Evaluation of viscoelastic parameters of surface layers by ultrasonic Love waves

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***Abstract*— Simultaneous determination of the rheological parameters of viscoelastic surface layers is very important in many applications such as: sensors, geophysics, seismology, and in the NDT of materials. Love wave energy is concentrated near the waveguide surface, so that Love waves are especially suited to study the material properties of surface layers. In this work, the Direct Sturm-Liouville Problem for the Love wave propagation in a layered viscoelastic waveguide have been presented and solved. Next, the Inverse Problem was created and solved as an Optimization Problem. The adequately formulated objective function that depends on the elastic and viscoelastic parameters of a waveguide of the Love wave and the experimental data was used. The solution of the Inverse Problem allows to determine unknown values of the viscosity and shear elasticity of a viscoelastic medium from measurements of the dispersion curves of Love waves.**

Keywords—Love waves; rheological properties; viscoelastic materials; inverse problems; viscosity; shear elasticity

I. INTRODUCTION

The objective of this work is to establish an Inverse Method for the simultaneous determination of the viscosity and shear elasticity of viscoelastic materials. Determination of the storage and loss modulus of viscoelastic media is very important in monitoring the technological parameters of e.g., viscoelastic polymers during the course of technological processes in the chemical industry. Classical mechanical methods for determining the rheological parameters of viscoelastic media are cumbersome and difficult to computerize. Thus, they cannot be applied on-line to control technological processes. To overcome the drawbacks of the mechanical methods, new ultrasonic methods that employ ultrasonic volume shear waves [1-3], and surface acoustic waves [4-9] were applied to investigate the rheological parameters of materials. Till present, rheological parameters of materials (viscosity and density) were measured simultaneously using ultrasonic two-element sensors [10,11] or special complex sensors that can be only used in laboratory [12,13]. Evaluation storage and loss modulus of viscoelastic surface sensing layers in bio and chemosensors is also of paramount importance in design and exploitation of these sensors [14]. The measured analyte influences directly the viscoelastic parameters of surface sensing layer. The determination of changes in the viscoelastic parameters of a sensing layer is a crucial problem in the theory, design and optimization of these sensors. This problem is also very important in geophysics [15], seismology [16], and in the NDT of materials.

Love waves are ideally suited to investigate the physical parameters of surface layers. The Kelvin-Voigt model of a viscoelastic layer was assumed ($\mu = \mu_B^0 - j\omega\eta$). In this study, to evaluate the viscoelastic parameters of an investigated surface layer, the following 3 steps were performed: 1) formulation and solution of the Direct Problem that describes propagation of Love waves in the layered structure: viscoelastic layer deposited on an elastic substrate, 2) experiment (numerical), and 3) formulation and solution of the Inverse Problem for Love waves propagating in this layered structure. This allows to determine the unknown viscoelastic parameters of the surface layers. This procedure employs the experimental dispersion curves of phase velocity and attenuation of Love waves (from 0.8 to 5 MHz) that propagate in the considered layered structure. The Inverse Problem was formulated and solved as a minimization problem. The objective function that depends on material parameters of the substrate, the frequency, experimental data (phase velocity and attenuation) and unknown viscoelastic parameters (μ_B^0, η) of the layer , has been constructed.

The results obtained are novel and original and can be of high relevance in industrial applications, for monitoring parameters of viscoelastic polymers.

II. DIRECT STURM-LIOUVILLE PROBLEM

A. *Dispersion relation*

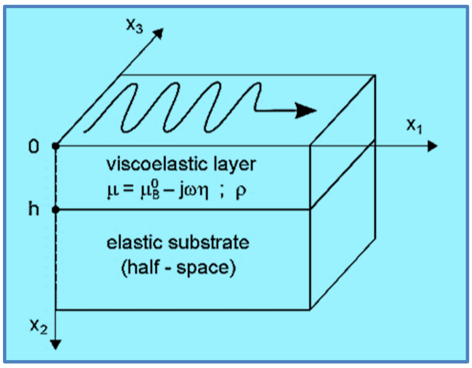

Fig.1. Structure of the surface Love wave layered waveguide. Viscoelastic layer is deposited over an elastic substrate.

The direct Sturm-Liouville problem relies on the determination of the dispersion curves and the mechanical displacement of the surface wave for given values of viscosity and elastic parameters of the surface layer and substrate, and frequency. Love wave propagates in a layered viscoelastic waveguide (a viscoelastic isotropic layer is rigidly attached to an isotropic and elastic substrate), see Fig.1.

Employing the continuity conditions of the mechanical displacement u_3 and shear stress τ_{23}, at the interfaces: 1) $x_2 = 0$ and 2) $x_2 = h$, we obtain the following analytical formula for the complex dispersion equation of the Love wave [17]:

$$\sin(q_B \cdot h) \cdot \mu_B \cdot q_B - \cos(q_B \cdot h) \cdot \mu_T \cdot b = 0 \qquad (1)$$

The complex wave number in Eq.1 is equal $k = k_0 + j\alpha$, where: $j = (-1)^{1/2}$.

The real part of the wave number k_0 describes the Love wave phase velocity. The imaginary part of the wave number α denotes an attenuation of the Love wave. In the dispersion equation (Eq.1), the quantities q_B, b and μ_B are complex:

$$q_B = \sqrt{\left(K_1^2 \frac{1}{(1+tan^2\delta)} - k_0^2 + \alpha^2\right) + j \cdot \left(K_1^2 \frac{tan\delta}{(1+tan^2\delta)} - 2 \cdot k_0 \cdot \alpha\right)} \quad (2)$$

$$b = \sqrt{(k_0^2 - \alpha^2 - k_2^2) + j \cdot 2 \cdot k_0 \cdot \alpha} \qquad (3)$$

$$\mu_B = \mu_B^0 - j\omega\eta \qquad (4)$$

where: $K_1 = \omega / v_1^0$; $k_2 = \frac{\omega}{v_2}$; $k_0 = \frac{\omega}{v_p}$; $tan\delta = \left(\frac{\omega\eta}{\mu_B^0}\right)$

After separating the real and imaginary parts of Eq. 1, we arrive at the following system of nonlinear algebraic equations with unknowns: k_0 and α.

$$C(\mu_B^0,\ \rho_B,\ \mu_T,\ \rho_T, \eta,\ h,\ \omega;\ k_0, \alpha) = 0 \qquad (5)$$

$$D(\mu_B^0,\ \rho_B,\ \mu_T,\ \rho_T, \eta,\ h,\ \omega;\ k_0, \alpha) = 0 \qquad (6)$$

Here: $\mu_B^0,\ \rho_B,\ \mu_T,\ \rho_T, \eta,\ h$ and angular frequency ω are parameters. The unknowns are: k_0 and α.

The system of nonlinear equations (5,6) was solved employing the numerical procedures from a computer package Scilab. Subsequently, the dispersion curves of Love waves are evaluated.

III. Rheological Model

In this study, to characterize the rheological properties of viscoelastic surface layer, the Kelvin-Voigt model has been chosen. Employing the constitutive equations for this model we arrive at the following formula for the complex shear elastic modulus:

$$\mu_B = \mu_B^0 - j\omega\eta \qquad (7)$$

where: μ_B^0 is the elastic shear modulus, and η is the viscosity of a viscoelastic medium.

IV. Inverse Sturm-Liouville Problem

The determination of the unknown rheological parameters from the measured dispersion curves of phase velocity and attenuation of Love waves propagating in a considered layered viscoelastic structure constitutes the Inverse Problem. In this work, the inverse problem was formulated and solved as an optimization problem with an appropriately defined objective function [18-20].

In order to solve the inverse problem the three steps were performed:

1. formulation and solution of the direct problem

2. carrying out numerical experiment

3. formulation and solution of the inverse procedure

a. inverse problem is formulated as an optimization problem

b. determination of an objective function $Q(\eta, \mu_B^0)$

c. application of optimization procedures

$$\min Q(\eta, \mu_B^0) \Rightarrow \left(\eta_{opt}, (\mu_B^0)_{opt}\right)$$

A. *Objective function Q*

The objective function $Q(\eta, \mu_B^0)$ (see Eq.8) is used in the inverse problem that relies on simultaneous determination of two unknown operational variables, i.e., the viscosity η and shear elasticity μ_B^0, of a viscoelastic medium.

$$Q(\eta, \mu_B^0) = \sum_{j=1}^{N_e} \left\{ \left(\frac{v_j^m - v_j^c(\eta, \mu_B^0)}{v_j^m}\right)^2 + \left(\frac{\propto_j^m - \propto_j^c(\eta, \mu_B^0)}{\propto_j^m}\right)^2 \right\} \qquad (8)$$

Here: N_e is the number of experimental points, v_j^c and α_j^c are the phase velocity and attenuation calculated from the direct problem that depend on both unknown values of the viscosity η and shear elasticity μ_B^0, known “a priori”: material parameters of an elastic waveguide, and experimental angular frequency ω_j. v_j^m and α_j^m are experimental quantities of the phase velocity and attenuation evaluated in Sect. V.

Using the optimization methods, minimum of the objective function $Q(\eta, \mu_B^0)$ was evaluated. It enabled the determination of the optimum values of the viscosity η and shear elasticity μ_B^0 simultaneously. These optimum values of the viscosity and shear elasticity constitute the solution of the Inverse Problem.

To minimize the appropriate objective function, optimization procedures of the Nelder-Mead type from Scilab computer package were applied.

V. Numerical Experiment

In numerical experiments we evaluated the dispersion curves numerically. At the beginning, by solving the direct problem (Eqs.5 and 6), we calculated the phase

TABLE I. Exact values of the phase velocity and attenuation at various frequencies for $\eta^{exact} = 0.37\ Pas$, $(\mu_B^0)^{exact} = 1.43 \times 10^9\ N/m^2$.

Frequency [MHz]	Phase velocity [m/s]	Attenuation [Np/m]
0.8	2005.64	4.82
1.0	1495.46	6.12
2.0	1171.42	19.70
3.0	1130.75	42.86
4.0	1117.42	75.33
5.0	1111.40	117.07

velocity and attenuation curves for the exact values of viscosity $\eta^{exact} = 0.37\ Pas$ and shear elasticity $(\mu_B^0)^{exact} = 1.43 \times 10^9\ N/m^2$, see TABLE I. Applied in the numerical calculations material parameters of the waveguide structure (PMMA on Quartz) are given in Section VI. Next, we added random errors (i.e., 1%, 2%, 5%, 10%) to these obtained values of the phase velocity and attenuation (columns 2 and 3 in TABLE I). Received in this way dispersion curves are regarded in the inverse procedure as simulated experimental curves.

VI. NUMERICAL RESULTS

Numerical calculations were performed for the following waveguide structure: PMMA – Poly(methyl methacrylate) surface layer attached to the quartz substrate (Fig.1). For PMMA: $\mu_B^0 = 1.43 \times 10^9\ N/m^2$, and $\rho_B = 1.18 \times 10^3\ kg/m^3$. For quartz: $\mu_T = 5.4 \times 10^{10}\ N/m^2$, and $\rho_T = 2.2 \times 10^3\ kg/m^3$. The thickness h of the surface layer is 0.4 mm. The viscosity and shear elasticity of the viscoelastic layer were assumed as $\eta^{exact} = 0.37\ Pas$; $(\mu_B^0)^{exact} = 1.43 \times 10^9\ N/m^2$ respecitively. Losses in the quartz substrate are omitted. The origin of losses is the viscosity of the viscoelastic surface layer.

We formulated and solved the inverse problem to evaluate simultaneously two unknown variables, i.e., the viscosity η, and shear elasticity μ_B^0. The Inverse Problem was solved for several values of added (to exact values of the phase velocity and attenuation) random errors in the range 1% to 10%, see TABLE I.

The relative error RE1 for a series of N measurements of the viscosity of a viscoelastic medium was defined as follows:

$$RE1 = \left\{\left(\frac{|\eta_1^{calc} - \eta^{exact}|}{|\eta^{exact}|} + \frac{|\eta_2^{calc} - \eta^{exact}|}{|\eta^{exact}|} + \cdots + \frac{|\eta_{N_e}^{calc} - \eta^{exact}|}{|\eta^{exact}|}\right)\right\}/N \quad (9)$$

An analogous formula is valid also for the relative error RE2 for a series of N measurements of the shear modulus μ_B^0 of a viscoelastic material.

$$RE2 = \left\{\left(\frac{\left|(\mu_B^0)_1^{calc} - (\mu_B^0)^{exact}\right|}{\left|(\mu_B^0)^{exact}\right|} + \frac{\left|(\mu_B^0)_2^{calc} - (\mu_B^0)^{exact}\right|}{\left|(\mu_B^0)^{exact}\right|} + \cdots + \frac{\left|(\mu_B^0)_{N_e}^{calc} - \eta(\mu_B^0)^{exact}\right|}{\left|(\mu_B^0)^{exact}\right|}\right)\right\}/N$$

(10)

The results of numerical calculations of the Inverse Method for determining the unknown values of the rheological parameters of an investigated viscoelastic body are inserted in TABLE II.

TABLE II. Relative error of the simultaneous evaluation of the viscosity and shear elasticity of the viscoelastic material determined from the inverse method employing objective function $Q(\eta, \mu_B^0)$, for the maximum random errors equal 1%, 2%, 5%, and 10%. Each evaluation of (η, μ_B^0) results from the minimization of the objective function $Q(\eta, \mu_B^0)$, (Eq.4), for consecutive simulated dispersion curves of phase velocity and attenuation.

Max Random error	1%	2%	5%	10%
Relative error (η) Eq.9 [%]	0.40	0.81	2.21	6.63
Root Mean Square Error(η) [%]	0.51	1.06	2.54	7.53
Relative error (μ_B^0) Eq.10 [%]	0.27	0.47	1.21	3.72
Root Mean Square Error (μ_B^0) [%]	0.30	0.57	1.49	4.56

VII. CONCLUSIONS

In this work, the Sturm-Liouville Direct Problem for the Love wave propagating in a layered waveguide with a viscoelastic layer has been formulated and solved. Then, the Inverse Problem for the ultrasonic Love wave propagating in the considered waveguide structure was also formulated and solved. The Inverse Problem was formulated as an Optimization Problem. To this end the appropriate objective function has been created. Minima of this objective function were evaluated by using the numerical optimization procedures (Nelder-Mead - nonlinear simplex). These minima specify simultaneously sought values of the viscosity η and shear elasticity μ_B^0. These values of the viscosity and shear elasticity form the solution of the Inverse Problem.

Objective function contains experimental data (v_j^m and α_j^m) and values of phase velocity and attenuation of Love waves (v_j^c and α_j^c) evaluated by solving the direct problem. Calculated values (v_j^c and α_j^c) are functions of unknown viscosity η and shear elasticity μ_B^0 of a viscoelastic material respectively. These values (v_j^c and α_j^c) depend also on the material parameters of the waveguide, that are known "a priori". Objective function is a measure of the distance between experimental data and values obtained from the theoretical model. Minimization of this distance enables to determine simultaneously the unknown values of the viscosity and shear elasticity of the surface layer.

The solution of the Inverse Problem allows to determine simultaneously with good accuracy (of the order of 5%), both the shear elasticity and viscosity of a viscoelastic material. The accuracy of the obtained results can be ameliorated by averaging, i.e., by solving the Inverse Problem for a series of simulated experimental dispersion curves, and subsequently, by calculating the mean value of the obtained values of the viscosity η and shear elasticity μ_B^0.

Simultaneous evaluation of the viscosity and shear elasticity of viscoelastic materials is very important in the monitoring of technological processes in many branches of industry, e.g., chemical, food, plastics, and polymer industries.

The results obtained in this study are original and fundamental and can provide useful data for the design and fabrication of Love-wave based miniaturized liquid sensing devices, such as biosensors and chemosensors. Results of the study can also be applied in seismology and geophysics (investigations of earthquakes).

According to the best authors knowledge, formulation and solution of the Inverse Problem for the ultrasonic Love wave propagation in a layered viscoelastic waveguide and subsequently, simultaneous determination of the viscosity η and shear modulus μ_B^0 of viscoelastic materials is a novelty.

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