NUMERICAL STUDY OF GAS PHASE PRODUCTION OF CERAMIC COATINGS MODIFIED BY CARBON NANOTUBES

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Coating of surfaces of components is an efficient method to increase their life time. For many applications, of great importance is the problem of hard coatings with improved damping and vibrated characteristics. Perspective way of solving this problem is creation of coatings with nano-sized structures, in particular with carbon nanotubes. Unique properties of carbon nanotubes such as high chemical and thermal stability, unusual electronic properties, excellent thermal conductivity and extraordinary mechanical characteristics allow designing on their base composite materials and coatings of a new generation.

It is known that straight multiwall carbon nanotubes oriented perpendicular to the surface of a substrate can be grown by catalytic pyrolysis of gaseous hydrocarbons. Such layer of nanotubes can be considered as reinforcement for special composite coatings. In the coating, nanotubes must be bound by a matrix material which provides solidity, hardness and strength of the coating. Such ceramic materials as SiC, BN and Al_2O_3 can be used for the matrix fabrication.

The major technological problem of production of such coatings is difficulty of combining the matrix with layer of nanotubes which represents a nano-sized porous medium. One of the most attractive methods for solving this problem is Chemical Vapour Infiltration (CVI) [1]. This method is based on deposition of the ceramic matrix on the heated carbon nanotubes by chemical decomposition of gaseous precursors infiltrating through a porous preform. An advantage of this method is a possibility to obtain composites with matrices having very high melting temperatures under minimum thermal, mechanical and chemical impact on the reinforcing nanotubes. One of the most important problems in CVI research is finding the optimal values of technological parameters providing minimal infiltration time and required quality of the coating. The main requirement is maximal and uniform densification of the layer of nanotubes by the ceramic matrix. Lately, numerical modelling has become a helpful tool for the CVI study and optimization [2-3].

In the present work, numerical study of production of dense coatings by isothermal and thermal gradient CVI has been performed. In the Isothermal CVI (ICVI), preform is kept at a constant temperature, while Thermal Gradient CVI (TG CVI) is characterized by a temperature gradient across the preform bulk, which is provided by placing the preform on a heated substrate.

One-dimensional (along the coating thickness) model is used for numerical experiments. Preform formed by carbon nanotubes grown perpendicular to the surface of a substrate is considered (Fig. 1). The model describes heat and mass transport in the porous medium and deposition of the matrix material in heterogeneous chemical reactions. It accounts for the convective mass transport due to the phase transitions and simulates the evolution of the porous medium. The model of heat and mass transport is a set of differential equations including the following equations [4]: (i) conservation of condensed product, allowing evaluation of evolution of porous medium geometry; (ii) gas mixture continuity; (iii) transport of individual gas species; (iv) momentum conservation (Darcy's law); (v) energy conservation. The quasi-steady-state approach is used to simulate a long-term CVI process.

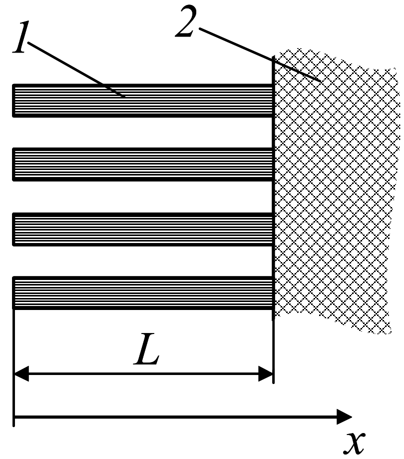

Fig. 1. Structure of porous medium of the coating: 1 – carbon nanotubes; 2 – substrate.

In this work, we consider production of coatings with SiC matrix from methyltrichlorosilane (MTS) CH_3SiCl_3 diluted by hydrogen. We assume SiC deposition to occur in the following reversible reaction of the MTS decomposition.

$$SiCl_3CH_3 \leftrightarrow SiC + 3HCl.$$

Effective characteristics of the porous medium (heat conductivity, permeability, diffusion coefficients and viscosity of the vapour in the pores) are computed from local values of porosity, vapour composition, pressure and temperature. Nanotube diameter used in the computations was equal to 80 nm. Volume fraction of carbon nanotubes in the layer was 0,18. The coating thickness (nanotube length) was varied in the range L = 100…1000 μm.

We have analysed the influence of the ICVI and TG CVI operating conditions (the MTS concentration in the ambient gas, the pressure in the reactor, temperature and temperature gradient) on the process duration (densification time) and the composite quality, i.e. the average residual porosity and residual porosity distribution along the coating thickness. Fig. 2 presents an example of computed residual porosity for different temperature values of ICVI process.

It was shown that the process temperature and the temperature gradient (for TG CVI) have the major effect on the coating quality. At the same time, the pressure and the precursor concentration influence primarily the densification time. It was found that extremely low temperature is needed to provide production of dense and uniform coatings. This effect is related to very high specific surface of nano-sized porous medium. This leads to high rate of condensed material formation, even at a low temperature, and, hence, to significant gradient of MTS concentration along the coating thickness. TG CVI process allows obtaining material of higher quality but its practical realization may be a serious technical problem.

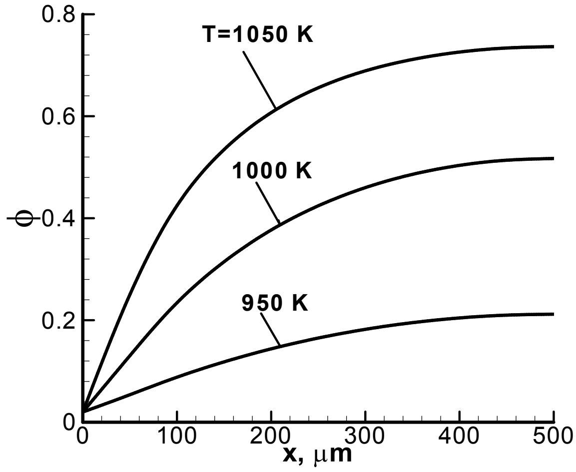

Fig. 2. Distribution of residual porosity along the coating thickness at various temperatures of ICVI process.

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