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3D THERMAL MODELING OF A PLASMA ASSISTED CHEMICAL VAPOR DEPOSITION PROCESS

S. Rouquette, L. Autrique, C. Chaussavoine, L. Thomas

IMP-CNRS, Université de Perpignan, 52 avenue de villeneuve, 66860 Perpignan cedex,
tel: +33 468 662 239, fax: +33 468 672 166, email : rouquett@univ-perp.fr

ABSTRACT : Due to unique properties such as hardness, or a low friction coefficient, amorphous silicon carbide films are attractive for mechanical applications. Even if Plasma Assisted Chemical Vapor Deposition processes are now commonly used for the growth of such coatings, mechanisms leading to their formations are not completely understood. However, the substrate temperature is considered as one of the key-parameters for this technique. Then in order to provide an effective predictive simulation tool or to determine optimal control procedures, a 3D thermal modeling of the Plasma Assisted Chemical Vapor Deposition process has to be investigated.

KEYWORDS : Chemical Process, Partial differential equation, Sensitivity analysis, Parameter estimation

1. INTRODUCTION

Surface treatments of materials offer an extended field of industrial applications, in particular in the thermo mechanical field (high hardness, low friction-low wear thin films). In order to obtain such specific coatings (amorphous silicon carbide films), a microwave Plasma Assisted Chemical Vapor Deposition (PACVD) reactor has been developed in the IMP CNRS Institute in France. In previous studies (considering similar technologies), it has been shown that the temperature of the substrate (where the growth of the film occurs) is one of the most important parameter for the elaboration process.

Thus a thermal modeling can provide an efficient predictive tool and can lead to the determination of optimal control, diagnosis or supervision procedures in order to control the temperature evolution of the substrate. In such a framework, a classical system of non linear partial differential equation and boundary conditions is considered and a finite element method in a three-dimensional geometry is implemented. Then, the model has to be validated and the effect of the uncertainty of parameters has to be investigated. Thus, an identification problem can be stated in order to determine the influent parameters.

The microwave plasma chemical vapor deposition reactor is briefly exposed in the following paragraph. Then, the model describing the temperature evolution is written. Direct problem is solved by a finite element method. Several numerical results are given. In the fourth paragraph, the determination of unknown parameters is considered and leads to the formulation of an inverse problem (least square estimation). The minimization of the quadratic criterion is achieved by a conjugate gradient algorithm.

2. PLASMA ASSISTED CHEMICAL VAPOR DEPOSITION PROCESS

Chemical Vapour Deposition (CVD) techniques are developed since the early eighties in many high technological domains such as microelectronics, optics or aeronautics. Specifically For PACVD techniques, the energy required for the activation of the chemical reactions, is generally provided by cold plasmas, sustained by an electrical field, which allows medium substrate temperature range ($300K \leftrightarrow 1100K$). Such a technique is developed in the IMP CNRS Institute in Perpignan (France) for

the elaboration of amorphous silicon carbide films for tribological applications. The experimental set-up is detailed elsewhere (Thomas et al., 2001). In our process the hidden backside of the substrate is heated by a circular thermal source (connected to a PID controller) set in a ceramic support. For technological reasons, the temperature measurements can not be obtained on the substrate surface. This is an important limitation of the PID efficiency.

In this study, we are mainly interested in the thermal behavior of the surface of the substrate (where the growth of the film will occur) before the plasma ignition. In that preliminary heating, the relevant domain is the heating element (PID controlled), the ceramic (support of the heating element) and the substrate holder. Several substrate shapes can be used in this reactor and avoid an axisymmetric assumption and a simplification of the geometry.

3. 3D THERMAL MODELING

The heat exchanges are presented on figure 1.

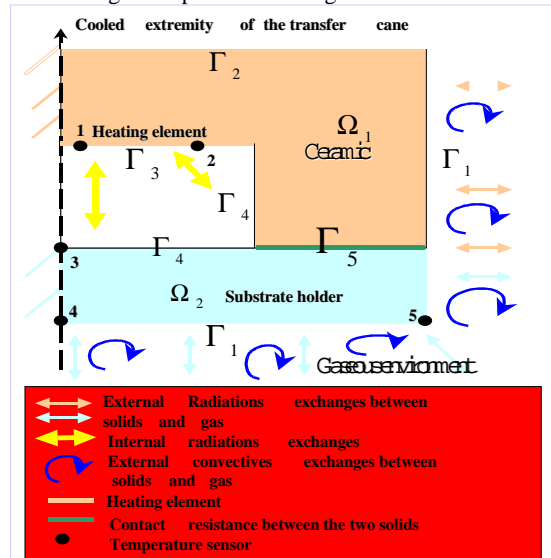


Figure 1 : description of the heat exchanges

Let us denote by :

- $x \in \Omega_1 \cup \Omega_2$, the space variable, where $\Omega_i \subset \mathbb{R}^3$ ($i=1$ corresponds to the ceramic and $i=2$ corresponds to the substrate holder). The surface of Ω is : $\Gamma = \bigcup_{j=1}^5 \Gamma_j$.
- $t \in T = [0, t_f]$ is the time variable.
- $\theta(x, t)$ is the temperature and the initial temperature is constant : $\theta_0 = 293K$,
- physical and thermal characteristics of the two considered materials ($i=1,2$) are denoted by ρ_i (the mass density), c_i (the specific heat), ε_i (the emissivity), $\lambda_i(\theta)$ (the thermal conductivity),

- $h(\theta)$ the convective exchange coefficient on boundary Γ_1 ,
 σ the stefan constant,
- R_c the thermal resistance describing the contact between Ω_1 and Ω_2 .

The thermal evolution of the material during the process is described by the following equations :

- state equations : $\forall (x,t) \in \left(\bigcup_{i=1}^2 \Omega_i \right) \times T$

$$\rho_i c_i \frac{\partial \theta}{\partial t} - \text{div}(\lambda_i(\theta) \overline{\text{grad}}(\theta)) = 0 \quad (1i)$$

- initial condition : $\forall x \in \Omega$

$$\theta(x,0) = \theta_0 \quad (2)$$

- heat exchange conditions : $\forall (x,t) \in \Gamma_1 \times T$

$$-\lambda_i(\theta) \frac{\partial \theta}{\partial \bar{n}} = h(\theta)(\theta - \theta_0) + \varepsilon_i \sigma (\theta^4 - \theta_0^4) \quad (3i)$$

where \bar{n} is the normal vector exterior to the surface.

- cooling condition : $\forall (x,t) \in \Gamma_2 \times T$

$$\theta(x,t) = \theta_c \quad (4)$$

where $\theta_c = 288K$ is the temperature of the cooled extremity of the transfer cane.

- heating condition : $\forall (x,t) \in \Gamma_3 \times T$

$$\theta(x,t) = \theta_h \quad (5)$$

where $\theta_h = 1000K$ is the temperature of the heating element (assumed to be constant and uniform).

- heat exchange conditions : $\forall (x,t) \in \Gamma_4 \times T$

$$-\lambda_i(\theta) \frac{\partial \theta}{\partial \bar{n}} = \varepsilon_i \sigma (\theta^4 - \theta_0^4) \quad (6i)$$

where Γ_4 is the boundary of the cylindrical cavity situated within the substrate holder and the ceramic .

- contact conditions : $\forall (x,t) \in \Gamma_5 \times T$

$$-\lambda_i(\theta) \frac{\partial \theta}{\partial \bar{n}} = \frac{1}{R_c} \delta_i(\theta) \quad (7i)$$

where $\delta_i(\theta)$ is the temperature gap between the ceramic and the substrate holder on boundary Γ_5 .

According to the previous notations, direct problem can be formulated as follows :

Problem P_{dir} :

find the temperature $\theta(x,t)$ solution of the non linear distributed parameter system :

$$(S) \begin{cases} \text{state equations} & (1i) \\ \text{initial condition} & (2) \\ \text{boundary conditions} & (3i)(4)(5)(6i)(7i) \end{cases}$$

Except for well-known problems (involving specific non linearities and boundary conditions), existence and uniqueness of the solution of problem P_{dir} can not be stated. Nevertheless, numerical methods such as finite element method can lead to a numerical determination of state $\theta(x,t)$ in an adapted function

space. Thermophysical properties of the materials are given in table 1.

Table 1 Thermophysical characteristics

	Ceramic	Steel 304 L
ρ_i mass density ($kg.m^{-3}$)	2500	$-0.346\theta + 7957$
c_i specific heat ($J.kg^{-1}.K^{-1}$)	853.8	$0.275\theta + 376$
λ_i thermal conductivity ($W.m^{-1}.K^{-1}$)	2.67	$0.0143\theta + 11.1$
ε_i emissivity	0.85	0.6

The heat exchanges are simulated with the following values :

- the unknown convective exchange coefficient is arbitrary fixed at $5 W.m^{-2}.K^{-1}$ for this simulation,
- the stefan constant is : $5.67 10^{-8} W.m^{-2}.K^{-4}$,
- the thermal resistance is about : $R_c = 1.85 10^{-3} m^2.K.W^{-1}$,

The time interval is $T = [0; t_f]$ where $t_f = 1800s$ since thermal equilibrium is expected by experimenters. Problem P_{dir} is solved by a finite element method in space (space step is about $1.5 10^{-3} m$) and finite differentiation in time (time step is $15s$). On the following figures (figure 2 and 3), the thermal behavior is presented :

- figure 2 : temperature evolution at points 3, 4 and 5 (see figure 1),
- figure 3 : temperature spatial distribution at $t_f = 1800s$.

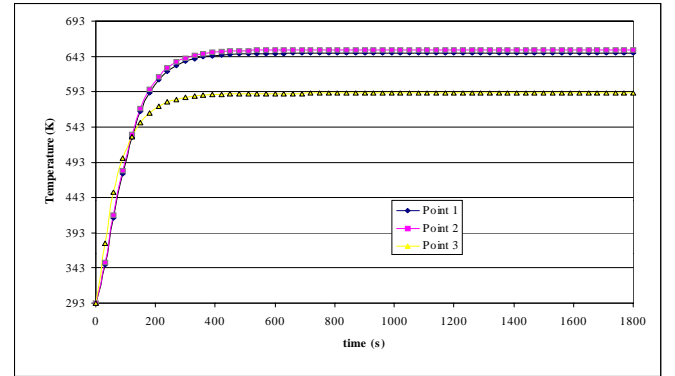


Fig. 2 : temperature evolution at points 3, 4 and 5.

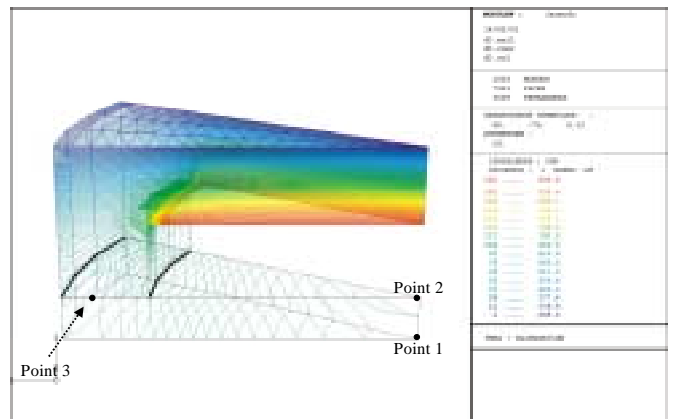


Fig. 3 : temperature distribution at $t_f = 1800s$.

Analysis of these numerical results leads to the following remarks:

- thermal equilibrium is reached after about 600s ,
- at points 3 and 4, temperature evolutions are quite similar (difference is lower than 4K), thus temperature gradient in the thickness of the substrate holder can be neglected,
- at the thermal equilibrium, temperature difference at points 4 and 5, is about 70K . Then, on the substrate surface, temperature is not uniform. Since this is one of the most important parameter (for the deposition process), a crucial aspect is then pointed out.

For further applications, an efficient predictive model is essential to achieve an optimal sensors location and to ensure the control of the process. Thus, the model has to be validated and the effect of the uncertainty of parameters has to be investigated. An identification problem can be stated in order to determine the influent parameters.

Effect of the uncertainty of parameters is investigated using a Design Of Experiment (DOE) procedure, see (Box and Draper, 1987) and (Montgomery, 1997) for example. Two-level factorial design is particularly useful in the early stages of experimental work, when there are likely to be many factors to be investigated. The aim of this approach is to identify the critical model factors and to provide information about which factors should be more carefully controlled to prevent high numerical variation of the model responses. DOE method is usually implemented in experimental situations where a model is not available. In the specific framework of this communication, a physical model is already established. This work is focused on the numerical sensitivity of the model. DOE strategy is applied to numerical results (obtained from several simulations based on a finite element method in a 3D geometry). Each run leads to a simulation and numerical results are obtained without experimental unknown disturbances (noises). Thus, statistical analysis (for example, significance tests determination of confidence intervals) are meaningless.

Several physical coefficients in the non linear partial differential equation or in the boundary conditions are considered :

- thermophysical parameters known with a given indeterminacy : $\rho_1 c_1$, $\rho_2 c_2$, λ_1 , λ_2 .
- thermophysical and experimental parameters which are not well-known : convective exchange coefficient h , emissivity ε_1 , ε_2 , thermal resistance R_c ,
- experimental conditions : heating element temperature θ_h (cooling temperature θ_c is assumed to be perfectly known).

However, it is obvious that these factors have to be taken into account in different ways. For example, based on the experimentation and the process knowledge, it seems that factors (θ_h) is crucial. Moreover, thermal characteristics ($\rho_1 c_1$) , ($\rho_2 c_2$) , (λ_1) , (λ_2) are generally well known and lead to narrow confidence interval (defined between two levels). Influence of factors (h) , (ε_1) , (ε_2) , (R_c) , is quite difficult to estimate (a priori). In (Rouquette *et al.*, 2001), based on the methodology exposed above, it has been shown that temperature simulations sharply depends on :

- θ_h , the heating element temperature,
- h the heat exchange coefficient with the gaseous environment (boundary Γ_1),

- R_c the thermal resistance which has an effect on the dynamics of the temperature transfers on Γ_5 .

The heating element temperature can be measured or controlled. Then, in the following the identification of the unknown parameters (heat exchange coefficient and thermal resistance) is investigated.

1. INVERSE PROBLEM

Measurements of the temperature are required in order to identify the unknown parameter. In the following, the identification of the heat exchange on Γ_1 is considered. However the methodology exposed is quite general. Let us denote by $\varphi(\theta)$ the heat flux on Γ_1 , equations (3i) become :

$$-\lambda_i(\theta) \frac{\partial \theta}{\partial n_i} = \varphi(\theta) \quad (8i)$$

where $\varphi(\theta) = \sum_{i=1}^{N-1} \varphi_i \xi_i(\theta)$ is a continuous piecewise linear function.

According to this notation $\varphi(\theta)$ is known when $\bar{\varphi} = (\varphi_i)_{i=1, \dots, N-1}$ is identified. Then the following inverse problem is considered :

Problem P_{inv} :

find $\bar{\varphi} \in \mathbb{R}^{N-1}$ which minimizes the cost function :

$$J(\bar{\varphi}) = \frac{1}{2} \int_T \left(\sum_{j=1}^{n_{sens}} (\theta(x_j, t; \bar{\varphi}) - \hat{\theta}_j(t))^2 \right) dt$$

where n_{sens} is the number of sensor, $\hat{\theta}_j$ is the temperature measured at sensor j located on point x_j ; with the constraint : the temperature $\theta(x, t)$ is solution of the non linear distributed parameter system (S) :

$$(S) \begin{cases} \text{state equations} & (1i) \\ \text{initial condition} & (2) \\ \text{boundary conditions} & (8i) (4) (5) (6i) (7i) \end{cases}$$

The minimization of $J(\bar{\varphi})$ is performed by the following regularizing algorithms, see (Alifanov, 1994).

General conjugate gradient algorithm :

- a) initialisation $k=0$; let us denote by $\bar{\varphi}_0$ the given initial approximation of $\bar{\varphi}$ and the initial descent direction

$$\bar{d}_0 = -\nabla J(\bar{\varphi}_0) = - \left(\left(\frac{\partial J}{\partial \varphi_i} \right)_{i=1, \dots, N-1} \right) (\bar{\varphi}_0) ,$$

- b) at iteration k, from point $\bar{\varphi}_k$, the next point is obtained :

$$\bar{\varphi}_{k+1} = \bar{\varphi}_k + \gamma_k \bar{d}_k \text{ where :}$$

$$\gamma_k = \arg \min_{\gamma \in \mathbb{R}} \left(J(\bar{\varphi}_k + \gamma \bar{d}_k) \right) .$$

- c) the next direction is defined by : $\bar{d}_{k+1} = -\nabla J(\bar{\varphi}_{k+1}) + \beta_k \bar{d}_k$

$$\text{with : } \beta_k = \frac{\|\nabla J(\bar{\varphi}_{k+1})\|^2}{\|\nabla J(\bar{\varphi}_k)\|^2} .$$

- d) stopping of the iterative process if $J(\bar{\varphi}_{k+1}) \leq J_{stop}$ or :
 $k \leftarrow k+1$ and go to (b).

Calculation of the gradient is performed by introducing a Lagrangian formulation and solving an adjoint problem, see (Abou Khachfe and Jarny, 2001).

Calculation of the descent depth is performed by minimizing :

$$\min_{\gamma \in \mathbb{R}} \left(\frac{1}{2} \int_T \sum_{j=1}^{n_{\text{sens}}} (\theta(x_j, t; \bar{\varphi}_k + \gamma \bar{d}_k) - \hat{\theta}_j(t))^2 dt \right)$$

In order to minimize the previous function, sensitivity function are considered, see (Beck and Arnold, 1977). Then, it is shown that at iteration k of the conjugate gradient algorithm, the descent depth is :

$$\gamma_k = \frac{\int_T \left(\sum_{j=1}^{n_{\text{sens}}} \delta\theta(x_j, t; \bar{\varphi}_k) (\theta(x_j, t; \bar{\varphi}_k) - \hat{\theta}_j(t)) \right) dt}{\int_T \left(\sum_{j=1}^{n_{\text{sens}}} (\delta\theta(x_j, t; \bar{\varphi}_k))^2 \right) dt}$$

where $(\delta\theta)_j(x_j, t; \bar{\varphi}_k)$ is the solution of the sensitivity problem in the direction $\delta\bar{\varphi} = \bar{d}_k$.

Sensitivity problem is easily derived from the direct problem described by system (S).

Then the following algorithm is available for solving the inverse problem P_{inv} :

General conjugate gradient algorithm :

- initialisation $k = 0$; $\bar{\varphi}_0$ the given initial approximation,
- solve the direct problem (S) to compute $\theta(x, t; \bar{\varphi}_k)$ and the cost function $J(\bar{\varphi}_k)$,
- solve the adjoint problem to compute the components of the gradient in order to know the descent direction \bar{d}_k ,
- solve the sensitivity problem to compute the descent depth γ_k , and $\bar{\varphi}_{k+1}$
- stopping of the iterative process if $J(\bar{\varphi}_{k+1}) \leq J_{\text{stop}}$ is close to zero or : $k \leftarrow k + 1$ and go to (b).

Remarks :

- J_{stop} is a positive scalar which can be chosen according to the variance of the temperature measurement errors in order to avoid unstable solutions ; see the iterative regularizing principle (Alifanov, 1994).
- Direct problem, adjoint problem and sensitivity problem are established from non linear partial differential equations. The same numerical method (finite element) and software can be implemented.
- Whatever the numerical approach is, identification can lead to erroneous estimates due to sensor errors, see (Emery and Fadale, 1997). Methods for optimum sensor locations are generally based upon sensitivity analysis, see (Fadale et al., 1995). The problem under interest is to locate one or several sensors such that the estimation of $\varphi(\theta)$ is not adversely affected by errors in the measured state θ . In order to determine spatial location where sensitivity function is maximum, sensitivity equations can be considered, see an application in (Autrique et al., 2000).

5. CONCLUSION

In this communication, a Plasma Assisted Chemical Vapor Deposition reactor has been briefly presented. Since the temperature is one of the most important parameter of the elaboration process, a 3D modeling of the thermal evolution has been established. Direct problem, solved by a finite element method, leads to temperature evolution simulations whom analysis show on one hand that the thermal equilibrium time seems to be over estimated by experimenters, and on the other hand that substrate surface temperature are not uniform. This crucial information has to be carefully investigated before a technological transfer into industrial scaled reactors. Then, numerical design of experiments methodology is briefly exposed and used in order to determine which unknown parameters are influent and have to be identified. In such a way, an inverse problem has been stated (for the identification of the heat transfer occurring on one of the domain boundary). The conjugate gradient method is presented in a quite general formulation and requires the gradient calculation (Lagrangian formulation) and the descent depth calculation (sensitivity equations). It is important to notice that the sensitivity problem provides optimal sensor location in order to ensure the efficiency of the minimization algorithm. Then, several perspectives can be considered. We are mainly interested by the implementation of an adaptive control procedure in order to improve the heating controller (a classical PID is used for) during the plasma ignition phase.

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