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MEASUREMENT OF LOCAL MASS-TRANSFER COEFFICIENTS BY HOLOGRAPHIC INTERFEROMETRY

H. GROSSE-WILDE* and J. UHLENBUSCH†
Physikalisches Institute II der Universität Düsseldorf, Universitätsstr. 1,4, Dusseldorf, West Germany

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Abstract—Double exposure holography as a tool for profilometric measurements on coated surfaces is described. The coating material is camphene, a transparent, crystalline substance of high vapour pressure at room temperature. The application of the technique to measurements of local mass-transfer coefficients under the condition of forced and free convection along a flat plate is shown. The results agree very well with the theoretically predicted values. The technique can also be used to analyze the flow field by means of the local mass transfer. As an example the longitudinal vortex instabilities were visualized and their influence on mass transfer was measured.

NOMENCLATURE

- D, diffusion coefficient of camphene vapour in air;
- Gr_x, local Grashof number;
- h', change in the thickness of the evaporated layer between two adjacent interference fringes;
- $h(x)$, total thickness of evaporated layer at position x;
- k_x, local mass-transfer coefficient;
- M_c, molecular weight of camphene;
- n, refractive index of camphene;
- p, vapour pressure of camphene;
- R, molar gas constant;
- Re_x, local Reynolds number;
- Sc, Schmidt number;
- Sh_x, local Sherwood number;
- St_x, local Stanton number;
- T, thermodynamic temperature;
- t_1, t_2, moments of exposures of the holographic plate;
- x, coordinate in the plane of the flat plate parallel to flow direction, measured from the leading edge;
- x_0, mass transfer free starting length of the flat plate;
- Δn, difference between refractive index of camphene and air;
- Δt, mass-transfer time.

Greek symbols

- λ, laser wavelength;
- ρ_c^s, density of solid camphene.

1. INTRODUCTION

THE MEASUREMENT of local mass-transfer coefficients related to stationary flow problems may be carried out in two ways: (i) the first method is to measure the local mass flux from the surface. This is done for example using the electrochemical method [1], where in a suitable electrolyte under certain conditions the mass flux density is proportional to the current density. (ii) A second way is to measure the deformation of a surface in a given time interval, when the deformation is induced by sublimation from the surface. This method is used for example in profilometric measurements on naphthalene coated surfaces [2].

An improvement of the profilometric method may be obtained, when the local surface deformation is measured by optical interference. The advantage of this method would be a higher resolution in the deformation measurement up to a tenth of a wavelength, contact free measurements and a large visual field in one interferogram.

Classical interferometers of Michelson or Mach–Zehnder type are not applicable for those measurements, because the good optical quality coating of a body with a sublimating substance is not feasible. In holographic interferometry however, there is no need for such a quality of the coating. Using for example, the double exposure technique the fringes in the resulting interferogram are only due to the changes of the object during the time between the two exposures. Optical inhomogeneities in the light path do not produce fringes as in classical interferometers.

Kapur and MacLeod [3] first used holographic interferometry for measurements of mass-transfer coefficients. They coated the surface under study with an elastomer, which was charged with a volatile swelling agent. The shrinkage of the elastomer caused by mass transfer was measured by holographic interferometry. However it is supposed that there is a systematic error in their measurements, as the position of the interference fringes is effected by diffusion of the swelling agent parallel to the surface from regions of low mass transfer to regions of high mass transfer.

In order to avoid this systematic error in this work a coating material was used which sublimated by itself from the surface similar to naphthalene in the naphthalene sublimation technique. As an application of this diagnostic method the local mass transfer of camphene from a flat plate due to forced and free

*Present address: Siemens AG, 852 Erlangen, West Germany.

convection was investigated. Additionally it was studied, how longitudinal vortex instabilities in the transition region from laminar to turbulent boundary layer influence the mass transfer.

2. MEASURING TECHNIQUE

Local changes in the thickness of a naphthalene layer induced by sublimation can hardly be measured with holographic interferometry. The reason is that also the polycrystalline structure of the surface is changed by the sublimation process; thus the information of the surface deformation is buried in the speckle pattern of the reconstructed image.

Therefore we looked for a substance, which is transparent in the visible, optically isotropic, easy to handle for coating of a test body and with a vapour pressure at room temperature of about 150 N/m^2. We found that camphene, a hydrocarbon with the chemical formula $C_{10}H_{16}$, met all desired conditions. Camphene is in the solid phase a transparent colourless substance with a wax-like feeling. It crystallizes below 49°C in the cubic form, so that it is optically isotropic.

The best results in coating we get by vapour deposition in vacuo. For this purpose the test body was cooled down to 0°C. Then it was placed into a vacuum vessel with a small reserve of camphene. After heating the vessel up to 35°C, the test body was uniformly coated with a transparent layer of thickness 0.5–1 mm within 35–45 min.

Measurements of deformations on the camphene coated surface of the body caused by sublimation were performed by the double exposure holographic technique. Changes in the optical path of the object beam between the first exposure at time t_1 and the second exposure at time t_2 give reason to interference fringes.

The optical arrangement for recording the holograms is shown in Fig. 1. The laser beam after passing a shutter S was divided by the beamsplitter BS into the object and the reference beam. The reference beam was directed to the photographic plate by the mirrors M_1 and M_2 and expanded by the lens L_1. The object beam illuminated the transparent camphene coated object using the mirror M_3 and the lens L_2. In our measurements the object was a flat glass plate. In order to localize the interference fringes on the plate, the illuminated side of the plate was sandblasted. The other side of the plate was coated with camphene. The diffuse light passed through the layer of camphene and was stored on the photographic plate as a hologram by interference with the reference beam.

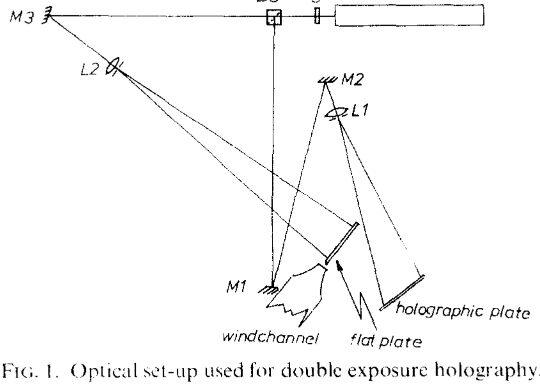

FIG. 1. Optical set-up used for double exposure holography.

By making a double exposure hologram, the first exposure stores light, which passed the camphene coating at time t_1. The second exposure stores light, which passed the coating at time t_2. Both light fields differ by having passed a coating of different thickness. Therefore the phase of the light field is different in both exposures and the phase difference is proportional to the thickness of the evaporated sheet.

By illuminating the developed photographic plate with the reference beam, both light fields are reconstructed simultaneously. The reconstructed object appears with interference fringes, which show the local phase differences between both exposures. From these interference fringes the thickness of the evaporated camphene layer can be calculated. Between two fringes the variation h' in the thickness of the evaporated coating is given by

$$h' = \frac{\lambda}{\Delta n} \tag{1}$$

A conservative value for the resolution in an interferogram is 0.1 fringe. Assuming $\lambda = 0.5145\ \mu m$ and $\Delta n = 0.500$ one yields a resolution of 0.1 μm for this technique.

3. PHYSICAL PROPERTIES OF CAMPHENE

Sufficient results we get with camphene in the grade "for synthesis" delivered by Merck–Schuchardt. The physical properties of camphene, needed to evaluate the interferograms and to compare the experimental results with theory, are listed in Table 1.

The refractive index at the wavelength of the argon-ion laser of $\lambda = 0.5145\ \mu m$ was measured using a camphene coating on a glass substrate. By measuring the angle of total reflection at the boundary between glass and camphene, the refractive index of camphene was found. The temperature dependence of the vapour pressure of camphene was derived from the vapour pressure at 5°C (133.3 N/m^2) and at 47.2°C (1333 N/m^2) [6], using the interpolation formula $\log_{10} p = -A/T + B$. The diffusion coefficient of the system camphene–air was calculated from kinetic theory [7] and in a second independent way by using an empirical formula from Fuller, cited in [8]. The mean value of both results is listed in Table 1.

Table 1. Physical properties of camphene [4–6]

Chemical formula	$C_{10}H_{16}$
Molecular weight	$M_c = 136.24 \cdot 10^{-3}$ kg/mol
Refractive index (22°C, $\lambda = 0.5145\ \mu m$)	$n = 1.500$
Density of solid (22°C)	$\rho_s = 0.87 \cdot 10^3$ kg/m^3
Vapour pressure	$\log_{10} p(T) = 9.7161 - (2111.5/T)$ $p[N/m^2]$, $T[K]$
Diffusion coefficient (in air, 22°C)	$D = 6.13 \cdot 10^{-6}$ m^2/s
Schmidt number (in air)	$Sc = 2.46$

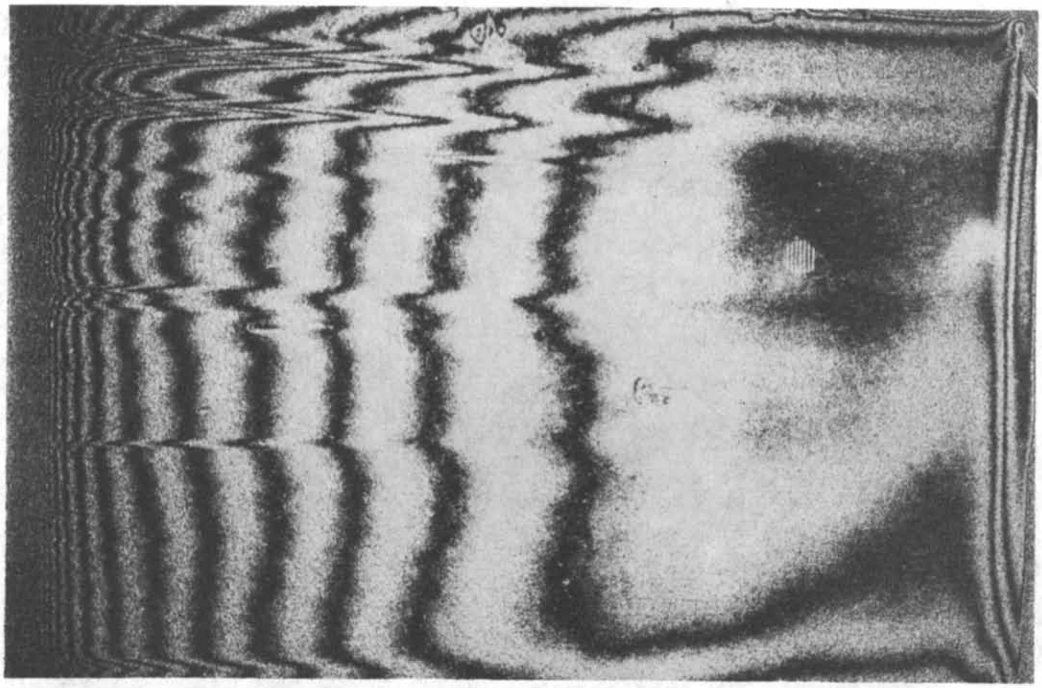

FIG. 2. Holographic interferogram of forced convection mass transfer from a flat plate. Flow direction from the left with a velocity of 9 m/s.

4. MEASUREMENTS AND RESULTS

This diagnostic method was used to study mass transfer under the following three experimental situations:

4.1. *Forced convection*

The measurements of the mass transfer with forced convection were performed with a free stream of air along a flat plate. The flow velocity was variable up to 15 m/s. The turbulence level was measured to be below 1%. Following Thomas [9] a level below 2.8% gives the same mass-transfer coefficient as the turbulence free case.

The flat plate consisted of a glass plate which was held by a frame of steel with a sharp leading edge. Only the glass plate was coated with camphene. Therefore the concentration boundary layer began after a mass transfer free starting length $x_0 = 0.0144$ m from the leading edge.

Interferograms were taken with mass-transfer times from 15 to 40 s and flow velocities of 9 and 14 m/s. The mass-transfer time is defined as the time interval during which the plate is exposed to the flow field. During the exposures the wind channel was switched off to drop changes in the optical path due to density variations in the surrounding air of the holographic set up.

A double exposure hologram is shown in Fig. 2. The glass plate is blown from the left with a velocity of 9 m/s. The mass-transfer time was 20 s. The intervals between successive interference fringes are growing downstream as predicted by mass-transfer theory of the laminar boundary layer on a flat plate [see below equation (2)]. In the leading part of the coated plate the fringes are buried in speckles. At the edges and at the end of the plate the fringes show effects due to the boundary between the glass plate and the frame.

A general problem is to recognize the ordinal number of the fringes. Here we proceeded in the following way: The shift of the fringe of lowest order—the last fringe at the trailing edge of the interferogram—was calculated using equation (2). Starting from this fringe the thickness profile of the sublimated layer could be determined for all fringes using equation (1). Interferograms with different mass-transfer times but the same flow velocity are taken as a control for the right choice of the ordinal number of the fringe of lowest order. This is reasonable because the ordinal number of fringes at a fixed position on the plate is proportional to the mass transfer time. In Fig. 3 the results from three interferograms are drawn in a similarity plot. The flow velocity was 9 m/s, the mass-

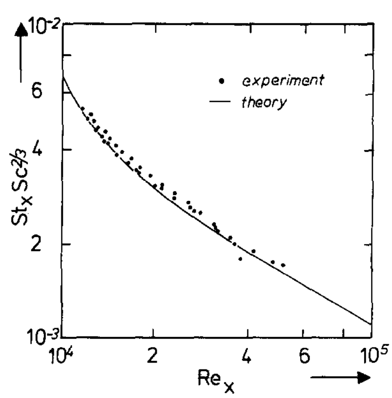

FIG. 3. Similarity plot of the results derived from three interferograms with forced convection mass transfer. ——, theory [equation (2)].

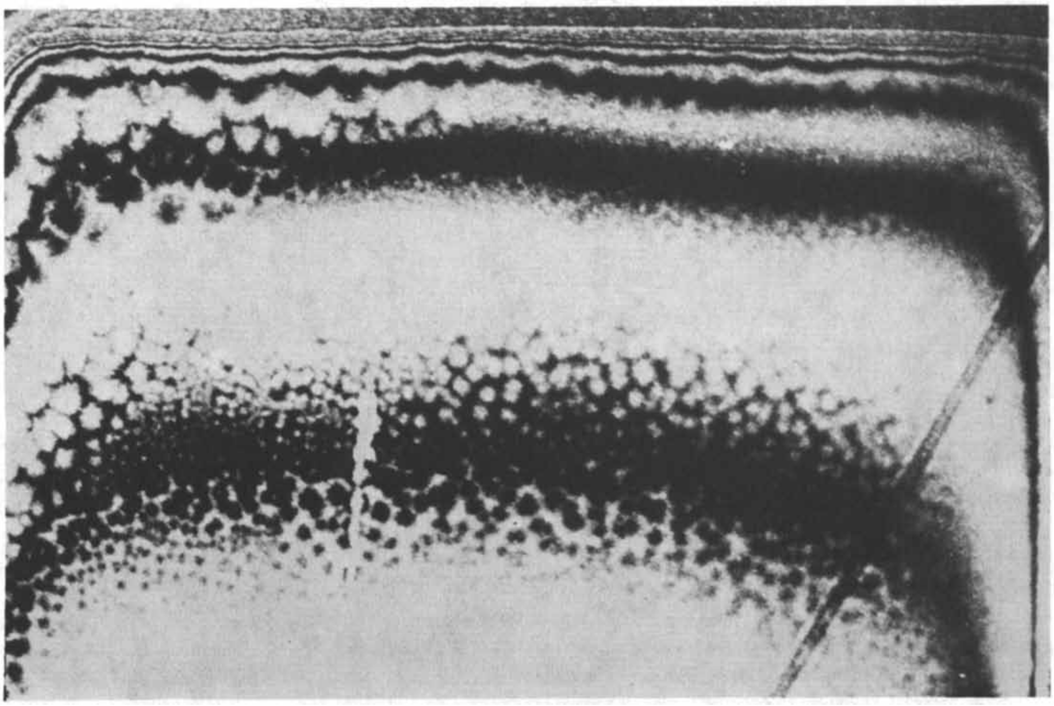

FIG. 4. Holographic interferogram of free convection mass transfer.

transfer times were 20, 30 and 40 s. Each dot represents the evaluated mass-transfer for one fringe.

The theoretical curve was calculated by the formula

$$St_x \cdot Sc^{2/3} = \frac{0.332}{Re_x^{1/2}}\left[1-\left(\frac{x_0}{x}\right)^{3/4}\right]^{-1/3} \quad x > x_0 \quad (2)$$

which is analogous to the formula for the corresponding heat-transfer problem [10]. The correction factor $[1-(x_0/x)^{3/4}]^{-1/3}$ is due to the fact that the hydrodynamic boundary layer starts with $x = 0$, while the concentration boundary layer starts with $x = x_0$.

This correction factor is derived from the von Karman–Pohlhausen approximation for the mass transfer from the flat plate, see e.g. [8].

The experimental results agree very well with the theoretical curve after equation (2) within the error limits of, say, 6.5%. They systematically exceed the theoretical values within a few percent. Probably the reason for this is an error in the diffusion coefficient or in the vapour pressure of camphene.

4.2. *Free convection*

Experiments were also performed to measure the free convection mass-transfer from a vertical flat plate. In front of the surface of a camphene coated plate a downward convection is induced by sublimated camphene leading to a locally varying mass transfer. The thickness of the sublimated layer during a given time was measured by the technique described above. In order to get a sufficient number of fringes mass-transfer times were chosen between 120 s and 195 s. As in the experiments with forced convection the interferograms with different mass-transfer times served to approve the calculated fringe shift of the fringes of lowest order in the interferograms. Figure 4 shows a double exposure hologram with a mass-transfer time of 120 s. At the edges of the plate an enhanced mass transfer can be observed in comparison with the middle of the plate. This is due to the fact that air without any contamination by camphene streams from the sides to the surface of the plate. The larger density gradient at the boundary of the plate induces an enhanced mass transfer.

To evaluate the holograms the thickness of the sublimated layer $h(x)$ at the position x was calculated from the order of the fringes as in the case of forced convection described above. In analogy to Section 4.1 all results can be summarized by using dimensionless numbers. With the mass-transfer time Δt the local Sherwood number $Sh_x = k_x \cdot x/D$ is given by

$$Sh_x = \frac{\rho_c^s \cdot R \cdot T \cdot h(x)}{M_c \cdot p \cdot \Delta t} \cdot \frac{x}{D}. \quad (3)$$

In Fig. 5 the results of four interferograms are plotted vs the local Grashof number Gr_x.

An approximate solution of mass transfer from a vertical flat plate, which is given by Eckert [11] for the corresponding heat-transfer problem gives the following relation between Grashof and Sherwood number

$$Sh_x = 0.508 \cdot Sc^{1/2}[0.952 + Sc]^{-1/4} \cdot Gr_x. \quad (4)$$

The evaluation of this formula is drawn in Fig. 5 as a solid line.

4.3. *Boundary-layer instabilities*

In the transition region from the laminar to the turbulent boundary layer of a flat plate periodical instabilities are observed. Predominantly

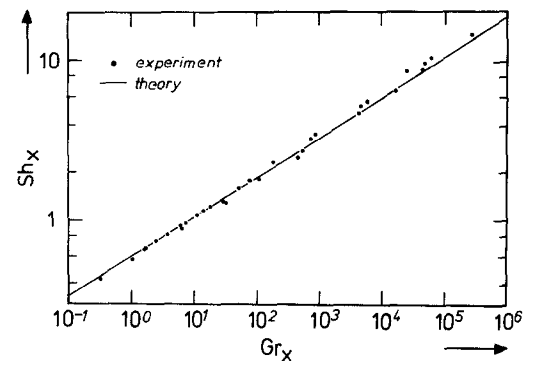

FIG. 5. Similarity plot of the results derived from four interferograms with free convection, ——, theory [equation (4)].

Tollmien–Schlichting waves [10(pp. 438–446)] and longitudinal vortex instabilities [12] occur. The Tollmien–Schlichting waves are periodical in the direction of the outer flow. The longitudinal vortices have their axis parallel to the flow and they arise at higher Reynolds numbers than the Tollmein–Schlichting waves, that means at larger distance from the leading edge [13]. With our experimental set up the mass transfer in the transition region could only be measured by inclining the plate against the outer flow. By this a pressure gradient was induced which shifted the transition region towards the leading edge of the plate [10] and into the field of view of the interferograms.

Interferograms with an inclination angle of the flat plate between 1° and 3° show a periodical variation in the fringe pattern. As an example an interferogram with an inclination angle of 2° is shown in Fig. 6. The structure in the fringe pattern is corresponding to a variation in the mass-transfer coefficient vertical to the flow. For interpretation of these interferograms a model was made to calculate the influence of the longitudinal vortices on mass transfer [14]. The results approve that the periodical structure in the interferograms had to be accounted for the longitudinal vortices in the boundary layer.

Going beyond an angle of 3° the periodical structure vanished and the mass-transfer rate grew. This indicates a higher degree of instability and thus a further step to the turbulent boundary layer.

5. CONCLUSION

All examples of mass transfer at forced and free convection given here show that the camphene sublimation technique in connection with holographic interferometry is very well suited for measuring local transport coefficients. The main advantage compared with other techniques is the fact that the thickness of the sublimated layer can be measured with an accuracy of 0.1 μm continuously over the entire holographically observed field. Additionally the boundary layer is not disturbed by probes.

Because the measurement is carried out over the entire plate, local deviations of the mass-transfer coefficient from the mean value are easily recognized. As an obvious example for this the longitudinal vortex instabilities can be visualized as shown in Fig. 6.

The application of this technique to mass-transfer measurements on bodies of complicated size is straightforward. However the evaluation of the holograms will not be as easy to handle as in the case of the flat plate.

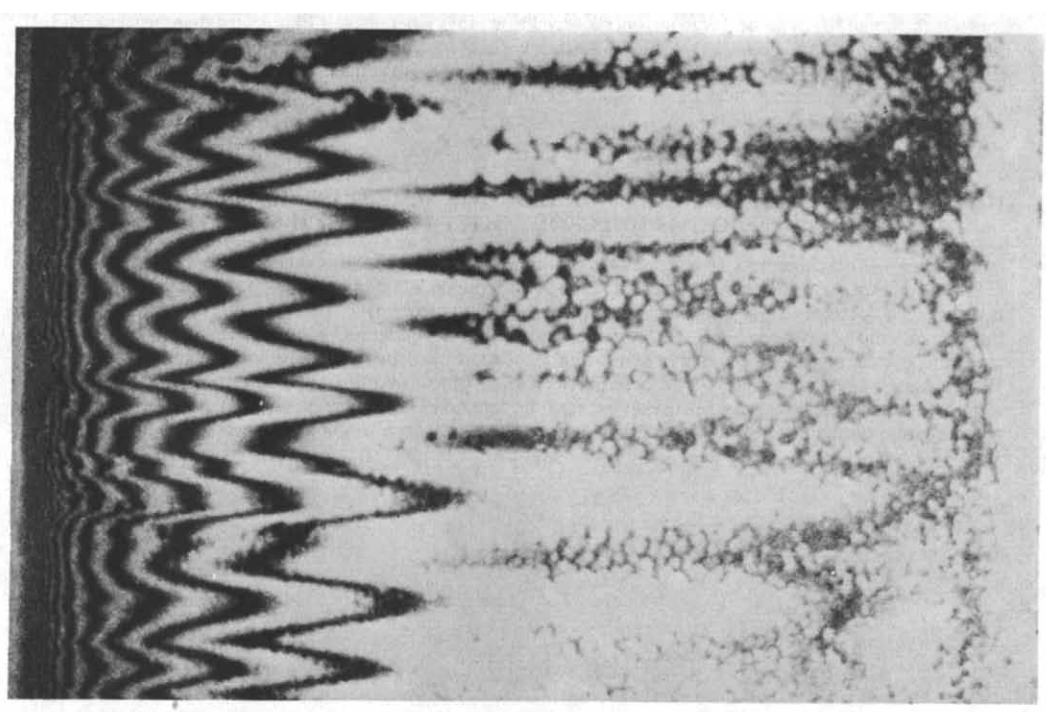

FIG. 6. Holographic interferogram of forced convection mass transfer from an inclined flat plate. The periodical structure shows the influence of longitudinal vortices on mass transfer.

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MESURE PAR HOLOGRAPHIE INTERFEROMETRIQUE DES COEFFICIENTS LOCAUX DE TRANSFERT MASSIQUE

Résumé—On décrit l'holographie à double exposition utilisée pour des mesures de profils sur des surfaces couvertes. Le matériau de recouvrement est le camphène, substance cristalline, transparente à grande tension de vapeur à la température ambiante. On montre l'application de la technique à la mesure du coefficient local de transfert massique dans les conditions de convection naturelle ou forcée le long d'une plaque plane. Les résultats s'accordent très bien avec les valeurs théoriques. La technique peut aussi être utilisée pour analyser le champ de vitesse par l'intermédiaire du transfert massique local. Par exemple, les instabilités de tourbillon longitudinal sont visualisées et leur influence sur le transfert massique est mesurée.

Zusammenfassung—Es wird ein Verfahren beschrieben, die holographische Doppelbelichtungsinterferometrie zur Messung der Schichtdickenänderung einer beschichteten Oberfläche zu benutzen. Das Beschichtungsmaterial ist Camphen, eine transparente, kristalline Substanz mit hohem Dampfdruck bei Zimmertemperatur. Die Anwendung des Verfahrens zur Messung des lokalen Stofftransportkoeffizienten bei freier und erzwungener Konvektion längs einer ebenen Platte wird demonstriert. Die Meßergebnisse bastätigen die theoretisch erwarteten Werte. Das Verfahren kann auch benutzt werden, um ein Strömungsfeld mit Hilfe des Stofftransports zu untersuchen. Als Beispiel dafür wurden die Längswirbel der Strömungsgrenzschicht sichtbar gemacht und ihr Einfluß auf den Stofftransport gemessen.

ИЗМЕРЕНИЕ КОЭФФИЦИЕНТОВ ЛОКАЛЬНОГО МАССОПЕРЕНОСА С ПОМОЩЬЮ ГОЛОГРАФИЧЕСКОЙ ИНТЕРФЕРОМЕТРИИ

Аннотация — Описывается голография с двойным экспонированием, используемая для профилометрических измерений поверхностей с покрытием. В качестве материала покрытия применялся камфен (прозрачное кристаллическое вещество) с большим давлением паров при комнатной температуре. Излагается методика измерения коэффициентов локального массопереноса в условиях вынужденной и свободной конвекции вдоль плоской пластины. Результаты хорошо согласуются с теоретическими расчетами. Данная методика приемлема и для анализа поля скоростей течения при локальном массопереносе. В качестве примера визуализировались продольные вихревые неустойчивости и измерялось их влияние на перенос массы.