Surface & Coatings Technology 237 (2013) 176–181

Contents lists available at ScienceDirect

Surface & Coatings Technology

journal homepage: www.elsevier.com/locate/surfcoat

Continuum and kinetic simulations of the neutral gas flow in an industrial physical vapor deposition reactor

Kirsten Bobzin [a], Ralf Peter Brinkmann [b], Thomas Mussenbrock [b], Nazlim Bagcivan [a], Ricardo Henrique Brugnara [a], Marcel Schäfer [a], Jan Trieschmann [a,b,*]

[a] RWTH Aachen University, Faculty of Mechanical Engineering, Surface Engineering Institute, D-52056 Aachen, Germany
[b] Ruhr University Bochum, Faculty of Electrical Engineering and Information Technology, Institute of Theoretical Electrical Engineering, D-44780 Bochum, Germany

ARTICLE INFO

Available online 19 August 2013

Keywords:
CFD
DSMC
Neutral gas dynamics
Rarefied gas flow

ABSTRACT

Magnetron sputtering used for physical vapor deposition processes often requires gas pressures well below 1 Pa. Under these conditions the gas flow in the reactor is usually determined by a Knudsen number of about one, i.e., a transition regime between the hydrodynamic and the rarefied gas regime. In the first, the gas flow is well described by the Navier–Stokes equations, while in the second a kinetic approach via the Boltzmann equation is necessary. In this paper the neutral gas flow of argon and molecular nitrogen gas inside an industrial scale plasma reactor was simulated using both a fluid model and a fully kinetic Direct Simulation Monte Carlo model.
By comparing both model results the validity of the fluid model was checked. Although in both models a Maxwell–Boltzmann energy distribution of the neutral particles is the natural outcome, the results of the gas flow differ significantly. The fluid model description breaks down, due to the inappropriate assumption of a fluid continuum. This is due to exclusion of non-local effects in the multi dimensional velocity space, as well as invalid gas/wall interactions. Only the kinetic model is able to provide an accurate physical description of the gas flow in the transition regime. Our analysis is completed with a brief investigation of different definitions of the local Knudsen number. We conclude that the most decisive parameter — the spatial length scale L — has to be very careful chosen in order to obtain a reasonable estimate of the gas flow regime.

1. Introduction

Physical Vapor Deposition (PVD) processes, such as the well established cathodic arc evaporation (CAE) and DC Magnetron Sputtering (DC-MS), as well as the promising High Power Pulsed Magnetron Sputtering (HPPMS, often referred to as HiPIMS) technology can be used for the production of hard protective coatings in corrosion and wear resistance applications [1–3]. In this context a uniform layer of coating material is an essential requirement. To optimize industrial PVD processes in terms of the quality of the obtained coatings, it is important to understand not only the discharge characteristics, but moreover to obtain a detailed picture of the neutral gas flow inside the reactor chamber.

The governing parameter commonly used for the analysis and the characterization of the gas flow regime is the Knudsen number Kn [4,5]. Kn allows to approximately estimate the flow regime in a given setup by specifying the degree of gas rarefaction [5]. It is commonly defined as the ratio of the mean free path λ to a representative (but local) spatial scale L,

$$Kn = \frac{\lambda}{L}. \tag{1}$$

In a gas in an equilibrium state with number density n, the mean free path can be estimated by $\lambda = \left(n\sigma\sqrt{2}\right)^{-1}$, where the hard sphere collision cross section $\sigma = \pi d^2$ may be used. A representative length scale L can be chosen based on geometric considerations. Moreover, following Boyd et al. [6] it can be defined through the normalized gradient of a local flow property Q by $L = |Q|/|\nabla Q|$. Bird suggests a choice of Q based on the mass density $Q = \rho = mn$ [4]. This is unfeasible for incompressible flows, which can be be assumed in the case investigated (with a low Mach number of at maximum $M = |\vec{v}|/c \approx 0.15$). Thus we choose the momentum as the characteristic flow property $Q = \rho\vec{v}$. This choice of Kn based on either the geometric or the gradient approach is addressed in a later context.

Regarding the analysis of the flow regime based on Kn, the following distinctions can be made: for $Kn \leq 0.1$, continuum models based on the Navier–Stokes equations — usually implemented in computational fluid dynamic (CFD) simulations — allow for a precise description of the gas flow. Such CFD models have found widespread applications in manifold areas of aerospace and automotive engineering [7,8]. In contrast, in situations where $Kn > 0.1$ the Navier–Stokes equations prove inadequate for the description of rarefied gas flows, e.g., in micro/nano scale gas flows [9,10], or in gas flows commonly used in low pressure PVD applications. This is due to non-local effects in the multi dimensional velocity space, as well as inappropriate treatment of gas/wall interactions.

* Corresponding author. Tel.: +492343229475.
E-mail address: jan.trieschmann@rub.de (J. Trieschmann).

0257-8972/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.surfcoat.2013.08.018

Moreover, in plasma processing a continuum representation (implying a Maxwell–Boltzmann energy distribution) is likely not valid, due to the interaction of neutral gas particles with non-equilibrium species (heavy particle, or electrons), leading to an overall non-equilibrium situation. Under rarefied conditions, in general, only kinetic models based on the Boltzmann equation provide an accurate description. Such scenarios are often solved by means of the Direct Simulation Monte Carlo (DSMC) method proposed by Bird [4]. At very low pressures where a stochastic description of the particle interaction with the background gas is justified, the Test Particle Monte Carlo (TPMC) method [11] is most commonly used. When using the DSMC method the Boltzmann equation is directly solved by means of following the trajectory of a sufficiently large number of pseudo-particles subject to collisions among themselves, as well as with the surrounding walls. It is interesting to note that, although hydrodynamic and Monte Carlo methods have been studied extensively [12–16], generalized limitations for the validity of the different models in the transition regime $0.01 < Kn < 2$ are not given.

The aim of this work is to discuss the validity of a conventional continuum model in the transition regime. For the analysis we apply the commercially available continuum fluid solver *FLUENT* [17]. In order to allow for a detailed comparison, we have modified — with respect to boundary conditions — the DSMC solver *dsmcFoam* [18–20]. In this work we analyze the neutral gas flow inside a reactor chamber used for DC-MS and HPPMS processes. We present a brief description of the investigated reactor system and motivate our analysis of the two different numerical algorithms. We give a short review of both numerical models. A detailed discussion of simulation results of the identical vacuum setup obtained via the two models is provided. Finally, the results are summarized and a conclusion is drawn. We provide suggestions on the validity of either continuum solution based CFD simulations and kinetic models.

2. Setup

The CemeCon CC800/9 Custom coating unit, investigated in this work, is typically used for DC-MS and HPPMS processes. As illustrated in Fig. 1, the main processing chamber has a floor space of 85×85 cm ^{2}. Additionally, the main chamber is extended towards the pump by a narrowing pump chamber. While the gas inlets (A) are along two of the corners of the main chamber at the left side, the pump (E) is mounted at the large flange at the right side. Inside the main chamber there are six sample holders (C). These sample holders can be static or rotating using a planetary gearing. Behind the sample holders two magnetron cathodes (B) are mounted. The main chamber and the pump chamber are separated by a heater (D). The coating unit is typically operated at pressures around 0.5 Pa. For this study we assume that argon is used as process gas at a flow rate of $F_{Ar} = 200$ sccm, while (molecular) nitrogen is used as reactive gas at a flow rate of $F_{N2} = 40$ sccm. All walls are assumed to have a constant temperature of $T = 300$ K.

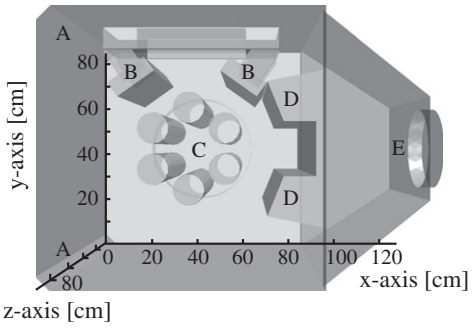

Fig. 1. 3D top view of the PVD coating unit CC800/9 Custom. Gas inlets (A) in both corners at the far left side, pump flange (E) at far right side. Two cathodes (B) on one side of the substrate holders (C). The heater (D) separates the main from the pump chamber.

From geometric considerations, the typical geometric dimension L ranges from about one centimeter (e.g., at small features, the substrates and cathodes) up to a few tens of centimeters (at open space in the vacuum chamber). Additionally, for the pressure of 500 mPa, a mean free path of $\lambda \approx 1.39$ cm can be approximated for an equilibrium gas assuming a molecular diameter of $d = 3.664$ Å for argon [21]. In consequence, one finds the Knudsen number $Kn = \lambda/L$ in the limits $0.05 < Kn < 1.5$. While the lower limit suggests that continuum models can be readily used for a numerical analysis, the upper limit enforces that only a kinetic treatment of particles is valid for a description of the gas flow. In the transition regime, however, no definite statement can be made. Therefore, to gain insight in the gas flow — and the models — an investigation of results from a continuum model based on the Navier–Stokes equations, as well as a kinetic DSMC simulation model, is desired.

3. Numerical models

The modeling of magnetron sputtering processes, involving the interaction of plasma with neutral background gas (in this work argon and molecular nitrogen), as well as the interaction of heavy particles with target materials (sputtering) and walls/substrates (deposition) is of very complex nature. Several authors have investigated the theoretical background of sputtering processes in terms of the plasma/wall interaction [22–24], the description of the deposition of sputtered material on substrates and walls (including the chemical interaction with gas phase species) [25–27], as well as the analysis of the plasma, particularly for HPPMS processes [28–31]. The numerical investigations are often based on particle based models [32–39]. In this work, we concentrate on the neutral gas flow; the interaction of the neutral gas with charged particles from the plasma, energetic heavy particles sputtered off the targets (e.g., gas rarefaction effects) [40], as well as the resulting interaction with the walls (i.e., the deposition process) are intentionally left for a later analysis. For our analysis we employ the CFD software *FLUENT* and a modified version of the DSMC implementation *dsmcFoam* provided with the freely available *OpenFOAM* simulation package [20].

For the CFD simulations, the *FLUENT* software release 14 [17] is used. We simulate the gas flow using a pressure based fluid model using the PISO method [8]. Additionally we use an RNG k-epsilon turbulence model [41] for robust convergence. For further improvement of the numerical solution scheme, two neighbor and two skewness correction iterations are applied, respectively. The ideal gas law is used to obtain the gas density from the pressure and the gas temperature. The walls are described by slip boundary conditions. As for the parameters, we set a mass flux and the wall temperature corresponding to the flow rate and temperature specified in the setup description. We further set the boundary condition at the pump to $p = 430$ mPa.

The DSMC method is based on the idea that a sufficiently large number of pseudo-particles (also referred to as simulators) is kinetically simulated, interacting among each other by means of a given set of collision processes [4]. In this ensemble (here in the converged state approximately 8 million simulators), each pseudo-particle represents a large number of physical particles, in our case 10^{13}. By the original authors of *dsmcFoam*, the solver was benchmarked against a number of examples from the literature [18] and was used for various studies of rarefied gas flow [9,10]. In version 2.1.1 used for this analysis, the implementation provides a simulation tool with the capabilities for arbitrary 2D/3D geometries, an arbitrary number of gas species, variable hard sphere (VHS) collisions, Larsen–Borgnakke internal energy redistribution, and it allows for unlimited parallel processing [18,4]. The original code has been modified to allow for appropriate boundary conditions for both gas species in the present problem: at the gas inlets mass flow rates are specified and at the outlet/pump an absorption probability allows to indirectly assign the pressure inside the vacuum