Parametric CFD Optimization of an APCVD Glass Coating Deposition Module

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ABSTRACT

Atmospheric Pressure Chemical Vapor Deposition (APCVD) thin film coating process is one of the most cost efficient large area thin film coating solutions presently available on the market and can be up to 2.5 times lower in cost compared with a low pressure sputtering system. Advanced materials such as transparent conductive oxides (TCO) used for solar panel manufacturing and for energy saving (Low-E) windows already have been deposited with APCVD Tools incorporating one or more deposition modules. Thin films such as SiO_2, TiO_2 and F: SnO_2, etc have been successfully deposited onto glass sheets. However further improvement in material efficiencies and operational cost reductions are needed to satisfy the growing demand for such highly customized materials. It is also desirable in the future to be able to deposit other material films which traditionally have not yet been available on this lower cost APCVD manufacturing platform, such as zinc oxide (ZnO).

To investigate in a quantitative manner the improvement potential for the traditional APCVD deposition module design solution we performed a multidimensional computational fluid dynamics (CFD) parametric study using ANSYS FLUENT V12. As a baseline deposition module design we used a commercially available APCVD deposition module originally developed by Watkins-Johnson for SiO_2 deposition trench fill of Si wafers from TEOS, O_2 and Ozone from which we could locate in previously published papers for both experiment and CFD modeling results. The CFD software enabled us to perform a full parametric APCVD deposition module design study and allowed us to quantify the efficiency and throughput gains/losses of a wide variety of design change options. The main driver for this study was to learn in a quantitative, cost efficient and time efficient manner about what system design modifications have the potential for significant precursor efficiency increase and/or deposition throughput gain for a particular APCVD deposition process. The results of this study will be utilized to accelerate our proprietary, next generation Off-line and On-line *CVDgCoat*™ APCVD platform development.

INTRODUCTION

Transparent conductive oxides coatings are thin films deposited on regular glass, which are transparent to visible light and conductive to electricity. Generally, they are in the forms of indium tin oxide (ITO), fluorine doped tin oxide (SnO_2: F), and doped zinc oxide (ZnO). TCO coatings have wide applications in both semiconductor and photovoltaic industries. For instance, ITO can be used as infrared-reflective coating to make energy-conserving windows as well as be applied in flat panel displays and solar cells.

There are various methods to deposit TCO films on a substrate, including metal organic molecular beam deposition, spray pyrolysis, pulsed laser deposition, magnetron sputtering and chemical vapor deposition [1]. Due to easy operation and continuous prodution, atmospheric pressure chemical vapor deposition (APCVD) becomes a cost-effective and thus popular method for TCO coating on substrates [2-4]. However, since it is operating near one atmosphere pressure, inside the deposition module the fluid flow, heat and mass transportation, and chemical reactions become very complicated. Instead of using a try-and-error experiment method, Computation fliud dynamics (CFD) modeling has been utilized as a time and cost effective method to reveal the detailed transport and reaction mechanisms in the reactor so as to improve the system design and optimize operating conditions for a uniform and high quality film deposition [5-9].

The purpose of this work is to perform parametric CFD study to investigate the improvement potential for a traditional APCVD deposition module. Since a SiO_2 or SiC_xO_y sodium diffusion barrier layer is typically deposited by an APCVD method between the glass substrate and the TCO layer so as to improve the life time, transmission and/or quality of it, in our model all the transport phenomena mentioned above are considered to predict SiO_2 deposition rate and rate distribution. A commercially available APCVD deposition module design originally developed by Watkins-Johnson for SiO_2 deposition has been selected as a base case. Same operating conditions as experiments are first utilized and the simulation results are compared with the published experiment data. Very good agreements are achieved between them. Based on this geometric and operating conditions are modified systematically to identify their effects on the final deposition rate and quality. Optimized conditions are therefore revealed. The results obtained in this work can then be used to to investigate if the same deposition module design is also compatible to deposit other coatings, for example fluorine doped tin oxide by an APCVD methodsor if a different geometry need to be selected for the respective APCVD deposition module.

CFD MODELING

Figure 1(a) shows the schematic of a traditional APCVD deposition module developed by Watkins Johnson Company, which mainly consists of inlet slot, injector head, nitrogen curtain, glass substrate and exhaust. The reactant gas is Silane and Oxygen diluted by Nitrogen. In the baseline case of CFD simulation, the glass is assumed motionless. Since the geometry and boundary condition is symmetric about the centerline of inlet slot, half of the geometry is used in CFD modeling to save computational expense. Figure 1(b) presents the grids distribution in the fluid part of half system.

Gas flow in the channel between injector head roof and substrate is laminar due to the low Reynolds number caused by small inlet flow rate. We only studied a steady state process condition. The governing equations to describe fluid motion, temperature distribution and mass transport are listed as follows

Continuity
$$\nabla \cdot (\rho \vec{u}) = 0 \tag{1}$$

Momentum
$$\nabla \cdot (\rho \vec{u}\vec{u}) = -\nabla p + \mu \nabla^2 \vec{u} + \rho \vec{g} \tag{2}$$

Energy
$$\nabla \cdot (\rho C_p T \vec{u}) = \nabla \cdot (k \nabla T) + S \tag{3}$$

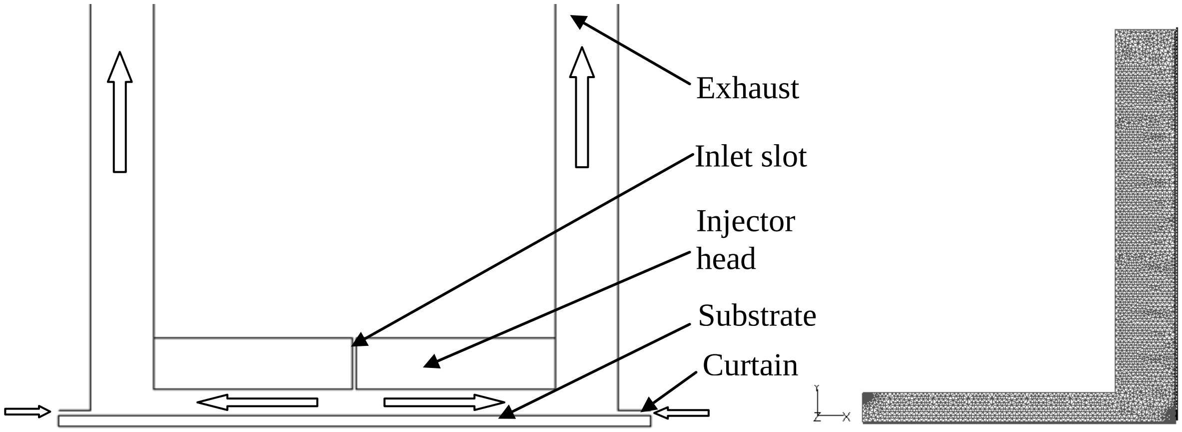
Species
$$\nabla \cdot (\rho \vec{u} Y_i) = -\nabla \cdot \vec{J}_i + R_i \tag{4}$$

where ρ, μ, C_p, k are the density, dynamic viscosity, specific heat and thermal conductivity of gas, respectively; $\vec{u}$, T, p are the velocity vector, temperature and pressure, respectively; S is source term due to chemical reaction; Y_i is the mass fraction of species i, $\vec{J}_i$ is the diffusion flux of species i, R_i is the net rate of production of species i by chemical reaction.

Gas phase chemical reaction mechanism for Silane combustion with Oxygen has been studied by many researchers since 1970's [10-14]. In this paper, the mechanism proposed by Britten et al. [14] is used, which is also called BTW model in literature. It consists of 70 elementary reactions and most of them are reversible. Both forward and reverse chemical reactions are considered in this work. Although gas phase reaction mechanism between Silane and Oxygen was studied in details, surface reaction for SiO_2 deposition on substrate surface was not well developed yet. Little reference about surface reaction mechanism could be found. However some experimental results are available in literatures [15]. In this work, a sticking coefficient method is used to simulate the deposition of SiO_2 on substrate. It is assumed that all SiO_2 on-containing species except Silane will deposit a certain amount of SiO_2 based on the specified sticking coefficient. Then the total deposition rate of SiO_2 can be obtained by summing up all of SiO_2 deposited from different Silicon-containing species. The correlation between deposition rate and sticking coefficient is described as follows:

$$R_S = \gamma_i C_i \sqrt{\frac{RT_S}{2\pi M_i}} \tag{5}$$

where R_S is the surface reaction rate, γ_i is the reactive sticking coefficient for species i with $\gamma_i \le 1$; C_i is the molar concentration of species i on substrate surface; R is the gas constant; T_S is the temperature of substrate surface; M_i is the molecular weight of species i.



(a) (b)

Figure 1. (a) Sketch of system geometry used in CFD modeling, (b) Grids distribution

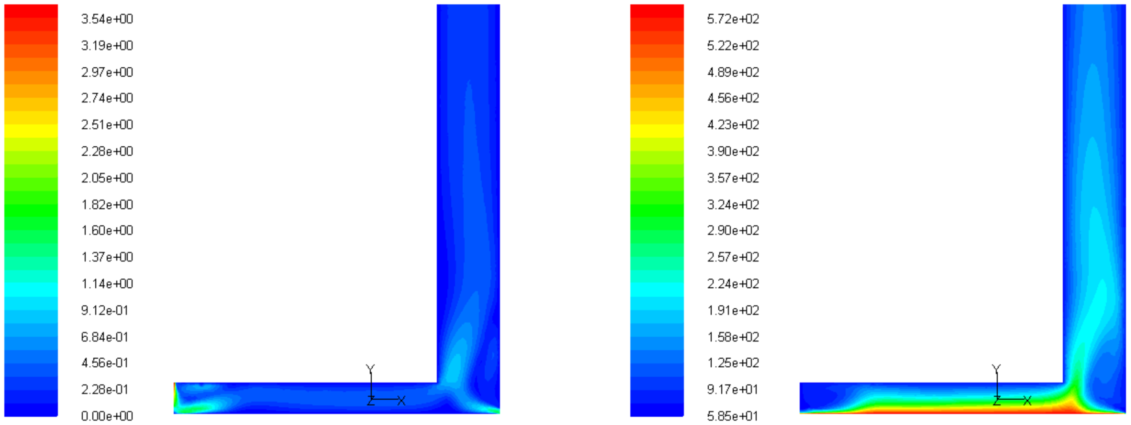
RESULTS & DISCUSSION

Velocity and temperature distribution

Velocity and temperature distribution is obtained by ANSYS FLUENT 12.0. Figure 2(a) shows the contours of velocity magnitude in the channel. Highest velocity occurs in the inlet slot because of the small gap. After a certain distance from the inlet slot, gas flow is well developed. Recirculation is formed near the gas inlet regions. Figure 2(b) presents the temperature distribution. It is clearly seen that the gas is heated up by the hot substrate. In the region where gas flow is well developed, temperature profile along gas moving direction is very uniform.

Analysis of simulation result to experiment data

Through the analysis of molar concentration of different Silicon-containing species simulated by ANSYS FLUENT, we found that only three gas species, i.e., HSiOOH, SiOOH and SiO_2 make the major contributions to the deposition rate of Silicon dioxide on the glass. By tuning the sticking coefficient of them, good agreement between simulation result and experiment data [15] was achieved. Figure 3 shows the comparison of deposition rate by CFD simulation and experiment. The run conditions in the experiment are as follows: total flow rate is 6.0 SLM with 0.23% of Silane, 1.7% of Oxygen and the balance of the flow is nitrogen; and the temperature of injector head and substrate are 60 °C and 572 °C, respectively. In our 2D CFD modeling, the same condition is used. The two profiles match well except the region near the inlet. This might be caused by the difference of flow condition and temperature boundary condition between CFD modeling and experiment. This case was used as a baseline for our parametric studies which investigated the total deposition thickness change in dependence of various geometric and operating conditions. In the baseline case, the distance between injector head roof and glass is 6mm and the width of injector head is 50mm.



(a) (b)

Figure 2. (a) Contours of velocity magnitude, and (b) Temperature distribution of baseline case

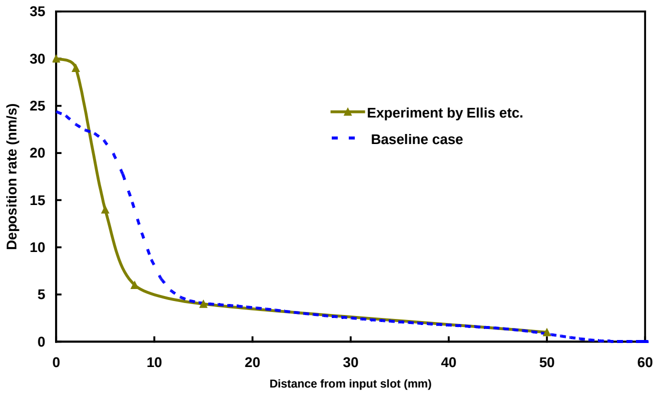


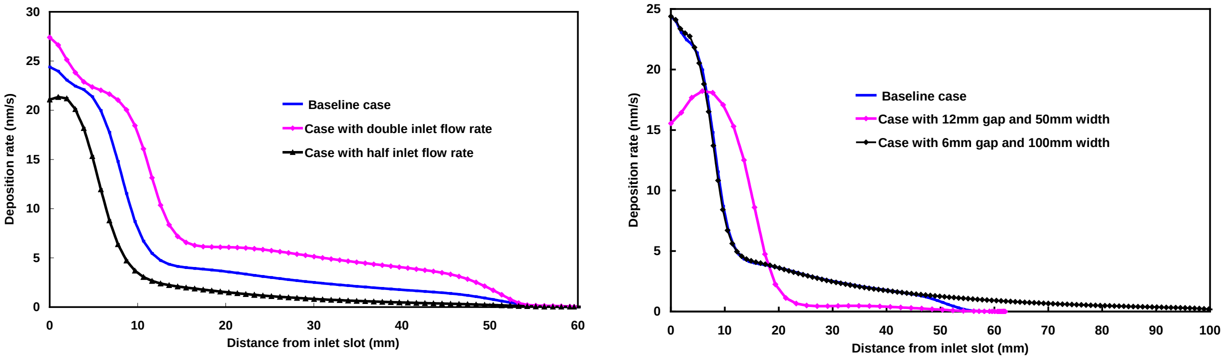
Figure 3. Comparison of deposition rate profile of silicon dioxide between CFD modeling and experiment

Effect of inlet flow rate

The effect of inlet flow rate on deposition rate is presented in Figure 4(a). With the increase of inlet flow rate, the maximum and average deposition rate is increased correspondingly, but the shape of deposition profile is very similar. The conversion efficiency of Silane to SiO_2 for the three cases are 12.8%, 17.2% and 20.6% respectively, which means the efficiency reduced with increase of inlet flow rate. It indicates that the increase of deposition rate is achieved by the sacrifice of conversion efficiency when a larger inlet flow rate is used. Therefore a balance between deposition rate and conversion efficiency need to be made.

Effect of reactor geometry variation on deposition

As expected, geometry change of injector head will influence both the deposition rate and conversion efficiency. The gap between injector head roof and glass, and the width of injector head are two important parameters in practical APCVD process and system optimization developments. Two more cases were simulated to reveal the size of the effect of geometry on deposition rate and conversion efficiency. In one case the gap between injector roof and glass is increased to 12mm (double width), and in the other one the injector width is extended to 100mm (double length). Figure 4(b) presents the deposition rate profiles compared with the baseline case. We observe that the maximum deposition rate is shifted to the right for the case with larger gap. After a 20mm distance from the gas injection port, the deposition rate decreases rapidly. For the case with doubled width, deposition rate within 50mm is almost the same as that of baseline case. After 50mm, it is reduced gradually. Both the average deposition rate and conversion efficiency for the case with larger gap are lower than those of baseline case because of diffusion boundary layer thickness increase on the substrate. However, for the double length injector head case, the average deposition rate is decreased, while total efficiency is increased.



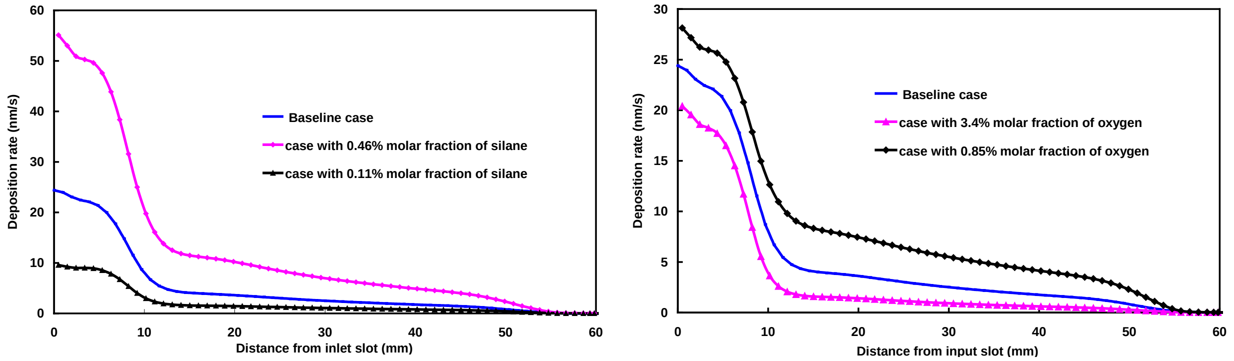
(a) (b)

Figure 4. (a) Comparison of deposition rate for three cases with different inlet flow rates, (b) Change of deposition rate when gap between injector roof and substrate, and width of injector head are modified

Effect of Silane and Oxygen molar fraction

Molar fraction of Silane and Oxygen in the baseline case is 0.23% and 1.7%, respectively. Two more cases are conducted in which the molar fraction of Silane is doubled and then reduced to half, respectively. Effect of molar fraction change of Silane on deposition rate is indicated in Figure 5(a). We found that both the average and maximum deposition rate is increased with the increase of molar fraction of Silane. This observation is consistent with the experimental data [15]. We found that deposition rate of SiO_2 depends on the concentration of several radicals generated by gas phase free-radical chain reactions. The conversion efficiency of Silane for the three cases are 13.8%, 17.2% and 20.8%, respectively, which means that increase of Silane molar fraction will augment the conversion of Silicon-containing species to SiO_2. However, molar fraction of Silane should be lower than a critical value (4%) to prevent explosion of Silane in present of Oxygen.

Similarly, molar fraction of Oxygen is doubled or decreased to half in another two cases studied. The profile of deposition rate for the three cases with different molar fraction of Oxygen is shown in Figure 5(b). We observe that both maximum and average deposition rates decrease with the increase of molar fraction of Oxygen. In the experimental results published[15], the authors reported that the peak growth rate is shifted with larger molar fraction of Oxygen, which demonstrated that Oxygen might hinder the gas phase chemical reactions and thus reduce the growth rate.



(a) (b)

Figure 5. Comparison of deposition rate for cases (a) with different molar faction of Silane, (b) with different molar faction of Oxygen

Summary of conversion efficiency

Table 1 summarizes the conversion efficiency (~total deposition efficiency) obtained for all of the run cases. We found that the case with double molar fraction of Oxygen has the lowest conversion efficiency and the case with half molar fraction of Oxygen resulted of the highest conversion efficiency of all cases studied.

Table 1 Summary of conversion efficiency for all cases

Case description	Conversion efficiency	Ratio to baseline case
Baseline case	17.2%	
Double inlet flow rate	12.8%	74.4%
Half inlet flow rate	20.6%	120.3%
Double molar fraction of Oxygen	10.6%	61.7%
Half molar fraction of Oxygen	25.2%	146.5%
Double molar fraction of Silane	20.8%	121.5%
Half molar fraction of Silane	13.8%	13.8%

CONCLUSIONS

This work performed parametric CFD modeling of SiO_2 by an APCVD process using Silane and Oxygen as CVD precursors. The spatial varying deposition rate distribution matches the experimental date well when a suitable sticking coefficient is chosen. Effects of inlet gas flow rate, gap between injector roof and substrate, width of injector head, molar fraction of Silane and Oxygen on the deposition rate and conversion efficiency where investigated. We observed that the most effective way to increase conversion efficiency was to decrease the molar fraction of Oxygen and to increase the deposition head width. The results obtained by current CFD modeling can be used to optimize the next generation Off-line and On-line *CVDgCoat*™ APCVD platform.

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