Probing flexible thermoplastic thin films on a substrate using ultrasonic waves to retrieve mechanical moduli and density: Inverse problem

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Abstract. Flexible, supple thermoplastic thin films (PVB and PET) placed on elastic substrates were probed using ultrasonic waves to identify their mechanical moduli and density. The composite medium immersed in a fluid host medium (water) was excited using a 50 MHz transducer operating at normal incidence in reflection mode. Elastic wave propagation data from the stratified medium was captured in the host medium as scattered field. These data were used along with theoretical fluid-solid interaction forward models for stratified-media developed using elasticity theory, to solve an inverse problem for the recovery of the model parameters of the thin films. Two configurations were modeled, one considering the substrate as a semi-infinite elastic medium and the second the substrate having a finite thickness and flanked by a semi-infinite host medium. Transverse slip for the sliding interface between the films and substrate was chosen. This was found to agree with the experiments whereby the thin films were just placed on the substrate without bonding. The inverse problems for the recovery of the mechanical parameters were successful in retrieving the thin films’ parameters under the slip boundary condition. The possible improvements to the new method for the characterization of thin films are discussed.

1. Introduction

This study reports the development of new methods for the characterization of two types of thin flexible films made from thermoplastic polymers by solving an inverse problem using real ultrasound reflection data and theoretical interaction models. The aim of this study is to:

(i) develop new methods/techniques and corresponding geometric configuration for the characterization of flexible, supple thermoplastic thin films (PVB and PET),
(ii) Identify their mechanical moduli (Youngs modulus, Poisson coefficient) and density.

The knowledge of mechanical properties is necessary to understand mechanical behavior and improve the design of thermoplastic thin films.

Thin flexible thermoplastic polymer resin films are employed in a variety of applications

• Thermoplastic thin made from polyEthylene terephthalate (PET) [1] are employed for the reinforcement of glass windscreens.
Polyvinyl butyral (PVB) is used as protective interlayer films that help hold shattered glass windows together during severe weather conditions, accidental impact or vandalism.

The popular methods for the characterization of thin films are based on scanning acoustic microscopy [2]. These methods have mainly allowed access to the measured velocity of sound waves propagating in the thin films and their V(z) curves [3] at a frequency range from 170-450 MHz. Lavrentyev et al [4] proposed a method for determining the properties of a coating on a thin plate. The method allows simultaneous determination of the coating thickness, density, elastic moduli and attenuation from both the normal and oblique incidences reflection frequency spectra. Phase-shift shadow Moiré optical methods have also been employed to recover Young’s moduli of composite systems composed of various epoxy coated polyethylene terephthalate (PET) micro-cantilevers [5].

2. Materials and methods

2.1. Geometry of the experimental configuration and probing frequency

The films to be characterized were placed on elastic substrates and were then probed using ultrasonic waves to identify their mechanical moduli and density.

The geometry of the acoustic wave reflection configuration by the layered media composed of the thin film (Ω₁) placed on an elastic substrate (Ω₂) and immersed in water (Ω₀) are depicted on Fig. (1). The mode conversions are also represented on the figure. The geometry is represented with an oblique angle of incidence (θ ≠ 0) for the acoustic wave impinging on the film. The measurements were done at normal incidence (θ = 0) only. The theoretical models herein were developed using elasticity theory (starting from Hooke’s law up to the derivation of the elastic wave propagation equations in the elastic solid as in reference [6]). The general case of an oblique angle of incidence θ was chosen for the model.

![Figure 1. The geometry of the reflection configuration of ultrasonic waves impinging on a thin film (domain Ω₁) placed on an elastic substrate (Ω₂). The semi-infinite fluid media (water) domain is depicted by Ω₀. The boundary between the fluid and the film is Γ₁, that with the substrate Γ₂ and that between the substrate and the fluid Ω₃, is Γ₃. The mode conversions of the incident wave from P-wave into P-wave and shear waves in the thin film (Ω₁) are not shown but are indicated in the substrate.](image)

The probing frequency is imposed by the film thickness and the number of desired modes propagating in the film, \( f = n v_L / 2 L \) (n is the mode number of longitudinal modes, \( v_L \) is the wave velocity, \( L \) is the thickness of the film).
2.2. The experimental film-substrate configurations studied

Two configurations of the substrate were studied, in which

(i) the film was placed on a substrate with the latter considered as a semi-infinite half space i.e no reflected wave from the second interface \( F_3 \) of the elastic medium substrate,

(ii) the substrate is of finite thickness (infinite elastic plate) with the composite structure (film on the substrate) bounded on both sides by an infinite fluid medium. A pressure wave is transmitted at the second interface of the substrate \( F_3 \).

The film and the substrate were not glued together. The film was allowed to slide on the substrate.

3. Modeling the configurations

3.1. The domain equations - frequency domain

The plane wave harmonic solutions of the wave equations for each elastic domains are written using potentials. The acoustic pressure wave in the fluid domain is,

\[ p_f(x, z) = (e^{\kappa z} + Re^{-\kappa z}) e^{\kappa f x} p_i \quad z \in \Omega_f, \]

where \( R \) is the reflection coefficient, \( p_i \) is the amplitude of the incident wave in the fluid, the complex wavenumber in the fluid \( \kappa z_f = i k_f \cos(\theta_i) \) \( (k_f = \omega/c_f \) and \( c_f \) is the wave velocity in the fluid) and the subscript \( f \) stand for fluid.

In the film layer, the potentials of elastic waves have the following form (for the incident and reflected waves),

\[ \Psi_{s1}(x, z) = (\Phi_{1f} e^{\kappa z_{1s} z} + \Phi_{12} e^{-\kappa z_{1s} z}) e^{\kappa f x} \quad z \in \Omega_1, \]

\[ = (A_{1f} \cosh (\kappa z_{1f} z) + A_{12} \sinh (\kappa z_{1s} z)) e^{\kappa f x} \quad z \in \Omega_1, \]

where the compressional wave potential amplitudes \( A_{1f} = \Phi_{11} + \Phi_{12}, A_{12} = \Phi_{11} - \Phi_{12} \) and \( A_{11} \) and \( A_{12} \) (after conversion of the exponential expressions into trigonometrical functions). The superscript \( s \) stands for solid.

\[ \Phi_{s1}(x, z) = (B_{1f} \cosh (\kappa z_{s1} z) + B_{12} \sinh (\kappa z_{s1} z)) e^{\kappa f x} \quad z \in \Omega_1, \]

\[ \Psi_{s2}(x, z) = \Phi_{21} e^{\kappa z_{s2} (z+L)} e^{\kappa f x} \quad z \in \Omega_2, \]

\[ \Psi_{s2}(x, z) = \Psi_{21} e^{\kappa z_{s2} (z+L)} e^{\kappa f x} \quad z \in \Omega_2, \]

where the amplitudes of the shear waves are \( B_{1f} = \Psi_{11} + \Psi_{12} \) and \( B_{12} = \Psi_{11} - \Psi_{12} \).

The amplitudes \( \Phi_{11} \) and \( \Psi_{11} \) are for the incident P-wave and shear-wave in the thin film respectively, while \( \Phi_{12} \) and \( \Psi_{12} \) are amplitudes of the reflected P-wave and shear-wave respectively and \( L \) is the thickness of the film, \( \Phi_{21} \) and \( \Psi_{21} \) are the amplitudes of the incident P-wave and shear-wave in the substrate respectively.

3.2. The complex wavenumbers

The complex wave-numbers \( \kappa f = i \gamma_f \) \( (i = \sqrt{-1}), \) \( \gamma_f = k_f \sin(\theta) \) and the compressional complex wave numbers \( \kappa z_{1n} \) and the shear ones \( \kappa z_{3n} \) \( (n = 1, 2 \) represents the medium \( \Omega_n) \) are given by

\[ \kappa z_{11} = i \sqrt{k_{11}^2 - \gamma_f^2}, \quad \kappa z_{12} = i \sqrt{k_{12}^2 - \gamma_f^2}, \]

\[ \kappa z_{21} = i \sqrt{k_{31}^2 - \gamma_f^2}, \quad \kappa z_{32} = i \sqrt{k_{32}^2 - \gamma_f^2}. \]
3.2.1. The relationship between wave numbers with the elastic constants

The wave numbers are given by

\[ k_{11} = \omega \sqrt{\frac{\rho_1 M_1}{\mu_1}}, \quad k_{31} = \omega \sqrt{\frac{\rho_1 \mu_1}{M_1}} \]

\[ k_{12} = \omega \sqrt{\frac{\rho_2 M_2}{\mu_2}}, \quad k_{32} = \omega \sqrt{\frac{\rho_2 \mu_2}{M_2}} \]  

(5)

where \( \omega \), angular frequency of the wave, \( \rho_n \), \( \mu_n \) and \( M_n \) are the density, shear modulus and the P-wave modulus of the layer respectively. The inherent damping of the films were taken into account by introducing a scalar-valued loss factor \( \eta_n \) into Young’s modulus, such that \( E_s^n = E_s^n(1 + i\eta_n) \) (\( E_s^n \) is the dynamic Young’s modulus of the layer). The relationships between \( M_n \) and \( E_s^n \) are given in [6].

3.3. Thin flexible film resting on a substrate and both immersed in a fluid - the boundary conditions (BC)

3.3.1. BC between the thin film and the fluid

The thin film is bounded on one side by semi-infinite fluid half space (\( \Omega_0 \)) while the other side is bounded by a semi-infinite elastic half space.

The boundary conditions at the interface \( \Gamma_1 \) between the thin film and the fluid are: continuity of the velocity and the normal stress while the tangential stress \( (\sigma_{xz}) \) is zero across the interface at \( z \in \Gamma_1 \)

\[ v_f(x, 0, s) = v_s^1(x, 0, s), \quad p_f(x, 0, \omega) = \sigma_{zz}^s(x, 0, s), \quad \sigma_{xz}^s(x, 0, s) = 0, \]  

(6)

where the superscript and subscript \( s \) and \( f \) stand for solid and fluid respectively.

3.3.2. BC between the thin film and the substrate - Sliding boundary conditions

The thin film was placed on the substrate without gluing. The water between them was squeezed out by sliding the edge of a flat ruler over the film. The measurement was taken immediately before water infiltrated between the film and substrate interface (a slow process). The boundary conditions between the thin film and the elastic substrate at the interface \( \Gamma_2 \) was chosen to be that of transverse slip (sliding can occur). Overall, there is: continuity of the normal velocity, the normal stress, while the tangential stress \( (\sigma_{xz}) \) is zero across the interface at \( z \in \Gamma_2 \)

\[ v_z^1(x, -L, s) = v_z^2(x, -L, s), \quad \sigma_{zz}^1(x, z, s) = \sigma_{zz}^2(x, -L, s), \]

\[ \sigma_{xz}^1(x, -L, s) = \sigma_{xz}^2(x, -L, s) = 0. \]

4. The direct and inverse problem for the retrieval of mechanical parameters of thin films

4.1. The direct problem

The relationships employed for the displacement \( (u) \), velocity \( (v) \) with the wave potentials in the Laplace domain are

\[ u_x^{sn} = \frac{\partial}{\partial x} \Psi^{sn}(x, z) - s \frac{\partial}{\partial z} \Phi^{sn}(x, z), \quad n = 1, 2, \]

\[ u_z^{sn} = s \frac{\partial}{\partial x} \Psi^{sn}(x, z) - \frac{\partial}{\partial z} \Phi^{sn}(x, z), \]

\[ u_x^{sn} = \frac{\partial}{\partial z} \Psi^{sn}(x, z) + \frac{\partial}{\partial x} \Phi^{sn}(x, z), \]

\[ u_z^{sn} = s \frac{\partial}{\partial z} \Psi^{sn}(x, z) + s \frac{\partial}{\partial x} \Phi^{sn}(x, z) \]  

(7)
4.1.1. The mechanical stresses

The stresses in the elastic solid medium are given by

\[ \sigma_{zz}^{sn} = \lambda_n \left( \frac{\partial}{\partial x} u_z^{zn} (x, z) + \frac{\partial}{\partial z} u_z^{zn} (x, z) \right) + 2 \mu_n \frac{\partial}{\partial z} u_z^{zn} (x, z) \, , \quad n = 1, 2, \]

\[ \sigma_{zz}^{sn} = \mu_n \left( \frac{\partial}{\partial x} u_z^{zn} (x, z) + \frac{\partial}{\partial z} u_z^{zn} (x, z) \right) , \tag{8} \]

4.2. The matrix equation of the stratified media - film-substrate interface are sliding

The matrix equation was obtained by combining the domain potential equations, the mechanical stress relationships and the boundary equations when the substrate is a semi-infinite half-space then extracted from the solution of this equation. This constitutes the interaction model (IM).

4.3. The inverse problem - construction of the cost functions

The viscoelastic interaction model of the acoustic wave reflection \( R^{IM} \) of the setup geometry is obtained from the matrix equation developed in the previous subsection (viscoelastic by the introduction of a complex Young’s modulus [6]). An error functional expressing the distance between the experimental data reflection coefficient from that of the interaction model was computed for each set of trial values of the parameters.

\[ \mathfrak{J}(E_1, \nu_1, \rho_1, E_2, \nu_2, \rho_2) = \sum_{n=1}^{N_s} \left( |R^{IM}(\omega_n, E_1, \nu_1, \rho_1, E_2, \nu_2, \rho_2)| - |R^{\text{exp}}_{IR}(\omega_n)| \right)^2 , \tag{10} \]

where \( |R^{\text{exp}}_{IR}| \) is the amplitude of the reflection coefficient of the thin film on its substrate obtained from the tank experimental data and \( |R^{IM}| \) is the amplitude of the interaction model reflection coefficient. The method of signal processing for obtaining the reflection coefficient from the measured incident and reflected signals from the thin film by computing the transfer function, are explained in detail in reference [7]. The parameters were retrieved by minimizing the objective functional \( \mathfrak{J} \). The substrate parameters \( E_2, \nu_2, \rho_2 \) were supposed known.

The problem was split into the computation of three cost functions \( \mathfrak{J}_E, \mathfrak{J}_\nu, \mathfrak{J}_\rho \) where each of the model parameters \( (E_{1s_m}, \nu_{1s_m}, \rho_{1s_m}) \), where \( s \) is the step, \( m = 1 \ldots p \) and \( p \) is the number of intervals) was varied while the other two were fixed. This was done in various intervals not too wide, e.g. Poisson ratio interval was chosen from 0.1 to 0.5, density from 800 to 2000, etc. Three initial values of the sought-for film parameters \( (E_{1s_0}, \nu_{1s_0}, \rho_{1s_0}) \) were chosen at the initial step. Some of these values, found in the literature (e.g. for PET [8, 9], for both PET and PVB [10])
were obtained using different methods. The solution for each of the parameters was found from the position of the global minimum of the cost function (CF) exhibiting a parabolic shape. The values corresponding to the three CF minima constituted the solution to the inverse problem for the step. The three new minima values were then used to update the initial values for the next step. The search for the optimized values was a manual iterative process repeated for each parameter at a time, at each iteration/step, until convergence was achieved for all the three sought for parameters. The objective functional curves for each parameter were then reported after assuring that all the three parameters had converged to their optimal values. The curves were plotted with respect to one of the three problem variables, the others being fixed at their computationally-optimal values, i.e., as 2-D plots [11].

5. Results
The captured temporal incident and reflected signals and their calculated spectra are presented in Fig. (2). A correction was undertaken by shifting the signals so as to assure artificially that the transducer-film and transducer-reflector distances were the same. The opposite face of the substrate was used as mirror to obtain the incident signal. The spectrum of the reflected (incident) signal shows the useful bandwidth.

![Figure 2](image-url)

Figure 2. (a) The reflected temporal signals, lower panel, the shifted signals (b) Their frequency spectra for the 750 µm thick PVB film.

5.1. The substrate is a semi-infinite half-space elastic medium
5.1.1. Retrieval of mechanical parameters - cost functions The cost functions are computed using relation in Eqn. (10). They are depicted in Fig. 3.

Measurements of the reflections coefficients were done for a 100µm PET film placed on substrates made of different elastic materials and geometries (glass plate, aluminium slab, aluminium cylinder). This was to analyze the influence of different elastic materials on the reflection coefficient.

5.2. Summary of the results
The results are summarized in Table (1). The densities were compared with those obtained from small specimens of the film whose weights were measured on an analytic electronic balance (Fisher scientific model PAS214C, Pittsburgh, PA USA). The velocities were measured using the time of flight and then compared with those computed using the recovered parameters. The
Figure 3. The cost functions for the recovery of the three mechanical parameters for a 450 µm PVB film (a) the density $\rho$ (kg/m$^3$), (b) Young’s modulus, (c) Poisson ratio. (d) The comparison theoretical/experimental reflection coefficients using the recovered parameters.

Figure 4. The comparison between the model (solid line) and the measured reflection coefficients for the 100µm PET film resting on the glass plate substrate (dotted line), aluminium slab substrate (long-dash) and aluminium cylinder (dash-dot), with the interface is considered as sliding.

densities were recovered from the electronic balance measurements results were within 3 percent difference with the ones recovered by solving the inverse problems.

The apparent dispersion of properties for different thicknesses of the PVB in Table 1 stems from the structural differences with the thickness of the film as shown in Fig. (5) of the Scanning electron microscopy (SEM) images of the PVB films of different thicknesses. X-ray diffraction (XRD) and reflectivity methods were also employed to reveal the density as an absolute value and also the surface and interface roughness of the PVBs but whereas these results are yet to be exploited, they showed the same dispersion in the density with the thickness
Table 1. Summary of the identified mechanical parameters with the measured velocity recovered from the time of flight of the longitudinal wave in the film layer.

| Thickness (µm) | Velocity m/s measured | Young’s modulus C_L1 GPa | Poisson ratio | Loss η_L s | Density (kg/m³) |
|----------------|-----------------------|---------------------------|---------------|-----------|----------------|
| 750            | 1904                  | 1850                      | 0.8           | 0.455     | 0.085          | 980            |
| PVB 450        | 1991                  | 1979                      | 0.77          | 0.465     | 0.095          | 1015           |
| 380            | 1603                  | 1581                      | 0.8           | 0.45      | 0.08           | 1217           |
| PET 100        | 1727                  | 1788                      | 1.175         | 0.456     | 0.025          | 1560           |

variation as those shown in the table.

In order to validate the values using the same specimens, the mechanical behaviour of the PVB films will be investigated in future using the small-strain behaviour through dynamic mechanical analysis (DMA) at frequencies from 1 to 100 Hz.

![Scanning electron microscopy images of the PVB films of thickness (a) 450 µm (b) 750 µm](image)

**Figure 5.** Scanning electron microscopy images of the PVB films of thickness (a) 450 µm (b) 750 µm

6. **Improved modeling - substrate of finite thickness**

In this case the substrate is of finite thickness (L2) and the stratified media is considered as bounded on both sides by the fluid. The procedure is the same as that of the substrate considered as a semi-infinite half-space. The changes in the domain potentials are only for the substrate and the presence of a transmitted pressure wave in the fluid (z ≤ -(L + L2)):

\[
\Psi^{s2}(x, z) = (A_{21} \cosh(\kappa z_{12} (z + L)) + A_{22} \cosh(\kappa z_{12} (z + L))) e^{\kappa f x} \quad z \in \Omega_2,
\]

\[
\Phi^{s2}(x, z) = (B_{21} \cosh(\kappa z_{22} (z + L)) + B_{21} \cosh(\kappa z_{22} (z + L))) e^{\kappa f x} \quad z \in \Omega_2,
\]

\[
p_{T}^{f}(x, z) = e^{(\kappa z_{11}L + \kappa z_{12}L_2) + \kappa z_{3}(L + L_2 + z)} T e^{\kappa f x} p_1 \quad z \leq -(L + L_2),
\]

where L2 is the thickness of the substrate, T is the transmission coefficient of the stratified medium. A matrix equation is obtained as previously and the reflection coefficient computed.
Figure 6. The comparison between the theoretical model (continuous line) and the measured (dash) reflection coefficients of thin PVB film placed on the glass substrate with the finite thickness and bordered by a semi-infinite fluid half space (a) 750µm (b) 450µm (c) 380µm.

When the measurement window captures the temporal reflected waves from the face at \( z = -(L + L_2) \) the reflection coefficient is perturbed as the computed ones in Fig. (6) but the modes do not change.

7. Conclusion
A method for the recovery of the mechanical parameters of flexible thermoplastic thin films (PETs and PVBs) on elastic substrates using reflected ultrasonic acoustic waves and interaction models has been developed. Two models were developed in this study, one in which the substrate was considered as a semi-infinite half-space and in the second one it had a finite thickness. In both configurations the sliding boundary contact condition between the thin film and substrate were considered as sliding. The stratified medium was immersed in a water tank where it was bounded on both sides by semi-infinite half spaces. The results of the recovered parameters agree well with those in the literature.

References
[1] Toufalli F A E 2006 Catalytic and Mechanistic Studies of Polyethylene Terephthalate Synthesis Ph.D. thesis Technische Universität Berlin URL https://doi.org/10.14279/depositonce-1333
[2] Briggs A and Kolosov O 2009 Acoustic Microscopy (Oxford University Press) ISBN ISBN-13:9780199232734 URL http://dx.doi.org/10.1093/acprof:oso/9780199232734.001.0001
[3] Maebayashi M, Matsuoka T, Koda S, Hashitani R, Nishio T and ichi Kimura S 2004 Polymer 45 7563 – 7569 ISSN 0032-3861 URL http://www.sciencedirect.com/science/article/pii/S0032386104008705
[4] Lavrentyev A I and Rokhlin S I 2001 Ultrasonics 39 211 – 221 ISSN 0041-624X URL http://www.sciencedirect.com/science/article/pii/S0041624X00000664
[5] Lim J, Ratnam M, Azid I and Mutharasu D 2011 Optics and Lasers in Engineering 49 1301 – 1308 ISSN 0143-8166 URL http://www.sciencedirect.com/science/article/pii/S0143816611001710
[6] Ogam E, Fellah Z and Ogam G 2016 *Composite Structures* **135** 205 – 216 ISSN 0263-8223 URL http://www.sciencedirect.com/science/article/pii/S026382231500865X

[7] Ben Mansour M, Ogam E, Fellah Z E A, Soukaina Cherif A, Jelidi A and Ben Jabrallah S 2016 *J. Acoust. Soc. Am.* **139** 2551–2560 URL http://scitation.aip.org/content/asa/journal/jasa/139/5/10.1121/1.4948573

[8] Li T C, Wu B H and Lin J F 2011 *Thin Solid Films* **519** 7875 – 7882 ISSN 0040-6090 URL http://www.sciencedirect.com/science/article/pii/S0040609011005682

[9] Bistričić L, Borjanović V, Leskovac M, Mikac L, McGuire G E, Shenderova O and Nunn N 2015 *Journal of Polymer Research* **22** 39 ISSN 1572-8935 URL http://dx.doi.org/10.1007/s10965-015-0680-z

[10] Shim G I, Kim S H, Ahn D L, Park J K, Jin D H, Chung D T and Choi S Y 2016 *Composites Part B: Engineering* **97** 150 – 161 ISSN 1359-8368 URL http://www.sciencedirect.com/science/article/pii/S1359836816305571

[11] Ogam E, Fellah Z, Sebaa N and Groby J P 2011 *J. Sound Vibr.* **330** 1074 – 1090