



SHEAR MODULUS ANALYSIS OF A MODIFIED JEFFREYS MODEL

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Abstract. *Thixotropic materials have economically important industrial applications. Recently proposed models describing its behavior are formulated by means of a two coupled equations system: the constitutive equation (based on viscoelastic classic models) and the rate equation (in which the microstructural evolution is described). The shear modulus and the viscosity coefficient(s) are considered, in such a class of models, as functions of the structural parameter. The expressions used for the formulation of such functions are defined by satisfying some asymptotic limits, such that the model is physically consistent. However, there is no agreement as to the form in which the expressions are constructed. The Tikhonov regularization method together with the L-curve parameter-choice criterion is applied to determine the form of shear modulus function, using some experimental data from rheological tests. The function is obtained using a model that formally considers the microstructural dependence of the shear modulus and viscosity coefficients in the dynamic principle that origins the constitutive equation. The model, presented in a recent work, is deduced from the Smoluchowski coagulation theory and a modified Jeffreys model. It is considered the use of a multi-objective framework, for a more general solution of the problem. The algorithm is implemented through a MATLAB developed code. It is evidenced the necessity of regularization methods and the feasibility of the Tikhonov/ L-curve method and a new function for the shear modulus is proposed.*

Keywords: *Shear modulus, Thixotropic materials, Tikhonov regularization method, L-curve criterion, Multi-objective framework*

1 INTRODUCTION

Thixotropic materials are applied in many important industrial sectors such as the food, oil-extracting and coating industries, among others (Mewis & Wagner, 2009; Barnes, 1997). Commonly, it is aimed to propose a model that predicts the behavior of such materials in a robust and precise manner. However, there are still doubts associated with the approaches established in the literature (Mujumdar et al., 2002; Coussot et al., 2002; Dullaert & Mewis, 2006; Mewis, 1979; Barnes, 1997).

Recently proposed models are formulated from two coupled differential equations (Azikri de Deus et al., 2016; de Souza Mendes & Thompson, 2013; de Souza Mendes, 2009, 2011; Mujumdar et al., 2002; Azikri de Deus & Dupim, 2013): the constitutive and rate equations. The structural evolution of the material is considered in rate equation which is commonly proposed in the following form

$$\dot{\lambda} = \dot{\lambda}(\dot{\gamma}, \tau) \quad (1)$$

where τ stands for the shear stress of the material in a instant of time, $\dot{\gamma}$ for the shear strain rate and λ for the structural parameter, a variable that accounts the structural level of the material.

The constitutive equation is usually established in the general form

$$\tau = \tau(\dot{\gamma}, \lambda). \quad (2)$$

The explicit form of Eq. (2) is obtained by modifying classical viscoelastic models such as the Maxwellian, Jeffreys or Kelvin-Voigt models (Truesdell & Noll, 2004; Macosko & Larson, 1994; Bird et al., 1977): the classical models constants, such as the shear modulus G and the viscosity coefficients η_c , are considered as structural dependent functions in the modified models, i.e. $G \equiv G(\lambda)$ and $\eta_c \equiv \eta_c(\lambda)$. Despite the fact that these functions are defined by satisfying some physically consistent asymptotic limits, they may not have the more adequate form for a specific problem intermediately to those limits, contributing to errors in the response of the model. A possible way to work around this problem is to obtain the shape of the curves $G \times \lambda$ and $\eta_c \times \lambda$ from experimental data, in order to propose a consistent form for each function. This problem can be approached in a class commonly treated in the scope of the applied mathematics: the inverse problems.

Inverse problems are usually treated when it is aimed to determine the cause of some specific effect and conversely, for a direct problem, the effect due to a specific cause is intended (Lebedev et al., 2003; Kirsch, 2011; Isakov, 1998; Engl et al., 1996). These problems can be reduced, after some linearization process, to operator equations like:

$$Ax = y, \quad (3)$$

where it is aimed to determine x from the operator A and y . The problem (3) can be seen in a difference form as

$$\min_x \|Ax - y\|. \quad (4)$$

It might be intended to solve more than one problem in the form of Eq. (4), i.e., to solve problems like

$$\min_x \sum_{i=1}^N \vartheta_i \|A_i x - y_i\|, \quad (5)$$

where ϑ_i accounts the weight associated with each problem i . In recent works (Gong et al., 2016), it is shown that a multi-objective framework can be used to solve problems in the form of Eq. (5).

Inverse problems might be ill-posed and, therefore, an effort was made by the scientific community in order to develop more robust and fast convergence methods to solve them (Alves, 2005; Borges, 2010; Coliboro, 2011; Margotti, 2011; Ruiz Quiroz, 2014; Ramsay et al., 2007; Li et al., 2012; Liu et al., 2013). It has been shown that regularization methods could yield good results when applied in problems of physics and engineering (Azikri de Deus et al., 2012; Bazán et al., 2016; Eilks & Elliott, 2008). Among regularization methods, the Tikhonov regularization method is considered well established in the literature (Isakov, 1998; Engl et al., 1996; Lebedev et al., 2003; Hansen, 1998).

The aim of the present work is to determine the form of the shear modulus function $G(\lambda)$, used in the formulation of a thixotropic model (Azikri de Deus et al., 2016). In order to achieve this goal, it is considered a crude oil data under a constant shear strain rate test. Also, a multi-objective framework is adopted within the Tikhonov/L-curve regularization method in the treatment of ill posed problems involved. In the next section, the model is briefly presented. Some aspects about the Tikhonov regularization method and the L-curve criterion are succinctly treated in the section 3. The problems to be solved and their treatments are formalized at section 4. The multi-objective formulation of the problem is presented at section 5 and finally, at section 6, the numerical results obtained are presented.

2 A MODIFIED JEFFREYS MODEL

The model presented by Azikri de Deus et al. (2016) was proposed considering a isotropic thixotropic material under a isothermal process. From a microscopic point of view, the material can be seen as a continuum system composed of large chains of particles, with only one type of structure. The structural level is accounted by a variable λ . Right before the beginning of a shear load test, the material is considered to be fully structured ($\lambda = 1$). During the application of the load, the large chains might be decomposed into structures constituted of smaller chains, frequently named "flocs". As the load is maintained, these flocs might be decomposed into isolated particles (completely unstructured material, $\lambda = 0$). On the other hand, due to collisions of the particles associated with the Brownian motion and the laminar flow of the material, the above mentioned flocs can coalesce resulting in a restructuring process (Azikri de Deus & Dupim, 2013, 2012; Silva et al., 2014). In the following, the constitutive and the rate equations are succinctly described and the main aspects associated with those equations are exalted. Then, the asymptotic limits related with the shear modulus and the viscosity coefficients are treated and the equations of the model are summarized. At the end of the section, the main objective of this work is presented.

2.1 Constitutive and rate equations

It is considered a modified Jeffreys model adapted for thixotropic substances, as can be seen in Fig. 1, where the shear modulus G and the viscosity coefficients η_ν and η_μ are defined as functions of the structural parameter λ . Since G and η_μ are structure dependent properties,

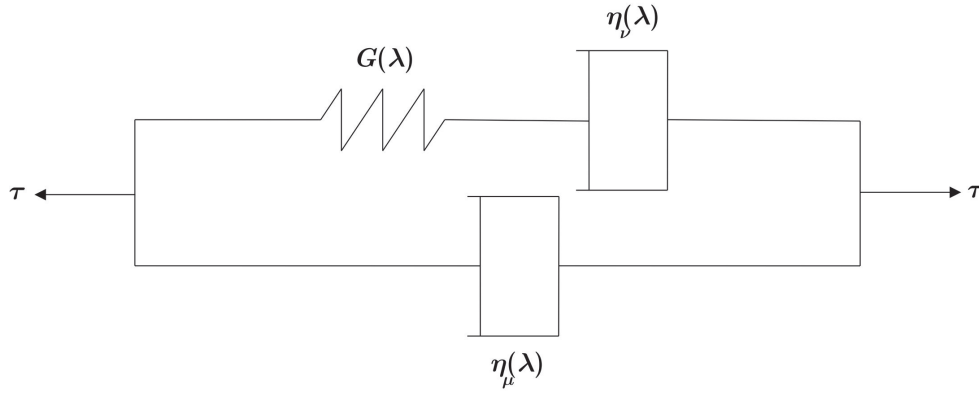


Figure 1: Sketch of the Modified Jeffreys model. Reprinted from the work of Azikri de Deus et al. (2016).

its rates ($\dot{G}$ and $\dot{\eta}_\mu$) are not necessarily null and they are considered at the explicit form of the constitutive equation:

$$\frac{2\eta_\nu}{G}\dot{\tau} + \left(1 - \frac{2\eta_\nu\dot{G}}{G^2}\right)\tau = \left[2\eta_\nu + 2\left(1 - \frac{2\eta_\nu\dot{G}}{G^2}\right)\eta_\mu + \frac{4\eta_\nu\dot{\eta}_\mu}{G}\right]\dot{\gamma} + \frac{4\eta_\nu\eta_\mu}{G}\ddot{\gamma}. \quad (6)$$

The development of the rate equation was based on the Reptation model, the Brownian motion and the Smoluchowski coagulation theory (de Gennes et al., 1971; Mazo, 2008) and the final form obtained was

$$\frac{d\lambda}{dt} = \frac{k_0^*}{t_f}(1 - \lambda)^\beta - \frac{(K_\psi\Theta\lambda^6\ddot{\gamma} + \tau)\lambda\dot{\gamma}}{\varsigma}, \quad (7)$$

where the parameter, k_0^* , β and t_f are related to the Brownian motion and laminar flow effects, θ is the absolute temperature, ς is the chemical potential and K_ψ^* incorporates some average micro-structural effects related to the relative motion between long chains of particles. In the following, the functions associated with the properties of the adapted Jeffreys model (i.e. the shear modulus $G(\lambda)$ and the viscosity coefficients $\eta_\nu(\lambda)$ and $\eta_\mu(\lambda)$) are presented.

2.2 Shear modulus

The definition of a consistent function of $G(\lambda)$ must take into account some restrictions. As $G(\lambda)$ tends to infinite and the viscosity coefficient associated with the Maxwell element $\eta_\nu(\lambda)$ assumes a finite value, then the material will exhibit a viscous behavior. On the other hand, if both G and η_ν assumes finite values, then a viscoelastic behavior is observed. Formally, it is expected that $\lim_{\lambda \rightarrow 0} G(\lambda) = \infty$ (de Souza Mendes, 2011; de Souza Mendes & Thompson, 2013; Macosko & Larson, 1994). It must be assumed also that as G assumes unbounded values, the solutions associated with the equations of the model must preserve stability. Therefore, another condition is commonly imposed: $\lim_{\lambda \rightarrow 0} \frac{\dot{G}}{G^2} = 0$. A function that satisfies both restriction is

$$G = G_0 \exp(m\lambda^{-1}), \quad (8)$$

where the shear modulus assumes a finite value ($G(1) = G_0 \exp(m)$) for $\lambda = 1$, and satisfies $\lim_{\lambda \rightarrow 0} G = \infty$.

It is considered that thixotropic materials will have its absolute viscosity¹ reduced as the structural level decays (de Souza Mendes, 2011). When considering a purely viscous material ($G(\lambda) \rightarrow \infty$), it can be checked from the constitutive equation (Eq. 6) that the absolute viscosity assumes the form

$$\eta = 2\eta_\mu + 2\eta_\nu. \quad (9)$$

From the Clausius-Duhem inequation (Azikri de Deus & Dupim, 2013; Truesdell & Noll, 2004) it is necessary that $\eta > 0$. Considering this restriction and the above mentioned considerations, the viscosity coefficients can be formulated in the following manner:

$$\begin{aligned} \eta_\mu &= \eta_0 \exp(\alpha_2 \lambda), \\ \eta_\nu &= \eta_0 \exp[(\alpha) \lambda] - \eta_\mu. \end{aligned} \quad (10)$$

$2\eta_0 > 0$ is the absolute viscosity when the material is completely unstructured ($\lambda = 0$) and $\alpha > \alpha_2 \geq 0$ are the parameters that accounts the structural influence in the viscous response of the material. The equations of the model are summarized next.

2.3 Equations of the model

The model is defined, in a general form by the following equations:

$$\frac{2\eta_\nu}{G} \dot{\tau} + \left(1 - \frac{2\eta_\nu \dot{G}}{G^2}\right) \tau = \left[2\eta_\nu + 2 \left(1 - \frac{2\eta_\nu \dot{G}}{G^2}\right) \eta_\mu + \frac{4\eta_\nu \dot{\eta}_\mu}{G}\right] \dot{\gamma} + \frac{4\eta_\nu \eta_\mu}{G} \ddot{\gamma}; \quad (11)$$

$$\frac{d\lambda}{dt} = \frac{k_0^*}{t_f} (1 - \lambda)^\beta - \frac{(K_\psi \Theta \lambda^6 \ddot{\gamma} + \tau) \lambda \dot{\gamma}}{\varsigma}; \quad (12)$$

$$G = G_0 \exp(m \lambda^{-1}); \quad (13)$$

$$\eta_\mu = \eta_0 \exp(\alpha_2 \lambda); \quad (14)$$

$$\eta_\nu = \eta_0 \exp[(\alpha) \lambda] - \eta_\mu, \quad (15)$$

Considering $\kappa \equiv \frac{k_0^* \varsigma}{t_f}$ and $K_\psi^* \equiv K_\psi \Theta$, the rate equation can be rewritten in a more compact manner

$$\frac{d\lambda}{dt} = \frac{1}{\varsigma} [\kappa (1 - \lambda)^\beta - (K_\psi^* \lambda^6 \ddot{\gamma} + \tau) \lambda \dot{\gamma}], \quad (16)$$

With the rate equation written in the above form, the model have nine positives parameters K_ψ^* , κ , ς , G_0 , m , η_0 , α , α_2 and β .

The form of the function $G(\lambda)$ defined in Eq.(13) satisfies the asymptotic limits mentioned at the beginning of the previous subsection. However, it can be verified that such form is not the only one with such property. Therefore, it is aimed to *determine* the form of G that fits best the

¹Defined by the ratio $\eta \equiv \frac{\tau}{\dot{\gamma}}$.

model with respect to experimental data. In this sense, an inverse problem approach is proposed at section 4 and the results of its application using crude oil data are presented at section 6. The Tikhonov regularization method and the L-curve criterion were considered for the treatment of the related ill-posed problems. Therefore, before presenting the formulation, a discussion with respect to these techniques are presented in the following section.

It is important to mention that, as for the function associated with the shear modulus, a similar approach can be made regarding the viscosity coefficients. Although it is considered only the shear modulus in the present work as a "trial procedure".

3 TIKHONOV METHOD AND L-CURVE CRITERION

The inverse problems which are aimed to be solved at the present work, can be described in the following general form²: find $\underline{\Delta x}_s \in \mathbb{R}^m$ such that

$$\underline{\Delta x}_s = \min_{\underline{\Delta x} \in \mathbb{R}^m} \|\underline{\Delta} \underline{\Delta x} - \underline{\mathbf{f}}\|_2, \quad \underline{\Delta} \in \mathbb{R}^{n \times m}, \quad \underline{\mathbf{f}} \in \mathbb{R}^n. \quad (17)$$

The standard solutions of problems with the form of Eq. (17) are the least square (LS) solutions, which can be represented as $\underline{\Delta x}_{LS} = \underline{\Delta}^\dagger \underline{\mathbf{f}}$ (where $\underline{\Delta}^\dagger$ denotes the pseudoinverse of $\underline{\Delta}$) (Hansen, 1998). These kind of solutions may presents numerical spurious error, which might contributes to solution instability and therefore to ill-posedness problems. In this sense, the Tikhonov regularization method is frequently adopted as a well-established form to reach a stable solution. It can be generally described as (Lebedev et al., 2003; Hansen, 1998): find $\underline{\Delta x}_\aleph \in \mathbb{R}^m$ such that

$$\underline{\Delta x}_\aleph = \min_{\underline{x}^{(j+1)} \in \mathbb{R}^m} \|\underline{\Delta} \underline{\Delta x} - \underline{\mathbf{f}}\|_2^2 + \aleph \|\underline{\Delta x}\|_2^2 \quad (18)$$

where $\aleph > 0$ is the regularization parameter. Analogously, the problem proposed in Eq.(18) can be described as to find the solution of

$$(\underline{\Delta}^T \underline{\Delta} + \aleph \underline{\mathbb{I}}) \underline{\Delta x} = \underline{\Delta}^T \underline{\mathbf{f}}; \quad (19)$$

$$\therefore \underline{\Delta x}_\aleph = (\underline{\Delta}^T \underline{\Delta} + \aleph \underline{\mathbb{I}})^{-1} \underline{\Delta}^T \underline{\mathbf{f}}, \quad (20)$$

where $\underline{\mathbb{I}}$ is the identity matrix $m \times m$. Another way to write the problem is considering the singular-value decomposition (SVD) of $\underline{\Delta}$: $\underline{\Delta} = \underline{\hat{\mathbb{S}}}_1 \underline{\hat{\mathbb{S}}}_2 \left(\underline{\hat{\mathbb{S}}}_3 \right)^T$, where $\underline{\hat{\mathbb{S}}}_2 \in \mathbb{R}^{n \times m}$ is a singular value diagonal matrix, and $\underline{\hat{\mathbb{S}}}_1 \in \mathbb{R}^{n \times n}$, $\underline{\hat{\mathbb{S}}}_3 \in \mathbb{R}^{m \times m}$ are unitary matrices, with $\underline{\hat{\mathbb{S}}}_3$ non singular matrix. Regarding the SVD, the Thikhonov problem (18) can be rewritten as

$$(\underline{\Delta}^T \underline{\Delta} + \aleph \underline{\mathbb{I}}) \underline{\Delta x}_\aleph = \underline{\Delta}^T \underline{\mathbf{f}}; \quad (21)$$

$$\therefore \underline{\Delta x}_\aleph = \underline{\hat{\mathbb{S}}}_3 \left(\left(\underline{\hat{\mathbb{S}}}_2 \right)^2 + \aleph \underline{\mathbb{I}} \right)^{-1} \underline{\hat{\mathbb{S}}}_2 \left(\underline{\hat{\mathbb{S}}}_1 \right)^T \underline{\mathbf{f}}, \quad (22)$$

or

$$\underline{\Delta x}_\aleph = \sum_{i=1}^n \frac{\hat{\mathbb{S}}_{2_i}^2}{\hat{\mathbb{S}}_{2_i}^2 + \aleph^2} \frac{\left(\underline{\hat{\mathbb{S}}}_{1_i}^T \underline{\mathbf{f}} \right)}{\hat{\mathbb{S}}_{2_i}} \underline{\hat{\mathbb{S}}}_{3_i}, \quad (23)$$

²See sections 4 and 5.

considering $\hat{S}_{2_i}^2$ as the i -th singular value, $\hat{\underline{S}}_{1_i}$ the i -th column vector of $\hat{\underline{S}}_1$ and $\hat{\underline{S}}_{3_i}$ the i -th column vector of $\hat{\underline{S}}_3$. As expected (Kirsch, 2011), the solution of the problem Eq.(18) converges to the solution of the original problem $\underline{A}\underline{\Delta}x = \underline{f}$ as $\aleph$ tends to zero.

In order that the solution $\underline{\Delta}x_{\aleph}$ be the closest solution to one not influenced by the spurious errors, it is important to note that the criterion for the determination of $\aleph$ plays an important role. From the criterion techniques related to the choice of regularization parameter presented in the specialized literature (Engl et al., 1996; Hansen, 1998; Isakov, 1998), the L-curved method (Hansen, 1999) was chosen due to its versatility.

Let $\underline{\Delta}x_{\aleph}$ be the solution of

$$\min_{\underline{\Delta}x \in \mathbb{R}^m} \|\underline{A}\underline{\Delta}x - \underline{f}\|_2^2 + \aleph \|\underline{\Delta}x\|_2^2 \quad (24)$$

and let $s_{1_{\aleph}} := \sqrt{\mathbb{k}_{1_{\aleph}}}$ and $s_{2_{\aleph}} := \sqrt{\mathbb{k}_{2_{\aleph}}}$, where

$$\mathbb{k}_{1_{\aleph}} := \|\underline{A}\underline{\Delta}x_{\aleph} - \underline{f}\|_2^2 \text{ and } \mathbb{k}_{2_{\aleph}} := \|\underline{\Delta}x_{\aleph}\|_2^2. \quad (25)$$

It can be verified that if the function $\aleph \mapsto s_{1_{\aleph}}$ is increasing, $\aleph \mapsto s_{2_{\aleph}}$ is decreasing and vice versa. Furthermore, it can be shown that the bounded set

$$\mathfrak{B} := \{(b_1, b_2) \in \mathbb{R}^2 \mid \exists \underline{\Delta}x_{\aleph} \in \mathbb{R}^m \text{ with } s_{1_{\aleph}} \leq b_1 \text{ and } s_{2_{\aleph}} \leq b_2\},$$

is a convex set with boundary described by $\aleph \mapsto (s_{1_{\aleph}}, s_{2_{\aleph}})$ curve.

It is expected that an optimal $\aleph$ value must minimize both $s_{1_{\aleph}}$ and $s_{2_{\aleph}}$ (as it can be seen from the Tikhonov problem). However, when plotted the curve $\ell : \aleph \in (0, \infty) \mapsto (\ln(s_{1_{\aleph}}), \ln(s_{2_{\aleph}})) \in \mathbb{R}^2$ for ill-posed problems, a typical L-shaped curve is encountered (Hansen, 1999; Azikri de Deus et al., 2012; Hansen, 1998): at the vertical region of ℓ -curve, small changes in $\aleph$ values correspond to large varying of $s_{2_{\aleph}}$ and small change in $s_{1_{\aleph}}$; on the other hand on horizontal part of the graph large variations of $\aleph$ corresponds to a small perturbation of $s_{2_{\aleph}}$ value, where ℓ -curve exhibits a flat (slowly decreasing) behavior. The transitions between these two regions is located a corner. The L-curve criterion consists in determining the regularization parameter $\aleph$ which maximizes the curvature of the ℓ -curve, i.e., it is aimed to determine $\aleph$ that corresponds to the corner of the typical L-shaped curve.

Let $\underline{R}_{\perp}$ be defined as the component of $\underline{f}$ orthogonal to $\hat{\underline{S}}_{1_1}, \dots, \hat{\underline{S}}_{1_n}$ (i.e. the least squares residual). Then it follows that

$$\begin{aligned} \mathbb{k}_{1_{\aleph}} &= \sum_{i=1}^n \frac{\aleph^4 \left(\hat{\underline{S}}_{1_i}^T \underline{f} \right)^2}{\left(\hat{S}_{2_i}^2 + \aleph^2 \right)^2} + \|\underline{R}_{\perp}\|_2^2 \therefore \frac{d\mathbb{k}_{1_{\aleph}}}{d\aleph} = 4\aleph^3 \sum_{i=1}^n \frac{\hat{S}_{2_i}^2 \left(\hat{\underline{S}}_{1_i}^T \underline{f} \right)^2}{\left(\hat{S}_{2_i}^2 + \aleph^2 \right)^3}; \\ \mathbb{k}_{2_{\aleph}} &= \sum_{i=1}^n \frac{\hat{S}_{2_i}^2 \left(\hat{\underline{S}}_{1_i}^T \underline{f} \right)^2}{\left(\hat{S}_{2_i}^2 + \aleph^2 \right)^2} \therefore \frac{d\mathbb{k}_{2_{\aleph}}}{d\aleph} = -4\aleph \sum_{i=1}^n \frac{\hat{S}_{2_i}^2 \left(\hat{\underline{S}}_{1_i}^T \underline{f} \right)^2}{\left(\hat{S}_{2_i}^2 + \aleph^2 \right)^3}; \\ \therefore \frac{d\mathbb{k}_{2_{\aleph}}}{d\mathbb{k}_{1_{\aleph}}} &= -\aleph^{-2}. \end{aligned} \quad (26)$$

Since the main problem associated with the L-curve criterion is to obtain the maximum curvature point of the curve ℓ (L-curve), the curvature $\mathfrak{J}$, as regularization parameter ($\aleph$) function, can be computed, in a continuous sense, as

$$\mathfrak{J}(\aleph) = \frac{s'_{1\aleph} s''_{2\aleph} - s''_{1\aleph} s'_{2\aleph}}{\left((s'_{1\aleph})^2 + (s'_{2\aleph})^2 \right)^{\frac{3}{2}}}, \quad (27)$$

where $(\cdot)' \equiv \frac{\partial(\cdot)}{\partial \aleph}$. In the present work, the L-curve consists of a finite (discrete) number of pairs $(s_{1\aleph}, s_{2\aleph})$ corresponding to different regularization parameter ($\aleph$) values. Therefore, a smooth β -cubic spline is fitted on discrete L-curve points. This procedure holds twice differentiability, stability in numerical differentiation, besides local shape characteristics preserving. The main steps involved in the Tikhonov/L-curve approach are related below.

The Tikhonov/L-curve Procedure

- i. From a few regularization parameter values $\aleph^{(i)}$, $i = 1, \dots, n_i$, range from higher until smaller values, generate points $\left(\ln(s_{1\aleph}^{(i)}), \ln(s_{2\aleph}^{(i)}) \right)$;
- ii. Compute a β -cubic spline curve $\mathfrak{S}$, with L-curve fitting algorithm, from $\left(\ln(s_{1\aleph}^{(i)}), \ln(s_{2\aleph}^{(i)}) \right)$ points, $i = 1, \dots, n_i$;
- iii. Determine the maximum curvature point on $\mathfrak{S}$, restricted to the $\left(\ln(s_{1\aleph}^{(i)}), \ln(s_{2\aleph}^{(i)}) \right)$ -plan, and the corresponding optimal regularization parameter $\aleph_*$;
- iv. Solve the regularization problem for $\aleph = \aleph_*$ and add the new point $\left(\ln(s_{1\aleph}^{(*)}), \ln(s_{2\aleph}^{(i)}) \right)$, $\aleph_{(*)}$ to the L-curve;
- v. Repeat from step ii until convergence.

The procedure described above will be used in the treatment of badly conditioned matrix. In the following, the problems to be solved at the present work are defined.

4 NUMERICAL APPROACH

4.1 Shear modulus curves

It is considered, as a first approximation, the expression $G = G_0 \exp\left(\frac{m}{\lambda}\right)$ for the shear modulus. As mentioned above, the model parameters are K_ψ^* , κ , ς , G_0 , m , η_0 , α , α_2 and β . In the present work, it will be considered data of a constant shear rate test of crude oil, already presented in a previous work (Tarcha et al., 2015). Since for these tests $\ddot{\gamma} \approx 0$, the definition of K_ψ^* is not necessary and will be disregarded in the present analysis. Therefore, a vector containing the parameters related to constant shear rate tests is defined as

$$\underline{W} = [\eta_0 \ \alpha \ \alpha_2 \ \kappa \ \beta \ \varsigma \ G_0 \ m]^T. \quad (28)$$

From physical considerations (Silva, 2015), it is possible to make a good estimate $\underline{W}_0$ for the parameters in a constant shear rate test, such that the model response $\tau(\underline{W}) \in \mathbb{R}^{N_t}$ is

approximately close to the experimental data $\underline{\tau}^{exp} \in \mathbb{R}^{N_t}$, for N_t intervals of time³. Considering that $\underline{W}_0$ in fact represents a good choice for the model response, it is aimed to determine the shape of the curve " $G \times \lambda$ " that fits best with respect to the experimental data. In this sense, it is defined the following vector (with model parameters not related with G):

$$\underline{W}_c = [\eta_0 \ \alpha \ \alpha_2 \ \kappa \ \beta \ \varsigma]^T. \quad (29)$$

Fixing the values of $\underline{W}_c$, such that $(\underline{W}_c)_i = (\underline{W}_0)_j$, for every integer $j \leq 6$, the first problem to be solved in this work is defined: considering $\underline{W}_c$ given and a fixed value of $\dot{\gamma}$, determine $\underline{G} \in \mathbb{R}^{N_t}$ and $\underline{\lambda} \in \mathbb{R}^{N_t}$ that satisfies, for every $i \in \{1, 2, 3 \dots N_t\}$, the following set of equations:

$$\left(\frac{2\eta_\nu}{G_i} \right)_i \dot{\tau}_i^{exp} + \left(1 - \frac{2\eta_\nu \dot{G}}{G^2} \right)_i \tau_i^{exp} = \left[2\eta_\nu + 2 \left(1 - \frac{2\eta_\nu \dot{G}}{G^2} \right) \eta_\mu + \frac{4\eta_\nu \dot{\eta}_\mu}{G} \right]_i \dot{\gamma}, \quad (30)$$

and

$$\dot{\lambda}_i = \frac{1}{\varsigma} \left[\kappa(1 - \lambda_i)^\beta - \tau_i^{exp} \lambda_i \dot{\gamma} \right], \quad (31)$$

where

$$(\eta_\mu)_i = \eta_0 \exp(\alpha_2 \lambda_i); \quad (32)$$

$$(\eta_\nu)_i = \eta_0 \exp[(\alpha) \lambda_i] - (\eta_\mu)_i, \quad (33)$$

and $(\cdot)_i$ represents the value of $(\cdot)$ at the i -th interval of time. It is important to mention that this problem is solved using an explicit Euler method.

Once $\underline{G}$ and $\underline{\lambda}$ are determined, it is possible to trace the curve $\ell_G : t_i, i \in \{1, 2, 3 \dots, N_t\} \mapsto (\lambda_i, G_i) \in \mathbb{R}^2$. Depending on the shape of ℓ_G , a new form for the function related with the shear modulus $G^N(\lambda)$ and a new set of model parameters $\underline{W}$ are proposed, in order that

$$\underline{W} = [\eta_0 \ \alpha \ \alpha_2 \ \kappa \ \beta \ \varsigma \ GP_1 \ GP_2 \ \dots]^T. \quad (34)$$

where $GP_1, GP_2 \dots$, are the parameters associated with the new shear modulus function G^N . In the following the procedure to determine $\underline{W}$, considering the new proposed form $G^N(\lambda)$ is considered.

4.2 New set of parameters

The model presented in section 2 is redefined, since the shear modulus function were changed to a new form $G^N(\lambda)$. Thus, the new model is constituted of Eqs. (11), (16), (14) and (15), but Eq. (13) is redefined as $G^N(\lambda)$. The aim is, in this part of the work, to define the best set of parameters that fits this new proposed model to experimental data. This problem is explicitly non-linear and therefore a linearization process is needed. In the sequence, it is described the Gauss- Newton approach related to the linearization of the problem.

³It is considered that the N_t intervals and experimental data $\underline{\tau}^{exp} \in \mathbb{R}^{N_t}$, refers to the results of a treatment and interpolation process of the direct obtained data. This process is necessary in order that $\dot{\tau}_i^{exp}$ may be determined: see Eq. (30).

It is aimed to determine $\underline{W}$ such that

$$\underline{W} = \operatorname{argmin} \mathbb{J}(\underline{W}), \quad (35)$$

with $\mathbb{J}(\underline{W}) = \sum_{i=1}^{N_t} \|\underline{\tau}_i(\underline{W}) - \underline{\tau}_i^{exp}\|_2^2$, where $\underline{\tau}_i(\underline{W})$ is the shear stress vector computed from the new model. At the j -th step of the Gauss-Newton method, the term $\mathbb{J}$ may be rewritten in the following way

$$\therefore \mathbb{J}(\underline{W}^{(j)}) = \|\underline{r}(\underline{W}^{(j)})\|_2^2, \quad (36)$$

with

$$\underline{r}_i(\underline{W}^{(j)}) = (\underline{\tau}_i(\underline{W}^{(j)}) - \underline{\tau}_i^{exp}); \quad (37)$$

$$\therefore \underline{r}(\underline{W}^{(j)}) = \begin{pmatrix} \underline{r}_1(\underline{W}^{(j)}) \\ \underline{r}_2(\underline{W}^{(j)}) \\ \vdots \\ \underline{r}_n(\underline{W}^{(j)}) \end{pmatrix}; \quad (38)$$

Considering a first order expansion,

$$\begin{aligned} \underline{r}_i(\underline{W}^{(j+1)}) &\approx \underline{r}_i(\underline{W}^{(j)}) + \frac{\partial \underline{r}_i}{\partial \underline{W}}(\underline{W}^{(j)}) (\underline{W}^{(j+1)} - \underline{W}^{(j)}); \\ &= \underline{A}^{(j)} \underline{\Delta W}^{(j)} - \underline{b}^{(j)}, \end{aligned} \quad (39)$$

where

$$\underline{A}^{(j)} = \frac{\partial \underline{r}_i}{\partial \underline{W}}(\underline{W}^{(j)}), \quad \underline{b}^{(j)} = -\underline{r}_i(\underline{W}^{(j)}) \text{ and } \underline{\Delta W}^{(j)} = \underline{W}^{(j+1)} - \underline{W}^{(j)}, \quad (40)$$

at j -th iteration, one has

$$\|\underline{r}(\underline{W}^{(j+1)})\|_2^2 \approx \|\underline{A}^{(j)} \underline{\Delta W}^{(j)} - \underline{b}^{(j)}\|_2^2. \quad (41)$$

Thus, the problem at j -th iteration step corresponds to solve the following problem

$$\min_{\underline{\Delta W}^{(j)} \in \mathbb{R}^m} \|\underline{A}^{(j)} \underline{\Delta W}^{(j)} - \underline{b}^{(j)}\|_2, \quad \underline{A}^{(j)} \in \mathbb{R}^{n \times m}, \quad \underline{b}^{(j)} \in \mathbb{R}^n. \quad (42)$$

In the case that one faces convergence problems due to high condition number, the Tikhonov/L-curve method is used, so that one aims to solve the problem (see section 3)

$$\min_{\underline{\Delta W}^{(j)} \in \mathbb{R}^m} \|\underline{A}^{(j)} \underline{\Delta W}^{(j)} - \underline{b}^{(j)}\|_2 + \aleph \|\underline{\Delta W}\|_2, \quad \underline{A}^{(j)} \in \mathbb{R}^{n \times m}, \quad \underline{b}^{(j)} \in \mathbb{R}^n. \quad (43)$$

It is important to mention that the above problem is associated with only one test. In the following, it is considered a multiobjective framework, in order that more than one test can be considered simultaneously at the minimization process.

5 MULTI-OBJECTIVE FRAMEWORK

It is considered N_e tests. For each test, equations of the form of Eq.(42) is obtained, and therefore, the operators and vectors associated with each tests are enumerated with an index i , in order that the functional

$$\mathcal{F}_i(\underline{\Delta W}^{(j)}) = \|\underline{A}_i^{(j)} \underline{\Delta W}^{(j)} - \underline{b}_i^{(j)}\|_2^2, \quad \underline{A}_i^{(j)} \in \mathbb{R}^{n \times m}, \underline{\Delta W}^{(j)} \in \mathbb{R}^m \quad (44)$$

will be minimized at the j -th Newton step for the i -th test.

The multiobjective strategy consists in minimize the functional

$$\mathcal{F}(\underline{\Delta W}^{(j)}) = \sum_{i=1}^{N_e} \vartheta_i \mathcal{F}_i(\underline{\Delta W}^{(j)}) \quad (45)$$

where ϑ_i represents the weight associated with i -th test, i.e. it is aimed to solve the problem

$$\min_{\underline{\Delta W}^{(j)} \in \mathbb{R}^m} \mathcal{F}(\underline{\Delta W}^{(j)}). \quad (46)$$

The functional $\mathcal{F}(\underline{\Delta W}^{(j)})$ can be written in a more explicit manner

$$\mathcal{F}(\underline{\Delta W}^{(j)}) = \|\underline{A}^* \underline{\Delta W}^{(j)} - \underline{b}^*\|_2^2 \quad (47)$$

where

$$\underline{A}^* = \begin{bmatrix} \sqrt{\vartheta_1} \underline{A}_1^j \\ \sqrt{\vartheta_2} \underline{A}_2^j \\ \vdots \\ \sqrt{\vartheta_{N_e}} \underline{A}_{N_e}^j \end{bmatrix}; \quad \underline{b}^* = \begin{bmatrix} \sqrt{\vartheta_1} \underline{b}_1^j \\ \sqrt{\vartheta_2} \underline{b}_2^j \\ \vdots \\ \sqrt{\vartheta_{N_e}} \underline{b}_{N_e}^j \end{bmatrix}. \quad (48)$$

(49)

The Thikhonov regularization for the multiobjective framework defined above consists in obtaining the solution of the problem

$$\min_{\underline{\Delta W}^{(j)} \in \mathbb{R}^m} \left\{ \mathcal{F}(\underline{\Delta W}^{(j)}) + \aleph \|\underline{\Delta W}^{(j)}\|_2^2 \right\} \quad (50)$$

with $\mathcal{F}$ defined in Eq.(47).

The procedure adopted, in order that a new form of the shear modulus function can be obtained, is summarized below.

Determination of the shear modulus procedure

- i. An estimate for the model parameters W_0 is obtained using physical assumptions as those considered in the work of Silva (2015). The shear modulus function considered is the one defined in the original model.

- ii. Considering the parameters not related with the shear modulus function $\underline{W}_c$, solve the Eqs. (30) to (33) and, for each of the N_e strain rate test, plot the curve ℓ_G and define the most convenient new proposal to the shear modulus function, $G^N(\lambda)$.
- iii. Set $j = 0$ and define $\underline{W}^{(0)}$.
- iv. Solve for each j the problem (43), when one test is considered at a time; or solve the multiobjective problem (Eq.(50)) if considered more than one test at a time;
- v. Repeat from step iv until convergence.

The procedure described above were implemented in a MATLAB developed code and its results are treated next.

6 RESULTS

6.1 $G \times \lambda$ curves

It was considered as a first estimate ($\underline{W}_0$), the following parameters: $\eta_0 = 0.09 \text{ Pa.s}$, $\alpha = 15.00$, $\alpha_2 = 9.59$, $\kappa = 0.11 \text{ J/m}^3$, $\beta = 1$, $m = 10^{-6}$, $G_0 = 6.5 \text{ Pa}$ and $\varsigma = 5 \times 10^5 \text{ J s/m}^3$. The results of the simulation of the original model (considering G defined at Eq. (13)) for a set of constant strain rate tests are shown at Figs. 2 and 3, for the steady and transient regimes, respectively.

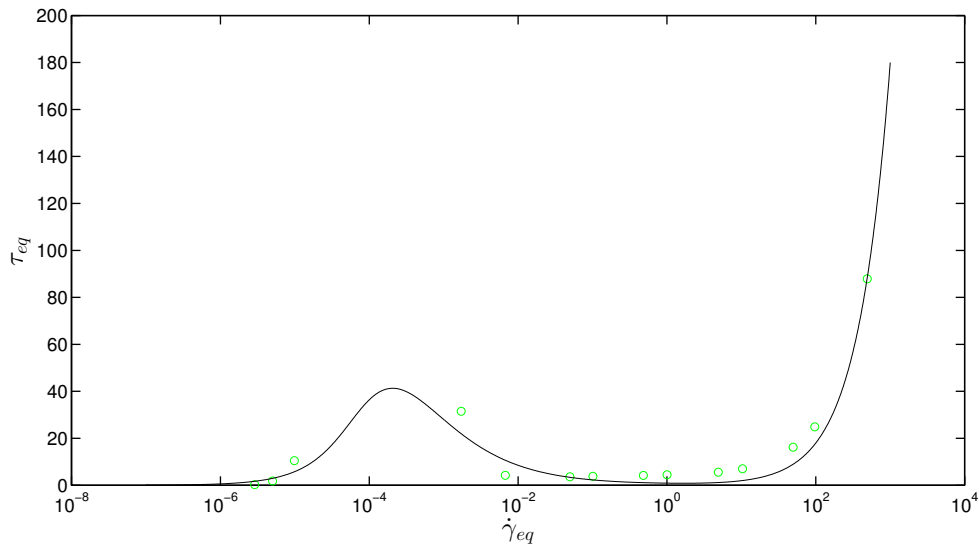


Figure 2: Flow curve: model response (solid lines) vs. experimental data (circles) for constant shear strain rate test, at steady state regime.

The trial estimate is in good agreement with the experimental data, as it can be seen. However, for large strain rate imposed values, there is a significant discrepancy between the response of the model and the experimental data. In this sense, the ℓ_G curves for these tests were obtained using the methodology defined in the previous section. The results for shear strain rate of 10 and 100 s^{-1} are shown in Figs. 4 and 5.

The ℓ_G curves obtained do not corresponds to the original form of the shear modulus, i.e. $G(\lambda) \neq G_0 \exp \frac{m}{\lambda}$. A new propose that satisfies the limits mentioned at subsection 2.2 and

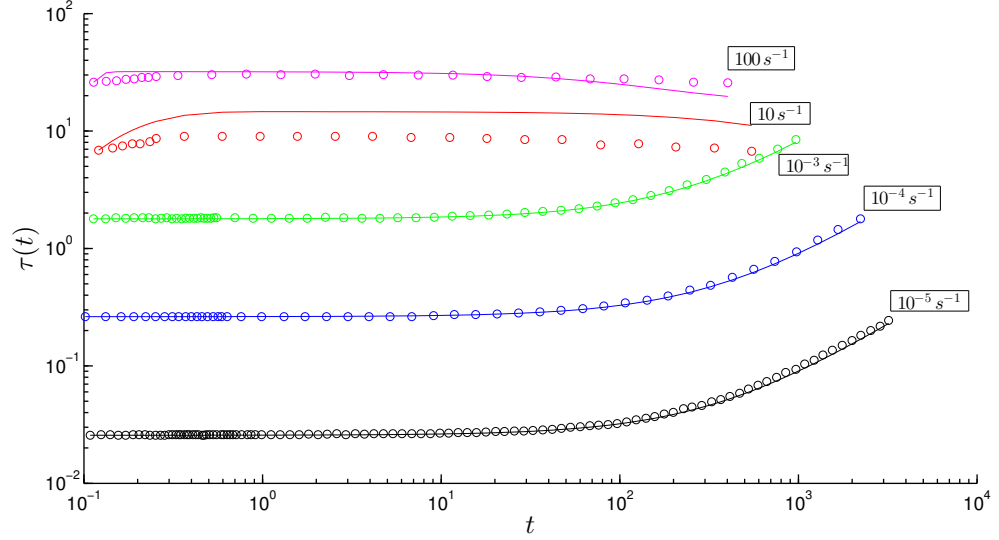


Figure 3: Constant shear rate test: model response (solid lines) vs. experimental data (circles) for constant shear strain rate test - $G = G_0 \exp \frac{m}{\lambda}$

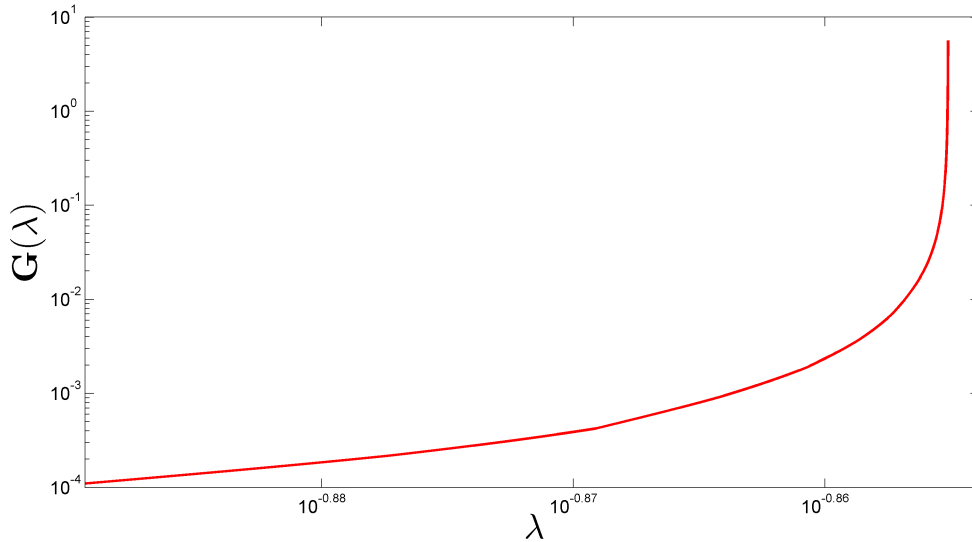


Figure 4: Shear modulus obtained from constant strain rate data, $\dot{\gamma} = 10 \text{ s}^{-1}$.

may exhibit the shape of the curves shown above is $G^N(\lambda) = \lambda G_0^* \exp \frac{m^*}{\lambda}$. The next step is to determine the parameters associated with the modified model:

$$\underline{W} = [\eta_0 \ \alpha \ \alpha_2 \ \kappa \ \beta \ \varsigma \ G_0^* \ m^*]^T. \quad (51)$$

6.2 Parameter estimation: new form for $G(\lambda)$

The Tikhonov regularization together with L-curve criterion were used to obtain the values of parameters for the new proposed form for the shear modulus. The computational parameters obtained are exposed at Table 1: the condition number were calculated as the ratio between the the greatest and smallest eigenvalue and the "time" represents the processing time. The

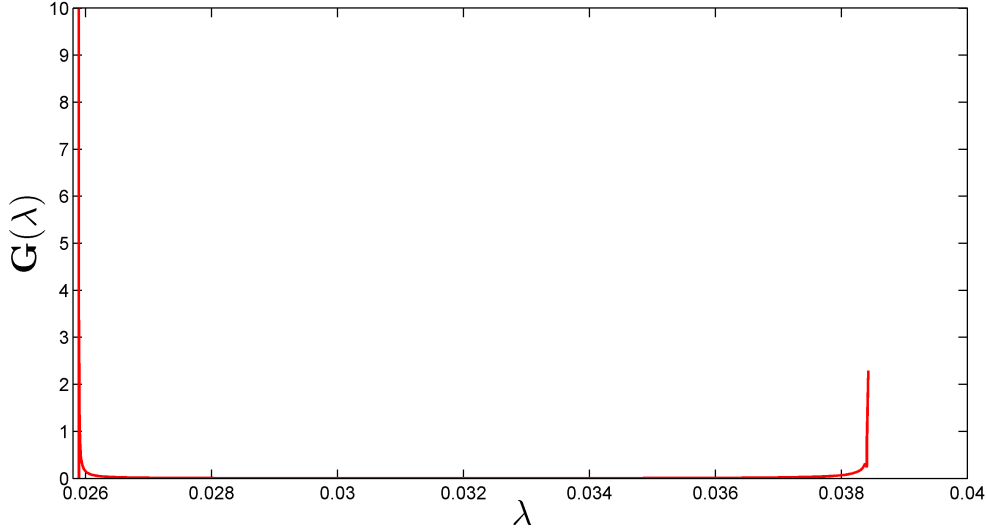


Figure 5: Shear modulus obtained from constant strain rate data, $\dot{\gamma} = 100 \text{ s}^{-1}$.

initial model parameters considered $\underline{W}^0$ (step iii of the procedure presented at the final of the previous section) was the same as the considered above: $\eta_0 = 0.09 \text{ Pa.s}$, $\alpha = 15.00$, $\alpha_2 = 9.59$, $\kappa = 0.11 \text{ J/m}^3$, $\beta = 1$ and $\varsigma = 5 \times 10^5 \text{ J s/m}^3$, with $m^* = 10^{-6}$, $G_0^* = 6.5 \text{ Pa}$ initially. For all results obtained it was considered a time discretization of $\Delta_t = 10^{-3} \text{ s}$ (although similar results were obtained for $\Delta_t = 10^{-4} \text{ s}$).

Table 1: Computational parameters associated with the Tikhonov/ L-curve method: $G^N = \lambda G_0^* \exp \frac{m^*}{\lambda}$.

Strain rate [s^{-1}]	$\frac{\ \Delta W\ }{\ W\ }$	Cond. Number	$\aleph$	Time [s]
10^{-5}	2.49×10^{-6}	3.93×10^{23}	0.3125	17.4255
10^{-4}	6.53×10^{-6}	3.92×10^{23}	1.7999	12.1981
10^{-3}	1.69×10^{-7}	2.11×10^{22}	686.9803	5.4354
10	7.83×10^{-6}	3.75×10^7	21.8151	3.1493
100	3.69×10^{-6}	5.11×10^7	58.8218	2.5480
$[10^{-5}, 10^{-4}, 10^{-3}, 10, 100]$	9.39×10^{-8}	2.02×10^7	25069.7863	45.2883

It can be seen from Table 1 that high condition numbers were obtained, which leads to the conclusion that the problem is effectively an ill-posed one. An important aspect to be mentioned is that low processing time were necessary to obtain the solution.

The model parameters obtained were the same as the initial one: at the first Newton step, the convergence criterion ($\frac{\|\Delta W\|}{\|W\|} \leq 10^{-5}$) was already satisfied, which is expected if it is a minimum of the function. It is important to mention that for more restricted criteria, the problem converges to approximately the same values and, as expected, the same procedure using the least square solution has not been able to circumvent the ill-posedness of the problem, i.e., there was

no convergence.

It is exposed at Fig. 6 the L-curve obtained for the multi-objective problem: the L-shaped form of the curve is evident. It is necessary to evident that similar curves were obtained consid-

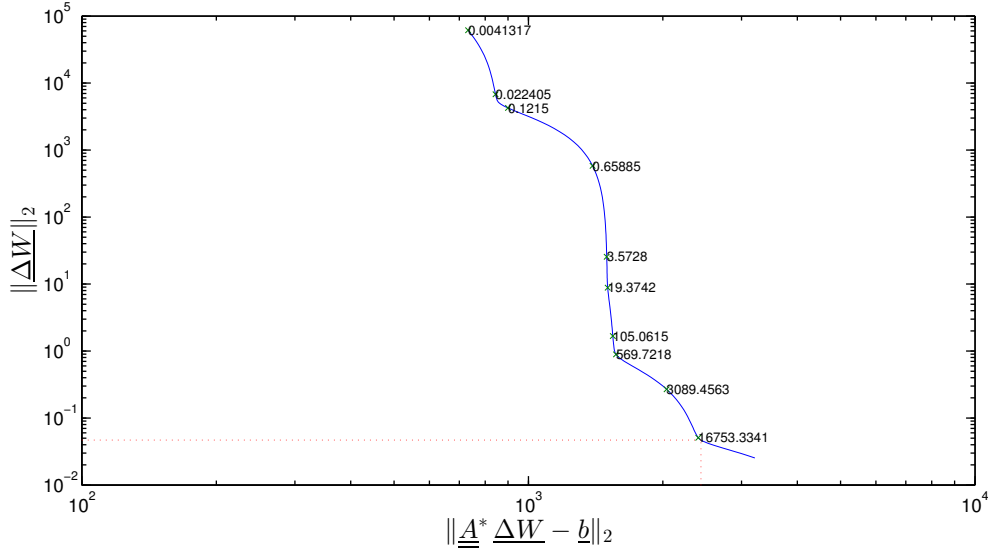


Figure 6: L-curve using the multiobjective framework, for the tests $\dot{\gamma} = 10^{-5}, 10^{-4}, 10^{-3}, 10$ and 100 s^{-1} - $G = G_0 \lambda \exp\left(\frac{m}{\lambda}\right)$.

ering the tests individually. As an example it is shown at Fig. 7 the L-curve of the $\dot{\gamma} = 10^{-5} \text{ s}^{-1}$, where larger condition number was observed. It can be noted that the L-shaped form for this larger condition number problem is more pronounced.

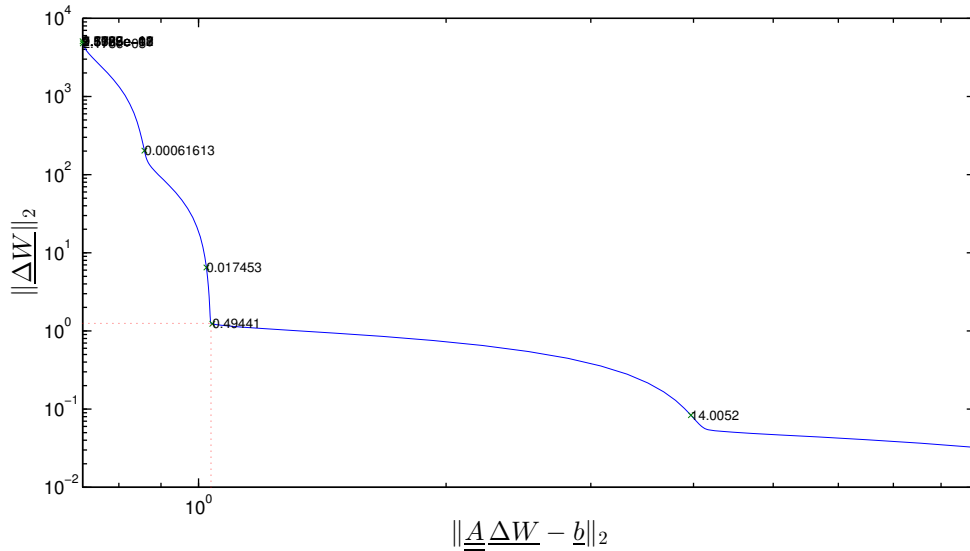


Figure 7: L-curve, $\dot{\gamma} = 10^{-5} \text{ s}^{-1}$, $G = G_0 \lambda \exp\left(\frac{m}{\lambda}\right)$.

At Fig. 8 it is compared the simulation of the model considering the form G^N for the shear modulus and the experimental data.

It is important to note the difference between Figs. 8 and 3: although they present similar results after long times, at the first seconds of large strain rates imposed tests, the new pro-

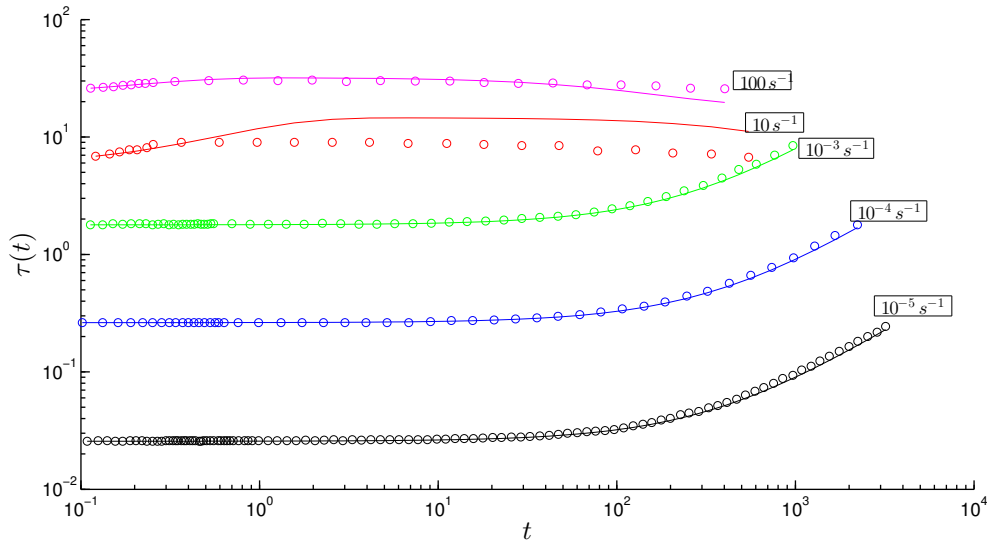


Figure 8: Constant shear rate test: model response (solid lines) vs. experimental data (circles) for constant shear strain rate test: $G = G_0^* \lambda \exp \frac{m^*}{\lambda}$

posed form for shear modulus contributes to a significant better agreement between the model response and the experimental data.

It can be concluded from the above exposed results that the methodology presents consistent solutions and contributes to a better agreement between the experimental data and the values obtained from the simulation of the model. It is important to note that the new form proposed at the present work is not the only possible one that satisfies the requirements mentioned throughout the text. However, it is an achievement of this work that a new form for the shear modulus can (and from the above results, must) be obtained, meaning that the study of the form for such functions can be a fruitful possible research area and can yields better agreement with respect to the experimental data.

FINAL REMARKS

The main objective of the present work was to analyze the form of the function associated with the shear modulus of a thixotropic model (Azikri de Deus et al., 2016). It can be mentioned that some goals were achieved:

- i The context in which the shear modulus functions are defined were presented and a brief description of the model proposed by Azikri de Deus et al. (2016) was made;
- ii The Tikhonov regularization method and the L-curve criterion were presented and adopted in the treatment of badly conditioned matrices;
- iii The numerical approach was formally defined and an algorithm were constructed in order to deduce the form of the shear modulus function that fits best with experimental data. The multi-objective framework was adopted together with crude oil constant strain rate tests data. The implementation of the algorithm was made through a MATLAB developed code;
- iv The obtained results show that the matrices associated were badly conditioned ones and that the Tikhonov method together with the L-curve criterion were able to work around the associated problems;

- v A new proposed form for the shear modulus was presented and it was shown that the model associated with this new form presents better agreement with the experimental data.

It is important to mention that a similar approach can be made with respect to the viscosity coefficients and it is hoped that such treatment can be considered for future works.

ACKNOWLEDGEMENTS

This work was financially supported by CNPq.

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