Multiple scale modeling of Al_2O_3 thin film growth in an ion beam sputtering process

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ABSTRACT

A multiple scale model approach is presented in order to investigate Al_2O_3 thin film growth in the framework of an existing Ion Beam Sputtering (IBS) coating process. Therefore, several simulation techniques are combined via optimized interfaces for realizing the concept of a virtual coater. Characteristic coating process parameters of the IBS coating plant are applied as input parameters to model the material transport in the chamber, the energy and angular distribution of the coating material at the substrate, the formation of structural thin film properties, and the optical as well as the electronic layer properties. The resulting thin film properties are validated to the data of an experimental IBS Al_2O_3 single layer prepared applying the underlying coating facility. The comparison accounts for a good agreement between the modeled layer properties using the virtual coater concept and the experimental characterization data.

Keywords: Modeling, thin film growth, Direct Simulation Monte Carlo, Molecular Dynamics, Density Functional Theory, Ion Beam Sputtering

1. INTRODUCTION

Thin film growth simulation approaches are a reliable tool to investigate in fundamental mechanisms of layer formation and to support experimental optimization procedures for the realization of high quality optical coatings. Since the computing power increases continuously, it became possible to simulate more complex and larger atomistic systems matching well the structural data of common optical thin films [1,2]. In order to perform representative calculations, which can be used to improve the resulting layer properties, the underlying parameters of the coating process have to be taken into account in the growth model. Especially, the generation of the coating material, the transport of the material inside the coating chamber, and, as a result, the characteristic energy and angular distributions of the coating material at the substrates define the growth of the thin films. Furthermore, the resulting optical layer properties, e. g. index of refraction, optical bandgap, extinction coefficient, are of particular interest, because this is the class of properties that is most easily accessible for the coating manufacturer.

In the present contribution, the growth of an Al_2O_3 single layer is studied on a theoretical level applying the deposition parameters and geometry of an experimental Ion Beam Sputtering (IBS) coating plant. To perform a full simulation of the entire coating process, starting with the generation of coating material, the modeling of thin film growth and finally, the calculation of the optical properties of the realized thin film structures, several different simulation techniques were combined to a multiple scale model approach. A Direct Simulation Monte Carlo (DSMC) approach is applied to model the transport of the coating material from the location of generation at the sputter target to the substrates. The resulting characteristic energy and angular distributions of the coating material at the substrates are used in the following to simulate the layer formation with a developed thin film growth approach, which is based on classical Molecular Dynamics (MD). From the classically grown films the structural layer properties, e. g. film density, surface roughness, and stoichiometry are evaluated. Finally, the optical properties are calculated by ab-initio quantum mechanical (QM) simulation techniques on the basis of Density Functional Theory (DFT). In this approach, the determined structural properties are used to prepare small supercells which represent well the modeled thin film and serve as the main input for the DFT simulation. Resulting, the imaginary part of the dielectric function and the frequency dependent index of refraction are modeled. Applying the presented concept of a "virtual coater", a full simulation of the coating process in dependence of characteristic process parameters is carried out.

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2. ION BEAM SPUTTERING FACILITY

The investigated Ion Beam Sputtering coating facility is located at the Laser Zentrum Hannover e.V. (LZH). The setup mainly consists of an ion source, a metallic aluminum target, and a rotatable calotte in which the substrates are mounted. The RIM10 ion source is inductively RF coupled and has a three grating extraction system. Here, the emitted ion current is up to 200 mA. The acceleration voltage can be varied between 200 and 2000 V. In addition, the IBS facility is equipped with an oil-free high vacuum TPH 2301 turbo molecular pump system, having a pumping speed of ~1900 l/s. The acceleration voltage was set to ~1.6 keV during the deposition of the Al_2O_3 single layer and the background pressure was 4.15×10^{-4} mbar. The coating process was operated in a reactive sputtering mode by the supply of Oxygen via a gas inlet, which is located close to the Aluminum target.

The coating chamber geometry and the characteristic process conditions were used in the following to simulate the material transport inside the plant and to calculate the energy and angular distribution of the coating material at the substrate position.

3. TRANSPORT SIMULATIONS

For the modeling of material transport inside the IBS plant by Direct Simulation Monte Carlo, a software package was applied, which has been developed at the Fraunhofer IST. The code is massively parallelized in order to reach feasible computation times and to enable the simulation of ambitious setups as for example the underlying IBS coater. For more detailed information about further investigated topics and the simulation model, the reader is referred to references [3,4]. The DSMC method allows for simulating the transport of neutral species, whereby a statistical approach is employed to solve the Boltzmann equations.

To consider the characteristic setup and dimensions of the IBS coating plant, the geometry is assembled in GMSH, a three dimensional finite element mesh generator [5]. The resulting surface mesh is overlain with a rectangular simulation grid, whereby each grid cell can comprise several particles. In order to reduce the noise and to observe global trends, time averages are taken at certain time intervals for each grid cell. Additionally, the energy and angular distributions of the particles hitting the substrate surface are sampled and accumulated for selected geometric surfaces and allocated as input files for the thin film growth simulations using classical MD. The objective of the performed calculations was to investigate the transport of sputtered coating material and reactive gas species. Therefore, the ion source was idealized by a single surface emitting Ar atoms with energies between 800 and 850 eV perpendicular to the surface. During the interaction of the Ar atoms with the metallic Al target, they were allowed to sputter Al atoms as well as Al_2O_3 with different energy dependent sputter yields. The sputter emission, the transport of sputtered as well as reactive gas species, and the scattering of the particles were monitored in the DSMC simulations, and the characteristic distributions at the substrate surface were evaluated subsequently.

The energy distributions for the Aluminum atoms and the reactive gas Oxygen at the substrate position for different simulation times between 100 and 500 ms is shown in figure 1 and 2, respectively. The Al distribution in figure 1 has a global maximum intensity at energy of approximately 25 meV and a local maximum between 1 and 2 eV. From this it can be assumed, that the first peak corresponds to thermalized Al atoms, whereby the second is due to the Thompson energy distribution of the sputtered particles at the target. Furthermore, it can be observed, that the distributions show a clear dependence as a function of simulation time. Nevertheless, the system reaches a stable configuration after already 500 ms, and this configuration was applied to the subsequently performed thin film growth simulations. The O_2 energy distribution displayed in figure 2 shows a maximum intensity at energy of about 25-30 meV and therefore, only thermalized O_2 molecules reach the substrate surface. Again, a stable state can be observed after a simulation time of 500 ms.

The angular distributions of Aluminum atoms and O_2 molecules at the substrate surface are presented in figure 3 and 4, respectively. In particular, it could be observed that both distributions are independent of simulation time.

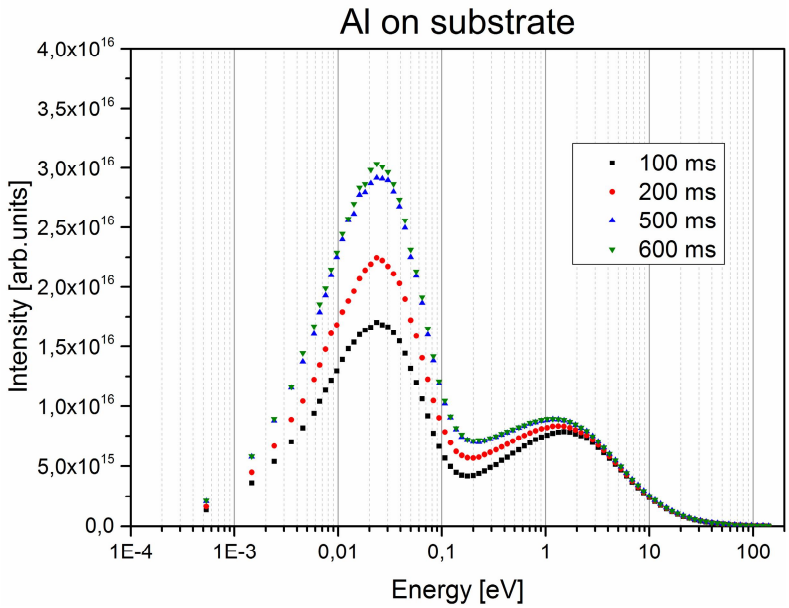

Figure 1. Energy distribution of sputtered Al at the substrate surface.

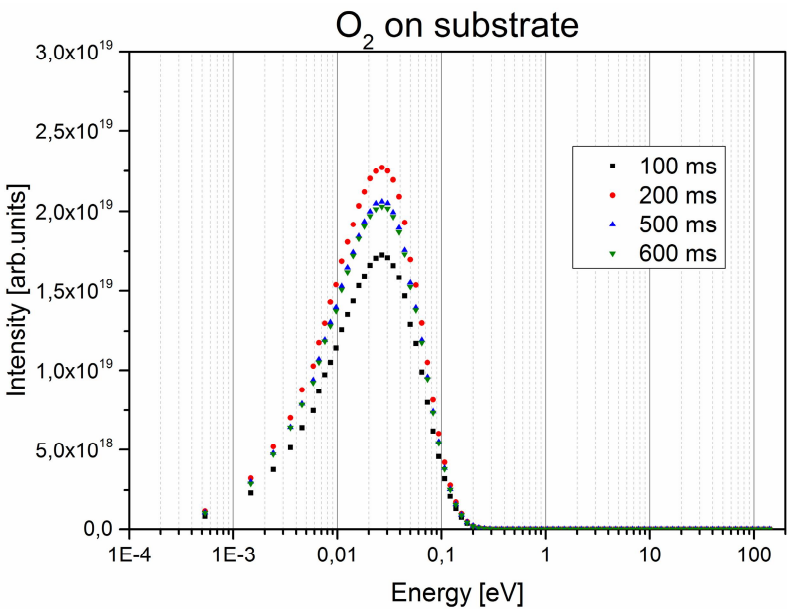

Figure 2. Energy distribution of O_2 molecules at the substrate surface.

The Aluminum distribution shows a shift of the maximum to a polar angle of approximately 30° (azimuth angle ~180°). At this position, the distribution is quite symmetric for the azimuth angle. The reason for the shift can be explained by the geometry of the IBS coating plant, whereby the ion source, the target, and the substrate are arranged in the yz-plane. The angular distribution for the O_2 molecules is almost symmetric in the xy-plane, whereby no shift of the maximum can be observed. This can also be declared by the geometrical position of the gas inlet in the chamber, which is located close to the target and with perpendicular orientation to the substrate surface.

In the next section, the thin film growth simulation according to the calculated coating material distributions is presented.

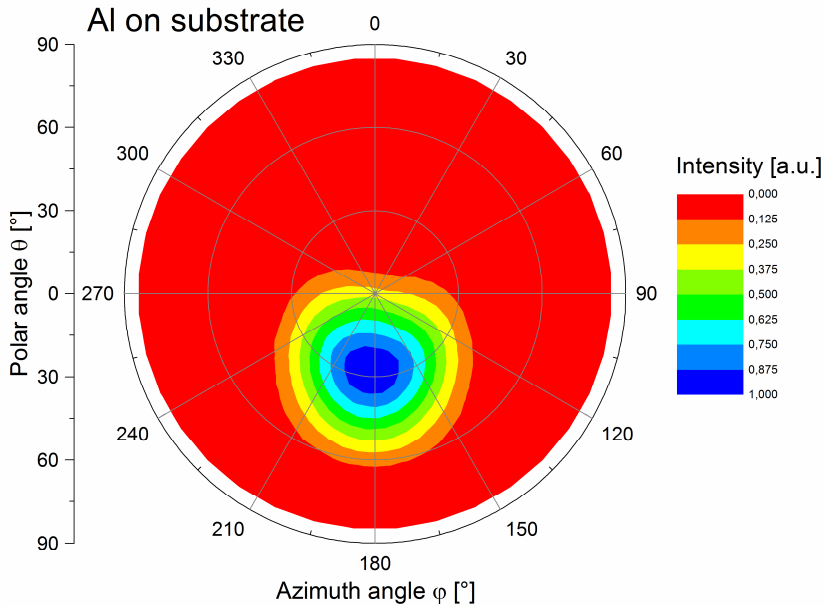

Figure 3. Angular distribution of sputtered Al at the substrate surface.

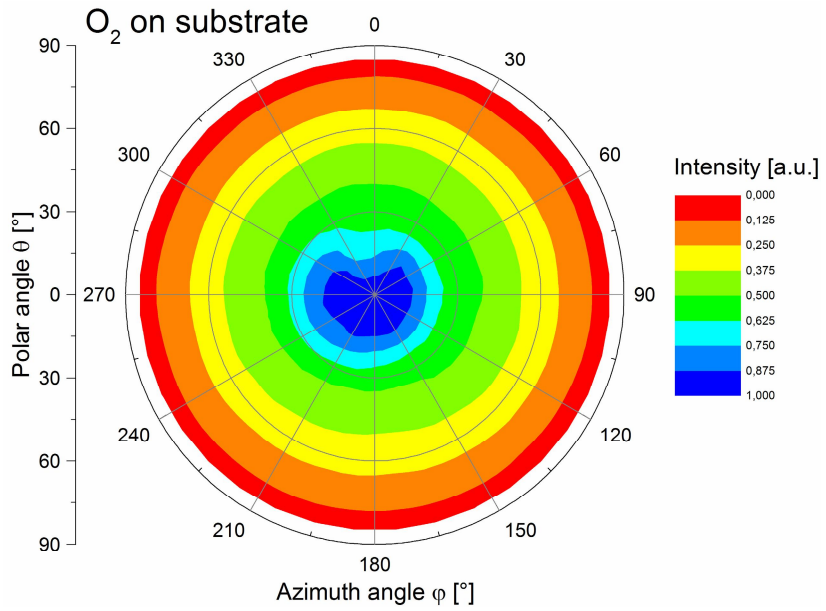

Figure 4. Angular distribution of O_2 at the substrate surface.

4. THIN FILM GROWTH SIMULATIONS

Thin film growth simulations were carried out applying a developed theoretical model based on classical Molecular Dynamics [2]. In this atomistic simulation technique, the particle positions are developed in time by solving the Newton´s equation of motion. Hence, a classical interaction potential has to be defined in advance, describing well the interactions between all involved particles in the system of interest. In the case of amorphous alumina, the classical potential predicted by Gutiérrez et. al. [6] is often applied. This potential consists of a Coulomb part and a Buckingham part, which is further divided into repulsive and van der Waals term. The analytical form of the potential and the

necessary fitting parameters (A, ρ, and C) for calculating the interactions between the different species are given in equation 1 and table 1, respectively. Here, q_i and q_j are the effective charges (q_{Al} = 1.4175 and q_O = -0.9450), ε_0 is the electrical permittivity, r_{ij} is the distance between atom i and j. Obviously, the classical potential depends only on the distance r_{ij} between atom i and j.

$$V(r_{ij}) = \frac{q_i q_j}{4\pi\varepsilon_0 r_{ij}} + A\exp\left(-\frac{r_{ij}}{\rho}\right) - \frac{C}{r_{ij}^6} \tag{1}$$

Table 1. Calculated parameters in accordance to the predicted potential by Gutiérrez et. al. [6]

i-j	**A**	**ρ**	**C**
	[eV]	[Å]	[Å^6 eV]
Al - Al	31557350.85	0.068	14.04518
Al - O	28465.10	0.172	34.56365
O - O	6460.00	0.276	85.05738

For the deposition of an alumina thin film, an Al_2O_3 base structure was created, in the following referred to as substrate, having 9600 atoms and a ground area (x,y) of approximately 56 nm^2. Periodic boundary conditions were only applied in the ground area directions in order to enable thin film growth in z direction. The deposition of each coating atom is modeled by a single MD calculation, which is subdivided in two parts using first a microcanonical and subsequently a canonical ensemble for the time development. This procedure secures the physically correct modeling of the energy input in the film structure during deposition and the subsequent cooling of the structure to a user defined substrate temperature.

For the deposition of the Al atoms a random position above the surface is determined and the kinetic energy as well as the angle of incidence were selected from the characteristic distributions calculated by the DSMC simulation of the IBS coating chamber (figure 1 and 3). Since Oxygen is provided as reactive gas in the IBS coating process, the supply of Oxygen in the growth simulation is modeled in a developed saturation event [2]. The saturation event ensures the realistic modeling of the reactive sputtering process using a metallic target including high energetic target atoms and thermalized Oxygen atoms. In this procedure, the whole surface is saturated with low energetic Oxygen atoms after a specified number of deposited Al atoms. In the following, the system is developed in time applying a MD simulation. Afterwards all Oxygen atoms, which are not deposited on the surface, are removed from the total system and the next Al deposition sequence is started. In this way it became possible to investigate also metal oxide thin film growth applying a metallic target material.

The Molecular Dynamics calculations were carried out applying the simulation package Dl_poly [7]. The time integration of the atomic ensemble was performed applying the Velocity Verlet algorithm [8]. Here, a timestep of size 2 fs was used and the total integration time for each deposition event was approximately 1 ps. The substrate temperature in the growth simulations was set to 300 K. The calculation procedure consisting of Aluminum deposition and saturation with low energetic Oxygen was carried out until a number of 10^5 deposited atoms was reached.

In the following, the structural layer properties, e.g., film density, layer stoichiometry, and surface roughness were calculated applying suitable analysis algorithms. The density of the resulting film structure was evaluated to be 3.383 g/cm^3 and the surface roughness was 0.195 nm rms. The results are of same magnitude comparing to the modeled data presented in [2], where the structural layer properties were investigated in dependence of discrete deposition energies and perpendicular angle of incidence. The ratio between Aluminum and Oxygen atoms in the film was calculated to be Al:O = 2:2.986, hence it can be stated that a stoichiometric thin film was realized. Since the optical properties are more easily accessible for the coater manufacturer, the index of refraction was calculated in the following and compared to the data of an IBS alumina single layer.

5. OPTICAL PROPERTIES

For the determination of the optical and electronic properties of the modeled thin film structures quantum mechanical simulation techniques based on Density Functional Theory were applied. Since the DFT calculations are extremely computation time consuming, the structure sizes to be modeled are limited to few hundred of atoms. Therefore, it is not possible to model the optical properties of the full thin film structure consisting of ~10^5 atoms with the current state of the art computation power. To solve this problem a small idealized supercell was prepared, which represents well the structural properties of the large thin film structure. Subsequently it became possible to calculate the optical and electronic properties of interest applying DFT. Three important structural properties were defined in advance, which were transferred to the idealized supercell, namely the density of the thin film and the stoichiometry as well as the non-periodic, amorphous structure. The schematically drawn interface between classical thin film growth structure and quantum mechanical calculation is shown in figure 5. The atom positions in the supercell were chosen randomly to avoid influences of user defined settings on the results.

The calculation procedure to determine the optical and electronic properties is divided into two parts. A more detailed description of the procedure is published in [1,9]. In the first part the supercell is subjected to a relaxation of the atomic positions applying QM algorithms until an equilibrium state is reached. In the second part, the electronic and optical properties are calculated applying a suitable functional. For the presented simulations, the Vienna ab initio simulation package VASP [10] was used. Here, the calculations were performed on the basis of Density Functional Theory using the PAW [11] method. The plane wave cutoff energy was set to 400 eV and the HSE06 hybrid functional [12] was applied to calculate the optical properties.

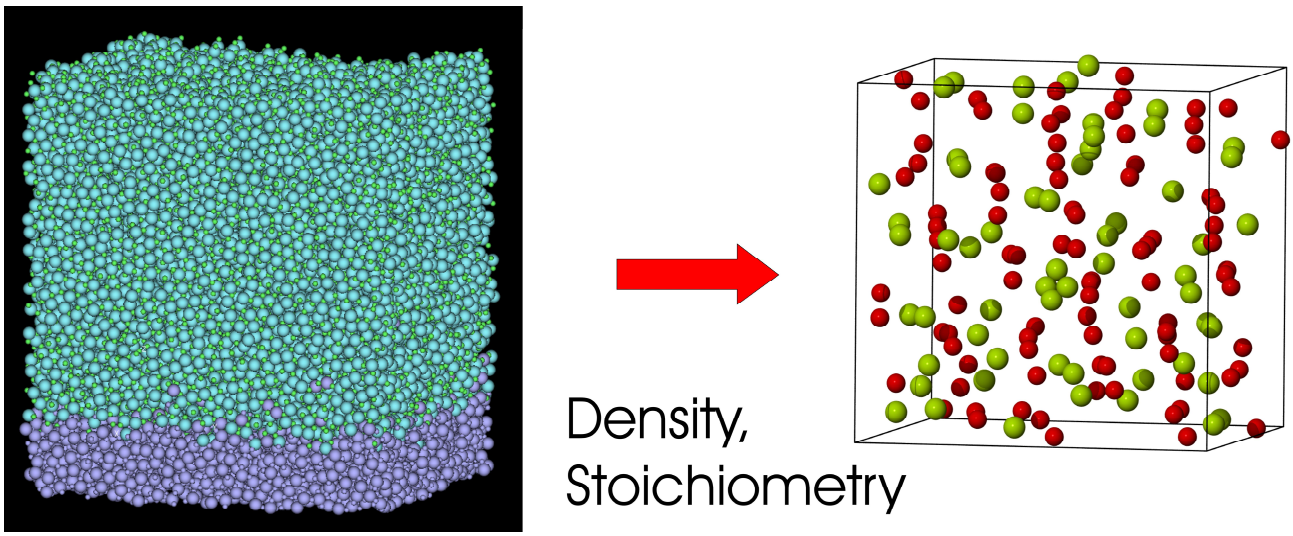

Figure 5. Schematical representation of interface between classical growth simulation and DFT calculation of optical and electronic film properties.

The result of the calculation is the frequency dependent imaginary part of the dielectric function. Via the well-known Kramers-Kronig relation also the real part is accessible. Having the full expression of the dielectric function, optical properties like the index of refraction, the extinction coefficient, the absorption coefficient, and the reflectivity can be calculated. For the validation to experimental results, the index of refraction was evaluated as the average value of the refractive index for each major axis of the supercell.

For comparison, the IBS coated Al_2O_3 single layer was characterized by spectral photometric measurement according to ISO 15368 [13] and the wavelength dependent refractive index was evaluated using the Sellmeier equation as implemented in the thin film software "SPECTRUM" [14]. In figure 6, the calculated and characterized index of

refraction dispersion data is shown as a function of wavelength. Obviously, a very good agreement between theoretical and experimental results can be obtained with a small offset of approximately 0.02. In future studies it is intended to compare also the absorption coefficient as well as the bandgap. Here, especially a more detailed investigation in k-point sampling and the choice of the functional have to be performed in more detail.

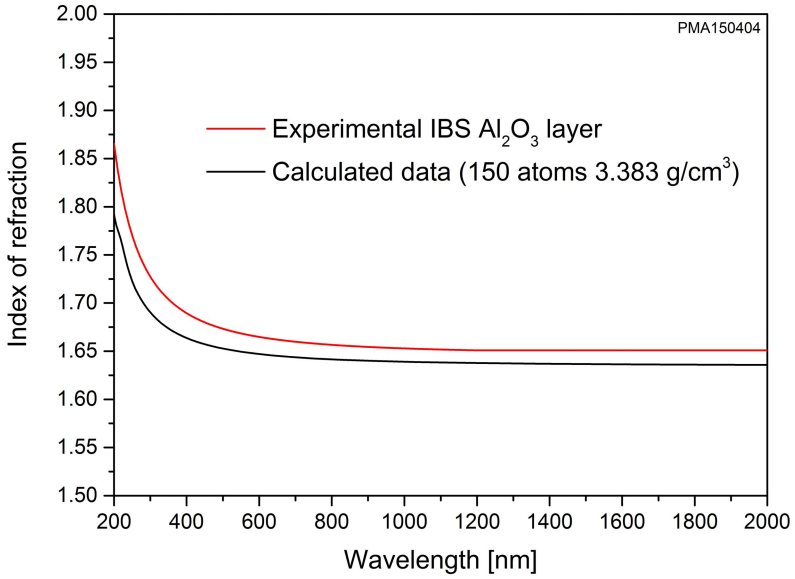

Figure 6. Index of refraction for the modeled Al_2O_3 structure and an experimental IBS single layer.

6. SUMMARY

A multiple atomistic scale model was applied to investigate Al_2O_3 thin film growth in the framework of an existing IBS coating process. The developed "virtual coater" concept combines DSMC, classical MD and DFT techniques via appropriate interfaces in order to investigate material transport, thin film growth, and optical as well as electronic layer properties. The calculated energy distribution of the Aluminum atoms at the substrate shows a clear subdivision in a thermalized and a high energetic part. Furthermore, the maximum of the angular distribution for Aluminum is shifted to a polar angle of about 30°, which is due to the geometrical orientation of the different components inside the chamber. The simulated structural layer properties are in agreement to previous studies on Al_2O_3 thin film growth [2] and the calculated index of refraction data matches very well with experimental results of a prepared IBS alumina single layer. These results show the excellent performance of the developed virtual coater concept and hence, it is intended to apply this method to other layer materials and coating processes of interest.

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