



Modeling reactive magnetron sputtering: a survey of different modeling approaches

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Abstract. The paper focuses on providing an insight into the current state of computational modeling regarding reactive magnetron sputtering systems. A detailed compilation of developed models is gathered and grouped into categories based on the phenomena being modeled. The survey covers models developed for the analysis of magnetron discharges, particle -surface interactions at the target and the substrate, as well as macroscopic models. Corresponding software packages available online are also presented. After gaining the necessary insight into the current state of research, a list of the most challenging tasks is given, comparing different approaches, that have been used to combat the encountered difficulties. The challenges associated with modeling tasks range from analytical complexity, mathematical know-how used for model approximation and reduction, as well as optimization between computational load and result accuracy. As a conclusion, the future challenges are compiled into a list and a probable direction in modeling is given, that is likely to be further pursued.

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1 Introduction

The goal of the survey is to offer a brief insight into the current state and results of different mathematical and computer modeling techniques and approaches used to investigate reactive magnetron sputtering.

Sputtering is a subclass of PVD - Physical Vapor Deposition, which is an essential process in manufacturing integrated circuits, photovoltaic modules, specialized optical and mechanical equipment. It is a process used for the creation of extremely thin (even down to atomic) coating layers on different surfaces in order to achieve better physical, chemical, optical or electrical characteristics, than those of the coated material itself. Among others, the use of this technique allows the formation of transparent but also conducting layers, which are essential in optics. The process essentially consists of the removal of material from one or more target surfaces and their deposition onto a surface that is required to have improved characteristics, also known as the substrate. This removal process is accomplished with the help of high energy noble gas ions which bombard the surface of the target due to the presence of an electric field. With the addition of a magnet behind the target one can achieve magnetron sputtering, greatly increasing the ionization efficiency and the sputtering yield.

With the introduction of one or more reactive gases into the vacuum chamber, reactive magnetron sputtering is achieved, which allows the formation of different compounds like oxides and nitrides. Reactive magnetron sputtering has been extensively studied and modeled in the past few decades.

There are several approaches and technologies used for magnetron sputtering, such as DC (direct current) sputtering, pulsed DC sputtering, AC (alternating current) sputtering, HiPIMS (high power impulse magnetron sputtering) and RF (radio frequency) sputtering. All of the different coating techniques mentioned require highly specialized operating environments in order to function correctly. The creation of a high vacuum environment (pressure in the range of a few Pascals) is mandatory. At such low pressures, different metals begin to evaporate, hindering the insertion of any nonspecialized equipment into the sputtering chamber. Simple questions as measuring the total or partial pressure inside the chamber become ever more difficult, requiring special processes to be put in place and the indirect calculation of the desired quantity from the gathered data. In situations like these, the use of a state identification algorithm becomes favored. From the control perspective, the system quickly becomes complex when one simply attempts to count the number of inputs, outputs and their non-linear cross coupled effect on each other. Each

of the magnetron sputtering processes set particular challenges from the point of view of modeling. This paper concentrates on gathering and presenting only computer modeling approaches used for DC reactive magnetron sputtering.

Due to the specifics of the environment, the overwhelming amount of process values to which attention has to be paid, the lack of quick and reliable access to measurable data, control of the process becomes complex enough that many laboratory experiments are conducted in an open-loop fashion. Therefore, reliable reproduction of experiments is still nearly impossible.

These constraints have compelled researchers to develop, test and validate complex models of the process. Most of the time, financial reasons are the main driving force behind the need for modeling, especially in industrial applications. The simplest desire that can be placed is the optimization of the process to achieve maximum yield. Another desire, which is far more complex is the achievement of required stoichiometric ratios between the deposited materials on the surface.

The paper is structured as follows: chapter 1 gives a brief introduction to the field of study, chapter 2 gives an overview of several prominent research results in the field of modeling, chapter 3 presents the different modeling tasks and challenges, and finally, chapter 4 outlines directions that are likely to be further pursued.

2 Literature survey of modeling approaches

Reactive magnetron sputtering is a complex process with several interacting subprocesses, representing difficult modeling tasks on their own. One of the possible approaches is the development of multi-physics models by coupling the models which describe the plasma, the magnetic field, the transportation phenomena in the chamber, the interaction of particles with the target, the thin film deposition on the substrate, etc. Such a model based on the perfect understanding of the physical phenomena would represent a perfect tool for the industry. However, the feasibility of a "virtual sputter tool" [58] resulting from this bottom-up approach is low, mainly because the level of understanding of different phenomena is not the same, and there will always be a weak element in the chain which might vitiate the overall results. A top-bottom model development, leading to a "holistic model" [58], seems to be more feasible. According to this approach, the development starts from a simplest possible model which describes "well enough" the phenomena of interest, and further on this model suffers extensions according to the necessities to cover other phe-

nomena as well. Contrary to these "white box" approaches, the input-output modeling of the system, which has little to do with the understanding of the details of its physics, can be very useful for control purposes [62].

According to the bottom-up approach, one may study the phenomena that are taking place inside the plasma discharge between charged particles. Such a model must also consider the magnetic field present in the chamber and its effects on the movement of charged particles.

A very intriguing phenomenon that can be modeled is the interaction between particles at the surface level of the target, where collisions either lead to implantation or material removal.

Particle-surface interactions are also studied at the substrate, where the formation of crystallographic structures is analyzed.

The top-bottom approach is represented in this paper by the extensions of the Berg model, the simplest one, which can efficiently describe the hysteresis phenomena in case of a single target, single reactive gas magnetron sputtering. These are macroscopic models based on equilibrium equations, and are not more complicated than necessary to reproduce the essential behavior of the system, trying to remain simple enough to facilitate control.

2.1 Modeling of magnetron discharges

An electric discharge taking place in vacuum can be divided into the following categories: dark, glow and arc discharge, each with a higher discharge current than the former.

According to Bogaerts et al. [9] there are three types of model categories for plasma discharges:

1. Fluid models, which are modeling species in the discharge as ones being in hydrodynamic equilibrium. The equations that govern are the continuity equations (fluxes and densities of the species) and the Poisson equation for the electric field. The fluid model is considered to be the simplest but also the most inaccurate one.

2. Kinetic models, which describe the plasma species as beams of particles. The movement of particles is described by the Boltzmann transport equations, combined with the Poisson equation.
3. Monte Carlo simulations, that describe the different plasma particles separately on a statistical basis. This is considered to be the most accurate method, but it requires considerable computational effort.

Depla and Mahieu compiled a comprehensive study on modeling the electric discharge in reactive magnetron sputtering [26]. They have grouped models into two main categories i.e. analytical and numerical. They further split numerical into fluid and kinetic ones. In this approach, kinetic types are either based on numerical solutions of the Boltzmann equation or on Monte Carlo simulations, which are also called particle simulations.

Wendt and Lieberman [60, 61] developed a two dimensional analytical model of the discharge process, taking into account both magnetic and electric fields.

Fluid models [14, 22, 23] have also been presented. These models calculate the electric field distribution by coupling the Poisson equation to the continuity and conservation of energy and momentum equations.

The solution of the Boltzmann equation has been widely used for modeling plasma discharges. The addition of the Lorentz force term complicates the solver, as presented by Porokhova et al. in [49, 50, 51].

In Monte Carlo simulations the path of a small number of particles is analyzed, their movement and their collisions being modeled using probabilities and random numbers [53, 29].

Hybrid models have been conceived with the assumption that heavy ions are practically not affected by the magnetic field, as opposed to the relatively lightweight electrons. Therefore, ions can be modeled by a fluid model and electrons can be modeled as particles. This assumption combines high precision of particle simulation codes with lower computational efforts of fluid codes [52, 35]. Bogaerts et al. have presented several papers modeling the magnetron discharge process using Monte-Carlo and hybrid Monte-Carlo simulations [11, 9, 10, 12].

Particle in Cell-Monte Carlo Collision (PIC-MCC) simulations are considered to be the most powerful available numerical tools. PIC-MCC simulations combine MC methods with the simulation of the electric field being produced by an external power supply and with the spatial distribution of the charged particles in the plasma [8].

2.2 Modeling particle-target interaction

When studying particle-target interactions at the surface of the target, one may find in the literature that depending on the kinetic energy and the incident angle of impact, some ions reach the surface and through chemisorption get chemically attached to the outermost layer. Others may effectively transfer their kinetic energy and lead to material removal from the target, and some may be so energetic that they get implanted into the subsurface and stay there until enough material is removed for them to resurface. Depla, Strijckmans and separately Kubart extended existing models by also considering chemisorption, knock-on yield, ion implantation, redeposition and reaction rate, leading to far more accurate but also far more complex models [25, 27, 56, 57, 36, 6].

According to [48] Monte Carlo type simulation codes, which have been proven to be the most effective, have the following major components:

1. the cross-section data for all processes being considered,
2. the algorithms used for the particle transport,
3. the methods used for geometry representation and to handle particle passage through volumes,
4. the methods to determine quantities of interest and analysis of the information obtained during the simulation.

Such an example is TRIM, which is a Monte Carlo computer program that calculates the interactions of energetic ions with amorphous targets (see Fig. 1). SRIM is a collection of software packages that calculate many features of the transport of ions in matter (see Fig. 2). Both programs were developed and are maintained by James F. Ziegler and can be downloaded from <http://www.srim.org/>.

Other MC based simulation packages are TRIDYN [44, 45], KALYPSO [32] and ACAT [68]. A comprehensive compilation and review of many other simulation codes for surface-ion interactions can be found in Chapter 1.2 of [26].

2.3 Modeling crystal formation on the substrate surface

Modeling the growth of the deposited layer on the substrate is one of the most complex aspects of modeling the sputtering process. The set of particles arriving at the substrate include not only outspattered particles from the target, but also electrons, gas ions and neutrals [26]. The incoming particles either settle at the surface by forming chemical bonds depending on the sticking coefficient of the atomic pair in discussion, or they migrate on the surface until

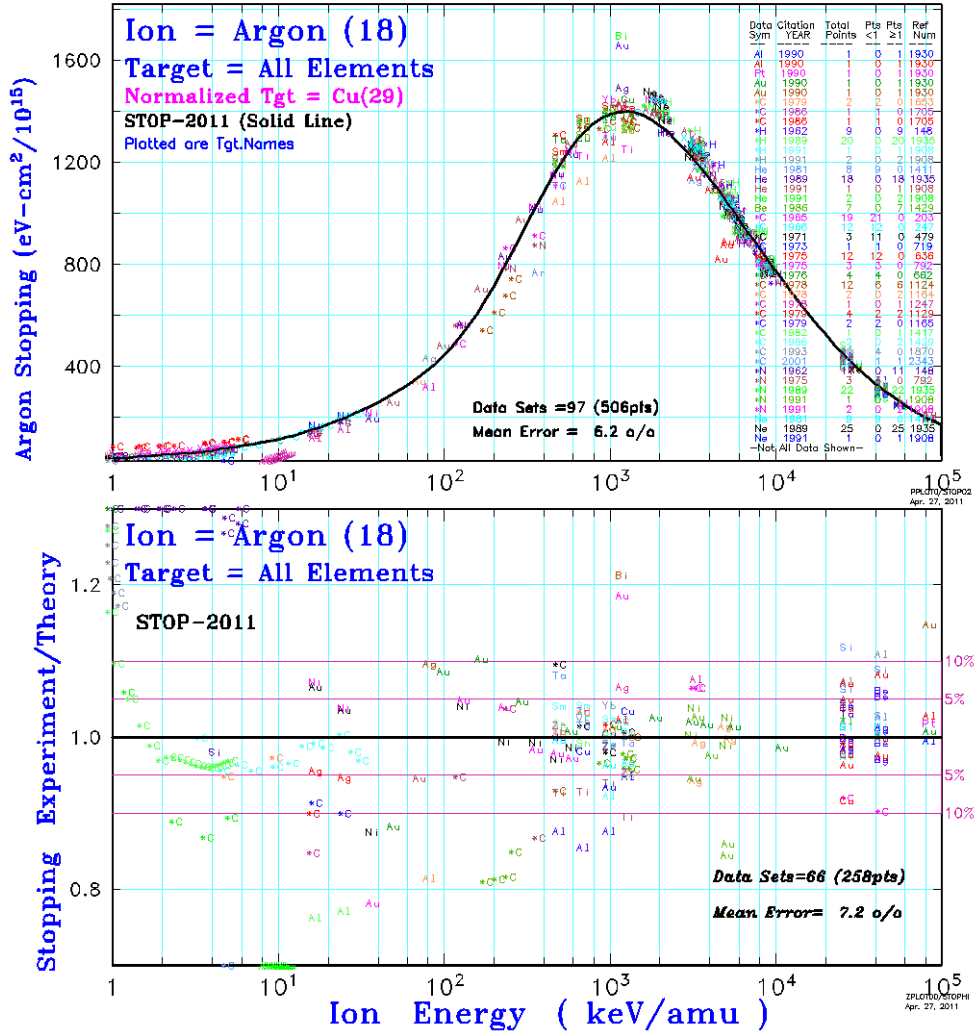


Figure 1: Application example of the TRIM program. Stopping range as a function of accelerated Argon ion energies using different targets; comparison of historically measured and simulated data (from <http://www.srim.org/>).

they reach a large enough crystal lump where they can take part in crystal formation on the grain boundary [30]. Because of this, the formation of a large area, even layer is not too common, rather ragged microstructures are formed [47]. The modeling of such microstructures is essential for example in the fab-

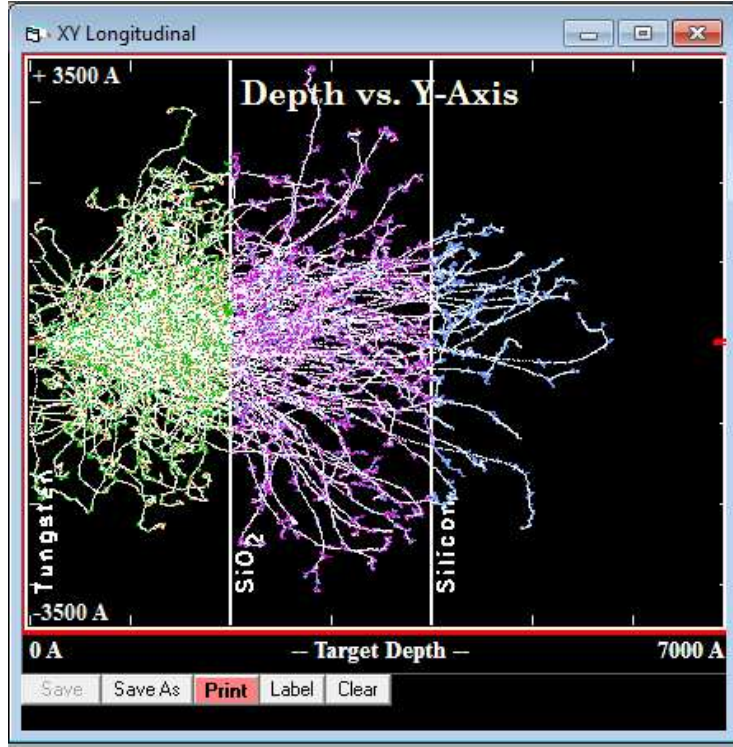


Figure 2: Application example of the SRIM program. Simulation of implantation depth of Boron ions into a multilayered W/SiO₂/Silicon target using SRIM-2013.00 software (from <http://www.srim.org/>).

rication of microelectronics, where one may want to fill a narrow trench or via. In [7] the formation of some crystal structures is predicted based on operating conditions.

An example software is SiMTra, which is a binary collision Monte Carlo program (see Fig. 3) that allows the user to simulate the transport of sputtered particles through the gas phase flux during sputtering.

2.4 Macroscopic modeling of the reactive sputtering process

Similarly to those described in the previous sections, it is obvious that one must find a balance between complex accurate modeling and simple straightforward usability when developing a model. Berg et al. developed a model that found a balance between simplifications whilst still being accurate enough to

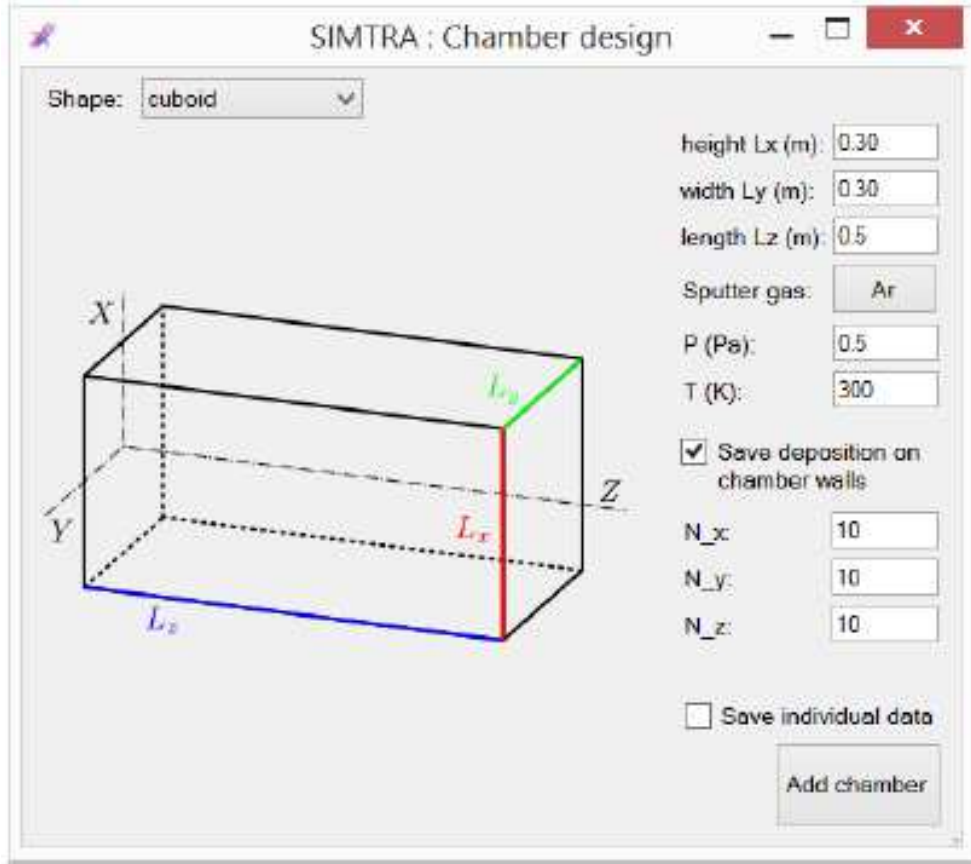


Figure 3: An input dialog screen of the SIMTRA software package used to configure the simulation setup (from <http://www.draft.ugent.be/>).

predict outcomes of experiments [3, 5, 4]. This model is commonly referred to in the literature as "Berg's Model". The model was constructed by focusing on the equilibrium equations, studying independent contributions to different quantities used to describe the process (see Figure 4). The main area where this model proved to be more successful than others is in the modeling of the hysteresis behavior. There have been a number of models [31, 1, 54] developed before Berg's Model that had problems reproducing the hysteresis behavior.

Berg employed variables θ_i to denote the coverage of the surfaces of interest (target and substrate) with compound molecules (the normalized amount of reacted surface area). The coverage takes values in the interval of $[0, 1]$, where

a value of 1 denotes a fully covered surface. This model is a monolayer surface model which only considers a chemisorption mechanism on the target (and substrate) surfaces. No redeposition can be considered.

The equation that defines the balance between supply and consumption of the reactive gas is given in Eq. (1), with the definitions of gas consumption on the surface of the target Q_t in Eq. (2), gas consumption on the surface of the substrate Q_c in Eq. (3) and of pumping flow Q_p in Eq. (4). The supply and usage of reactive gas is also presented in Fig. 4. The sticking coefficients are denoted with α , the flux of reactive gas atoms with F and the corresponding areas of the studied surfaces with A . S represents the pumping speed while P represents the partial pressure.

$$Q_{\text{tot}} = Q_t + Q_c + Q_p \quad (1)$$

$$Q_t = \alpha F(1 - \theta_t)A_t \quad (2)$$

$$Q_c = \alpha F(1 - \theta_c)A_c \quad (3)$$

$$Q_p = SP \quad (4)$$

The balance equations describing the interactions on the target and substrate surface are given in Eq. (5) and (6), respectively. The outspattered compound F_c and metallic F_m flux is given in Eq. (7) and (8), respectively. Y denotes the sputtering yield, q is the electron charge and J denotes the ion current density of the background gas. Equations (1-8) have been presented in this form in [26].

$$\frac{J}{q}Y_c\theta_t = \alpha 2F(1 - \theta_t) \quad (5)$$

$$F_c(1 - \theta_c) + 2Q_c = F_m\theta_c \quad (6)$$

$$F_c = \frac{J}{q}Y_c\theta_tA_t \quad (7)$$

$$F_m = \frac{J}{q}Y_m(1 - \theta_t)A_t \quad (8)$$

Due to the reactive gas poisoning of the target, a process that tends to reduce the sputtering yield, a hysteresis can be observed due to the positive

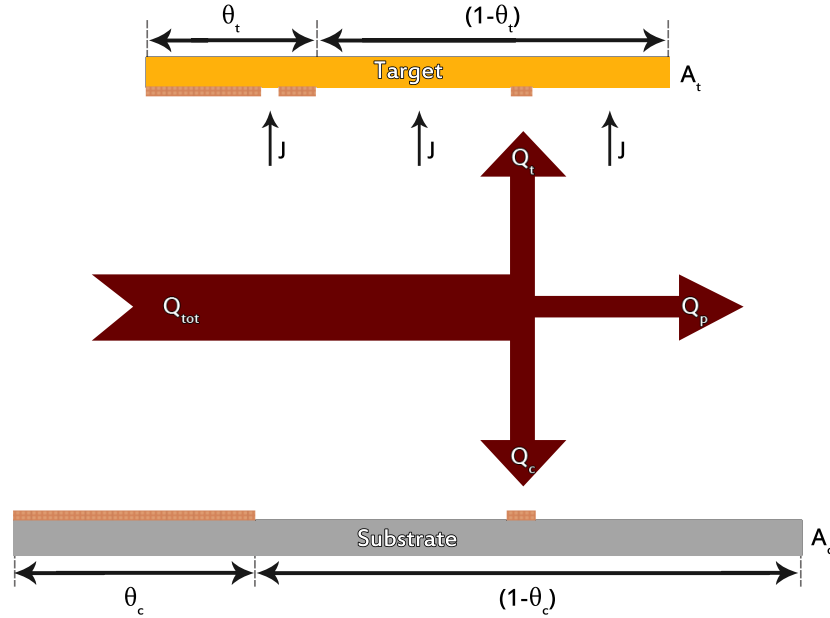


Figure 4: The supply and consumption of reactive gas during sputtering [5].

feedback between the partial pressure of the reactive gas and the target surface coverage (see Fig. 5). Since the publication of the Berg model, several extensions based on equilibrium equations have been developed in order to include modeling of multiple targets, multiple reactive gases, ion implantation, particle redeposition, multiple deposited layers, deposition of multiple compounds, spatial resolution and dynamics of phenomena, etc.

Given that many times the formation of complex films is required, which in turn require more than one reactive gas, Carlsson et al. further developed the model to work with two reactive gases as inputs, though only considering binary compounds as result of the interaction processes [15].

The research group named [DRAFT](http://www.draft.ugent.be/), led by Diederik Depla, made their work of more than a decade available free of charge as a downloadable package from their university website <http://www.draft.ugent.be/>. The package contains their model named RSD2013 and it also comes with a detailed PDF guide to help the researcher quickly get started. The package also includes a piece of software called SIMTRA, which aids the simulation of deposition profiles. This model is an extension of the Berg model, having the most im-

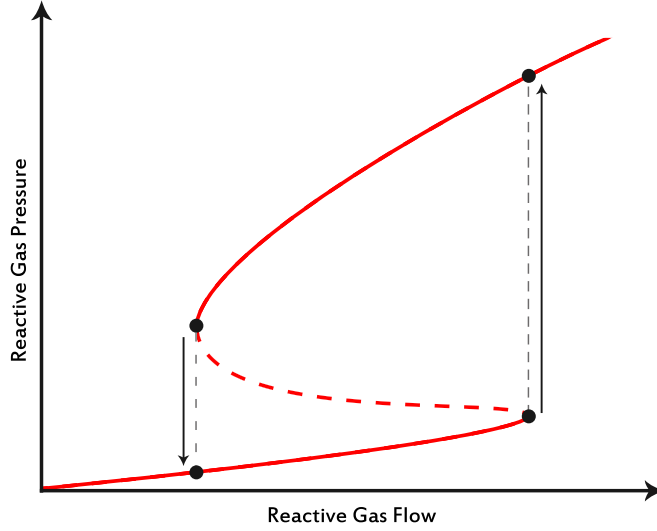


Figure 5: The observed hysteresis behavior of the system on a p-q plot [5].

portant extra feature of subsurface implantation of reactive gas ions/atoms and the compound forming 2nd order reaction mechanism in the subsurface. A chemisorption mechanism governs the reaction on the surface and redeposition of sputtered material back on the target is taken into account [58].

3 Modeling tasks and challenges

Throughout the following subsections a comparative study analysis is given under each modeling task.

3.1 Parameter identification approaches

One of the biggest, if not the biggest challenge in modeling reactive magnetron sputtering is the lack of readily available parameters or tools to obtain those parameters.

The lack of identified sticking coefficients led Bogaerts et al. [13] to attempt to identify them by trying to select the ones that would have had resulted in correct estimation of the process, after the experiment was undertaken. The chosen experimental setup was one, in which the sticking coefficient played

a significant role. The ion density at a certain distance from the substrate was measured and simulated. They performed the simulation for every possible value of the sticking coefficient and later checked which ones were in accordance with the measured data. What they found is that two given circumstances resulted in different parameters, meaning that the model wasn't detailed enough and some other effect also had to be considered. Their result, however, narrowed the plausible range for the sought parameter.

Leroy et al. attempted the parameter identification by analyzing samples of deposited films and estimating the actual content of a given material in them [37, 38]. Having this estimate, it was possible to calculate what the missing parameter should have had been. An extensive guide has been given on how this should be undertaken. The process requires several specialized equipment and techniques (XPS - X-ray Photoelectron Spectroscopy, EPMA - Electron-probe microanalysis) and complex calculations to be able to reach a result.

Strijckmans et al. also undertook this challenge of parameter identification in [56]. They used their most detailed RSD2013 model, missing only 3 parameters (see Fig. 6), and wrote a parallel C++ program to run the model on a supercomputer, to see which parameter sets would be usable.

The parameter fitting algorithm has two main components: a parameter set selection component and a parameter set evaluation component. A 2-D representation of the parameter set selection component can be seen in Fig. 7. Increasing the dimensionality of the parameter space is feasible, though with increased computational load. Three ordered lists have been employed to differentiate between unfinished (U), finished (F) and rejected (R) parameter sets. A parameter set is considered rejected (R) when the result of the model simulation does not agree with the measured data.

The evaluation component has an acceptance criterion which is determined by the PQ - pressure-flow relation of the simulation result. The critical flow values (the limits of the unstable region) are observed at three different current values and the maximum of the six squared errors is considered.

Before this study there was complete disagreement among researchers, many of them having measured completely different values for the same parameter. The results of this attempt further narrowed down the possible range of parameter sets.

3.2 Modeling the produced compounds

Berg et al. laid down the foundation for macroscopic modeling of magnetron sputtering with one reactive gas [3, 4, 5]. When the need arose to model binary or ternary compounds this model has been expanded.

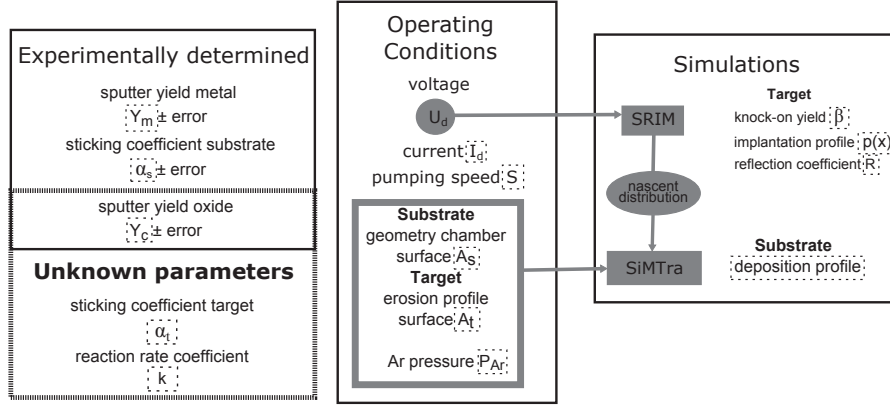


Figure 6: A scheme visualizing the input parameters of the RSD2013 model [58]

Carlsson et al. published an article where two reactive gases were used and binary compounds were modeled [15]. This model neglected ternary compounds for the sake of simplicity. When the growth of a ternary compound such as of TiO_xNy is the requirement, one must find a different model.

Kelemen et al. published a macroscopic model that studies the formation of ternary compounds on the substrate [33]. The model follows the same approach as the two mentioned above, allocating 8 regions on the substrate covered with different active, passive, binary and ternary compounds.

3.3 Modeling different magnetron types

Dual magnetron co-sputtering allows the deposition of alloy material without having to manufacture alloy targets [55, 43, 34]. Multicomponent models have also been developed and analyzed in [17, 18, 46]. This setup requires two separate magnetrons to be installed, each of them becoming the anode of the other during sputtering. Such a setup requires a power supply that can generate pulsed bipolar signals. Given the fact, that each magnetron is switched between anode and cathode roles, the typical charge build-up on poisoned areas is neutralized due to sequential target bombing with ions as well as electrons. The absence of local charge build-up also leads to an arc free operation, which is mandatory for consistent large area, even layer deposition.

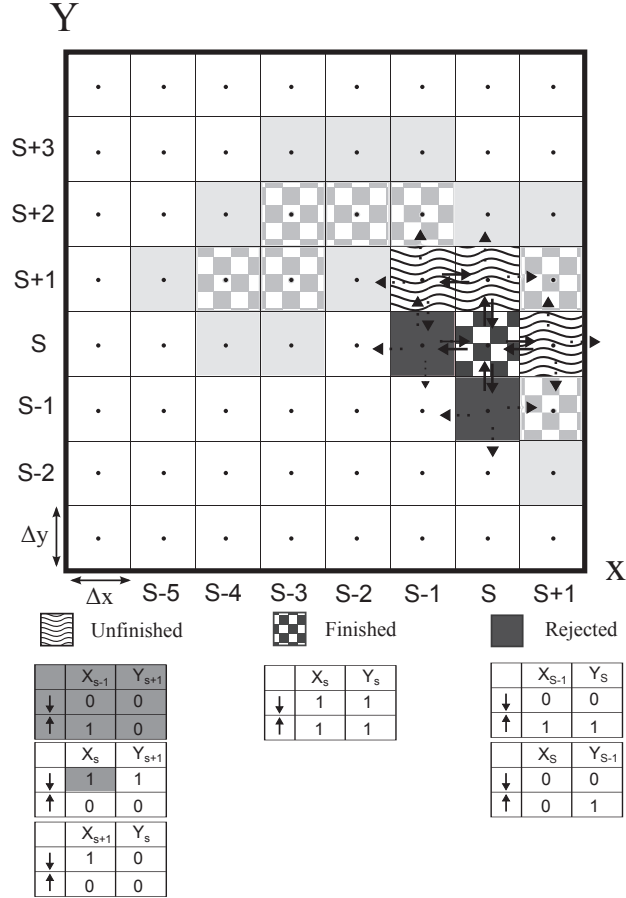


Figure 7: A 2-D illustration of the parameter selecting component of the searching algorithm [58].

Cylindrical rotating magnetron sputtering was introduced to reduce the operational costs of large scale industrial coaters, due to the inherent disadvantages of sputtering of stationary planar magnetrons. One of these disadvantages is the well-known formation of a so-called "race track", a high erosion zone on the surface of the target, that is caused by the inhomogeneous magnetic field that "shapes" the plasma above the target. By rotating the target with respect to the magnetic field no high erosion zone is formed. This leads

to a very efficient process due to the uniform wear the target experiences. The uniform wear maximizes target usability by avoiding the formation of a localized deep race track. The effects of rotation have been modeled and the influences of rotation speed have been studied in [27, 24, 42, 39]. It was observed that the rotation speed highly influences the shape and location of the hysteresis curve.

3.4 Modeling for control structure synthesis

The study of the system dynamics and the control structure synthesis require the development of time dependent, dynamic models. Based on macroscopic models, several authors have developed time dependent models, usually in state-space formulation [33, 57, 19, 20, 21, 2]. When derived from the original Berg model, the dynamic model represents a control-affine system of third order with linear and bilinear terms [62]. Woelfel et al. also presented the Berg model in a state-space form along with a detailed explanation of its construction (Eq. (9) and (10)). Coefficients $a_{i,j}$ and $b_{i,j}$ include parameters like sticking coefficients, sputtering efficiencies, sputtering current density, gas constant, temperature, etc. The state variables are the partial pressure of the reactive gas and the coverages of the target and of the substrate, while the input of the system is the input flow rate of the reactive gas.

$$\begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix} = \begin{pmatrix} P \\ \theta_t \\ \theta_s \end{pmatrix} \quad (9)$$

$$\begin{aligned} \dot{x}_1(t) &= x_1(t)(-a_{1,1} + a_{1,2}x_2(t) + a_{1,3}x_3(t)) + bu(t) \\ \dot{x}_2(t) &= a_{2,12}x_1(t)(1 - x_2(t)) - a_{2,2}x_2(t) \\ \dot{x}_3(t) &= a_{3,13}x_1(t)(1 - x_3(t)) + a_{3,23}x_2(t)(1 - x_3(t)) - a_{3,32}x_3(t)(1 - x_2(t)) \end{aligned} \quad (10)$$

The magnetron sputtering system is essentially nonlinear, but several studies attempt to use linearization around the equilibrium points to study the stability and controllability of the system. Due to the difficulties to apply the results of control theory, there have been studies and even patents on how to control a system of such dynamics through heuristic synthesis of complex control structures. In [59] a control structure was presented in which the chosen manipulated quantities influence the controlled variable indirectly, through a chain of effects in the system.

Woelfel had a different approach by studying the essential input-output interactions in different operating conditions in order to develop a simplified model that was conceived with the aim of being practical for control purposes [62, 63, 64, 67]. Woelfel et al. presented a control design method that is based on artificial neural networks and ordinary differential equations [66, 65]. The goal of using neural networks was to decouple the plasma, electrical and gas subsystems from each other. Two multilayer perceptron type networks have been used, both employing a hidden layer of 10 neurons with sigmoid activation functions.

Our previous work also concentrated on control structure synthesis [40]. It started with the assumption that an on-line state identification algorithm can be developed, and with its use, the stoichiometry of the process can be controlled. The input-output characteristics have been plotted to observe the location of the peak of the substrate coverage with the desired compound (see Fig. 8). By selecting a preferred subsurface by formulating first order equations to delimit a monotone region (see Fig. 9) it was possible to conceive a state-dependent state limiting algorithm that can operate the system with two independent continuously pumped reactive gas inputs. This came as a proposed alternative for RGPP - Reactive Gas Pulsed Process [16, 41] which for the sake of stability had to compromise on stoichiometry.

One would be mistaken by thinking that better control of the process can only be achieved by ever more complicated control structures. Some researchers focused on model reduction, in order to facilitate the implementation of less complex control algorithms. The work done by Woelfel et al. concentrates on developing models that suffice the control goal [67]. It was mentioned that most results in the field were provided from either a vacuum science or a thin solid film approach, therefore most models developed were not usable for control purposes. It was emphasized that not enough work has been done to identify the input-output behavior of the reactive sputtering process.

They recognized [62] that for control purposes the Berg model can be reduced to the Abel differential equation (see Eq. (11)). In Eq. (11) the system input $u(t)$ is the gas flow and the system output $y(t)$ is the sputtering voltage.

$$\dot{y}(t) = Ay^3(t) + By^2(t) + Cy(t) + D + Eu(t) \quad (11)$$

Using this model it was later demonstrated that a venerable PID (proportional–integral–derivative) type controller can be used to control and stabilize the complex system. Detailed description has been given for the necessary preconditions that are needed to apply the given tuning rules [64].

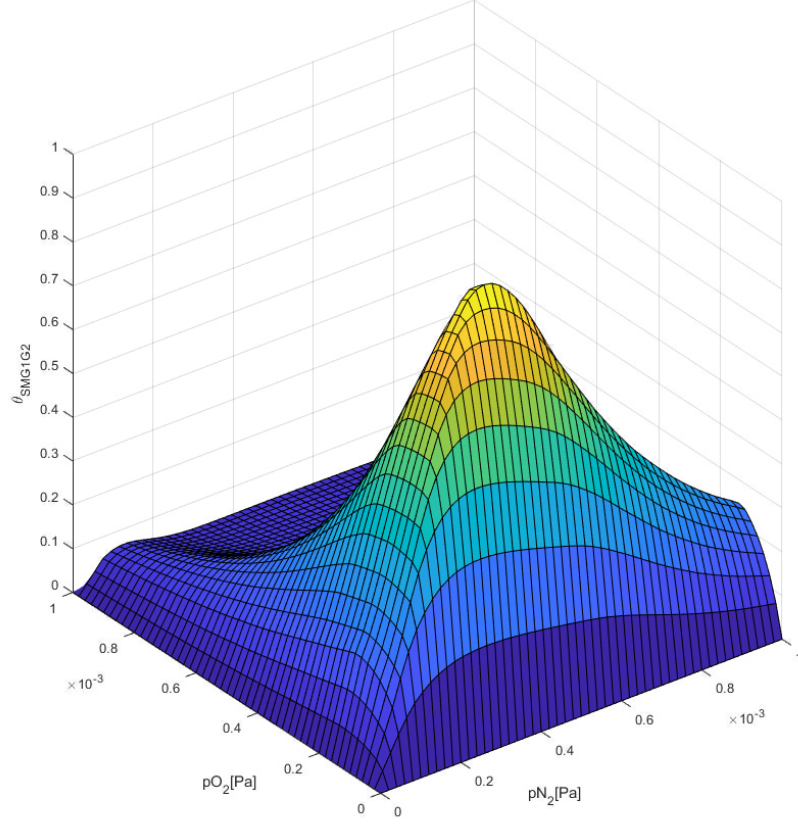


Figure 8: Substrate coverage with $\theta_{MG_1G_2}$ at $I = 0.1[A]$ for different reactive partial pressure combinations [40].

In [62], the parameter identification of the reduced model in Eq. (11) has been presented. The identification process begins with the construction of a matrix using the inputs and outputs of the system (see Eq. (12)). According to this approach, the parameters are identified from measured samples of the system input, system output and of the derivative of the system output.

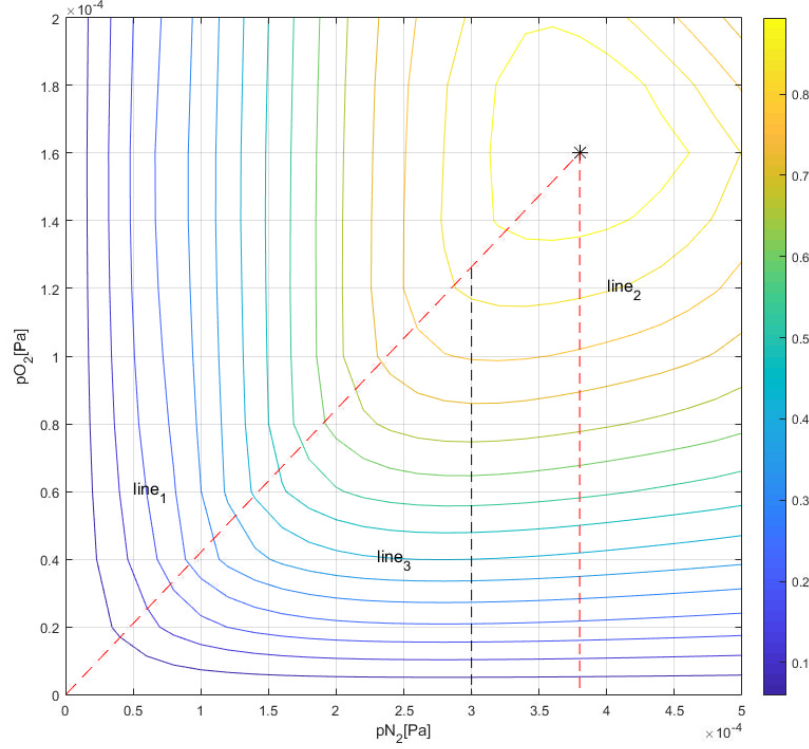


Figure 9: The contour plot of the substrate coverage and the lines used to delimit the preferred area [40].

$$\underbrace{\begin{pmatrix} y_{m1}^3 & y_{m1}^2 & y_{m1} & 1 & u_{m1} \\ y_{m2}^3 & y_{m2}^2 & y_{m2} & 1 & u_{m2} \\ y_{m3}^3 & y_{m3}^2 & y_{m3} & 1 & u_{m3} \\ y_{m4}^3 & y_{m4}^2 & y_{m4} & 1 & u_{m4} \\ y_{m5}^3 & y_{m5}^2 & y_{m5} & 1 & u_{m5} \\ \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix}}_M \underbrace{\begin{pmatrix} A \\ B \\ C \\ D \\ E \end{pmatrix}}_n = \underbrace{\begin{pmatrix} \dot{y}_{m1} \\ \dot{y}_{m2} \\ \dot{y}_{m3} \\ \dot{y}_{m4} \\ \dot{y}_{m5} \\ \vdots \end{pmatrix}}_r \quad (12)$$

Calculating the Moore-Penrose type left pseudoinverse matrix gives the parameter vector $\mathbf{n}$ of the system under examination (see Eq. (13)). The parameter vector obtained by Eq. (13) can be used to calculate the parameters of

the original state-space system $(\mathbf{a}_{i,j})$. Equations (11-13) have been presented in this form in [67].

$$\mathbf{n} = \mathbf{M}^+ \mathbf{r} \quad (13)$$

4 Conclusion and future challenges

In 2019, Depla et al. published an article on the current opportunities and challenges in modeling reactive magnetron sputtering [28]. They mentioned four areas that still need considerable attention:

1. redeposition of sputtered atoms,
2. studying I-V characteristics,
3. sample rotation,
4. pulsing discharge currents.

In our opinion, in future research on this topic emphasis should and probably will be placed on the following key points:

1. delimitation of ranges for some parameters in macroscopic models with the aid of the results obtained from detailed kinematic modeling,
2. precise identification of parameters with the help of the macroscopic dynamic models using measureable data such as partial pressures, gas flows, substrate temperatures, spectrum intensities, voltages and current values,
3. the use of online parameter identification algorithms to identify real-time substrate coverage and composition, mainly for control purposes,
4. development of macroscopic models that, from the perspective of process control, adequately integrate the models used to describe phenomena on the surfaces of both target and substrate as well as models used to describe the state of the plasma discharge.

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References

- [1] T. Abe, T. Yamashina. [The deposition rate of metallic thin films in the reactive sputtering process.](#) *Thin Solid Films*, **30**, 1 (1975) 19–27. [⇒ 120](#)
- [2] Z. Ahmad, B. Abdallah. [Controllability analysis of reactive magnetron sputtering process.](#) *Acta Physica Polonica, A.*, **123**, 1 2013. [⇒ 127](#)
- [3] S. Berg, H.-O. Blom, T. Larsson, C. Nender. [Modeling of reactive sputtering of compound materials.](#) *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **5**, 2 (1987) 202–207. [⇒ 120, 124](#)
- [4] S. Berg, C. Nender. [Modeling of mass transport and gas kinetics of the reactive sputtering process.](#) *Le Journal de Physique IV*, 5 (C5):C5–45, 1995. [⇒ 120, 124](#)
- [5] S. Berg, T. Nyberg, H.-O. Blom, C. Nender. [Computer modeling as a tool to predict deposition rate and film composition in the reactive sputtering process.](#) *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **16**, 3 (1998) 1277–1285. [⇒ 120, 122, 123, 124](#)
- [6] S. Berg, T. Nyberg. [Fundamental understanding and modeling of reactive sputtering processes.](#) *Thin solid films*, **476**, 2 (2005) 215–230. [⇒ 117](#)
- [7] M.-M.-M. Bilek, D.-R. McKenzie. [Predicting the structure of plasma deposited materials.](#) *Czechoslovak Journal of Physics* **52** (2002) 905–920. [⇒ 119](#)
- [8] C.-K. Birdsall, A.-B. Langdon. *Plasma Physics Via Computer Simulation, Bristol, UK*. IOP Publishing, 1991. [⇒ 116](#)
- [9] A. Bogaerts, M. van Straaten, R. Gijbels. [Monte Carlo simulation of an analytical glow discharge: motion of electrons, ions and fast neutrals in the cathode dark space.](#) *Spectrochimica Acta Part B: Atomic Spectroscopy*, **50**, 2 (1995) 179–196. [⇒ 115, 116](#)
- [10] A. Bogaerts, R. Gijbels, W.-J. Goedheer. [Hybrid Monte Carlo-fluid model of a direct current glow discharge.](#) *Journal of Applied Physics*, **78**, 4 (1995) 2233–2241. [⇒ 116](#)
- [11] A. Bogaerts, M. van Straaten, R. Gijbels. [Description of the thermalization process of the sputtered atoms in a glow discharge using a three-dimensional Monte Carlo method.](#) *Journal of applied physics*, **77**, 5 (1995) 1868–1874. [⇒ 116](#)
- [12] A. Bogaerts, R. Gijbels, W.-J. Goedheer. [Two-dimensional model of a direct current glow discharge: Description of the electrons, argon ions, and fast argon atoms.](#) *Analytical Chemistry*, **68**, 14 (1996) 2296–2303. [⇒ 116](#)
- [13] A. Bogaerts, J. Naylor, M. Hatcher, W.-J. Jones, R. Mason. [Influence of sticking coefficients on the behavior of sputtered atoms in an argon glow discharge: Modeling and comparison with experiment.](#) *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **16**, 4 (1998) 2400–2410. [⇒ 123](#)
- [14] J.-W. Bradley. [The plasma properties adjacent to the target in a magnetron sputtering source.](#) *Plasma sources science and technology*, **5**, 4 (1996) 622. [⇒ 116](#)
- [15] P. Carlsson, C. Nender, H. Barankova, S. Berg. [Reactive sputtering using two reactive gases, experiments and computer modeling.](#) *Journal of Vacuum Science*

- Technology A: Vacuum, Surfaces, and Films*, **11**, 4 (1993) 1534–1539. [⇒ 122, 125](#)
- [16] J.-M. Chappé, N. Martin, J. Lintymer, F. Sthal, G. Terwagne, J. Takadom. [Titanium oxynitride thin films sputter deposited by the reactive gas pulsing process](#). *Applied Surface Science*, **253**, 12 (2007) 5312–5316. [⇒ 128](#)
- [17] D.-J. Christie, W.-D. Sproul, D. Carter. [Mid-frequency dual magnetron reactive co-sputtering for deposition of customized index optical films](#). In *Society of Vacuum Coaters 46 th Annual Technical Conference*, 2003 pp. 393–398. [⇒ 125](#)
- [18] [D.-J. Christie](#). [Power conversion and control for pulsed magnetron reactive sputtering](#). PhD thesis, Colorado State University, 2004. [⇒ 125](#)
- [19] D.-J. Christie. [Making magnetron sputtering work: Modelling reactive sputtering dynamics, part 1](#). *SVC Bulletin*, 2014. pp. 24–27. [⇒ 127](#)
- [20] D.-J. Christie. [Making magnetron sputtering work: Modelling reactive sputtering dynamics, part 2](#). *SVC Bulletin*, 2015, pp. 30–33. [⇒ 127](#)
- [21] D.-J. Christie. [Making magnetron sputtering work: Modelling reactive sputtering dynamics, part 3](#). *SVC Bulletin*, 2015, pp. 38–41. [⇒ 127](#)
- [22] C. Costin, L. Marques, G. Popa, G. Gousset. [Two-dimensional fluid approach to the dc magnetron discharge](#). *Plasma Sources Science and Technology*, **14**, 1 (2005) 168. [⇒ 116](#)
- [23] N.-F. Cramer. [Analysis of a one-dimensional, steady-state magnetron discharge](#). *Journal of Physics D: Applied Physics*, **30**, 18 (1997) 2573. [⇒ 116](#)
- [24] D. Depla, J. Haemers, G. Buyle, R. De Gryse. [Hysteresis behavior during reactive magnetron sputtering of Al₂O₃ using a rotating cylindrical magnetron](#). *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **24**, 4 (2006) 934–938. [⇒ 127](#)
- [25] D. Depla, S. Heirwegh, S. Mahieu, R. De Gryse. [Towards a more complete model for reactive magnetron sputtering](#). *Journal of Physics D: Applied Physics*, **40**, 7 (2007) 1957. [⇒ 117](#)
- [26] D. Depla, S. Mahieu, et al. [Reactive Sputter Deposition](#), volume 109. Springer, 2008. [⇒ 116, 117, 121](#)
- [27] D. Depla, X.-Y. Li, S. Mahieu, K.-V. Aeken, W.-P. Leroy, J. Haemers, R. De Gryse, A. Bogaerts. [Rotating cylindrical magnetron sputtering: Simulation of the reactive process](#). *Journal of Applied Physics*, **107**, 11 (2010) 113307. [⇒ 117, 127](#)
- [28] D. Depla, K. Strijckmans, A. Dulmaa, F. Cougnon, R. Dedoncker, R. Schelfhout, I. Schramm, F. Moens, R. De Gryse. [Modeling reactive magnetron sputtering: Opportunities and challenges](#). *Thin Solid Films*, 2019. [⇒ 131](#)

-
- [29] J. Goree, T.-E. Sheridan. [Magnetic field dependence of sputtering magnetron efficiency](#). *Applied physics letters*, **59**, 9 (1991) 1052–1054. [⇒116](#)
 - [30] T. Hammerschmidt, A. Kersch, P. Vogl. [Embedded atom simulations of titanium systems with grain boundaries](#). *Physical Review B*, **71**, 20 (2005) 205409. [⇒118](#)
 - [31] J. Heller. [Reactive sputtering of metals in oxidizing atmospheres](#). *Thin Solid Films*, **17**, 2 (1973) 163–176. [⇒120](#)
 - [32] M.-A. Karolewski. Kalypso: a software package for molecular dynamics simulation of atomic collisions at surfaces. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, **230**, 1-4 (2005) 402–405. [⇒117](#)
 - [33] A. Kelemen, D. Biró, A.-Zs. Fekete, L. Jakab-Farkas, R.-R. Madarász. [Macroscopic thin film deposition model for the two-reactive-gas sputtering process](#). *Acta Universitatis Sapientiae Electrical and Mechanical Engineering*, **8** (2016) 62–78. [⇒125](#), [127](#)
 - [34] S. Kikkawa, M. Fujiki, M. Takahashi, and F. Kanamaru. [Reactive co-sputter deposition and successive annealing of fe-al-n thin film](#). *Journal of the Japan Society of Powder and Powder Metallurgy*, **44**, 7 (1997) 674–677. [⇒125](#)
 - [35] R.-L. Kinder, M.-J. Kushner. [Wave propagation and power deposition in magnetically enhanced inductively coupled and helicon plasma sources](#). *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **19**, 1 (2001) 76–86. [⇒116](#)
 - [36] T. Kubart, O. Kappertz, T. Nyberg, S. Berg. [Dynamic behaviour of the reactive sputtering process](#). *Thin Solid Films*, **515**, 2 (2006) 421–424. [⇒117](#)
 - [37] W.-P. Leroy, S. Mahieu, R. Persoons, D. Depla. [Method to determine the sticking coefficient of o2 on deposited al during reactive magnetron sputtering, using mass spectrometry](#). *Plasma Processes and Polymers*, **51**, 6 (2009) S342–S346. [⇒124](#)
 - [38] W.-P. Leroy, S. Mahieu, R. Persoons, D. Depla. [Quantification of the incorporation coefficient of a reactive gas on a metallic film during magnetron sputtering: The method and results](#). *Thin Solid Films*, **518**, 5 (2009) 1527–1531. [⇒124](#)
 - [39] W.-P. Leroy, S. Mahieu, D. Depla, A.-P. Ehiasarian. [High power impulse magnetron sputtering using a rotating cylindrical magnetron](#). *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **28**, 1 (2010) 108–111. [⇒127](#)
 - [40] R.-R. Madarász and A. Kelemen. [Stoichiometry control of the two gas reactive sputtering process](#). In *2019 IEEE 19th International Symposium on Computational Intelligence and Informatics and 7th IEEE International Conference on Recent Achievements in Mechatronics, Automation, Computer Sciences and Robotics (CINTI-MACRo)*, pp. 000217–000222. IEEE, 2019. [⇒128](#), [129](#), [130](#)
 - [41] N. Martin, R. Sanjines, J. Takadoum, F. Lévy. [Enhanced sputtering of titanium oxide, nitride and oxynitride thin films by the reactive gas pulsing technique](#). *Surface and Coatings Technology*, **142** (2001) 615–620. [⇒128](#)

- [42] H.-E. McKelvey. [Rotatable sputtering apparatus](#), May 1 1984. US Patent 4,445,997. [⇒127](#)
- [43] C. Misiano and E. Simonetti. [4.4 co-sputtered optical films](#). *Vacuum*, 27, 4 (1977) 403–406. [⇒125](#)
- [44] W. Möller, W. Eckstein, J.-P. Biersack. [Tridyn-binary collision simulation of atomic collisions and dynamic composition changes in solids](#). *Computer Physics Communications*, **51**, 3 (1988) 355–368. [⇒117](#)
- [45] W. Möller, M. Posselt. [TRIDYN_FZR User Manual](#). FZR Dresden, 2001. [⇒117](#)
- [46] M. Moradi, C. Nender, S. Berg, H.-O. Blom, A. Belkind, Z. Orban. [Modeling of multicomponent reactive sputtering](#). *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, **9**, 3 (1991) 619–624. [⇒125](#)
- [47] E. Penilla, J. Wang. [Pressure and temperature effects on stoichiometry and microstructure of nitrogen-rich tin thin films synthesized via reactive magnetron dc-sputtering](#). *Journal of Nanomaterials*, 2008. [⇒118](#)
- [48] L.-M. Popescu. [A computer code package for Monte Carlo photon-electron transport simulation: Comparisons with experimental benchmarks](#). *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, **161** (2000) 318–322. [⇒117](#)
- [49] I.-A. Porokhova, Y.-B. Golubovskii, J. Bretagne, M. Tichy, J.-F. Behnke. Kinetic simulation model of magnetron discharges. *Physical Review E*, **63**, 5 (2001) 056408. [⇒116](#)
- [50] I.A. Porokhova, Y.-B. Golubovskii, J.-F. Behnke. [Anisotropy of the electron component in a cylindrical magnetron discharge. i. theory of the multiterm analysis](#). *Physical Review E*, **71**, 6 (2005) 066406. [⇒116](#)
- [51] I.-A. Porokhova, Y.-B. Golubovskii, J.-F. Behnke. [Anisotropy of the electron component in a cylindrical magnetron discharge. ii. application to real magnetron discharge](#). *Physical review E*, **71**, 6 (2005) 066407. [⇒116](#)
- [52] R.-K. Porteous, D.-B. Graves. [Modeling and simulation of magnetically confined low-pressure plasmas in two dimensions](#). *IEEE transactions on plasma science*, **19**, 2 (1991) 204–213. [⇒116](#)
- [53] T.-E. Sheridan, M.-J. Goeckner, J. Goree. [Model of energetic electron transport in magnetron discharges](#). *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, 8, 1 (1990) 30–37. [⇒116](#)
- [54] F. Shinoki, A. Itoh. [Mechanism of rf reactive sputtering](#). *Journal of Applied Physics*, 46, 8 (1975) 3381–3384. [⇒120](#)
- [55] W.-D. Sproul, D.-J. Christie, D.-C. Carter. [Control of reactive sputtering processes](#). *Thin solid films*, **491**, 1-2 (2005) 1–17. [⇒125](#)
- [56] K. Strijckmans, W.-P. Leroy, R. De Gryse, D. Depla. [Modeling reactive magnetron sputtering: Fixing the parameter set](#). *Surface and Coatings Technology*, 206, 17 (2012) 3666–3675. [⇒117](#), [124](#)
- [57] K. Strijckmans and D. Depla. [A time-dependent model for reactive sputter deposition](#). *Journal of Physics D: Applied Physics*, 37, 23 (2014) 235302. [⇒117](#), [127](#)

- [58] K. Strijckmans. *Modeling the reactive magnetron sputtering process*. PhD Thesis, Ghent University, 2015. [⇒114, 123, 125, 126](#)
- [59] R. Terry, K. Gibbons, S. Zarrabian. *Method and apparatus for reactive sputtering employing two control loops*, August 22 2000. US Patent 6,106,676. [⇒127](#)
- [60] A.-E. Wendt, M.-A. Lieberman, H. Meuth. *Radial current distribution at a planar magnetron cathode*. *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, 6, 3 (1988) 1827–1831. [⇒116](#)
- [61] A.-E. Wendt, M.-A. Lieberman. *Spatial structure of a planar magnetron discharge*. *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, 8, 2 (1990) 902–907. [⇒116](#)
- [62] C. Woelfel, P. Awakowicz, J. Lunze. *Model reduction and identification of nonlinear reactive sputter processes*. *IFAC-PapersOnLine*, 50, 1 (2017) 13728–13734. [⇒115, 127, 128, 129](#)
- [63] C. Woelfel, P. Awakowicz, J. Lunze. *Robust high-gain control of nonlinear reactive sputter processes*. In *2017 IEEE Conference on Control Technology and Applications (CCTA)*, IEEE, 2017, pp 25–30. [⇒128](#)
- [64] C. Woelfel, P. Awakowicz, J. Lunze. *Tuning rule for linear control of nonlinear reactive sputter processes*. In *2017 21st International Conference on Process Control (PC)*, IEEE, 2017, pp. 109–114. [⇒128](#)
- [65] C. Woelfel, S. Kockmann, P. Awakowicz, J. Lunze. *Model identification of nonlinear sputter processes*. In *2017 17th International Conference on Control, Automation and Systems (ICCAS)*, IEEE, 2017, pp. 182–187. [⇒128](#)
- [66] C. Woelfel, S. Kockmann, P. Awakowicz, J. Lunze. *Neural network based linearization and control of sputter processes*. In *2017 11th Asian Control Conference (ASCC)*, IEEE, 2017, pp. 2831–2836. [⇒128](#)
- [67] C. Woelfel, D. Bockhorn, P. Awakowicz, J. Lunze. *Model approximation and stabilization of reactive sputter processes*. *Journal of Process Control*, 2018. [⇒128, 131](#)
- [68] Y. Yamamura and M. Ishida. *Monte Carlo simulation of the thermalization of sputtered atoms and reflected atoms in the magnetron sputtering discharge*. *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, 13, 1 (1995) 101–112. [⇒117](#)

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