Experimental Study of Vapor Deposition and Dew Points in Combustion Gases by Interferometric Technique*

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An investigation has been made of the relationship between optical parameters such as incident angle, surface roughness, and polarization state, which has led to the development of an improved optical method. Utilizing these enhanced experimental techniques, the deposition rates of rapidly growing films consisting of either Na₂SO₃ or K₂SO₃ on a metal surface exposed to flowing combustion product gases has been measured. The experiments were performed with a film thickness less than the critical thickness for the onset of complications due to run-off. At lower metal surface temperatures, condensation of vapors to aerosols occurs before the vapor reaches the surface. Under these conditions, the mechanism of deposition is no longer determined by vapor diffusion alone but is heavily influenced by the combined action of vapor and aerosol transports. At higher surface temperatures, only the vapor deposition effect is predominant. This study also includes measurements of the dew points and evaporation rates.

Key Words: Vapor Deposition, Mass Transfer, Diffusion, Dew Point, Combustion, Interferometry, Polarization

1. Introduction

The deposition and behavior of inorganic compounds from combustion gases are considered to be relevant to the problems of 'hot corrosion' on gas turbine blade surfaces. The presence of sulfur in fuels and the ingestion of various inorganic salts into the combustion chambers along with the intake air have been related to hot corrosion attack in marine, industrial, and aircraft turbine engines\(^{(4)-(9)}\).

There are some basic mechanisms by which condensable vapors in high-temperature combustion gas streams can be deposited onto the cooled turbine blades immersed in the gas stream. These are as follows:

1. Fick diffusion—molecules are transported through the mass boundary layer to condense on the cooled surface.
2. Thermal diffusion—inorganic compounds are driven down the temperature gradient to the cooled surface.
3. Variable property effects—gas properties vary across the boundary layer, thereby affecting the magnitude of the dimensionless mass transfer coefficients.
4. Inertial effects— as large aerosols or particles are concerned, inertia becomes a significant factor in the deposition.

At lower surface temperatures, fog formations within the boundary layer occur, and this vapor-condensation phase transport exhibits different deposition processes from those of vapor phase transport alone.

Conventional experimental measurements of the deposition rates have been carried out by the gravimetric method (e.g., measuring the mass of deposited materials); these experiments need more than 12 hours for each measurement\(^{(6),(9)}\). However, as the thickness of the condensed liquid film increases,
motion is induced in the liquid film by the effects of surface tension and shear-induced forces. Excluding the complications due to run-off, we needed a real-time measuring system and so developed an improved optical interferometric technique. By using this interferometric technique, Seshadri and Rosner carried out measurements on the growth rate of condensate liquid films of $B_2O_3$. The main thrust of the present study is to develop a technique which is capable of taking measurements in the range of lower target temperatures. This is made possible by employing a beam of higher intensity, which we have achieved by the effective use of polarization control. A discussion on polarization control will be given in more in section 3.2.

Nomenclature

$D$: Fick (or Brownian) diffusion coefficient

$j_v$: diffusion mass flux for vapor

$M$: molecular weight

$R$: universal gas constant

$T_e$: temperature at wall

$u$: fluid velocity component parallel to the wall

$v$: fluid velocity component normal to the wall

$x$: streamwise distance along interface

$y$: normal distance to the interface

$\gamma$: activity coefficient

$\lambda$: molar heat of evaporation or sublimation

$\rho$: mass density of the prevailing mixture

$\omega_v$: mass fraction of vapor

$\omega_v^0$: mass fraction of vapor at equilibrium

2. Experiments

2.1 Experimental apparatus

The apparatus (see Fig. 1) consists of the combustion system, the target and temperature measuring systems, a nebulizer aerosol-generating system, and the optical systems.

2.1.1 Combustion system To avoid horizontal temperature variations at the target as much as possible and to obtain a laminar flow of the premixed gases with a uniform velocity above the burner, we make a laminar premixed flat flame burner on which honeycombs are placed. The premixed gas is $C_3H_8-O_2-N_2$. The flame temperature and stability are controlled by providing the proper combination of gases. To improve the stability of the flame and to prevent the influx of atmospheric $O_2$ into the combustion products, shielding flows of $N_2$ are maintained around the burner.

2.1.2 Target and temperature measurement systems The target is a 5 mm wide, 0.127 mm thick Pt ribbon. It is supported above the burner by two copper rods connected to a D.C. power supply which continuously controls the target temperature. The target temperature can be determined by measuring the electrical resistance. The calibrations of the relation between the electrical resistance and the temperature are implemented in the following fashion. The electrical resistance of the target is measured just when the chemical compounds, whose melting temperatures are known, start to melt. Two Pt wires of 0.127 mm dia., spot-welded on the central section of the Pt ribbon, are used to measure the electrical resistance.

2.1.3 Nebulizer-aerosol generating system

To generate highly dispersed aqueous solution aerosols which are transported into the burner via a carrier stream of $N_2$, an ultrasonic nebulizer which consists of a vertical glass cylinder, at the base of which is mounted a piezoelectric transducer, is used. Aqueous solution droplets (e.g., $NaSO_4$, $KSO_4$) produced by this nebulizer are small (3-4 µm dia.) enough to evaporate completely before the combustion gas stream encounters the target.

2.1.4 Optical systems Laser 1. A plane polarized He–Ne laser beam with an output power of 4 mW is focused on the center of the target where deposition is occurring. Incident light (Fig. 2) is reflected from the deposited film surface and, after being transmitted through the film, from the metal surface during the
growth of deposited film. These two divided beams of light undergo constructive and destructive interference due to the optical path differences, causing modulated intensity in the collected beam.

The film thickness increment, $\Delta f_1$, associated with a change from the intensity maximum to minimum is calculated from

$$\Delta f_1 = (1/4)(N^2 - \sin^2 \theta)^{-1/2} \lambda,$$

where $\theta$ is the angle of incidence, $\lambda$ is the wavelength of light and $N$ is the refractive index of the film.

Laser 2- To determine the concentration of the seeding aerosols in the precombustion gas feed stream, four ports are drilled at 90° intervals. Through one opposite pair of ports, covered by thin glass windows, a beam from laser 2 (He-Ne laser, 10 mW) is passed (see Fig. 1). The presence of any seeded material aerosols in the unburned gas mixture through which the beam passes as it crosses the burner causes the light from the beam to be scattered. The light-scattering signals are transmitted through the optical fibers, amplified by the photomultiplier tube, and monitored by the recorder.

2.2 Experimental procedures

While the electronic equipment is stabilized, premixed gases are fed to the burner and ignited. Initially, a high target temperature is maintained so that the initially deposited materials are evaporated and the target surface becomes clean. After all experimental conditions are stabilized, the aerosols of the inorganic compounds with their carrier gas, $N_2$, are directed into the flame. At the same time, we reduce the amount of $N_2$ in the premixed gas by reducing the flow rate of carrier $N_2$ so that the ratio of $N_2$ in premixed gas can be maintained constant at all the times. While passing through the flame zone, aerosols of inorganic compounds are vaporized. These vapor-state inorganic compounds are transported through the boundary layer and finally deposited onto the target surface. As the thickness of deposited materials increases, the light signals of reflected beams from the film and metal surfaces undergo interference (7)-(10). These signals are monitored by the recorder. Figure 3 shows these interference signals. The growth rate of deposited film can be estimated from the peak to peak intervals.

The measuring procedures for the evaporation rates are as follows: $N_2$ in the premixed gas is changed from carrier gas to pure $N_2$ gas. Therefore, the aerosols of the seeded materials are not fed and then the deposited materials are evaporated by abruptly increasing the target temperature. As the evaporation process is started, the film thickness decreases and the interference signals are determined as the deposition process continues. We repeat these experiments changing both the target temperature and the concentration of aerosols in the seeded materials.

3. Optical Parameters

According to the target temperature, the phase of the films is either liquid or solid (e.g., the melting point of Na$_2$SO$_3$ is 884°C at atmospheric pressure). In each case, the interference signals monitored by the recorder are different, as shown in Fig. 3. In order to measure the growing film thickness (deposition rate measurement) and the decreasing film thickness (evaporation rate measurement) we require optical techniques. Using various measuring techniques, we investigate the following optical parameters in interferometry: 1) incident angle, 2) polarization state of incident beam, and 3) state of film (liquid or solid).

3.1 Incident angle

As Fresnel's law of reflection states, the intensity of reflected light is a function of incident angle and the state of incident polarization. We increase the incident beam angle as much as possible in order to increase the intensity of reflected light. Increasing the incident angle results in an increase in the optical path difference and an enlargement of the amplitudes of the interference signals.

3.2 Polarization state

Changes in the polarization state show the variation of reflected beam intensities and the mode of interference signals. Figures 3 and 4 show these variations as a change of two different incident polarization states. In the case of an S-polarized incident beam, the phase of a light wave changes by $\lambda/2$ at the film surface and by $\lambda/2$ at the metal surface, so that a total phase shift $\lambda$ occurs. This is initially accomplished by destructive signal interference. In the case of P-polarization, the $\lambda/2$ phase change occurs only at the film surface, and not at the metal surface. This phase change is initially accompanied by constructive signal

![Fig. 3 P-polarized interference signals (Na$_2$SO$_3$). Target temperature: a) $T_1=950°C$, b) $T_1=1130°C$, c) $T_1=1120°C$](image-url)
interference. A P-polarized incident beam always shows larger interference signals than those of an S-polarized incident beam. This P-type of polarization also enables the effect of target surface roughness to be reduced, i.e., we can measure the deposition and evaporation rates independent of surface roughness. As a result, we choose the P-polarized incident beam in this experiment.

3.3 State of film (liquid or solid)

As shown in Fig. 4, in the case of the liquid state, only one interference signal peak can be seen, i.e., beams become extinct in liquid film. In the case of the solid state, the interference signal peaks are repeated. This can be explained by noting that the extinction of the beam in the solid phase is smaller than that in the liquid phase.

4. Experimental results and Discussion

4.1 Deposition rates as a function of target temperature

By adopting P-polarization, a beam of higher intensity was attained. This enables deposition rate measurements in the range of lower target temperatures. These are shown clearly in Figs. 5 and 6, which display the deposition rate versus the target temperature. Figure 5 shows that the deposition rates are independent of the target temperature at very low target temperatures. However, in Fig. 6, the deposition rate decreases slowly with increasing target temperature up to the dew point and then sharply falls to zero. (We define this temperature as the dew point). We divide these curves into three regions according to the range of target temperature.

4.1.1 Range of very low target temperatures

The temperatures in this range are so low that the inorganic vapors are condensed to aerosols before reaching the target surface\(^{10(12)14}\). When this occurs, the mechanism of deposition is no longer vapor diffusion alone; the deposition is assisted by thermal diffusion of aerosols. The effect of thermal diffusion is substantially less than that due to Fick diffusion. Therefore, the contribution of the thermal diffusion effects does not outweigh the reduction of the vapor diffusion effect by phase change. There are two reasons for the flatness of the deposition curve: first, the condensed phase transport (e.g., Fick diffusion, thermal diffusion) reduces the deposition rate more than theoretical vapor diffusion alone, and second, in the vapor transport mechanism, the driving force is the difference between the partial pressure of the vapor at the mass boundary layer edge and the saturation pressure at the target temperature. On lowering the target temperature, the saturation pressure, being an exponential function of the target temperature, approaches zero (cf. Clapeyron equation), and the driving force becomes independent of the wall temper-

![Fig. 5 Na$_2$SO$_4$ deposition rates vs. target temperature ($T_{\text{sat}}$=1100°C)](image)

![Fig. 6 Na$_2$SO$_4$ deposition rates vs. target temperature ($T_{\text{sat}}$=1100°C). Solid lines represent the predictions by CFBL theory](image)


*JSME International Journal*
ature\textsuperscript{[14]}. Hence, a local thermochemical equilibrium calculation at the target surface requires simultaneous consideration of the transport laws.

4.1.2 Range of temperatures lower than the dew point
In this target temperature range, the deposition rates decrease with increasing target temperature as shown in Figs. 6 and 7. The trends of these curves can be predicted by the following theoretical analysis known as the CFBL theory\textsuperscript{[10][12]}. The assumptions of this analysis are listed below.

1) The mass fractions of inorganic compounds in the combustion gases are so small that the effects of inorganic vapors are neglected in the momentum and thermal boundary layers.

2) The seeded vapor is undersaturated in the mainstream. Furthermore, within the boundary layer, there are no chemical reactions and no phase changes.

3) The properties concerned are all constant.

4) Gas flow is laminar and the flow field can be described by the Hiemenz solution.

As long as the vapors in the mass boundary layer are undersaturated, Fick diffusion and phase change at the interface adjacent to the target surface are the basic mechanisms of deposition in the target temperature range. The following equations and boundary conditions can explain the trends of the experimental results.

○ Species conservation equation:

\[ u \frac{\partial \omega_y}{\partial x} + v \frac{\partial \omega_y}{\partial y} = D \frac{\partial^2 \omega_y}{\partial y^2} \]  

(1)

○ Boundary conditions:

\[ \omega_y = \omega^\infty(T_w) \text{ at } y = 0, \]

\[ \omega_y = \omega_y, y, x \text{ at boundary layer edge,} \]

where \[ \omega^\infty(T_w) = \frac{P_0 M}{\rho R T_w} \exp \left( \frac{-A}{RT_w} \right) \]

○ Deposition rate: \[ j_v'' = \rho D \frac{\partial \omega_y}{\partial y} \]  

(2)

In Eq. 1, the diffusion term in the \( x \)-direction is negligible compared to the diffusion term in the \( y \)-direction.

There are no sink or source terms due to assumption 2. Equation 2 is Fick's law.

The results of this theory for Na\textsubscript{2}SO\textsubscript{4} and K\textsubscript{2}SO\textsubscript{4} are represented by curves in Figs. 6 and 7, respectively, and compared with the experimental results. In these figures, the comparison of absolute values between the experimental and theoretical results is not possible because the experimental data consist of relative values. In this context, we feel that only qualitative comparisons are meaningful in the present study, and further detailed measurements would be required for a quantitative analysis. The apparent discrepancies between the experimental findings and the predictions in the temperature range of 1100\textdegree - 1170\textdegree C, as discernible in Fig 6, are unexpected results. These discrepancies are obvious regardless of whether these results are examined qualitatively or quantitatively. We tend to believe that these discrepancies point to the existence of condensate aerosols in the mainstream. Therefore, the possibility that the inertial effects of the condensate might be responsible for the deposition rate increment cannot be excluded.

Despite these discrepancies, we find that the trend of the experimental results fits well with expected ones. In the case of K\textsubscript{2}SO\textsubscript{4}, we cannot point out any outstanding discrepancies. From these comparisons, we ascertain that the interferometric technique is a powerful method that can be used to measure the growth of liquid films.

4.1.3 Range of temperatures near the dew point
In this target temperature region, the deposition rate sharply decreases. The cause of this sharp reduction in the deposition rate is caused by the fact that \( \omega^\infty \) is an exponential function of \( T_w \); consequently the curve of the deposition rate decreases rapidly as \( \omega^\infty \) increases exponentially.

We define the dew point as \( j_v'' = 0 \) and from Eq. 2 this condition is satisfied when \( \omega_{v,e} = \omega^\infty(T_w) \). Therefore, we can predict the dew point from the following equation:

\[ \omega_{v,e} = \frac{P_0 M}{\rho R T_{dp}} \exp \left( \frac{-A}{RT_{dp}} \right) \]  

(3)

When the target temperature is higher than the dew point, evaporation phenomena take place rather than deposition. The evaporation rates versus target temperature are represented in Fig. 8. The dew points in Figs. 6 and 8 are different because during the experiment illustrated in Fig. 8, the carrier gas of inorganic compounds is shut down (the seeded material aerosols are not supplied into the burner).

4.2 Deposition rates as a function of mass fractions
Mass fractions of inorganic compounds are deter-
Fig. 8 Na$_2$SO$_4$, K$_2$SO$_4$ evaporation rates vs. target temperature ($T_{\text{set}}=1100^\circ\text{C}$)

determined by a light-scattering method. Because mass fraction cannot be measured quantitatively by this method, we measure the mass fraction relatively. Equation 2 includes the fact that the predicted deposition rates have linear relations with the main stream mass fraction, $\omega_{V,e}$, for fixed target temperatures. Experimental results (see Figs. 6 and 7) show that the deposition rates increase with the increasing mass fraction of inorganic compounds (large intensities of the light scattering signal means high mass fraction of the seeded materials).

5. Conclusions

(1) Measuring the growth rates of condensed liquid film by interferometric technique, we could determine the deposition rates, on a 30-second time scale, well before the onset of surface flow phenomena.

(2) Optical parameters are important in evaluating the variations in the amplitude of the interference signal and the effects of target surface roughness.

(3) There are three different trends for deposition rates, according to the surface temperature. Especially at lower surface temperatures, the condensation of vapor to droplets occurs before the vapor reaches the surface. Thermal diffusion of condensed aerosols results in low deposition rates and tends to reduce the deposition rates below the theoretical level.

(4) Dew points are shifted with changes in the densities of inorganic compounds and evaporation rates.

The authors acknowledge the Korea Science and Engineering Foundation for their support of this research.

References


