

Modeling a Discontinuous CVD Coating Process: II. Detailed Simulation Results

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Abstract: The atmospheric chemical vapor deposition process on continuous glass sheets is a well developed one and the parameters that affect it are relatively well understood. When this process is converted to coat discrete glass plates it introduces a new variable, the gap between the glass plates, which can significantly impact the quality of the coatings. In this study a 2D pseudo steady state model of the process was developed to study the effect of the gap, and the ratio of outlet to inlet gas flow rates (called the bias), on the coating quality. The model was solved with the commercially available computational fluid dynamics program FIDAP which employs a finite element scheme. An earlier study had shown the validity of the pseudo steady state model and the use of FIDAP for this problem [Lawrence, J.G.; Nadarajah, A. (2007): Fluid Dynamics & Materials Processing, vol.3,no.3, pp.247-254]. The simulations showed that the gas flows were always well ordered. The value of the bias and the size of the gap were found to have a significant effect on the coating rate, but had only a small effect on coating uniformity. Lower biases and gaps produced the highest coating rates and coating uniformities, but the overall coating rate will still be lower than that for the continuous glass ribbons. The results suggest that this CVD process can be adapted for discrete glass plates to produce close to uniform coatings of required thicknesses if the plate motion can be suitably adjusted.

Keyword: Glass plates, Simulation, CVD, Gap, Uniformity.

1 Introduction

CVD coatings on sheets of glass are commercially achieved by using an online CVD coater. Coatings of particular interest to the glass industry include coatings of silicon, titanium nitride, and the oxides of silicon, aluminum, tin, zinc and transition metals, which can add very useful electrical and optical properties to the glass [Gordon (1997)]. Use of atmospheric pressure chemical vapor deposition (APCVD) process is now widespread [Sheel and Pemble (2002), O'Neill, Parkin, Clark, Mills and Elliott (2003)]. APCVD processes have very high deposition rates and are insensitive to small variations in temperature. Our interest here is in the APCVD glass coating process for consumer appliances, such as oven doors. The essential functions of CVD equipment are to create an appropriate vapor or gas mixture and to make it flow over a glass surface, to achieve the coating [Gordon (1997)]. To facilitate uniform coating and a continuous coating process, the online CVD coaters have been developed. One such online CVD coater was jointly developed by Gordon (1997) and McCurdy (1999) and was first implemented at the Libbey Owens Ford (LOF) Corporation. Later, Pilkington LOF designed an online coater for silicon deposition from gaseous silane [McCurdy (1999)]. The process is discussed in detail by Zhu (2000). In this type of coater, the mass flow rate is used to control the flow of gases in the upstream and downstream sides. We refer the ratio of gas flow rate at the outlet vent to the inlet gas flow rate as the bias.

The continuous coating process is a well developed one and integrated with float glass production lines producing continuous glass ribbons. Continuous sheets of glass are coated by moving the glass along the length of the coater dur-

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ing which time the reactant gases deposit the coating on the surface. The reactant gases flow slowly through an inlet funnel into the reactor, travel horizontally above the moving glass and exit through two exhaust vents, one upstream and other downstream. This provides more complete isolation of the bi-directional laminar reactant gas flow from the ambient atmosphere. The continuous sheet of glass moves horizontally on rollers into the reaction chamber. This process produces glass coatings of exceptional uniformity.

Currently, this process is being adapted for coating previously manufactured pieces of cut glass run on belt or roller furnaces in an off-line coating process. It is this off-line process that is of interest in this study. In continuous CVD process the uniformity and thickness of the coating are dependent on various parameters such as coater alignment, coater height, carrier gas concentrations, chemistry of carrier gases, precursor chemistry and exhaust temperature. These parameters have been studied and modeled previously [Meyyappan (1995), Sheel and Pemble (2002)]. The gap distance is an additional parameter that must be accounted for when adapting this process to coat discrete glass plates for use in consumer appliances. The quality of the coating is determined by the velocity, concentration and temperature profiles in the reactor. By its effect on these parameters the gap can significantly affect the quality of the coating, but it is difficult to predict this without costly experiments. However, these effects can be readily studied in a mathematical model of the problem, but to date such analyses have not been carried out for this process.

In order to study these issues, the model developed has to be solved using a numerical technique. A finite difference method can be used for this and we developed one for determining the velocity profile in the simplified model of this problem [Lawrence and Nadarajah (2006)]. However, extending the model to include features such as temperature and concentration effects and complex geometries is a time consuming undertaking. Commercially available computational fluid dynamics (CFD) programs can readily be adapted for this task and this approach was taken here with

the program FIDAP [FIDAP (1998)].

However, before commercial CFD programs can be used they need to be tested for their applicability to the particular problem. FIDAP was used for a simplified version of the discontinuous coater problem and compared with a finite difference numerical solution that we had developed [Lawrence and Nadarajah (2006)]. Results for the velocity profile with the FIDAP program showed excellent agreement with that from the finite difference scheme. This validates its use in the more comprehensive simulations done here to examine the effect of discontinuity of the glass surface on the atmospheric CVD coating process.

2 Formulation of the Problem

The presence of the gap between the plates makes this a time dependent problem. In an earlier study a 2D pseudo steady state model of the problem was developed, based on the much faster gas flow rates compared with the plate motion [Lawrence and Nadarajah (2006)]. In this formulation a steady state simulation was carried out for a fixed location of the gap. The simulations from this model suggested that this approach was valid, with each simulation providing a snapshot of the process for the given location of the gap. This approach will be adopted here as well, along with the 2D formulation. This means that most of the model equations developed earlier will still be valid. However, the earlier formulation employed only a small section of the coater surrounding the gap. Such a restricted geometry will not allow for the verification of the uniformity of the deposition process through the entire coater.

The geometry of the coater used in this study is shown in Fig. 1. As can be seen the gas flow region of the reactor has a very small height to length ratio of 1:64. At this aspect ratio the introduction of the gap between the plates should have almost no effect upstream of it. This fact, coupled with negligible effect of the plate motion, means that the process upstream of the gap is almost perfectly symmetric about the axis of the inlet funnel. Based on this, half of the entire coater will be modeled here, with the downstream end chosen arbitrarily. The model geometry will extend from

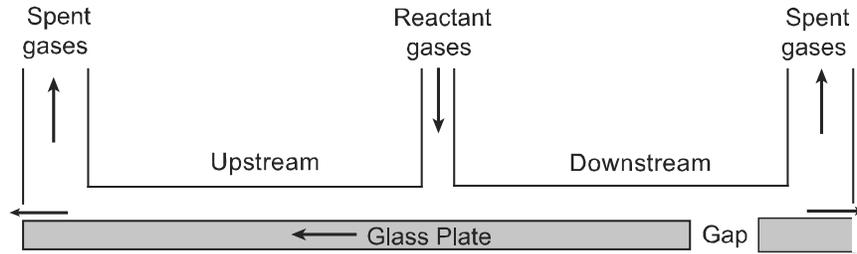


Figure 1: Reactor geometry used for model simulations.

half the inlet funnel to the entire down stream outlet. However, this symmetry assumption will fail when the gap is near the inlet. As a result, in order to examine the coating uniformity on an entire glass plate, some simulations were carried out for the complete coater.

The dimensions of the reactor shown in Fig. 1 are as follows [Zhu (2000)]. The reactor is 16 inches long, $1/4$ inch high and the conveyor system passing through it is 34 inches wide. The glass load consists of pieces of glass 13 inches long and 21 inches wide, with 1 inch gap between the glass plates. There is also a $5/32$ inch gap above the plates in the plate entrance and exit into the reactor. The inlet width is $3/8$ inch and the outlets are 1 inch wide.

The domain equations and boundary conditions [see Lawrence and Nadarajah (2006) for details], were incorporated into FIDAP and solutions to the problem were obtained. Unlike for the earlier simplified formulation, the energy and species mass conservation equations were solved as well to obtain the temperature and concentration profiles. The bias is defined as the ratio of volumetric gas flow rate at the outlet and inlet. If the outlet gas flow rate at the downstream and upstream vents are Q_d and Q_u respectively, for an inlet flow rate of Q_p , the bias is $Q_d/(Q_p/2)$ at the downstream vent and $Q_u/(Q_p/2)$ at the upstream vent. When the bias is less than 1 there is outflow of gases through the gap between the plates and the plate entrance and exit to the reactor, and when it is greater than 1 there is an intake of gases.

The physical properties used for the simulation are given in Table 1 of the earlier study [Lawrence and Nadarajah (2006)]. The input parameters for

Table 1: Input parameters for model simulations

Parameter	Value
Volumetric flow rate at the inlet Q_p	4.78 slpm
Glass plate velocity V	0.08 m/sec
Inlet gas temperature T_1	400°F
Gap gas uptake temperature	400°F
Glass plate temperature T_{ref}	1100°F
Number of nodes, course mesh	1305
Number of nodes, fine mesh	4809
Residual vector for convergence	10^{-5}

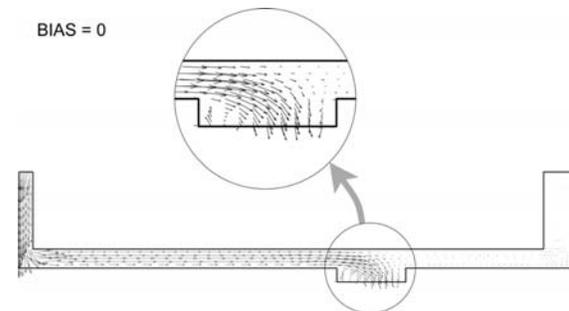


Figure 2: Velocity vector plot of the coating process with bias of zero and a 1 inch gap between the plates. For this bias all the inlet gases exit through the gap. The figure shows the velocities at the instant the gap is near the center. The flow profile near the gap is also shown in an enlarged view.

the simulations are given in Table 1. Most simulations were carried out twice, once with a courser mesh and then with a finer mesh and the invariance of the solution was checked.

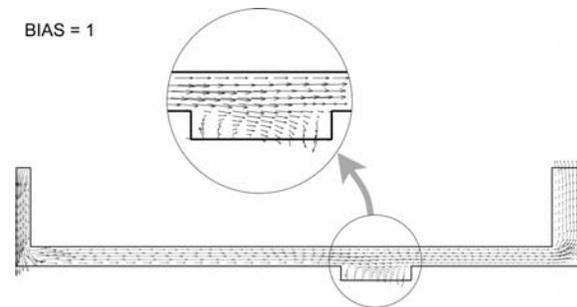


Figure 3: Velocity vector plot of the coating process with a bias of 1. For this bias all the inlet gases flows out through the outlet vent. The enlarged view shows a circulation pattern near the gap with significant intake of external gases.

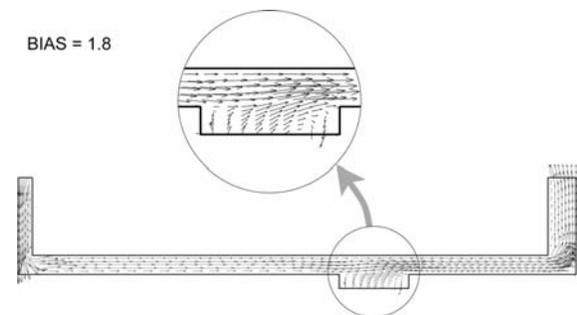


Figure 4: Velocity vector plot of the coating process with a bias of 1.8. For this bias the gas flow through the outlet vent is higher than that the inlet flow by 80%, with intake of external gases through the gap accounting for the difference.

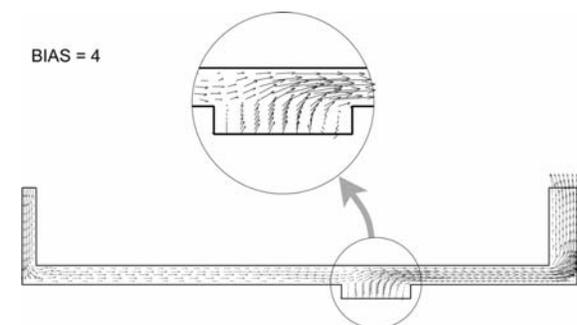


Figure 5: Velocity vector plot of the coating process with a bias of 4. For this bias the gas flow through the outlet vent is higher than that the inlet flow by 300%, with intake of external gases through the gap accounting for the difference.

3 Results and Discussion

Unlike for the continuous CVD process, new phenomena were expected to occur due to the presence of the gap between the plates in the discrete coating process. The first concern was that the existence of the gap could lead to turbulent flow patterns in the coater. The second was the effect of the flow pattern on the uniformity of the coatings. The gap introduces additional edges in the glass which may affect the uniformity. Thirdly, the effects of changing the bias and the gap size and position on the flow profile and coating uniformity need to be studied. The simulations that were carried out were mostly focused on these issues.

3.1 Effect of the Gap on the Velocity Profile

The velocity vector plots were obtained for the model using the symmetry assumption. Simulations were performed for different biases of 0, 1.0, 1.8 and 4.0. The velocity vector plots for the different biases are shown in Figs. 2 to 5. For all these cases it is clear that turbulent flow patterns do not occur. A low bias means that the inlet gases will escape through the gap between the plates. This is undesirable as it will result in the coating materials being deposited on the rollers moving the plates and on other components. An extreme example of this is the bias of zero shown Fig. 2. As can be seen in this case all the coating gases escape through the gap. This also means that the gases will not be available for coating the plate downstream of the gap.

The flow pattern for a bias of 1.0 is shown in Fig. 3. This represents the neutral case with no net outflow or inflow of the coating gases expected through the gap, i.e. the entire coating gases should exit through the outlet. As a result, the flow is well ordered through the entire coater. This should produce uniform coatings on the plates upstream and downstream of the gap. However, the magnified inset in Fig. 3 shows that there is significant loss of coating gases and significant intake of external inert gases into the coater at the gap. This is caused by a circulation pattern at the gap.

For biases higher than 1 there is a net inflow of external inert gases into the coater through the gap. The advantage of higher biases is that they can minimize the leakage of coating gases through the gap. However, this would be offset by the lower deposition rates in plates downstream of the gap due to the intake of inert external gases. The result for a bias of 1.8 is shown in Fig 4 and for this case as much as 44% of the gases in the coater near the outlet are external gases. Although the flow pattern is still ordered, similar to that in Fig. 3, the external gas intake is beginning to modify the flow profile downstream of the gap. This may also begin to affect the quality of the coating process beyond the gap. When the magnified inset is examined it suggests that there is still a small amount of coating gases leaking out through the gap and a diminished circulation pattern. While this leakage may be negligible it does suggest why high biases may be needed to ensure minimal leakage of coating gases through the gap.

Fig. 5 shows the extreme case of a bias of 4. The flow here is still an ordered one without any pockets of turbulence and will produce uniform films on the glass plates. While the flow is slightly non-uniform just downstream of the plate, it is uniform everywhere else. This should produce uniform coatings through most of the coater. However, the thickness of the coatings and the efficiency of the coating process will be low due to the large intake of exterior gases through the gap, which in turn will result in greatly diluting the coating gases downstream of the gap.

3.2 Edge Effects

Edge effects are variations in coater uniformity near the edges of the glass plates. To detect the presence of any significant edge effects, concentration contour plots for the simulations in Figs. 2-5 were analyzed and they are shown in Fig. 6. Of particular interest in these plots is the concentration near glass surface. Since the first order reaction

rate is proportional to this concentration, it is a direct measure of the coating rate or efficiency. Variations of this concentration, or its lack thereof, are also an indication of the uniformity of

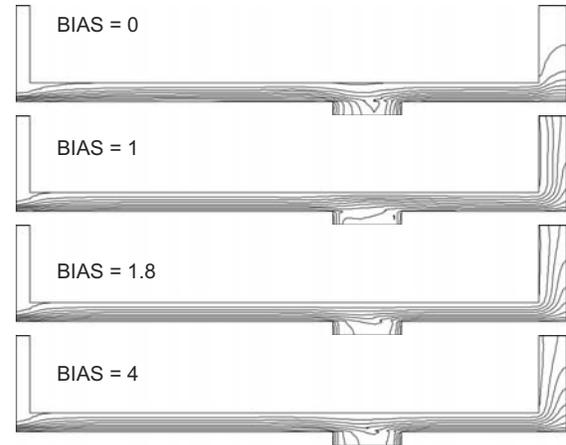


Figure 6: Concentration contour plots for gas flows with biases of 0, 1, 1.8 and 4 (from top to bottom), corresponding to the velocity plots shown in Figs. 2-5.

the coating.

In these plots there is little concentration variation across single glass plates, particularly when the biases are low to moderate. For these cases the contour lines near each plate are parallel to the plate. This uniformity is lost only near the inlet and outlet, but this is not due to the gap and is likely to occur even in the continuous coating process as will be discussed later. For the case of a bias of 1.8 and 4 some deviation from parallel contour lines occur near the gap, suggesting a mild edge effect. This suggests that the presence of the gap has only a small effect on the uniformity of the coating near the gap edges.

The effect of the relative position of the gap on edge effects was also studied. Concentration contour plots for the different positions of the gap are shown in Fig. 7. Once again the plots show that the contour lines are parallel to the coating surface for each glass plate, meaning that the concentrations are constant along the surface. Similar to the effect of the bias, the position of the gap does not seem to cause significant edge effects, particularly when the gap is near the outlets and inlet. The coatings are mostly uniform across a glass plate for most flow conditions for any position of the gap at any instant of time. While there was little variation across a single plate, there were signif-

icant variations between two adjacent plates depending on the bias as will be discussed later.

3.3 Position of the Gap and Effect of Bias

To study the effect of the bias on the position of the gap, the symmetry assumption may not be valid and so the entire reactor was simulated. This model will also verify the results obtained from simulations using the symmetry assumption. Previous simulations indicated that the bias of 1.0 could be considered as the neutral bias since the gas inflow or outflow through the gap was minimal. The gaps were positioned at three different locations in the downstream section of the reactor and a bias of 1.0 was applied at both the outlets. The gaps were positioned near the inlet, at the center of the downstream section and near the outlet of the downstream section. The effect on uniformity in coatings due to the presence of large gaps is shown in Fig. 8 and the presence of narrow gaps is shown in Fig. 9. The narrow gaps were one-fifth in size of the large 1 inch gap.

The concentration contour plots in Figs. 8 and 9 validate the symmetry assumption employed in the half reactor simulations shown in Figs. 2-7. The contour plots are perfectly symmetric about the inlet axis upstream of the gap. This symmetry is broken only when the gap is near the inlet, but this case was not considered in the half reactor simulations.

When the gap is near the outlet there is only a single glass plate inside the reactor. This produces a uniform coating across this plate. At the other extreme, placing the gap near the inlet also produces a uniform coating on both plates in the reactor. The uniformity is lost somewhat only near the inlet and outlet for both of these cases. This variation will not impact the overall coating uniformity as all points on a plate will be equally affected as they successively pass through the inlet and the outlets. In contrast to these cases, when the gap is at the center of the downstream section of the reaction chamber, there was a drastic drop off of nearly 80% in the coating efficiency in the downstream plate. The upstream plate mostly maintains the uniformity and efficiency of the coating, but near the edge there is a moderate edge effect

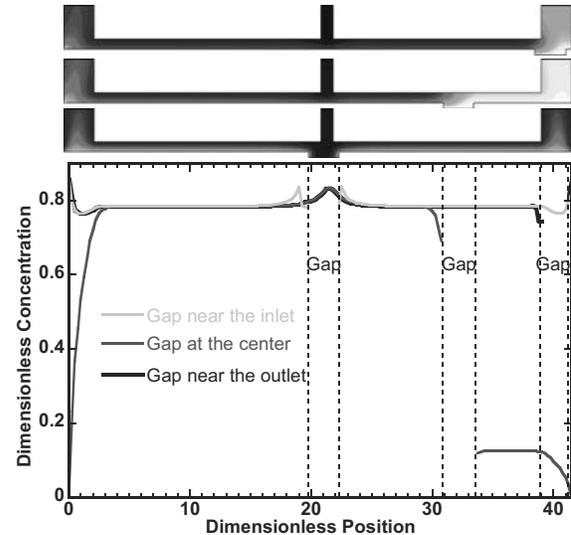


Figure 8: Concentration contour plots (above) and the corresponding plot of the reactant concentration near the glass surface as a function of position (below). Results are shown for a large gap in three positions in the downstream section of the reactor with a bias of 1.0. The dark area in the contour plots corresponds to the maximum concentration and the light area corresponds to the minimum concentration.

with a small drop in coating uniformity.

The reason for this drop in the coating rate at the downstream plate is the leakage of coating gases through the gap and the corresponding intake of inert gases. As shown in Fig. 3, a bias of 1 means there is not net flow of gas through the gap, but this does not prevent offsetting leakage and intake. This causes the coating gases to be significantly diluted downstream of the gap resulting in decreased coating efficiencies on the downstream plate.

While there are variations in coating efficiency between the two plates in the reactor, the coating uniformity across a single plate is mostly maintained as shown in Figs. 8 and 9, with only a small edge effect near the gap in the upstream plate. This result is in agreement with the concentration contour plots shown in Figs. 6 and 7. However, the drop off in coating efficiency between the plates observed when the gap is between the

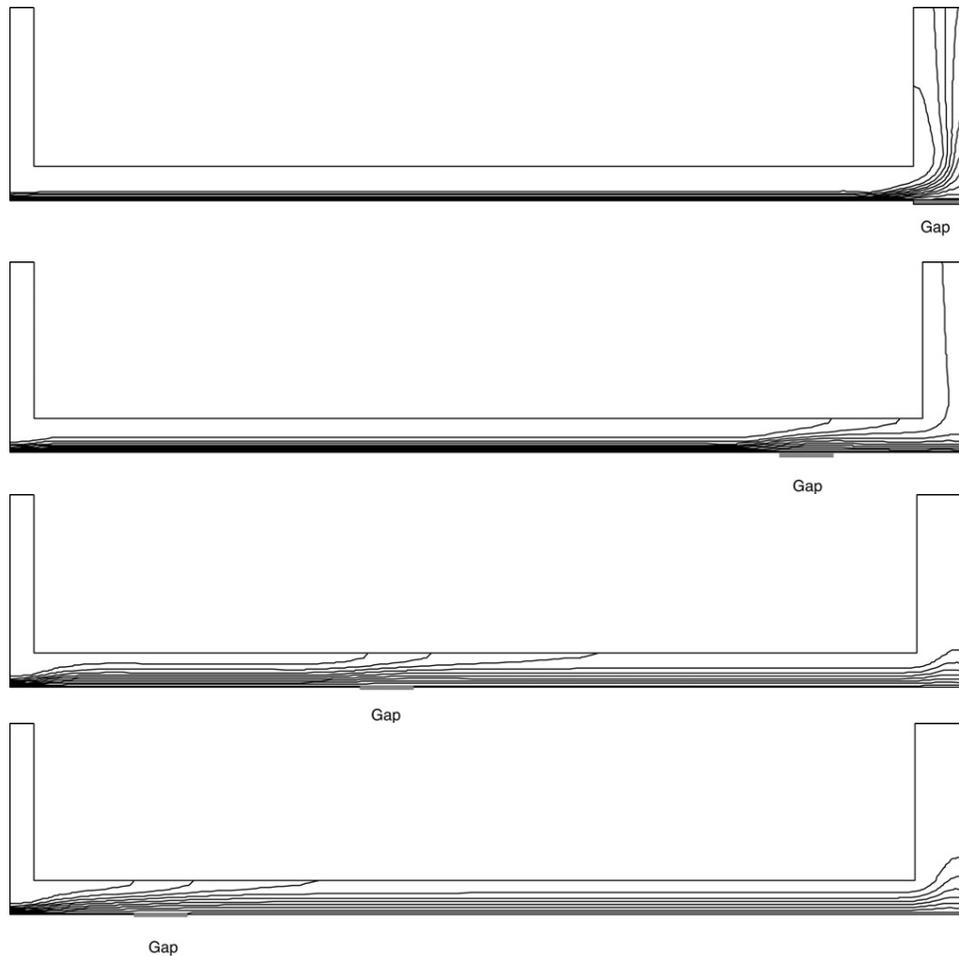


Figure 7: Concentration contour plots for gas flows with a bias of 1 when the gap between the plates is at different locations. The area of the bottom section highlighted shows the location of the gap.

inlet and outlet is not seen when the gap is directly below the inlet. This is because the down flow pattern from the inlet and its symmetry prevents the formation of a circulation pattern in the gap. As a result there is no coating gas leakage and corresponding inert gas intake at the gap in this position and no loss in coating efficiency.

When Figs. 8 and 9 are compared, when the gap is between the inlet and outlet, the drop off in coating efficiency in the downstream plate is not so drastic when the size of the gap between the plates is reduced. This is because a smaller gap reduces the circulation pattern there and the corresponding mixing between the coater gases and the inert external gases. As can be expected, when the gap was near the inlet and the outlets the gap size had

little effect on the coating efficiency.

Given the uniformity of the coating when the gap is near the inlet and outlet, the remaining simulations will focus on the results when the gap is in-between. The contour plots in Figs. 6 had already indicated that changing the bias would affect the difference in coating efficiency between the two plates in the reactor. This can be seen more clearly when the concentration near the surface is plotted as a function of position on the glass surface as shown in Figs. 10 and 11 for biases of 0.5, 1.0 and 1.5. They show that decreasing the bias reduces the drop off in coating efficiency significantly. This reduction is magnified for smaller gaps between plates. Additionally, decreasing the bias also minimizes the edge effect in

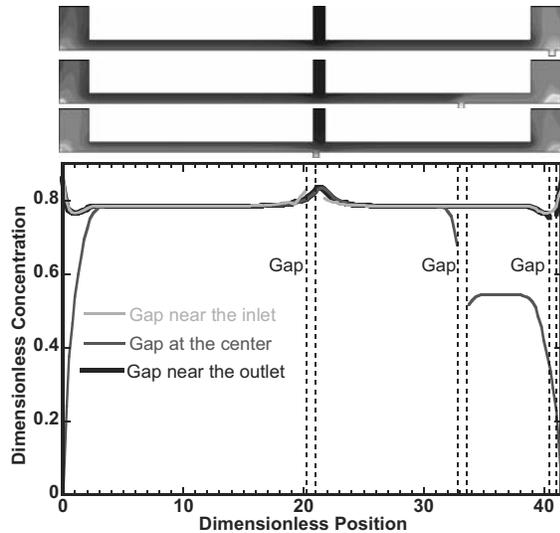


Figure 9: Concentration contour plots (above) and the corresponding plot of the reactant concentration near the glass surface as a function of distance from the upstream outlet (below). Results are shown for a narrow gap in three positions in the downstream section of the reactor with a bias of 1.0. The dark area in the contour plots corresponds to the maximum concentration and the light area corresponds to the minimum concentration.

the upstream plate.

The reason for this dependence on the bias was seen from the velocity plots in Figs. 2 to 5. For biases less than 1 some of the coating gases will flow out through the gap. For the 0.5 bias shown, half of the coating gases will flow through the gap with the remainder exiting through the outlets. Such losses mean that there is less coating gas available for the downstream plate. However, this is offset by the decrease of inert gases flowing into the reactor. As a result this bias produced the least drop off of coating efficiency in the downstream plate. When the gap was narrowed there was very little intake of inert gases as shown in Fig. 11, and the drop off in efficiency was less than 10%.

At higher biases significant amounts of inert gases flow into the reactor through the gap and it is impossible to avoid significant decreases in coating efficiency at the downstream plate. As a result, as

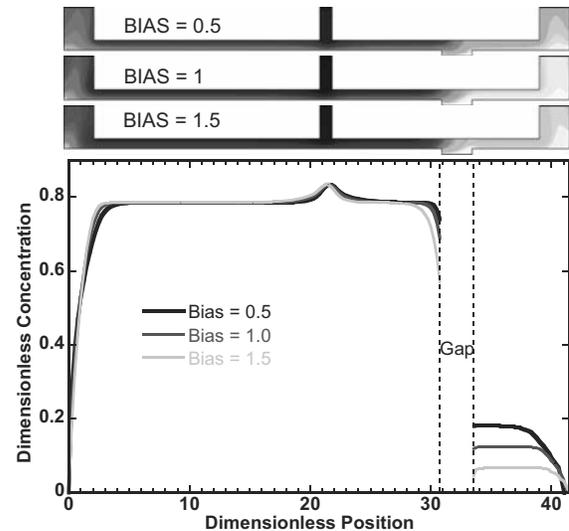


Figure 10: Concentration contour plots (above) and the corresponding plot of the reactant concentration near the glass surface as a function of distance from the upstream outlet (below). Results are shown for a large gap with biases of 0.5, 1.0 and 1.5. The dark area in the contour plots corresponds to the maximum concentration and the light area corresponds to the minimum concentration.

the bias increases to 1.5 the coating efficiency in the downstream plate is over 90% lower than the upstream one. However, for these cases it is possible to significantly increase coating efficiencies on the downstream plate by narrowing the gap as shown in Fig. 11.

Increasing the biases also increases the edge effect on the upstream plate as seen in Figs. 10 and 11. Narrowing the gap caused the area of this coating nonuniformity to narrow as well. For a bias of 1.5, the large gap between the plates (1 inch) caused an edge effect up to 0.75 inches from the gap, while for a narrow gap (0.2 inches) this effect was less than 0.5 inches. Smaller biases produce even smaller edge effects. This effect is most likely caused by the intake of external gases through the gap and by some of the mixing occurring near the upstream plate. Interestingly, this edge effect is not significant on the downstream plate. This again suggests that the mixing of gases occurs near the upstream plate while near

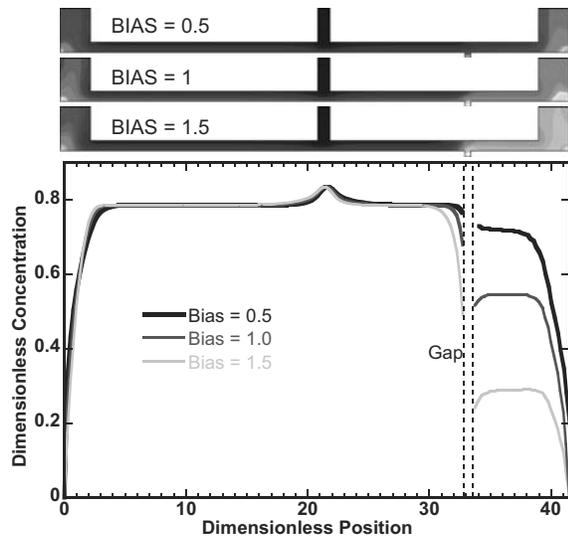


Figure 11: Concentration contour plots (above) and the corresponding plot of the reactant concentration near the glass surface as a function of distance from the upstream outlet (below). Results are shown for a narrow gap with biases of 0.5, 1.0 and 1.5. The dark area in the contour plots corresponds to the maximum concentration and the light area corresponds to the minimum concentration.

the downstream plate the mixing is complete. The contour plots in Figs. 10 and 11 show that the area of rapid concentration variation is indeed mostly near the edge of the upstream plate at the gap.

The above results suggest that the uniformity of the coatings is only moderately affected by the presence of gaps between the plates in the coating process. A small edge effect does occur near at the downstream edge of the plates. The only other variations occur when the plates pass the outlets and the inlet. As mentioned before, unlike the edge effect, these variations will not affect the coating uniformity as all points on the plate sequentially pass through these stages. In other words, the uniformity of the coating with discrete glass plates seems to resemble that for the continuous plates. To verify this some simulations were carried out with the continuous process under similar conditions and the results are shown in Fig. 12. Comparing these results with those in Figs. 10 and 11 it is easy to see the similar-

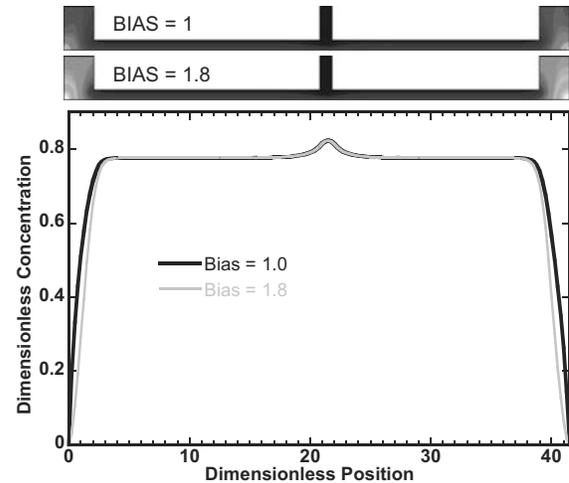


Figure 12: Concentration contour plots (above) and the corresponding plot of the reactant concentration near the glass surface as a function of distance from the upstream outlet (below). Results are shown for the continuous coating process with biases of 1.0 and 1.8. The dark area in the contour plots corresponds to the maximum concentration and the light area corresponds to the minimum concentration.

ties in coating uniformity in the discrete and continuous processes. However, it is also clear that the continuous coating process maintains a uniformly high coating efficiency unlike the discrete process.

The simulations strongly suggest that the quality of the coating with this discrete process should be comparable with those from the continuous one. However, the efficiency of the coating process with the discrete glass plates is much lower than that for the continuous process. When the plate enters the coater the coating efficiency will be quite low. This will remain low until the leading edge of the plate passes under the inlet funnel. At this point the coating efficiency jumps to the value expected for the continuous process. This high coating efficiency remains until the trailing edge of the plate passes under the inlet funnel. At this point the coating efficiency again drops to the previous low value until the plate passes out of the coater. This means that for a plate the size of the reactor length, the high efficiency coating rate will

occur only 50% of the time in the coater. Longer plates will have higher fractions of times at the high efficiency coating rate and smaller plates will have lower fractions. As a result the plate motion through the coater will have to be slowed appropriately to achieve the same level of coating as the continuous coating process.

The results also show that the critical parameters controlling the coating efficiency and uniformity of this process are the bias and the gap between the plates. Careful control of these parameters is important for obtaining optimal coating efficiencies and uniformities. In particular, narrowing the gap between the plates as much as possible and lowering the bias to as close to 1 as possible minimizes the drop offs in coating efficiencies in downstream plates. Lowering the bias below 1 will improve this further, but it will come at the cost of losing coating gases through the gap which is undesirable.

4 Conclusions

The simulations of the discrete coating process with the 2D pseudo steady state model with detailed reactor geometry were carried out employing the FIDAP program. The velocity profiles obtained showed that the gas flows for this process will be well ordered without any turbulent effects. However, the simulations showed that the value of the bias can affect the flow of inert external gases into the reactor through the gap between the discrete glass plates.

The concentration profiles from the simulations showed that the gap between the plates had a small effect on the uniformity of the coatings, producing only a small edge effect at the downstream edge. This nonuniformity increases modestly with the size of the gap and the value of the bias. These parameters had a much stronger effect on the coating rate or efficiency, with the intake of inert external gases into the coater through the gap contributing to lowered coating efficiencies. Lowering the bias and reducing the gap size will lower the reduction in coating efficiencies for this process. The simulations also showed that the reduction in coating efficiencies was not uniform throughout the reactor, but occurred at particular

points. They also suggested that by appropriately adjusting the plate motion through the reactor it will be possible to compensate for reductions in coating rates and to produce coatings of almost equal uniformity and thickness for the discrete glass plates as those for the continuous glass ribbons.

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