

A REGULARIZED SOLUTION FOR THE INVERSE PROBLEM OF PHOTOACOUSTIC SPECTROSCOPY

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Abstract - In the present work we propose the use of weighted Bregman distances in the construction of regularization terms for the Tikhonov's functional applied for the formulation and solution of the inverse problem of photoacoustic spectroscopy. Test case results demonstrate that better estimates were obtained for the simultaneous estimation of the thermal diffusivity and optical absorption coefficient using as synthetic experimental data the information on both the amplitude and phase-lag of the temperature at the interface sample-gas between the material under analysis and the air chamber of the closed photoacoustic cell.

1. INTRODUCTION

The Photoacoustic Spectroscopy (PAS) [24], or more generally the Photothermal Spectroscopy [17], are non-destructive testing methodologies that have been applied for the thermal and optical characterization of materials [13, 23, 27, 30, 32]. There are other applications under development, such as gases monitoring [11] and investigation on thermal contact resistance for copper coatings on carbon surfaces [21].

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 an acoustic 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 [25], known as RG theory, this is the only phenomenon taken into account in the PAS signal.

In a previous work [28] we used an implicit inverse problem formulation, and the Levenberg-Marquardt method, for the PAS with the direct problem modeled with the RG theory. As 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, α , the thermal conductivity, k , and the optical absorption coefficient, β , of the material under analysis. However, it was not possible to estimate any pair of coefficients simultaneously.

In [4] we extended our previous results [28] by considering also as 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 (α, β) was performed but the estimated values for the unknowns were corrupted by the amplification of the error present in the experimental data. For a set of experimental data with 3% noise, the confidence bounds for the estimates were of the order of 8%. Using also a set of 3% noisy experimental data, we attempted to estimate simultaneously (α, k) or (β, k) but the confidence bounds were, respectively, of the order of 14% and 7%. The range of the modulated frequency for external illumination was shifted from 5-17 Hz (in [28]) to 1-8 Hz (in [4]) in order to have a higher sensitivity to the parameters to be estimated.

In both works [4,28], it was required, for most of the test cases, the use of the damping factor in the Levenberg-Marquardt method in order to achieve convergence.

In order to deal with the effects of the noise present in the experimental data, Tikhonov's regularization [29] is the most well known approach used for the solution of ill-posed problems, and it has been used in numerous different areas of application [5, 8, 9, 18, 19]. Much work has been done on the analysis and proposition of regularization terms for Tikhonov's functional [10, 14, 20, 26, 31], and the proper choice of the regularization parameter is of key importance for the implementation of such an approach for the solution of inverse problems [3, 12, 15].

Cidade *et al.* [6, 7] proposed the use of Bregman distances [2] as Tikhonov's regularization terms for one application in the restoration of atomic force microscopy nanoscale images. Using Csiszár's measure [16], called q -discrepancy, a family of regularization terms was constructed. Berrocal Tito *et al.* [1] and Pinheiro *et al.* [22] extended this idea by using moments of the q -discrepancy. The former work [1] is related to the estimation of parameters in an environmental model, and the latter [22] deals with an inverse problem of radiative properties estimation.

In the present work a one parameter family of regularization terms constructed with Bregman distances based on the q -discrepancy function is implemented in the formulation and solution of PAS as an inverse problem. The original idea [6,7] was improved by the proper weighting of the unknowns to be determined, and we use here the denomination weighted Bregman distances. We have focused on the simultaneous estimation of the sample thermal diffusivity, α , and optical absorption coefficient, β . The results are significantly improved in comparison to our previous works [4, 28].

The effects of the parameter q used in the construction of the regularization terms, as well as those of the regularization parameter λ are investigated. Some test case results are presented.

As real experimental data is not yet available, we have used synthetic experimental data. The experimental apparatus is available in our institution, and in the near future we will be able to acquire real experimental data. Before dealing with the difficulties associated with the real experiments, we decided to perform the numerical simulations in order to evaluate the best conditions in which the experiments will be performed.

2. MATHEMATICAL FORMULATION AND SOLUTION OF THE 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 adjoint to the air chamber of the cell, is exposed to an incident modulated light with intensity

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

where I_0 is the maximum intensity of the incident light, and ω is the angular frequency of the chopping mechanism.

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

$$I_s(x, t) = e^{\beta x} I(t) \quad (2)$$

where β is the optical absorption coefficient.

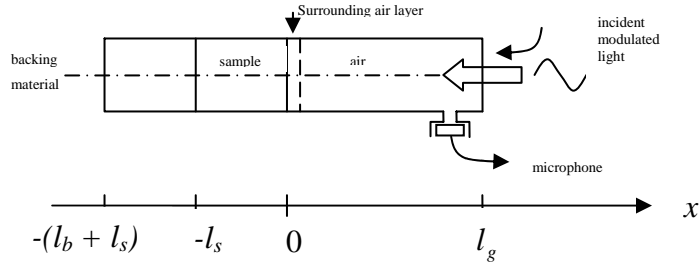


Figure 1. Schematical representation of the closed photoacoustic cell.

The volumetric heat generation at the sample due to the light absorbed is given by

$$S(x, t) = \frac{dI_s(x, t)}{dx} = \frac{1}{2} \beta I_0 e^{\beta x} (1 + \cos \omega t) \quad (3)$$

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

$$\frac{\partial^2 \theta_g(x, t)}{\partial x^2} = \frac{1}{\alpha_g} \frac{\partial \theta_g(x, t)}{\partial t}, \quad 0 < x < l_g \quad (4a)$$

$$\frac{\partial^2 \theta_s(x, t)}{\partial x^2} = \frac{1}{\alpha_s} \frac{\partial \theta_s(x, t)}{\partial t} - \frac{\beta I_0}{2k_s} e^{\beta x} (1 + e^{j\omega t}), \quad -l_s < x < 0 \quad (4b)$$

$$\frac{\partial^2 \theta_b(x, t)}{\partial x^2} = \frac{1}{\alpha_b} \frac{\partial \theta_b(x, t)}{\partial t}, \quad -(l_b + l_s) < x < -l_s \quad (4c)$$

with the interface conditions given by

$$\theta_s(0, t) = \theta_g(0, t), \quad \theta_s(-l_s, t) = \theta_b(-l_s, t) \quad (4d, e)$$

$$k_s \frac{\partial \theta_s(x,t)}{\partial x} \Big|_{x=0} = k_g \frac{\partial \theta_g(x,t)}{\partial x} \Big|_{x=0}, \quad k_s \frac{\partial \theta_s(x,t)}{\partial x} \Big|_{x=-l_s} = k_b \frac{\partial \theta_b(x,t)}{\partial x} \Big|_{x=-l_s} \quad (4f, g)$$

and the initial conditions given by

$$\theta_g(x,0)=0, \quad 0 \leq x \leq l_g, \quad \theta_s(x,0)=0, \quad -l_s \leq x \leq 0, \quad \theta_b(x,0)=0, \quad -(l_s+l_b) \leq x \leq -l_s \quad (4h-j)$$

where j is the imaginary number $\sqrt{-1}$, θ is the complex valued temperature, k represents the thermal conductivity, α 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 [24, 25, 28]. Here we are interested only in the temperature at the sample-gas interface, i.e. $x = 0$,

$$\theta(0,t) = F_0 + \theta_0 e^{j\omega t} \quad (5)$$

where F_0 is the time-independent (dc) component of the solution at $x = 0$, and θ_0 is a complex valued number given by

$$\theta_0 = \left[\frac{p_1 - p_2 + p_3}{p_4 - p_5} \right] H \quad (6a)$$

$$p_1 = (r-1)(b+1)e^{\sigma_s l_s}, \quad p_2 = (r+1)(b-1)e^{-\sigma_s l_s}, \quad p_3 = 2(b-r)e^{-\beta l_s} \quad (6b-d)$$

$$p_4 = (g+1)(b+1)e^{\sigma_s l_s}, \quad p_5 = (g-1)(b-1)e^{\sigma_s l_s}, \quad (6e,f)$$

$$H = \frac{\beta I_0}{2k_s(\beta^2 - \sigma_s^2)}, \quad b = \frac{k_b a_b}{k_s a_s}, \quad g = \frac{k_g a_g}{k_s a_s}, \quad r = \frac{(1-j)\beta}{2a_s} \quad (6g-j)$$

$$a_s = \left(\frac{\omega}{2\alpha_s} \right)^{1/2}, \quad a_b = \left(\frac{\omega}{2\alpha_b} \right)^{1/2}, \quad a_g = \left(\frac{\omega}{2\alpha_g} \right)^{1/2}, \quad \sigma_s = (1+j)a_s \quad (6k-n)$$

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

$$\text{Re}[\theta(0,t)]_{ac} = |\theta_0| \cos(\omega t + \phi) \quad (7)$$

Writing,

$$\theta_0 = \theta_1 + j\theta_2, \quad \theta_1 = \text{Re}[\theta_0] \quad \text{and} \quad \theta_2 = \text{Im}[\theta_0] \quad \text{and} \quad \theta_0 = |\theta_0| e^{j\phi} \quad (8, 9)$$

we obtain the amplitude

$$A = |\theta_0| = \sqrt{\theta_1^2 + \theta_2^2} \quad (10)$$

and the phase-lag

$$\phi = \arctan\left(\frac{\theta_2}{\theta_1}\right) \quad (11)$$

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 eqns (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 $x = 0$.

3. MATHEMATICAL FORMULATION AND SOLUTION OF THE INVERSE PROBLEM

Consider a vector of unknowns,

$$\vec{Z} = \{Z_1, Z_2, \dots, Z_{N_u}\}^T \quad (12)$$

where Z_i , $i=1,2,\dots,N_u$, are thermal or optical properties of a sample of the material being tested by Photoacoustic Spectroscopy (PAS), and N_u represents the total number of unknowns.

For each modulation frequency used in the PAS experiment, i.e. f_i , $i=1,2,\dots,N_f$, 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

of the steady periodic temperature at $x=0$, i.e. A_{exp_i} , $i=1,2,\dots,N_f$, and the phase-lag ϕ_{exp_i} , $i=1,2,\dots,N_f$.

The inverse problem is then formulated as an optimization problem in which we seek to minimize Tikhonov's regularization functional

$$T(\bar{Z}) = \sum_{i=1}^{2N_f} [C_i(\bar{Z}) - E_i]^2 + \lambda S(\bar{Z}, \bar{Z}^R) = \bar{R}^T \bar{R} + \lambda S(\bar{Z}, \bar{Z}^R) \quad (13)$$

where $C_i(\bar{Z})$ and E_i represent the calculated and experimental values of the amplitude, when $i=1,2,\dots,N_f$, and the calculated and experimental values of the phase-lag, when $i=N_f+1, N_f+2, \dots, 2N_f$, \bar{R} is the vector of residues given by

$$\bar{R} = \{A_{\text{calc}_1} - A_{\text{exp}_1}, \dots, A_{\text{calc}_{N_f}} - A_{\text{exp}_{N_f}}, \phi_{\text{calc}_1} - \phi_{\text{exp}_1}, \dots, \phi_{\text{calc}_{N_f}} - \phi_{\text{exp}_{N_f}}\}^T \quad (14)$$

λ is the regularization parameter, S represents the regularization terms, and \bar{Z}^R is a vector of reference values for the unknowns we want to determine, \bar{Z} . We have made an adjustment by a constant factor in the calculated and experimental values for the amplitudes, A_{exp_i} and A_{calc_i} , $i=1,2,\dots,N_f$, to make them of the same order of magnitude as the phase-lag values.

Using Csiszár's measure [16], here called q -discrepancy [6, 7], we have

$$\eta_q(\bar{Z}) = \frac{1}{1+q} \sum_{i=1}^{N_u} Z_i \frac{Z_i^q - m^q}{q} \quad (15)$$

where m is a measure associated with a prior information (it will cancel out in the calculations to be done next), and the Bregman distance [2, 6,7]

$$D_q(\bar{Z}, \bar{Z}^R) = \eta_q(\bar{Z}) - \eta_q(\bar{Z}^R) - \langle \nabla \eta_q(\bar{Z}^R), \bar{Z} - \bar{Z}^R \rangle \quad (16)$$

a family of one parameter regularization terms can be constructed. From eqns (15) and (16), we obtain

$$D_q(\bar{Z}, \bar{Z}^R) = \frac{1}{1+q} \sum_{i=1}^{N_u} \left\{ Z_i \frac{Z_i^q - Z_i^{Rq}}{q} - Z_i^{Rq} (Z_i - Z_i^R) \right\} \quad (17a)$$

We must stress that varying the parameter q , with $q \geq 0$, then a family of regularization terms is obtained.

By taking the limit $q \rightarrow 0$ in eqn (17), one gets the cross-entropy regularization term

$$D_0(\bar{Z}, \bar{Z}^R) = \sum_{i=1}^{N_u} \left\{ Z_i \ln \left(\frac{Z_i}{Z_i^R} \right) - (Z_i - Z_i^R) \right\} \quad (17b)$$

and with $q=1$ the usual energy regularization term is derived, namely

$$D_1(\bar{Z}, \bar{Z}^R) = \frac{1}{2} \sum_{i=1}^{N_u} (Z_i - Z_i^R)^2 \quad (17c)$$

As the unknowns Z_i , $i=1,2,\dots,N_u$, may be of different orders of magnitude, we propose a modification to eqn. (17a) by introducing a weighting factor $1/f_{Z_i}$, $i=1,2,\dots,N_u$, such that

$$\bar{D}_q(\bar{Z}, \bar{Z}^R) = \frac{1}{1+q} \sum_{i=1}^{N_u} \frac{1}{f_{Z_i}} \left\{ Z_i \frac{Z_i^q - Z_i^{Rq}}{q} - Z_i^{Rq} (Z_i - Z_i^R) \right\} \quad (18a)$$

$\bar{D}_q(\bar{Z}, \bar{Z}^R)$ denominated weighted Bregman distances, for the particular case of $q \rightarrow 0$ results in

$$\bar{D}_0(\bar{Z}, \bar{Z}^R) = \sum_{i=1}^{N_u} \frac{1}{f_{Z_i}} \left\{ Z_i \ln \left(\frac{Z_i}{Z_i^R} \right) - (Z_i - Z_i^R) \right\} \quad (18b)$$

In the present work we consider the weighting factor

$$f_{Z_i} = Z_i^{R^{q+1}}, \quad i = 1, 2, \dots, N_u \quad (19)$$

The regularization term in eqn. (13) may then be either

$$S(\bar{Z}, \bar{Z}^R) = D_q(\bar{Z}, \bar{Z}^R) \quad \text{or} \quad S(\bar{Z}, \bar{Z}^R) = \bar{D}_q(\bar{Z}, \bar{Z}^R) \quad (20a,b)$$

In order to minimize the cost function given by eqn. (13), we write the critical point equation as

$$\frac{\partial T(\bar{Z})}{\partial \bar{Z}} = 0, \quad \text{i. e.} \quad \frac{\partial T(\bar{Z})}{\partial Z_j} = 0, \quad j = 1, 2, \dots, N_u \quad \text{resulting} \quad J^T \bar{R} + \lambda \bar{S}_{\bar{Z}} = 0 \quad (21a,b, 22)$$

where the elements of the Jacobian matrix J are given by

$$J_{s,t} = \frac{\partial C_s}{\partial Z_t}, \quad s = 1, 2, \dots, 2N_f \quad \text{and} \quad t = 1, 2, \dots, N_u \quad \text{and} \quad \bar{S}_{\bar{Z}} = \left\{ \frac{\partial S}{\partial Z_1}, \frac{\partial S}{\partial Z_2}, \dots, \frac{\partial S}{\partial Z_{N_u}} \right\}^T \quad (23, 24)$$

The elements of the Jacobian matrix J , given by eqn. (23), were calculated numerically by using a central finite-difference approximation. By using the Taylor expansions

$$\bar{R}(\bar{Z}^{n+1}) = \bar{R}(\bar{Z}^n) + J^n \Delta \bar{Z}^n, \quad \bar{S}_{\bar{Z}}(\bar{Z}^{n+1}) = \bar{S}_{\bar{Z}}(\bar{Z}^n) + J_s^n \Delta \bar{Z}^n \quad (25,26)$$

where n is used as the iteration index in the iterative procedure that will be constructed for the estimation of the vector of unknowns \bar{Z} ,

$$\bar{Z}^{n+1} = \bar{Z}^n + \Delta \bar{Z}^n \quad (27)$$

and the elements of the Jacobian matrix J_s are given by

$$J_{s,u,v} = \frac{\partial S_{Z_u}}{\partial Z_v} = \frac{\partial}{\partial Z_v} \left(\frac{\partial S}{\partial Z_u} \right), \quad u = 1, 2, \dots, N_u \quad \text{and} \quad v = 1, 2, \dots, N_u \quad (28)$$

Introducing eqns (25) and (26) into eqn. (22) results in the following:

$$\left(J^{nT} J^n + \lambda J_s^n \right) \Delta \bar{Z}^n = - \left(J^{nT} \bar{R}^n + \lambda \bar{S}_{\bar{Z}}^n \right) \quad (29)$$

We are now in a position to construct an iterative procedure for the estimation of the vector of unknowns \bar{Z} . We first choose an initial guess \bar{Z}^0 , which may be for example,

$$\bar{Z}^0 = \bar{Z}^R \quad (30)$$

and then we calculate the Jacobian matrices J^0 and J_s^0 , whose elements are given by eqns (23) and (28), respectively, as well as the elements of the vector of residues given by eqn. (14). Next, the vector of corrections $\Delta \bar{Z}^0$ is calculated by solving the system of algebraic linear eqns (29). The vector, with new estimates for the unknowns, \bar{Z}^1 , is obtained using eqn. (27). The iterative procedure for calculating the corrections $\Delta \bar{Z}^n$ with eqn. (29) and new estimates for the unknowns \bar{Z}^{n+1} with eqn. (27) is continued until a convergence criterion such as

$$\left| \frac{\Delta Z_i}{Z_i} \right| < \varepsilon \quad \text{for} \quad i = 1, 2, \dots, N_u \quad (31)$$

is satisfied, where ε is a given tolerance.

Before we proceed, we must show how the elements of the vector $\bar{S}_{\bar{Z}}$, and the elements of the Jacobian matrix J_s , are calculated.

From eqns (20a,b) and (24) we observe that

$$\frac{\partial S}{\partial Z_j} = \frac{\partial D_q}{\partial Z_j} \quad \text{or} \quad \frac{\partial S}{\partial Z_j} = \frac{\partial \bar{D}_q}{\partial Z_j}, \quad j = 1, 2, \dots, N_u \quad (32a,b)$$

and from eqns (28) and (32a,b) we obtain

$$J_{S_{u,v}} = \begin{cases} \frac{\partial^2 D_q}{\partial Z_u^2} & \text{or} \quad \frac{\partial^2 \bar{D}_q}{\partial Z_u^2} & \text{if } u = v \\ 0 & & \text{if } u \neq v \end{cases} \quad u = 1, 2, \dots, N_u \quad \text{and} \quad v = 1, 2, \dots, N_u \quad (33a,b)$$

The first and second derivatives of the Bregman distance in eqns (32a,b) and (33a,b) are obtained from eqns (17a,b) or (18a,b). These derivatives, as well as the Bregman distances, are shown in Table 1 for both situations: (a) regular Bregman distances, and (b) weighted Bregman distances.

Table 1: Bregman distances used as regularization terms, and the first and second derivatives for both cases: (a) regular Bregman distances, and (b) weighted Bregman distances.

(a) regular	(b) weighted
$D_q = \frac{1}{1+q} \sum_{i=1}^{N_u} \left\{ Z_i \frac{Z_i^q - Z_i^{R^q}}{q} - Z_i^{R^q} (Z_i - Z_i^R) \right\}$ $D_0 = \sum_{i=1}^{N_u} \left\{ Z_i \ln \left(\frac{Z_i}{Z_i^R} \right) - (Z_i - Z_i^R) \right\}$	$\bar{D}_q = \frac{1}{1+q} \sum_{i=1}^{N_u} \frac{1}{f_{Z_i}} \left\{ Z_i \frac{Z_i^q - Z_i^{R^q}}{q} - Z_i^{R^q} (Z_i - Z_i^R) \right\}$ $\bar{D}_0 = \sum_{i=1}^{N_u} \frac{1}{f_{Z_i}} \left\{ Z_i \ln \left(\frac{Z_i}{Z_i^R} \right) - (Z_i - Z_i^R) \right\}$
$\frac{\partial D_q}{\partial Z_j} = \frac{Z_j^q - Z_j^{R^q}}{q}, \quad j = 1, 2, \dots, N_u$ $\frac{\partial D_0}{\partial Z_j} = \ln \left(\frac{Z_j}{Z_j^R} \right), \quad j = 1, 2, \dots, N_u$	$\frac{\partial \bar{D}_q}{\partial Z_j} = \frac{1}{f_{Z_j}} \frac{Z_j^q - Z_j^{R^q}}{q}, \quad j = 1, 2, \dots, N_u$ $\frac{\partial \bar{D}_0}{\partial Z_j} = \frac{1}{f_{Z_j}} \ln \left(\frac{Z_j}{Z_j^R} \right), \quad j = 1, 2, \dots, N_u$
$\frac{\partial^2 D_q}{\partial Z_j^2} = Z_j^{q-1}, \quad j = 1, 2, \dots, N_u$ $\frac{\partial^2 D_0}{\partial Z_j^2} = \frac{1}{Z_j}, \quad j = 1, 2, \dots, N_u$	$\frac{\partial^2 \bar{D}_q}{\partial Z_j^2} = \frac{1}{f_{Z_j}} Z_j^{q-1}, \quad j = 1, 2, \dots, N_u$ $\frac{\partial^2 \bar{D}_0}{\partial Z_j^2} = \frac{1}{f_{Z_j}} \frac{1}{Z_j}, \quad j = 1, 2, \dots, N_u$

4. RESULTS AND DISCUSSION

As a continuation of our previous works [4,28] we are here interested in the numerical evaluation of the PAS before running the real experiments. Therefore, we use in the solution of the inverse problem synthetic experimental data generated with

$$E_i = C_i \left(\bar{Z}_{exact} \right) + 2.576 r_i \sigma, \quad i = 1, 2, \dots, 2N_f \quad (34)$$

where r_i is a random number in the range [-1, 1], and σ represents the standard deviation of the measurement errors. The level of noise in the experimental data that will be reported next for the numerical test cases is computed by

$$noise_i(\%) = \left| \frac{2.576 r_i \sigma}{C_i} \right| \times 100\%, \quad i = 1, 2, \dots, 2N_f \quad (35)$$

and we take the maximum value of the $noise_i(\%)$, $i = 1, 2, \dots, 2N_f$.

In order to compare the performance of the approach developed in the present work with that presented in [4, 28], we used the same geometry, process, thermal and optical parameters for the photoacoustic cell and for the sample of the material under analysis. In the test cases performed we have used frequencies of the modulated

incident light in the range [1,8] Hz which may yield higher sensitivities to the parameters we want to determine [4]. In Table 2 is presented a summary of the process, thermal and optical parameters used.

Table 2: Process, thermal and optical parameters for the photoacoustic cell.

backing material	sample	gas	light
Aluminum, $\alpha_b = 0.82 \times 10^{-4} \text{ m}^2 / \text{s}$, $k_b = 201 \text{ W} / \text{mK}$, $l_b = 5 \times 10^{-3} \text{ m}$	Opaque glass, $\alpha_s = 5.286 \times 10^{-7} \text{ m}^2 / \text{s}$, $\beta = 10^3 \text{ m}^{-1}$, $k_s = 1.047 \text{ W} / \text{mK}$, $l_s = 5 \times 10^{-3} \text{ m}$	Air, $\alpha_g = 0.19 \times 10^{-4} \text{ m}^2 / \text{s}$, $k_g = 0.0239 \text{ W} / \text{mK}$, $l_g = 1.2 \times 10^{-3} \text{ m}$	Laser HeNe or other monochromatic light, $I_0 = 100 \text{ W} / \text{m}^2$ modulation frequencies used $f = 1, 2, \dots, 8 \text{ Hz}$

In Figure 2 are presented the results for the simultaneous estimation of the thermal diffusivity, α_s , and optical absorption coefficient, β , of opaque glass, using three approaches: (i) without regularization, (ii) with regularization using the regular Bregman distances, and (iii) with regularization using weighted Bregman distances. The first approach corresponds to $\lambda = 0$ in eqn. (13), and for the second and third approaches $\lambda = 0.03$ and $q = 1$ with the regularization terms given, respectively, by eqns (20a) and (20b). The reference values and the initial estimates for the thermal diffusivity and optical absorption coefficients were taken as: $\alpha_s^R = 4.0 \times 10^{-7} \text{ m}^2 / \text{s}$, $\beta^R = 9.5 \times 10^2 \text{ m}^{-1}$, $\alpha_s^0 = 5.0 \times 10^{-6} \text{ m}^2 / \text{s}$, and $\beta^0 = 0.9 \times 10^3 \text{ m}^{-1}$. The exact values which we want to recover are shown in Table 2.

Five computations were performed for each approach, using for each computation a different set of pseudo-random numbers, simulating therefore five different experiments. The value of $\sigma = 0.5$ in eqn. (34) led to synthetic experimental data with noise up to 4%.

In order to achieve convergence the full Newton correction step in eqn. (27) was not used. Instead we have applied a gain factor γ , with $0 \leq \gamma \leq 1$, such that

$$\bar{Z}^{n+1} = \bar{Z}^n + \gamma \Delta \bar{Z}^n \quad (36)$$

To obtain the results presented in Figure 2 we used $\gamma = 0.5$.

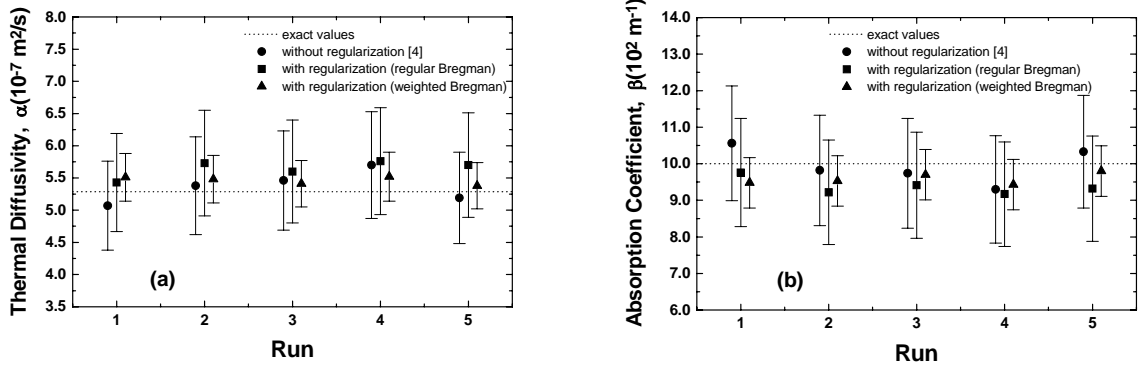


Figure 2. Estimates and confidence bounds for (a) thermal diffusivity, and (b) optical absorption coefficient using three approaches: without regularization; with regularization using regular Bregman distances; and with regularization using weighted Bregman distances.

From Figure 2 we may conclude that the use of the regularization terms with weighted Bregman distances yielded better results, i.e. smaller confidence bounds. Nonetheless we must be careful because the confidence bounds were calculated using the inverse of the matrix $J^T J + \lambda J_s$, and therefore higher values of λ could yield smaller confidence bounds. This subject deserves further investigation.

In Figure 3 are shown the values of the confidence bounds as a percentage of the estimated values of the unknowns, and the values of the percentage difference between the estimated and exact values of the unknowns. Five different runs were performed for each of the two following approaches: (i) without regularization, i.e. $\lambda = 0$ in eqn. (13), and (ii) with regularization using the weighted Bregman distances. For the second approach it was considered $\lambda = 0.03$ and $q = 1$. Here we have used the following reference values and initial estimates for the unknowns: $\alpha_s^0 = \alpha_s^R = 4.0 \times 10^{-7} \text{ m}^2 / \text{s}$ and $\beta^0 = \beta^R = 9.5 \times 10^2 \text{ m}^{-1}$. The exact values which we

want to recover are the same used for the test case whose results are presented in Figure 2, i.e. $\alpha_s^{exact} = 5.286 \times 10^{-7} m^2 / s$ and $\beta^{exact} = 10^3 m^{-1}$. It should be noted that in order to obtain the results shown in Figure 3 the reduction on the Newton correction step was not necessary, and we have therefore used $\gamma = 1$ in eqn. (36), going back to eqn. (27). It was also observed that the use of two different initial guesses led to very similar estimates for the unknowns, i.e. within the same order of confidence bounds and deviation of the exact values.

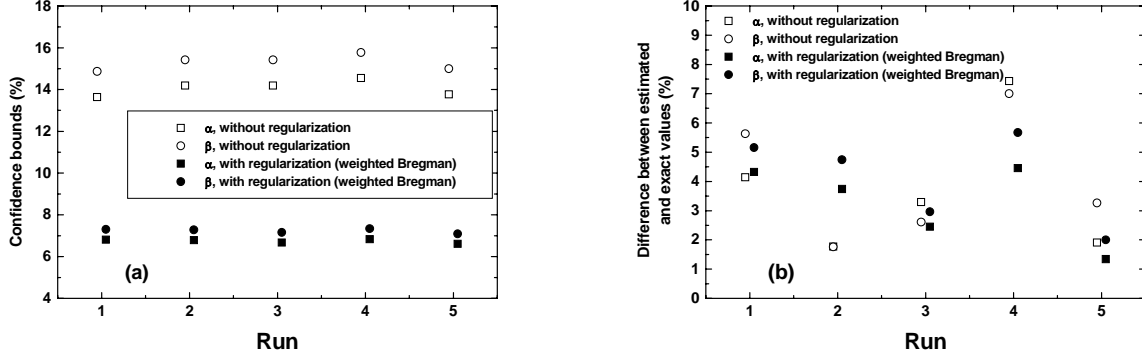


Figure 3. (a) Confidence bounds as a percentage of the estimated values for the unknowns, and (b) the percentage difference between the estimated and exact values of the unknowns.

From the results shown in Figure 3 we observe a reduction in the values of the confidence bounds when the regularization with the weighted Bregman distances is used, but as mentioned before this reduction may have been caused by the terms added to the diagonal of the information matrix. The difference between the estimated and exact values do not present a clear trend to be smaller when the regularization is used, but as shown next in Figure 4 the dispersion of this difference changes with the variation of the parameter q as well as with the variation of the regularization parameter λ .

In Figure 4 are presented the variation of the average for the confidence bounds as a percentage of the estimated values of the unknowns, and the variation of the average values of the differences between the estimated and exact values, with the regularization parameter λ and with the parameter q .

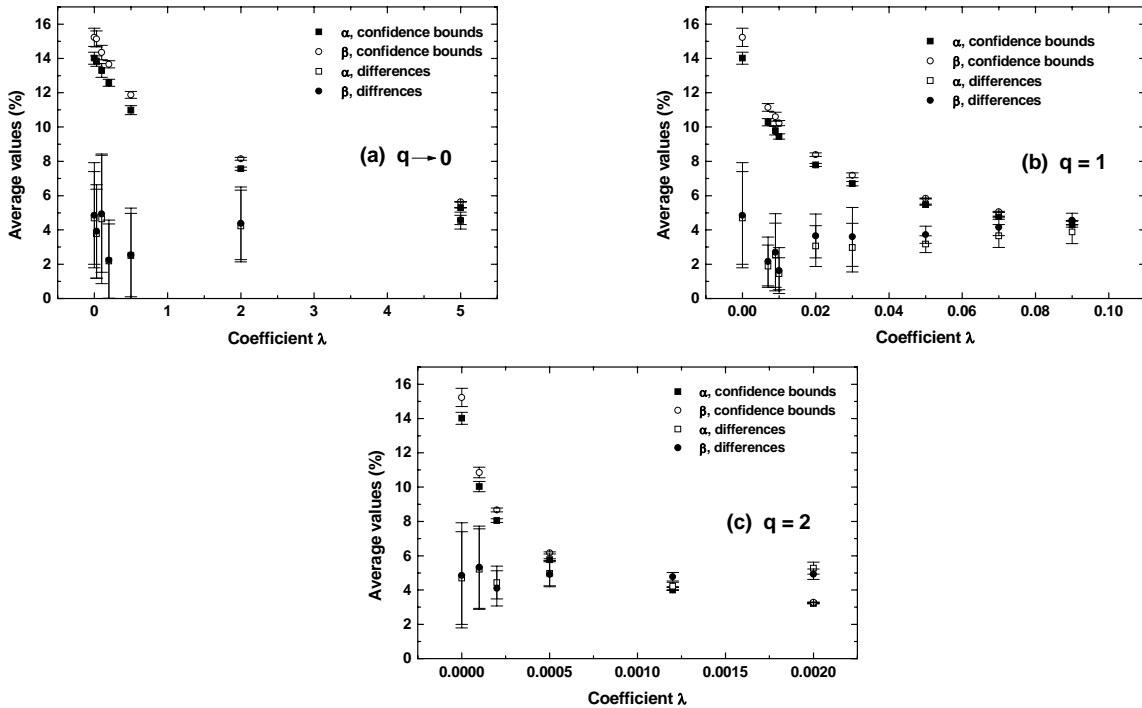


Figure 4. Average values, in 10 runs, for the confidence bounds and difference between estimated and exact parameters as a function of the regularization parameter λ and for (a) $q \rightarrow 0$, (b) $q = 1$, and (c) $q = 2$.

It must be stressed that in Figure 4 the error bars correspond to one standard deviation from the average values of both the percentage differences and confidence bounds (as a percentage of the estimated values) obtained in 10 runs for each value of q and for each value of λ . The standard deviation was calculated from the distribution of the results obtained in the 10 runs. Further, from the observation of the error bars shown in Figure 4 we conclude that the deviation of the estimates for α and β become smaller as higher values of λ are considered for a given value of q .

The average values presented in Figure 4 were obtained from 10 runs of the algorithm using experimental data with noise up to 4% and the regularization with the weighted Bregman distances.

From the results presented, we conclude that for different values of the pair (λ, q) the error in the estimates of the unknowns are of the order of, or smaller than, the level of the noise present in the experimental data, which represents an improvement on the results presented in [4].

5. CONCLUSIONS

In this work we have used an inverse problem approach for the Photoacoustic Spectroscopy (PAS) in which we were interested in estimating simultaneously the thermal diffusivity and optical absorption coefficient of a sample.

The use of weighted Bregman distances constructed with the q -discrepancy functional as regularization terms in Tikhonov's functional appears to yield better estimates for the unknowns. We have varied both the regularization parameter λ and the parameter q in the weighted Bregman distance in an attempt to find optimal values for such parameters. We have observed that by increasing λ we obtain a smaller dispersion of the estimates for the unknowns.

In real applications, the exact values for the parameters may be far from the available reference values, and therefore we have also implemented a feed-back approach in which the estimates at the end of one cycle of iterations are used as the new values for the reference parameters in the subsequent cycle of the iterations. This procedure will be the subject of further evaluation in a future work. Further, in the future we will also perform some real experiments in an experimental apparatus that is available at our institution.

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