An inverse problem for thermal diffusivity estimation with the
photoacoustic spectroscopy

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Abstract. In the present work we investigate the estimation of the thermal diffusivity with an
inverse problem approach for photoacoustic spectroscopy (PAS). The direct problem is solved
analytically using Rosencwaig-Gersho (RG) theory and the inverse problem is solved using the
method of Levenberg-Marquardt. A sensitivity analysis is presented. We used real
experimental data for the amplitude and phase-lag of the photoacoustic signal, acquired with a
PAS experimental apparatus. A discussion is presented on the results obtained for the thermal
diffusivity of a glass sample.

1. Introduction
The Photoacoustic Spectroscopy (PAS) [1], or more generally the Photothermal Spectroscopy [2-4],
are non-destructive testing methodologies that have been applied for the thermal and optical
characterization of materials [5-7].

The photoacoustic effect is the basic phenomenon upon which PAS is built, and it occurs when a
material sample placed inside a closed cell filled with air is illuminated with periodically interrupted
light. The light absorbed by the sample is converted into heat through a nonradiative de-excitation
process. The periodic flow of heat into the air chamber of the cell produces, as a thermal piston,
pressure disturbances in it, which can be detected by a microphone mounted at the cell wall. In the
model developed by Rosencwaig and Gersho, known as RG theory [8], this is the only phenomenon
taken into account in the photoacoustic (PA) signal.

The periodic heat flow from the sample to the surrounding gas, in the photoacoustic cell, depends:
on the amount of the incident radiation absorbed by the sample; on the light-into-heat conversion
efficiency; and on the heat diffusion through the sample. In this way, PAS can be used not only as a
thermal and optical characterization technique, but also as a technique for photosynthesis [9,10] and
photovoltaic [11,12] studies, where the detected PA signal is a result of concurrent processes between
light-into-heat conversion and photochemical and photoelectrical activities, respectively.

Other phenomena besides the thermal diffusion, photochemical and photoelectrical activities, may
be responsible for the signal generation detected by the microphone. The cyclical expansion and
contraction of the solid sample is described by the thermal expansion model (13), and the cyclical
thermoelastic bending of the solid sample due the variable absorption of the light with the depth of the
sample is described by the thermoelastic bending model (14).
Even when only one of the phenomena described above is taken into account, as is the case of the thermal diffusion in the RG theory, the analytical solution of the problem, if available, may become difficult to obtain and interpret. To make the problem more tractable special cases usually are examined for the sake of acquiring physical insight, but in such cases the applicability of the technique may be limited or even impaired. An alternative approach may be the use of numerical methods.

The inverse problem of optical and thermal properties estimation relies heavily on accurate physical, mathematical and computational modeling of the phenomena involved. Several methods have been developed for the solution of inverse problems and some of them are described in the books [15-20], just to name a few.

In a previous work [21] we used an implicit inverse problem formulation, and the Levenberg-Marquardt method, for the PAS with the direct problem modeled with the full analytical expression derived by RG theory. As synthetic experimental data it was used only the amplitude of the steady periodic temperature established at the surface of the material sample that is next to the air chamber of the closed photoacoustic cell. We were able to estimate, separately, the thermal diffusivity, $\alpha$, the thermal conductivity, $k$, and the optical absorption coefficient, $\beta$, of the material under analysis. However, it was not possible to estimate any pair of coefficients simultaneously.

In another work [22] we have extended our previous results by considering also as synthetic experimental data the phase-lag between the temperature at the sample-gas interface and the modulated light source. An improvement on the solution of the inverse problem was observed (smaller confidence bounds) when each parameter was estimated separately, except for the thermal conductivity, due to the null sensitivity of the phase-lag with respect to this parameter. The simultaneous estimation of ($\alpha, \beta$) was performed but the estimated values for the unknowns were corrupted by the amplification of the error present in the synthetic experimental data.

In the following works [23,24] we constructed a one parameter family of regularization terms with Bregman distances based on the $q$-discrepancy function, which was implemented in the formulation and solution of PAS as an inverse problem. The original idea was improved by the proper weighting of the unknowns to be determined. We have focused on the simultaneous estimation of the sample thermal diffusivity, $\alpha$, and optical absorption coefficient, $\beta$. The results were significantly improved in comparison to our previous works [21,22].

In the present work we present a sensitivity analysis, and real experimental data acquired with a PAS experimental apparatus. Using the real experimental data on the amplitude of the PA signal we were able to estimate the thermal diffusivity of a glass sample.

2. Direct problem – RG Theory

Consider the cylindrical closed photoacoustic cell represented schematically in figure 1. The sample of the material under analysis is placed upon a backing material, and the other boundary of the sample adjacent to the air chamber of the cell, is exposed to an incident modulated light with intensity

$$I(t) = \frac{1}{2} I_0 [1 + \cos(\omega \cdot t)]$$

(1)

where $I_0$ is the maximum intensity of the incident light, $\omega$ is the angular frequency of the chopping mechanism, and $t$ represents time.

It is assumed that the light doesn’t go through any interaction within the air chamber and is fully absorbed by the material sample according to Beer’s law

$$I_s(x,t) = e^{-\beta x} I(t)$$

(2)

where $\beta$ is the optical absorption coefficient, the subscript $s$ represents the sample, and $x$ is the space coordinate representing the depth in the sample starting at the interface between the sample and the air chamber, as shown in figure 1.
The volumetric heat generation at the sample due to the light absorbed is given by

$$S(x,t) = \frac{dI_g(x,t)}{dx} = \frac{1}{2} \beta \cdot I_0 e^{\beta x} [1 + \cos(\omega \cdot t)]$$

(3)

and the mathematical formulation of the heat conduction problem in the photoacoustic cell is given by

$$\frac{\partial^2 \theta_g(x,t)}{\partial x^2} = \frac{1}{\alpha_g} \frac{\partial \theta_g(x,t)}{\partial t}, \quad 0 < x < l_g$$

(4a)

$$\frac{\partial^2 \theta_s(x,t)}{\partial x^2} = \frac{1}{\alpha_s} \frac{\partial \theta_s(x,t)}{\partial t} - \frac{\beta \cdot I_0}{2k_s} e^{\beta x} [1 + e^{i\omega t}], \quad -l_s < x < 0$$

(4b)

$$\frac{\partial^2 \theta_b(x,t)}{\partial x^2} = \frac{1}{\alpha_b} \frac{\partial \theta_b(x,t)}{\partial t}, \quad -(l_b + l_s) < x < -l_s$$

(4c)

with the interface conditions given by

$$\theta_g(0,t) = \theta_s(0,t), \quad \theta_s(-l_s,t) = \theta_b(-l_s,t)$$

(4d-e)

$$k_s \frac{\partial \theta_s(x,t)}{\partial x} \bigg|_{x=0} = k_g \frac{\partial \theta_g(x,t)}{\partial x} \bigg|_{x=0}, \quad k_s \frac{\partial \theta_s(x,t)}{\partial x} \bigg|_{x=-l_s} = k_b \frac{\partial \theta_b(x,t)}{\partial x} \bigg|_{x=-l_s}$$

(4f-g)

and the initial conditions given by

$$\theta_g(x,0) = 0, \quad 0 \leq x \leq l_g, \quad \theta_s(x,0) = 0, \quad -l_s \leq x < 0, \quad \theta_b(x,0) = 0, \quad -(l_i + l_b) \leq x \leq -l_s$$

(4h-j)

where $j$ is the imaginary number $\sqrt{-1}$, $\theta$ is the complex valued temperature, $k$ represents the thermal conductivity, $\alpha$ the thermal diffusivity, and the subscripts $g$, $s$, and $b$ denote air (gas), sample and backing material, respectively.

The complete solution for problem (4) is given in [1,8,21]. Here we are interested only in the temperature at the sample-gas interface, i.e. $x = 0$,

$$\theta(0,t) = F_0 + \theta_0 e^{j\omega t}$$

(5)

where $F_0$ is the time-independent (d.c.) component and $\theta_0 e^{j\omega t}$ is the time-dependent (a.c.) component of the solution at $x = 0$, where $\theta_0$ is a complex valued number given by

$$\theta_0 = \left[ \begin{array}{c} P_1 - P_2 + P_3 \\ P_4 - P_5 \end{array} \right] H$$

(6a)

$$p_1 = (r-1)(b+1)e^{\sigma_1 l_s}, \quad p_2 = (r+1)(b-1)e^{-\sigma_1 l_s}, \quad p_3 = 2(b-r)e^{-\beta l_s},$$

(6b-d)

$$p_4 = (g+1)(b+1)e^{\sigma_1 l_s}, \quad p_5 = (g-1)(b-1)e^{-\sigma_1 l_s},$$

(6e-f)

Figure 1. Schematically representation of the closed photoacoustic cell.

The surrounding air layer is made up of a backing material, a sample and modulated light incident over a microphone.
\[
H = \frac{\beta \cdot I_0}{2k_s} \left( \beta^2 - \sigma_s^2 \right), \quad b = \frac{k_s a_b}{k_s a_g}, \quad g = \frac{k_s a_g}{k_s a_s}, \quad r = \frac{(1 - j) \beta}{2a_s} \tag{6g-j}
\]

\[
a_s = \left( \frac{\omega}{2a_s} \right)^{1/2}, \quad a_b = \left( \frac{\omega}{2a_b} \right)^{1/2}, \quad a_g = \left( \frac{\omega}{2a_g} \right)^{1/2}, \quad \sigma_s = (1 + j) a_s \tag{6k-n}
\]

With the PAS experimental apparatus, we measure the a.c. component of the temperature (second term on the right hand side of equation (5)), and only the real part is of physical interest. Therefore, we choose only the terms

\[
\text{Re\left[\theta(0,t)_{ac}\right]} = |\theta_0| \cos(\omega \cdot t + \phi) \tag{7}
\]

Writing,

\[
\theta_0 = \theta_1 + j \theta_2, \quad \text{where} \quad \theta_1 = \text{Re}[\theta_0] \quad \text{and} \quad \theta_2 = \text{Im}[\theta_0] \tag{8}
\]

and

\[
\theta_0 = |\theta_0| e^{j\phi} \tag{9}
\]

we obtain the amplitude

\[
A^\theta = |\theta_0| = \sqrt{\theta_1^2 + \theta_2^2} \tag{10}
\]

and the phase-lag

\[
\phi^\theta = \arctan\left(\frac{\theta_2}{\theta_1}\right) \tag{11}
\]

The superscript \(\theta\), in equations (10) and (11), denotes that the amplitude and phase-lag are related to temperature at \(x = 0\).

If we know the optical and thermal properties of the sample, the thermal properties of the other materials in the photoacoustic cell, the physical dimensions \(l_s, l_b\) and \(l_g\) represented in figure 1, the frequency of the chopping mechanism, and the intensity of the incident light, then equations (10) and (11) provide the calculated values for the amplitude and phase-lag of the temperature at the interface sample-gas between the material and the air chamber at \(x = 0\).

As, in this work, we are using real experimental data acquired with the PAS experimental apparatus shown schematically in figure 2, it is necessary to have the amplitude and phase-lag related to the PA signal detected by the microphone, i.e. an analytical expression related to the voltage measured.

As presented by RG theory [1,8], the acoustic pressure wave produced in the photoacoustic cell is

\[
\delta P(t) = \frac{\gamma P_0 \theta_0}{a_g l_g T_0 \sqrt{2}} e^{j(\omega t - \pi/4)} \tag{12}
\]

where \(\gamma\) is the ratio of specific heats, \(P_0\) is the ambient pressure and \(T_0\) is the ambient temperature of the photoacoustic cell. However, the PA signal measured by the microphone and the synchronous amplifier is

\[
\delta V(t) = G \cdot \frac{\gamma P_0 \theta_0}{a_g l_g T_0 \sqrt{2}} e^{j(\omega t - \pi/4)} = |V| e^{j(\omega t - \pi/4)} \tag{13}
\]

where \(G\) is the pressure to the voltage transducer gain factor. In this way, similarly to the equations (7)-(11), we obtain the PA signal amplitude

\[
A^v = |V| = \left| G \cdot \frac{\gamma P_0 \theta_0}{a_g l_g T_0 \sqrt{2}} \right| = \sqrt{V_1^2 + V_2^2} \tag{14}
\]

and the PA signal phase-lag

\[
\phi^v = \arctan\left(\frac{V_2}{V_1}\right) \tag{15}
\]
The superscript \( v \), in equations (14) and (15), denotes that the amplitude and phase-lag are related to voltage measured with the microphone and synchronous amplifier.

Figure 2. PA experimental apparatus.

Now, with the experimental data, we brought more parameters for the direct problem, as compared with the previous works [21-24], i.e. \( \gamma, P_0, V, G, I_0, I_g \) and \( a_g \). It increased the already complicated problem. However, we provide a solution which is based on using the normalized PA signal, where all the acquired (and calculated) PA signal is divided by the first experimental data (related to the smallest frequency). We will refer to the normalized amplitude and to the relative phase-lag of the PA signal as amplitude, \( A \), and phase-lag, \( \phi \), respectively, in the next sections.

3. The inverse problem

We are interested in investigating the feasibility of the estimation of the following vectors of unknowns

\[
\begin{align*}
\vec{Z}_1 &= \{\alpha\}, \\
\vec{Z}_2 &= \{\beta\}, \\
\vec{Z}_3 &= \{k\}, \\
\vec{Z}_4 &= \{\alpha, \beta\}^T, \\
\vec{Z}_5 &= \{\alpha, k\}^T, \\
\vec{Z}_6 &= \{\beta, k\}^T, \\
\vec{Z}_7 &= \{\alpha, \beta, k\}^T
\end{align*}
\]  
(16a-g)

where the superscript \( T \) denotes transpose.

For each modulation frequency used in the PAS experiment, i.e. \( f_i \), with \( i = 1,2,\ldots,N_f \), where \( f_i = \omega_i/2\pi \) and \( N_f \) is the total number of frequencies considered, we acquire the experimental data on both the amplitude, i.e. \( A_{\text{exp},i} \), \( i = 1,2,\ldots,N_f \), and the phase-lag, i.e. \( \phi_{\text{exp},i} \), \( i = 1,2,\ldots,N_f \).

As the number of available data is larger than the number of unknowns, the inverse problem is formulated implicitly as an optimization one [25] in which we seek to minimize the squared residues cost functional

\[
S(\vec{Z}) = \sum_{i=1}^{2N_f} \left[ C_i(\vec{Z}) - E_i \right] \left[ C_i(\vec{Z}) - E_i \right]^T = \vec{R}^T \vec{R}
\]  
(17)

where \( C_i(\vec{Z}) \) represents the calculated values for the amplitude, \( A_{\text{calc},i} \), when \( i = 1,2,\ldots,N_f \), and the calculated values for the phase-lag, \( \phi_{\text{calc},i} \), when \( i = N_f + 1, N_f + 2,\ldots,2N_f \), and \( E_i \) are the respective experimental data, i.e. \( A_{\text{exp},i} \), for \( i = 1,2,\ldots,N_f \), and \( \phi_{\text{exp},i} \), for \( i = N_f + 1, N_f + 2,\ldots,2N_f \). Therefore the vector of residues is given by

\[
\vec{R} = \{ A_{\text{calc}_1} - A_{\text{exp}_1}, \ldots, A_{\text{calc}_{N_f}} - A_{\text{exp}_{N_f}}, \phi_{\text{calc}_1} - \phi_{\text{exp}_1}, \ldots, \phi_{\text{calc}_{N_f}} - \phi_{\text{exp}_{N_f}} \}^T
\]  
(18)
We have made an adjustment by a constant factor in the calculated and experimental values for the amplitudes to make them of the same order of magnitude of the phase-lag values.

Applying the Levenberg-Marquardt method [26] we start the iterative procedure with an initial guess for the vector of unknowns, \( \bar{Z}^0 \), and new estimates are obtained with

\[
\bar{Z}^{n+1} = \bar{Z}^n + \Delta \bar{Z}^n
\]

where \( n \) is the iteration index. The corrections \( \Delta \bar{Z}^n \) are obtained from the solution of the following system of linear equations

\[
(J^n)\bar{Z}^n + (\lambda^n)I\Delta \bar{Z}^n = -J^n \bar{R}^n
\]

\( I \) is the identity matrix, \( \lambda^n \) is a damping parameter, and \( J \) is the Jacobian matrix whose elements are given by

\[
J_{pq} = \frac{\partial C_p}{\partial Z_q}, \quad p = 1, 2, ..., 2N_f \quad \text{and} \quad q = 1, 2, ..., M
\]

where \( M \) is the total number of unknowns. For \( \bar{Z}_1, \bar{Z}_2 \) and \( \bar{Z}_3 \) we have \( M = 1 \), for \( \bar{Z}_4, \bar{Z}_5 \) and \( \bar{Z}_6 \), \( M = 2 \), and for \( \bar{Z}_7 \), \( M = 3 \).

The parameter \( \lambda^n \) is reduced from one iteration to the next according to the procedure proposed by Marquardt [26].

The Levenberg-Marquardt iterative procedure is interrupted when a stopping criterion such as

\[
\frac{\Delta Z_q^n}{Z_q^n} < \varepsilon, \quad q = 1, 2, ..., M
\]

where \( \varepsilon \) is a tolerance, say \( 10^{-5} \).

The elements of the Jacobian matrix given by equation (21) as well as the vector of residues given by equation (18) were calculated using the direct problem solution described in the previous section. For the former a central finite difference approximation was used in the computation of the derivatives.

4. Results and discussions

As far as we know, only partial formulations of the RG model has been used previously for thermal and optical parameter estimation where special cases of opaqueness and thermal behavior of material are considered. In the present work we are interested in the use of the general RG model for such parameter estimation. The full analytical expression for the PA signal, as shown before, is quite complex and to interpret it is necessary to carry out a sensitivity study of the parameters with respect to the overall experimental conditions. In table 1 is presented a summary of the process, thermal and optical parameters used (some of them was varied during the sensitivity study). We choose, as a sample, the commercial glass used as an eye protection for electrical welding workers. The thermal diffusivity of the opaque glass was determined indirectly using the literature value of the density for a crown glass and two others experimental apparatus (not presented here) where we measured the thermal conductivity and the optical absorption coefficient. These values are in agreement with the range of these values we can observe in the literature for crown and Pyrex glasses.

Sensitivity analysis

Therefore, we decided to calculate the modified sensitivity coefficients [27] for both amplitude, \( X^A \), and phase-lag, \( X^\phi \), data with respect to the unknowns \( \alpha_i \), \( \beta \), and \( k_i \), i.e. unknown parameters \( \bar{Z}_j \).
where \( i \) represents amplitude or phase-lag.

The variation of the modified sensitivity coefficients related to the amplitude and phase-lag data, with the frequency of the modulated light source, are presented in figures 3, and 4, respectively, for sample thickness equal to \( l_s = 1,50 \times 10^{-3} \text{ m} \), and in figures 5 and 6 for \( l_s = 0,20 \times 10^{-3} \text{ m} \). All the other parameters were those given in table 1.

### Table 1. Process, thermal and optical parameters for the photoacoustic cell.

| Backing material | \( \alpha_b = 3,4 \times 10^{-5} \text{ m}^2/\text{s} \) | \( k_b = 108,8 \text{ W/mK} \) |
|------------------|----------------------------------|-----------------|
| Sample           | \( \alpha_s = 1,127 \times 10^{-7} \text{ m}^2/\text{s} \) | \( \beta = 4,0 \times 10^3 \text{ m}^{-1} \) |
| Gas              | \( \alpha_g = 1,9 \times 10^{-5} \text{ m}^2/\text{s} \) | \( k_g = 0,0239 \text{ W/mK} \) |
| Light source     | \( I_0 = 100 \text{ W/m}^2 \) | \( f = 1,2,3,...,50 \text{ Hz} \) |

As can be seen in the figures 3-6, the sensitivity response of the amplitude and phase-lag with modulation frequency improves when we decrease both the frequency and the sample thickness. However, unfortunately, the best conditions we found were not possible to be put in practice because we have instrumental limitations for frequencies below 4 Hz and also thin samples became transparent. It means that too much light arrived on the backing material which was not supposed to absorb light in the RG model, what proved not to be true in real experiments.

### Real experiment

Therefore, we decide to work with the sample thickness equal to \( l_s = 1,50 \times 10^{-3} \text{ m} \) and frequency range from 4 up to 25 Hz. In figures 7 and 8 are presented three real experimental runs and the corresponding simulation for PA amplitude and PA phase-lag. In the simulation we used all the parameters given in table 1 including also \( P_0 \), \( V \), the literature values for \( \gamma \), and the approximate value of \( G \) that we obtained experimentally. As can be seen in figures 7 and 8, there is a good agreement with respect to amplitude data and a very poor agreement with respect to phase-lag data. Up to now we do not have an adequate explanation for this PA phase-lag behavior. As we showed in the previous work [22-24], where we used synthetic data, the phase-lag data improved the inverse problem solutions. In this way, with those experimental data, we return back to the case of one parameter estimation, since once without phase-lag data is not possible to obtain two parameters simultaneously. Therefore, we decided to estimate only the thermal diffusivity of the glass sample, using the real experimental data of the amplitude of the PA signal.
Figure 3. Variation of the modified sensitivity coefficients related to the amplitude with the frequency of modulation. \( l_s = 1.50 \times 10^{-3} m \)

Figure 4. Variation of the modified sensitivity coefficients related to the phase-lag with the frequency of modulation. \( l_s = 1.50 \times 10^{-3} m \)

Figure 5. Variation of the modified sensitivity coefficients related to the amplitude with the frequency of modulation. \( l_s = 0.20 \times 10^{-3} m \)

Figure 6. Variation of the modified sensitivity coefficients related to the phase-lag with the frequency of modulation. \( l_s = 0.20 \times 10^{-3} m \)

Thermal diffusivity

Taking the experimental data measured in one of the experimental runs shown in figures 7 and 8, we applied the inverse problem approach, using the full expression of the RG model, as presented before. Here we used the normalized PA signal. The results for both experimental and calculated PA amplitude and for the residues are presented in figures 9 and 10, respectively. The thermal diffusivity value obtained was \( \alpha_s = (15.4 \pm 0.3) \times 10^{-7} m^2 / s \), which is within the range of values given in the literature for crown and Pyrex glasses.
**Figure 7.** Experimental and simulated results for the PA amplitude as a function of the frequency.

**Figure 8.** Experimental and simulated results for the PA phase-lag as a function of the frequency.

**Figure 9.** Experimental and inverse problem calculated results for the normalized PA amplitude as a function of the frequency.

**Figure 10.** Residues of the experimental and inverse problem calculated results for the normalized PA amplitude as a function of the frequency.

### 5. Conclusions
This is a ongoing work. In previous works [21-24] we obtained good results in applying the inverse problem approach to photoacoustic spectroscopy, when we used synthetic experimental data. Here, the main goal was to use the real experimental data in such approach. We tried to show how the sensitivity analysis can help us in the inverse approach once we can find the best process and parameters conditions for the real experimental apparatus in order to acquire the real experimental data. Due to the fact that the results predicted by the full analytical expression, derived by RG theory, did not agree with the real experimental data for the PA phase-lag, we decided to estimate only the thermal diffusivity of an opaque sample glass using the PA amplitude data. We are still looking for the simultaneous estimations of two or more parameters. Therefore, more investigation, both theoretical and experimental, must be performed.

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