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Photoacoustic measurement of thermal diffusivity of polymer foils

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The photoacoustic measurement of polymer foils, typically 170–200 μm thick, is discussed. It is shown that the measurement based upon the phase lag between the front and rear illuminations is applicable only in a limited range of frequencies from 6 to 12 Hz. The dominant mechanism responsible for the photoacoustic signal, in almost the entire frequency range 10–100 Hz, is proven to be the thermoelastic bending of the foil samples. The thermal diffusivity is then obtained from the frequency dependence of the front-phase illumination data.

I. INTRODUCTION

In a recent paper¹ we have demonstrated the usefulness of a single modulation frequency photoacoustic (PA) method for measuring the thermal diffusivity of solid samples. The method consisted in measuring the relative phase lag $\Delta\phi = \phi_F - \phi_R$, at a single modulation frequency, between the rear-surface illumination (R) and the front-surface illumination (F). This method is an alternative to the one proposed by Yasa and Amer,² in which the attenuation of the rear-illumination signal amplitude (S_R) relative to the front-illumination signal amplitude (S_F) is measured. In this case, as well as in all other conventional photoacoustic and photothermal techniques,^{2–8} the thermal diffusivity is measured by recording the PA signal as a function of the modulation frequency. In contrast, in the two-beam phase-lag method the thermal diffusivity is obtained from a single chopping frequency measurement as follows.

Using the thermal diffusion model of Rosenzweig and Gersho⁹ (RG) for the production of the PA signal, the ratio S_F/S_R of the signal amplitude and phase lag $\Delta\phi$ for front- and rear-surface illuminations (cf. Fig. 1) are given by

$$S_F/S_R = I_F/I_R [\cosh^2(l\alpha_s) - \sin^2(l\alpha_s)]^{1/2} \quad (1)$$

and

$$\tan(\Delta\phi) = \tanh(l\alpha_s)\tan(l\alpha_s), \quad (2)$$

where l is the sample thickness, $\alpha_s = (\pi f/a_s)^{1/2}$ is the thermal diffusion coefficient, f is the modulation frequency, α_s is the sample thermal diffusivity, and I_F (I_R) is the absorbed light intensity for the front (rear) illumination. In principle, either Eq. (1) or (2) would give us the value of α_s from a single modulation frequency measurement. However, since Eq. (1) depends explicitly on the ratio I_F/I_R (i.e., one needs precise power monitoring and identical surface conditions on both sides of the sample), the value of the thermal diffusivity in the signal amplitude ratio measurement is obtained from the slope of the curve S_F/S_R as a function of the modulation frequency. In contrast, Eq. (2) exhibits no explicit dependence on the absorbed power and surface conditions, so that a single modulation frequency measurement is sufficient to derive the thermal diffusivity. Furthermore, the fact

that the phase-lag method [cf. Eq. (2)] is independent of power calibrations and surface conditions renders it as a more precise technique than the amplitude ratio method.

In Ref. 1 we have successfully applied the phase-lag method to several samples ranging from optically opaque (e.g., semiconductors) to optically transparent (e.g., glasses). When applying this technique to the case of polymer foils, a few hundred microns thick, it worked only at very low modulation frequencies, typically of the order of 6–12 Hz. In this paper we address ourselves to this problem. We show that in the case of polymer foils, the phase-lag method based upon the simple RG thermal diffusion model, as given by Eq. (2), is not straightforwardly applicable. As will be demonstrated, the reason for the failure of Eq. (2) to explain the experimental data is that in this expression for $\Delta\phi$ the bending of the sample due to the thermoelastic coupling¹⁰ is neglected. The thermoelastic bending is essentially due to a temperature gradient normal to the sample plane (z axis). The existence of this temperature gradient causes the thermal expansion of the sample to depend on z . This z dependence of the sample displacement along its plane induces a bending of the sample in the z direction.

II. EXPERIMENT

The experimental arrangement of our two-beam experiment is basically the same as the one described in Ref. 1. The light from a 120-W tungsten lamp, after being chopped, is divided by a beam splitter, and each resulting beam is directed to opposite sides of the photoacoustics cell (cf. Fig. 1). The PA cell is a conventional PA brass cell in which a $\frac{1}{2}$ -in. BK condenser microphone is mounted in one of its walls. The samples, in the form of 8-mm-diam disks, were flushed against the back wall of the PA cell, which has a 4-mm-diam hole through which the rear beam is incident. The polymer samples studied were the following: Teflon, 180 μm thick; polyvinyl chloride (PVC), 180 μm thick; cellulose acetate, 170 μm thick; polypropylene, 200 μm thick; low-density polyethylene, 190 μm thick. To ensure the optical opacity condition implicit in Eqs. (1) and (2), a thin circular Al foil (20 μm thick) of 3 mm diam was attached to each side of

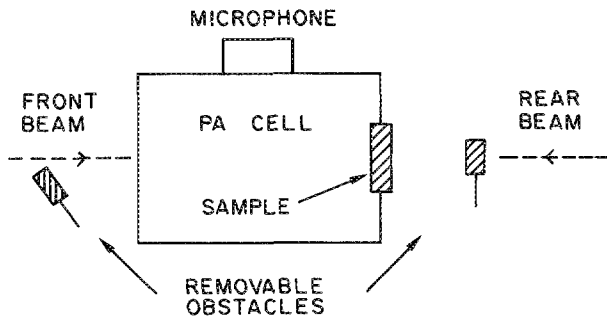


FIG. 1. Schematic arrangement for the two-beam photoacoustic measurement of the thermal diffusivity.

the sample using a thin layer of diffusion pump oil. In this way, we ensured the optical surface absorption condition, as discussed in Ref. 1.

III. RESULTS AND DISCUSSION

As mentioned in the Introduction, the thermal diffusion model applied to the phase-lag method worked only at low frequencies (6–12 Hz range). This is seen in Fig. 2, in which the semilog plot of the rear-signal amplitudes for the low-density polyethylene (LDPE) and the Teflon (PTFE) samples are shown as a function of the modulation frequency. In the 6–12-Hz frequency range, the rear-signal amplitude is dominated by an exponential behavior [$\sim \exp(-a\sqrt{f})$], as predicted by the thermal diffusion model, for a thermally thick sample. However, for frequencies greater than 15 Hz the rear signal behaves approximately as $1/f$, as shown by the solid curves in Fig. 3. This $1/f$ dependence for the rear-signal amplitude of a thermally thick sample means that, in this frequency range, the observed signal is certainly not described by the thermal diffusion model.

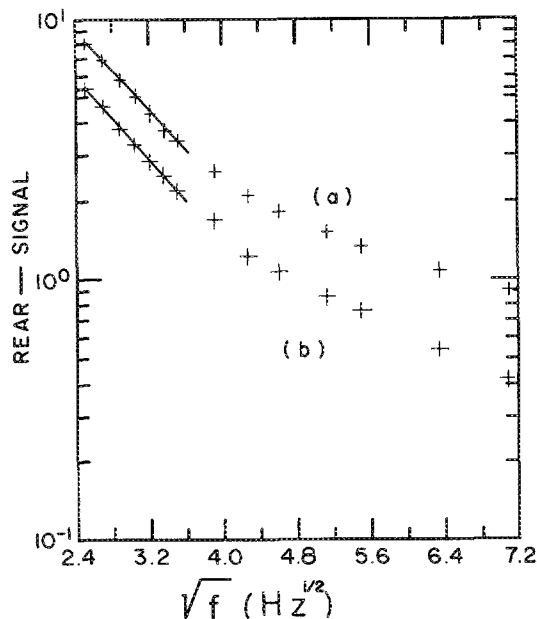


FIG. 2. Semilog plot of the rear-signal amplitude vs \sqrt{f} for (a) LDPE and (b) PTFE.

To explain the observed behavior of the front and rear signals of our polymer samples, we resort to the composite piston model of Rousset, Lepoutre, and Bertrand.¹⁰ According to this model, the PA signal is due to two contributions: One is the sample to gas thermal diffusion and the other is the thermoelastic bending. Using Eqs. (16)–(20) of Ref. 10 and carrying out some straightforward calculations for the different illumination configurations, the pressure in the PA cell can be written as

$$P_R = \frac{\gamma P_0 I_R}{T_0 l_g k_s \sigma_s \sigma_g} \left[\frac{1}{\sinh(l_s \sigma_s)} + \frac{3R^4 \alpha_T T_0}{R_c^2 l_s^2 l_s \sigma_s} \left(\frac{\alpha_s}{\sigma_g} \right)^{1/2} \times \frac{\cosh(l_s \sigma_s) - (l_s \sigma_s / 2) \sinh(l_s \sigma_s) - 1}{\sinh(l_s \sigma_s)} \right], \quad (3)$$

$$P_F = \frac{\gamma P_0 I_F}{T_0 l_g k_s \sigma_s \sigma_g} \left[\frac{\cosh(l_s \sigma_s)}{\sinh(l_s \sigma_s)} + \frac{3R^4 \alpha_T T_0}{R_c^2 l_s^2 l_s \sigma_s} \left(\frac{\alpha_s}{\sigma_g} \right)^{1/2} \times \frac{(l_s \sigma_s / 2) \sinh(l_s \sigma_s) - \cosh(l_s \sigma_s) + 1}{\sinh(l_s \sigma_s)} \right], \quad (4)$$

for the rear and front illumination, respectively. In Eqs. (3) and (4), P_0 , T_0 , and γ are the ambient pressure, temperature, and specific-heat ratio of air. l_i is the length of material i ($i = s, g$; s -sample, g -air), k_s is the thermal conductivity of material i , $\sigma_i = (1 + j)a_i$ with $a_i = (\pi f / \alpha_i)^{1/2}$, α_i is the thermal diffusivity of material i , R_c is the PA cell radius, R is the support radius for the sample, and α_T is the thermal expansion coefficient of the sample. The first terms in the brackets of Eqs. (3) and (4) correspond to the thermal diffusion contributions whereas the last terms are due to the thermoelastic bending. For a thermally thick sample ($l_s a_s \gg 1$), as in the case of our experiment, the thermal diffusion model predicts an exponentially decreasing signal for the rear signal and a $1/f$ dependence for the front signal; i.e.,

$$P_{R,th} \sim (1/f) e^{-a\sqrt{f}}, \quad P_{F,th} \sim 1/f, \quad (5)$$

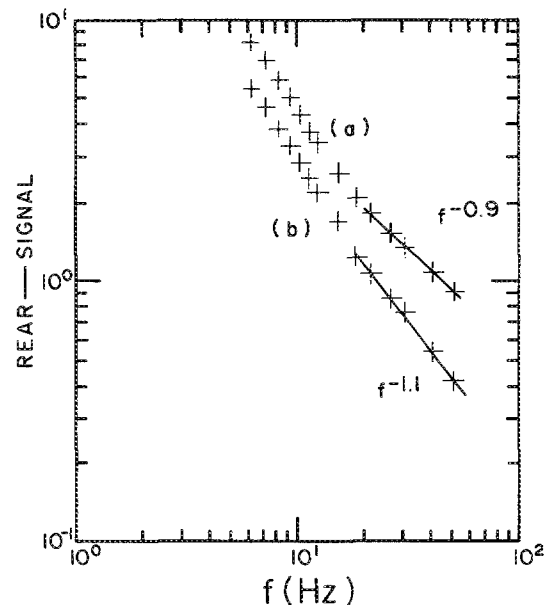


FIG. 3. Log-log plot of the rear-signal amplitude vs the modulation frequency f for (a) LDPE and (b) PTFE.

TABLE I. Thermal diffusivity values obtained from the front-phase data fitting.

Material	Front-phase data fitting (cm ² /s)	Literature values ^a (cm ² /s)
Teflon	0.0014	0.0011
PVC	0.0006	0.0008
Cellulose acetate	0.0008	0.0009
Polypropylene	0.0007	0.0008
Low-density polyethylene	0.0016	0.0016

^a Reference 11.

where $a = l_s(\pi/\alpha_s)^{1/2}$. In contrast, the thermoelastic contribution predicts a $1/f$ frequency dependence for both rear and front illumination. The ratio of the thermoelastic to thermal diffusion contributions depends on the thermal properties α_T and α_s of the sample, as well as on geometrical factors. Nevertheless, it should dominate at high frequencies where the thermal diffusion contribution is exponentially damped out.

It follows from the above discussion that at low frequencies (6–12 Hz) the thermal diffusion model accounts very well for our data, and the phase-lag method is justified for determining the thermal diffusivity. In the frequency range above 15 Hz, where S_R behaves as $1/f$, the thermal diffusivity may be obtained from the thermoelastic phase contribution. To check this, we have fitted our phase data, for $f > 15$ Hz, with the corresponding expressions for the thermoelastic phase contribution of a thermally thick sample, namely [cf. Eqs. (3) and (4)],

$$\phi_F = -\pi/2 + \arctan[1/(z-1)] \quad (6)$$

and

$$\phi_R = \pi/2 + \arctan[1/(z-1)], \quad (7)$$

where $z = l_s(\pi f/\alpha_s)^{1/2}$. We note from Eqs. (6) and (7) that the phase lag between the front and rear signals is now a

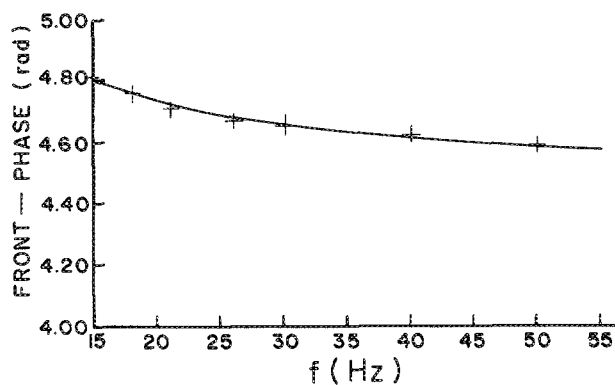


FIG. 4. Front-signal phase dependence on the modulation frequency for the PTFE sample. The solid curve is the result of the data fitting to the thermoelastic contribution to the front phase.

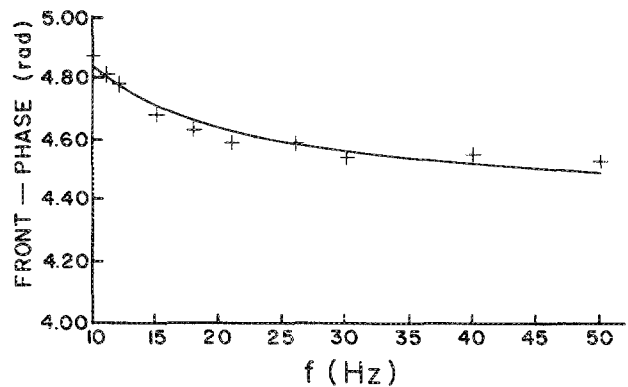


FIG. 5. Front-signal phase dependence on the modulation frequency for the LDPE sample. The solid curve is the result of the data fitting to the thermoelastic contribution to the front phase.

constant. This means that the thermal diffusivity in this case is obtained from the frequency dependence of one of the signal phase measurements. In Table I we show the results of the front-phase data fitting using Eq. (6). In Figs. 4 and 5 we show the frequency dependence of the front-signal phase for two typical samples, namely, PTFE and LDPE, respectively, as well as the curves resulting from the data fitting. Also listed in Table I are the values of the thermal diffusivity quoted in the literature.¹¹

The above results tell us that some caution should be exercised when measuring the thermal diffusivity of foil samples with large thermal expansion coefficients, as in the case of polymers. In this case, the simple RG thermal diffusion model is no longer straightforwardly applicable to describe the PA signal at all frequencies. This was manifested by the failure of the phase-lag method in determining the thermal diffusivity of our polymer foils in the frequency range of 6–100 Hz. For these samples, the thermoelastic bending was proven to be the dominant mechanism responsible for the PA signal in almost the entire frequency range, except at a limited range, in the low-frequency region (6–12 Hz). The thermal diffusivity is then obtained from the fitting of the front-phase data to the corresponding expression due to the thermoelastic contribution, namely Eq. (6).

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