Estimating Response Time of Polymer/Ceramic Pressure Sensitive Paint

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Abstract Experiments and numerical simulations have been used in this work to obtain insight into the response time scales of Polymer/Ceramic Pressure Sensitive Paint (PC-PSP). A recently improved analytical model describing the essential physics in PC-PSP quenching kinetics is used which includes the effect of both diffusion time scale and luminescent lifetime on the response of PC-PSP. Experimental results from a shock tube are presented wherein roughness and pressure jump magnitude have been varied to provide a comparison with the model simulations. Model parameters are varied to obtain a good fit to experimental results and this optimized model is then used to obtain response time for a step decrease in pressure, an estimate of which is currently not obtainable from experiments.

Keywords: Pressure Sensitive Paint, Polymer/Ceramic, Step Response

1 Introduction

Pressure-sensitive paint (PSP) is an optical method for determining the qualitative and quantitative description of surface pressure distribution on aerodynamic bodies. Due to its ability to provide a high resolution pressure field, PSP has found applications in a myriad of studies [1] for calculating pressure loads, validating CFD results and for inferring flow physics. With conventional PSP well established as a sensor technology in industries and research labs around the world for steady state testing, recent research and development in PSP has focused on its improvement for unsteady applications. Tremendous improvements in physical structure of the PSP has led to development of fast-responding versions of PSP (Fast-PSP) [3] which are suitable for measuring frequencies above 1 kHz.

Polymer/Ceramic PSP (PC-PSP) is a sprayable form of Fast-PSP that can be readily applied on an aerodynamic model before testing. It consists of a polymer/ceramic basecoat which is first applied on the model as a thin layer and is subsequently used to host the luminophore molecules. The intensity of the excited-state luminescence is modulated based on the concentration of oxygen in the vicinity of the excited luminophores (Figure 1). This dependence of intensity on local pressure is exploited to obtain the pressure information. For improved response characteristics, PC-PSP uses heavy loading of ceramic particles which creates voids in the continuum of the polymer/ceramic coating. This leads to improved porosity and larger scattering of excitation light, which decreases the effective thickness of the PC-PSP coating, thereby decreasing the response times to pressure change. Since PC-PSP imposes no limitation on the model material, it has received wide interest among researchers to characterize unsteady flows.



Fig. 1 Schematic of Polymer/Ceramic Pressure Sensitive Paint

As PC-PSP is increasingly used to study unsteady problems, it is necessary to ascertain the dynamic characteristics of this sensor technology in order to quantify the limits of its application and to develop compensation methods for applications beyond it. Experimental frequency response measurements have shown that at 7-8 kHz, a typical PC-PSP coating has amplitude attenuation of -3 dB and a phase lag of 35° [4]. These studies have also improved the physical understanding of PC-PSP quenching dynamics. Unlike conventional and other Fast-PSPs, the thickness of the coating does not affect the response of PC-PSP [5] which is due to only a very small top layer of the coating (about the roughness of the paint) actively participating in the response characteristics. It has been demonstrated [6] that this thickness-independent response is a characteristic of adsorbed-type PSPs where the luminophores are adsorbed onto a pre-coated base layer. This roughness region of the paint coating is easy to maintain during spray coating and coatings can have different thicknesses but have same roughness and hence same response [4]. Increasing the roughness improves the response characteristics [4][7] by increasing the diffusion coefficient of O_2 in the active part of PC-PSP coating and by improving the scattering of the excitation light. Analytical modelling of frequency response [4][9] has also been used in conjunction with these experimental results to gain further understanding of the dynamic response mechanisms. Comparison of simulated frequency response with experimental data has been done [4] to obtain preliminary quantitative estimates of physical parameters like diffusion coefficient and optical depth.

Though a great deal of quantitative information and physical understanding has been achieved from the frequency response studies, the response characteristics of PC-PSP at frequencies above 10 kHz are still not well understood. This has been due to physical limitations in creating repeatable frequency fluctuations at higher frequencies in those studies. To understand the response time scales, step response studies are needed since it subjects the PSP to much higher frequency content. Previously shock tubes or solenoid-valve driven pressure jump devices have been used to quantify response times of conventional PSPs [10][12] and Anodized-Aluminum PSP (AA-PSP) [13]-[15]. In [16], an earlier version of PC-PSP [17] was tested and its response time was documented as 253 μ s using a first order fit (time constant). The physical structure of PC-PSP has undergone further improvements since then through decrease of polymer amount and replacement of Al₂O₃ ceramic particles with TiO₂ particles [18]. This adsorbed-type PC-PSP is currently the most used type of PC-PSP but its response timescales have not been quantified, though it is suggested to be as low as 25 μ s [19], [20] which compares well with AA-PSP. This fast responding PC-PSP is also commercially available from Innovative Scientific Solutions, Inc. (ISSI product ID – FP-XXXX).

The objective of the current work is to obtain an estimate of the response time for this PC-PSP and to understand the physical parameters that affect it. For this purpose, experimental response time measurements were performed in a shock tube and an analytical model (originally developed in [9] and later used in [4] for direct comparison with experimental frequency response data) was used to describe the observed response. Model parameters were tailored to provide a best fit to the observed response functions. In order to understand how our knowledge of limited frequency response studies extrapolates to higher frequencies, the parameters obtained in current study were compared with those of the previous frequency response study [4]. To begin with, the model is briefly described in the next section along with its numerical implementation and is used in the next section to obtain estimates of response times. In the subsequent sections, the shock tube setup used to obtain step change in pressure is described and corresponding step response results are presented and discussed with model fits.

2 Analytical Model and Numerical Implementation

A simplified analytical model was developed in [9] which accounts for the essential physics in PC-PSP quenching kinetics: one-dimensional diffusion of O_2 into the paint coating and first order population dynamics of excited-state luminophores. The model was further developed in [4] to account for attenuation of excitation light and heterogeneity of paint layer and it was tailored to adsorbed-type PC-PSP by using a two-layered structure. It is briefly described here for a homogeneous coating.

Since thickness of a paint coating is much smaller than the surface area of application, one-dimensional diffusion is assumed to model the permeation of O_2 molecules over time after a pressure change.

$$\frac{\partial \left[O_{2}\right]_{i}}{\partial t} = D \frac{\partial^{2} \left[O_{2}\right]_{i}}{\partial x^{2}}$$
(1)

Eq. (1) can be solved over a discretized paint coating to obtain the local O_2 concentration, $[O_2]_i$ of oxygen in the *i*th layer (of discretization), where *x* is along the depth of the paint and *D* is the diffusion coefficient. For the adsorbed-type PC-PSP, a two-layered structure is assumed since they do not show variation in response with thickness. The bottom layer is assumed to be of a constant diffusion coefficient and is about the thickness of the coating whereas the top layer corresponds to the final deposition of basecoat which constitutes the roughness of the paint. Since this top layer has a much more open structure for rapid diffusion, it is assumed to have a much higher diffusion coefficient (D_1) than the lower layer.

The first order population conservation equation for the number of excited state luminophores can be reduced (see [4, 8] for details) to the following equation which models the generation of intensity ratio in the i^{th} layer,

$$\tau_{ref,i} \frac{d}{dt} \left(\frac{I_i(t)}{I_{ref,i}} \right) + \left(\left(1 - B_i \right) + B_i \frac{\left[O_2 \right]_i(t)}{\left[O_2 \right]_{ref,i}} \right) \left(\frac{I_i(t)}{I_{ref,i}} \right) = 1$$
(2)

where τ_{ref} is the luminescent lifetime at ambient pressure and $B_i = k_Q [O_2]_{ref,i} / (k_R + k_{NR} + k_Q [O_2]_{ref,i})$, is the sensitivity of the *i*th layer. Both these physical parameters can in general vary over the thickness of coating but are assumed to be constant throughout for a homogeneous coating. Given $[O_2]_i(t)$, Eqn. (2) can be numerically integrated for each layer to obtain the corresponding intensity ratio which can then be summed over all layers to obtain the observed intensity response using fraction f_i .

$$\left(\frac{I(t)}{I_{ref}}\right)_{observed} = \sum_{i=1}^{m} \left(\frac{I_i(t)}{I_{ref,i}}\right) f_i$$
(3)

Once light is integrated through the thickness of the paint, a static calibration is then used to convert the intensity ratio to measured pressure fluctuation.

$$\left(\frac{P(t)}{P_{ref}}\right)_{measured} = \frac{1}{B} \left\{ \left(\frac{I_{ref}}{I(t)}\right)_{observed} - (1-B) \right\}$$
(4)

In the end, this measured pressure ratio is compared to the step change used in the boundary condition of the diffusion equation to obtain the simulated response time.

To numerically implement the above model, Eq. (1) is discretized over the thickness of the coating using a finite difference scheme and the interface between the top and bottom layers is modeled as described in [21]. Finer discretization steps were used for the top layer (active part of the coating): a typical sample of 40 μ m thickness and 3 μ m roughness was discretized into 2000 layers of which the top 1000 were assigned to the top 5 μ m while the lower 1000 were assigned to the rest of the thickness. Boundary conditions for the model are a step change in pressure (magnitude and direction of which was varied) at the top and an impermeable wall at the bottom. Initial conditions of uniform oxygen concentration (atmospheric pressure) throughout the paint layer was assumed. Once the local oxygen concentration as a function of time is known at each layer, Eq. (2) is used to determine the local emission intensity ratio. To obtain the observed intensity ratio from Eq. (3), the fraction f_i was defined as

$$f_{i} = \frac{e^{-c(i-1)}}{\sum_{i=1}^{m_{active}} e^{-c(i-1)}}.$$
(5)

This fractional contribution of each layer is based on the assumption that attenuation of incident light with

depth can be described by the Beer-Lambert Law, i.e. $I_x = e^{-(hf)x}$, where I_x is the irradiance at depth x, and hf is the exponential attenuation factor (described hereafter as the hiding factor). In Eq. (5), c is the product of hiding factor (hf) of the Beer-Lambert Law with the thickness of each layer i.e. c is dimensionless and m_{active} is the number of active layers. For higher values of c (or hf), more generation of excited state luminophores happens in upper layers and hence these layers play a larger role in the observed dynamics of PC-PSP as per Eq. (3).

In the next section, we use the parameters obtained by comparing the experimental results (described in section 5) with the model, to obtain an estimate of response time scale of a typical PC-PSP coating for both pressure increase and decrease.

3 Estimate of Response Time

Simulations using the above analytical model were compared with experimental measurements in section 5 and optimized curve fitting was used to estimate the value of diffusion coefficient and hiding factor of a typical PC-PSP coating. Diffusivity of the top layer was found to be $5 \times 10^{-8} \text{ m}^2/\text{s}$, with the lower layer assumed to have a 1000 times smaller value. Hiding factor for violet excitation was found to be 1.67×10^6 per m. An active thickness the same as the roughness of the paint was found to be sufficient for a good curve fit. For a typical sample, an ambient pressure lifetime of 7.30 µs and sensitivity of 0.65 were observed. These parameters have been used to estimate the observed intensity response (*P* increase) and recovery (*P* decrease) times for a step change in pressure between vacuum and 1 Atm in Figure 3(a). A very important use of this optimized model is the estimation of recovery times since experimentally obtaining an estimate of the same is very challenging. Figure 3(b) shows the corresponding measured pressure response (from Eq. (10)). All the figures have been normalized using,

$$I_n(t) = \frac{I(t) - I_{initial}}{I_{final} - I_{initial}} ; \quad P_n = \frac{P(t) - P_{initial}}{P_{final} - P_{initial}}$$
(2)

In this paper, response time is defined as the time taken for intensity or pressure to reach 85% of the final value (τ_{85}). It can be seen in Figure 3 that a pressure increase has an intensity response over three times faster than for pressure decrease for this pressure difference. This arises from longer relaxation time (lifetime) of excited state molecules at lower pressure. However, non-intuitively this difference in intensity response time does not persist when static calibration is used to convert the intensity response to pressure. This opposite behavior of intensity and pressure response is due to the inherent non-linearity in Stern-Volmer equation which produces a higher intensity change for same pressure change if the mean is at a lower pressure. In Figure 3(b), we see that measured pressure decrease shows a faster component initially but slows down later with almost similar times to reach 85% of final value. The initial faster response for pressure decrease is similar to a previous modelling study [22] in which population dynamics was not considered and where it was shown that response time for pressure decrease is smaller than pressure increase. However inclusion of population dynamics leads to a smaller difference in the intensity and pressure response to the finite time taken by intensity to respond to any pressure change, unlike the instantaneous luminophore response considered in [22].



Fig. 3 Observed intensity (a) and measured pressure (b) response (from simulations) for a typical sample for a step change between vacuum and 1 Atm.



Fig. 4 Observed intensity (a) and measured pressure (b) response (from simulations) for a typical sample for a step change between 1 Atm. and 2 Atm.

Corresponding calculations for a different pressure difference – 1 atm. to/from 2 atm. are shown in Figure 4. The pressure increase simulations are similar to the experimental results presented later where almost comparable pressure change is enforced on PC-PSP samples. However using the model, we also obtain the response for a pressure decrease which was not possible experimentally. It can be seen that the trends are similar to Figure 3. We still observe an almost two times faster intensity response for pressure increase; however the 90% intensity response times are not same as Figure 3. Trends for pressure response are also similar for this pressure difference but the difference between step response of increase and decrease is much less. From these two simulations we can estimate the 85% response time for step increase and decrease to be about 65 μ s and 58 μ s, respectively, for this pressure change. This is slightly slower than the previously suggested time constant of 25 μ s which would give an 85% response time of 47.3 μ s.

4 Experimental Framework

The experimental results presented in this study were conducted with samples prepared using the standard formulation of PC-PSP used by many researchers [3]. Readers are referred to [4] for details on how the samples were prepared and the thickness and roughness measurements were performed. Static calibrations of

the samples were performed using a calibration chamber which can achieve set points in temperature and pressure independently of one another. A pulsed LED (400 nm) array (ISSI LM2x-DMHP) was used to obtain lifetime scans at steps of 20.6 kPa from 20.6 kPa to 206 kPa while the temperature was fixed at 298 K. Five hundred scans were averaged and a single exponential curve fit was applied to obtain the lifetime at each pressure.



Figure 5 Shock tube setup

To validate the numerical simulations in previous section, experimental step response data for PC-PSP is required. Shock tubes are the preferred tools in Fast-PSP research due to their ability to create a step change of any magnitude in less than 1 µs. However shock tubes can only be used for creating a pressure increase; in fact obtaining step pressure decrease experimentally is very challenging. A schematic diagram of the setup used in this work is shown in Figure 5. In this study, an end-wall configuration of shock tube is used to exploit the combined power of initial and reflected shock to generate a large change in pressure. It also enables face-based measurement which provides a larger area for light integration and lower photodegradation than in a point-based measurement. The shock tube is constructed of a 0.05 m inlet diameter PVC tube and consists of a driver section (1.83 m) and a driven section (0.75 m). An acrylic tube is used to provide optical access to the setup and at its end, an aluminum plug coated with PC-PSP sample is placed. The driver section was pressurized using air from a high pressure line while a transducer was used to monitor the progress of pressure rise. The driven section was maintained at ambient pressure prior to bursting the diaphragm. Once the desired driver section pressure was reached, a valve was closed to isolate the shock tube. The diaphragm which separates the high pressure driver section from the driven section was constructed of only a 0.1 mm acetate sheet which was burst during the runs using a heated Nichrome wire. A 24V DC supply was used to pass current through the Nichrome wire to heat it. A continuous LED (400 nm) array (ISSI LM2x-DM) with an overall power output of 3 W was used to excite the PC-PSP samples. Emission was captured using a photomultiplier tube (PMT) (Hamamatsu h9505-03) fitted with a long-pass filter (620 nm) to block excitation light. A 300 Ω resistor was used to convert the current output of the PMT to voltage, with the value of the resistor selected to ensure that the dynamic response of the measurement system does not affect the response time of PC-PSP. An oscilloscope (Lecroy Waverunner 44Xi) was used to capture a record 10⁵ samples long of the PMT output at a sample rate of 2 GHz. In order to trigger this data collection, a transducer collocated at the end of the shock tube was used. Post processing of collected time scans involved digital filtering, which was performed using a low-pass FIR filter with passband frequency of 1 MHz. Figure 6 shows the normalized recorded PMT data along with the filtered data which was used for comparison with the model. For normalization, the mean of PMT values before t = 0 was used as a minimum and the mean from $t = 1.5 \,\mu s$ until the end of the acquisition was used as the maximum. We observe that the shock tube setup produces a clean initial rise in pressure but has some fluctuations in the steady state region. Response simulations presented below were however done using a step change in pressure instead of this actual pressure change.



Fig. 6 Normalized PMT signal and the corresponding filtered data.

4 Results and Discussions

The primary objective of this work is to estimate the response time of PC-PSP and to understand how our current understanding of the paint response extends to higher frequencies. To this end, step response measurements for a typical sample are shown while simulated response obtained by varying the parameters of the model are fit to these experimental measurements. The parameters obtained from this work are then compared with parameters obtained from a previous frequency response study. Also shown are the experimental effects of varying the final pressure of pressure change and roughness to validate the model.

Measured values of sample characteristics for a typical and rough coating are listed in Table 1. We observe that sensitivity of the paint, which is defined in Eq. 2 as the slope of the static calibration, is only slightly affected by roughness of coating. However the lifetime at ambient pressure is smaller for a rougher sample since a more open structure provides for a faster quenching by ambient O_2 . Uncertainty in these measurements has been discussed in [4].

Sample	Thickness	Roughness	Lifetime	Sensitivity
Typical	44.12 μm	3.16 µm	7.30 µs	0.65
Rough	104.2 μm	23.85 µm	6.75 µs	0.64

 Table 1. Sample characteristics

Figure 7 shows the normalized intensity response for two typical samples each evaluated twice for their step response characteristics in the shock tube setup. The shock tube was run with a driven pressure of 213 kPa which subjected the sample to a step change in pressure from 101.3 kPa to 205.6 kPa. The repeatability of the setup is demonstrated by the similar response over two runs for each of the two samples. Repeatability across samples is also demonstrated by this figure as two similarly prepared samples have almost the same step response characteristics. Also shown in the figure is the simulated response for this step change in pressure while the parameters for the same are listed in Table 2. We see that with these parameters, the simulated response produces a very good match to the experimental results (ignoring the fluctuations after 85% rise time). During simulations, hiding factor was fixed at the previously modeled value [4] of 1.67 x 10⁶ per m, whereas diffusivity of the top layer was allowed to vary. The best fit value of diffusion coefficient is well within an order of magnitude of the value obtained from frequency response work and thus provides confidence to the modelling efforts. Lifetime value input into the model was slightly different from the previous work; however it is known from [4] that such small a difference will not produce any observable

change in response time. Static calibration was used to convert the simulated and experimental intensities to pressure which are shown in normalized form.



Fig. 7 Measured and simulated intensity (a) and pressure (b) step response over different runs and samples.

To validate the obtained values of parameters, an experiment with larger pressure change was conducted to exploit the effect of non-linearity in intensity response. The driver section was pressurized to 275.8 kPa which produced a step change from 101.3 kPa to 257.4 kPa (more than 50 kPa higher than a typical run) at the PC-PSP location. As shown in Figure 8, we obtain a faster intensity response for this larger pressure change due to smaller luminescent lifetime at higher pressure. Larger fluctuations are also observed in the steady state region. For simulating this higher pressure run, all the parameters were fixed for that of the typical sample and only the final pressure in the diffusion equation was changed to this higher value. We observe that the simulations correspond very well with the experimental measurements. Figure 8 (b) shows that translation to pressure diminishes the difference in intensity response as seen previously in (Figure 4).



Fig. 8 Measured and simulated intensity (a) and pressure (b) step response for different final pressure.

Roughness of the PC-PSP basecoat has been found [7][4] to affect the frequency response of the paint coating by improving the permeability of O₂. To confirm this at higher frequencies, a rough sample (in fact the same as was used in previous work [4]) was evaluated in the shock tube setup. Figure 9 compares the step response for typical and rough samples when both are subjected to the same pressure change. We observe that intensity response of the rough sample is much faster than a typical one. The simulated response which provided the best curve fit to the rough sample response is also shown. The same methodology as a typical case was used; lifetime and hiding factor were fixed at the values known from the frequency response study while diffusion coefficient was varied to obtain a good fit. This exercise gives an estimate of the largest uncertainty in our knowledge of this physical parameter. Table 2 lists the parameters, and it can be seen that diffusion coefficient is again well within an order of magnitude of the previous study. Figure 9 (b) shows that this fast intensity response of a rough coating also translates into fast pressure response since the pressure change and sensitivity are same as the typical case. We observe about 30% improvement in the response time as the roughness was increased which can be exploited in applications where increased flow intrusion by larger roughness in not an issue.



Fig. 9 Measured and simulated intensity (a) and pressure (b) step response for a typical and a rough sample.

Sample	Parameter	Step Response Study	Frequency Response Study
Typical	Lifetime (µs)	7.30	7.04
roughness	D ₁ (m ² /s)	5 x 10 ⁻⁸	9.78 x 10^{-8}
with PtTFPP	Hiding Factor (1/m)	1 67 x 10 ⁶	1.67 x 10^{6}
Larger	Lifetime (μ s)	6.75	6.75
roughness	D ₁ (m ² /s)	2.5 x 10 ⁻⁶	7.51 x 10 ⁻⁶
with PtTFPP	Hiding Factor (1/m)	3.13 x 10 ⁵	3.13 x 10 ⁵

Table 2. Model Parameters for Curve Fits

Conclusions

Response time scale to step change in pressure has been estimated for a typical TiO2 and PtTFPP based PC-

PSP using experiments and numerical simulations. Parameters of the PC-PSP dynamic model (diffusion coefficient and hiding factor) were varied to obtain a good fit to measured step rise data from a shock tube experiment. The model was then used to estimate response time for a step decrease in pressure which is currently not obtainable experimentally. Due to the non-linear (hyperbolic) nature of Stern-Volmer equation, step decrease in pressure will have a shorter response time than corresponding step rise across same pressure difference. Model parameters from current work were also compared with those obtained from a previous limited frequency response study and this comparison provides a more confident bound on the estimates of diffusion coefficient. Though the values were comparable and within an order of magnitude of previous work, the difference might be attributed to the limited frequencies at which the measurements were conducted in the earlier work and the limited ability to achieve a step pressure change in the current experiments. However it can now be said with confidence that the diffusion coefficient for the top active layer (roughness region) of a typical PC-PSP is about 5 x 10^{-8} to 10^{-7} m²/s.

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