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Modeling of forced flow-thermal gradient CVI: effects of control parameters and processing schemes on a densification

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Abstract: A two-channel temperature-control infiltration reactor aiming at fabricating thicker and more uniform composites was designed. Unsteady numerical simulations containing hydrodynamics, mass transfer, heat transfer and pore structure evolution were carried out. By analyzing the effects of concentration, velocity and temperature on the porosity distribution, different densification schemes were proposed and compared. Results show that a preform of thickness 0.04 m and initial porosity 0.8 can be densified to a uniform and compact one with the maximum porosity of 0.1 within 300 hours through the three-step densification scheme. This scheme not only assures a low and uniform porosity over the whole preform but also avoids the huge pumping pressure which may cause the preform cracking.

Keywords: ceramic matrix composite; chemical vapor infiltration; deposition; modeling

1 Introduction

The ceramic matrix composites (CMCs) have continued to be of special concern that promises high safety and economic benefits in the fields of aeronautic and aerospace due to their excellent thermal and mechanical properties [1-2]. Its main fabrication technique is chemical vapor infiltration (CVI), whereby silicon carbide

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(SiC) matrix infiltrates into the porous preform and deposits on the fiber to form fiber-reinforced composites [3]. The most widely used CVI technique is isothermal isobaric CVI (ICVI), in which the densification is carried out at uniform temperature and the reactant gases are transported by concentration-gradients-induced diffusion. The ICVI was first introduced in the 1960s [4], and it is still widely used today [5-8] because it requires a rather simple technology and procedure. The preforms with complex shapes can be easily densified in the same run. However, its densification rate is very low, and the infiltration always lasts for hundreds of hours [7] due to the limitation of diffusion transport mode; besides, the surface of porous preform is often sealed due to overcoating, while the inner preform is still coarse, thus the infiltration process must be interrupted to machine impermeable skin [9].

A forced flow and an appropriate temperature gradient may fix the problems of transportation limitation and nonuniform deposition. Actually, since the 1980s the forced flow-thermal gradient CVI (F-TGCVI) has been developed to improve the densification quality [10-12]. The F-TGCVI reactor mainly consists of a bottom water-cooled holder and a top hot furnace. The reactant gases are forced into the fibrous preform under pressure and flowed from the cooled side to the hot side. The rapid flow can significantly accelerate the densification, for instance, preform densification that previously required weeks to densify now requires ~24 h [12]. The highest deposition rate of 3 $\mu\text{m/h}$ was reported in Ref. [13]. The initial steep temperature gradient makes the deposition mainly occur at the hot portion of the preform, which will increase the density and thermal conductivity of the preform. Consequently, the hot region expands progressively from the top hot side to the bottom cooled side of the preform, so does the densification. This deposition feature avoids the frequent interruption of machining impermeable skin, but with the

densification going on, the top hot side of the preform may be quite dense while other portions are still coarse. For instance, the coating thickness is about 8 μm at the cold side while 22-24 μm at the hot side [14]. This phenomenon gives rise to two problems. One is that the preform cannot be thick. The densified preform reported in Refs. [13-16] are quite thin, only 0.64-1.27 cm. The other one is that as the infiltration progressed, the back pressure increased as the pores in the preform were filled by the deposited matrix, the back pressure increased from the initial 3.4 KPa to a final 170 KPa in Ref. [13]. The huge pump pressure may cause preform cracking [17].

In order to solve these problems, we designed a two-channel F-TGCVI reactor, which is expected to obtain a thicker and more uniform composite material. Since running timescale experiments to test this design or optimize the parameters can be very expensive [18], the numerical simulation might be an economically viable way. In F-TGCVI, the control parameters, such as temperature, pressure, the gas composition and the flow rate, all affect chemical reaction and furtherly change the pore structure. The evolution of pore structure, in turn, will affect the species transport and temperature distribution. Therefore, it is not appropriate to simulate the infiltration process with an oversimplified mathematic model or assumption of a steady state of temperature and concentration [19-20]. In the present paper, a two-dimensional unsteady numerical simulation was carried out, in which hydrodynamics, mass transfer, heat transfer and pore structure evolution were considered. The effects of inlet reagent concentration, inlet gas velocity and temperature on the porosity distribution were analyzed and an improved three-step densification scheme was also given.

2 Numerical scheme

In the present paper, precursor methyltrichlorosilane (CH_3SiCl_3 , MTS), carrier gas hydrogen and carbon fiber are adopted to fabricate C_f/SiC composites through the two-channel F-TGCVI reactor. Since the complex relationships among the time-dependent variables, such as chemical reaction, concentration, temperature and porosity, will make it difficult to model the deposition in the preform, some necessary assumptions are considered:

- The pyrolysis of MTS is regarded as a first order reaction [21-22], and the reaction is simplified as $\text{CH}_3\text{SiCl}_3 \rightarrow \text{SiC}_{(s)} + 3\text{HCl}$. The heat generation by the chemical reaction is neglected.
- The porosity is considered uniform in the porous preform before infiltration, and the model of the surface area distribution between and within bundles proposed in Ref. [19] is also applicable to this work.
- All of the gases are considered as incompressible ideal gas, and the variation of their certain properties, such as density, dynamic viscosity, diffusion coefficient with temperature are neglected.

2.1 Physical model

The physical model, as shown in Fig. 1, contains two rectangle channels (the horizontal one $0.1 \times 0.5 \text{ m}^2$, the vertical one $0.1 \times 0.25 \text{ m}^2$) and a $0.1 \times 0.04 \text{ m}^2$ rectangle porous preform locating at the junction. A, B and C are velocity inlet or pressure outlet, reactor walls are thermal insulation and a certain temperature is assigned to the top and bottom edges of the preform (in practice, this can be achieved by the focused IR heating technology). Based on this two-channel F-TGCVI reactor, the flow control and the temperature control can be closely matched to optimize the deposition process, thus fabricating thick and uniform composite, simultaneously protecting the fiber preform from cracking.

Fig. 1 Schematic of physical model. For the convenience of results display, the vertical midline of preform is marked as l_v , the coordinate origin is set at the top-left end point of the preform and the point (0.05, -0.0374) is marked as M, the temperature at the top and bottom edges of the preform are marked as T_{top} and T_{bottom} , respectively.

2.2 Mass transport

According to mass conservation, chemical species transport through diffusion and convection can be expressed as [23]:

$$\frac{\partial \varepsilon c_i}{\partial t} + \mathbf{u} \cdot \nabla c_i - \nabla \cdot D_{\text{eff}} \nabla c_i = R_i \quad (1)$$

where ε denotes the porosity of preform; c_i is the concentration of species i , mol/m³; t is the infiltration time, s; $\mathbf{u}$ is the velocity field, m/s; D_{eff} denotes the effective diffusion coefficient, m²/s, $D_{\text{eff}} = \varepsilon D_{AB} / \tau$, τ is the tortuosity factor, $\tau = (\varepsilon_0 - 0.03) / (\varepsilon - 0.03)^{0.95}$ [6], D_{AB} is the Fick diffusion coefficient [24],

$$D_{AB} = \frac{10^{-7} T^{1.75} (1/M_A + 1/M_B)^{1/2}}{p \left[\sum V_A^{1/3} + \sum V_B^{1/3} \right]^2} \quad (2)$$

the subscript A, B represents MTS and H₂; T is the temperature, K; M is the molecular weight, kg/mol; p is the absolute pressure, atm; $\sum V$ is the diffusion volume, a sum of atomic volumes in each molecule. R_i is the reaction rate expression for the species i , mol/(m³·s), for the reagent (MTS, $i = 1$), for example, R_1 can be expressed as:

$$R_1 = -k_s c_1 S_v \quad (3)$$

k_s is the reaction rate constant, m/s, $k_s = k_0 \exp(-E_r/RT)$, k_0 is the preexponential factor, m/s, E_r is the activation energy, J/mol, and R is the gas constant, J/(K·mol); c_1 is the concentration of MTS, mol/m³, initial MTS concentration $c_{1,0} = \chi p/RT$, χ is the molar fraction of MTS; S_v is the surface area per unit volume, 1/m, $S_v = 2/r_0 \sqrt{(1-\varepsilon_0)/\varepsilon_0} \sqrt{1-\varepsilon} \varepsilon$ [19], r_0 and ε_0 are the initial pore radius (m) and porosity, respectively. The deposition of SiC ($i = 2$) causes the progressive plugging of the preform, therefore, the preform structural evolution can be written as:

$$\frac{d\varepsilon}{dt} = -\frac{M_2}{\rho_2} k_s c_1 S_v \quad (4)$$

M_2 is the molecular weight of SiC, kg/mol; ρ_2 is the density of SiC, kg/m³.

2.3 Flow

Flow in the free and porous domain are modeled using the Navier-Stokes equation and modified Navier-Stokes equation (Brinkmann equation) [25], respectively,

$$\rho \frac{\partial \mathbf{u}}{\partial t} = \nabla \cdot \left[-p \mathbf{I} + \mu \left(\nabla \mathbf{u} + \nabla \mathbf{u}^T \right) \right] \quad (5)$$

$$\frac{\rho}{\varepsilon} \frac{\partial \mathbf{u}}{\partial t} = -\nabla p + \nabla \cdot \left[\frac{1}{\varepsilon} \left(\mu \left(\nabla \mathbf{u} + \nabla \mathbf{u}^T \right) - \frac{2}{3} \mu \nabla \cdot \mathbf{u} \mathbf{I} \right) \right] - \frac{Q_m}{\varepsilon^2} \mathbf{u} - \frac{\mu}{\kappa} \mathbf{u} \quad (6)$$

where ρ is the density of gas mixture, kg/m³; μ is the dynamic viscosity of gas mixture, kg/(m·s); $\mathbf{I}$ denotes the identity matrix; Q_m denotes the mass deposit, kg/(m³·s), $Q_m = \rho \nabla \cdot \mathbf{u}$; κ is the permeability, m², $\kappa = D_p^2 \varepsilon^3 / [150 (1-\varepsilon)^2]$ [25], D_p is the average particle diameter, m, $D_p = 4\varepsilon/S_v$ [25].

2.4 Heat transfer

According to local thermal equilibrium, heat transfer equation in the porous domain is [26]:

$$\rho C_{p, \text{eff}} \frac{\partial T}{\partial t} + \rho C_p \mathbf{u} \cdot \nabla T + \nabla \cdot \mathbf{q} = Q \quad (7)$$

where $(\rho C_p)_{\text{eff}}$ is the effective volumetric heat capacity, consisting of porous medium term and gas mixture term, $(\rho C_p)_{\text{eff}} = (1 - \varepsilon) \rho_p C_{p,p} + \varepsilon \rho C_p$; $\mathbf{q}$ is the conductive heat flux, W/m, $\mathbf{q} = -k_{\text{eff}} \nabla T$, k_{eff} is the effective thermal conductivity, W/(m·K), $k_{\text{eff}} = 1 - \varepsilon k_p + \varepsilon k$, the ‘irradiative conductivity’ [27-28] is added to the k_p ; Q is the heat source, W/m. Heat transfer equation in the free domain is similar except ruling out the porous medium term.

2.5 Numerical solver

The COMSOL Multiphysics 5.5 was adopted to conduct the present two-dimension transient Multiphysics modeling. The transport of diluted species, Brinkman equation and heat transfer in porous media modules were activated to deal with the flow, mass transport and heat transfer problems, respectively. The issue of porosity evolution was settled by defining custom variables and a partial differential equation. A physics-controlled mesh (the element size is finer) was built, hour was selected as time unit and time step was 0.1 h. The requirement of mesh and time step independence were all met. Parameters used in the present simulation are summarized in Table 1.

Table 1 List of parameters used in the simulation

Parameter	Value	Parameter	Value
Total pressure p	10 kPa	Initial porosity ε_0	0.8
H ₂ /MTS molar ratio χ	6	Initial pore radius r_0	7×10^{-6} m
Gas flow F	1×10^{-4} m ³ /s	Specific Heat of gas C_p	14990 J/(kg·K)
Dynamic viscosity μ	5.4641×10^{-4} Pa·s	Specific Heat of preform $C_{p,p}$	2020 J/(kg·K)
Density of gas ρ	29.596 kg/m ³	Thermal conductivity of gas k	0.5 W/(m·K)

Density of preform ρ_p	1760 kg/m ³	Thermal conductivity of preform k_p	140 W/(m·K)
Activation energy E_r	2.96×10^5 J/mol	Preexponential factor k_0	3.89×10^9 m/s

3 Results and discussions

3.1 The effect of inlet MTS concentration

Fig. 2 Porosity variations over time along line l_v under different inlet MTS concentration. (a) 0.086 mol/m³; (b) 0.136 mol/m³; (c) 0.186 mol/m³; (d) 0.236 mol/m³. $T_{top} = 900$ K, $T_{bottom} = 1150$ K, inlet velocity 0.0127 m/s, A in C out.

The effect of inlet MTS concentration on the preform porosity was first explored. The gas mixture flows from A to C (Fig. 1), inlet velocity 0.0127 m/s, T_{top} 900 K and T_{bottom} 1150 K. Fig. 2 shows the porosity variations over time along the line l_v under different inlet MTS concentration. Generally, the porosity distribution of the four cases in Fig. 2 is similar, that is, the porosity drops rapidly at the bottom of the line l_v while remains unchanged at the top side; given that the fresh precursors flow from the

top to the bottom of the preform, it can be deduced that the temperature T_{top} 900 K is too low for the chemical reaction. One can find that the deposition rate increases as the increase of inlet MTS concentration. For instance, it takes 75 h for porosity at the point M to decrease to 0.3 in the case of inlet MTS concentration 0.136 mol/m^3 (Fig. 2b), while it just needs 45 h in the case of inlet MTS concentration 0.236 mol/m^3 (Fig. 2d). One can also notice that the gap between two adjacent lines gradually decreases over time in each case (Fig. 2), which could be caused by a continuous shrink of MTS concentration over time (Fig. 3a). Besides, under a certain inlet MTS concentration, the position of the largest slope moves upward over time, for instance, in Fig. 2b the largest slope of the curve $t = 60 \text{ h}$ is at $y = -0.0374 \text{ m}$, while on the curve $t = 90 \text{ h}$ it is at $y = -0.0234 \text{ m}$. Likewise, the largest slope position moves upward as the increase of inlet MTS concentration, for example, the largest slope of the curve $t = 60 \text{ h}$ in the case of inlet MTS concentration 0.136 mol/m^3 is at $y = -0.0374 \text{ m}$ (Fig. 2b), while on the curve $t = 60 \text{ h}$ in the case of inlet MTS concentration 0.186 mol/m^3 it is at $y = -0.0234 \text{ m}$ (Fig. 2c). These may mainly attribute to the deposition, that is, the increase of infiltration time or inlet MTS concentration results in more deposition, which will raise the thermal conductivity and induce more heat transferring upward from the bottom high-temperature zone (Fig. 3b). Given that the precursor flows downwards, the upward heat gives rise to the moving upward of the largest slope position.

Fig. 3 MTS concentration (a) and temperature (b) variations over time along line l_v in the case of inlet MTS concentration 0.086 mol/m^3 , $T_{\text{top}} = 900 \text{ K}$, $T_{\text{bottom}} = 1150 \text{ K}$, inlet velocity 0.0127 m/s , A in C out.

3.2 The effect of inlet velocity

Fig. 4 shows the porosity and velocity at point M under different inlet velocity at $t = 50 \text{ h}$. It presents a simple linear relationship, that is, as the inlet velocity increases, the velocity at the point M synchronously increases (the red line in Fig. 4), and the porosity lineally decreases (the black line in Fig. 4). When inlet velocity is 0.0027 m/s , the velocity at point M is just 0.0016 m/s , a decay about 40 percentage, and after an infiltration of 50 h the porosity is still as high as 0.463 . On the other hand, as the inlet velocity increase by 0.005 m/s (a corresponding total gas flow of 2.34 l/min), the porosity just decreases by about 0.06 , which is far from economical.

Fig. 4 Porosity and velocity at the point M under different inlet velocity. Inlet MTS concentration 0.186 mol/m^3 , $T_{\text{top}} = 900 \text{ K}$, $T_{\text{bottom}} = 1150 \text{ K}$, A in C out, infiltration time $t = 50 \text{ h}$.

Furtherly, we checked the MTS concentration and temperature distribution on the line l_v to find more information about the effect of inlet velocity. As shown in Fig. 5a, the increase of inlet velocity can improve the MTS concentration on the whole line l_v , but more obvious at the upside (top portion of the preform). Inlet velocity also has positive effect on the temperature distribution, but in a tiny degree (Fig. 5b). To sum up, the increase of inlet velocity will accelerate the deposition by increasing the concentration of MTS in the preform, but it is neither effective nor economic due to the large H_2 /MTS molar ratio. Hence, a moderate inlet velocity will be better for guaranteeing a forced-flow and an economic densification.

Fig. 5 MTS concentration and temperature distribution along line l_v under different inlet velocity. Inlet MTS concentration 0.186 mol/m^3 , $T_{\text{top}} = 900 \text{ K}$, $T_{\text{bottom}} = 1150 \text{ K}$, A in C out, infiltration time $t = 50 \text{ h}$.

3.3 The effect of temperature

The effect of temperature was also investigated. Under conditions of A in C out, inlet MTS concentration 0.186 mol/m^3 and inlet velocity 0.0127 m/s , a number of cases with different top-bottom temperature pairs were simulated. Fig. 6 presents four typical cases: in the case one $T_{\text{top}} = 900 \text{ K}$ and $T_{\text{bottom}} = 1150 \text{ K}$, the bottom preform was firstly densified (Fig. 6a) due to the excessively low top temperature; things are changed in the case of $T_{\text{top}} = 1100 \text{ K}$, $T_{\text{bottom}} = 1200 \text{ K}$. The bottom temperature is still higher than the top temperature, but the top temperature 1100 K is high enough for

reaction, thus densification mainly occurs at the top portion (Fig. 6b) where the precursor concentration is higher; when T_{bottom} remains at 1200 K, but the T_{top} is lowered to 1050 K. The porosity distribution over the whole preform notably changes once again. As shown in Fig. 6c, the most distinguished porosity decrease occurs at the middle portion; when the preform is kept iso-thermal, 1000 K, the densification over the whole preform is quite uniform, but the deposition is very slow (Fig. 6d).

Fig. 6 Porosity variations over time along line l_v under different temperature pairs. (a) $T_{\text{top}} = 900$ K, $T_{\text{bottom}} = 1150$ K; (b) $T_{\text{top}} = 1100$ K, $T_{\text{bottom}} = 1200$ K; (c) $T_{\text{top}} = 1050$ K, $T_{\text{bottom}} = 1200$ K; (d) $T_{\text{top}} = 1000$ K, $T_{\text{bottom}} = 1000$ K mol/m³. Inlet MTS concentration 0.0186 mol/m³, inlet velocity 0.0127 m/s, A in C out.

In the cases of Fig. 6b and Fig. 6c, the only difference is the top temperature, i.e., 1100 K in the case b and 1050 K in the case c, but this small temperature difference causes quite different porosity distribution. Initially, as shown by Fig. 7, the porosity difference of the two cases is quite small in magnitude (about 0.003) at $t = 1$ h. The distribution trend, however, is already quite evident. That is, in the case b, porosity at

the top portion of the preform is smaller due to the higher top temperature (more intense reaction); while reaction at the top portion in the case c is less intense, thus more precursors reaching to the middle-lower portion where the temperature is higher, as a result, the middle-lower portion of the preform has the lowest porosity (Fig. 7).

Fig. 7 Porosity distribution in the case b and c at $t = 1$ h.

Fig. 8 Relationship of surface area with porosity.

The initial porosity distribution will strongly affect the successive distribution of some parameters, such as surface area per unit volume, MTS concentration and temperature. According to the relationship between surface area per unit volume and porosity [19], the decrease of porosity will cause the surface area to increase first and then decrease (Fig. 8). Therefore, the porosity distribution trend in Fig. 7 will lead to a different surface area distribution in the two cases, i.e., larger surface area at the top portion in the case b while larger surface area at the middle portion in the case c, as shown in Fig. 9a and Fig. 9d. Likewise, the MTS concentration also closely relates to the porosity distribution: in the case b, the porosity at the top portion of the preform drops rapidly, which means a gradually plugging at the top portion and results in more precursors staying at the top portion (Fig. 9b); while lower top temperature and coarser top preform in the case c induce a higher MTS concentration through the whole preform (Fig. 9e). In terms of the temperature, as explained in section 3.1, the porosity relates to the thermal conductivity and furtherly affects the temperature distribution. In the case b, the thermal conductivity at the top portion is higher,

corresponding to the smaller temperature gradient at the top portion (Fig. 9c), while in the case c, the smaller temperature gradient occurs near the middle portion (Fig. 9f). As we should remember, the temporal distribution of porosity is proportional to the reaction rate R_1 , more specifically, it is proportional to the surface area, MTS concentration and temperature ($R_1 = k_s c_1 S_v$). That is to say, the initial parameters bring about an initial porosity distribution trend, this initial porosity distribution furtherly induces a corresponding parameter change, which in turn, will enhance the porosity distribution trend.

Fig. 9 Surface area per unit volume, MTS concentration and temperature variations over time along the line l_v corresponding to the case b (a-c) and the case c (d-f).

3.4 The two-step densification scheme

According to Fig. 6a and Fig. 6b, a two-step densification scheme which has the possibility to simply densify the preform was proposed: The gas mixture first flows from A to C (Fig. 10a) to densify a half of the preform, and then the gas flows from C to B (Fig. 10b) to finish the densification of the remaining half.

Fig. 10 Streamlines distribution at the last hour of each step in a simple two-step densification scheme. The detailed parameters are listed in table 2. The vectors give the direction, and the color gives the magnitude in m/s.

Cases with different parameters were tried, and table 2 lists a viable one. In this case, porosity at the upper portion quickly drops due to the suitable temperature and fresh precursors, and the other portion of the preform is also densified to some degree with the aid of high bottom temperature 1200 K (Fig. 11a). In the next step, the top and bottom temperature arrangement is reversed. The gas mixture flows from C to B, and the lower portion of the preform is densified (Fig. 11b).

Table 2 List of parameters used in the two-step densification scheme

Steps	Temperature (K)		Inlet MTS concentration (mol/m ³)	Inlet velocity (m/s)	Inlet and outlet	Infiltration time (h)
	T_{top}	T_{bottom}				
step I	1100	1200	0.186	0.0127	A in C out	45
step II	1200	1100	0.186	0.0127	C in B out	45

Fig. 11 Porosity variations over time along line l_v in a simple two-step densification scheme. (a) - (b) Step I - Step II. The detailed parameters are listed in table 2.

As can be noticed, the infiltration time in the two steps are short and the final porosity is rather high (Fig. 11b). Actually, cases with longer infiltration time were tried but they all failed. The reason lies in the high reaction rate at a specific region. When the gas flows from A to C, the upper left corner of the preform has the thinnest concentration boundary layer, i.e., the highest concentration. Moreover, the temperature there (1100 K) is suitable for the pyrolysis, thus the overlay of high concentration and temperature giving rise to an extremely low porosity zone at the upper left corner (Fig. 12a). The porosity of somewhere at the upper left corner decreases to zero when the total infiltration time exceeds 90 hours. As a result, the simulation cannot go on even if the maximum porosity over the entire preform is still as high as 0.5 (Fig. 12b).

Fig. 12 Contour of porosity variations over the whole preform at the last hour of each step in a simple two-step densification scheme. (a) - (b) Step I - Step II. The detailed parameters are listed in table 2. Porosity in the two steps differs a lot, for the convenience of viewing, different bars and color systems were used in the porosity contours.

3.5 The three-step densification scheme

To avoid the overlay of high concentration and high temperature, a three-step densification scheme was proposed: In the first step the gas flows from A to C (Fig. 13a), and deposition mainly occurs at the bottom portion where temperature is high; in the middle step the gas still flows from A to C (Fig. 13b), but the top and bottom temperature is adjusted so that the middle portion can be densified; due to the

plugging of most of the preform, if we still pump all the gas out from the outlet C, the huge stress may cause preform cracking [17], thus in the final step the gas mixture flows from A to B (Fig. 13c), and precursors diffuse into the top portion to densify the remaining coarse top preform.

Fig. 13 Streamlines (a-c) and MTS concentration (d-f) distribution at the last hour of each step in the three-step densification scheme. The detailed parameters are listed in table 3. The vectors give the direction, and the color gives the magnitude in m/s or mol/m³.

Table 3 List of parameters used in a three-step densification scheme

Steps	Temperature (K)		Inlet MTS concentration (mol/m ³)	Inlet velocity (m/s)	Inlet and outlet	Infiltration time (h)
	T_{top}	T_{bottom}				
step I	900	1150	0.186	0.0127	A in C out	85
step II	1050	1200	0.186	0.0127	A in C out	80
step III	1125	1050	0.186	0.0127	A in B out	35

Cases with different parameters were simulated. By comparing the final porosity and total infiltration time, table 3 lists a relatively good one. In the step I, due to the low top temperature 900 K, the precursor can reach far deep to the lower middle preform (Fig. 13d), and porosity near the bottom preform decreases significantly as expected (Fig. 14a). In the next step, with higher top and bottom temperature, the reaction zone moves upward, the precursor is almost depleted at the middle portion

(Fig. 13e), and the middle portion of the preform is densified (Fig. 14b). In these two steps, as the lower part of the preform first gets plugged, the porosity gradient induces an oscillatory flow in the preform (Fig. 13a-b). In the final step, the precursor diffuses into the top preform (Fig. 13f) and the remaining coarse top preform is also densified (Fig. 14c)

Fig. 14 Porosity variations over time along line l_v in a three-step densification scheme. (a) - (c) step I - step III. The detailed parameters are listed in table 3.

The three-step densification scheme significantly lowers the porosity of the whole preform, but the uniformity still needs to be improved. In the first two steps the deposition zone gradually moves from the bottom to the top. There is an obvious stratification and porosity in each layer is quite uniform, as shown by Fig. 15a and Fig. 15b. In the step III, however, the unwanted impermeable skin arises at the upper left corner of the preform while porosity under the right surface is still as high as 0.19 (Fig. 15c). Likewise, the two blue high-porosity strips near the bottom preform are also very eye-catching.

Fig. 15 Contour of porosity variations over the whole preform at the last hour of each step in the three-step densification scheme. (a) - (c) Step I - Step III. The detailed parameters are listed in table 3.

Three measures were taken to improve the three-step densification scheme. 1. In the step I and II, extend the infiltration time to eliminate the high-porosity strips near

the bottom preform. 2. In the step III, pump part of gases out from the outlet C to increase the MTS concentration in the preform. 3. In the step III, uniform the deposition (reaction rate) at the top portion by applying a horizontal linearly increasing temperature on the top edge of the preform to offset the effect of horizontally decreasing concentration. The measure of horizontal temperature gradient fixes the problem of the rapid sealing of the upper left corner, so the simulation can run longer in the step III. Additionally, the gas pumping from the outlet C obviously increases the concentration near the top portion (Fig.16). The detail parameters of improved three-step densification scheme are given in table 4. As can be seen from Fig. 17, the densification situation is significantly improved, i.e., the porosity over whole preform is not only lower but also more uniform (Fig. 17b).

Fig. 16 Contour of MTS concentration distribution at $t = 1$ h under different outlet condition. (a) A in B out; (b) A in, B and C out.

Table 4 List of parameters used in the improved three-step densification scheme

Steps	Temperature (K)		Inlet MTS concentration (mol/m ³)	Inlet velocity (m/s)	Inlet and outlet	Infiltration time (h)
	T_{top}	T_{bottom}				
step I	900	1150	0.186	0.0127	A in, C out	90
step II	1050	1200	0.186	0.0127	A in, C out	90
step III	$80x+1075$	1050	0.186	0.0127	A in, B C out	120
	$0 \leq x \leq 0.1$

Fig. 17 Contour of porosity distribution (a) before and (b) after some improvements being taken in the three-step densification scheme.

4 Conclusions

In the present paper a two-channel temperature-control infiltration reactor aiming at fabricating thicker and more uniform composites was designed. The effects of concentration, velocity and temperature on the porosity distribution were first analyzed, based on which different densification schemes were proposed and compared. By adopting improving measures, an optimal densification scheme was given.

(1) The densification rate can be accelerated by increasing the inlet MTS concentration (MTS molar ratio) or inlet velocity (total gas flow), while the former is more effective. The deposition behavior is mainly determined by the top and bottom temperature, a small temperature difference can dramatically change the porosity distribution. Temperature, concentration and velocity determine the porosity evolution, the porosity evolution, in turn, also affects the distribution of these parameters.

(2) A three-step densification scheme based on the two-channel reactor is proposed: In the first step the gas flows through the preform whose top temperature is not suitable for reaction, and deposition mainly occurring at the high-temperature bottom portion; in the middle step the top and bottom temperature is adjusted to assure certain deposition occurring at the middle portion; in the final step, the two outlets (pump)

are all open, and a linearly increasing temperature is applied to the top edge of the preform, whereby the coarse top portion is furtherly densified. This scheme avoids the unwanted sealing of preform by separating the high precursor concentration region from the high temperature region. Besides, in the step III a part of the gas is pumped out from the outlet C, and the main flow doesn't go through the dense preform, which not only assures the precursor supply in the preform but also protects the fiber preform from cracking due to the huge pumping pressure.

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Figures

Figure 1

Schematic of physical model. For the convenience of results display, the vertical midline of preform is marked as l_v , the coordinate origin is set at the top-left end point of the preform and the point (0.05, -0.0374) is marked as M , the temperature at the top and bottom edges of the preform are marked as T_{top} and T_{bottom} , respectively.

Figure 2

Porosity variations over time along line lv under different inlet MTS concentration. (a) 0.086 mol/m³; (b) 0.136 mol/m³; (c) 0.186 mol/m³; (d) 0.236 mol/m³. $T_{top} = 900$ K, $T_{bottom} = 1150$ K, inlet velocity 0.0127 m/s, A in C out.

Figure 3

MTS concentration (a) and temperature (b) variations over time along line lv in the case of inlet MTS concentration 0.086 mol/m³, T_{top} = 900 K, T_{bottom} = 1150 K, inlet velocity 0.0127 m/s, A in C out.

Figure 4

Porosity and velocity at the point M under different inlet velocity. Inlet MTS concentration 0.186 mol/m³, T_{top} = 900 K, T_{bottom} = 1150 K, A in C out, infiltration time t = 50 h.

Figure 5

MTS concentration and temperature distribution along line lv under different inlet velocity. Inlet MTS concentration 0.186 mol/m³, T_{top} = 900 K, T_{bottom} = 1150 K, A in C out, infiltration time t = 50 h.

Figure 6

Porosity variations over time along line lv under different temperature pairs. (a) $T_{top} = 900$ K, $T_{bottom} = 1150$ K; (b) $T_{top} = 1100$ K, $T_{bottom} = 1200$ K; (c) $T_{top} = 1050$ K, $T_{bottom} = 1200$ K; (d) $T_{top} = 1000$ K, $T_{bottom} = 1000$ K mol/m³. Inlet MTS concentration 0.0186 mol/m³, inlet velocity 0.0127 m/s, A in C out.

Figure 7

Porosity distribution in the case b and c at $t = 1$ h.

Figure 8

Relationship of surface area with porosity.

Figure 9

Surface area per unit volume, MTS concentration and temperature variations over time along the line lv corresponding to the case b (a-c) and the case c (d-f).

Figure 10

Streamlines distribution at the last hour of each step in a simple two-step densification scheme. The detailed parameters are listed in table 2. The vectors give the direction, and the color gives the magnitude in m/s.

Figure 11

Porosity variations over time along line Iv in a simple two-step densification scheme. (a) - (b) Step I - Step II. The detailed parameters are listed in table 2.

Figure 12

Contour of porosity variations over the whole preform at the last hour of each step in a simple two-step densification scheme. (a) - (b) Step I - Step II. The detailed parameters are listed in table 2. Porosity in the two steps differs a lot, for the convenience of viewing, different bars and color systems were used in the porosity contours.

Figure 13

Streamlines (a-c) and MTS concentration (d-f) distribution at the last hour of each step in the three-step densification scheme. The detailed parameters are listed in table 3. The vectors give the direction, and the color gives the magnitude in m/s or mol/m³.

Figure 14

Porosity variations over time along line lv in a three-step densification scheme. (a) - (c) step I - step III. The detailed parameters are listed in table 3.

Figure 15

Contour of porosity variations over the whole preform at the last hour of each step in the three-step densification scheme. (a) - (c) Step I - Step III. The detailed parameters are listed in table 3.

Figure 16

Contour of MTS concentration distribution at $t = 1$ h under different outlet condition. (a) A in B out; (b) A in, B and C out.

Figure 17

Contour of porosity distribution (a) before and (b) after some improvements being taken in the three-step densification scheme.