Solution of the inverse problem of estimating particle size distributions

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In this work, we describe two alternative methods for solving the ill-conditioned inverse problem that allows estimating the particle size distribution (PSD) from turbidimetry measurements. The first method uses the inverse Penrose matrix to solve the inverse problem in its discrete form. The second method consists of replacing an ill-posed problem with a collection of well-posed problems, penalizing the norm of the solution, and it is known as the Tikhonov regularization. Both methods are used to solve a synthetic application of the inverse problem by solving the direct problem using a theoretical expression of the distribution of particles sizes function $f(D)$ and considering soft industrial latex particles (NBR), with average particle diameters of: 80.4, 82.8, 83.6, and 84.5 nm; and three illumination wavelengths in the UV-Vis region: 300, 450, and 600 nm The estimated solution obtained by the inverse Penrose matrix is different from the original solution due to the inverse problem is ill-conditioned. In contrast, when using Tikhonov's regularization, the estimate obtained is close to the original solution, which proves that the particle size distribution is adequate.

1. Introduction

It is well-known that a particle's scattering properties depend on its size, shape, the real and the imaginary part of its refractive index, and the particle size distribution (PSD) [1]. The PSD is an important physical feature in particulate colloidal systems such as aerosols, emulsions^i, suspensions^{ii}, dispersions^{iii}, powders, etc. [2]. PSD influences the rheological performance and chemical stability of emulsions and dispersions, the rate of reaction and diffusion, and magnetic and optical properties [2]. In industrial applications, PSD can affect the taste and texture of certain foods, the properties of automotive paints, inks, and toners, ceramic manufacturing processes, and the consumption rate of fuels and explosives [2]. Usually, turbidimetry techniques are employed to obtain the PSD by using a spectrophotometer that projects a beam of monochromatic light through a sample and measures the amount of light absorbed. According to Hadamard, estimating the PSD from turbidimetry measurements involves the resolution of an ill-conditioned inverse problem [1]. From this, it is possible to calculate the turbidity spectrum, which is related to the PSD by Mie's theory. Different methods based on regularization techniques have been proposed to solve this problem from the knowledge of turbidity [3]. In 2016, González-Garcés analyzed the particle size distribution

of blends of spices and scents to enhance and optimize it by using a laser diffraction technique, focused on the food industry [4]. In 2017, Galpin *et al.* recovered refractive index values of spherical polystyrene particles using illumination wavelengths corresponding to 220 and 420 nm by employing a differential optical absorption spectrophotometer and Mie's theory [5]. Also, previous reports found in the literature have described the solving of the inverse problem of the estimation of particle size distributions by using the least squares approximation and neural networks. In particular, this work studies the inverse problem of estimating particle size distributions theoretically and numerically from turbidimetry measurements, and its novelty and originality lies in the fact that the inverse Penrose matrix and the Tikhonov regularization are used to solve it. In addition, we present the deduction of the mathematical model that describes the inverse problem, which may be written in matrix form. In this way, the inverse problem for estimating particle size distributions is given by the system of linear equations $\tau = Af$; where τ is the turbidimetry measurements, A is the matrix obtained from Mie's theory, and f is the distribution of size of particles vector to be estimated. The inverse problem consists in finding f from knowing τ and the ill-conditioned matrix A.

2. Approach to the inverse problem of estimating particle size distributions (PSD)

This work uses turbidimetry to estimate the particle size distribution from Mie's theory. Turbidimetry is an optical measurement technique based on the scattering of light that occurs when solid particles appear in a homogeneous solution [7]. The loss of homogeneity means that the light that passes through the solution is not the same intensity as before turbidity appeared [6]. Turbidity is defined as the decrease in a liquid's transparency caused by undissolved particles [7]. A turbidimetry experimental test is carried out using a spectrophotometer and consists of measuring at different wavelengths λ_i with $i = \overline{1, m}$; the attenuation of light when passing through a set of spherical particles immersed in a non-absorbent medium [1, 2]. The principle of operation comprises a light beam that passes through a monochromator which in turn divides the incident light beam into different wavelengths, producing a band of colors called a spectrum due to light scattering; then, a certain wavelength is selected, which travels an optical path length $\mathscr{L}$ and impinge the sample to be analyzed (See Fig. 1. Finally, the photometer or detector measures the intensity $I(\lambda_i)$ of the transmitted light produced by the spectrum when passing through the sample with turbidity [1].

In general, the turbidity spectrum measurement using turbidimetry is based on the relationship between the incident light intensity and the light intensity scattered by the particles in the medium through Lambert-Beer's law, in which turbidity is proportional to particle concentration [7]. According to Mie's theory, for a monodisperse[iv] suspension[v] of non-absorbing spherical isotropic[vi] particles, in the absence of multiple scattering, it is possible to define the relationship between the turbidity spectrum $\tau(\lambda_i)$ and the particle size distribution $f(D)$ through the following equation [8, 9]:

$$\tau(\lambda_i) = \frac{\log_{10}(e)\pi\mathscr{L}}{4} \int_0^\infty f(D)Q_{ext}(x_i, m_i)D^2 \, dD, \quad (1)$$

where $f(D)$ is the continuous normalized particle size distribution (number of particles per cm^3 vs particle diameter D); Q_{ext} is the efficiency of the extinction of light passing through the sample; m_i is the relative refractive index; and x_i is the particle size parameter [8, 9]. In addition, the attenuation of an electromagnetic wave in a turbidimetry experiment is based on a physical phenomenon called light extinction due
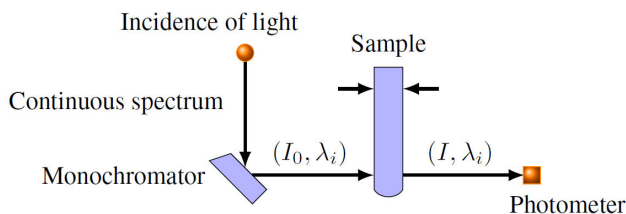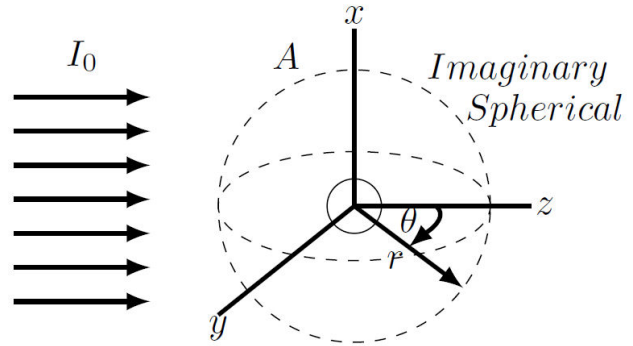

FIGURE 2. Extinction of light by an arbitrary particle in a non-absorbent medium.

to light absorption and scattering as it passes through a particulate medium [10]. Considering the extinction of light by a single arbitrary particle illuminated by a plane wave (See Fig. 2, it is possible to consider an imaginary spherical surface of radius r surrounding the particle in such a way that if the particle is in a non-absorbing medium, the rate of energy inside the particle can be neglected. Thus we will have the net ratio at which electromagnetic energy is extinguished when passing through surface A, denoted by W_{ext} is the sum of the energy absorption rate W_a and the energy dispersion rate W_s [10].

It is known that the incident irradiance I_0 passes through the cross-section of the imaginary sphere that contains the particle. Thus, the relationship between W_{ext} and I_0 is a quantity with area dimensions. This quantity is called the cross-section of light extinction and to denote by C_{ext} [10].

On the other hand, based on Mie's theory, it is possible to theoretically model the physical phenomena that occur in a turbidimetry experiment based on the Q_{ext} coefficients [2]. The theory is only valid under certain conditions of simple scattering, that is, under the hypothesis that the light scattered by a particle does not interact with any other particle in the system [2]. According to Bohren and Huffman, the extinction light efficiency factor is defined as $Q_{ext} = C_{ext}/G$, where G is the cross-section area of the projected particle in a plane perpendicular to the direction of the incident beam. If the particles are spherical, then the cross-section of the particle is a circle, so it is possible to rewrite the factor Q_{ext} as:

$$Q_{ext}(x_i, m_i) = \frac{2}{x_i^2} \sum_{n=1}^\infty (2n+1)$$
$$\times Re[a_n(x_i, m_i) + b_n(x_i, m_i)], \quad (2)$$

where a_n and b_n are Mie's coefficients. Even more, if they meet the Rayleigh conditions; that is, if $x_i \ll 1$ and $|m_i x_i| \ll 1$, then the dispersion coefficients a_n and b_n of the series expansion of Eq. (2) containing functions $(x_i)^t$ with $t > 7$, may be neglected. Furthermore, the light extinction efficiency function reduces to the following expresion [2, 8, 9]:

$$Q_{ext}(x_i, m_i) = \frac{8}{3}x_i^4 \left(\frac{m_i^2 - 1}{m_i^2 + 2}\right)^2. \quad (3)$$

FIGURE 1. Diagram of a spectrophotometer for turbidity measurement.

Incidence of light
Sample
Continuous spectrum
Monochromator
(I_0, λ_i)
(I, λ_i)
Photometer

However, since there is a finite number of measurements, the problem is posed on its discrete form in such a way that if $f(D_j)$ is considered as a discrete function equivalent to the continuous function $f(D)$, that depends on different mean diameters D_j $(j = \overline{1, n})$ taken at regular intervals ΔD, and considering a finite set of wavelengths λ_i $(i = \overline{1, m})$, then it is possible to rewrite the Eq. (1) as:

$$\tau(\lambda_i) = \frac{\log_{10}(e)\pi\mathscr{L}}{4} \sum_{j=1}^{n} f(D_j)Q_{\text{ext}}(x_{ij}, m_i)D_j^2, \quad (4)$$

where,

$$m_i = \frac{n_p(\lambda_i)}{n_m(\lambda_i)}; \quad (5)$$

and where x_{ij} is the size parameter defined as:

$$x_{ij} = \pi D_j \left(\frac{n_m(\lambda_i)}{\lambda_i} \right), \quad (6)$$

in this case, $n_m(\lambda_i)$ is the refractive index function of the medium and $n_p(\lambda_i)$ is the refractive index function of the particle. Equation (4) may be written in its matrix form:

$$\tau = Af; \quad (7)$$

where $A \in M_{m \times n}(\mathbb{K})$, $\tau \in \mathbb{K}^m$, $f \in \mathbb{K}^n$ ($\mathbb{K} = \mathbb{R}$ o $\mathbb{C}$) and the coefficients of matrix A are given by [8]:

$$a_{ij} = \frac{\log_{10}(e)\pi\mathscr{L}}{4}Q_{\text{ext}}(x_{ij}, m_i)D_j^2. \quad (8)$$

Thus, the direct problem of estimating particle size distributions consists in finding the value of the turbidity spectrum τ, based on knowing the particle size distribution vector f and the matrix A. Consequently, the inverse problem consists in finding the vector f from knowing the turbidity spectrum τ and the matrix A. For the case in which $m > n$ and $rank(A) = \min(m, n)$, the solution of the inverse problem is given by $f = [A^t A]^{-1} A^t \tau$. However, the inverse PSD estimation problem is ill-posed in the Hadamard sense; therefore, small deviations in the measurements of τ result in large deviations in the solution f. Hence, methods of solving the inverse problem are needed, which allow finding a better approximation to the original solution. The following section presents two solutions that solve the problem and demonstrate what is stated above.

3. Solution methods of the inverse problem

Once the matrix form (7) of the inverse problem has been established, two solution methods are presented: the inverse Penrose matrix and the Tikhonov regularization. Two definitions equivalent to the definition of the Penrose inverse matrix are the Moore inverse matrix and the pseudoinverse matrix. We have implemented the Penrose definition for solving a synthetic application for this work. If the reader requires more information about the Moore inverse, he can refer to [11].

3.1. Inverse Penrose matrix

For the case where $m < n$ and matrix $A \in M_{m \times n}(\mathbb{C})$ is row-full-rank, there always exists a unique matrix $Y \in M_{n \times m}(\mathbb{C})$ such that $A^* = (AYA)^* = (YA)^*A^*$ and $(YA)^* = YA$. Then,

$$A^* = YAA^*. \quad (9)$$

Also, since $rank(AA^*) = rank(A)$; if $rank(A) = m$, then $N(AA^*) = 0$; therefore, AA^* is invertible, then by multiplying the inverse on both sides of Eq. (9), the Penrose inverse of A is:

$$Y = A^* [AA^*]^{-1}. \quad (10)$$

From Eq. (10), an approximation of the solution to the problem given by (7) is obtained. However, when inverse problems are ill-conditioned, they cannot be solved using only the Penrose inverse method, so it is necessary to apply a regularization method. In this work, Tikhonov regularization is implemented.

3.2. Tikhonov regularization

It is well-known that a regularization method consists of replacing an ill-posed problem with a collection of well-posed problems so that the solution is a good approximation to the original solution [3]. The collection will depend on the regularization parameter; for each value of the parameter, there exists a different problem, and its solution is called a regularized solution.

Formally, from the point of view of free linear operators, given $K : X \to Y$ linear and bounded, in search for a x_α which satisfies $\|Kx_\alpha - y\| \le \|Kx - y\|$ for all $x \in X$ and $y \in Y$, such that it is a solution of the normal equation $K^*Kx_\alpha = K^*y$, where K^* is the adjoint operator of K. Tikhonov's regularization method consists of penalizing the norm $\|x\|$ of the solution in the least squares problem to minimize the effect of the changes [3,12]. That is, we want to obtain the regularized solution x_α that minimizes the Tikhonov functional defined by:

$$J_\alpha(x) := \|Kx - y\|^2 + \alpha^2 \|x\|^2,$$
$$\forall x \in X, \quad \alpha > 0, \quad (11)$$

where α is the regularization parameter, and it must be satisfied that $\|x_\alpha\|$ tends to zero as α tends to infinity and $\|x_\alpha\|$ tends to infinity as α tends to zero.

In Ref. [3], the existence of a minimum for the Tikhonov functional is proven, although it is obvious to the reader that several steps have been neglected; in this work, we show the complete sequence of the missing steps in the Appendix A.

The regularized solution (A.22) minimizes the Tikhonov functional, so it is possible to apply said regularized solution to solve the ill-conditioned discrete inverse problem of PSD estimation.

4. Solution of the problem and numerical experiments

Due to the lack of data, a synthetic example is built, in which the direct problem is solved, so then, to solve the inverse problem of estimating the particle sizes distributions by turbidimetry in the UV-Vis spectrum near the Rayleigh region based on the theory according to Ref. [8], for soft industrial latexes (NBR) with particle sizes in the range of 80 nm to 85 nm . The inverse problem is solved using the Penrose inverse and the Tikhonov regularization method.

We want to solve the discrete inverse problem given by Eq. (4) where x_{ij}, m_i and Q_{ext} are given by Eqs. (3), (5), (6) with $\mathscr{L} = 1$ cm. It is assumed that the refractive index functions of the medium $n_m(\lambda_i)$ and the particles $n_p(\lambda_i)$ are defined as [8]:

$$n_m(\lambda_i) = 1.324 + \frac{3046}{\lambda_i^2},$$

$$n_p(\lambda_i) = 1.494 + \frac{6284}{\lambda_i^2}.$$

4.1. Solution of the direct problem

First, we consider four different mean diameters of soft industrial latex (NBR) particles immersed in water, given by: $D_1 = 80.4$ nm, $D_2 = 82.8$ nm, $D_3 = 83.6$ nm, and $D_4 = 84.5$ nm; and three different illumination wavelengths: $\lambda_1 = 300$ nm, $\lambda_2 = 450$ nm, and $\lambda_3 = 600$ nm. In this way, and using matrix notation, Eq. (4) is as follows:

$$\tau = Af, \tag{12}$$

where $\tau \in \mathbb{R}^3$, $f \in \mathbb{R}^4$, and $A \in M_{3\times4}(\mathbb{R})$, with coefficients given by the Eq. (8) for $i = \overline{1,3}$ and $j = \overline{1,4}$. To solve the direct problem, the matrix A and the function f are assumed to be known in order to calculate the values of λ. For this, an approximation of the continuous function $f(D)$ is considered, given by [8]:

$$f(D) = \frac{N_p}{D\sigma\sqrt{2\pi}} \exp\left\{ \frac{-[\ln(D) - \ln(D_g)]^2}{2\sigma^2} \right\}, \tag{13}$$

with $N_p = 1.2 \times 10^{12}$ part./cm^3; $D_g = 80$ nm; and $\sigma = 0.1$. The continuous distribution of Eq. (13) may be changed by the equivalent discrete function $f(D_j)$ given by Eq. (14), taking into account the diameters D_j and the wavelengths λ_i. The $f(D_j)$ values are then calculated using the following expression:

$$f(D_j) = \frac{1.2 \times 10^{12} \ \#\text{part./cm}^3}{D_j(0.1)\sqrt{2\pi}}$$

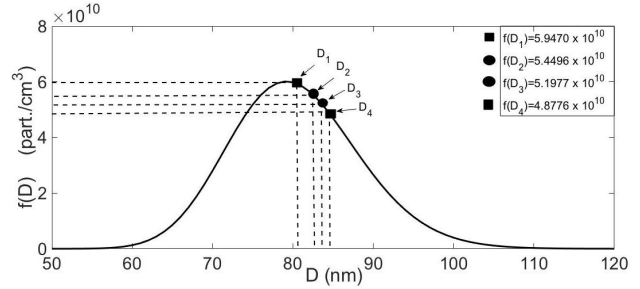$$\times \exp\left\{ \frac{-[\ln(D_j) - \ln(80)]^2}{0.02} \right\}. \tag{14}$$

FIGURE 3. Particle size distribution function $f(D)$.

For the particular case in which $j = \overline{1,4}$,

$$f(D) = \begin{bmatrix} 5.9470 \times 10^{10} \\ 5.4496 \times 10^{10} \\ 5.1977 \times 10^{10} \\ 4.8776 \times 10^{10} \end{bmatrix}. \tag{15}$$

Now, to find the coefficients of matrix A, we first calculate the refractive indices of the particles, $n_p(\lambda_i)$, the refractive indices of the medium $n_m(\lambda_i)$, and the relative refractive indices m_i, considering the given wavelengths λ_i (see Table I.

The particle size parameters x_i and the light extinction coefficients $Q_{ext}(x_i, m_i)$ is calculated from the above data y are shown in Table II.

TABLE I. Values of the refractive index of the medium $n_m(\lambda_i)$, values of the refractive index of the particle $n_p(\lambda_i)$ and relative refractive index values m_i.

λ_i	$n_m(\lambda_i)$	$n_p(\lambda_i)$	m_i
300 nm	1.3578	1.5638	1.1517
450 nm	1.3390	1.5250	1.1389
600 nm	1.3325	1.5115	1.1343

TABLE II. Particle size parameter values x_{ij} and the light extinction coefficients $Q_{ext}(x_{ij}, m_i)$.

x_{ij}	$Q_{ext}(x_{ij}, mi)$
$x_{11} = 1.1432$	$Q_{ext}(x_{11}, m_1) = 0.0439$
$x_{12} = 1.1774$	$Q_{ext}(x_{12}, m_1) = 0.0493$
$x_{13} = 1.1887$	$Q_{ext}(x_{13}, m_1) = 0.0513$
$x_{14} = 1.2015$	$Q_{ext}(x_{14}, m_1) = 0.0535$
$x_{21} = 0.7516$	$Q_{ext}(x_{21}, m_1) = 0.0069$
$x_{22} = 0.7740$	$Q_{ext}(x_{22}, m_1) = 0.0078$
$x_{23} = 0.7815$	$Q_{ext}(x_{23}, m_1) = 0.0081$
$x_{24} = 0.7899$	$Q_{ext}(x_{24}, m_1) = 0.0084$
$x_{31} = 0.5609$	$Q_{ext}(x_{31}, m_1) = 0.0020$
$x_{32} = 0.5777$	$Q_{ext}(x_{32}, m_1) = 0.0023$
$x_{33} = 0.5833$	$Q_{ext}(x_{33}, m_1) = 0.0023$
$x_{34} = 0.5895$	$Q_{ext}(x_{34}, m_1) = 0.0025$

In this way, using Eq. (8) and the values found previously, the a_{ij} coefficients of matrix A are found given by:

$$A = \begin{bmatrix} 96.7045 & 115.3699 & 122.2216 & 130.3319 \\ 15.2341 & 18.1745 & 19.2538 & 20.5315 \\ 4.4296 & 5.2846 & 5.5984 & 5.9699 \end{bmatrix}.$$

With the obtained results in this section, the direct problem of estimating the particle sizes distribution has been solved, and its solution is given by:

$$\tau = Af = \begin{bmatrix} 24.7479046889131 \times 10^{12} \\ 3.89858946984298 \times 10^{12} \\ 1.13358678358970 \times 10^{12} \end{bmatrix}.$$

The solution of the direct problem will be used as input data to solve the inverse problem.

4.2. Solution to the inverse problem

Let $\tau \in \mathbb{R}^3$ and the matrix $A \in M_{3 \times 4}(\mathbb{R})$ known data; also, a relative error ϵ is considered, such that $\tau = Af + \epsilon$. In this section, we implement the Penrose inverse and the Tikhonov regularization method to find the exact solution $f \in \mathbb{R}^4$.

4.2.1. Solution obtained by the Penrose inverse

Equation (10) is employed to find the Penrose inverse matrix of A. In this case, $m = 3$ and $n = 4$; furthermore, matrix A is of full range by rows, which means that $dim R_A = rank(A) = 3$. Since $A \in M_{3 \times 4}(\mathbb{R})$, then $A^* = A^t$. Therefore, doing the calculations for matrix A, the Penrose inverse matrix of A is given by:

$$Y = A^t \left[AA^t \right]^{-1} = \begin{bmatrix} -0.0925 & 0.6226 & -0.1210 \\ -0.0514 & -0.4166 & 2.5555 \\ 0.2348 & -1.4884 & -0.0064 \\ -0.1060 & 1.3026 & -2.1663 \end{bmatrix} \times 10^4.$$

The matrix Y satisfies the Penrose conditions. Then, the matrix Y is used to find an approximation to the solution of the inverse problem given by:

$$\hat{f} = Y\tau = \begin{bmatrix} 0.000182477455386 \\ 0.000084237889068 \\ 0.001238859978678 \\ 0.000527147719822 \end{bmatrix}.$$

Note that the estimated solution $\hat{f}$ is different from the original solution f. In practice, measurement errors are generally common, so it is necessary to determine their effect on the solution. For this, the vector $\tau(\lambda_i)$ changes by adding a term with a small value, *i.e.*, $\epsilon = 1 \times 10^{-4}$, which represents the error in the measurements, thus:

$$\hat{\tau}(\lambda_i) = \tau(\lambda_i) + \epsilon = \begin{bmatrix} 0.247579046889131 \\ 0.0390858946984298 \\ 0.0114358678358970 \end{bmatrix}.$$

Now, we calculate the estimation of the vector with parameters $\hat{\hat{f}}$ by considering the vector τ:

$$\hat{\hat{f}} = Y\hat{\tau} = \begin{bmatrix} 0.409293417919107 \\ 2.087583172718300 \\ -1.258845877485565 \\ -0.969210418586275 \end{bmatrix}.$$

The errors are $\left\| \hat{f} - \hat{\hat{f}} \right\| = 2.65579$ and $\| \tau - \hat{\tau} \| = 1 \times 10^{-4}$. Note that small variations in the vector of measurements τ cause large errors in the estimated solution $\hat{f}$ since the inverse problem is ill-conditioned, as explained next. We consider the matrix $A \in M_{3 \times 4}(\mathbb{R})$ as the matrix representation of the linear operator $K : \mathbb{R}^4 \to \mathbb{R}^3$ and the transposed matrix $A^t \in M_{4 \times 3}(\mathbb{R})$ as the matrix representation of the adjoint linear operator $K^* : \mathbb{R}^3 \to \mathbb{R}^4$, on the canonical bases of $\mathbb{R}^3$ and $\mathbb{R}^4$. To find the singular values σ_k ($k = \overline{1,4}$) of matrix A, singular value decomposition is applied, and we obtain:

$$\sigma = \begin{bmatrix} 236.760785418670 \\ 60.5937041072596 \times 10^{-6} \\ 27.6654149258401 \times 10^{-6} \\ 0 \end{bmatrix}. \quad (16)$$

Only the non-zero singular values are to be considered; therefore, we have that $\max_k |\sigma_k| = 236.76079$ and $\min_k |\sigma_k| = 27.66541 \times 10^{-6}$. Hence, the condition number of matrix A is:

$$c(A) = \frac{\max_k |\sigma_k|}{\min_k |\sigma_k|} = 8.558006 \times 10^6.$$

Since $c(A) \gg 1$, the inverse PSD problem is ill-conditioned. Consequently, calculating only the Penrose inverse is not enough; it is necessary to regularize the problem. For this, a new well-posed problem is solved such that its solution correctly approximates the original solution.

4.2.2. Solution obtained by the Tikhonov regularization

In order to apply the Tikhonov regularization method, we choose a collection of well-posed problems that depend on the regularization parameter α. For each value of the parameter α, different problems are obtained and, consequently, a different regularized solution. The solutions of well-posed problems are in terms of the regularization parameter, which may be chosen by applying Morozov's discrepancy principle or the L-curve method.

In this case, the data given by the turbidity spectrum vector τ and the matrix A are known, so the regularization problem consists in finding the regularized solution $f_\alpha \in \mathbb{R}^4$ that minimizes the Tikhonov functional given by:

$$J_\alpha(f) := \| Af - \tau \|^2 + \alpha^2 \| f \|^2,$$
$$\forall f \in \mathbb{R}^4, \tau \in \mathbb{R}^3 \ y \ \alpha > 0. \quad (17)$$

Appendix A demonstrates the existence of the minimum of the Tikhonov functional, which will be used to solve the synthetic application shown below. Let the exact solution $f(D) \in \mathbb{R}^4$ given by:

$$f(D) = \begin{bmatrix} 5.9470 \times 10^{10} \\ 5.4496 \times 10^{10} \\ 5.1977 \times 10^{10} \\ 4.8776 \times 10^{10} \end{bmatrix},$$

which we want to approximate by the regularized solution f_α; in addition, it is true that the smaller the size of the regularization parameter α, the regularized solution f_α is closer to the exact solution $f(D)$; that is, $\|f_\alpha - f(D)\| \to 0$ when $\alpha \to 0$. In general, it is known that the regularized solution f_α can be obtained using the singular value decomposition of the matrix A, given by $A = USV$, where:

$$U = \begin{bmatrix} -986.80841 & 160.03908 & 24.42638 \\ -155.45419 & -894.59020 & -418.97778 \\ -45.20122 & -417.24798 & 907.66788 \end{bmatrix} \times 10^{-3},$$

$$S = \begin{bmatrix} 236.76079 & 0 & 0 & 0 \\ 0 & 60.59370 \times 10^{-6} & 0 & 0 \\ 0 & 0 & 27.66541 \times 10^{-6} & 0 \end{bmatrix},$$

$$V = \begin{bmatrix} -0.413908 & -0.315888 & -0.103176 & -0.847496 \\ -0.493799 & -0.425291 & 0.689643 & 0.315727 \\ -0.523125 & 0.831208 & 0.172510 & -0.075330 \\ -0.557838 & -0.168630 & -0.695692 & 0.419991 \end{bmatrix}.$$

The regularized solution that minimizes the function (17) is given by Eq. (39). In this case, it is true that $r = rank(A) < n$, so the sum of the regularized solution goes from $i = \overline{1, r}$. Given that $rank(A) = 3$, then it is possible to write the regularized solution to the inverse problem of estimating the PSD using the SVD of matrix A as:

$$f_\alpha = \sum_{i=1}^{3} \left[\frac{\sigma_i}{\alpha^2 + \sigma_i^2} u_i^t \tau \right] v_i. \tag{18}$$

Since the matrix $A \in M_{3 \times 4}(\mathbb{R})$ is the matrix representation of the compact linear operator $K : \mathbb{R}^4 \to \mathbb{R}^3$, $A^t \in M_{4 \times 3}(\mathbb{R})$ is the matrix representation of the adjoint operator $K^* : \mathbb{R}^3 \to \mathbb{R}^4$, and the vectors $\sigma_1 \geq \sigma_2 \geq \sigma_3 > 0$, which are given by Eq. (16), are the non-zero singular values of the matrix A, then, there exist orthonormal sets $\{u_i\} \subset \mathbb{R}^3$ and $\{v_i\} \subset \mathbb{R}^4$ given by the columns of the matrix U and matrix V, respectively. Said orthonormal systems satisfy that $Av_i = \sigma_i u_i$ and $A^t u_i = \sigma_i v_i$ with $i \in I = \{1, 2, 3\} \subset \mathbb{N}$.

Similarly, it is known from the singular value decomposition of A that there are more vectors to the right v_i than vectors to the left u_i to know; vectors v_i on the right are used to find the regularized solution f_α, and they must satisfy that $u_i = Av_i/\sigma_i$, with $i = \overline{1, 3}$. In this case, the condition is true for columns v_1, v_2, and v_3, and for column v_4, $Av_4 \approx 0$. Therefore, the orthonormal system (v_i) is formed by the first three columns of the matrix V of the DVS of A. The regularization parameter is chosen adequately to calculate the

solution f_α. If we give values to α in such a way that each case it is smaller, the following is obtained:

- Para $\alpha = 1$:

$$f_\alpha = \begin{bmatrix} 4.38421797581138 \times 10^{10} \\ 5.23043693814276 \times 10^{10} \\ 5.54106671580672 \times 10^{10} \\ 5.90875761045696 \times 10^{10} \end{bmatrix}.$$

- Para $\alpha = 0.5$:

$$f_\alpha = \begin{bmatrix} 4.38427660522684 \times 10^{10} \\ 5.23050687725977 \times 10^{10} \\ 5.54114092962522 \times 10^{10} \\ 5.90883665273824 \times 10^{10} \end{bmatrix}.$$

- Para $\alpha = 0.3$:

$$f_\alpha = \begin{bmatrix} 4.38428905001994 \times 10^{10} \\ 5.23052170833471 \times 10^{10} \\ 5.54115692854422 \times 10^{10} \\ 5.90885348533342 \times 10^{10} \end{bmatrix}.$$

With this, we have found the approximate solution to the inverse problem of estimating the PSD using the optical technique of turbidimetry.

5. Conclusions

The function $f(D)$ and the matrix A of the system are assumed to be known to solve the direct problem related to the inverse problem of estimating the particle size distribution. Then, using the inverse Penrose matrix, the inverse problem is solved, and is obtained that the estimated solution is different from the exact solution $f(D)$. Therefore, the problem is regularized by implementing Tikhonov's regularization. Singular value decomposition is applied to matrix A, and values are given to the regularization parameter α. With this, an adequate approximation to the original solution $f(D)$ is obtained, which verifies that the particle size distribution is of the logarithmic-normal type. There are specific methods to obtain the regularization parameter α in such a way that it is optimal; the study of these methods is beyond the scope of this article, so it is left as a pending problem.

This work establishes a precedent for the use of the inverse Penrose matrix and the Tikhonov regularization to solve the inverse problem of estimating the particle size distribution since this problem has been studied in the literature using least squares and neural networks. The limitations that they found when solving the inverse problem are the lack of a data bank that adjusts to the studied model and the lack of a spectrophotometer to validate the results. For this reason, a synthetic application has been solved.

Appendix

A. Existence of a minimum for the Tikhonov functional

To prove the existence of the minimum of the Tikhonov functional, it needs to be proved that it is the limit of a Cauchy sequence. To do this, we consider a minimizing sequence $\{x_n\} \subset X$, which satisfies that:

$$\lim_{n\to\infty} J_\alpha(x_n) = I = \inf_{x\in X} J_\alpha(x). \tag{A.1}$$

Let $m, n \in \mathbb{N}$, then for x_n and x_m it is true that:

$$J_\alpha(x_n) + J_\alpha(x_m) = \|Kx_n - y\|^2 + \|Kx_m - y\|^2$$
$$+ \alpha^2 \|x_n\|^2 + \alpha^2 \|x_m\|^2. \tag{A.2}$$

Applying the binomial formula, we obtain:

$$\frac{\alpha^2}{2}\left(\|x_n - x_m\|^2 + \|x_n + x_m\|^2\right)$$
$$= \alpha^2 \|x_n\|^2 + \alpha^2 \|x_m\|^2. \tag{A.3}$$

Then, by substituting Eq. (A.3) into Eq. (A.2), we get:

$$J_\alpha(x_n) + J_\alpha(x_m) = \|Kx_n - y\|^2 + \|Kx_m - y\|^2$$
$$+ \frac{\alpha^2}{2}\|x_n - x_m\|^2 + 2\alpha^2 \left\|\frac{x_n + x_m}{2}\right\|^2. \tag{A.4}$$

On the other hand, we know that:

$$2\left\|K\left(\frac{x_n + x_m}{2}\right) - y\right\|^2$$
$$= 2\left\|K\left(\frac{x_n + x_m}{2}\right) - \frac{1}{2}y - \frac{1}{2}y\right\|^2$$
$$= \frac{1}{2}\|(Kx_n - y) + (Kx_m - y)\|^2. \tag{A.5}$$

Furthermore, applying the binomial formula again to the last term of (A.5), it is obtained:

$$\|(Kx_n - y) + (Kx_m - y)\|^2 + \|(Kx_n - y) - (Kx_m - y)\|^2$$
$$= 2\|Kx_n - y\|^2 + 2\|Kx_m - y\|^2. \tag{A.6}$$

Then, by substituting Eq. (A.5) into Eq. (A.6), we get:

$$2\left\|K\left(\frac{x_n + x_m}{2}\right) - y\right\|^2 + \frac{1}{2}\|Kx_n - Kx_m\|^2$$
$$= \|Kx_n - y\|^2 + \|Kx_m - y\|^2. \tag{A.7}$$

By substituting Eq. (A.7) into Eq. (A.4), we obtain:

$$J_\alpha(x_n) + J_\alpha(x_m) = 2\left\|K\left(\frac{x_n + x_m}{2}\right) - y\right\|^2$$
$$+ \frac{1}{2}\|K(x_n - x_m)\|_2^2 + \frac{\alpha^2}{2}\|x_n - x_m\|^2$$
$$+ 2\alpha^2 \left\|\frac{x_n + x_m}{2}\right\|^2. \tag{A.8}$$

Given that:

$$2J_\alpha\left(\frac{x_n + x_m}{2}\right) = 2\left\|K\left(\frac{x_n + x_m}{2}\right) - y\right\|^2$$
$$+ 2\alpha^2 \left\|\frac{x_n + x_m}{2}\right\|^2,$$

the Eq. (A.8) can be rewritten as:

$$J_\alpha(x_n) + J_\alpha(x_m) = 2J_\alpha\left(\frac{x_n + x_m}{2}\right)$$
$$+ \frac{1}{2}\|K(x_n - x_m)\|^2 + \frac{\alpha^2}{2}\|x_n - x_m\|^2.$$

Moreover, as

$$\lim_{n,m\to\infty} J_\alpha\left(\frac{x_n + x_m}{2}\right) = I,$$

then is must be satisfied that $J_\alpha(x_n) + J_\alpha(x_m) \geq 2I + (\alpha^2/2)\|x_n - x_m\|^2$. Also, $J_\alpha(x_n) + J_\alpha(x_m) \to 2I$, when $n, m \to \infty$. Therefore, we have that $0 \geq \|x_n - x_m\| \geq 0$. This means that the sequence of vectors $\{x_n\}$ is a Cauchy sequence. Therefore the sequence $\{x_n\}$ converges to some $x_\alpha \in X$. Since the Tikhonov functional is continuous, then $J_\alpha(x_n) \to J_\alpha(x_\alpha)$; so, for Eq. (A.1), it is obtained that $J_\alpha(x_\alpha) = I$, and this proves the existence of a minimum of J_α.

On the other hand, if x_α is considered to be the minimum of J_α, then it must be fulfilled that:

$$J_\alpha(x_\alpha) \leq J_\alpha(x), \quad \forall x \in X. \tag{A.9}$$

In addition, from Eq. (11), we have that:

$$J_\alpha(x) - J_\alpha(x_\alpha) = \left(\|Kx - y\|^2 - \|Kx_\alpha - y\|^2\right)$$
$$+ \alpha^2 \left(\|x\|^2 - \|x_\alpha\|^2\right).$$

Given that:

$$\|Kx - y\|^2 - \|Kx_\alpha - y\|^2$$
$$= 2Re(\langle K^*(Kx_\alpha - y), x - x_\alpha\rangle)$$
$$+ \|K(x - x_\alpha)\|^2;$$

then,

$$J_\alpha(x) - J_\alpha(x_\alpha) = 2Re(\langle K^*(Kx_\alpha - y), x - x_\alpha\rangle)$$
$$+ \|K(x - x_\alpha)\|^2 + \alpha^2 \left(\|x\|^2 - \|x_\alpha\|^2\right). \tag{A.10}$$

By the binomial formula, we have that:

$$\|x\|^2 = \|x_\alpha + x - x_\alpha\|^2 \tag{A.11}$$

$$= \|x_\alpha\|^2 + \|x - x_\alpha\|^2 + 2Re\left(\langle x_\alpha, x - x_\alpha \rangle\right). \tag{A.12}$$

By substituting Eq. (A.11) into Eq. (A.10) and factoring terms, we obtain:

$$J_\alpha(x) - J_\alpha(x_\alpha) = 2Re\left(\langle K^*(Kx_\alpha - y) + \alpha^2 x_\alpha, x - x_\alpha \rangle\right)$$
$$+ \|K(x - x_\alpha)\|^2 + \alpha^2 \|x - x_\alpha\|^2. \tag{A.13}$$

Then, by substituting Eq. (A.12) into inequality (A.9), we have:

$$0 \leq 2Re\left(\langle K^*(Kx_\alpha - y) + \alpha^2 x_\alpha, x - x_\alpha \rangle\right)$$
$$+ \|K(x - x_\alpha)\|^2 + \alpha^2 \|x - x_\alpha\|^2, \quad \forall x \in X. \tag{A.14}$$

In particular, if $x = x_\alpha + \alpha z$, for any $z \in X$ we have that:

$$0 \leq 2Re\left(\langle K^*(Kx_\alpha - y) + \alpha^2 x_\alpha, \alpha z \rangle\right) + \|K(\alpha z)\|^2$$
$$+ \alpha^2 \|\alpha z\|^2 = 2\alpha Re\left(\langle K^*(Kx_\alpha - y) + \alpha^2 x_\alpha, z \rangle\right)$$
$$+ \alpha^2 \|Kz\|^2 + \alpha^4 \|z\|^2.$$

Dividing by $\alpha > 0$ and taking the limit as $\alpha \to 0$, we conclude that $Re(\langle K^*(Kx_\alpha - y) + \alpha^2 x_\alpha, z \rangle) \geq 0$, for all $z \in X$, hence $K^*(Kx_\alpha - y) + \alpha^2 x_\alpha = 0$; that is, x_α is a solution to the normal equation:

$$\alpha^2 x_\alpha + K^*Kx_\alpha = K^*y. \tag{A.15}$$

Furthermore, the solution x_α is unique. That is, if x_l is a solution of the normal Eq. (A.15), then $\alpha^2 x_\alpha + K^*Kx_\alpha = \alpha^2 x_l + K^*Kx_l$ is obtained, which is only true if $x_\alpha = x_l$. Thus, the following result is proven.

Theorem 1. *Let $K : X \to Y$ be a linear and bounded operator on Hilbert spaces and $\alpha > 0$. Then the Tikhonov functional J_α has a unique minimum $x_\alpha \in X$. This minimum x_α is the only solution of the normal equation:*

$$\alpha^2 x_\alpha + K^*Kx_\alpha = K^*y.$$

In particular, if matrix A is the matrix representation of the linear and bounded operator $K : \mathbb{R}^n \to \mathbb{R}^m$ and matrix A^t is the matrix representation of the adjoint operator $K : \mathbb{R}^m \to \mathbb{R}^n$, on the canonical basis of $\mathbb{R}^n$ and $\mathbb{R}^m$, then the normal equation given in Eq. (A.15) may be written as:

$$\left(\alpha^2 I_n + A^t A\right) x_\alpha = A^t y, \quad \alpha > 0, \tag{A.16}$$

where $A \in M_{m \times n}(\mathbb{R})$ and I_n denotes the $n \times n$ identity matrix. If we consider the singular value decomposition of the matrix $A \in M_{m \times n}(\mathbb{R})$ given by $A = U\Sigma V^t$, we have:

$$A^t = V\Sigma^t U^t. \tag{A.17}$$

Then, $A^t A = (V\Sigma^t U^t)(U\Sigma V^t)$. Since the matrices U and V are orthogonal, that is, $U^t U = I_m$ and $VV^t = I_n$, then $A^t A = V\Sigma^t \Sigma V^t$. From here, we have:

$$\alpha^2 I_n + A^t A = V(\alpha^2 I_n + \Sigma^t \Sigma)V^t; \tag{A.18}$$

however, $\alpha^2 I_n + \Sigma^t \Sigma = diag(\alpha^2 + \sigma_1^2, \ldots, \alpha^2 + \sigma_n^2)$; then, Eq. (A.18) is equivalent to:

$$\left(\alpha^2 I_n + A^t A\right) V$$
$$= V diag\left(\alpha^2 + \sigma_1^2, \ldots, \alpha^2 + \sigma_n^2\right). \tag{A.19}$$

Since $\alpha > 0$, then the eigenvalues of $\alpha^2 I_n + A^t A$ are positive, that is, the matrix $\alpha^2 I_n + A^t A$ is positive definite. Since every positive defined matrix is invertible, then $\alpha^2 I_n + A^t A$ is invertible, and from Eq. (A.19), its inverse is given by:

$$\left(\alpha^2 I_n + A^t A\right)^{-1}$$
$$= V diag\left(\frac{1}{\alpha^2 + \sigma_1^2}, \ldots, \frac{1}{\alpha^2 + \sigma_n^2}\right) V^t. \tag{A.20}$$

Then, from Eq. (A.16), we obtain:

$$x_\alpha = \left(\alpha^2 I_n + A^t A\right)^{-1} A^t y; \tag{A.21}$$

and by substituting Eqs. (A.20) and (A.17) into Eq. (A.21), we have that the regularized solution x_α is given by:

$$x_\alpha = V \, diag\left(\frac{1}{\alpha^2 + \sigma_1^2}, \ldots, \frac{1}{\alpha^2 + \sigma_n^2}\right) V^t V\Sigma^t U^t y.$$

Since the matrix V is orthogonal and

$$\Sigma^t U^t y = \begin{bmatrix} \sigma_1 u_1^t y \\ \vdots \\ \sigma_n u_n^t y \end{bmatrix},$$

then it must fulfill that:

$$x_\alpha = \begin{pmatrix} v_1 & \ldots & v_n \end{pmatrix}$$
$$\times diag\left(\frac{1}{\alpha^2 + \sigma_1^2}, \ldots, \frac{1}{\alpha^2 + \sigma_n^2}\right) \begin{bmatrix} \sigma_1 u_1^t y \\ \vdots \\ \sigma_n u_n^t y \end{bmatrix}.$$

Therefore, the regularized solution using decomposition in singular values is:

$$x_\alpha = \sum_{i=1}^{n} \left[\frac{\sigma_i}{\alpha^2 + \sigma_i^2} u_i^t y\right] v_i. \tag{A.22}$$

i. Type of heterogeneous mixture made up of small particles of a solid, dispersed in a liquid medium in which they cannot dissolve.

ii. Heterogeneous system consisting of two immiscible liquids.

iii. System in which distributed particles of one material is dispersed in a continuous phase of another material.

iv. Heterogeneous mixture formed by a solid powder or by small insoluble particles that are dispersed in a liquid medium.

v. Characterized by particles of uniform size in a dispersed phase.

vi. Isotropic materials have identical material properties in all directions at every given point.

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