

# Optimization of Operating Conditions in a Single-Zone Drier for Two-Layer Polymer Coatings

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**ABSTRACT:** In industrial dryers, hot air is blown over wet coatings either from top or from both top and bottom sides to remove the solvent. Drying of multilayer coatings is a complicated process that involves simultaneous heat and mass transfer, transport, and thermodynamic behavior of polymer solutions. Airflow and its temperature are important operating conditions in a dryer. We report optimization of the operating conditions to minimize residual solvent without inducing blisters for two-layer coatings using mathematical modeling. Our results reveal that optimal air flow

on the bottom side of the coating is always greater than or equal to that on its top side—an agreement with the common notion of “backside drying.” Furthermore, the effect of coating thickness, residence time, individual layer thickness, initial solvent concentration, and reversal of the two layers on the optimal conditions is discussed. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 111: 308–316, 2009

**Key words:** multilayer coatings; drying; optimization; blisters

## INTRODUCTION

Many commercial coatings are made by spreading thin layer(s) of a polymer solution(s), prepared by dissolving a polymer in a solvent(s), on a moving substrate and then drying the layer(s) to remove the solvent. Alsoy and Duda<sup>1</sup> noted that multilayer coatings have gained importance to make films with unique properties. All the layers are usually coated simultaneously before they are dried. Jets of hot air blown either across or onto the coated substrate, both from top and bottom sides, dry the coating. The chief goal of drying is to remove the solvent without creating defects such as blisters and cracking in the coating. Typical operating conditions that can be manipulated to achieve the goal are air flow rate and its temperature.

During drying of a single-layer coating, solvent departs only from top of the coating if the substrate is impermeable; the polymer being nonvolatile does not leave the coating. This leads to concentration gradient both for the solvent and the polymer; the concentration is lowest at the top and highest at the bottom for the solvent and *vice versa* for the polymer. As drying progresses, the solvent concentration falls everywhere in the coating with a concomitant rise in the polymer concentration. At the top, with falling solvent concentration, the diffusion coefficient, which is a strong

function of concentration and temperature,<sup>2,3</sup> also falls. Hence, to match the external flux of solvent due to airflow, a steep concentration gradient of the solvent develops at the top. Concomitantly, a steep concentration gradient of the polymer also develops, with its concentration being highest at the top. For two-layer coatings, as shown by Alsoy and Duda,<sup>1</sup> steep gradient for the solvent can develop not only at the top but also at the interface between the layers.

At any instant of time during drying, bubble point temperature corresponding to solvent concentration at a location in the coating can be estimated by equating solvent partial pressure to ambient pressure (usually 1 atm). Partial pressure equals vapor pressure multiplied by solvent activity; the latter can be calculated by Flory–Huggins theory. Rising solvent activity through the coating thickness reduces the bubble point temperature from the top to the bottom of the coating. Hence, the temperature at which the coating boils or blisters are generated is taken as the bubble point temperature at the bottom.

Solvent concentration at the bottom does not drop for a certain time, the length of which depends on the operating conditions. During this time, bubble point temperature remains constant at the temperature corresponding to initial solvent concentration. As the solvent concentration at the bottom starts to fall, the bubble point temperature of the coating rises. Being heated from the top and the bottom, the coating temperature, however, rises to the air temperature. Price and Cairncross<sup>4</sup> stated that blisters form when the coating temperature exceeds the

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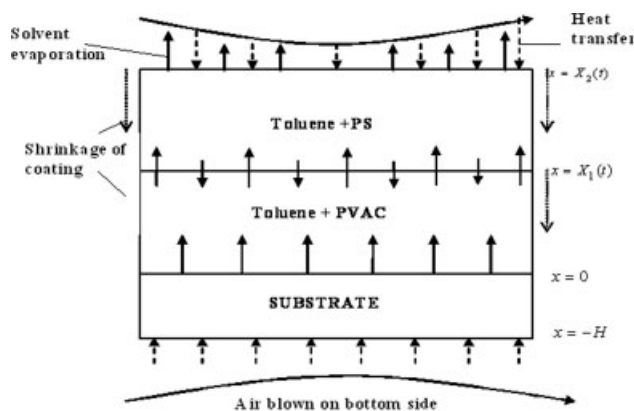
bubble point temperature anytime during drying. Whether or not blisters are induced depends on the operating conditions.

Intense conditions imply faster rise in the coating temperature and it may cross the bubble point temperature causing blisters. These conditions, however, lead to faster solvent removal. Mild conditions result in slower rise in the coating temperature making the blister formation less likely, but more solvent would be retained in the coating. In between the two are optimum conditions, which ensure that the coating is dried adequately fast to minimize residual solvent without creating blisters. Conditions milder than optimum means that residual solvent is more because of slow heating; those intense than optimum means that blisters are induced. Price and Cairncross<sup>4</sup> found the optimum conditions by minimizing the residual solvent and by forcing the coating temperature to equal bubble point temperature sometime during drying.

Mohammadi<sup>5</sup> showed through bench-top experiments that blisters or bubbles can be generated at temperatures well below bubble point even at room temperature. It was observed that bubbles form when either air flow or its temperature is raised. It was hypothesized that blisters form when the coating becomes supersaturated with air during drying; degassing the solution prior to casting reduces the propensity of blister formation. Air solubility in coatings is high when they are applied because of high solvent content and as the solvent departs during drying its concentration, and, hence, the solubility of air reduces leading to saturation and blisters. In this article, we do not consider the effect of dissolved air recognizing that dissolved air produces blisters at milder operating conditions than does boiling.

Aust et al.<sup>6</sup> did not perform optimization of operating conditions but showed that air flows should be reduced as its temperature is raised to prevent blister formation. For a given coating thickness, initial solvent concentration, air temperature, solvent, polymer, and substrate physical properties, and solvent and polymer diffusion properties, Price and Cairncross<sup>4</sup> found that the coatings should be dried at low air flows (or heat-transfer coefficients) on the top and the bottom as the air temperature is raised. Air flows on the top are always higher than those on the bottom. Their results contradict prevalent backside drying approach, which suggests that the air flow (or heat-transfer coefficient) on the bottom should be higher.

In this article, we describe optimization of operating conditions for two-layer coatings. Mass and energy transport equations including thermodynamic equilibrium and transport at interface between the two layers have been described. The solution of these equations has been used for the optimization along same lines as Price and Cairncross.<sup>4</sup> Our optimization results and their analysis indicated



**Figure 1** Schematic of a two-layer coating showing the phenomena during drying. The coating is heated from the top and bottom sides, and the solvent departs only from the top. As the solvent leaves, the coating shrinks. PS and PVAC stand for polystyrene and poly(vinyl acetate), respectively.

that the air flow on the top should always be lower than that on the bottom—a disagreement with the results of Price and Cairncross<sup>4</sup> and an agreement with the backside drying approach. The effect of air temperature, coating thickness, reversal of layers, and change of concentration on optimum operating conditions is discussed.

## GOVERNING EQUATIONS

Figure 1 shows schematic of a drying two-layer coating. The top layer is cast from polystyrene (PS)/toluene solution and the bottom layer from poly(vinyl acetate) (PVAC)/toluene. These coatings have been chosen because their properties to estimate diffusion coefficients are well characterized. The coating is heated from the top and the bottom by hot air; the solvent leaves only from the top. As the solvent departs, the thickness of the two layers changes; sometimes the bottom layer may swell if solvent diffuses into it from the top layer. Both the layers, however, shrink eventually. Even though Alsoy and Duda<sup>1</sup> explained the equations for mass and heat transport and change in thickness of the two layers, these are described here for the sake of completeness.

### Mass transport

The mass transport equations within the coatings were written with respect to volume average velocity. The volume average velocity is zero in the bottom layer and, therefore, the convection term does not appear in the transport equation. The rate of change of the solvent concentration at any point equals the gradient of the solvent flux at that point.

$$\frac{\partial c_1}{\partial t} = \frac{\partial}{\partial x} \left( D_1 \frac{\partial c_1}{\partial x} \right) \quad (1)$$

**TABLE I**  
Free-Volume Parameters for Estimating  
Diffusion Coefficients<sup>1</sup>

Parameter	PVAC toluene	PS toluene
$D_o$ (cm <sup>2</sup> /s)	$4.82 \times 10^{-4}$	$4.82 \times 10^{-4}$
$E_a$ (J/mol)	0	0
$K_{11}/\gamma$ (cm <sup>3</sup> /(g K))	0.00145	0.00145
$K_{12}/\gamma$ (cm <sup>3</sup> /(g K))	0.000433	0.000582
$K_{21}$ (K)	-86.32	-86.32
$K_{22}$ (K)	-258.2	-327
$T_{g1}$ (K)	0	0
$T_{g2}$ (K)	0	0
$K$ (K)	0	0
$\hat{V}_1^*$ (cm <sup>3</sup> /g)	0.917	0.917
$\hat{V}_2^*$ (cm <sup>3</sup> /g)	0.728	0.85
$\xi$	0.82	0.58
$\chi$	0.393	0.354

where  $c_1$  is the concentration of the solvent in the bottom layer and  $D_1$  is the mutual diffusion coefficient of polymer/solvent system in the bottom layer; it is estimated by Vrentas and Duda's free-volume theory. The parameters of the theory, listed in Table I, were taken from the work of Alsoy and Duda,<sup>1</sup> who had obtained them from Zielinski.<sup>7</sup>

In the top layer, the mass-transfer equation involves the convection term because the volume average velocity in this layer is not zero.

$$\frac{\partial c_2}{\partial t} + v \frac{\partial c_2}{\partial x} = \frac{\partial}{\partial x} \left( D_2 \frac{\partial c_2}{\partial x} \right) \quad (2)$$

where  $c_2$  is the concentration of the solvent in the top layer,  $D_2$  is the mutual diffusion coefficient in the top layer, and  $v$  is the volume average velocity. It is obtained by mass balance at the interface between the layers:

$$v = D_1 \frac{\partial c_1}{\partial x} \frac{V_s}{(1 - c_1 V_s)} - D_2 \frac{\partial c_2}{\partial x} \frac{V_s}{(1 - c_2 V_s)} \quad (3)$$

where  $V_s$  is the specific volume of the solvent.

#### Boundary and initial conditions

There is no flux of the solvent into the substrate. Hence,

$$x = 0, \quad \frac{\partial c_1}{\partial x} = 0 \quad (4)$$

There is a thermodynamic equilibrium at the interface between the layers. Therefore,

$$x = X_1(t), \quad \mu_1 = \mu_2 \quad (5)$$

where  $X_1$  is the thickness of the bottom layer,  $\mu_1$  is the chemical potential of the solvent at the interface on the bottom layer side, and  $\mu_2$  is the same on the top layer side. These were estimated by using Flory-Huggins theory.

The continuity equation at the interfaces between the layers gives

$$x = X_1(t), \quad \frac{dX_1}{dt} (c_2 - c_1) = c_2 v - D_2 \frac{\partial c_2}{\partial x} + D_1 \frac{\partial c_1}{\partial x} \quad (6)$$

At the air-coating interface, the boundary condition involves a mass-transfer coefficient

$$x = X_2(t), \quad c_2 v - D_2 \frac{\partial c_2}{\partial x} - c_2 \frac{dX_2}{dt} = k_1^G (p_i^G - p_b^G) \quad (7)$$

where  $X_2$  is the thickness of both the layers,  $k_1^G$  is the mass-transfer coefficient, which was calculated from a known heat-transfer coefficient on the top side using Chilton-Colburn analogy. The solvent partial pressure at the surface,  $p_i^G$ , is product of vapor pressure of the pure solvent at the coating temperature and activity of the solvent at the air-coating interface. Partial pressure of the solvent in bulk gas phase  $p_b^G$  was set to zero in this work.

The initial condition for eqs. (1) and (2) was the uniform solvent concentration in both the layers. Use of rigorous approach as suggested by Alsoy and Duda<sup>1</sup> to compute solvent concentration near the interface on both its sides, at the beginning of drying, by thermodynamic equilibrium and unsteady state mass diffusion into semi-infinite slab improves by the accuracy of the results.

#### Heat transport

Coating temperature was modeled by a lumped rather than a distributed approach because the resistance to heat transfer due to conduction in the polymer layers and the substrate is much lower when compared with that due to convection from air.<sup>8</sup> For some cases, we solved for temperature distribution in the coating and the substrate and found that the temperature varied insignificantly across their thickness. Hence, the temperature was considered to be a function of time only and not of position.

The energy balance includes the heat transferred from the air on the top and the bottom sides of the coating-substrate system and the heat of vaporization of the solvent leaving the coating.

$$\frac{dT}{dt} = \frac{1}{A} [h^g (T^g - T) + h^G (T^G - T) - Q \Delta \hat{H}_{vs}] \quad (8)$$

$$Q = c_{2s} v - c_{2s} \frac{dX_2}{dt} \quad (9)$$

$$A = \rho_{\text{sub}} \hat{C}_{\text{psub}} H + \rho_1 \hat{C}_{p1} X_1(t) + \rho_2 \hat{C}_{p2} (X_2(t) - X_1(t))$$

where  $\Delta \hat{H}_{vs}$  is the heat of vaporization of the solvent,  $c_{2s}$  is the solvent concentration at the surface of the coating,  $A$  is the thermal mass of system,  $Q$  is

the rate of solvent evaporation,  $\hat{C}_p$  is the heat capacity, and  $\rho$  is the density. Subscripts "sub," "1," and "2" refer to substrate, bottom, and top layers, respectively.  $H$  is the thickness of the substrate. For the two layers, average values for densities and heat capacities were used. Initial temperature of the coating-substrate system was set to 298 K.

### Layer thickness

The solvent diffuses from the bottom layer to the top layer or *vice versa* depending on its concentration in the two layers. This causes the thickness of both layers to change with time. Eventually, the solvent departs from the surface to the air causing the total thickness of the coating and the thickness of each layer to reduce with time. The thickness of the two layers can be predicted by mass balance of the solvent at the two interfaces:

$$\frac{dX_1}{dt} = \left[ \frac{D_1 \hat{V}_s}{(1 - c_1 \hat{V}_s)} \frac{\partial c_1}{\partial x} \right]_{x=X_1(t)} \quad (10)$$

$$\frac{dX_2}{dt} = \left[ v + \frac{D_2 \hat{V}_s}{(1 - c_2 \hat{V}_s)} \frac{\partial c_2}{\partial x} \right]_{x=X_2(t)} \quad (11)$$

Initial thicknesses of the two layers were input to the drying code.

Equations (1), (2), (6), (8), (10), and (11) are coupled nonlinear partial and ordinary differential equations. These were solved using finite difference method, which transforms the partial differential equations into ordinary differential equations (ODEs). Then the ODEs were integrated with ode15s of MATLAB.

### OPTIMIZATION

For a given set of coating parameters, the problem of optimization is to find the heat-transfer coefficients on both the top and the bottom sides, which minimize the final residual solvent content without generating blisters. Our objective function,  $\Phi$ , defined below, was the final residual solvent.

$$\Phi = \int_0^{X_1(t)} c_1 dx + \int_{X_1(t)}^{X_2(t)} c_2 dx \quad (12)$$

First term on the right-hand side gives the residual solvent in the bottom layer and the second term in the top layer. Generation of blisters is related to solvent partial pressure during drying. Pressure condition was imposed as a constraint,  $\xi$ .

$$\xi = 1 - P_{\max} \quad (13)$$

$P_{\max}$  is the maximum solvent partial pressure during drying. When the condition in eq. (13) is satis-

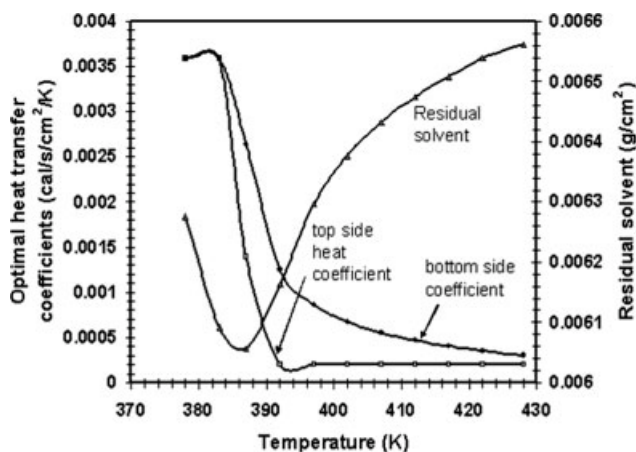
TABLE II  
Model Parameters and Operating Conditions  
for the Base Case<sup>1</sup>

Initial conditions	
Temperature	298 K
Thickness of lower layer	0.01 cm
Thickness of upper layer	0.01 cm
Initial composition of solvent in lower layer	0.642 g/cc
Initial composition of solvent in upper layer	0.722 g/cc
Substrate parameters	
Heat capacity	1.25 J/(g K)
Density	1.37 g/cm <sup>3</sup>
Base thickness	0.00356 cm
Coating parameters	
Heat capacity of lower layer	1.933 J/(g K)
Heat capacity of upper layer	1.653 J/(g K)
Heat of vaporization of the solvent	360 J/(g K)
Operating conditions	
Base side heat transfer coefficient	0.00018 W/(cm <sup>2</sup> K)
Coat side heat transfer coefficient	0.00018 W/(cm <sup>2</sup> K)
Bottom air supply temperature	353 K
Top air supply temperature	353 K
Mass transfer coefficient	0.31 × 10 <sup>-9</sup> s/cm
Mole fraction of solvent in the gas	0.00
Other parameters	
Density of PVAC	1.19 g/cm <sup>3</sup>
Density of toluene	0.862 g/cm <sup>3</sup>
Density of PS	1.05 g/cm <sup>3</sup>

fied, the solvent partial pressure becomes equal to ambient pressure (1 atm). This is tantamount to making the coating temperature and the bubble point temperature equal sometime during drying. The constraint optimization routine, *fmincon*, of MATLAB was used to solve for optimum heat-transfer coefficients.

### RESULTS AND DISCUSSION

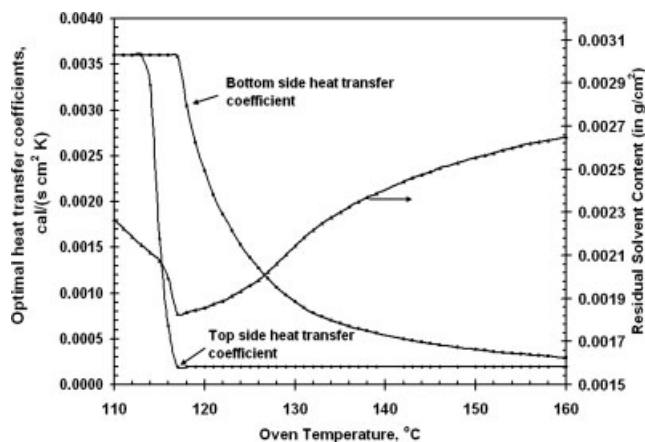
The base case for optimization was a two-layer coating with PVAC-toluene solution as the lower layer and PS-toluene as the upper layer. The physical parameters and operating conditions are listed in Table II. Figure 2 shows the effect of changing the air temperature on residual solvent and optimal heat-transfer coefficients for the base case. Residence time was 30 s, and the initial solvent concentration in the lower and upper layers was 0.722 and 0.642 g/cc, respectively. The results show that the lower heat-transfer coefficients are needed at higher air temperatures to minimize residual solvent without creating blisters. The two coefficients reach their maximum value at air temperatures less than or equal to the bubble point temperature corresponding to initial composition. Also, the top-side heat-transfer coefficient is always lower than or equal to that on the bottom side. Residual solvent depends on air temperature and its flow rate. The residual solvent



**Figure 2** The effect of changing air temperature on the residual solvent and optimal heat-transfer coefficients. The lower layer is PVAC-toluene and the upper layer is PS-toluene. The coating thickness is 200  $\mu\text{m}$ , with the ratio of top to bottom layer thickness being 1 : 1. Residence time is 30 s, and the initial solvent concentration in lower and upper layers are 0.722 and 0.642 g/cc, respectively.

decreases and then rises back as the air temperature is raised. The decrease is due to rise in air temperature and the increase is due to slower rate of heating. The difference between minimum to maximum residual solvent is found to be about 10%.

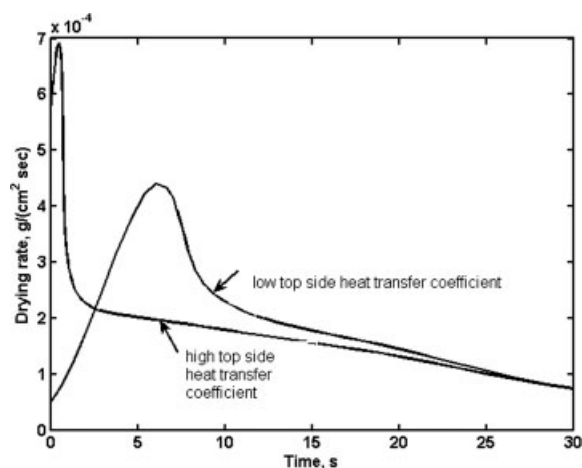
The trends of heat-transfer coefficients obtained by us are the same as those obtained by Price and Cairncross,<sup>4</sup> but their top-side heat-transfer coefficients were always higher than those on the bottom side. Intrigued by the difference between their and our results, we optimized heat-transfer coefficients for a single-layer PVAC/toluene coating of about 150  $\mu\text{m}$  initial thickness containing 30% polymer by weight when coated. Figure 3 was generated by



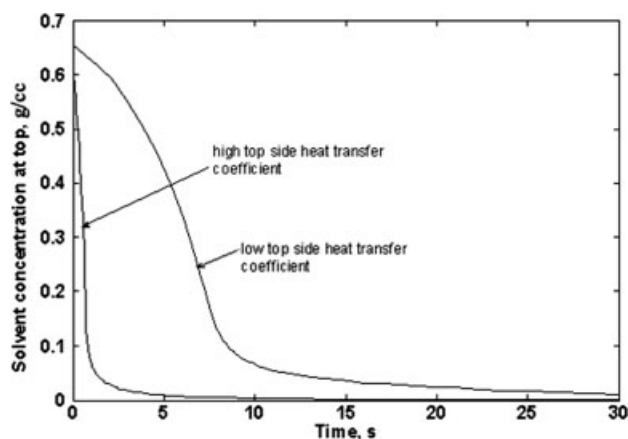
**Figure 3** Optimal top- and bottom-side heat-transfer coefficients at several different air temperatures for a single-layer PVAC-toluene coating. Residence time is 30 s, initial coating thickness is 150  $\mu\text{m}$ , and initial solvent concentration is 0.65 g/cc.

using the same physical and diffusional properties as those used by Price and Cairncross<sup>4</sup> and it shows that even for a single-layer coating the top-side heat-transfer coefficient is always lower than or equal to that on the bottom side. The trends in residual solvent are the same as those for two-layer coatings.

For an air temperature of 120°C, we considered two cases: in the first case, the optimum heat-transfer coefficients obtained by us were used; in the second case, keeping the sum of the coefficients of the first case the same, the top-side coefficient was made higher. In other words, the rate of energy delivery to the coating in both the cases is the same, but mass-transfer coefficient would be higher in the first case than that in the second case. When we ran our drying code (not optimization) for the two cases, it was observed that the residual solvent is higher in the second case. This observation was analyzed by plotting drying rates for the two cases. Figure 4 shows that, for the second case, the drying rates are faster initially but fall precipitously during later stages of drying. This is because of drastic fall in solvent concentration at the top (Fig. 5). Drying rates level off and it leads to what is sometimes called as “figurative skinning.”<sup>9</sup> For the first case, the drying rates are slower initially, but they remain significantly high during the later stages resulting in higher overall drying rate. This exercise was done at other temperatures also and the same result was found. It was noted that the difference in the final residual solvent for the two cases decreases as the air temperature is raised.

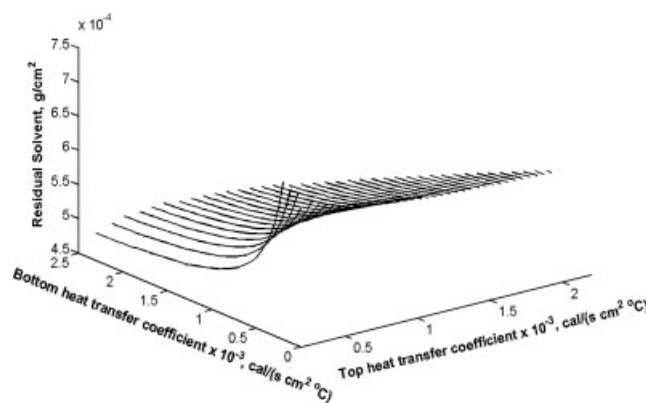


**Figure 4** Drying rates before and after the interchange of optimal heat-transfer coefficients at 120°C for a single-layer PVAC-toluene coating. The top curve corresponds to heat-transfer coefficients before the interchange, which were  $2 \times 10^{-4}$  and  $2.33 \times 10^{-3}$  cal/(s cm<sup>2</sup> K) on the top and the bottom sides. The bottom curve corresponds to heat-transfer coefficients after the interchange. The initial coating thickness and the solvent concentration were 150  $\mu\text{m}$  and 0.65 g/cc, respectively.



**Figure 5** Solvent concentration at the top of the coating before and after the interchange of optimal heat-transfer coefficients at 120°C for a single-layer PVAC-toluene coating. The top curve corresponds to heat-transfer coefficients before the interchange, which were  $2 \times 10^{-4}$  and  $2.33 \times 10^{-3}$  cal/(s cm<sup>2</sup> K) on the top and the bottom sides. The bottom curve corresponds to heat-transfer coefficients after the interchange. The initial coating thickness is 150 μm and the solvent concentration is 0.65 g/cc.

The optimization results were further analyzed by plotting residual solvent as a function of top-side and bottom-side heat-transfer coefficients at 120°C (Fig. 6). Both the heat-transfer coefficients were varied from lowest to highest allowable values in small increments and those coefficients for which the solvent partial pressure was less than 1 atm were recorded. The residual solvent fell as one or both the coefficients were raised. For all the coefficients at the lower end of the curves, the constraint of solvent partial pressure equal to 1 atm was met within a small tolerance, and sum of these coefficients is nearly the same. The residual solvent was found to be lowest



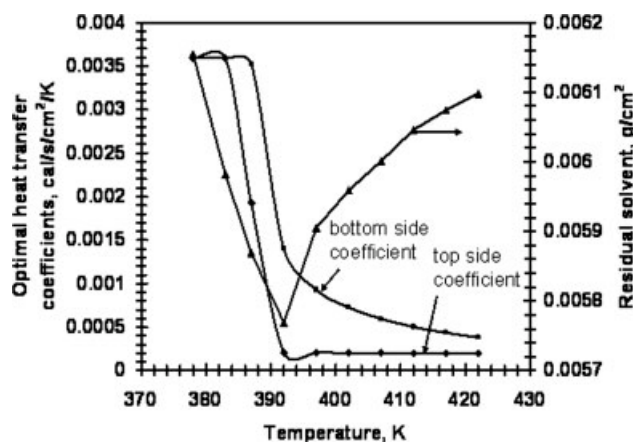
**Figure 6** Residual solvent as a function of top- and bottom-side heat-transfer coefficients at 120°C for a single-layer PVAC-toluene coating. The curves were generated by varying the coefficients in small increments and recording those for which maximum solvent partial pressure is less than 1 atm. Residence time is 30 s, initial coating thickness is 150 μm, and solvent concentration is 0.65 g/cc.

when the top-side and bottom-side coefficients were  $2 \times 10^{-4}$  and  $2.3 \times 10^{-3}$  cal/(s cm<sup>2</sup> K), respectively.

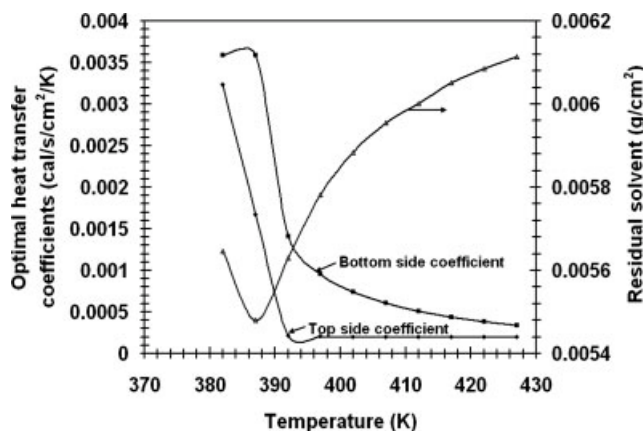
The optimization results of Price and Cairncross<sup>4</sup> converged to a higher top-side coefficient than that on the bottom side. Their formulation of optimization problem is different from ours. They minimized two objective functions: first one had two parts; one part was the difference between maximum solvent partial pressure and ambient pressure and the other part sets limits on heat-transfer coefficients. The second objective function was the residual solvent. The cause for discrepancy between our results and theirs could be the different mathematical formulations of optimization.

A closer look at Figure 6 shows that optimization routine is likely to converge to a higher top-side coefficient if its starting value for the routine is higher. They could have started with a higher top-side heat-transfer coefficient for their routine; the choice of starting values is not discussed in their work. It is to be noted that they mention in their paper that, in some cases, near optimum results are obtained for lower top-side coefficients. Our optimization routine always converged to a lower top-side coefficient for several different initial values of the coefficients. Our results with polystyrene/ethylbenzene system also showed the same trends for heat-transfer coefficients. Those on the top side were always lower than those on the bottom side and both of them fell as air temperature was raised. The optimum temperature shifted to a higher value than for PVAC/toluene system.

Figure 7 shows the optimal heat-transfer coefficients and residual solvent content as a function of air temperature when the top layer concentration is



**Figure 7** The optimal heat-transfer coefficients and residual solvent as a function of air temperature when the top-layer concentration is greater than the bottom-layer concentration. The coating thickness is 200 μm, with the ratio of top to bottom layer thickness being 1 : 1. Residence time is 30 s, and the initial solvent concentration in the upper and lower layer is 0.722 and 0.642 g/cc, respectively.



**Figure 8** The optimal heat-transfer coefficients and residual solvent as a function of air temperature when the polymer layers are reversed (i.e., PS in the lower layer and PVAC in the upper layer). The coating thickness is 200  $\mu\text{m}$ , with the ratio of top to bottom layer thickness being 1 : 1. Residence time is 30 s, and the initial solvent concentration in the lower and upper layer is 0.722 and 0.642 g/cc, respectively.

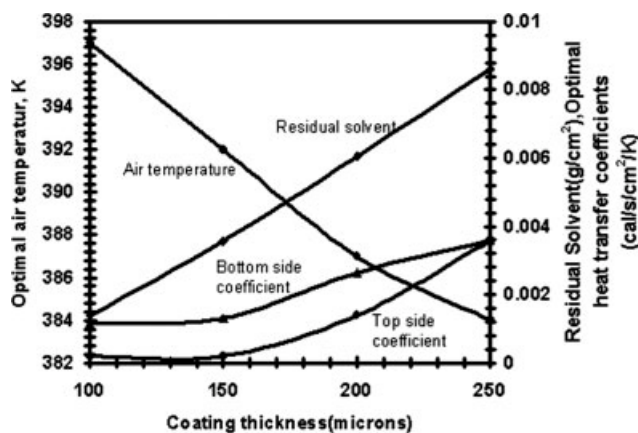
greater than the bottom layer concentration. The trends for the optimal heat-transfer coefficients and residual solvent content remained the same as those of base case. The global optimum oven temperature was 387 K when the solvent concentration in the top layer was less than that in the bottom layer; it rose to 392 K when solvent concentration in the top layer was made higher. For a given oven temperature, the optimal heat-transfer coefficients in this case were always higher than those in the base case where the bottom layer solvent concentration was more than the upper layer. The residual solvent at any temperature was lower than the base case. Again, the top-side coefficient was less than or equal to that on the bottom side.

Figure 8 shows the optimal heat-transfer coefficients and residual solvent content as a function of air temperature when the polymer layers are reversed. That is, PVAC-toluene solution was made the upper layer and PS-toluene the lower layer. The optimal heat-transfer coefficients at any air temperature were higher and the residual solvent content was lower than those for the base case. Alsoy and Duda<sup>1</sup> also showed that the residual solvent is lower if PS is in the lower layer. This is because of reduction of the commonly known "skinning" effect. They did not optimize the operating conditions but fixed all operating parameters and conditions and reversed the polymer layers. The heat-transfer coefficients on the top and the bottom sides were kept the same when PS was in the upper and the lower layers.

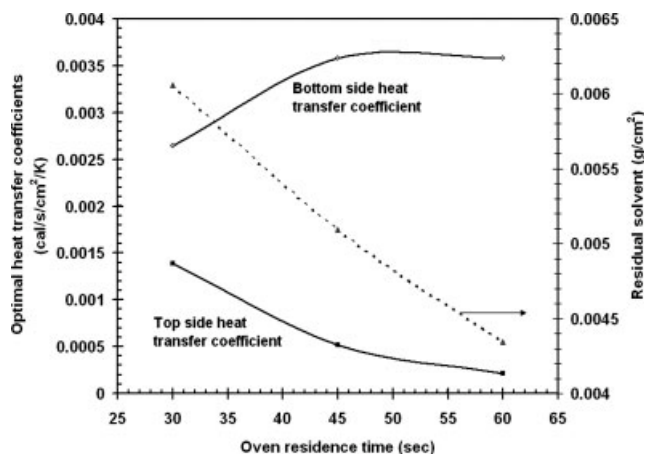
Optimization results show that the air flows are higher when PS is in the bottom layer than when it is in the upper layer, the other conditions being kept the same. For the same concentration, activity of

toluene, and hence its partial pressure, is lower in PS layer than in PVAC layer. All conditions remaining the same, an implication of lower activity in PS is that higher temperatures are needed for single-layer PS-toluene coatings than those for single-layer PVAC-toluene coating to make partial pressure of toluene equal to ambient pressure and, therefore, for blisters to develop. Also, for the same air temperature, PS-toluene coatings can be dried at higher air flows than PVAC-toluene coatings without inducing blisters. Hence, for the same air temperature, two-layer coatings with PS in the lower layer can be heated at a faster rate without the risk of generating blisters. This results in faster rise in coating temperature and faster drying rates because diffusion coefficient rises steeply with rise in temperature. Hence, higher air flows in addition to reduced skinning effect cause lower residual solvent in coatings with PS in the bottom layer.

Our model assumes that both the layers remain in rubbery state through the drying period. This assumption is questionable and it is possible that the regions near the interface in both the top and the bottom layers could become glassy. The transport in coatings that turn glassy during drying is phenomenologically different from that in rubbery systems and cannot be fully described by Fick's law of diffusion.<sup>10</sup> Several papers in the literature describe the drying of the systems under consideration, PS-toluene and PVAC-toluene, by Fick's law of diffusion. We followed their approach. Diffusion in polymer solvent systems that become glassy can also be described by modifying the equation for self-diffusion coefficient derived from the free-volume theory. This modification needs more parameters, which are estimated for few systems. Our results change



**Figure 9** Global optimal heat-transfer coefficients and air temperature as a function of coating thickness. Residence time is 30 s, the initial solvent concentration in lower and upper layer is 0.722 and 0.642 g/cc, respectively, and the ratio of top to bottom layer thickness is 1 : 1.



**Figure 10** Sensitivity of the residual solvent and the global optimal heat-transfer coefficients to the residence time. The coating is initially 200  $\mu\text{m}$  thick, and the initial solvent concentration in the lower and upper layer is 0.722 and 0.642 g/cc, respectively.

quantitatively but not qualitatively if the changed equation is used.

Figure 9 shows the global optimum operating conditions (air temperature and upper and lower heat-transfer coefficients) and residual solvent as a function of coating thickness, which was varied from 100 to 250  $\mu\text{m}$  keeping the proportion of the two layers the same. For each coating thickness, the air temperature at which the residual solvent minimized was recorded as the global optimum temperature. As expected, the residual content was lower in thinner coatings and it rose with the thickness. Thicker coatings required low oven temperatures and consequently high heat-transfer coefficients for optimal drying. These results are similar to those of Price and Cairncross.<sup>4</sup> The thickest coating considered in this work required both the heat-transfer coefficients to be maximum, i.e.,  $3.6 \times 10^{-3}$  cal/(s  $\text{cm}^2$  K) and a low oven temperature of 384 K. As the coating thickness was decreased, the optimum top-side heat-transfer coefficient fell to its minimum allowable value,  $2 \times 10^{-4}$  cal/(s  $\text{cm}^2$  K), and the optimum bottom-side heat-transfer coefficient decreased gradually.

Figure 10 shows the sensitivity of global optimal heat-transfer coefficients and residual solvent content to the residence time. As the residence time was increased, the optimal top-side heat-transfer coefficients fell and those on the bottom-side remained the same. The bottom-side coefficients are expected to fall as the residence time is increased. With increasing residence time, coating temperature reaches bubble point temperature later during drying and lower air flows were needed. The residual solvent, as expected, decreased with rise in residence time.

Optimization results for another two-layer system made from poly(methyl methacrylate)/poly(*p*-methyl

styrene)/ethylbenzene showed the same trends as those shown in Figures 7–10. For example, the top-side heat-transfer coefficients were always lower than or equal to those on the bottom side; the global optimum temperature decreased and the heat-transfer coefficients increased as the coating thickness was increased; the residual solvent was lower when poly(methyl methacrylate) was in the bottom layer because of lower ethylbenzene activity in this layer; global optimum temperature increased and residual solvent decreased when the concentration of the solvent in the top layer was higher than that in the bottom layer.

## CONCLUSIONS

The goals of removing solvent quickly from a coating and drying it without generating blisters conflict with each other. This article discusses optimization of air flow and its temperature for two-layer coatings to avoid the conflict. Each of PS-toluene and PVAC-toluene solutions forms a layer of a two-layer coating. Optimization involves minimizing residual solvent without inducing blisters during drying given coating parameters such as thickness of each layer, initial solvent concentration and residence time. Residual solvent is calculated by solving governing equations for drying of two-layer coatings and, the maximum solvent partial pressure during drying is forced to become equal to bubble point temperature to ensure that the coating dries adequately fast without blister formation. The governing equations are complicated with simultaneous heat and mass transport, thermodynamic equilibrium between the layers, and transport in each of the layers.

For a given air temperature, we found that the top-side air flow (or heat-transfer coefficient) was always lower than that on the bottom side, which contradicts published results for a single-layer coating. The optimization results were analyzed for a single-layer coating. Drying rates are higher when the heat-transfer coefficient is higher on the top side because of the higher mass-transfer coefficient. The rates, however, drop severely because the solvent concentration, and hence the diffusion coefficient, fall at the top of the coating as drying progresses. At lower heat-transfer coefficients (or air flows), however, the drying rates remain significantly high for longer periods, leading to more solvent removal.

Thicker coatings need to be heated at lower air temperatures and higher air flows. The residual solvent was found to be lower in thinner coatings and it increased with total thickness of the coating. The residual solvent in the coating decreased when the initial concentration in the top layer exceeded that in the bottom layer and the coating can be dried at



higher air flows without boiling. As the residence time is raised, the optimal heat-transfer coefficients fall; the bottom-side air flow fell more gradually than that on the top side.

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