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Application of PSP to Low Density Gas Flows

Mori, H.*, Niimi, T.*, Yoshida, M.*, Kondo, M.*, and Oshima, Y.*

* Department of Mechanical Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya, 464–8603, Japan.
E-mail: hmori@mech.nagoya-u.ac.jp

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Abstract : Recently optical pressure measurement systems using pressure sensitive paints (PSP) have actively developed to measure continuous pressure distributions on solid surfaces. However, the pressure range has been almost limited above 130 Pa (about 1 Torr) and there is no application to lower pressure range because the pressure sensitivity seems to be not so high in that range. In this study, we have applied three types of PSP [two types are composed by organic dye and polymer (luminophore/binder); PtOEP/GP197 and PtTFPP/poly(TMSP), and another one is Bath-Ru adsorbed on anodized aluminum] to the rarefied gas flow mainly lower than 130 Pa and examine those fundamental properties such as pressure sensitivity, leading to selection of the most suitable PSP among them for the low-pressure range. Since PtTFPP/poly(TMSP) has the highest sensitivity, it is applied successfully to the measurement of the two-dimensional pressure distribution on the surface interacting with a low density supersonic free jet.

Keywords : PSP, Low density gas flow, Stern-volmer plot, Compressible flow

1. Introduction

In the recent development of nanotechnology, rarefied gas flows become more important, related to the aerospace science, formation of the thin film for semiconductors and so on. Rarefied gas flows with high Knudsen number cannot be analyzed by similar methods used for analyses of continuum flows, because the intermolecular collision rate is extremely small and effects caused by molecular motions are significant. Therefore, diagnostic tools based on process in the molecular level are required for analyses of rarefied gas flows. Moreover, flows in microsystems with small dimension have also high Knudsen number and they must be treated as rarefied gas flows. Especially, there may be no detailed information yet about the behavior of a molecule in the vicinity of the solid surface and the energy exchange between molecules and the surface, motivating development of new measurement techniques in the molecular level.

Recently optical pressure measurement systems using pressure sensitive paints (PSP) have actively developed to measure two-dimensional pressure distributions on solid surfaces (Kavandi et al., 1990, Engler et al., 1991, Liu et al., 1997). PSP is composed of luminescent molecules (luminophore) whose luminescence is easily quenched by oxygen and a binder to fix the luminescent molecules to a solid surface. Because the luminescence intensity depends on the oxygen partial pressure on the surface, the surface pressure can be measured by analyzing the luminescence intensity of PSP. The PSP technique has enabled to measure two-dimensional pressure distributions on solid surfaces with extremely low cost and also pressure on objects with complex shape, which cannot be measured by using pressure taps and transducers. Up to now, the pressure range has been almost limited above 1 Torr (130 Pa) and there is no application to lower pressure range because the pressure sensitivity seems to be not so high in that range. It is because polymers used as binders do not have sufficient oxygen permeability, and the change of luminescent intensity of the PSPs is small for the small change of surface pressure. However, conventional method using pressure taps and transducers cannot be utilized for measurements of pressure distributions on solid surfaces interacting with low density gas flows, because the conductance of the pressure taps limits the sensitivity and accuracy. On the other hand, PSP techniques are based on the process in the molecular level such as quenching of luminescence of organic luminophores, and they have potential for applications in high Knudsen number regime (low density gas flows).

For applications of PSP techniques to low-pressure range, it is important to develop a new PSP using a binder with extremely high oxygen permeability. In this study, three types of PSPs are examined in low-pressure range below 150 Pa, and the most suitable PSP is proposed by clarifying the feasibility and problems of PSPs. Moreover, the pressure distribution on the surface interacting with an impinging supersonic free jet is measured by the PSP.

2. Properties of Pressure Sensitive Paints

Optical pressure measurement systems using PSP are based on oxygen quenching of luminescent molecules (Engler et al., 1991, Liu et al., 1997). A PSP is composed of luminescent molecules and a binder material to fix the luminescent molecules to a solid surface. The luminescent molecules at the ground singlet state can be excited by absorption of photon energy to a higher singlet state. After the transition of the excited molecules from the singlet state to the lowest triplet state by an intersystem crossing, the molecules emit phosphorescence and transfer to the ground singlet state. Because the transition between the triplet state and the singlet state is spin forbidden, the lifetime of the phosphorescence is long. On the other hand, oxygen molecules with triplet ground state act as a quencher of the luminescence. As a result, the phosphorescence intensity decreases as an increase in partial pressure of oxygen.

The dependence of the luminescence intensity on partial pressure of oxygen is described by the Stern-Volmer relation. The ratio of the luminescence intensity I to I_0 in the absence of oxygen is given by (Liu et al., 1997)

$$\frac{I_0}{I} = 1 + KP, \tag{1}$$

where K is the coefficient depending on the temperature, and P is the pressure. It is usually difficult to measure I_0 to calibrate the PSP, but if the luminescence intensity of the PSP I_{ref} at the reference pressure P_{ref} is known, the pressure P can be obtained by the relation between inverse intensity ratio I_{ref}/I and P/P_{ref} that is deduced from Eq. (1) as

$$\frac{I_{ref}}{I} = A_0(T) + A_1(T)\frac{P}{P_{ref}}. \tag{2}$$

In practice, the following equation taking the nonlinearity into consideration is used:

$$\frac{I_{ref}}{I} = \sum_{n=0}^{N} A_n(T)\left(\frac{P}{P_{ref}}\right)^n. \tag{3}$$

The coefficients A_n can be deduced by calibration tests, and usually a second-order polynomial ($N = 2$) is used. The coefficients A_n in Eq. (3) have the dependence on temperature, which may cause errors in pressure measurements using PSP. The temperature dependence of A_n is due to the luminescence quenching of the luminophore and the dependence of the oxygen permeability of the polymer binder on the temperature (Liu et al., 1997).

3. Compositions of PSPs

To select a PSP suitable for low-pressure conditions, three types of PSPs are examined. Two of them are composed by organic dyes and polymers (luminophore/binder); PtOEP/GP197 and PtTFPP/poly(TMSP), and the other one is Bath-Ru adsorbed on an anodized aluminum (AA) surface developed by Asai et al. (1997).

PtOEP/GP197 is one of the first-generation PSPs developed at the University of Washington and it has been widely used (Kavandi et al., 1990, Fujimoto et al., 2001). However, it has many disadvantages such as low sensitivity and slow response; that is mainly because the polymer GP197 has low permeability of oxygen. PtOEP/GP197 is tested only to clarify that it is not suitable for applications in low pressure.

Bath-Ru/AA (anodized aluminum) is prepared by dipping an anodized sample made of aluminum into a solution of Bath-Ru [Ruthenium II tris (4,7- diphenyl-1, 10-phenanthrolin) chloride] in an organic solvent. Bath-Ru is adsorbed on a porous surface of anodized aluminum. Bath-Ru/AA has many advantages compared to some primitive PSPs such as PtOEP/GP197; it has fast response time and capability even in a cryogenic conditions (Sakaue et al., 2001, Asai et al., 2002). However, it can be applied only to aluminum or aluminum alloys.

For PtTFPP/poly(TMSP), organic dye PtTFPP is fixed by a glassy polymer poly(TMSP) with extremely high oxygen permeability. The PSP has high advantages such as high sensitivity, fast response time, and capability of measurement in cryogenic conditions (Asai et al., 2002, Egami et al., 2001). Moreover, PtTFPP/poly(TMSP) can be applied to any materials, unlike Bath-Ru/AA.

4. Experimental Apparatus

Figure 1 shows the experimental apparatus composed for this study. The PSPs are painted on aluminum plates (50mm×25mm×1mm) and then the plates are set inside a vacuum chamber evacuated by a rotary pump (ULVAC D-950) and a turbo molecular pump (ULVAC UTM-300). In this study, pure oxygen is used as a test gas to acquire high sensitivity. The test gas is supplied into the chamber and the pressure is monitored by a capacitance manometer (ULVAC CCMT-10A) and an ionization vacuum gauge (ULVAC GI-1000). The temperature of the PSP sample is controlled by a Peltier thermo-controller and is monitored by a thermocouple. A sonic nozzle can be attached in the chamber for measurements of the pressure distributions on the surfaces interacting with supersonic free jets, and the source pressure P_0 is measured by a U-shaped mercury manometer.

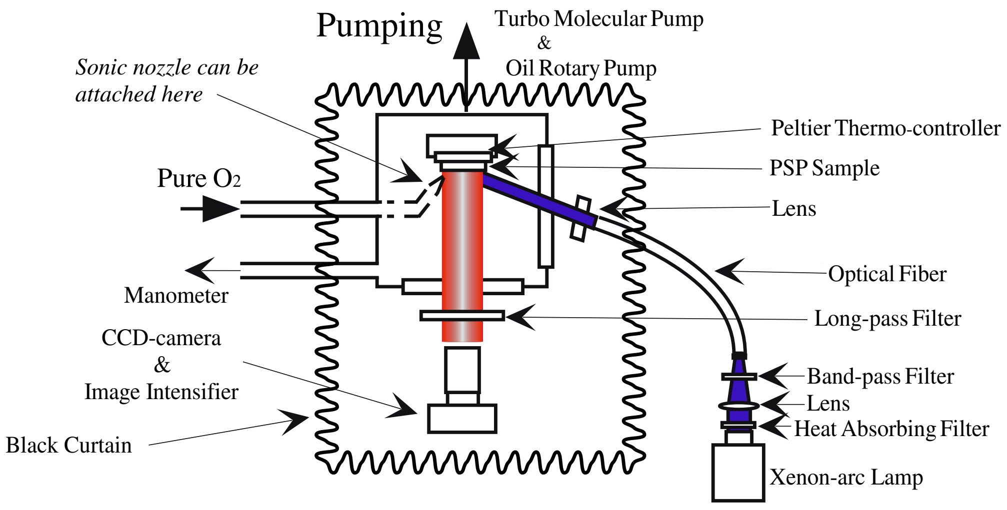


Fig. 1. Experimental apparatus.

A xenon-arc lamp (Ushio UXL-500SX) with a band-pass filter is used as an excitation light source, and the light is transmitted via an optical fiber to illuminate the sample in the vacuum chamber. The wavelength ranges of band-pass filters are 380±20nm for PtOEP/GP197, 450±10nm for Bath-Ru/AA, and 400±10nm for PtTFPP/poly(TMSP), which cover one of the intense absorption bands of each luminophore.

The luminescence is filtered by a long-pass filter (600nm) to eliminate the light from the xenon lamp, and is detected by a CCD camera (Hamamatsu, C7300-10) with an image intensifier (Hamamatsu, C6653). The optical system including the vacuum chamber and the CCD-camera is covered with a black curtain to shut out stray lights from the outside of the system. The image of the luminescence is processed by a personal computer. The CCD camera has spatial resolution of 1280×1024 pixels and intensity resolution of 12bit. To obtain the Stern-Volmer plot of each PSP, the luminescence intensity is averaged over 200×200 pixels of the luminescence image, which is equivalent to the area of 10mm×10mm of each sample.

5. Results and Discussions

5.1 Stern-Volmer Plots of the PSPs

Figure 2 shows the Stern-Volmer plots of PtOEP/GP197, Bath-Ru/AA, PtTFPP/poly(TMSP) at low pressure below 150 Pa. The horizontal axis is the pressure on the sample surface, and the vertical axis is I_{ref}/I. The surface temperature of the samples is kept at 300 K (but PtOEP/GP197 is kept at 293 K). The pressure ranges are 1.0×10^{-2}-151 Pa for PtOEP/GP197, 1.0×10^{-2}-152.5 Pa for Bath-Ru/AA, 2.7-153.6 Pa for PtTFPP/poly(TMSP). The increment of the pressure follows "1, 1.5, 2, 3, 5, 8" sequence, except 40 and 60 Pa instead of 50 Pa for PtOEP/GP197. The reference pressure P_{ref} of each datum is set at the lower limit of the pressure range.

It is clearly seen from Fig. 2 that the luminescence intensity of PtOEP/GP197 has no dependence on the pressure. This may be attributed to insufficient gas permeability of the polymer GP197, indicating that PtOEP/GP197 is not suitable for pressure measurement in the low-pressure condition below 150 Pa.

The luminescence intensity ratio I_{ref}/I of Bath-Ru/AA depends linearly on the pressure ratio P/P_{ref} at the pressure above 20 Pa, but the nonlinear dependence appears below 20 Pa. The nonlinear dependence makes the calibration of Bath-Ru/AA difficult. It should also be mentioned that the absolute luminescence intensity of Bath-Ru/AA is relatively low, resulting in the low signal-to-noise ratio. For these reasons it cannot be applied to quantitative measurements in low-pressure regime.

PtTFPP/poly(TMSP) has the highest sensitivity among the three types of paints examined in this study, and the luminescence intensity depends linearly on the pressure in the whole range of the pressure. Moreover, the absolute luminescence intensity of PtTFPP/poly(TMSP) is the highest

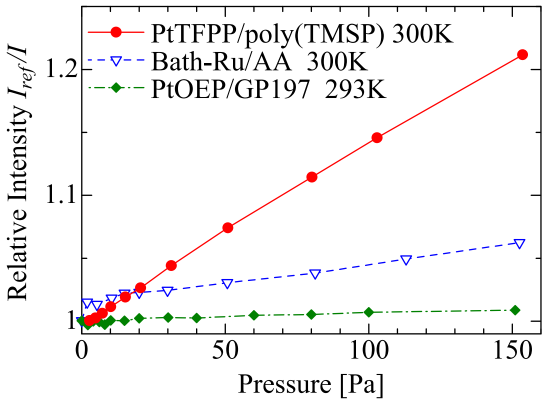


Fig. 2. Stern-Volmer plots for PtOEP/GP197, Bath-Ru/AA, PtTFPP/poly(TMSP).

among the three types of PSPs, resulting in the highest signal-to-noise ratio. These properties enable the accurate measurements of pressure distribution by the PSP in such a low-pressure region. Because of the large free volume (Nakagawa, 1995), oxygen molecules can permeate more easily through polymer matrix. From the results we have concluded that PtTFPP/poly(TMSP) is the most suitable PSP for quantitative measurements of pressure distribution on surfaces interacting with rarefied jets.

5.2 Measurement of Pressure Distribution on Jet-impinging Surface

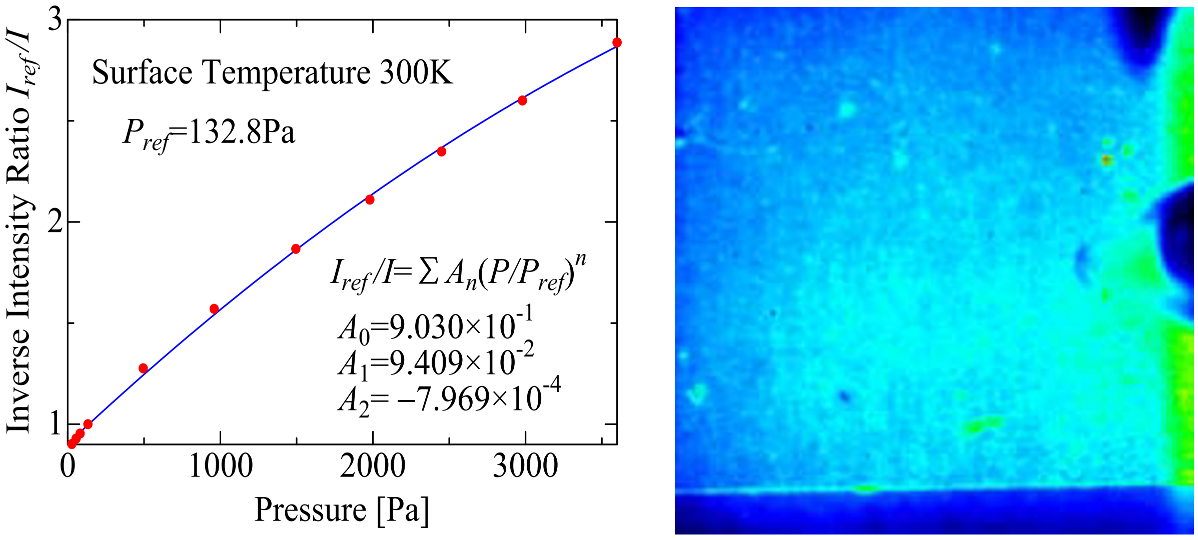
We have applied PSPs to measurement of pressure distribution on a solid surface interacting with a supersonic free jet. In the experiments, a sonic nozzle (simple convergent nozzle) made of glass was used to generate supersonic free jet of oxygen gas. The exit diameter of the nozzle was 0.3 mm. The impinging angle and the distance from the nozzle exit to the surface were set to 60° and 2 mm, respectively. PtTFPP/poly(TMSP) was used for the experiment and the temperature of the solid surface was kept at 300 K. We did not compensate for the luminescence non-uniformity caused by the nonuniform temperature distribution on the surface, because the density of the jet impinging on the surface is low enough and the temperature change caused by energy exchange between jet molecules and the surface can be ignored. The effect of the displacement of the surface was also ignored in this study, because rarefied jets with extremely low density cause very little aerodynamic forces.

The procedure to obtain pressure distribution map is shown in Fig. 3. First, the Stern-Volmer plot of the PSP at 300 K in the pressure range of 9.3 Pa-3.6 kPa was examined for calibration as shown in Fig. 3(a). The reference luminescence intensity I_{ref} was measured at $P_{ref} = 132.8$ Pa. The solid line is given by the least-square fitting with the second-order Stern-Volmer equation (Eq. (3) with $N = 2$), and the Stern-Volmer coefficients A_n are obtained as $A_0 = 9.030\times10^{-1}$, $A_1 = 9.409\times10^{-2}$, and $A_2 = -7.969\times10^{-4}$. Second, the reference image was taken (Fig. 3(b)) in the wind-off condition whose pressure P_{ref} is known. In this case, P_{ref} was set to 132.8 Pa. The output of each pixel of the image was used as the reference intensity I_{ref}. After the preparations, the luminescence image was taken in the wind-on condition. The image of Fig. 3(c) was taken for the source pressure P_0 of 53.9 kPa and the background pressure P_B of 133.8 Pa. By dividing the intensity I of each pixel of the wind-on image by that of the wind-off image, the image shown in Fig. 3(d) was obtained. The raw image of Fig. 3(c) is affected by the roughness of the surface, the non-uniformity of the paint layer, the non-uniformity of the light source and so on. However, it is clearly seen that they are eliminated by dividing the intensity I of each pixel by I_{ref} of the corresponding pixel, and the processed image of Fig. 3(d) depends only on the pressure.

By using the Stern-Volmer plot shown in Fig. 3(a), the two-dimensional pressure distribution of the surface can be obtained. Figure 4 shows the pressure distribution on the solid surface obtained from the luminescence distribution of Fig. 3(d) and the Stern-Volmer plot of Fig. 3(a). In this figure, the quantitative pressure distribution is shown in pseudo-color, along with the black-and-white image of luminescence intensity (equivalent to the image of Fig. 3(d)).

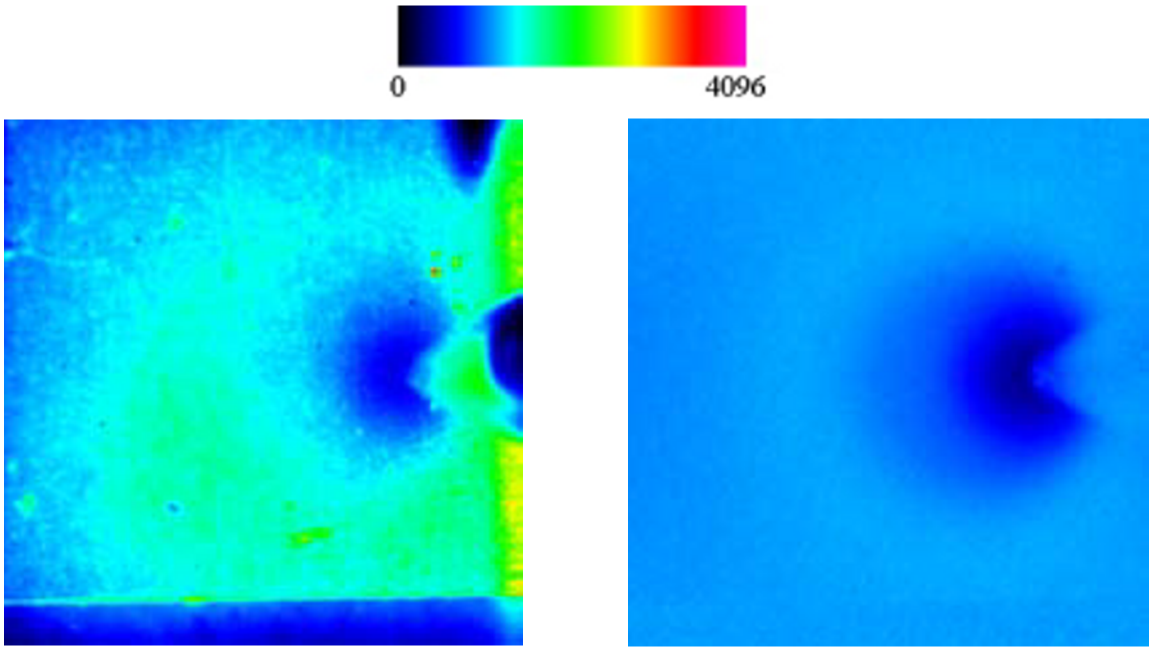
One-dimensional pressure distributions for various background pressure conditions are shown in Fig. 5, which were measured along a projection line of the centerline of the jet shown in Fig. 4. The horizontal axis of Fig. 5 is the position normalized by the whole length of the line in Fig. 4, and the origin is placed downstream. The position 1 is placed at the right edge of the picture. The data in the region over 0.8 are distorted by the refraction by the glass nozzle, and are invalid. The conditions of the source pressure P_0 were 53.9 kPa, 31.7 kPa, 17.7 kPa and 9.3 kPa. In these cases, the background pressures P_B were 133.8 Pa, 80.2 Pa, 50.9 Pa, and 31.2 Pa, respectively, and the highest one among them is as low as about 1 Torr. It is shown in Fig. 5 that the pressure far from the nozzle exit is close to the background pressure in every case. These results indicate that quantitative measurements are possible by the use of PtTFPP/poly(TMSP) in the low-pressure range.

We have also tried to measure the pressure distribution on the surface using Bath-Ru/AA, but the measured pressure far from the nozzle exit did not coincide with the background pressure, although qualitative pressure distribution map could be obtained. From the results we have concluded that it is impossible to measure pressure distributions on solid surfaces by Bath-Ru/AA in low-pressure region quantitatively.



(a) Stern-Volmer plot

(b) Reference image at P_{ref} = 132.8 Pa



(c) Raw image of on-wind condition at P_0 = 53.9 kPa, depending on the pressure distribution

(d) Processed image obtained by dividing image (c) by (b)

Fig. 3. Process to obtain two-dimensional pressure distribution map. In the figures (b)-(d), the intensity of each pixel is shown in pseudo-color. Dark-blue color corresponds to low intensity (high pressure) and red color corresponds to high intensity (low pressure). Cone shape shown in the right part of each image is the shadow of the sonic nozzle made of glass.

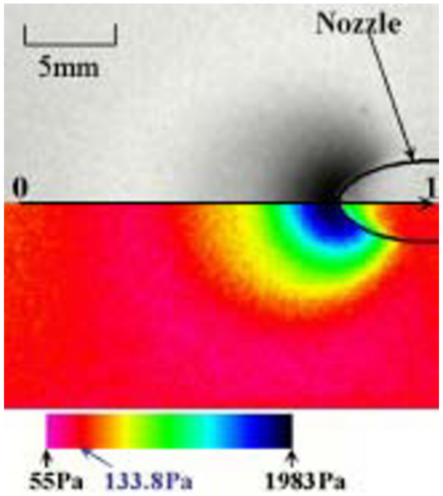


Fig. 4. Pressure distribution in pseudo-color on a solid surface interacting with an impinging supersonic free jet using PtTFPP/poly(TMSP) at P_0 = 53.9 kPa and P_B = 133.8 Pa.

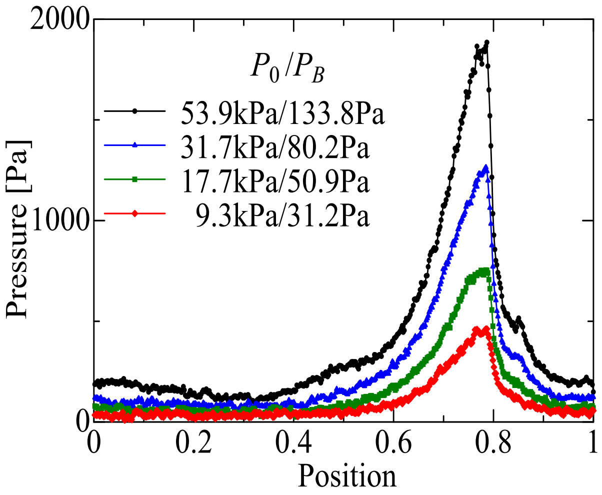


Fig. 5. Pressure distribution along a centerline of a solid surface interacting with an impinging supersonic free jet using PtTFPP/poly(TMSP).

6. Summary

In this study, we have inspected the fundamental properties of three types of PSPs [PtOEP/GP197, Bath-Ru/AA, PtTFPP/poly(TMSP)] in low-pressure conditions below 150 Pa (about 1 Torr), which had not been clarified. We also applied PSP to measurements of pressure distributions on solid surfaces interacting with low density jets. The concluding remarks are as follows:

(1) PtOEP/GP197, which is one of the first generation of the PSPs, shows no pressure sensitivity in the range of pressure below 150 Pa.

(2) Bath-Ru/AA has pressure sensitivity in the range of pressure below 150 Pa, but its Stern-Volmer plot has strong nonlinearity below 20 Pa. Moreover, the absolute luminescence intensity of Bath-Ru/AA is relatively low, resulting in the low signal-to-noise ratio. Therefore, Bath-Ru/AA can hardly be used for quantitative measurement of pressure with high accuracy.

(3) PtTFPP/poly(TMSP) has high and linear sensitivity to pressure change, and the absolute luminescence intensity of PtTFPP/poly(TMSP) is the highest among the three types of PSPs tested in this study, resulting in the highest signal-to-noise ratio. These features of PtTFPP/poly(TMSP) enable quantitative measurement of pressure with high accuracy.

(4) We have succeeded in obtaining a pressure distribution on a solid surface interacting with a low-density supersonic free jet by using PtTFPP/poly(TMSP). From this result, we can see that PtTFPP/poly(TMSP) is very useful for measuring quantitative pressure distribution on solid surfaces in low-pressure range.

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References

Asai, K., Amao, Y., Iijima, Y., Okura, I. and Nishida, H., Novel Pressure-sensitive Paint for Cryogenic and Unsteady Wind-tunnel Testing, J. of Thermophysics and Heat Transfer, 16-1 (2002), 109–115.

Asai, K., Kanda, H., Cunningham, C., Erausquin, R. and Sullivan, J. P., Surface Pressure Measurement in a Cryogenic Wind Tunnel by using Luminescent Coating, ICIASF '97 Record, (1997), 105–114.

Egami, Y., Iijima, Y. and Asai, K., Optimization of Polymer-based PSP for Cryogenic Wind Tunnels, ICIASF 2001, (2001), 177–185, IEEE.

Engler, R. H., Hartmann, K., Troyanovski, I. and Vollan, A., Description and Assessment of a New Optical Pressure Measurement System (OPMS) Demonstrated in the High Speed Wind Tunnel of DLR in Göttingen, DLR–FB (1991).

Fujimoto, T., Sato, K., Naniwa, S., Inoue, T., Nakashima, K. and Niimi, T., Visualization of Impinging Supersonic Free Jet on a Tilt Plate by LIF and PSP, J. of Visualization, 4-2 (2001), 151–158.

Kavandi, J., Callis, J., Gouterman, M., Khalil, G., Wright, D., Green, E., Burns, D. and McLachlan, B., Luminescent Barometry in Wind Tunnels, Rev. Sci. Instrum., 61-11 (1990), 3340–3347.

Liu, T., Campbell, B. T., Burns, S. P. and Sullivan, J. P., Temperature- and Pressure-sensitive Luminescent Paints in Aerodynamics, Appl. Mech. Rev., 50-4 (1997), 227–246.

Nakagawa, T., Gas Permeability of Poly [1-(trimethylsilyl)-1-propyne] Membrane, MEMBRANE, 20-3 (1995), 156–168, in Japanese.

Sakaue, H., Sullivan, J. P., Egami, Y., Iijima, Y., Asai, K., Engler, R. H., Beifuss, U. and Döring, F., Open-system Pressure Sensitive Paint for Surface Pressure Measurements in a Cryogenic Wind Tunnel, ICIASF 2001, 186–195, IEEE (2001).

Authors' Profiles

Hideo Mori: He received his M.S. degree in Electronic-Mechanical Engineering from Nagoya University in 1997 and his Dr. (Eng) degree in Electronic-Mechanical Engineering from Nagoya University in 2000. He worked in Nagoya University as a postdoctoral research fellow of Japan Society for the Promotion of Science (JSPS) from April 2000 to March 2002. During the postdoctorate he worked at the DLR-Göttingen in Germany as a guest scientist from April 2001 to March 2002 to study for analyses of thruster jets by REMPI. He became a research associate in 2003 at Nagoya University. His research interests are spectroscopic measurement techniques of rarefied gas flows.

Tomohide Niimi: He received his M.S. degree in Mechanical Engineering from Nagoya University in 1974 and his Dr. (Eng) degree in Mechanical Engineering from Nagoya University in 1989. He became an associate professor in 1990 and a full professor in 2002 at Nagoya University. His research interests are rarefied gas flow, gas-surface interaction, laser-based measurement technique for highly rarefied gas flow such as LIF and REMPI, optical pressure measurement system for a solid surface and simulation of rarefied gas flow using DSMC and MD.

Masaki Yoshida: He was enrolled in master's course of Electronic-Mechanical Engineering of Nagoya University from 2000 to 2002, and he has received M.S. degree in Electronic-Mechanical Engineering on March 2002. He works in Aisin AW since April 2002. He contributed to the work of this paper during his master's course.

Makoto Kondo: He was enrolled in master's course of Electronic-Mechanical Engineering of Nagoya University from 2000 to 2002, and he has received M.S. degree in Electronic-Mechanical Engineering on March 2003. He works in Omron since April 2003. He contributed to the work during his master's course.

Yusuke Oshima: He is enrolled in master's course of Electronic-Mechanical Engineering of Nagoya University since 2002, and he will receive M.S. degree in Electronic-Mechanical Engineering on March 2004.