IMECE 2004-59900

REDUCTION OF THERMOGRAPHIC PHOSPHORS DATA TO HEAT RATES

P.R. Crim and D.G. Walker*

Department of Mechanical Engineering
Vanderbilt University
VU Station B 351592
Nashville, Tennessee 37235
Email: greg.walker@vanderbilt.edu

S.W. Allison and S. Goedeke

Thermographic Phosphor Sensing Applications
Oak Ridge National Lab
2360 Cherahala Blvd.
Knoxville, Tennessee 37932

ABSTRACT

Thermographic phosphors have emerged as a new technique for measuring heat fluxes, which relies on the temperature dependent intensity decay of thermographic phosphors. However, instead of reducing the intensity data to temperatures, heating rate is estimated. It has been shown that the heating rate can provide significantly better heat flux estimates than temperature measurements. Because the technique is new, little is known about the quality of heating rate estimates. Further, the heating rate estimation depends on the introduction of additional free parameters, which increases the uncertainty of the estimates. The analysis presented here indicates that sample rates must be one to two orders of magnitude greater than the frequency at which the heat flux must be known. Also, the sensitivity of the intensity to higher-order derivatives is small suggesting that derivatives beyond the heating rate are not accessible with single-shot data.

NOMENCLATURE

γ intensity sample rate (s^{-1})
Γ experimental sample rate (s^{-1})
I emission intensity (V)
I_0 initial emission intensity (V)
ξ decay time derivative
n number of excited electrons
n_0 number of initial excited electrons
n^* number of excited luminescence centers

t time (s)
τ decay time (s)
τ_0 initial decay time (s)
τ_e electron lifetime (s)
W_R transition rate of radiative mechanisms (s^{-1})
W_{NR} transition rate of non-radiative mechanisms (s^{-1})

INTRODUCTION

In search of an accurate and stable heat flux determination technique, researchers have begun to investigate temperature-derivative measurement approaches. Heat flux determination is usually accomplished in one of two ways. 1) Heat flux can be measured directly with devices that are calibrated to return a voltage proportional to the heat heat flux. These devices are difficult to calibrate, expensive and tend to integrate high-frequency components in the data. 2) Heat fluxes can also be determined by measuring temperature and using a data reduction technique to discover the heat flux. Although temperature measurements are much more reliable and cheaper to obtain than heat fluxes, the data reduction of heat fluxes from temperatures is an ill-posed problem, and any uncertainty in the measurement becomes amplified during the data reduction.

Mathematically, the difficulty of determining heat fluxes from temperature measurements arises from the differentiation of data [Kress, 1989]. Frankel and Keyhani [1997] suggested the use of temperature derivatives to stabilize the inverse heat conduction solution. Since then, numerous conference proceed-

ings that identify the utility of heating rate measurement devices have appeared [Frankel and Keyhani, 1999, Frankel and Osborne, 2003, e.g.]. Walker and Schetz [2003] first identified thermographic phosphors as a legitimate technique to measure heating rate. Later, Walker [2003] provided the first evidence that heating rates and subsequent heat flux can be deduced stably from intensity measurements of thermographic phosphors. Despite this preliminary evidence, little is known about the sensitivity of the heat flux estimates from measured thermographic phosphor data.

The objective of the present work is to examine the temperature dependence of phosphor decay and to establish models for predicting time derivative decay from intensity measurements. With experimental data [Goedeke et al., 2002], we will show that the new approach can predict temperature signatures more accurately when transients occur during the decay measurements. Although not a primary focus of the present work, the change in decay time can be used to predict the heating rate directly, which will ultimately provide superior heat flux estimation capabilities [Walker, 2003].

1 THEORY

Thermographic phosphors are rare-earth doped ceramics that fluoresce when exposed to light. The intensity, decay rate and wavelength of the emission is temperature dependent. Therefore, each of these properties can be detected to predict temperature. The application and data requirements typically determine the type of phosphor and measurement technique to use [Allison and Gillies, 1997]. Because emission is inherently transient, intensity measurements can be used to extract transient temperature data (i.e., heating rate). The present work focuses on the nature of the temperature dependence of phosphor decay and using this temperature dependence to extract temperature signatures from intensity measurements during transient events.

When a phosphor is excited by incident photons, it will begin to reemit at a prescribed wavelength based on its electronic band structure [Shionoya and Yen, 1999]. The emission intensity is dependent on many factors including material properties, doping, temperature and the excitation source. Measured emission I is proportional to the rate of change of excited luminescence centers n^* [Shionoya and Yen, 1999], i.e.

$$I \propto \frac{dn^*}{dt}. \tag{1}$$

The number of luminescence centers (electron/hole pairs available for recombination) is governed by the radiative and non-radiative recombination of electrons with holes as

$$\frac{dn^*}{dt} = -(W_R + W_{NR})n^*, \tag{2}$$

where W is the transition rate of radiative and non-radiative mechanisms [Shionoya and Yen, 1999]. The transition rates are commonly combined into an overall lifetime τ such that [Shionoya and Yen, 1999]

$$\tau^{-1} = W_R + W_{NR}. \tag{3}$$

In general, this lifetime is temperature dependent. Because the emission is a function of the lifetime, emission measurements can be used to predict the temperature of the phosphor.

Three methods for extracting temperature information from intensity measurements are considered. The first approach is merely a presentation of standard data reduction techniques used to extract temperatures only from intensity measurements. Even though this technique has been described previously [Allison and Gillies, 1997], salient features are repeated here to provide a basis for discussion and comparison. The next two approaches mathematically introduce the decay derivative by assuming a varying time-dependent temperature in the governing equation. These methods are similar to the simple approach in that the decay time and a change in the decay time are predicted using parameter estimation techniques. Ultimately, these quantities are then converted to heating rates through the material properties of phosphors that have been characterized a priori. These heating rates have been shown to produce far more stable estimates of heat fluxes than temperature measurements alone [Walker, 2003]. However, the focus of the present work is to examine the decay in light of transient thermal behavior and to illustrate how the new models can provide superior estimates of the temperature when transients are present.

1.1 Simple Approach

When a phosphor is excited by incident photons, it will begin to reemit at a prescribed wavelength based on its electronic band structure [Shionoya and Yen, 1999]. The emission intensity is dependent on many factors including material properties, doping, temperature and the excitation source. Recall Eqns. 1 and 2 describing emission I and excited luminescence centers n^*, where the negative sign indicates emission. The number of excited electrons n after the excitation is turned off is governed by the differential equation

$$\tau(T)\frac{dn}{dt} + n = 0, \tag{4}$$

where $\tau_e[T(t)]$ is the electron lifetime, which in general is a function of temperature that can change with time. If the electron lifetime is assumed to be constant during the intensity decay, then

the resulting solution to Eqn. 4 is simply

$$\frac{n}{n_0} = \exp\left(-\frac{t}{\tau_e}\right), \qquad (5)$$

where n_0 is the number of electrons at $t = 0$ (when the excitation source is removed). In the foregoing analysis, the intensity decay time τ is assumed to be equivalent to the electron lifetime $\tau_e = \tau$. This decay time can be recovered by differentiating Eqn. 5 as prescribed by Eqn. 1.

$$\frac{I}{I_0} = \exp\left(-\frac{t}{\tau}\right), \qquad (6)$$

where I_0 is the initial intensity when the excitation source is removed ($t = 0$). This will be referred to as the *simple model*. The decay time can be recovered from a fit of the foregoing model to measured intensity data. Finally, the decay time is related to the temperature, which is determined from characteristic phosphor curves. (See Figure 1.)

1.2 Power Method

If the variation in temperature is of the order of the decay time, then the constant lifetime assumption is presumably invalid. In general, τ must be expressed as some function of time and the resulting nonlinear differential equation must be solved. However, the function is unknown and approximations must be made. Perhaps the simplest approximation is to assume a Taylor series expansion of the lifetime, such that

$$\tau(t) = \tau_0 + \frac{d\tau}{dt}t + O(t^2), \qquad (7)$$

where $\tau_0 = \tau\,(t = 0)$ is the lifetime at the beginning of the decay. With this linear approximation to the variation of τ in time, an additional parameter is introduced compared to the *simple model*. The solution to Eqn. 7 for intensity (called the *power model*) is then given as

$$\frac{I}{I_0} = \left(\frac{\tau_0}{\tau_0 + \frac{d\tau}{dt}t}\right)^{1/\frac{d\tau}{dt}}, \qquad (8)$$

where $I_0 = n_0$ at $t = 0$. For steady state, $d\tau/dt \to 0$, and the simplified form (Eqn. 6) is recovered. Now the model (Eqn. 8) can be compared to measured data to extract the two parameters. While an estimate for τ can be used to find the temperature, the estimate for $d\tau/dt$ will directly give the heating rate using the

chain rule.

$$\frac{dT}{dt} = \frac{dT}{d\tau}\frac{d\tau}{dt}. \qquad (9)$$

Note that the derivative of temperature with respect to τ can be found by differentiating the calibration curve. The calibration curve of any phosphor can be easily obtained by measuring the decay time of the phosphor at different steady state temperatures [Allison and Gillies, 1997].

1.3 Exponential Method

An alternate model suggested by Walker and Schetz [2003], is to retain the exponential approximation, but assume that τ is a linearly varying function of temperature (truncated Taylor series as before). The resulting model (called *exponential model*) is simply presented as

$$\frac{I}{I_0} = \exp\left(-\frac{t}{\tau_0 + \frac{d\tau}{dt}t}\right) \qquad (10)$$

The physical parameters are presumably identical and physically based, yet there is an apparent discrepancy between the forms of the two higher-order models.

However, the truncated higher-order terms affect the form of the solution in Eqn. 8. In other words, the form and validity of the solution is dependent on the type of measurements being made. Nevertheless, it is not clear whether this feature will influence the quality of heating rate estimates.

2 MEASUREMENTS

Data were collected at Oak Ridge National Lab on a piezoresistive microcantilever that has been described previously [Goedeke et al., 2002]. The cantilever was treated with Mg_4FGeO_6:Mn whose calibration curve is given in figure 1. The phosphor decay time is approximately 10 ms and heating was produced during the experiment (through Joule heating) for 10 ms as well. Several intensity measurements were made so that the heating pulse occurred at different points during the decay measurements. Therefore, the temperature was not constant during the intensity decay measurements suggesting that transient models are necessary.

3 RESULTS AND DISCUSSION

Walker [2003] has shown that a tremendous amount of error in the heating rate quantity can be tolerated by data reduction schemes. However, no work has been performed to predict the types and level of errors that can be expected in the estimation
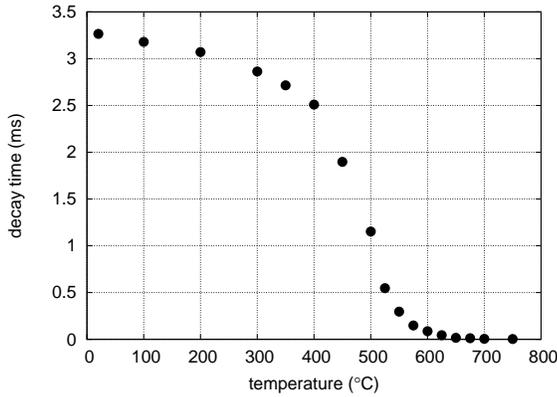
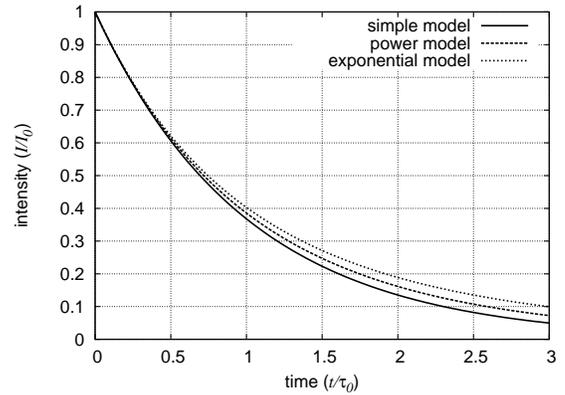Figure 1. CALIBRATION CURVE FOR Mg_4FGeO_6:Mn PHOSPHOR.

Figure 2. THE EXPONENTIAL MODEL AND POWER MODEL DO NOT MATCH THE SIMPLE MODEL BECAUSE ξ IS NON-ZERO ($\xi = 0.1$). WE CAN SEE THE EFFECTS OF THE NON-PHYSICAL APPROXIMATION OF THE EXPONENTIAL MODEL FOR LARGE ξ.
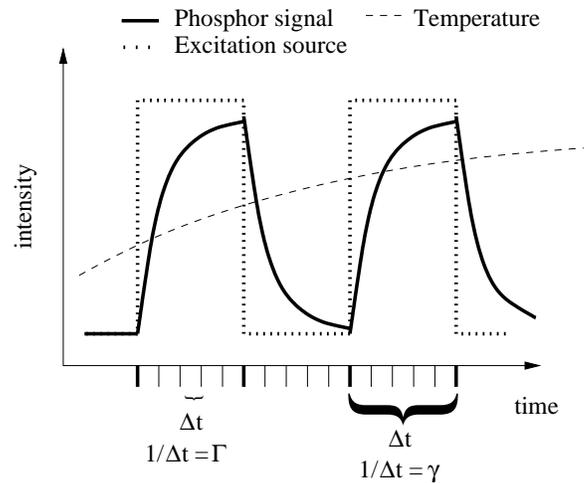

Figure 3. THE TWO TIME SCALES CORRESPONDING TO THE INTENSITY SAMPLE RATE γ AND THE EXPERIMENTAL SAMPLE RATE Γ. NOTE THAT THE VARIATION IN TEMPERATURE (OR HEAT FLUX OR HEAT RATE) IS SMALL BUT NOTICEABLE COMPARED TO Γ, BUT INSIGNIFICANT COMPARED TO γ.

of the heating rate. Furthermore, the validity of different decay models has not been explored. For the purposes of discussion, the decay time derivative is written as $d\tau/dt = \xi$. Subscripts e and p will be used to designate the exponential model or power model respectively.

The exponential model has the advantage that it can incorporate higher orders of τ derivatives by simply extending the Taylor series expansion. To include higher-order terms in the power model, a new differential equation must be solved. In addition, the exponential model does not suffer from computational problems as ξ becomes small (i.e. the temperature is not changing). Despite these advantages, the exponential model is not as physically accurate, especially as ξ increases, and contains a singularity at $\xi = -\tau_0/t$. Figure 2 shows the three models at $\xi = 0.1$. Here we can see that the non-physical nature of the approximation used in the exponential model results in a deviation between the models. Note that we expect a deviation of both models from the simple model because the simple model does not include any change in the time constant. Therefore, as long as ξ is non-zero, neither the exponential nor the power model will match the simple model.

Relative magnitudes of the parameters of interest, τ and ξ, can be obtained by examining measured data and expected sample rates. There are fundamentally two characteristic rates in the measurement problem. The first is the experimental rate at which the heat flux is to be determined Γ, and the second is the sample rate for intensity measurements γ. Refer to Figure 3 for a pictorial representation. Note that the graph is a cartoon only, and the excitation duration and time between triggers are not necessarily correlated. Naturally, the intensity measurement sample rate should be larger than the experimental sample rate ($\gamma > \Gamma$). The excitation source is pulsed at the experimental rate Γ. For each pulse, many intensity measurements are taken to measure the decay of the phosphor emission. For a given application, Γ will be determined such that any high frequency components in-

herent to the experiment can be captured. This approach ensures that observations faithfully recreate physical features. Therefore, the magnitude of the change in the measured quantity between experimental times $1/\Gamma$ is expected to be relatively small. As a result, we can assume that ξ will be small. From order of magnitude estimates and examination of typical phosphor data, the normalized values of $\tau/\tau_0 = 1$ and $\xi = 0.1$ are representative of actual measurements.

Sensitivities computed by differentiating the model inten-

4

sity with respect to each free parameter can further elucidate differences between the models. The sensitivity is defined as the derivative of the measured quantity (intensity) as a function of the predicted parameter. Therefore, a large sensitivity suggests that the parameter can be predicted with a high degree of confidence. For the exponential model, the normalized sensitivity coefficients are given as

$$\frac{d(I/I_0)}{d\tau_0} = \frac{t}{(\tau_0 + \xi t)^2} \exp\left(-\frac{t}{\tau_0 + \xi t}\right) \quad \text{and} \qquad (11)$$

$$\frac{d(I/I_0)}{d\xi} = \frac{t^2}{(\tau_0 + \xi t)^2} \exp\left(-\frac{t}{\tau_0 + \xi t}\right). \qquad (12)$$

For the power model, the sensitivities are given as

$$\frac{d(I/I_0)}{d\tau_0} = \frac{\tau_0^{1/\xi - 1}}{(\tau_0 + \xi t)^{1/\xi + 1}} t \quad \text{and} \qquad (13)$$

$$\frac{d(I/I_0)}{d\xi} = -\frac{\tau_0^{1/\xi}}{(\xi)^2(\tau_0 + \xi t)^{1/\xi + 1}} \times$$
$$[\xi t + (\tau_0 + \xi t)(\ln(\tau_0) - \ln(\tau_0 + \xi t))]. \qquad (14)$$

Evaluated at the assumed values for τ_0 and ξ, like sensitivities are plotted against each other in Figure 4. First notice that the sensitivities are linearly independent for each model (Compare $\tau_0 d(I/I_0)/d\tau$ and $\xi d(I/I_0)/d\xi$ for each model in (a) and (b) of Figure 4.) This means that the two parameters τ_0 and ξ can be estimated independently for each model. The sensitivity of τ_0 indicates that both higher-order models can predict τ_0 with similar confidence to each other and the simple model. However, estimation of ξ can be performed with greater confidence in the exponential model. The sensitivities have been normalized by multiplying by the corresponding estimates. As such, the scale of Figure 4 (a) and (b) indicate that ξ can not be determined with as much confidence as the decay time τ_0. Moreover, this feature indicates that higher-order derivatives of the decay time will contain a tremendous amount of uncertainty and will likely be unusable for estimation purposes. This is not surprising, because the experimental sample rate Γ is chosen so that these higher-order derivatives are indeed small.

Consider thermographic phosphor measurements made on the piezoresistive microcantilever [Goedeke et al., 2002]. The electrical current was turned on at $t = 0\,\text{ms}$ for a duration of $10\,\text{ms}$. Temperature measurements using thermographic phosphors were made at $1\,\text{ms}$ intervals starting $6\,\text{ms}$ before the current was turned on. The phosphor has a characteristic decay time of the order of the heating duration. Therefore, the intensity will change as a result of the changing temperature during the measurement. From the standard temperature measurement
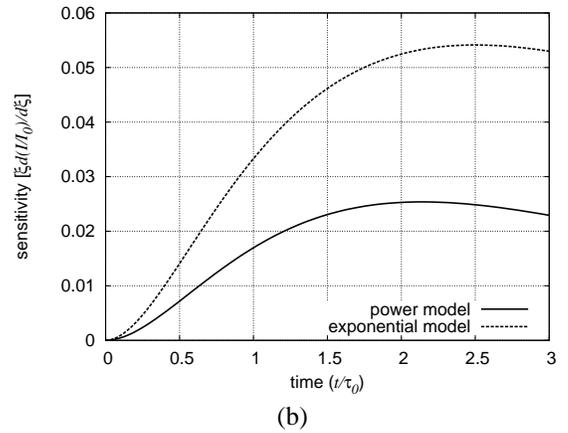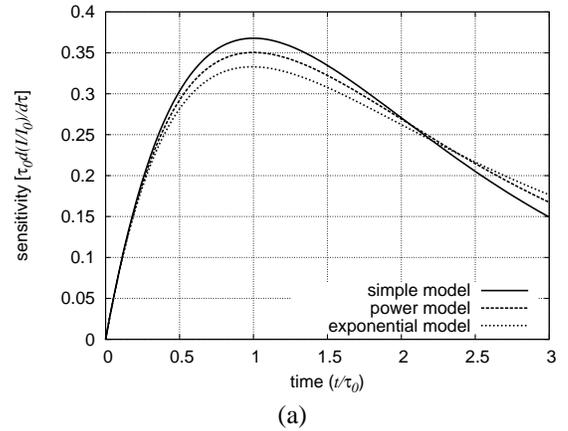

(a)

(b)

Figure 4. NORMALIZED SENSITIVITIES FOR (a) τ AND (b) ξ COMPARED BETWEEN MODELS.

technique (*simple model*), the decay time and temperature prediction can be seen in figure 5. Although we know the temperature does not rise until the heating is turned on, the temperature prediction begins to rise before the heating and begins to fall before the heating is turned off. The decay time of most phosphors is inversely proportional to temperature, so the decrease in decay time corresponds to the rise in temperature. The higher order models, shown in figure 6, show that the decay time is relatively constant before the heat is turned on, which better matches our intuition than the *simple model*. Further, the decay time continues to decrease during the heating phase and begins to increase much closer to the turn-off time than the *simple model*.

For the higher order models, the estimate of the derivative of decay time (shown in figure 7) should be near zero while temperature is not changing. If the temperature is rising, the value should be positive; for decreasing temperature the value should be negative. Unfortunately, the results do not display this behavior. There are several possible reasons for this. First, little is known about the real heating in the piezoresistive cantilever

5
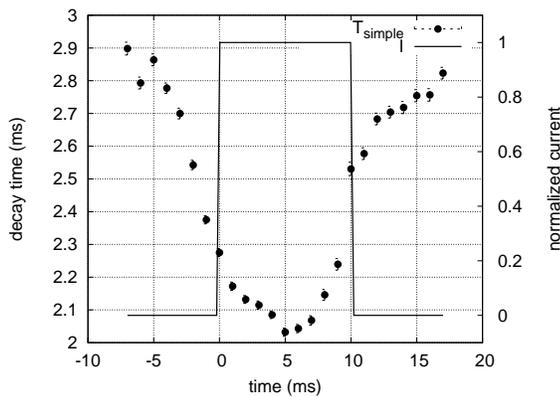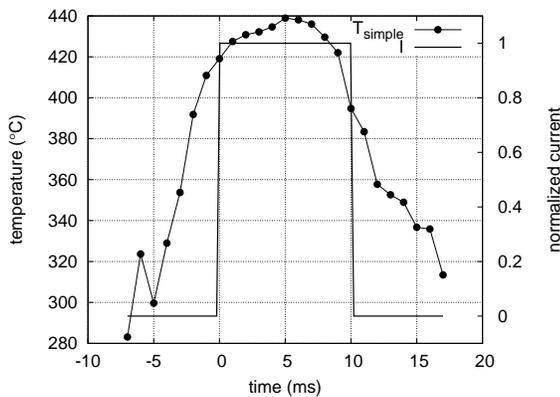
Figure 5. (a) DECAY ESTIMATES FROM SINGLE-SHOT TGP MEASUREMENTS DURING A HEATING EVENT USING THE SIMPLE MODEL. (b) TEMPERATURE REDUCED FROM THE DECAY TIME.
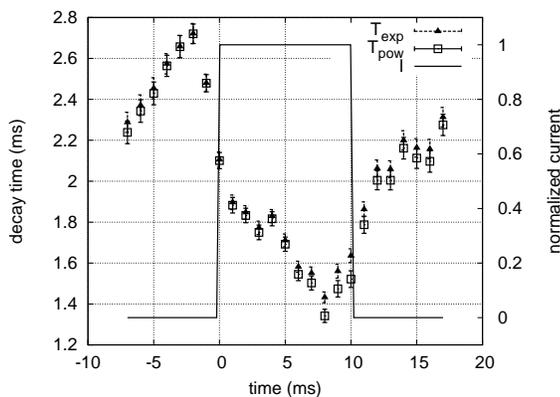

Figure 6. DECAY TIME FROM SINGLE-SHOT TGP MEASUREMENTS DURING A HEATING EVENT USING THE HIGHER-ORDER MODELS.
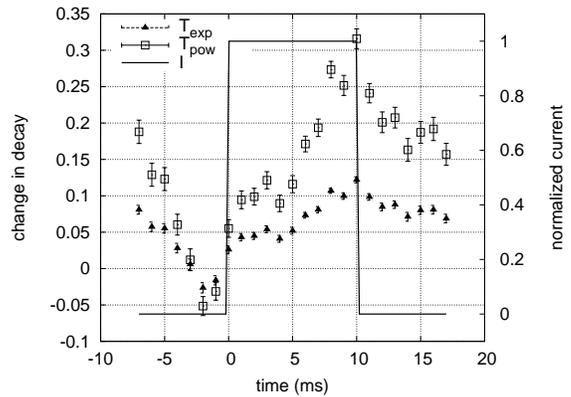

Figure 7. CHANGE IN DECAY TIME (ξ) ESTIMATED FROM TGP MEASUREMENTS DURING A HEATING EVENT.

and the boundary conditions that govern the actual temperature. In fact, we can not be completely sure that the temperature does not react as predicted. This suggests that a tightly controlled experiment be performed to evaluate the validity of the change in decay time. Interestingly, by the time this work is presented, data from just such experiments are expected to be available. Second, the emission from the piezoresistive cantilever is produced from clumps of TGP that may not be in intimate thermal contact with the device (see Goedeke et al. [2002]). Although the decay time for all models behaves roughly as expected, the estimates with relatively smaller sensitivities (see figure 4) accentuate flaws in the measurements.

4   CONCLUSIONS

Thermographic phosphors can be used to predict both temperature from decay time and heating rate from change in decay time. The higher-order models assume that the temperature varies during intensity decay measurements of the phosphors. Both higher-order models are approximately equivalent for modest temperature changes. However, the exponential model is likely not accurate for large temperature changes. Additional testing needs to be performed to establish the limits and accuracy of each model. Nevertheless, both models are able to predict a more accurate temperature signatures based on decay time compared to the *simple model* when transient features are present during the measurements.

ACKNOWLEDGMENT

REFERENCES

S. W. Allison and G. T. Gillies. Remote thermometry with thermographic phosphors: Instrumentation and applications. *Review of Scientific Instrumentation*, 68(7):2615–2650, July 1997.

J. I. Frankel and M. Keyhani. A global time treatment for inverse heat conduction problems. *Journal of Heat Transfer*, 119:673–683, November 1997.

J. I Frankel and M. Keyhani. Inverse heat conduction: The need for heat rate data for design and diagnostic purposes. In *proceedings of the 18th LASTED International Conference on Modeling, Identification and Control*, Innsbruck, Austria, February 1999.

J. I. Frankel and G. E. Osborne. The development of heating/cooling, $\partial T/\partial t$ and heat flux rate, $\partial q''/\partial t$ sensors for aerospace applications. Technical report, Propulsion Measurement Sensor Development Workshop, NASA Marshall Space Flight Center - Space Transportation Directorate, May 2003.

S. Goedeke, S. W. Allison, M. R. Cates, and P. G. Datskos. Determination of surface temperature on micrometer scaled objects. In *Proceedings of the 48th International Instrumentation Symposium of the ISA*, San Diego, CA, May 2002.

R. Kress. *Linear Integral Equations*, volume 82 of *Applied Mathematical Sciences*. Springer-Verlag, 1989.

S. Shionoya and W. M. Yen, editors. *Phosphor Handbook*. CRC Press, 1999.

D. G. Walker. A new technique for heat flux determination. *Journal of Heat Transfer*, September 2003. accepted.

D. G. Walker and J. A. Schetz. A new technique for heat flux determination. In *proceeding of the ASME Summer Heat Transfer Conference*, Las Vegas, NV, July 2003.