counters are divided by the total number of rays which left the face. The counters then represent the fraction of light emanating from the original surface which is incident on all of the other exposed surfaces. This is called the view factor \((F_{A_1-A_2})\) between surface 1 and surface 2. This process of finding the view factors is continued until all of the faces which may emit or absorb radiation have been selected.
Radiant Energy Exchange

Assuming that all radiating surfaces are "gray", the heat transfer rate at the ith radiating surface is given by an equation involving the surface temperature $T$, emissivity $\varepsilon$, and radiosity $B$:

$$\frac{Q_i}{A_i} = \varepsilon_i \left( \sigma T_i^4 - \sum_{j=1}^{N} B_j F_{Ai-Aj} \right)$$  \hspace{1cm} (5)

Notice that the heat transfer for surface $i$ depends upon the radiosity of all of the radiating surfaces, the sum over all $j$. Therefore, the heat flow for all of the surfaces must be solved simultaneously. The radiosity of a surface is the sum of the emitted and reflected radiant flux,

$$B_i = \varepsilon_i \sigma T_i^4 + (1-\varepsilon_i) \sum_{j=1}^{N} B_j F_{Ai-Aj}$$  \hspace{1cm} (6)

where $1-\varepsilon_i$ is the reflectance of surface $i$.

Equation (6) can be rewritten in matrix form,

$$X\vec{B} = \vec{\Omega}$$  \hspace{1cm} (7)

where

$$X_{ij} = \delta_{ij} - \frac{(1-\varepsilon_i) F_{Ai-Aj}}{\varepsilon_i}$$  \hspace{1cm} (8)

$$\Omega_i = \sigma T_i^4$$

and $\delta_{ij}$ is equal to 1 for $i=j$ and 0 for $i \neq j$.

Given that the view factors ($F$) have been calculated from the ray tracing portion of the program, the emissivity of the surfaces are known, and assuming the nodal temperatures are a good approximation for the surface temperatures, the radiosity at all faces within the simulation domain can be calculated. These are used with equation (5) to calculate the heat flux through each surface.

In the finite volume calculation of nodal temperatures the matrix form

$$A\vec{T} = \vec{b}$$  \hspace{1cm} (9)

is used, where $A$ is the matrix of interaction coefficients from the discretization equation, the
vector \( T \) contains the nodal temperatures and the vector \( b \) includes the source terms. To account for the thermal radiation, the heat source at each node is found by summing all of the heat transfers multiplied by the surface area of the exposed faces for the control volume. This sum gives the total amount of radiant energy which enters or leaves that volume. This then is included as a source term in the right hand side of equation (9).

During the initial testing of the model it was observed that, for large initial temperature gradients, the inclusion of the radiant source term sometimes causes divergence in the iterative temperature solution. In order to lessen the chance of divergence the source term was damped by the following equation

\[
S_{new} = \alpha S_{calculated} + (1 - \alpha)S_{old}
\]

(10)

Where the value of \( \alpha \) is generally between 0.01 and 0.1.

Since equation (7) is solved prior to solving for the nodal temperatures, old values of temperature are used in the solution. In order to obtain a self-consistant solution for the temperature several iterations of solving the radiant energy equations then solving for the nodal temperatures is required.

**CVI MODELING**

Our CVI model which includes gas flow, reactant transport and heat flow by conductance and convection, has been described previously\(^4\). The flow of the carrier gas and the concentrations of reacting species are determined by differential equations in the form of equation (1).

For pressure-driven gas flow,

\[
\nabla \cdot \left( \frac{k}{\mu} V P \right) = 0
\]

(11)

where \( k \) is the Darcy permeability for the material of each volume element, \( V \) is the gas molar volume and \( \mu \) is the gas viscosity. This formulation of gas transport does not include source or convective (inertial) terms. It will not be accurate for high velocity gas flow in open reactors but is suitable for the pressure-driven gas flow through semi-permeable materials as is the case for forced flow CVI.
The transport equation for the reacting species includes convection, diffusion and source terms,

\[ \nabla(u \ C_i) = \nabla \cdot (D_i \ \nabla C_i) + S \quad (12) \]

Two species are included in the simulation for SiC deposition, methyltricholorsilane (MTS) and hydrochloric acid (HCl). The MTS is the input species which decomposes into solid SiC which is deposited within the fibrous preform and gaseous HCl is a by-product. The matrix deposition rate depends on the concentrations of both species.

The coupled systems of differential equations for temperature, pressure and chemical species concentration are solved in the steady-state. For a selected time increment the local reaction rate is used to calculate a new density for each preform volume element. A new steady-state solution is then calculated and the density incremented again, producing a series of "snap shots" of the densification process.

Temperature Profile

Figures 3 and 4 show the calculated temperature profiles within the tube CVI system. Figure 3 does not include thermal radiation, including only conductive and convective heat transport. Figure 4 includes thermal radiation in addition to conduction and convection. The temperatures are very different for this furnace geometry. The radial temperature profile through the midpoint of the tube is plotted for each case in Figure 5 along with corresponding experimental measurements. Inclusion of radiation is clearly needed in order to match the actual temperature profile in the CVI system.
Figure 3. Temperature profile with no radiative heat transfer.

Figure 4. Temperature profile with radiative heat transfer.
Figure 5. Temperature profile matches experiment only when radiative heat transfer is included.

Densification

By using artificial, "effective" thermal conductivities for the gas spaces, the temperature profile without radiation can be forced to match the correct values for this simple furnace geometry. Two separate values of "effective" conductivity are needed for the gas space within the mandrel and the gas space outside of the preform. While this approach reduces the computational effort for the simulation, the method does not track the changing temperature profile during densification (Figure 6). Consequently, the densification behavior does not match that found with proper inclusion of radiation (Figure 7).
Figure 6. ID and OD temperatures change with densification. Using an "effective" conductivity, the temperatures diverge from the correct radiation-included result.

Figure 7. ID and OD densities increase at different rates. Density increase with "effective" thermal conductivity does not match that with radiation included.
CONCLUSIONS

Radiation is an important component of heat transfer in some configurations of the CVI process. Using a ray tracing procedure, radiative heating can be incorporated into the finite-volume method used previously for CVI modeling. While adding some computational burden this method allows accurate calculation of temperature profile for arbitrary furnace geometries and for the typical CVI situation where the thermal conductivity of the preform material changes during the process.

REFERENCES


## APPENDIX
### DISTRIBUTION

<table>
<thead>
<tr>
<th>Company Name</th>
<th>Address</th>
<th>Contact Person(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3M COMPANY</td>
<td>201-4N-01 3M Center, St. Paul, MN 55144</td>
<td>M. A. Leitheiser</td>
</tr>
<tr>
<td>A. P. GREEN REFRactories COMPANY</td>
<td>Green Blvd., Mexico, MO 65265</td>
<td>J. L. Hill</td>
</tr>
<tr>
<td>AIR PRODUCTS AND CHEMICALS</td>
<td>P.O. Box 538, Allentown, PA 18105</td>
<td>S. W. Dean</td>
</tr>
<tr>
<td>ALLISON GAS TURBINE DIVISION</td>
<td>P.O. Box 420, Indianapolis, IN 46206-0420</td>
<td>P. Khandelwal (Speed Code W-5), R. A. Wenglarz (Speed Code W-16)</td>
</tr>
<tr>
<td>AMA RESEARCH &amp; DEVELOPMENT CENTER</td>
<td>5950 McIntyre Street, Golden, CO 80403</td>
<td>T. B. Cox</td>
</tr>
<tr>
<td>ARGONNE NATIONAL LABORATORY</td>
<td>9700 S. Cass Avenue, Argonne, IL 60439</td>
<td>W. A. Ellingson, J. P. Singh</td>
</tr>
<tr>
<td>ARGONNE NATIONAL LABORATORY-West</td>
<td>P.O. Box 2528, Idaho Falls, ID 83403-2528</td>
<td>S. P. Henslee</td>
</tr>
<tr>
<td>ARMY MATERIALS TECHNOLOGY</td>
<td>Laboratory SLCMT-MCC, Watertown, MA 02172-0001</td>
<td>D. R. Messier</td>
</tr>
<tr>
<td>AVCO RESEARCH LABORATORY</td>
<td>2385 Revere Beach Parkway, Everett, MA 02149</td>
<td>R. J. Pollina</td>
</tr>
<tr>
<td>BABCOCK &amp; WILCOX</td>
<td>1562 Beeson St., Alliance, OH 44601</td>
<td>T. I. Johnson</td>
</tr>
<tr>
<td>BABCOCK &amp; WILCOX</td>
<td>Domestic Fossil Operations, 20 South Van Buren Avenue, Barberton, OH 44023</td>
<td>M. Gold</td>
</tr>
<tr>
<td>BRITISH COAL CORPORATION</td>
<td>Coal Research Establishment, Stoke Orchard, Cheltenham, Gloucestershire, England GL52 4RZ</td>
<td>J. Oakey</td>
</tr>
<tr>
<td>CANADA CENTER FOR MINERAL &amp; ENERGY TECHNOLOGY</td>
<td>568 Booth Street, Ottawa, Ontario, Canada K1A 0G1</td>
<td>R. Winston Revic, Mahi Sahoo</td>
</tr>
<tr>
<td>DOE OAK RIDGE OPERATIONS</td>
<td>P.O. Box 2001, Oak Ridge, TN 37831</td>
<td>E. E. Hoffman</td>
</tr>
<tr>
<td>DOE OAK RIDGE OPERATIONS</td>
<td>Building 4500N, MS 6269, Oak Ridge, TN 37831</td>
<td>E. E. Hoffman</td>
</tr>
</tbody>
</table>
