Multiscale Modeling and Run-to-Run Control of PECVD of Thin Film Solar Cells

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Abstract

In this work, we focus on the development of a multiscale modeling and run-to-run control framework with the purpose of improving thin film product quality in a batch-to-batch plasma-enhanced chemical vapor deposition (PECVD) manufacturing process. Specifically, at the macroscopic scale, gas-phase reaction and transport phenomena yield deposition rate profiles across the wafer surface which are then provided to the microscopic domain simulator in which the complex microscopic surface interactions that lead to film growth are described using a hybrid kinetic Monte Carlo algorithm. Batch-to-batch variability has prompted the development of an additional simulation layer in which an exponentially weighted moving average (EWMA) control algorithm operates between serial batch deposition sequences to adjust the operating temperature of the PECVD reactor to overcome drift in the electron density of the plasma. Application of the run-to-run (R2R) control system developed here is shown to reduce offset in the product thickness from 5% to less than 1% within 10 batches of reactor operation. Finally,

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Preprint submitted to Renewable Energy

June 29, 2016

we propose an extension of the EWMA algorithm to four independent, radial wafer zones in order to improve thickness uniformity in the presence of spatially non-uniform species concentrations. It is demonstrated that the produced thin films can be driven to the desired thickness set-point of 300 nm in less than 10 batches in the presence of both electron density drift and non-uniform deposition rate profiles.

Keywords:

Process modeling; plasma-enhanced chemical vapor deposition; thin film solar cells; multiscale modeling; parallel computing; run-to-run control.

1. Introduction

Plasma enhanced chemical vapor deposition (PECVD) remains the dominant mechanism by which silicon based thin films are produced in both the microelectronics and solar cell industries (Kern, 1991). Although moderate manufacturing costs and the possibility of extremely low operating temperatures (≤ 525 K) allow for the widespread use of batch PECVD processing (Kreiger et al., 2013; Yang et al., 2000), two often neglected problems are persistent in the production of high quality amorphous silicon (a -Si:H) layers: the influence of reactor conditioning on the plasma chemistry (Gabriel et al., 2014), and the spatial non-uniformity in the thin film thickness (Armaou and Christofides, 1999) owing to gas-phase transport phenomena across the wafer. Specifically, during the initial operation of a clean PECVD reactor the plasma phase exhibits transient behavior as the interior surfaces become coated by the deposition species causing drift in the electron density profiles and in the film thickness. Second, at the reactor length

47 scale (for example, a 20 cm wafer is used in this work) consumption and transport
48 of deposition species across the wafer surface have been shown to cause growth
49 rate differences greater than 19% (Stephan et al., 1999; Sansonnens et al., 2003).
50 Given that the efficiency of a photovoltaic absorber layer (e.g., an *a*-Si:H thin
51 film) is strongly dependent on the film thickness (Kabir et al., 2012), reduction of
52 growth rate non-uniformities is of paramount importance. While preconditioning
53 prior to thin film deposition (i.e., allowing the reactor surfaces to become fouled)
54 is a well-established practice, this represents significant waste in both valuable
55 resources and manufacturing time. In addition, the film uniformity within a sin-
56 gle batch cannot be corrected via preconditioning alone, and has been shown to
57 require advanced, in-situ processing techniques (Croese et al., 2015).

58 Recently much attention has been given to the development of model-based
59 control schemes with the goal of improving solar cell performance through the
60 production of textured surfaces (e.g., thin film grating) (Huang et al., 2012a,b).
61 These models typically rely on kinetic Monte Carlo (kMC) algorithms to simulate
62 deposition processes and therefore focus on the evolution of surface microstruc-
63 ture in nano- to micrometer length scales. As a result, these models are unable
64 to capture the reactor scale dynamics and cannot be applied to the problems men-
65 tioned previously which pertain to both the macroscopic and microscopic domains
66 of batch PECVD reactor operation. Fortunately, advances in high-performance
67 computing have made possible the modeling of multiscale processes with com-
68 plex behavior and large system sizes (Maroudas, 2000; Kevrekidis et al., 2004;
69 Vlachos, 2005). In a pervious work of our group (Croese et al., 2015), we de-

70 developed a multiscale model which provided fundamental understanding of the
71 dynamics involved in the PECVD of α -Si:H thin films and allowed for quantita-
72 tive prediction of product quality. In the present work, we present improvements
73 to our original model, as well as an additional simulation domain that allows for
74 run-to-run (R2R) control of the batch process in an effort to counteract both batch-
75 to-batch and spatial variations in the film thickness.

76 More specifically, this work proposes a multiscale modeling and operation
77 framework which is capable of not only capturing the interdependence of the gas-
78 phase and film growth phenomena, but also allows for multi-batch operation under
79 the implementation of a run-to-run (R2R) control algorithm. Within a single batch
80 simulation a standard gas-phase model is used; however, the microscopic model,
81 describing the α -Si:H thin film surface evolution, has been developed from the
82 work of Tsalikis et al. (Tsalikis et al., 2013) to be computationally efficient and
83 account for the four dominant microscopic processes: physisorption, surface mi-
84 gration, hydrogen abstraction, and chemisorption. As opposed to traditional kMC
85 formulations, surface migration has been decoupled from the other microscopic
86 surface processes owing to its faster rate to allow for simulations on the order of
87 thousands of monolayers without compromising fidelity to established chemical
88 models.

89 At the completion of each batch simulation, a novel R2R control algorithm
90 is applied in order to predict appropriate operating conditions for the upcoming
91 batch. In other words, post-batch measurements of the film thickness at various
92 radial positions are fed to an exponentially weighted moving average (EWMA)

93 algorithm which in turn updates the temperature of the PECVD reactor within
94 discrete zones. It is demonstrated that through appropriate tuning of the mul-
95 tiscale model, and application of the proposed R2R operation strategy, the thin
96 film product can be driven to the desired thickness set-point regardless of radial
97 position and drift within the PECVD reactor.

98 The structure of the manuscript is as follows: first, a detailed description of
99 both the macroscopic gas-phase and microscopic surface models are provided.
100 Next, extensive simulations demonstrate strong agreement between experimen-
101 tally grown *a*-Si:H films and those resulting from the multiscale model proposed
102 in this work. A total of 40 serial batch simulations conducted using the nomi-
103 nal process parameters (i.e., $T = 475$ K and $P = 1$ Torr) then reveal drift in the
104 product away from the desired film thickness of 300 nm. The proposed EWMA
105 algorithm is then applied within a single reactor zone and shown to successfully
106 reduce film offset to within the process noise level. Finally, four concentric reactor
107 zones are defined, each with a corresponding R2R controller and initial gas-phase
108 concentration. In the presence of both plasma variation caused by conditioning
109 of the reactor and spatial non-uniformity caused by consumption of the process
110 gas, the proposed control scheme demonstrates significantly improved thickness
111 uniformity regardless of radial position within the reactor.

112 **2. Process description and modeling**

113 The process under consideration in this work is a parallel plate PECVD reactor
114 utilizing two charged electrodes designed to deposit thin films onto a single wafer

Figure 1: Macroscopic (left) and microscopic (right) PECVD simulation regimes.

placed on top of the lower electrode; see Fig. 1 (left). We employ a showerhead
arrangement to distribute the influent gas stream consisting of a 9:1 mixture of hy-
drogen (H_2) and silane (SiH_4) throughout the chamber. Thin film growth proceeds
through the production of plasma by an radio frequency (RF) power source which
generates a chemically reactive mixture of radicals. These radicals (namely, SiH_3
and H) are transported to the surface via diffusion and convection where they react
to form amorphous silicon ($a\text{-Si:H}$). Successful deposition requires the uniform
growth of a 300 nm thick $a\text{-Si:H}$ film on a wafer with a diameter of 20 cm.

Two distinct simulation regimes exist within this framework: the macroscopic
gas phase which includes mass and energy balances, as well as the complex, mi-
croscopic surface interactions that dictate the structure of the silicon film of inter-
est. Fig. 1 highlights the multiscale character of this process and the need to cap-
ture the dynamics at both scales due to the codependency between the macroscopic
and microscopic regimes. The following sections detail both the macroscopic
gas-phase model and the microscopic surface model.

130 2.1. Gas-phase model

131 The twelve dominant species that lead to film growth and their corresponding
 132 thirty-four gas-phase reactions are accounted for throughout this work. A com-
 133 plete listing of the reactions, mechanisms and rate constants are available in Table
 134 1. The first set of results presented in this work, within the open-loop operation
 135 section, assume that the process gas is well mixed and the rate constants do not
 136 depend on temperature. For this case a differential mass balance is used to nu-
 137 merically integrate the species concentrations forward with time and the resulting
 138 values are used without reference to the spatial location within the PECVD reac-
 139 tor. The following differential equation demonstrates the possible contributions to
 140 the mass balance for a given species:
 141

$$\frac{dc_i}{dt} = \frac{c_i^{in}}{t_{in}} - \frac{c_i}{t_{out}} + \left[\sum_j \nu_i^j R^j \right] - \frac{1}{N_a V} r_{phys,i} + \frac{1}{N_a V} r_{abs,i}, \quad (1)$$

142 where c_i is the concentration in mol/cm^3 of species i , t_{in} is the inlet gas time
 143 constant defined as the reactor volume over the flow rate into the reactor, t_{out} is
 144 the outlet stream time constant defined as the reactor volume over the flow rate
 145 out of the reactor, ν_i^j is the stoichiometric coefficient for reactant i in reaction j
 146 as listed in Table 1, $r_{phys,i}$ is the rate of physisorption of species i , and $r_{abs,i}$ is the
 147 rate of abstraction of species i from the wafer surface.

148 The second set of results presented in this work, those referring to spatial non-
 149 uniformity in film growth, utilize a gas-phase model that includes energy and mo-
 150 mentum balances in addition to the differential mass balance presented above. Un-
 151 der the assumptions of axisymmetric flow and continuum in the gas phase, mass,

Table 1: Reactions included in the gas-phase model. Note: Rate constants have units of cm^3/sec and have been adopted from the collection prepared by Kushner et al. (Kushner, 1988).

Reaction	Mechanism	Rate constant
R^1	$e^- + H_2 \rightarrow 2H$	7.66×10^{12}
R^2	$e^- + SiH_4 \rightarrow SiH_3 + H$	9.57×10^{13}
R^3	$e^- + SiH_4 \rightarrow SiH_3^+ + H$	3.40×10^{12}
R^4	$e^- + SiH_4 \rightarrow SiH_2 + 2H$	1.13×10^{13}
R^5	$e^- + SiH_4 \rightarrow SiH + H_2 + H$	5.62×10^{12}
R^6	$e^- + SiH_4 \rightarrow Si + H_2 + 2H$	6.70×10^{12}
R^7	$e^- + Si_2H_6 \rightarrow SiH_3 + SiH_2 + H$	2.15×10^{13}
R^8	$e^- + Si_2H_6 \rightarrow H_3SiSiH + 2H$	7.41×10^{13}
R^9	$e^- + Si_3H_8 \rightarrow H_3SiSiH + SiH_4$	3.35×10^{14}
R^{10}	$H + SiH_2 \rightarrow SiH_3$	6.68×10^{11}
R^{11}	$H + SiH_2 \rightarrow SiH + H_2$	1.20×10^{13}
R^{12}	$H + SiH_3 \rightarrow SiH_2 + H_2$	1.20×10^{13}
R^{13}	$H + SiH_4 \rightarrow SiH_3 + H_2$	1.38×10^{12}
R^{14}	$H + H_2Si = SiH_2 \rightarrow Si_2H_5$	3.01×10^{12}
R^{15}	$H + Si_2H_6 \rightarrow SiH_4 + SiH_3$	4.03×10^{12}
R^{16}	$H + Si_2H_6 \rightarrow Si_2H_5 + H_2$	7.83×10^{12}
R^{17}	$H + Si_3H_8 \rightarrow Si_2H_5 + SiH_4$	1.19×10^{12}
R^{18}	$H_2 + SiH \rightarrow SiH_3$	1.20×10^{12}
R^{19}	$H_2 + SiH_2 \rightarrow SiH_4$	1.20×10^{11}
R^{20}	$SiH_2 + SiH_4 \rightarrow Si_2H_6$	6.02×10^{12}
R^{21}	$SiH_3 + SiH_3 \rightarrow SiH_4 + SiH_2$	4.22×10^{12}
R^{22}	$SiH_3 + SiH_3 \rightarrow Si_2H_6$	6.02×10^{12}
R^{23}	$SiH + SiH_4 \rightarrow Si_2H_5$	1.51×10^{12}
R^{24}	$SiH_2 + SiH_4 \rightarrow H_3SiSiH + H_2$	6.02×10^{12}
R^{25}	$SiH_2 + Si_2H_6 \rightarrow Si_3H_8$	7.23×10^{13}
R^{26}	$SiH_2 + SiH_3 \rightarrow Si_2H_5$	2.27×10^{11}
R^{27}	$SiH_3 + SiH_3 \rightarrow SiH_4 + SiH_2$	4.06×10^{13}
R^{28}	$SiH_3 + Si_2H_6 \rightarrow SiH_4 + Si_2H_5$	1.98×10^{13}
R^{29}	$Si_2H_5 + SiH_4 \rightarrow SiH_3 + Si_2H_6$	3.01×10^{11}
R^{30}	$SiH_3 + Si_2H_5 \rightarrow Si_3H_8$	9.03×10^{13}
R^{31}	$H_3SiSiH + SiH_4 \rightarrow Si_3H_8$	6.02×10^{12}
R^{32}	$Si_2H_5 + Si_2H_5 \rightarrow Si_3H_8 + SiH_2$	9.03×10^{13}
R^{33}	$H_3SiSiH \rightarrow H_2Si = SiH_2$	2.71×10^{13}
R^{34}	$H_2Si = SiH_2 \rightarrow H_3SiSiH$	2.29×10^{10}

energy and momentum balances allow for the detailed modeling of the gas phase required in order to investigate spatial non-uniformities. The governing equations have been developed at length for CVD-type applications (e.g., (Vlachos, 1997; Lam and Vlachos, 2001; Christofides and Armaou, 2006)); however, here we apply the formulation by Armaou and Christofides (Armaou and Christofides, 1999) as radial dependence of species concentrations is needed to be accounted for due to the strong dependence of thin film thickness on photovoltaic efficiency. The mathematical model of the PECVD reactor consists of a flow velocity profile and four nonlinear dynamic diffusion-convection-reaction equations in two (radial and axial) dimensions (a set of four coupled parabolic PDEs). By treating the gas-phase as a three-dimensional stagnation flow, the evolution of the flow velocity profile within the PECVD reactor is computed from the Navier-Stokes and continuity equations. The spatio-temporal evolution of the concentration of the species throughout the reactor is obtained by applying dynamic material balances to the gas phase and accounting for diffusive and convective mass transfer, and bulk and surface reactions. Finite-difference methods are then applied in order to discretize the spatial derivative of the species concentration in the r and z directions (z is defined as the direction normal to the wafer surface; see Fig. 1). Finally, time integration of the resulting ordinary differential equations is performed using the alternate direction implicit (ADI) method. More details on the model structure can be found in Armaou and Christofides (Armaou and Christofides, 1999), and calculated steady-state concentration profiles are discussed further in the spatial non-uniformity results section.

175 2.2. Surface microstructure model

176 The kMC methodologies applied in this work follow closely that of Crose
 177 et al. (Crose et al., 2015) and share many features common to standard kinetic
 178 Monte Carlo processes; however, simulation results generated by the multiscale
 179 model can vary widely due to discrepancies in the physical phenomena and model
 180 parameters used. As such, details of the microscopic surface interactions are pre-
 181 sented here in full, starting with the thin film chemistry and growth kinetics.

182 2.2.1. Thin film growth chemistry

183 At the operating conditions of interest ($T = 475$ K and $P = 1$ Torr) two species
 184 dominate the interactions on the growing film, SiH_3 and H ; the remaining species
 185 that exist in the gas-phase model are prohibited from depositing on the lattice sur-
 186 face and as a result are ignored for the remainder of the microscopic model. A
 187 schematic of the surface interactions is shown in Fig. 2, and has been verified
 188 experimentally by Perrin et al. (Perrin et al., 1998) and Robertson (Robertson,
 189 2000). Physisorption occurs as SiH_3 and H radicals encounter hydrogenated sili-
 190 con sites ($\equiv \text{Si} - \text{H}$) at the film surface according to the following reactions:

191 Migration involves the rapid diffusion of physisorbed radicals across the lattice
 192 surface:

193 eventually contributing to either hydrogen abstraction:

Figure 2: Chemical model illustration showing particle-surface interactions.

194 whereby a physisorbed SiH_3 radical removes a surface hydrogen forming SiH_4
 195 and creating a dangling bond ($\equiv \text{Si}^0$), or chemisorption at a preexisting dangling
 196 bond site according to the following reactions:

197 Chemisorption of SiH_3 at a dangling bond site permanently grows the lattice po-
 198 sition by one (i.e., the Si atom is fixed at that location and is no longer a candidate
 199 for migration), whereas chemisorption of H results in the surface site returning to
 200 a hydrogenated state.

201 2.2.2. Lattice Characterization

202 In our recent work (Croese et al., 2015), a solid-on-solid (SOS) lattice was
 203 implemented; specifically, particles in each successive monolayer were centered
 204 above those of the previous layer and the occupation of vacant postilions was en-
 205 forced in order for the height of a given lattice column to increase. Alternatively,
 206 a two-dimensional triangular lattice is used in this work which allows for the de-
 207 position of SiH_3 and H particles (each deposited species is modeled by a single
 208 particle) without the restriction of SOS behavior. While an SOS square lattice

209 does not allow for the creation of voids and porous structure within the film, a
 210 triangular lattice can produce overhangs that lead to voids while maintaining a
 211 minimum of two nearest neighbors per particle (see Fig. 3).

212 The size of the two-dimensional lattice is characterized by the number of lat-
 213 eral sites, L , and the number of monolayers, H . Using a silicon diameter of ~ 0.25
 214 nm, the physical length of the lattice can be calculated as $0.25 \times L$. Similarly, the
 215 thickness is calculated by:

$$\tau = 0.25 \cdot H \cdot \frac{\sqrt{3}}{2}, \quad (6)$$

216 where the factor $\sqrt{3}/2$ accounts for the reduction in thickness due to the offset
 217 monolayers (refer to Figs. 3 and 4). The number of lateral sites is chosen to be
 218 1200 in this work as it allows for the morphology of the film to be adequately
 219 captured without being so large as to necessitate spatial variations in the gas-
 220 phase. It is important to note here that although spatial variations in the gas-phase
 221 species concentrations are introduced in later sections, they are applied at the
 222 macroscopic scale (i.e., between reactor zones) not within individual microscopic
 223 kMC simulations.

224 In order to enforce the restriction that all physisorbed and chemisorbed parti-
 225 cles exist in stable, predetermined lattice locations, surface relaxation is included
 226 in the microscopic model. As shown in Fig. 4, an incident particle must first relax
 227 to a position that is centered within the defined triangular lattice. Additionally,
 228 each particle must meet the criteria of at least two nearest-neighbors to be con-
 229 sidered stable. Unstable particles continue to relax down the lattice until a stable

Figure 3: Triangular lattice representation showing four microscopic processes. Processes from left to right: migration, physisorption, chemisorption, and hydrogen abstraction.

Figure 4: Surface relaxation for physisorbed radicals. (a) Incident particle location. (b) Surface Si particle in lattice. (c) Predefined triangular lattice site with one nearest neighbor. (d) Stable position for incident particle (two nearest neighbors).

location is reached.

2.2.3. Relative rates formulation

A standard Arrhenius-type formulation can be used to estimate the rate of thermally activated kinetic events (e.g., migration and hydrogen abstraction):

$$r_{t,i} = v_i e^{-E_i/k_B T}, \quad (7)$$

where v_i is the attempt frequency prefactor (s^{-1}) and E_i is the activation energy of radical i . Frequency prefactor and activation energy values are drawn from Perrin et al. (Perrin et al., 1998) to correspond to the growth of a -Si:H films via the two

237 species deposition of SiH_3 and H.

238 Physisorption events follow an athermal or barrierless reaction model based on
239 the fundamental kinetic theory of gases which yields the following rate equation:

$$r_{a,i} = J_i s_c N_a \sigma, \quad (8)$$

240 where J is the flux of gas-phase radicals, s_c is the local sticking coefficient, N_a
241 is the Avogadro number, and σ is the average area per surface site. J can be
242 calculated via the following equations:

$$J_i = \eta_i \bar{u}_i, \quad (9)$$

243

$$\eta_i = \frac{p_i}{RT}, \quad (10)$$

244

$$\bar{u}_i = \sqrt{\frac{8k_B T}{\pi m_i}}, \quad (11)$$

245 where η_i is the number density of radical i (here the reactive gas-phase is assumed
246 to be ideal), $\bar{u}_i$ is the mean radical velocity, p_i is the partial pressure of i , R the
247 gas constant, T is the temperature, k_B is the Boltzmann constant, and m_i is the
248 molecular weight of radical i . Substitution of Eqs. (9)-(11) into Eqn. (8) provides
249 the overall reaction rate for an athermal radical i :

$$r_{a,i} = \frac{p_i}{RT} \sqrt{\frac{8k_B T}{\pi m_i}} s_c N_a \sigma. \quad (12)$$

250 Reaction rates and sticking coefficients for the nominal process conditions
251 (i.e., $T = 475$ K and $P = 1$ Torr) are given in Table 2. Sticking coefficients are re-
252 ported for athermal reactions rather than reaction rates as these are not dependent

Table 2: Sticking coefficients and reaction rates composing the chemical model.

Thermally activated		Athermal reactions	
Reaction type	Reaction rate ($\text{s}^{-1}\text{site}^{-1}$)	Reaction type	Sticking coefficient
hydrogen abstraction	3.35×10^5	SiH ₃ physisorption	0.5
hydrogen migration	1.58×10^{11}	hydrogen physisorption	0.8
SiH ₃ migration	1.16×10^{11}		

on the gas-phase composition which varies significantly from batch to batch. The microscopic surface kinetics presented in this work have been developed from the detailed work of Tsalikis et al. (Tsalikis et al., 2013).

2.2.4. Kinetic Monte Carlo implementation

The time evolution of the lattice microstructure is simulated using a standard n-fold kinetic Monte Carlo algorithm. The overall reaction rate is defined as

$$r_{\text{total}} = r_a^{\text{SiH}_3} + r_a^{\text{H}} + r_t^{\text{abs}}, \quad (13)$$

where $r_a^{\text{SiH}_3}$ is the rate of physisorption of SiH₃, r_a^{H} is the rate of physisorption of H, and r_t^{abs} is the rate of hydrogen abstraction forming SiH₄. Surface migration does not factor into the overall rate as it has been decoupled in the interest of computational efficiency and will be discussed at length in the next subsection. At the beginning of each kMC event a uniform random number, $\gamma_1 \in [0, 1]$ is generated. If $\gamma_1 \leq r_a^{\text{SiH}_3}/r_{\text{total}}$, then an SiH₃ physisorption event is executed. If $r_a^{\text{SiH}_3}/r_{\text{total}} < \gamma_1 \leq (r_a^{\text{SiH}_3} + r_a^{\text{H}})/r_{\text{total}}$, then a hydrogen radical is physisorbed. Finally, if $\gamma_1 > (r_a^{\text{H}} + r_t^{\text{abs}})/r_{\text{total}}$, then the abstraction of a surface hydrogen via SiH₃ occurs.

Both physisorption of SiH₃ and H start by selecting a random lattice site from a list of the available hydrogenated and dangling bond surface sites. If the chosen

270 site contains a dangling bond, the radical becomes instantaneously chemisorbed
 271 causing the lattice to grow by one; otherwise, the radical is deposited on the lattice
 272 surface where it relaxes to a stable position (see Fig. 4) before becoming a candi-
 273 date for migration events. Hydrogen abstraction proceeds by removing a random
 274 SiH_3 particle to form SiH_4 , which is returned to the gas-phase and execution con-
 275 tinues. Next, a second random number, γ_2 , is chosen and the time step for this
 276 kMC event is calculated as

$$\delta t = \frac{-\ln(\gamma_2)}{r_{\text{total}}}, \quad (14)$$

277 where $\gamma_2 \in (0, 1]$ is a uniform random number.

278 2.2.5. *Decoupling surface migration*

279 Figure 5 highlights the disparity in the frequency of migration events rela-
 280 tive to other event types. As a result, more than 99% of computational resources
 281 would be spent on migration alone in a traditional brute force kMC algorithm.
 282 At the standard operating conditions of $T = 475$ K and $P = 1$ Torr, it is clear
 283 that only a small fraction of simulation time contributes to events leading to film
 284 growth; consequently, the computation of rapid particle migration is decoupled
 285 from our standard n-fold kMC implementation using a two-dimensional lattice
 286 random walk process. A kMC cycle is defined by a single physisorption or ab-
 287 straction event. At the completion of each cycle, a propagator is introduced to
 288 monitor the motion of physisorbed radicals. The total number of propagation
 289 steps is $N_H + N_{\text{SiH}_3}$ where

$$N_H = \frac{r_t^H}{r_a^H + r_t^{\text{abs}} + r_a^{\text{SiH}_3}}, \quad N_{\text{SiH}_3} = \frac{r_t^{\text{SiH}_3}}{r_a^H + r_t^{\text{abs}} + r_a^{\text{SiH}_3}}, \quad (15)$$

Figure 5: Normalized frequency of reaction events within the present kMC scheme at $T = 475$ K, $P = 1$ Torr, and a SiH_4^{in} mole fraction of 0.9.

290 and r_t^H and $r_t^{\text{SiH}_3}$ are the thermally activated migration rates of hydrogen and
 291 silane radicals, respectively. The N_H and N_{SiH_3} propagation steps are split evenly
 292 among the n_H and n_{SiH_3} physisorbed radicals. Each radical then undergoes a
 293 two-dimensional random walk process that approximates the intricate movements
 294 of an individual particle with the bulk motion of the propagator. The procedure of
 295 the random walk process is as follows: a radical type is chosen, a random radical
 296 of the given type is selected, the weighted random walk with N_i/n_i propagation
 297 steps begins, propagation continues until either N_i/n_i steps have occurred or the
 298 radical becomes chemisorbed at a dangling bond site, the final position of the
 299 propagator is then stored as the radical's new position and this cycle continues for

all $n_H + n_{SiH_3}$ physisorbed species. It is important to note that the weighting of each propagation step is such that an exponentially higher probability exists for a particle to relax down the lattice as opposed to jumping up lattice positions. In a similar manner to physisorption and hydrogen abstraction, the time increment for an individual migration step is calculated via the following equations:

$$\delta t_H = \frac{-\ln(\gamma_i)}{r_t^H}, \quad \delta t_{SiH_3} = \frac{-\ln(\gamma_j)}{r_t^{SiH_3}}. \quad (16)$$

Therefore, the total time required for all migration events, δt , is determined to be

$$\Delta t = \sum_i^{N_H} \frac{-\ln(\gamma_i)}{r_t^H} + \sum_j^{N_{SiH_3}} \frac{-\ln(\gamma_j)}{r_t^{SiH_3}}. \quad (17)$$

Film growth continues to develop under the hybrid kMC algorithm until the time allotted for a given batch has elapsed, t_{batch} .

Our methodology of decoupling the diffusive processes from the remaining kinetic events has been validated by confirming that the underlying lattice random walk process results: (1) in surface morphologies and film porosities appropriate for the chosen process parameters, and (2) growth rates on par with experimental values. Details of our model validation strategy are given in the following sections.

3. Parallel computation

Due to the size of the multiscale simulations presented in this work, the computational demands are non-trivial. Specifically, deposition of a 300 nm thick film with a horizontal dimension of 300 nm requires a simulation time on the order of

318 days when using a single processor. The results presented in this work represent
319 many successive batches, as well as data that has been averaged over several re-
320 peated simulations in an effort to reduce noise that occurs due to the stochastic
321 nature of the kMC model. Consequently, computation on a single processor rep-
322 resents an impractical task; however, we present parallel computation here as a
323 viable solution.

324 The motivations behind the use of parallel computation are threefold. As men-
325 tioned previously, the reduction in simulation time for a serial task is significant
326 through the use of multiple processors. Second, kMC simulations inherently ex-
327 hibit noise due to the stochastic nature of the model. By repeating a simulation
328 with the same parameters numerous times we can reduce the noise and obtain
329 more accurate, averaged values. Finally, one might want to perform many simu-
330 lations at different conditions (e.g., to find suitable model parameters by testing
331 various deposition conditions and calibrating with known experimental data).

332 As a brief outline, the process of creating a parallel program can be broken
333 down into three elementary steps: (1) the original serial task is decomposed into
334 small computational elements, (2) tasks are then distributed across multiple pro-
335 cessors, and (3) communication between processors is orchestrated at the com-
336 pletion of each batch simulation. Here decomposition of the original program
337 is achieved by separating duplicate batch simulations only, as single batch op-
338 erations cannot be parallelized. The maximum achievable speedup can then be
339 calculated as follows:

$$S(N) = \frac{1}{(1 - P) + \frac{P}{N}} , \quad (18)$$

340 where S is the maximum speedup, P is the fraction of the program which is avail-
341 able for parallelization (i.e., the fraction of the serial task which involves repeated
342 batch simulations), and N is the number of processors utilized (Culler et al., 1999).

343 There are two general modes used in the assignment of tasks: synchronous and
344 asynchronous. Given the need for averaged data to be available before the next
345 batch can be initiated, a synchronous scheme is necessary despite a slight loss in
346 performance as compared to an asynchronous workflow in which processors are
347 not required to wait for a batch completion before starting the next task. In other
348 words, if it is desired to simulate n batches of PECVD reactor with ten redundant
349 computations per batch, all ten redundant tasks must complete before the next
350 batch in line can be initiated.

351 Orchestration of the assigned tasks and communication between processors is
352 achieved using a standard message-passing interface (MPI) structure and therefore
353 will not be discussed at this time. Further details of the parallel computation
354 strategy employed here can be found in the recent publication of Kwon et al.
355 (Kwon, JSI. et al., 2015a) on which the parallel computation is based.

356 To demonstrate the reduction in computational time, a sample batch-to-batch
357 simulation is run which includes 40 successive batches of PECVD of a 300 nm
358 thick film with 10 redundant calculations per batch. The resulting computational
359 requirements are shown in Table 3. It is clear that significant time savings are
360 achieved as the number of cores utilized is increased. Increasing the number of
361 cores also generates additional overhead costs due to the communication between
362 cores and the requirement that all cores wait for the completion of the slowest

363 redundant calculation; this is evident in the difference between the true and ideal
 speedup times when $n_{cores} > 1$.

Table 3: Required time to complete a sample batch-to-batch simulation and the speedup time achieved.

n_{cores}	time (hours)	speedup (times)	ideal speedup (times)
1	73.83	1.00	1
2	38.11	1.94	2
4	19.45	3.80	4
10	8.32	8.87	10

364

365 4. Open-loop results

366 Throughout this work, simulations are conducted using a 1200 particle length
 367 ($L = 1200$) lattice with a surface that is roughened using 2400 randomly dis-
 368 tributed deposition events to ensure that the results are not impacted by the initial
 369 configuration of the lattice. The inlet gas composition, temperature and pressure
 370 are chosen to represent industrially used PECVD parameters and to correspond to
 371 conditions for which reliable experimental data exist. In the following two sec-
 372 tions, simulations at the center of the PECVD reactor are used to determine the
 373 fidelity of the hybrid kMC formulation with respect to experimentally obtained
 374 film characteristics; later results will discuss operation in other reactor zones in
 375 order to highlight the importance of thickness uniformity on the efficiency of the
 376 thin film.

377 4.1. Plasma composition, film roughness and hydrogen content

378 The temporal profiles for the five species with leading concentration are shown
 379 in Fig. 6 (Note: this plot represents operation using typical process parameters,

Figure 6: Time evolution of dominant gas-phase species at $T = 475$ K and $P = 1$ Torr.

380 $T = 475$ K and $P = 1$ Torr). Here the inlet gas stream is maintained at 9:1 ratio
 381 of hydrogen to silane with a flow rate of $75 \text{ cm}^3/\text{s}$. When compared to the time
 382 scale of a complete batch ($t_{batch} \approx 285$ sec), the dynamics of the species within
 383 the plasma are relatively short lived: within the first few seconds of operation all
 384 five species quickly approach their respective steady state values. Detailed plasma
 385 models have been developed by Amanatides et al. (Amanatides et al., 2001) and
 386 Kushner, M. (Kushner, 1988) which predict similar behavior for the dominant
 387 species considered here and yields confidence in the plasma composition obtained
 388 in this work.

389 Scanning tunneling microscopy has been used by Tanenbaum et al. (Tanenbaum et al.,
 390 1997) to accurately map the surface morphology during the growth of a 50 nm
 391 thick amorphous silicon layer deposited at 523 K and 0.54 Torr. A 1200 particle

Figure 7: Comparison of experimental and simulated rms roughness at various film thicknesses.

392 wide lattice is grown under identical conditions and the root mean square (RMS)
 393 roughness sampled at various thicknesses. The RMS roughness is calculated via
 394 the following equation:

$$R_{rms} = \sqrt{\frac{1}{L} \sum_{i=1}^L (h_i - \bar{h})^2}, \quad (19)$$

395 where h_i is the height of the lattice at position i and $\bar{h}$ is the mean height of the
 396 lattice. As noted previously, due to the triangular arrangement of Si atoms, the
 397 height at a given position is calculated to be $\sqrt{3}/2 \cdot n$ where n is the number of
 398 layers. The results in Fig. 7 represent considerable consistency between the exper-
 399 imental work of Tanenbaum et al. (Tanenbaum et al., 1997) and the prediction of
 400 the multiscale model, owing further confidence to the hybrid kMC methodology
 401 used here.

402 Calibration of the hydrogen content within the film is accomplished by varying
 403 the deposition temperature in successive batches before postprocessing the film to

determine the atomic hydrogen fraction. Comparing these values to those reported in literature (Ance and Ravindra, 1983; Bertran et al., 1991; Korevaar et al., 2000) reveals three deposition regions with distinct behavior: (1) below 500 K the hydrogen content decreases linearly with increasing deposition temperature, (2) between 500 K and 575 K atomic hydrogen fraction remains relatively unchanged and (3) above 575 K the hydrogen capacity of the film begins to increase (see Fig. 8). While the observed range for the atomic hydrogen fraction falls within the accepted experimental region, the upturn above 575 K contradicts the expected behavior. The shift in hydrogen content at high deposition temperatures is believed to be due to competition between migration and hydrogen abstraction. When operating below 550 K, an increase in temperature will boost the migration rate for surface radicals resulting in a more dense lattice; however, at high temperatures migration down the lattice cannot overcome the rapid creation of dangling bonds leading to a porous film with increased hydrogen content. In this work the operating conditions of interest call for $T = 475$ K which lies within the linear region in which the model captures well the available experimental data.

Due to the complexity of the thin film microstructure, the hydrogen content alone cannot be used to determine the validity of the multiscale model developed here. Instead, the criteria of interest is the relationship between the porosity of the film and the hydrogen content. In this work, the site occupancy ratio (SOR) is used as a measure of porosity:

$$SOR = \frac{n}{LH}, \quad (20)$$

Figure 8: Hydrogen content dependence on deposition temperature.

where n is the number of occupied lattice sites and LH is the total number of sites within the lattice. Given that hydrogen persists on the interior surfaces of the film it is expected that a strong correlation exists between the hydrogen content and SOR.

Fig. 9 shows the details of the film microstructure produced by the model at $T = 475$ K and $P = 1$ Torr. The scale of the lattice has been reduced here in order to demonstrate the porosity of the film, a typical simulation used in this work produces a lattice with dimensions $L = 1200$ and $H \sim 1400$ particles. The lattice shown has an SOR of 86% and an f_H of 13.8%. While Fig. 9 provides insight into the morphology of the amorphous thin film, further simulation is necessary in order to determine the accuracy of the multiscale model. Consequently, the deposition temperature is again varied and the hydrogen content of the resulting films calculated; however, in this case the hydrogen content is plotted against the

Figure 9: Representation of voids within a typical simulated lattice. (Note: Only a fraction of the full size lattice is shown in order to highlight porosity.)

438 SOR as experimental data is readily available for comparison. In Fig. 10 it is
 439 clear that regardless of the SOR (i.e., porosity) of a given thin film, the multiscale
 440 model developed in this work accurately reproduces the hydrogen content. In the
 441 interest of clarity, consider two simulated thin films: the first contains a distribu-
 442 tion of small (e.g., mono- and di-) vacancies as well as less frequent, long range
 443 voids. The second simulated film may contain only a single, large pore. While
 444 the SOR of both films may be calculated to be 86%, the hydrogen content would
 445 vastly differ due to the degree of interior surface area available in each case. As
 446 such, the strong agreement in the relationship between the film SOR and hydrogen
 447 content yields confidence in the ability of the multiscale model developed in this
 448 work to reproduce the deposition of α -Si:H thin films with accurate structure and
 449 composition.

Figure 10: Relationship between film SOR and hydrogen content.

4.2. Batch-to-batch variability

Successive deposition sequences lead to fouling on the interior surfaces of the PECVD reactor causing rapid drift in the electron density of the plasma during the first several batches. The resulting drop in the concentration of SiH_3 causes the growth of the α -Si:H layer to slow yielding thin films which deviate significantly from the target thickness. This effect is demonstrated in Fig. 11 where the film thickness after 284 seconds of deposition is reported for 40 successive batches. By batch number 40, the reactor is considered to be fully conditioned and the electron density of the plasma is held constant thereafter. A deposition time of 284 seconds is utilized here as this represents the average time required for an α -Si:H layer to reach 300 nm thickness when simulated using the nominal process parameters discussed previously.

Figure 11: Batch-to-batch drift in the film thickness.

462 Films deposited in later batches show an offset of up to 5% from the target
 463 thickness of 300 nm. Due to a need for nanometer scale resolution in silicon
 464 processing industries, an offset of ~ 15 nm represents an unacceptable margin
 465 and has been shown to adversely affect the photovoltaic efficiency of the thin
 466 film (Kabir et al., 2012). Although the loss in efficiency due to batch-to-batch
 467 variations is relatively minor (e.g., on the order of 1%), the effect becomes greatly
 468 exaggerated when spatial variations within the PECVD reactor are accounted for.
 469 The following sections detail our efforts to reduce offset in the film thickness using
 470 a run-to-run control algorithm and spatially distributed operating temperatures.

471 5. R2R control of film thickness

472 Recently Crose et al. (Crose et al., 2015) demonstrated that the deposition rate
473 of α -Si:H can be regulated by adjusting the temperature of the parallel plate re-
474 actor to values determined a priori by a non-linear growth rate equation. They
475 have shown that through the use of an appropriate open-loop operation strategy,
476 films can be deposited with uniform thickness in order to optimize photovoltaic
477 efficiency; however, this strategy is limited to a single batch and therefore cannot
478 account for efficiency loss due to batch-to-batch variability in the product thick-
479 ness. In this work, we build upon the concept of regulated deposition temperatures
480 in order to develop an operation strategy which can drive the thickness of the film
481 to the desired set-point of 300 nm in the presence of drift in the electron density of
482 the plasma. As a result, it is first necessary to develop a relationship between the
483 thin film growth rate and the deposition temperature within the multiscale model.

484 The growth rate of the film is plotted against deposition temperature in Fig. 12.
485 The nominal process parameters call for a deposition temperature of 475 K. In the
486 vicinity of chosen operating conditions the growth rate is shown to be propor-
487 tional to the deposition temperature; consequently, a linear growth rate equation
488 is derived using standard least squares methods:

$$G(T) = 0.0365 T - 6.56 \text{ (nm} \cdot \text{K}^{-1} \text{s}^{-1}) . \quad (21)$$

489 Due to the brief startup time for the reactor, the thickness (τ) of a deposited layer
490 can be approximated as follows:

$$\tau = G(T) \cdot t_{batch} . \quad (22)$$

Figure 12: Relationships between growth rate and temperature for the nominal operation parameters.

Together Eqns. 21 and 22 allow for the conversion between thickness measurements and temperature units required for closed-loop operation. A detailed closed-loop calculation is provided in the following section.

5.1. EWMA formulation

Given the difficulty of continuous time measurement within a PECVD reactor, a run-to-run control strategy is utilized in this work which requires only post-batch measurement (Kwon, JSI. et al., 2015b,c). Specifically, at the completion of a batch deposition process (i.e., after 284 seconds of simulation time) the thickness of the amorphous silicon layer is measured and fed into an EWMA (exponentially weighted moving average) algorithm which updates the temperature of the reactor for the next batch in an effort to overcome the aforementioned thickness non-uniformity. A schematic of the closed-loop operation strategy is shown in Fig. 13. Details of the EWMA algorithm (i.e., the post-batch simulation regime) are

Figure 13: Run-to-run operation of PECVD reactor.

provided below:

$$\begin{aligned}\varepsilon_{k+1} &= (1 - \lambda)\varepsilon_k + \lambda(T_{sp} - T_{model}) \\ T_{k+1} &= T_k + \varepsilon_{k+1},\end{aligned}\tag{23}$$

where ε_{k+1} is the parameter correction for the $k + 1$ batch, λ is the learning factor, T_{sp} is the temperature that the model predicts will be necessary to reach the set-point given t_{batch} seconds of deposition, T_{model} is the temperature that the model predicts should lead to the measured thickness, and T_{k+1} is the updated deposition temperature for the $k + 1$ batch based on the previous deposition temperature and the parameter correction value, ε_{k+1} .

5.2. Closed-loop operation

Three data sets are shown in Fig. 14; the square data points represent successive batch operation without the application of the EWMA control algorithm.

Figure 14: Drift cancellation via a R2R controller.

As stated previously, after 40 batches the product is roughly 5% below the desired set-point of 300 nm. The spherical data points represent repeated simulation using the nominal process parameters (i.e., no drift in the electron density and no control strategy applied). This provides some insight into the level of process noise and represents the threshold for perfect operation of the controller. Finally, the triangular data points represent the simulated reactor under the influence of the proposed EWMA control algorithm. Similar to the case with no control, we see that the thickness of the product initially moves away from the set-point (batches 1-3). However, in the following batches the parameter error correction, ε , begins to counteract the effect of drift in the electron density. By batch 10 the controller has driven the product to the desired set-point and then holds the product thickness within the process noise level for all remaining batches.

As demonstrated in Fig. 15, the value of the learning factor affects the number of batches required for the thin film product to approach close to the desired set-

Figure 15: Comparison of different learning factor values.

point of 300 nm, however, in both cases ($\lambda = 0.75$ and $\lambda = 0.25$), the thickness value approaches the set-point after about 10 batches.

It is important to note here that each batch simulation treats the PECVD reactor as a single zone. In other words, the reported thickness measurements are for the entire thin film and the temperature parameters are applied equally throughout the reactor. In the following section we report an extension of the EWMA controller to multiple reactor zones in an effort to address the problem of spatial non-uniformity within a batch deposition process.

6. Spatial non-uniformity in PECVD systems

In the previous section, the discussion was focused on drift in the electron density in a batch-to-batch manufacturing process; in other words, the non-uniformity in plasma composition occurred between successive deposition sequences. Here, we address the additional problem of in-batch spatial variations (e.g., non-uniform

Figure 16: Radial non-uniformity in $[\text{SiH}_3]$ at $T = 475$ K and $P = 1$ Torr.

steady-state concentration profiles in SiH_3).

As discussed in the gas-phase model, a steady-state concentration gradient for SiH_3 above the wafer surface has been calculated (Armaou and Christofides, 1999) using finite differences methods. For a 20 cm diameter wafer, a radial non-uniformity of up to 17% in the $[\text{SiH}_3]$ is observed from the center to the edge of the silicon wafer which is expected to cause an efficiency loss of roughly 5.4% (Kabir et al., 2012); Fig. 16 demonstrates this effect for the nominal process parameters (i.e., $T = 475$ K and $P = 1$ Torr) at four distinct locations across the wafer surface.

This 5.4% level of efficiency drop corresponds to roughly the same level of thin film solar cell power output loss, as can be concluded from the dependence of voltage and current on thin film thickness reported in (Kabir et al., 2012). Carrying out a rough engineering estimate, for a typical utility-scale thin film solar cell system of 100 MW, a total installation cost of \$1.49/W–\$1.84/W is projected (Fu et al., 2015); this cost includes not only the photovoltaic modules themselves,

556 but also infrastructure, labor, environmental permits and other costs. Given an ef-
557 ficiency loss due to non-uniform deposition of thin film layers, as described above,
558 more modules will be required in order to meet the client specification of 100 MW
559 of power production. Assuming that the most economical features are chosen (i.e.,
560 using the lower bound for installation cost of \$1.49/W), and ignoring fixed costs,
561 the additional photovoltaic modules (resulting from non-uniform thin film thick-
562 ness) are expected to contribute on the order of \$8 million to the total plant cost
563 of roughly \$150 million (Fu et al., 2015). As such, significant economic benefits
564 are available through the use of controlled operation of the PECVD reactor.

565 Similar to the case of batch-to-batch drift, the concentration of the deposition
566 species of interest ($[\text{SiH}_3]$) is tied directly to the electron density. The resulting
567 effect on the uniformity of the film thickness can be seen in Fig. 17 by simulating
568 the deposition of 40 successive batches in which both batch-to-batch variation in
569 the electron density is allowed to occur, as well as spatial non-uniformity in the
570 steady-state concentration profiles.

571 The four curves in Fig. 17 represent batch-to-batch operation at four zones
572 within the reactor without the application of an EWMA control algorithm (Note:
573 each zone is simulated using a single lattice of length $L = 1200$). The effect of
574 spatial non-uniformity in the concentration of SiH_3 is evident in both the initial
575 thickness (i.e., thickness of batch 0) of each zone, as well as the total offset after
576 40 batches. While the product in zone 1 maintains the same 5% offset discussed
577 earlier, zones 2-4 exhibit increased offset. After 40 batches zone 4 has drifted
578 roughly 15% from the desired set-point of 300 nm despite the same drift in elec-

Figure 17: Open-loop drift and spatial non-uniformity in the four radial wafer zones.

tron density being applied.

6.1. EWMA extension to concentric reactor zones

Croese et al. (Croese et al., 2015) have shown that four concentric zones can be defined within the PECVD reactor as shown in Fig. 18 each with an associated heating element. Thus, the temperature in every zone can be adjusted independently in order to modulate the growth rate of the thin film. However, accurate prediction of the deposition temperature in each zone requires a corresponding growth rate relation.

Consequently, simulated films are deposited at various temperatures and the corresponding growth rates are plotted in Fig. 19. Each data set represents a different reactor zone in which the concentration of SiH_3 has been adjusted to account for the aforementioned spatial non-uniformity. The uppermost curve corresponds to the first zone and is therefore identical to the growth rate data presented in the

Figure 18: Discrete PECVD reactor schematic showing four independent, concentric substrate temperature control zones.

Figure 19: Relationships between growth rate and temperature for each reactor zone.

previous section. The remaining curves demonstrate the reduction in film growth rate that occurs due to the consumption of deposition species as process gas travels radially outward toward the edge of the wafer. Linear relations are developed for each zone using standard least squares methods:

$$\begin{aligned} G(T)^1 &= 0.0365 T - 6.56 \text{ (nm} \cdot K^{-1} s^{-1}), \\ G(T)^2 &= 0.0366 T - 6.74 \text{ (nm} \cdot K^{-1} s^{-1}), \\ G(T)^3 &= 0.0337 T - 5.54 \text{ (nm} \cdot K^{-1} s^{-1}), \\ G(T)^4 &= 0.0331 T - 5.54 \text{ (nm} \cdot K^{-1} s^{-1}). \end{aligned} \tag{24}$$

$G(T)^1$ through $G(T)^4$ correspond to zones 1-4, respectively.

At this point, we can define a set of EWMA algorithms using the developed growth rate relations:

$$\begin{aligned} \varepsilon_{k+1}^i &= (1 - \lambda) \varepsilon_k^i + \lambda (T_{sp}^i - T_{model}^i) \\ T_{k+1}^i &= T_k^i + \varepsilon_{k+1}^i. \end{aligned} \tag{25}$$

Recall, through the assumption of time invariant growth rates we can write the thickness as $\tau^i = G(T)^i \cdot t_{batch}$ to allow for conversion between post-batch measurements and temperature values necessary for the controlled input.

In the interest of clarity, Fig. 20(a) is provided which details the paths of the controlled inputs, T^i . In each zone, the deposition temperature increases from batch to batch to overcome the reduction in electron density that occurs as the reactor becomes conditioned. As expected, a significantly higher temperature must be applied in the outermost zones in order to accommodate for the reduced growth rate due to consumption of SiH_3 . The four matching curves in Fig. 20(b) show the performance of the extended EWMA control algorithm. By batch number 10, the thin film thickness in each reactor zone is driven to the desired set-point and then maintained within the process noise level for all successive batches.

(a) Control action computed by EWMA algorithm.

(b) Closed-loop operation via EWMA control algorithm.

Figure 20: R2R control of four independent wafer zones.

611 7. Conclusions

612 In this work, a multiscale modeling and run-to-run framework for the PECVD
613 of thin film solar cells with uniform thickness and reduced batch-to-batch vari-
614 ability was proposed. The macroscopic gas-phase utilizes mass, momentum and
615 energy balances, under the assumption of continuum to provide the plasma com-
616 position necessary in the calculation of deposition rate profiles. Additionally, the
617 proposed microscopic model is based on a novel hybrid kinetic Monte Carlo al-
618 gorithm that has been shown to be both computationally efficient and to repro-
619 duce thin film morphologies on par with experimental observations. Together
620 the macroscopic and microscopic simulation have yielded insights and data that
621 have been used to develop a closed-loop operation strategy which has been shown
622 to improve film quality and to reduce batch-to-batch variability caused by drift
623 during the conditioning phase of reactor operation. Specifically, an EWMA (ex-
624 ponentially weighted moving average) algorithm has been introduced which cal-
625 culates substrate temperatures that mitigate the effect of batch-to-batch reactor
626 variability. Simulations of open-loop operation suggest an offset of 5% from the
627 desired set-point of 300 nm film thickness due to reactor variability. The proposed
628 EWMA control strategy was applied first to a PECVD reactor with a single heat-
629 ing control element which allowed for the temperature to be adjusted post-batch
630 to counteract drift in the electron density, thereby reducing the product offset to
631 $<1\%$. Lastly, an extension of the proposed EWMA control algorithm was demon-
632 strated which allows for in batch variation in the deposition temperature within
633 concentric reactor zones in an effort to reduce spatial non-uniformity in the thin

634 film thickness. Simulations demonstrate both a reduction in the product offset to
635 less than 1% as well as radially uniform films within 10 batches of closed-loop
636 operation.

637 **8. Acknowledgements**

638 Financial support from the National Science Foundation (NSF), CBET-1262812,
639 is gratefully acknowledged.

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