Multiscale modeling and systemic analysis of chemical vapor deposition processes

Nikolaos Cheimarios

YIC2013
Second ECCOMAS Young Investigators Conference
2–6 September 2013, Bordeaux, France

Multiscale modeling and systemic analysis of chemical vapor deposition processes

N. Cheimarios

National Technical University of Athens, School of Chemical Engineering,
Zografou Campus, Athens 15780, Greece

nixeimar@chemeng.ntua.gr

Abstract. *Two multiscale computational frameworks are proposed for coupling/linking the co-existing scales in chemical vapor deposition (CVD)processes: From the macro-scale of a CVD reactor (~cm or m) to the transport inside micro-features (~μm) and kinetic Monte Carlo techniques in the nano-scale (~nm). To accelerate the computations parallel processing techniques are used. Moreover, a computational framework, based on projection type methods, is proposed to enable "black box" CFD codes to perform nonlinear systemic analysis tasks.*

Keywords: Mutliscale modeling; nonlinear analysis; parallel computing; chemical vapor derposition

1 INTRODUCTION

Chemical vapor deposition (CVD) is probably the most widely used process for producing thin solid films from gaseous reactants. CVD is performed in specially designed reactors equipped with heated surfaces, the wafers [see Fig. 1(a)], where the surface reactions occur, leading to the deposition of the solid film. The produced films via chemical vapor deposition (CVD) are utilized in a wide range of applications; from semiconductor devices to micro- and nano-electromechanical systems (MEMS and NEMS), and protein microarrays and chips.

1.1 Mutliscale modeling in chemical vapor deposition processes

Nowadays, the size of the fabricated devices or systems shrinks to lower scales and the specifications of the films, e.g. thickness, conformality (thickness uniformity on a patterned surface), surface morphology, refer to properties in micro- or nano-scale. Thus, the conventional single scale CVD modeling methods are not adequate and more advanced, multiscale modeling, methods are needed for studying the phenomena in the co-existing (multiple length) scales. For example, the filling of a micro-trench on the wafer and the pertinent film conformality come from the "interaction" of the macro- or reactor scale with the micro- or feature (trench) scale. Similarly, the nano-roughness developing on a film's surface during deposition comes from the interaction of the macro- or reactor scale with the submicro- or nano- or surface morphology scale.

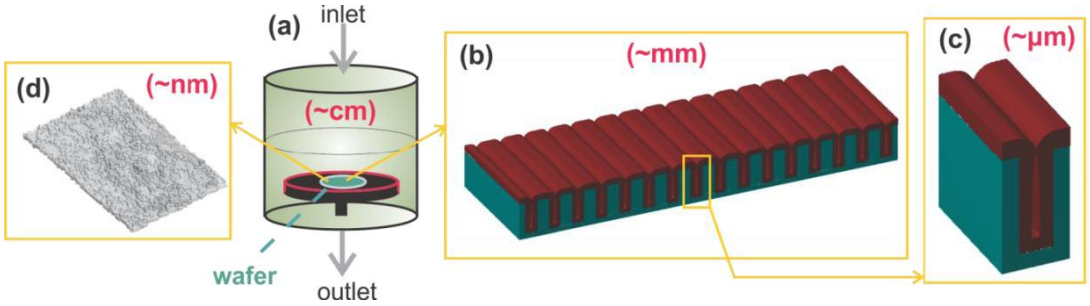

Figure 1: The length scales of a CVD process. (a) CVD reactor (macro-scale). (b) A cluster of trenches on the wafer. (c) A trench of the cluster (micro-scale). (d) Rough surface of a film deposited on an initially flat surface (submicro- or nano-scale).

The description of each scale requires a model: The reactor scale model (RSM), Fig.1(a), is used for the description of the transport phenomena in the bulk phase of the CVD reactor, the feature scale model (FSM), Figs.1(b), is used to describe the film deposition inside the features [e.g. trenches, Figs.1(c)] and the nano-morphology model (NMM), Fig.1(d), is used to trail the surface morphology of the deposited film. The effect of the operational parameters of a CVD reactor on the film profile evolution inside a trench on the wafer or on the film's surface nano-morphology evolution can be predicted by linking or coupling of RSM with FSM or NMM. The linking of models refers to their sequential use, while the coupling incorporates a two-way interaction of the models.

The RSM describes the transport phenomena in the macro-scale of the CVD reactor. The governing equations are the continuity, the momentum, the energy and the species transport equations. They are solved numerically at steady state to predict the velocity, pressure, temperature and species concentrations inside the bulk phase of the CVD reactor. The CFD code Fluent is used for the numerical solution of the aforementioned set of equations. The FSM results from coupling a local flux calculation model, a surface model and a profile evolution algorithm. The local flux calculation model is a ballistic model, which is formulated by a set of integral equations and it is valid at the high Knudsen number conditions occurring in the micro- trenches of the predefined topography. It links the fluxes of the species reaching the predefined topography on the wafer with the local fluxes inside the features (e.g. trenches) of the topography. The surface model describes the surface processes and quantifies the effect of species fluxes or concentrations on the local deposition rate. The profile evolution algorithm of the deposited film inside the features (moving boundary) uses the level set method. The NMM is based on the kinetic Monte Carlo method. All surface processes, i.e. adsorption, surface diffusion, surface reaction, desorption are modeled as a Markov process by transition probabilities per unit time. The transition probabilities per unit time are modeled by an Arrhenius expression and depend on local activation energies. The NMM is essentially a surface model as the surface model of the FSM; however, the surface model of the FSM is formulated by an analytical surface reaction rate expression (e.g. in terms of the reactive species concentration) and cannot predict the surface morphology.

The coupling of the RSM with FSM is based on the correction of consumption rates of each species on a predefined topography (e.g. an array of trenches) on the wafer. The aim of this correction is to take into account the increased consumption of species inside the topography, without the topography being included in the computational domain of macro-scale. A correction factor, ε_k [1], is applied to each surface reaction rate k, reflecting a change of the boundary condition of the species equation. The procedure is applied locally along the wafer radius, or in computational terms, on all the boundary cells of the RSM of the wafer. After convergence of the iterative scheme, film profile evolution inside the features is performed for a time step. The same procedure is followed for all time instances.

The linking [2] of the RSM with NMM starts with the numerical solution of the equations in the macro-scale at steady state. All species fluxes just above the wafer are fed to NMM. No bidirectional exchange of computational information between the scales is performed since the change of the surface nano-morphology is not expected to alter the species consumption on the wafer surface.

1.2 Accelerating multiscale computations in CVD processes

For solving the set of partial differential equations (PDEs) which describes the physical/chemical phenomena in the RSM the appropriate boundary conditions (BCs) are necessary. In the present multiscale computational framework, these BCs come from the solution of the problem in the different co-existing scales, i.e. the micro- or nano-scale on the wafer's surface. To handle the high computational demands, both in memory requirements and computational speed, a crossbred multi-parallel method (CMPM) [3] is proposed by combining (crossbreeding) different parallel techniques in the multiple scales of interest. In particular, domain decomposition techniques in the RSM are combined with a "master-worker" scheme to calculate the appropriate BCs for the aforementioned PDEs. These BCs come from computations with FSM or NMM and are changing during the simulation time. The use of master-worker parallel scheme is feasible since the computations with FSM or NMM can be performed independently for each boundary cell of the wafer. The multi-parallel term stems from the fact that different number of processors can be used to implement the domain decomposition and the master-worker schemes, an important factor since the computational demands in the different scales vary.

1.3 Systemic analysis of CVD processes

CFD codes are becoming standard in many fields of science and engineering involving flows of gases and liquids; numerical simulations are used from plasma processing and electromagnetics to catalysis and chemical vapor deposition processes. Despite their evolution, commercial CFD codes are practically unable to perform adequate systemic analysis, i.e. systematic detection and tracing of multiple solutions, to circumvent turning point singularities along solution branches and to compute unstable steady-state solutions as parameter vary. Therefore,

they fail to provide all the "pieces of the puzzle" i.e. of the dependence of solutions of nonlinear problems on key parameters. These shortcomings are serious limitations on the predictive capability of commercial CFD codes: missing pieces may hide crucial information about the limits of stability of solutions as well as entire branches of solutions which might be suggestive of advantageous operating "windows" of the process of concern.
A computational framework [4] is proposed for enabling a CFD "black box" code, such as Fluent, to converge past turning points on unstable steady states branches and therefore successfully trace an entire solution branch. The proposed framework is based on the so-called recursive projection method (RPM) which is implemented as a computational shell around the CFD commercial code Fluent. The main purpose is to enable Fluent to trace solution branches of nonlinear problems that have multiple solutions in a systematic and efficient way, and even induce convergence on unstable steady states. The RPM is combined with arc-length type continuation techniques in order to enable convergence on unstable solutions past turning points, offering therefore the possibility to trace entire solution branches. As a by-product it also delivers approximations of the critical eigenmodes of the discretized physical problem, through the solution of a low-dimensional eigenvalue problem. This proves useful in problems with bifurcation points, where the derived eigenvectors can be used for solution branch switching. Finally, it reduces the computational cost, when used in the context of parameter continuation.

2 REPRESENTATIVE RESULTS

Representative results from coupling RSM with FSM and linking and RSM with NMM are shown in Fig. 2. The effect of a predefined topography on the gas phase composition inside the CVD reactor is demonstrated in Fig. 2(a-b). In Fig. 2(a-b) conventional CVD computations are compared to multiscale computations from a macro-scale point of view [5]. In the multiscale computations, the consumption of the reactants in the wafer's surface increases due to the increased for deposition area owing to of the existence of the micro-topography. As a consequence, the mole fraction of the reactants decreases [Fig.2(b)], especially near the wafer's surface, compared to the conventional single scale computations [Fig.2(b)]. The results from the multiscale computations are shown for t = 0s where the trenches are empty. Figs. 2(c-d) shows two trenches upon filling [5]. Depending on the operating conditions of the CVD reactor conformal [Fig. 2(d)] or non-conformal (void formation) [Fig. 2(c)] deposition is observed. Concerning the linking of the RSM with NMM the results [2] focus on the epitaxial deposition of a crystalline film of Si on a Si (001)2x1 surface where dimmers are formed. In Fig. 2(e) the surface morphology at the center of the wafer after deposition for 1.5 s is shown. In Fig. 2(f) the same is shown at the edge of the wafer. The center and the edge positions correspond to the maximum and minimum deposition rate as computed by the RSM. Due to the difference in the deposition rates the number of monolayers deposited after 1.5 s differs and leads to surfaces with different cluster's orientation. The multiscale computations are accelerated by using the CMPM. Still, as a characteristic example, exploiting only the master-worker parallel technique the multiscale computations concerning RSM/FSM are accelerated from 2 ½ days to 5 hours [6].

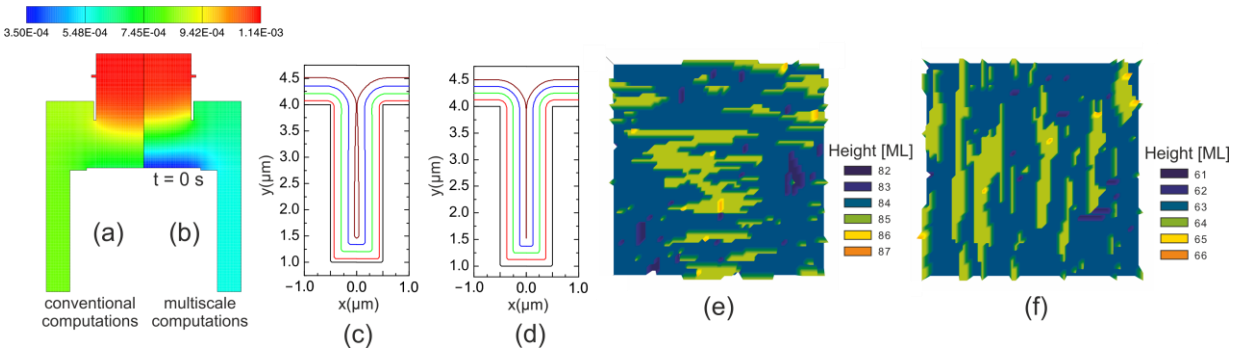

Figure 2: Case study: silicon (Si) CVD from silane (SiH_4) [2][7]. *Coupling RSM with FSM.* (a-b) Conventional vs multiscale computations. Contours of the SiH_4 mole fraction for (a) conventional (single scale) and (b) multiscale computations. (c-d) Repletion of trenches (c) void formation (high deposition temperatures) and (d) uniform (low deposition temperatures). *Linking RSM with NMM.* (e-f) Nano-morphology of an initially flat Si surface. Different dimmers orientation are observed depending on the computed deposition rate from the RSM (e) maximum deposition rate (wafer's center) and (f) minimum deposition rate (edge of the wafer).

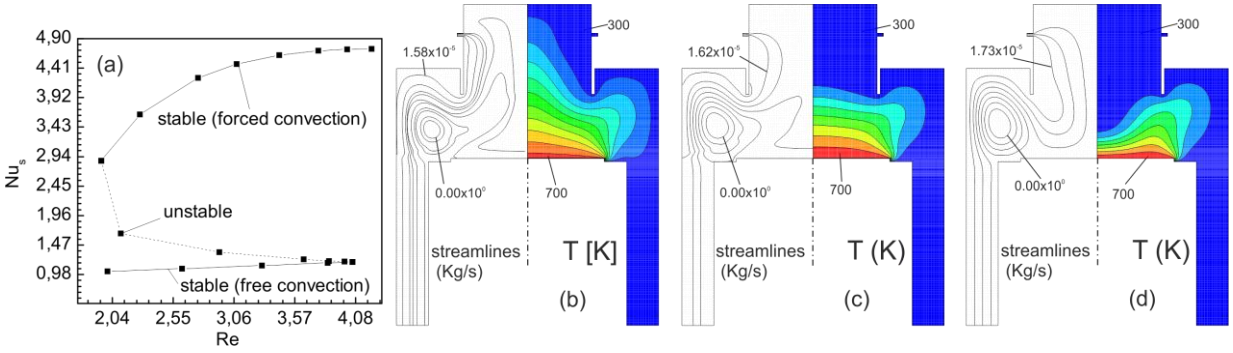

Figure 3: Arc-length RPM assisted Fluent computational framework. (a) Solution space and the corresponding solutions (streamlines and temperature contours) in the different solution branches (b) free convection (stable), (c) unstable and (d) forced convection (stable) for the same operating conditions of the reactor.

In Fig. 3 representative results, obtained by applying arc-length RPM assisted Fluent method, are presented. The upper stable branch contains solutions where the dominant mechanism is forced convection whereas the lower stable solution branch is dominated by free convection. It is through the competition between free and forced convection, that multiplicity arises and in practice this can influence greatly the end result, i.e. the film growth and uniformity [5][7]. As seen in Figs. 3(b)(c)(d), where the flow and temperature fields are plotted for the same parameter value, at different branches, the solution is affected significantly by the dominant mechanism.

3 CONCLUSIONS

Concerning the multiscale modeling aspects of my doctoral thesis, different models are combined to couple/link the physical/chemical phenomena in the multiple scales of interest in CVD: From the continuum regime (~cm/m) of the macro-scale of a CVD reactor to ballisctic transport inside micro-features (~μm) and kinetic Monte Carlo techniques in the nano-scale (~nm). The computations are performed in parallel in high performance computational clusters. Concerning the systemic analysis of CVD processes, a computational framework is proposed which efficiently combines projection type methods with "black box" CFD codes (such as Fluent) to enable them to perform nonlinear analysis tasks. Besides the computational challenges for developing the aforementioned computational frameworks, the application of the frameworks in the analysis of realistic CVD problems, yields a plethora of interesting physical results and leads to new perspectives for interpreting the physical/chemical mechanisms encounter in deposition processes.

ACKNOWLEDGEMENT

I am indebted to my doctoral thesis advisor A. G. Boudouvis, Professor and Dean of the School of Chemical Engineering. I am grateful to Dr. G. Kokkoris for his guidance in multiscale modeling, to Dr. E.D. Koronaki for her advice with RPM and to Dr. A.N. Spyropoulos for his help with parallel computing.

PUBLISHED WORK

[1] Cheimarios, N., Kokkoris G., Boudouvis A.G., Multiscale modeling in chemical vapor deposition processes: Coupling reactor scale with feature scale computations. *Chemical Engineering Science* 65:5018-5028, 2010.
[2] Cheimarios N., Garnelis S., Kokkoris G., Boudouvis A.G., Linking the operating parameters of CVD reactors with filling conformality and surface nano-morphology. *Journal of Nanoscience and Nanotechnology* 11: 8132-8137, 2011.
[3] Cheimarios N., Spyropoulos A.N., Kokkoris G., Boudouvis A.G., 'Parallel multiscale computations in Chemical Vapor Deposition processes." In: *Proceedings of the Seventh GRACM International Congress on Computational Mechanics,* Athens, Greece, 30 June-2 July 2011.
[4] Cheimarios N., Koronaki E.D., Boudouvis A.G., Enabling a commercial computational fluid dynamics code to perform certain nonlinear analysis tasks." *Computers & Chemical Engineering* 35:2632-2645, 2011.
[5] Cheimarios N., Kokkoris G., Boudouvis A.G., Multiscale computational analysis of the interaction between the wafer micro-topography and the film growth regimes in chemical vapor deposition processes." *ECS Journal of Solid State Science and Technology* 1:P197-P203, 2012.
[6] Cheimarios N., Kokkoris G., Boudouvis A.G., An efficient parallel iteration method for multiscale modeling of chemical vapor deposition processes. *Applied Numerical Mathematics* 67:78-88, 2013.
[7] Cheimarios N., Koronaki E.D., Boudouvis A.G., Illuminating nonlinear dependence of film deposition rate in a CVD reactor on operating conditions. *Chemical Engineering Journal* 181-182:516-523, 2012.