Thermal barrier coating deposition by rarefied gas jet assisted processes: Simulations of deposition on a stationary airfoil

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The uniform coating of a complex shaped substrate, such as a gas turbine airfoil, by collisionless physical vapor deposition processes requires rotation/translation of the substrate or sources and is inconceivable for regions on the substrate that are never in the line-of-sight of the vapor source. Recently developed directed vapor deposition processes use electron beam evaporation and inert gas jets to entrain, transport, and deposit metal oxide vapor in an environment where many vapor atom collisions occur prior to deposition. Direct simulation Monte Carlo simulations and experimental depositions of a rare earth modified thermal barrier coating are used to investigate fundamental aspects of the deposition process, including coating thickness and column orientation, over the surface of a nonrotated model airfoil substrate with substantial non-line-of-sight regions. The coating thickness uniformity was found to depend on the deposition chamber pressure and the pressure ratio between the low-pressure deposition chamber and high-pressure reservoir upstream of the gas jet forming nozzle. Under slow flow conditions, significant coating of the non-line-of-sight regions was possible. The growth column orientation is found to also vary over the substrate surface due to changes in the local incidence angle distribution of depositing vapor atoms. The variation in growth column orientation is not predictable by the Tangent rule widely used for predicting columnar growth orientation in physical vapor deposition processes. © 2013 American Vacuum Society.

I. INTRODUCTION

Gas turbine engines are widely used for aircraft and ship propulsion, as well as electrical power generation. The performance of these engines has been significantly improved through increases in the gas temperature at the inlet to the turbine section of the engine. In this region of the engine, the momentum of the high temperature combustion gases is partially converted to rotation of a disk by interaction with a set of turbine blades (airfoils) attached to the periphery of the disk. These turbine airfoils are subjected to one of the most extreme thermomechanical and chemically aggressive environments encountered by a materials system, and their durability in this environment paces further advances in engine performance.

Gas turbine airfoils are currently made from superalloys. Those in the hottest locations are cast in a single crystal form to reduce creep deformation rates under the severe centrifugally created stresses associated with engine operation. The airfoil interiors are hollow, and the thin metal wall is penetrated by many small holes so that compressed air can be used to reduce the airfoil’s temperature and rate of degradation. To further protect the superalloy airfoils, they are coated with a metallic bond coat whose composition is optimized to reduce the rates of both oxidation and hot corrosion (usually at the expense of bond coat creep resistance). Further durability is achieved by depositing a thermal barrier coating (TBC) on the bond coat. The most commonly used thermal barrier is a 100–150 μm thick layer of zirconia stabilized with 7 wt. % yttria. Such coatings reduce the bond coat surface temperature by 100–200°C. Other ceramic TBCs containing additional rare earth dopants are also being developed with even lower thermal conductivity.

The temperature drop across a ceramic thermal barrier coating is governed by both the coating’s thickness and its thermal resistance. The latter can be increased by using materials with low intrinsic thermal conductivity and by incorporating porosity within the coating to further impede thermal transport. Porosity is also used to reduce the elastic strain energy that develops in the coating system from differences in thermal expansion of the airfoil and the materials used to make the TBC system. Pores that fully penetrate the coating thickness are especially helpful for reducing strain energy during thermal cycling. Controlling the thickness, pore volume fraction, and pore morphology of a coating is therefore essential to fully realize the potential of this airfoil protection strategy.

Thermal barrier coatings are often deposited on gas turbine airfoils using an electron beam-physical vapor deposition (EB-PVD) process. In the traditional EB-PVD method, an intense electron beam is used to melt and evaporate a ceramic source material. The evaporated molecules travel to, and condense on the airfoil, in a high vacuum in which few gas phase collisions occur. The molecules therefore travel in essentially a straight line from the source to the airfoil, and only regions on the airfoil that are visible from the source are coated. Substrate rotation is required to coat the nonplanar airfoils (blades and vanes) of interest here. Some complex shaped turbine components, such as doublet

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guide vanes, have regions on their gas flow path surfaces that are never in the line-of-sight of the vapor source regardless of the manner of substrate rotation. These components are therefore impossible to uniformly coat using the EB-PVD approach. Several groups are exploring alternative methods of coating deposition on complex substrates including plasma spray and sputtering processes.

Here we investigate fundamental aspects of a modified EB-PVD process that uses an inert gas (typically He or Ar) jet to entrain electron beam evaporated molecules and redirect them toward a substrate. The vapor molecules in this EB-directed vapor deposition (EB-DVD) process undergo many collisions before reaching the substrate, and deposition occurs by scattering from the jet flow streamlines. As a result, deposition onto any surface over which a stream flows (including non-line-of-sight regions) is potentially viable, and has been experimentally demonstrated by uniformly coating a nonrotated cylindrical substrate oriented transversely to the gas jet. To ensure source material evaporation by the electron beam in a low vacuum environment (where a portion of beam energy is dissipated by electronically exciting and partially ionizing the carrier gas), a high voltage (70 kV) electron beam gun is used, as the scattering cross section of electrons with background gasses decreases with increasing electron accelerating voltage.

The trajectories of the molecules in a rarefied vapor deposition process can be numerically simulated using direct simulation Monte Carlo (DSMC) techniques. DSMC is a statistical simulation method developed to analyze rarefied gas environments for which continuum computational fluid dynamics methods are difficult to apply. It has been widely used to study conventional physical and chemical vapor deposition processes and has been employed to better understand the EB-DVD coating process. DSMC uses a subset of test particles within a grid to simulate the behavior of a much larger number of real gas particles (on the order of 1 test atom per billion real atoms). The simulation atoms interact with each other through binary collisions. Between collisions they can interact with electromagnetic fields and can be deposited by collisions with surfaces.

Here, the DSMC technique is used to investigate some of the factors controlling coating thickness uniformity over the surface of a model, nonrotated airfoil undergoing DVD coating. Some regions on the airfoil we investigate are never in line-of-sight of the vapor source. We explore the effects of process conditions including the deposition chamber pressure [which controls the intercollision mean free path (MFP)] and the pressure ratio upstream and downstream of the gas jet forming nozzle, which governs jet speed. The validity of the simulation approach is assessed by comparison of the predicted and experimentally deposited coating thickness distributions on the model airfoil. The predicted coating thickness distributions for the EB-DVD and EB-PVD processes are also compared, and the factors controlling coating uniformity identified. The experimental coatings contained through thickness, intergrowth-column pores whose orientation to the airfoil surface varied along the airfoil. The columnar growth angles are compared with results obtained from the Tangent Rule, which describes the correlation between vapor atom incident angle and resulting columnar growth angle. The rule was found to be an unreliable prediction tool for DVD as the incident flux strikes the surface with a broad distribution of angles, instead of the sharply defined single value assumed in the Tangent Rule. However, insight into the columnar growth process can be gained through computed incident angle distributions (IADs) of the vapor atom flux along the airfoil surface.

II. SIMULATION METHODOLOGY

In the EB-DVD process, a gas jet is established by allowing gas from a high-pressure reservoir to flow through a nozzle into a chamber maintained at a lower pressure. The nozzle is arranged so that it surrounds a water-cooled crucible containing an evaporation source, Fig. 1. Evaporation occurs by impingement of a high power electron beam on to the surface of the source material. By arranging for evaporation to occur in the throat of a nozzle, the gas expansion process entrains the evaporant in a trans-sonic jet that travels toward the airfoil substrate.

A model turbine blade airfoil was designed for both numerical simulations and experimental depositions in the EB-DVD process environment, Fig. 2. It was attached to a thin back-plate to facilitate mounting in the deposition chamber during experimental depositions. The airfoil’s shape varied in only two dimensions (it had no twist) to simplify the simulations. The airfoil’s convex and concave surfaces were defined by quadratic functions of the y-coordinate to simplify simulation grid design and test piece manufacture. The airfoils were oriented with the y-axis aligned with the central axis of the electron beam. The deposition chamber was pumped to a pressure of 10⁻³ mbar to facilitate heat transfer to the water-cooled crucible.

![Fig. 1. (Color online) Schematic illustration of a directed vapor deposition process showing the electron beam evaporated vapor plume and model airfoil sample geometry. A mixture of 90% He and 10% O₂ was used for experimental studies.](image)
axes of both the jet flow and evaporation source. The y-coordinate origin was placed closest to the source.

The DSMC method is well suited for analysis of the DVD process because of the rarefied gas conditions in the deposition chamber. This, together with the presence of large pressure variations, makes it very difficult to apply conventional computational fluid dynamics. The DSMC program, Icarus, developed by T. J. Bartel (Sandia National Lab) was used for the simulations. It is a two-dimensional code capable of simulations on either axisymmetric or Cartesian coordinate meshes. Icarus uses the variable hard sphere approximation to model particle–particle interactions.

The DSMC simulations were performed using a gas jet mixture consisting of 90 at. % helium and 10 at. % oxygen (identical to that used in subsequent experiments). Reflections from solid surfaces were modeled as fully diffuse. Elemental Zr atoms were used as the simulated vapor species as parameters for the rare earth metals and the metal oxide molecules also present in the experimental vapor plumes were not available. The Zr parameters were deduced from experimental measurements on vapors of alkali group metals using a procedure proposed by Fan. A recent evaluation of the approximation procedure for PVD simulations found that it uniformly underestimated the mass flux at the substrate, but closely matched experimental results when normalized with the maximum incident flux.

The DSMC calculations proceeded in time steps of $10^{-7}$ seconds each of which included a free propagation and a collision step. The mesh developed to simulate the EB-DVD process is shown in Fig. 3. The cell size in a DSMC mesh must be smaller than the local MFP between collisions to properly resolve flow gradients. The simulated zone grid was optimized by performing numerous trials until converged results were attained at minimized computational cost. The grid used here consisted of 28 regions with differing grid size to account for variations in the MFP. The grid spacing was optimized to reduce computational time following the suggestions of Kannenberg and Boyd, and a species-weighting scheme was used to enhance the number of zirconium atoms in critical regions. Due to the 2D planar simulation geometry, the circular annulus nozzle used in the experimental chamber must be modeled as a linear double slit. To verify the accuracy of the simulation geometry, cylindrical axisymmetric and planar XY simulations with a simple flat disk substrate geometry were performed and showed qualitatively similar flow fields.

Simulation flow parameters were calculated to closely model the conditions used in the experiments described later. The chamber pressure was fixed at the external grid boundaries. The upstream/downstream pressure ratio was set by the...
inlet carrier gas flow rate, which was adjusted through trial and error until the desired value was reached. The vapor flux emitted by the source was held constant for all the simulations at $8.8 \times 10^{20}$ atoms/m$^2$s. The simulations were executed for 325 000 time iterations to reduce statistical scatter. Between $1 \times 10^6$ and $10 \times 10^6$ particles of He, O$_2$, and Zr were simulated to ensure adequate populations of trace deposition atoms. The simulations were performed on a 16-core Linux cluster with Intel E5000 series processors and ran for approximately 40 h each.

III. SIMULATION RESULTS

A. Flow fields

If the nozzle design, upstream gas temperature and composition, and pressure within the deposition chamber are held constant, the flow field is controlled only by the ratio of pressures before and after the expansion nozzle.$^{45}$ The relationship between the gas velocity, measured at test locations 15 mm up- and downstream of the substrate (see Fig. 3), is shown for four nozzle pressure ratios in Table I. The velocities increase with pressure ratio and are much higher upstream of the substrate, indicating a significant flow resistance created by the substrate. The Zr vapor atom concentration in the gas jet is also influenced by the pressure ratio. The steady state vapor concentration distributions for three pressure ratios are shown in Fig. 4. The overall vapor concentration decreases with increasing pressure ratio due to the reduced residence time in the jet as velocity increases. Increasing pressure ratio also leads to a narrowing of the gas jet. This results from the higher axial jet velocity and vapor atom momentum, which reduces the transverse scattering effects of collisions with background gas atoms. A large vapor depleted (dark blue) region surrounds the substrate. This depleted region is a result of vapor atoms scattering from the gas jet onto the substrate surface where they are allowed to stick and are then removed from the simulation. As the pressure ratio was increased, the depleted region behind the substrate dramatically increased in area due to the higher jet momentum. However, at low jet speeds, transverse diffusion of the vapor had begun to significantly reduce the nonuniformity in evaporant concentration.

When the nozzle pressure ratio was held constant, the flow field was dependent only upon the deposition chamber

![Zr Concentration (atoms/m$^2$)]

Fig. 4. (Color online) Contour plots of Zr vapor atom concentration for a chamber pressure of 16 Pa and three nozzle pressure ratios: (a) 2.0, (b) 4.5, and (c) 7.5.
Increasing the chamber pressure reduced the jet velocities both before and after the substrate, as indicated in Table II. The velocity downstream of the substrate was again always much lower than that found upstream; especially at high chamber pressures. The effects of changing the chamber pressure on vapor concentration within a jet plume are shown in Fig. 5. As the pressure increased, the vapor in these “slow” jets became much more concentrated on the centerline of the carrier gas jet, and the vapor depleted region behind the substrate decreased in area due to the decrease in the axial velocity component of the jet. The slow jet’s increased vapor atom concentration was a consequence of the increased residence time of vapor atoms in the jet plume, while the increased chamber pressure (smaller mean free path) confined the vapor to the center of the jet by reducing the rate of lateral diffusion.

### Table I. He jet velocities upstream and downstream of the substrate with various pressure ratios. The velocity at both locations increases with increased pressure ratios.

<table>
<thead>
<tr>
<th>Pressure ratio</th>
<th>Velocity upstream of substrate (m/s)</th>
<th>Velocity downstream of substrate (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0</td>
<td>288</td>
<td>88</td>
</tr>
<tr>
<td>3.0</td>
<td>363</td>
<td>104</td>
</tr>
<tr>
<td>4.5</td>
<td>452</td>
<td>125</td>
</tr>
<tr>
<td>7.0</td>
<td>553</td>
<td>158</td>
</tr>
</tbody>
</table>

![Contour plots of Zr concentration](image)

**Fig. 5.** (Color online) Contour plots of Zr concentration at a nozzle pressure ratio of 2.0 and chamber pressures of (a) 16 Pa, (b) 30 Pa, and (c) 45 Pa.

#### B. Atom trajectories

To gain better insight into the scattering processes responsible for vapor depletion near the substrate, we calculated the He jet streamlines in the vicinity of the substrate by determining the average trajectories along the simulation grid. Those for a representative deposition configuration (a chamber pressure of 16 Pa and a pressure ratio of 2) are shown in Fig. 6 overlaying the local pressure distribution. The streamlines (the time averaged helium atom trajectories) generally curve toward regions of lower pressure and away from those of higher pressure. There are regions of high pressure at the airfoil’s leading edge and along its concave surface. Along the concave side, the streamlines are initially deflected toward the surface, and later bend away from the substrate. Along the convex surface, the local pressure was reduced and the collisional mean free path increased. As a result, the vapor was confined to the center of the jet, leading to increased vapor atom concentration.
result, fewer scattering collisions occur, and the He atoms travel a straighter path.

The vapor atom streamlines at the above conditions were also determined and are shown, along with the trajectories of three hypothetical Zr atom classes that impact the airfoil, in Fig. 7. Each solid circle represents a collision between a vapor atom and an atom in the jet flow. The dashed lines indicate the vapor atom’s trajectory between collisions. The mean free path between collisions for this example was 3–4 mm near the substrate, but varies with pressure within the deposition chamber. The three paths shown in Fig. 7 exemplify three classes of condensation. The center path results in deposition onto the leading edge of the airfoil at near normal incidence to the surface and with little deviation from the original direction of the jet. The atoms in the outer paths are knocked randomly left and right by collisions. In the gas jet far from the substrate, there are an equal number of scattering collisions to the left and right. However, near the substrate surface, a collision can scatter a vapor atom toward, and onto the substrate. This destroys the directional balance that exists far from the substrate, and results in more vapor atoms traveling toward the substrate than away from it. As a result, the vapor streamlines bend toward the substrate as shown in the figure. In both cases, atoms are deposited from streamlines onto surface regions that are not in the line-of-sight of the vapor source and enable the possibility of non-line-of-sight (NLS) deposition.

To investigate the effect of pressure ratio variation (at a fixed chamber pressure of 16 Pa) upon the deposition process, vapor atom streamlines for pressure ratios of 2, 4.5, and 7 are shown in Fig. 8. We define the capture width as the cross-section of the vapor jet that is eventually deposited on the substrate surface. The capture width is defined 13.5 mm upstream of the substrate’s leading edge, before the jet is significantly influenced by the substrate. The capture width is asymmetric around the centerline of the airfoil with the majority of the included streamlines terminating on the convex substrate surface. A large capture width increases the vapor flux incident upon the substrate surface and the deposition efficiency (the ratio of deposited atoms to atoms emitted by the source). The simulations show that vapor atom

### Table II. He jet velocities upstream and downstream of the substrate at various chamber pressures. The velocity decreases with increasing chamber pressure.

<table>
<thead>
<tr>
<th>Chamber pressure (Pa)</th>
<th>Velocity upstream of substrate (m/s)</th>
<th>Velocity downstream of substrate (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.5</td>
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<td>166</td>
</tr>
<tr>
<td>16</td>
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<td>88</td>
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<td>50</td>
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<tr>
<td>45</td>
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</tr>
</tbody>
</table>

![Fig. 6. (Color online) He streamlines and pressure contours near the substrate at a chamber pressure of 16 Pa and nozzle pressure ratio of 2.0.](image)

![Fig. 7. Schematic illustration demonstrating the paths of vapor atoms that deposit on the concave surface, leading edge, and convex surface. The random walk path of a hypothetical individual atom is overlaid with the average trajectory of vapor atoms in the region.](image)
streamlines nearest to the substrate’s centerline result in deposition at the front of the airfoil, while atoms in streamlines further from the centerline are more likely to deposit near the tail. The capture width in Fig. 8 decreases with increased pressure ratio due to the greater momentum of the vapor atoms. Fast moving vapor atoms are deflected less toward the substrate by scattering collisions than slower ones found at lower pressure ratios, resulting in fewer streamlines terminating on the substrate surface.

The effect of changing the deposition chamber pressure (at a fixed pressure ratio of 2.0) on the vapor atom trajectories is shown in Fig. 9. As the chamber pressure was increased from 7.5 to 45 Pa, the capture width decreased from 12.5 to 5.8 mm. At high pressures (i.e., shorter mean free path and lower jet velocity), the majority of the substrate intersecting streamlines end near the leading edge. Streamlines outside of this region are curved away from the substrate by the high pressure regions surrounding it. Although the carrier jet is moving slowly, the streamlines must make a large divergence from the jet axis. At lower chamber pressures, the mean free path is greater, and the capture distance is larger with streamlines intersecting the substrate uniformly. The reduced rate of scattering collisions results in vapor streamlines with more gradual curvature. The streamlines that do not terminate at the substrate are much less perturbed than those at higher pressures indicating a reduced influence of the substrate on the carrier gas jet.

It is interesting to note that while a larger capture width is correlated with a larger fraction (width) of the vapor plume impacting the substrate surface; it does not correlate with the largest vapor flux impacting the airfoil surface. This disparity arises because of the dependence of vapor concentration in the jet with flow conditions as discussed in Sec. III A. The condition with the largest capture widths in Figs. 8 and 9 is

![Figure 8](image1.png)  
**Fig. 8.** Zr vapor atom streamlines for at a fixed chamber pressure and pressure ratios of (a) 2.0, (b) 4.5, and (c) 7.0. The vapor atom capture distance is noted on each flow field. The capture distance decreases with increasing pressure ratio (jet speed), resulting in a reduction of deposited flux onto the substrate surface.

![Figure 9](image2.png)  
**Fig. 9.** Zr vapor atom streamlines at a fixed pressure ratio of 2.0 and chamber pressures of (a) 7.5 Pa, (b) 30 Pa, and (c) 45 Pa. The capture distance decreases with increasing chamber pressure.
Thus, although it captures a larger volume fraction of the vapor, the lower vapor concentration results in a smaller total flux. The consequences of this will be discussed below.

C. Coating thickness

By assuming a sticking factor of unity, the local flux deposited on the substrate surface can be determined from the DSMC simulations. If the deposited coating’s density does not change with position on the airfoil surface, this flux is then proportional to coating thickness. To deduce the vapor flux profile on the airfoil, the concave and convex sides of the simulation substrate were each divided into 40 surface elements that corresponded to the surrounding DSMC simulation grid, and the flux incident to each region was then calculated. Incident flux profiles for the concave and convex surfaces of the airfoil are shown for several pressure ratios (at a fixed chamber pressure of 16 Pa) in Fig. 10 [note the different ordinate scales in (a) and (b)]. Along the convex surface [Fig. 10(a)], the flux near the leading edge of the airfoil decreases with increasing pressure ratio. This decrease indicates that larger areas of the substrate become shadowed from the vapor in the jet due to increased vapor atom velocity. This is also observable in Fig. 8 where depositing streamlines at high pressure ratios gradually deflect toward the substrate. The flux profiles along the convex surface [Fig. 10(b)] show a much reduced dependence on the pressure ratio. For all ratios, the flux maximum is located at the leading edge of the airfoil. This results from the region near the leading edge being in the line-of-sight of the vapor source. The largest flux is recorded at the highest pressure ratio of 7.0. This arises because the vapor is highly collimated and does not need to undergo scattering collisions to be deposited on the leading edge.

The flux profiles also vary with the chamber pressure. The profiles for several chamber pressures (at a fixed pressure ratio of 2.0) are shown in Fig. 11. At the lowest chamber pressure, the concave surface flux, Fig. 11(a), is lowest at the leading edge and gradually increases toward the tail. At this chamber pressure, the mean free path is large (5 mm (7.5 Pa and pressure ratio of 2) has the lowest vapor density. Thus, although it captures a larger volume fraction of the vapor, the lower vapor concentration results in a smaller total flux. The consequences of this will be discussed below.
at 7.5 Pa), and the vapor atoms travel a significant distance along the substrate before scattering onto the surface. The vapor atom concentration in the jet is only slowly depleted due to the infrequent scattering, and additional deposition occurs due to the reduced diffusion distance needed to impact the protruding tail. As the chamber pressure rises, the location of highest flux on the concave side moves from the tail to the tip of the airfoil. The most uniform concave surface deposition (at a pressure ratio of 2) occurs at an intermediate pressure of 16 Pa. Along the convex surface [Fig. 11(a)], the flux profiles show little dependence upon the chamber pressure as in Fig. 10(b).

Examination of Figs. 10 and 11 reveals that the best coating uniformity occurs at moderate chamber pressures and pressure ratios. These slow flow conditions provide an optimum balance between mean free path and vapor velocity. Higher chamber pressures result in shorter mean free paths, and atoms experience more collisions, which direct them toward the substrate. However, the atoms also move with lower momentum, which allows scattering collisions to rapidly direct the atoms to the substrate. Lower pressures result in longer mean free paths, and atoms can travel greater distances without depositing onto the substrate. Changing the pressure ratio influenced the flux profile by controlling the momentum of the incident vapor atoms. At high pressure ratios, the vapor atoms had large momentums, which increased the distance atoms traveled before depositing onto the substrate. A very small pressure ratio (<1.5) resulted in atoms depositing quickly, near the leading edge of the substrate.

IV. EXPERIMENTAL COMPARISONS

To evaluate the validity of the coating flux and thickness predictions above, thermal barrier coatings were experimentally deposited upon identical airfoil substrates to those used in the simulations. The vapor source was a 1.25 cm diameter zirconia rod triply doped with yttria, gadolinia, and samaria obtained from TCI ceramics, Inc. (Bethlehem, PA). The surface of the source rod was evaporated using a 70 kV/2.45 kW electron gun in the EB-DVD system schematically illustrated in Fig. 1. Model airfoil-shaped substrates were milled from 303 grade stainless steel plates, grit blasted, and cleaned prior to deposition of the ceramic coating. No bond coat was used since we were not evaluating the durability of the coating. The substrates were first heated to 500°C at 16 Pa for 30 min to clean the surface and then heated to 1000°C during deposition. During deposition, the substrate was held at a fixed orientation (no rotation) with the airfoil leading edge nearest to the source as shown in Fig. 1. Coatings were deposited at a chamber pressure of 16 Pa using an upstream/downstream nozzle pressure ratio of 3.5. The carrier gas flow rate was set to 9.0 slm of helium and 1.0 slm of oxygen. After deposition, the samples were sectioned and the thickness and microstructure of the coating examined at various positions using a scanning electron microscope.

A. Experimental results

The experimentally measured coating thickness (normalized by the maximum value on each side) for the convex and concave airfoil surfaces is compared with simulation results in Fig. 12. SEM images of the coating at several locations around the substrate are shown in Fig. 13 for an experimental deposition performed at 16 Pa and a pressure ratio of 3.5. The coating was thickest on the airfoil’s leading edge, while the thinnest region was near the trailing edge. The experimental and simulated thickness profiles are very similar on both surfaces. On the convex surface, the trends were quite similar to those found in Figs. 10(b) and 11(b). Along the concave surface, the experimental profile was sensitive to the precise point on the leading edge where the first thickness measurement was recorded. Near the front of the airfoil, in the region that is within the line-of-sight to the source, the coating is thick. However, this thickness quickly decreases with increasing distance along the airfoil. The non-line-of-sight region that occupies most of the airfoils concave surface has a fairly uniform thickness coating that slowly increases down the length of the substrate. The profile demonstrates the vapor flux variation between line-of-sight and non-line-of-sight deposition.
The microstructure of the coating was also found to vary along the substrate (Fig. 13). At the leading edge, the columns are oriented with angles approximately equal to the local substrate surface normal. Along the convex and concave surfaces, the growth angles tilt away from the surface normal and toward the incident gas jet. This column orientation variation is usually thought to be governed by the incidence angle of the vapor with respect to the local surface normal.46,47

B. Incidence angle distribution

The IAD for a simulation corresponding to the experiment were determined from the DSMC results and were found to vary greatly along the airfoil, as shown in Fig. 14, which shows results at the same locations as Fig. 13. On the leading edge of the airfoil (not shown), the IAD was narrowly distributed around normal incidence, indicating that the vapor atoms were impacting the surface with minimal scattering from the main plume. In the less shadowed regions, [Figs. 14(a), 14(b) and 14(d)] the IADs were also narrow, but now skewed away from normal incidence. The broadest IADs [Figs. 14(c), 14(e) and 14(f)] were found in shadowed regions along the convex and concave sides. The broad IADs in these regions result from the scattering collisions the vapor must experience to deposit in these regions.

The IADs are also modified by the chamber pressure and pressure ratio. The effects of these variations are shown in Fig. 15 using the IAD experienced by the surface on the center of the concave side (point b in Figs. 13 and 14). Increasing the chamber pressure causes a broadening of the IAD in shadowed regions (due to increased scattering). The peak of the IAD moves away from the surface normal with decreasing chamber pressure. An extreme case is shown for a simulation using conventional PVD conditions (pressure of 0.02 Pa) in Fig. 15(b). The PVD angle of incidence is sharply peaked at an angle far from the local surface normal.

C. PVD versus DVD

All of the configurations of the DVD environment provided greatly improved coating uniformity compared to PVD. To demonstrate the improvement, simulations were conducted using an identical simulation grid, but with PVD-like deposition conditions. The chamber pressure was held at 0.02 Pa, while the carrier gas jet was turned off. The vapor evaporation rate was set identical to the DVD simulations. Figure 16 shows the flux along the concave surface for PVD and DVD simulations where the DVD simulation was

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Fig. 13. SEM images of the coating at various locations on the airfoil substrate with the average columnar growth angle indicated. Note the differences in magnification between images.
performed with a 16 Pa chamber pressure and a 3.0 pressure ratio. The flux impacting the airfoil surface was generally one to two orders of magnitude higher under DVD conditions than PVD. The mean free path under PVD conditions was approximately 0.5 m. This distance is greater than the length of the simulation domain (0.4 m), and thus the average vapor particle experienced no collisions during its flight. The lack of scattering is reflected on the plot of IADs at different chamber pressures in Fig. 15. The PVD conditions result in the narrowest distribution with a maximum far from the local substrate normal. The peak in the angular distribution is very close to the angle the substrate surface makes to the incident vapor rays.

V. DISCUSSION

Coating uniformity is determined by the binary scattering conditions in chamber regions near the substrate surface. The uniformity is controlled through both the rate of scattering collisions near the substrate and the momentum imparted from the collisions. In the substrate-adjacent regions, an incident vapor atom has the possibility of undergoing collisions with three classes of carrier gas atoms: (1) Type I: The vapor atom collides with a carrier gas atom from the main jet column. These collisions serve to knock the atom further away from the source and in a random direction perpendicular to the gas jet axis. (2) Type II: The vapor atom collides with a carrier gas atom that has struck and randomly scattered from the substrate surface. These collisions decrease the likelihood that a vapor atom will eventually reach the substrate surface by imparting momentum away from the substrate. These types of collisions are prevalent near the leading edge and along the convex surface of the airfoil (Fig. 6). Many carrier gas atoms strike this surface region and reflect with high velocities. These reflected atoms collide with the incident vapor atoms and deflect their trajectories away from the substrate surface. (3) Type III: The atom collides with a background atom of the carrier gas.
These background atoms have random motions, but their collisions will generally push the vapor atom toward the center of the carrier gas plume. Background gas atoms must travel through the midpoint of the vapor plume’s cross-section before they will scatter vapor atoms away from the substrate. Type I and III collisions can be considered as enabling deposition near the collision location, while type II collisions knock the vapor atoms away from the local substrate surface. The influence of type II collisions can be seen in the streamlines at different chamber pressures shown in Fig. 9. As the chamber pressure increases, the number of atoms scattering from the substrate surface also grows, which increases the number of type II collisions and causes the vapor atom streamlines to deflect farther from the substrate surface. As a result, vapor atoms either impact near the leading edge, or are likely to travel past the substrate.

The three classes of collisions can be manipulated with the chamber pressure and the pressure ratio. Modifying the chamber pressure does little to change the momentum imparted during the collisions, but it does adjust the frequency of collisions. Modifying the pressure ratio does not change the rate of collisions, but alters the momentum of the carrier gas jet. The higher jet momentum causes the atoms to travel further in the axial direction between collisions, and results in the vapor atoms experiencing fewer collisions as they travel past the substrate.

The results above show the importance of two fundamental quantities that determine coating uniformity: the vapor atom concentration and mean free path. Vapor atom concentration is of great importance for deposition onto macroscopic, non-line-of-sight substrates. Although the carrier gas may flow along the entire surface of a substrate, depletion of vapor atoms can prevent deposition onto a non-line-of-sight region. This vapor depletion can be controlled via the mean-free path near the substrate. With larger mean free paths, scattering collisions are less frequent, allowing the vapor atoms to travel further in a straight line. A large mean free path will allow deep penetration into a non-line-of-sight region, but may prevent the necessary collisions for eventual deposition onto the surface. Thus, ideal deposition conditions are found through balancing these two parameters, which occurs at moderately low pressure ratios (2–3) and moderate chamber pressures (~16 Pa) where the jet flows are slow.

The paths of three individual vapor atoms along with the average streamline trajectories in their vicinity are shown in Fig. 7. The mean free path distance that atoms travel between collisions varies along the streamline path. Each collision with the carrier gas knocks the vapor atom along the streamline, but also introduces random movement.
perpendicular to the carrier gas axis. Far away from the substrate the mean free path is large (several millimeters) but generally becomes shorter closer to the substrate due to the increasing local pressure. The mean free path remains comparably large compared to the substrate dimensions, which maintains the importance of the random motion imparted by collisions with the background gas. The random collisions cause incident atoms to impact the substrate at random angles and results in a broad IAD. The schematic in Fig. 7 also shows how a broad IAD is created through scattering collisions. The random direction of motion allowed between collisions enabled individual atom trajectories to deviate substantially from the average ones represented by the streamlines.

Further insight into the coating process can be obtained by comparing the measured angles of the growth columns with the Tangent Rule using the simulated IADs. Nieuwenhuizen and Haanstra first proposed the Tangent Rule for predicting the inclination angle of growth columns deposited from a monoangle incident flux in 1966. By measuring the growth angle of the deposited columns, one can determine the angle of incidence through the relationship

$$2\tan(\phi) = \tan(\theta),$$

where $\theta$ is the flux angle, and $\phi$ is the columnar growth angle (both measured from the local surface normal). To test its validity with non-line-of-sight deposition, the rule was evaluated at locations around the substrate to determine its applicability in non-line-of-sight regions. Table III shows the results of applying the tangent rule at various points on the substrate surface. The rule is accurate at locations in line-of-sight regions, but performs poorly in non-line-of-sight regions. This is not unexpected, as the Tangent Rule was not designed to evaluate coatings deposited from vapor fluxes with a wide IAD. Clearly, alternative methods are necessary to predict columnar growth characteristics in regions where condensation occurs with a broad IAD.

### VI. SUMMARY AND CONCLUSIONS

Coatings have been deposited onto a stationary airfoil substrate using a gas-jet assisted DVD technique. Complete coverage of the substrate was achieved, which is impossible using traditional PVD. DSMC simulations were found to accurately predict coating thickness trends with both substrate surface location and deposition conditions. The thickness uniformity and coating porosity were found to be sensitive to the deposition chamber pressure and gas jet velocity, both of which manipulate the mean free path of the vapor atoms. The optimum conditions for creating uniform thickness distributions have been identified.

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**Table III. Comparison of columnar growth angles predicted by the Tangent rule, and measured from experiments.**

<table>
<thead>
<tr>
<th>Measurement region</th>
<th>Peak vapor atom incident angle ($\theta$) (deg)</th>
<th>Predicted columnar growth angle ($\phi$) (deg)</th>
<th>Measured columnar growth angle ($\phi_m$) (deg)</th>
<th>Percent difference with simulation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Leading edge</td>
<td>3</td>
<td>1.5</td>
<td>2</td>
<td>7</td>
</tr>
<tr>
<td>Center of convex surface</td>
<td>–64</td>
<td>–45.7</td>
<td>–22</td>
<td>70</td>
</tr>
<tr>
<td>Center of concave surface</td>
<td>28</td>
<td>15</td>
<td>21</td>
<td>33</td>
</tr>
</tbody>
</table>

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