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# 1 Identification of viscoelastic material properties by ultrasonic angular 2 measurements in double through-transmission

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## 6 Abstract

7 Recent advances in additive manufacturing of viscoelastic materials have paved the way towards the de-  
8 sign of increasingly complex structures. In particular, emerging biomedical applications in acoustics involve  
9 structures with periodic micro-architectures, which require a precise knowledge of both longitudinal and  
10 transverse bulk properties of the constituent materials. However, the identification of the transverse prop-  
11 erties of highly soft and attenuating materials remains particularly challenging. Thereby, the present work  
12 provides a methodological framework to identify the frequency-dependent ultrasound characteristics (*i.e.*,  
13 phase velocity and attenuation) of viscoelastic materials. The proposed approach relies on an inverse proce-  
14 dure based on angular measurements achieved in double through-transmission, referred as  $\theta$ -scan. Towards  
15 this goal, a forward modeling of the double transmitted waves through a homogeneous solid is proposed  
16 for any incidence angle based on the global matrix formalism. The experimental validation is conducted  
17 by performing ultrasound measurements on two types of photopolymers commonly employed for additive  
18 manufacturing purposes: a soft elastomer (Elastico™Black) and a glassy polymer (VeroUltra™White). As  
19 a result, the inferred dispersive ultrasound characteristics are of interest for the computational calibration  
20 and validation of models involving complex multi-material structures in the MHz regime.

*Keywords:* viscoelasticity, transverse properties, photopolymer material, global matrix formalism, inverse  
problem.

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## 21 1. Introduction

22 Additive manufacturing (AM), also referred to as 3D printing, has increasingly expanded over the past  
23 decade in engineering and sciences. The development of accurate and versatile 3D printing technologies has  
24 paved the way to the design of customized and complex structures printed with a large choice of materials,  
25 ranging from elastomers and polymers to ceramics or metals [1, 2]. Among these, multi-material jetting, also  
26 known as inkjet technology, represents a powerful multi-material 3D printing process, which allows tailoring

27 the material composition from the micro to the macro scale. For instance, this process has not only enabled  
28 the fabrication of heterogeneous materials with dissimilar properties [3], such as periodic lattices or porous  
29 media [4], but also interpenetrating phase composites [5] or functionally graded materials [6, 7].

30 In this context, multi-material photopolymer-based AM has experienced a significant interest in acoustics  
31 to design and fabricate tailored tissue-mimicking phantoms [8] for calibrating ultrasound imaging systems [9,  
32 10] or for therapeutic ultrasound applications [11]. Interestingly, the achievement of controlled bio-inspired  
33 structures involving periodic micro-architectures has also experienced recent advances to design scaffolds  
34 for bone tissue repair [12]. As the prediction of the acoustic behavior of such structures is particularly  
35 challenging, recent modeling efforts have focused on disentangling the relative contributions of viscoelasticity  
36 and periodicity on ultrasonic signatures, such as dispersion, attenuation, and bandgaps localization [13, 14].  
37 In this respect, an accurate identification of both longitudinal and transverse ultrasound characteristics (*i.e.*,  
38 phase velocity and attenuation) of the constituent phases is typically required, together with their frequency  
39 dependence.

40 Notwithstanding, while several ultrasound characterization methods have been explored to measure  
41 longitudinal bulk properties of photopolymer materials, including broadband reflection and transmission  
42 techniques [9, 15, 10] only a limited number of works have been carried out to report on their transverse  
43 properties [16]. Interestingly, the latter study, which relies on the double through-transmission technique,  
44 exploits mode conversion at the liquid-solid interface in oblique incidence in order to measure the transverse  
45 properties of the wave propagating inside the material. This approach has been initially developed to measure  
46 the phase velocity of anisotropic elastic materials [17, 18] and was then extended to the measurement of the  
47 attenuation of viscoelastic materials [19]. Although this technique has the advantage to provide longitudinal  
48 and transverse bulk properties on a broad frequency range by windowing four measured echoes only (*i.e.*,  
49 front-face and back-face reflections at normal incidence, double through-transmission at normal and oblique  
50 incidences), it suffers from several drawbacks. First, the frequency-thickness product of the sample must be  
51 optimized to ensure a proper temporal separation of the echoes. Second, the identification of the transverse  
52 bulk properties relies on a measurement performed after the critical angle, above which only transverse waves  
53 propagate through the sample. In the case of soft and highly attenuating materials, the critical angle is high,  
54 and so the propagation distance is large, as a consequence of which the transverse wave retrieved in double  
55 through-transmission is typically drown in noise. Altogether, these limitations prevent straightforwardly  
56 applying the approach described in Gattin et al. [16] to highly viscoelastic materials.

57 Alternatively, another approach to retrieve the bulk properties of viscoelastic samples relies on the analy-

58 sis of the reflection and transmission coefficients obtained for different incident angles in simple transmission  
59 [20]. An optimization process, performed on the measurement of these coefficients and their modeled coun-  
60 terparts [21, 22], then allows retrieving apparent longitudinal and transverse properties. This approach has  
61 been widely applied to anisotropic and viscoelastic materials [23, 24] and has also been recently extended  
62 to evaluate the anisotropy of adhesive bonds [25]. Unlike for the *classical* double through-transmission  
63 technique, no temporal separation of the echoes is needed here. However, the application of this approach  
64 to highly viscoelastic materials is not straightforward since the previous studies have not considered dis-  
65 persive properties. Moreover, only the amplitude of the transmission and reflection coefficients was used in  
66 the optimization process, thereby neglecting their phases, which may also carry information on the bulk  
67 properties.

68 Here, we propose to leverage the methodology initially introduced by Castaings et al. [20] on composites,  
69 together with the advantages of the double through-transmission technique [16], to estimate the ultrasound  
70 characteristics of photopolymer materials. In particular, the purpose of this work is to provide a methodolog-  
71 ical framework towards the identification of the frequency-dependent bulk properties of highly viscoelastic  
72 materials, without the need to isolate the transverse waves after the critical angle. The originality of this  
73 work not only lies in the estimation of the longitudinal and transverse phase velocities and attenuations  
74 of photopolymer materials, but also of their frequency-dependence, as well as the thickness of the sample,  
75 by considering the complex transmission transfer function in the optimization process. To validate our  
76 approach, ultrasound measurements were performed in double through-transmission on viscoelastic sam-  
77 ples manufactured using a multi-material 3D printing technology. To this end, several combinations of two  
78 dissimilar photopolymers –a glassy polymer and a soft elastomer– were investigated.

79 The remainder of the paper is organized as follows: Sec. 2 describes the additively manufactured samples  
80 along with the experimental setup used to achieve the angular measurements in double through-transmission.  
81 Then, Sec. 3 states the principle of the double through-transmission modeling at normal and oblique in-  
82 cidence according to the global matrix formalism, considering both the wave amplitude and phase. The  
83 optimization strategy for material properties identification is detailed in Sec. 4. Eventually, the ultrasound  
84 characteristics of the photopolymer materials and, in particular, the influence of the volume fraction of each  
85 of the two studied materials are reported and discussed in Sec. 5.

86 **2. Experimental method**

87 The proposed ultrasound characterization method relies on the measurement of double through-transmitted  
 88 signals across the samples at various incident angles, as schematically depicted in Fig. 1. This angular mea-  
 89 surement will be referred as *angular scan* in what follows, and its time representation will be named  $\theta$ -scan.

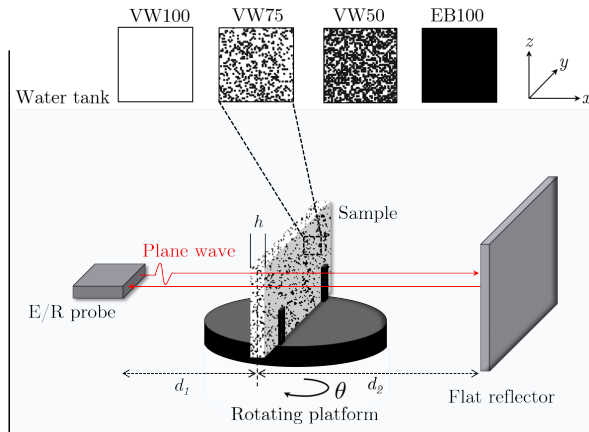


Figure 1: Schematic representation of the experimental setup used for measuring double through-transmitted signals at various incident angles, *i.e.*,  $0^\circ \leq \theta \leq 64^\circ$ , on photopolymer samples made of VeroUltra™White (VW) and Elastico™Black (EB).

90

91 **2.1. Additively manufactured samples**

92 Multi-material photopolymer samples were achieved using a commercial 3D printer (Polyjet Pro J35,  
 93 Stratasys, Los Angeles, CA). This additive manufacturing technology consists in projecting multiple drops of  
 94 UV-curable photopolymer on a building tray through printing nozzles according to a layer-by-layer process  
 95 [26]. At the microscopic scale (*i.e.*, voxel resolution around  $18 \mu\text{m}$ ), this inkjet technology allows for the  
 96 mixing of photopolymers with highly dissimilar viscoelastic properties thanks to the concurrent spreading  
 97 of multiple streams. At the macroscopic level (*i.e.*, millimetric wavelength in the ultrasound regime), this  
 98 process results in quasi-homogeneous materials obtained by randomly mixing two contrasted photopolymers.

99 In this study, two photopolymers were employed: a rigid glassy polymer (commercial name: VeroUL-  
 100 tra™White, denoted by VW) and a soft elastomer (commercial name: Elastico™Black, denoted by EB). The  
 101 two pure materials were investigated, 100% of VW (VW100) and 100% of EB (EB100), together with two  
 102 intermediate volume fractions  $V_f$  of VeroUltraWhite, 75% (VW75) and 50% (VW50). The samples were  
 103 manufactured according to a rectangular plate geometry with overall dimensions of  $35 \times 90 \times h \text{ mm}^3$ . The  
 104 length and the width of the samples were selected so that they cover the probe emission surface (recall

105 Fig. 1). The thickness  $h$  ranged between 2.5 and 4 mm to ensure a sufficient double through-transmission  
106 across the sample for all measurements below the critical angle [27], that is to avoid a too low signal-to-noise  
107 ratio (SNR) for the transverse waves because of the attenuation. Note that one sample was manufactured  
108 for each material, except for the VW100, for which two samples with two different thicknesses were tested:  
109  $h = 4$  mm (VW100) and  $h = 2.7$  mm (VW100<sub>t</sub>). The mass density  $\rho$  of each sample was determined from  
110 their mass measured with an electronic balance ( $\pm 0.01$  mg) and their dimensions measured with a caliper  
111 ( $\pm 0.02$  mm).

## 112 2.2. Experimental setup

113 All measurements were carried out using a multielement probe (Imasonic SAS, Voray sur l'Ognon, France)  
114 driven by a programmable multi-channel electronics (Advanced OEM Solutions, WestChester, USA). First  
115 developed for medical imaging applications (*e.g.*, elastography [28]), this type of device was recently used  
116 to characterize the bulk properties of both homogeneous [16] and heterogeneous photopolymer samples  
117 [15, 13] in the MHz regime. Measurements were performed using a linear transducer array that consists  
118 of 32 elements. Each element had a width and a height of 0.5 and 12 mm, respectively, leading to a total  
119 transmission surface of  $16 \times 12$  mm<sup>2</sup>. The transducer operated at a central frequency  $f_c = 2.25$  MHz ( $-6$   
120 dB frequency bandwidth from 1.5 to 3 MHz) and the pitch of the array was 0.5 mm.

121 As schematized in Fig. 1, measurements were carried out in a water tank at room temperature ( $22 \pm$   
122  $0.5$  °C) using a plane wave imaging mode, by concurrently emitting a pulse on all elements. The water  
123 temperature was controlled by a digital thermometer throughout the experiments (Zacro LCD digital ther-  
124 mometer, resolution of  $\pm 0.1$  °C), so that the water celerity could be estimated using Marczak's polynomial  
125 model [29]. The received signals  $s(t)$  were recorded during  $150 \mu\text{s}$  starting from  $300 \mu\text{s}$  after the pulse emission  
126 to target double-through transmitted waves only, using a sampling rate of 100 MHz and 12-bit resolution.  
127 The advantages of using a multielement probe both as emitter and receiver rather than a monoelement one  
128 are here twofold. First, as the wavelength in water at the central frequency was small compared to the  
129 lateral dimension of the probe array, a high directivity factor was assumed, and thus geometrical diffraction  
130 effects could be neglected in the further processing and modeling of the double through-transmitted signals.  
131 Second, the recorded signals could be averaged over the 32 elements of the probe, thus enhancing the SNR  
132 of the measurement [16]. To retrieve double through-transmitted waves, a large flat reflector was used,  
133 which was oriented parallel to the transducer. In this configuration, the backward propagating plane waves  
134 travel exactly along the same path as the forward incident wave [18]. Thereby, the transducer remains  
135 fixed, regardless of the incident angle, whereas in simple through-transmission the receiver has to be shifted

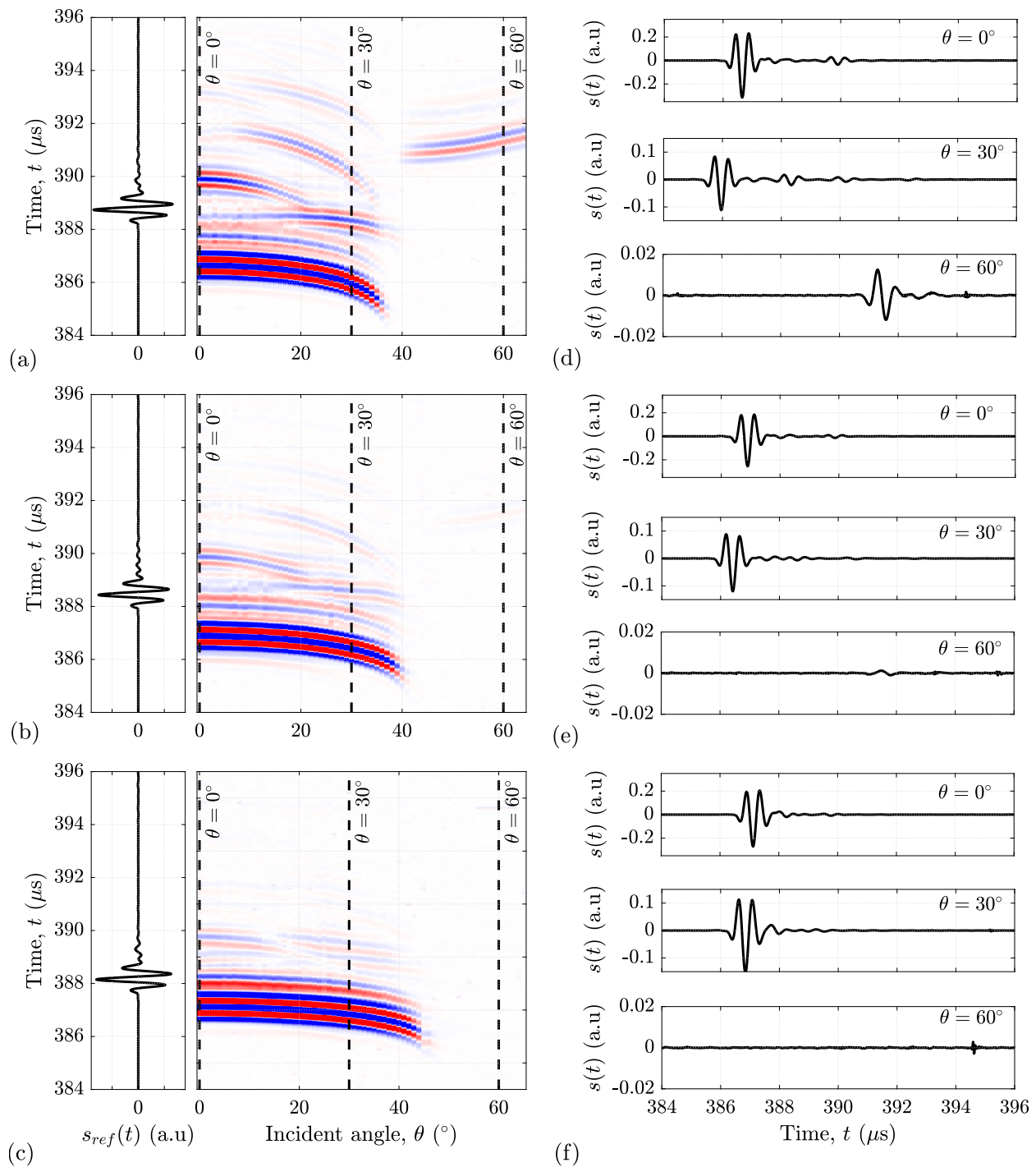


Figure 2: Examples of experimental  $\theta$ -scans of double through-transmitted signals measured on (a) VW100, (b) VW50, and (c) EB100 samples, along with their respective signal displacements  $s(t)$  ((d), (e) and (f), respectively) obtained at three specific incident angles  $\theta = 0^\circ$ ,  $30^\circ$  and  $60^\circ$ . The saturated colormap for each  $\theta$ -scan represents the normalized positive (in red) and negative (in blue) amplitude of the signals. The double through-transmitted signal measured in water only, denoted by  $\tilde{s}_{\text{ref}}(t)$ , is also shown for comparison.

136 depending on the angle of the incident wave [20, 17]. Another advantage of double through-transmission is  
 137 to reduce the amplitude differences between the different waves being compared, which is of paramount im-  
 138 portance for transverse wave measurement in highly attenuating materials, where this amplitude difference  
 139 could be in the same order of magnitude than the dynamic range of the transducer, leading to weak SNR  
 140 or signal distortion.

141 The samples were mounted on a motorized rotation stage ( $\pm 1^\circ$ ) and the signals  $s(t)$  were recorded for  
 142 each incident angle  $\theta$  between  $0^\circ$  and  $64^\circ$  with a  $1^\circ$  step. Note that the maximal incident angle was chosen  
 143 to ensure that the probe emission surface was still entirely aiming the sample. Eventually, the double  
 144 through-transmitted wave across the water only, denoted by  $s_{\text{ref}}(t)$ , was measured by removing the sample,  
 145 without moving the transducer and the flat reflector. To evaluate the reproducibility of the measurement  
 146 (*e.g.*, probe-sample-mirror alignment, parallelism of the sample's faces), the full *angular scan* measurement  
 147 was repeated three times on the VW100 sample.

148 Three examples of experimental  $\theta$ -scans are shown in Fig. 2 for the following materials: (a) VW100,  
 149 (b) VW50, and (c) EB100, together with the temporal signals  $s(t)$  obtained at three specific incident angles,  
 150 *i.e.*,  $\theta = 0^\circ, 30^\circ$  and  $60^\circ$ . As can be observed, the critical angle is visible on each  $\theta$ -scan at the angle for which  
 151 the signal vanishes, and its value increases when the volume fraction of VW decreases. In addition, while  
 152 the transverse wave is clearly visible after the critical angle for the VW100 sample (see Fig. 2(a) and (d) at  
 153  $\theta = 60^\circ$ ), its amplitude is much lower for the VW50 sample (see Fig. 2(b) and (e) at  $\theta = 60^\circ$ ) and totally  
 154 drowned in noise for the EB100 sample (see Fig. 2(c) and (f) at  $\theta = 60^\circ$ ). These observations overall support  
 155 the need of a specific methodology to retrieve the transverse properties of such attenuating materials.

### 156 3. Double through-transmission modeling

157 We consider the double through-transmission (forward path, reflection on a mirror then backward path  
 158 [18]) of a plane harmonic acoustic wave with angular frequency  $\omega$  and incidence angle  $\theta$  through an ho-  
 159 mogeneous and isotropic solid material (mass density  $\rho$ , longitudinal and transverse wave numbers  $k_\ell$  and  
 160  $k_t$ , respectively) immersed in a fluid (mass density  $\rho_0$ , wave number  $k_0$ ). The mirror surface is assumed  
 161 to be normal to the acoustic wave vector  $\mathbf{k}_0 = k_0 [\cos \theta \sin \theta]$ , so that the forward and backward paths are  
 162 *symmetrical*.

163 The method relies on the relation between the double through-transmitted signals measured *with* the  
 164 sample inserted and *without* it (so-called insertion/substitution method [30]). These are respectively denoted  
 165 by  $S(\omega)$  and  $S_{\text{ref}}(\omega)$  as the Fourier coefficients of  $s(t)$  and  $s_{\text{ref}}(t)$  at the frequency  $\omega$ . Dropping the dependence

166 of all variables on  $\omega$ , such relation can be expressed as

$$S = H_T^2 S_{\text{ref}} , \quad (1)$$

where

$$S_{\text{ref}} = S_{\text{in}} \overbrace{P(d_1 + d_2)}^{\text{forward}} \underbrace{R}_{\text{reflection}} \overbrace{P(d_2 + d_1)}^{\text{backward}} \quad (2)$$

and

$$S = S_{\text{in}} \overbrace{P(d_1) \underbrace{H_T}_{\text{transmission}} P(d_2)}^{\text{forward}} \underbrace{R}_{\text{reflection}} \overbrace{P(d_2) \underbrace{H_T}_{\text{transmission}} P(d_1)}^{\text{backward}} , \quad (3)$$

167 where  $S_{\text{in}}$  is the complex spectrum of the input signal,  $P(d_i) = e^{-ik_0 d_i}$  is the propagation *phase* associated  
 168 with the fluid path  $d_i$ , with  $i = 1, 2$ ,  $R$  is the transfer function associated with the reflection on the mirror,  
 169 and  $H_T$  is the transfer function corresponding to the *single* transmission of waves through the specimen.  
 170 The distances  $d_1$  and  $d_2$  respectively correspond to the probe-sample and sample-mirror distances, as shown  
 171 in Fig. 1. These are defined with respect to *the plane of symmetry of the sample*, so that  $H_T$  already takes  
 172 into account the phase  $P(-h)$  associated with the insertion of the sample in the propagating path.

173 It is thus clear that measuring the reference signal  $s_{\text{ref}}(t)$  presents several advantages: the transfer  
 174 function  $H_T$  can be estimated experimentally and used as the basis for the identification of the sample  
 175 properties without prior knowledge of the distances  $d_1$  and  $d_2$ , the input signal  $S_{\text{in}}$  and, more interestingly,  
 176 the mirror reflection transfer function  $R$ . Hence, the mirror can be of arbitrary material and/or thickness  
 177 [17], without constraint associated with the time separation of the successive trains of echoes.

### 178 3.1. Single transmission transfer function

179 As the relation between  $S$  and  $S_{\text{ref}}$  only involves the squared transfer function  $H_T$ , the model of the  
 180 experimental data is reduced to the calculation of the *single wave transmission* that corresponds to the  
 181 problem statement shown in Fig. 3. We thus derive here  $H_T$  as the ratio between the transmitted and  
 182 incident complex wave amplitudes, respectively denoted by  $a_T$  and  $a_I$ , for any incident angle  $\theta$  and frequency  
 183  $\omega$ . The 2D coordinate system  $(x, y)$  is chosen so that the two infinite solid–fluid plane interfaces are situated  
 184 at  $x = \pm h/2$  and their normal is along  $x$ . Hence, all wave phases are expressed *with respect* to the solid  
 185 layer plane of symmetry at  $x = 0$ .

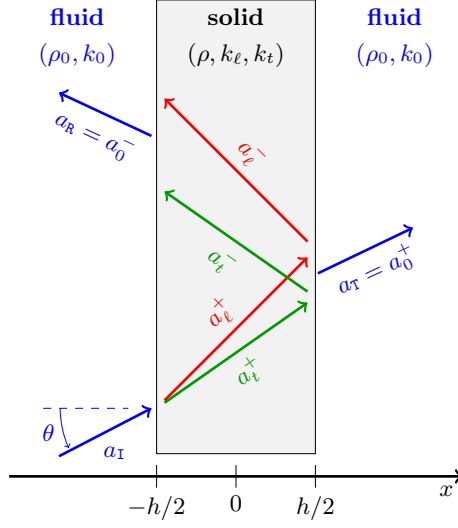


Figure 3: Scheme of the single wave transmission problem.

186 In each subdomain, the pressure field  $p$  or displacement field  $\mathbf{u}$  is assumed as a combination of plane  
 187 waves with amplitudes  $a_r$ , polarizations  $\mathcal{P}^{(r)}$  or  $\mathbf{U}^{(r)}$  and wavevector  $\mathbf{k}_r = [\xi_r \ \kappa_r] = k_r [\cos \theta_r \ \sin \theta_r]$  as

$$\begin{bmatrix} \mathbf{u}(x, y) \\ p(x, y) \end{bmatrix} = \sum_r a_r \begin{bmatrix} \mathbf{U}^{(r)} \\ \mathcal{P}^{(r)} \end{bmatrix} e^{-i(\xi_r x + \kappa_r y)}, \quad (4)$$

188 with  $r$  standing either for the wave propagating in water ( $r = 0$ ) or the *longitudinal* ( $r = \ell$ ) and *transverse*  
 189 ( $r = t$ ) waves in the solid.

The relation between wave amplitudes is governed by the continuity of the stress vector and normal displacements at both solid–fluid interfaces, which can be expressed as

$$\sigma_{xx}(x = \pm \frac{h}{2}, y) + p(x = \pm \frac{h}{2}, y) = 0, \quad (5a)$$

$$\sigma_{xy}(x = \pm \frac{h}{2}, y) = 0, \quad (5b)$$

$$\rho_0 \omega^2 u_x(x = \pm \frac{h}{2}, y) - \frac{\partial}{\partial x} p(x = \pm \frac{h}{2}, y) = 0. \quad (5c)$$

190 As these 6 continuity equations need to be fulfilled for any  $y$ , the wave modulation  $e^{-i\kappa_r y}$  along the vertical  
 191 axis must be constant across interfaces. This leads to the well-known Snell-Descartes law as

$$\kappa_r = k_r \sin \theta_r = k_0 \sin \theta = \kappa, \quad \forall r \quad (6)$$

192 where  $\kappa$  is the *vertical* component of the wavevector that is a constant of the problem. It is worth noting  
 193 that the wavevectors  $\mathbf{k}_r$  are generally complex and can have non-colinear real and imaginary parts; thus, the  
 194 angles  $\theta_r$  can take complex values in general. Here it is chosen to derive the following expressions using the  
 195 wavevector components  $(\xi_r, \kappa)$ . In particular, the *absolute* projection  $\chi_r$  of the wavevectors on the  $x$ -axis,  
 196 such that  $\xi_r = \pm\chi_r$ , is defined as

$$\chi_r = \sqrt{k_r^2 - \kappa^2}, \quad (7)$$

197 where  $\chi_r$  may take complex values when  $|k_r| < |\kappa|$ .

198 The 6 continuity equations in Eq. 5 can then be recast following a global matrix formalism [31] (see Ap-  
 199 pendix A). By taking into account the symmetries of the fluid-solid interfaces and of the forward-backward  
 200 waves, the matrix can be recast to obtain two decoupled  $3 \times 3$  systems, which can be subsequently analytically  
 201 inverted to recover the expressions of  $a_0^S$  and  $a_0^A$ , so that  $a_r^\pm = a_r^S \pm a_r^A$ , where the superscripts  $A$  and  $S$  stand  
 202 for antisymmetric and symmetric parts, respectively. At the end, using the fact that  $a_T = a_0^+ = a_0^S + a_0^A$ ,  
 203 the analytical expression of the complex transfer function associated with the single wave *transmission*  $H_T$   
 204 through the solid domain reads as

$$H_T = \frac{a_T}{a_I} = -4 \frac{\gamma}{\Pi_0} \frac{\alpha \Pi_\ell (\Pi_t^2 - 1) + \beta \Pi_t (\Pi_\ell^2 - 1)}{\Delta}, \quad (8)$$

where

$$\begin{aligned} \Delta &= (\alpha + \beta - \gamma)^2 (\Pi_\ell^2 - 1) (\Pi_t^2 - 1) \\ &\quad - 4\beta\gamma (\Pi_\ell^2 - 1) - 4\alpha\gamma (\Pi_t^2 - 1) \\ &\quad + 4\alpha\beta (\Pi_\ell - \Pi_t)^2, \end{aligned} \quad (9a)$$

$$\Pi_r = e^{-i\chi_r h}, \quad (9b)$$

$$\alpha = (\chi_t^2 - \kappa^2)^2, \quad (9c)$$

$$\beta = 4\kappa^2 \chi_\ell \chi_t, \quad (9d)$$

$$\gamma = \frac{\rho_0}{\rho} \frac{\chi_\ell}{\chi_0} (\chi_t^2 + \kappa^2)^2. \quad (9e)$$

205

206 *3.2. Frequency-dependence of the wave number*

207 The single transmission transfer function  $H_T$  given by Eq. (8) can be used to compute, from Eq. (1)  
 208 and the measured reference signal  $s_{\text{ref}}(t)$ , the  $\theta$ -scan corresponding to a sample with given wave dispersion  
 209 characteristics. In other words, the dependence of  $k_0$ ,  $k_\ell$  and  $k_t$  with the frequency  $\omega$  needs to be provided in  
 210 order to predict the double through-transmitted signal  $s(t)$ . In the following, we use the general formulation

$$k_r(\omega) = \frac{\omega}{v_r(\omega)} - i\alpha_r(\omega), \quad (10)$$

211 where  $v_r(\omega)$  and  $\alpha_r(\omega)$  respectively denote the phase velocity and attenuation as *real* functions of the  
 212 frequency.

213 *3.2.1. Non-dispersive media*

214 As a starting point, non-dispersive properties can be assumed for both the solid and the fluid media,  
 215 characterized with a *complex* and *constant* wave celerity  $c_r \in \mathbb{C}$  such that  $k_r(\omega) = \omega/c_r$ . Hence,

$$v_r(\omega) = \Re(c_r) \left( 1 + \frac{\Im(c_r)^2}{\Re(c_r)^2} \right) \quad \text{and} \quad \alpha_r(\omega) = \omega \frac{\Im(c_r)}{|c_r|^2}. \quad (11)$$

216 This assumption is known to provide a good approximation for the wave number  $k_0$  associated with pressure  
 217 waves in the fluid. In particular, as the temperature is known, the wave celerity  $c_0$  in water can be predicted  
 218 using Marczak's polynomial model [29]. In this work, the non-dispersive model from Eq. (11) is also used to  
 219 provide an initialization for the material wave numbers ( $k_\ell$ ,  $k_t$ ) needed in the inverse problem (see Sec 4.1).

220 *3.2.2. Szabo's rheological model*

Viscoelastic materials such as photopolymers exhibit dispersive losses that cannot be accurately cap-  
 tured by a classical rheological Voigt model (*i.e.*, attenuation proportional to  $\omega^2$ ), which may be due to  
 their heterogeneous nature involving multiple relaxation mechanisms. Nevertheless, earlier studies by our  
 group [15, 16] evidenced that the viscoelastic behavior of photopolymers may be well described by the  
 Szabo's model [32]. This phenomenological model has the advantage to describe not only the frequency-  
 dependence of the attenuation  $\alpha_r(\omega)$  with a power-law, but also the frequency variations of the phase velocity  
 $v_r(\omega)$ , because the model is causal in the sense that it satisfies Kramer-Kronig relationships [33]. In this  
 regard, convenient expressions of  $v_r(\omega)$  and  $\alpha_r(\omega)$  as a function of a reference frequency  $\omega_c$ , were proposed

by Bakaric et al. [10] as

$$v_r(\omega) = \left( \frac{1}{v_{cr}} + \frac{\alpha_{cr}}{\omega_c} \tan\left(\frac{\pi}{2} y_r\right) \left( \left( \frac{\omega}{\omega_c} \right)^{y_r-1} - 1 \right) \right)^{-1}, \quad (12a)$$

$$\alpha_r(\omega) = \alpha_{cr} \left( \frac{\omega}{\omega_c} \right)^{y_r}, \quad (12b)$$

where  $\omega_c = 2\pi f_c$  can be deliberately chosen as the central frequency of the available ultrasonic probe. With the Szabo model, the dependence of the wave number  $k_r(\omega)$  with the frequency is thus completely given by the three corresponding parameters  $v_{cr}$ ,  $\alpha_{cr}$  and  $y_r$ , which respectively denote the phase velocity and attenuation at the central frequency  $\omega_c$  and a dispersion coefficient.

#### 4. Identification of viscoelastic material properties

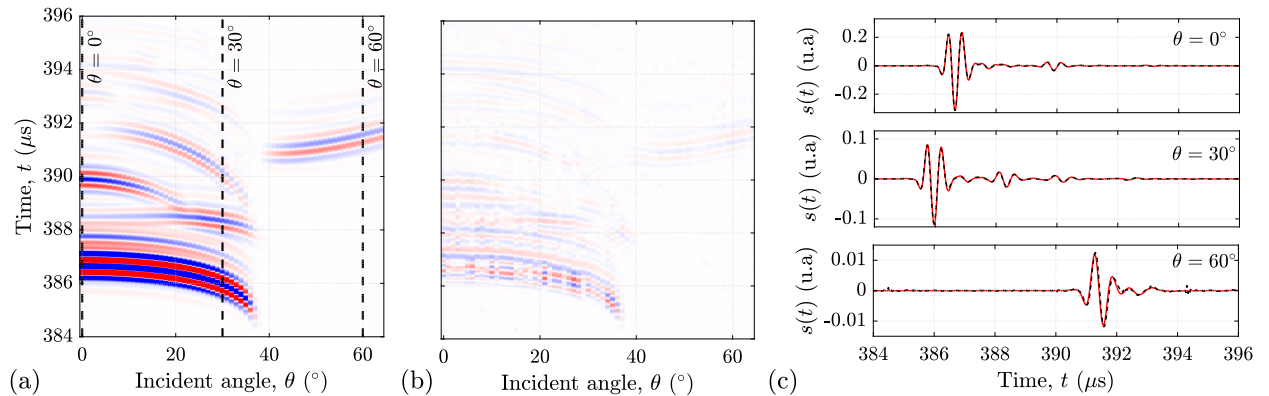


Figure 4: Illustration of the optimization procedure applied to one realization of the VW100 sample: (a) Modeled  $\theta$ -scan obtained with the optimized set of parameters  $\hat{\mathbf{p}}$ , (b) difference between the experimental  $\theta$ -scan shown in Fig. 2a and the modeled  $\theta$ -scan, and (c) comparison of experimental ( - - - ) and modeled ( — ) double through-transmitted signals at three specific incident angles  $\theta = 0^\circ$ ,  $30^\circ$  and  $60^\circ$ . The colormaps of (a) and (b) are ten times saturated to highlight the successive echoes arising from the mode conversion at the interfaces.

The inverse procedure used to identify the ultrasound characteristics (phase velocity  $v_r(\omega)$  and attenuation  $\alpha_r(\omega)$ ) from a measured  $\theta$ -scan  $s(t, \theta)$ , the reference signal  $s_{\text{ref}}(t)$  and the model  $H_{\text{T}}(\omega, \theta)$  is described in what follows. In practice, the wave dispersion  $k_r(\omega)$  needs to be given on the whole frequency domain in order to reconstruct a *candidate*  $\theta$ -scan using Eq. (1). However, the ultrasound probe is characterized by a *limited* bandwidth around  $\omega_c$ . Thereby, a rheological model such as Eq. (11) or Eq. (12) is used to regularize the inverse problem. In particular, the procedure is here divided into two steps [34]. First, an initial *guess* is provided using the estimation of a non-dispersive longitudinal celerity  $c_\ell$  via a direct problem. Second, an optimization procedure is implemented to estimate the model parameters of the Szabo model.

234 *4.1. Calibration of the model at normal incidence*

235 For a normally incident plane wave ( $\theta = 0$ ), the vertical component of the wavevector vanishes, thus  
 236  $\kappa = 0$ . Consequently, the expression of  $H_T$  in Eq. (8) can be simplified to

$$H_T = \frac{T_\ell^2}{\Pi_0} (\Pi_\ell^{-1} - R_\ell^2 \Pi_\ell)^{-1} , \quad (13)$$

where

$$T_\ell = \frac{2\zeta_\ell}{\zeta_\ell + 1} , \quad R_\ell = \frac{\zeta_\ell - 1}{\zeta_\ell + 1} , \quad \text{and} \quad \zeta_\ell = \frac{\rho}{\rho_0} \frac{k_0}{k_\ell} .$$

237 Under the hypothesis of a *non-dispersive* material behavior (recall Eq. (11)), it is noteworthy that  $\zeta_\ell$ , thus  $T_\ell$   
 238 and  $R_\ell$ , are constants. In such a case, this allows us to express the inverse of the double through-transmission  
 239 transfer function as

$$\frac{S_{\text{ref}}(\omega)}{S(\omega)} = [H_T(\omega)]^{-2} = \frac{\psi_0^\omega}{T_\ell^4} (\psi_\ell^{-\omega} - 2R_\ell^2 + R_\ell^4 \psi_\ell^\omega) , \quad (14)$$

240 where  $\psi_r = e^{-i\frac{2h}{c_r}}$ . Once the above expression is multiplied by  $\psi_0^{-\omega}$ , which can be computed from the wave  
 241 celerity  $c_0$  in water and the specimen thickness  $h$ , it takes the form of an harmonic polynomial in  $\psi_\ell$  only.  
 242 Hence, a Prony method is implemented here to provide an initial guess of the longitudinal wave number  $k_\ell$   
 243 associated with a constant celerity  $c_\ell \in \mathbb{C}$  (see Appendix B).

244 *4.2. Estimation of dispersive properties*

245 We propose an optimization procedure based on the full  $\theta$ -scan, to identify, for each sample, the pa-  
 246 rameters of the Szabo model (recall Eq. (12)), *i.e.*,  $\mathbf{p}_r = [v_{cr}, \alpha_{cr}]$  associated with longitudinal ( $r = \ell$ ) and  
 247 transverse ( $r = t$ ) waves. Note that the dispersion coefficient  $y_r$  involved in Szabo's model is assumed to be  
 248 equal to  $y_\ell = 1.1$  and  $y_t = 0.7$  for all the investigated materials, based on the results obtained in [15, 16].  
 249 In addition, the sample thickness  $h$  and a uniform angle correction  $\theta_0$ , which may be interpreted as an  
 250 experimental angle offset at the normal incidence, are included as *refinement* parameters in order to avoid  
 251 biases due to their experimental initial determination. The set of parameters  $\mathbf{p} = [\mathbf{p}_\ell, \mathbf{p}_t]$  is thus estimated  
 252 via the following minimization problem:

$$(\hat{\mathbf{p}}, \hat{h}, \hat{\theta}_0) = \arg \min_{\substack{\mathbf{p} \in \mathbb{R}^4 \\ h \in \mathbb{R} \\ \theta_0 \in \mathbb{R}}} \sum_{t_i, \theta_j} \Delta s(t_i, \theta_j; \mathbf{p}, h, \theta_0)^2 \quad (15)$$

253 with the residual  $\Delta s$  given by:

$$\Delta s(t, \theta; \mathbf{p}, h, \theta_0) = u(t, \theta; \mathbf{p}, h, \theta_0) - s(t, \theta) \quad (16)$$

254 and the signal *model*  $u$  computed as:

$$u(t, \theta; \mathbf{p}, h, \theta_0) = \mathcal{F}^{-1} \left[ H_T(\omega, \theta + \theta_0; \mathbf{p}, h) S_{\text{ref}}(\omega) \right] \quad (17)$$

255 where  $\mathcal{F}^{-1}$  denotes the inverse Fourier Transform. In contrast to an estimation that would be based on the  
 256 transfer function only, performing the minimization directly on the temporal data  $s(t, \theta)$  provides a natural  
 257 *weighting* of the error, associated with the reference signal  $s_{\text{ref}}$ , that accounts both for the probe's bandwidth  
 258 and losses/reflections in the mirror.

259 The optimization procedure was implemented using a gradient-based method implemented in Matlab  
 260 (The MathWorks Inc., Natick, MA) and finite differences for the numerical estimation of the gradient of the  
 261 residuals.

262 As required by such approach, a specific initialization of the set of model parameters  $\mathbf{p}$  was performed.  
 263 The longitudinal parameters of the Szabo model  $\mathbf{p}_\ell$  were initialized from the longitudinal wave number  
 264  $k_\ell$ , which is associated with the constant complex velocity  $c_\ell$  estimated at the normal incidence using the  
 265 model calibration described in Sec. 4.1. Likewise, the initialization of the transverse parameters  $\mathbf{p}_t$  was  
 266 subsequently determined from the complex transverse wave celerity  $c_t$ , which can be determined from the  
 267 relation

$$c_t = c_\ell \sqrt{\frac{1 - 2\nu_1}{2(1 - \nu_1)}}, \quad (18)$$

268 assuming an *initial* Poisson's ratio  $\nu_1$  equal to 0.38 for all samples [16]. Eventually, the angle correction  $\theta_0$   
 269 was arbitrary initialized to  $0.01^\circ$ , which corresponds to the uncertainty of the rotating platform and the  
 270 sample thickness  $h$  was initialized to the experimental value measured with a caliper, as described in Sec. 2.

271 To serve as an example, Fig. 4 depicts the optimal matching between the measured and modeled  $\theta$ -scans  
 272 for the VW100 sample. As can be observed, the measured and modeled signals are in excellent agreement,  
 273 which is shown herein for three specific incidence angles  $\theta$ .

274 4.3. Uncertainty of the identification procedure

275 The uncertainty associated with the inverse procedure, noted  $\sigma_{\hat{\mathbf{p}}}$ , was evaluated via error propagation  
 276 for the set of parameter  $\hat{\mathbf{p}}$ , assuming independent variables, as

$$\sigma_{\hat{p}_k} = \sqrt{\sum_{t_i, \theta_j} \left( \frac{\partial p_k}{\partial u} \Big|_{(t_i, \theta_j; \hat{\mathbf{p}}, \hat{h}, \hat{\theta}_0)} \right)^2 \Delta s(t_i, \theta_j; \hat{\mathbf{p}}, \hat{h}, \hat{\theta}_0)^2}, \quad (19)$$

277 where the terms between parentheses are obtained via an inversion of the model sensitivity matrix. At the  
 278 end, each parameter is associated to a confidence interval of 95% such that  $p_k \pm 1.96 \sigma_{\hat{p}_k}$ .

279 **5. Results**

280 The optimized Szabo's parameters  $\mathbf{p}_r = [v_{cr}, \alpha_{cr}]$ , for  $r = \{\ell, t\}$ , are reported in Table 1 for all samples,  
 281 together with the optimized sample thickness  $\hat{h}$  and the uniform incident angle correction  $\hat{\theta}_0$ . All values  
 282 are given with their respective uncertainty, which is related to the identification procedure as explained in  
 283 Sec. 4.3.

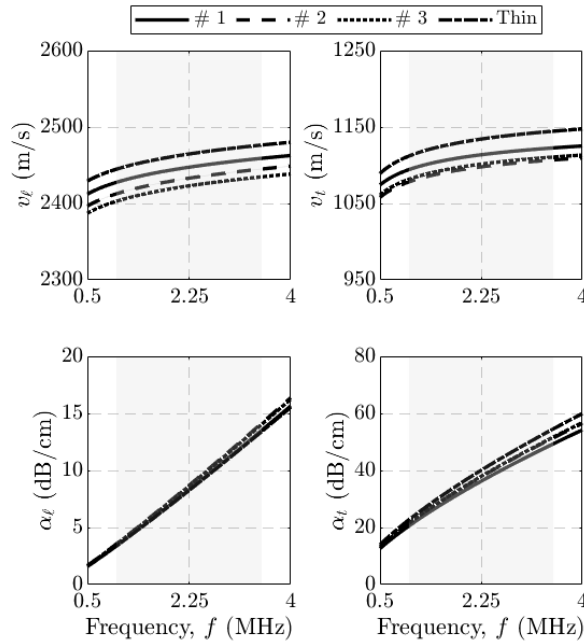


Figure 5: Ultrasound characteristics, *i.e.* the phase velocity  $v_r(\omega)$  and the attenuation  $\alpha_r(\omega)$ , obtained for three measurements performed on the VW100 sample. The light gray areas display the useful frequency bandwidth of the probe on which the inversion was conducted.

284 5.1. Measurement reproducibility

285 The ultrasound characteristics of VW are shown in Fig. 5 for three independent measurements performed  
 286 on the same sample, along with a measurement performed on a thinner sample.

287 On the one hand, at the probe's central frequency, *i.e.*,  $f_c = 2.25$  MHz, some discrepancies can be  
 288 observed across the three measurement repetitions. While the optimized longitudinal phase velocity  $\hat{v}_{cl}$   
 289 varies between  $2423.0 \pm 0.5$  m/s and  $2447.4 \pm 0.2$  m/s, the transverse phase velocity  $\hat{v}_{ct}$  varies between  
 290  $1097.9 \pm 0.3$  m/s and  $1113.7 \pm 0.1$  m/s, which represents less than 2% of relative error. Comparable results  
 291 are obtained for the attenuation, where the optimized longitudinal attenuation  $\hat{\alpha}_{cl}$  (respectively transverse  
 292 attenuation  $\hat{\alpha}_{ct}$ ) ranges between  $8.31 \pm 0.01$  dB/cm and  $8.69 \pm 0.04$  dB/cm (respectively between  $37.9 \pm 0.1$   
 293 dB/cm and  $36.2 \pm 0.1$  dB/cm), which stands for a relative error of about 5%.

294 On the other hand, although the results obtained on the two samples of VW named as VW100 and  
 295 VW100<sub>t</sub> in Table 1 are in relatively good quantitative agreement, the overall optimized Szabo's parameters  
 296  $[\mathbf{p}_\ell, \mathbf{p}_t]$  are slightly higher for the thinner sample. These variations across the samples and the measurements  
 297 will be further discussed in Sec. 6.

298 5.2. Influence of the material mixing

	$\rho$ (kg/m <sup>3</sup> )	Measure #	$\hat{\theta}_0$ (°)	$\hat{h}$ (mm)	$r$	$\hat{v}_{cr}$ (m/s)	$\hat{\alpha}_{cr}$ (dB/cm)
VW100	$1185.3 \pm 0.5$	1	$0.14 \pm 0.01$	$3.98 \pm 0.00$	$\ell$	$2447.4 \pm 0.2$	$8.31 \pm 0.01$
					$t$	$1113.7 \pm 0.1$	$36.2 \pm 0.1$
		2	$0.03 \pm 0.03$	$3.93 \pm 0.00$	$\ell$	$2433.1 \pm 0.9$	$8.69 \pm 0.04$
					$t$	$1097.9 \pm 0.3$	$37.8 \pm 0.11$
		3	$0.28 \pm 0.02$	$3.93 \pm 0.00$	$\ell$	$2423.0 \pm 0.5$	$8.63 \pm 0.02$
					$t$	$1101.5 \pm 0.2$	$37.9 \pm 0.1$
VW100 <sub>t</sub>	$1185.3 \pm 0.5$	1	$0.64 \pm 0.07$	$2.76 \pm 0.01$	$\ell$	$2464.7 \pm 1.7$	$8.25 \pm 0.11$
				$t$	$1134.2 \pm 0.4$	$40.1 \pm 0.1$	
VW75	$1162.9 \pm 0.4$	1	$0.01 \pm 0.02$	$3.67 \pm 0.00$	$\ell$	$2379.5 \pm 0.3$	$9.50 \pm 0.02$
				$t$	$1069.1 \pm 0.2$	$47.2 \pm 0.2$	
VW50	$1147.9 \pm 0.7$	1	$1.16 \pm 0.02$	$3.39 \pm 0.01$	$\ell$	$2255.8 \pm 0.6$	$11.5 \pm 0.1$
				$t$	$991.4 \pm 1.0$	$69.3 \pm 0.8$	
EB100	$1115.1 \pm 0.8$	1	$1.24 \pm 0.03$	$2.78 \pm 0.01$	$\ell$	$2035.1 \pm 0.9$	$15.4 \pm 0.03$
				$t$	$740.6 \pm 0.8$	$86.3 \pm 1.5$	

Table 1: Measured mass density  $\rho$  and optimized ultrasound parameters  $\hat{\mathbf{p}} = [\hat{v}_{cr}, \hat{\alpha}_{cr}, \hat{\theta}_0, \hat{h}]$ . For each quantity, the uncertainty associated with the measurement (for  $\rho$ ) or to the inverse method (for  $\hat{\mathbf{p}}$ ) is indicated, with 96% of confidence for the latter.

299 The influence of the volume fraction  $V_f$  of VW on the frequency-dependent ultrasound characteristics is  
 300 depicted in Fig. 6 for  $V_f = 100, 75, 50$  and  $0\%$ , recalling that  $0\%$  corresponds to the pure soft elastomer,  
 301 named as EB100. For the readability of the figure, only one out of the three measurements performed on  
 302 VW100 was arbitrarily chosen, *i.e.*, measurement #1. As expected, the optimized Szabo's parameters  $\mathbf{p}_r$  are  
 303 closely related to the volume fraction of VW. Indeed, when the volume fraction of VW decreases (from  $100\%$

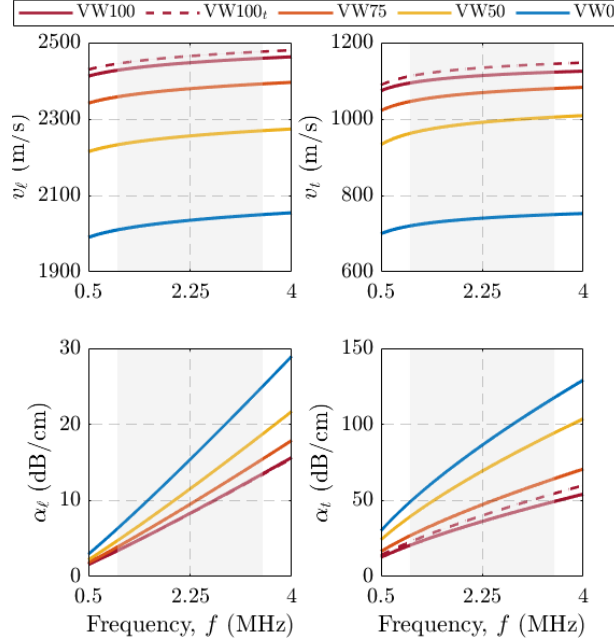


Figure 6: Ultrasound characteristics, *i.e.* the phase velocity  $v_r(\omega)$  and the attenuation  $\alpha_r(\omega)$ , of VW100 (red), VW75 (orange), VW50 (yellow) and EB100 (blue) samples. The light gray areas display the useful frequency bandwidth of the probe on which the inversion was conducted.

304 to 0%), the phase velocity decreases, and the attenuation increases. In particular, the optimized longitudinal  
 305 parameters, *i.e.*,  $[v_{cl}, \alpha_{cl}]$ , range from  $[2447 \pm 0.2 \text{ m/s}, 8.31 \pm 0.01 \text{ dB/cm}]$  for VW100 to  $[2035.1 \pm 0.9 \text{ m/s},$   
 306  $15.4 \pm 0.03 \text{ dB/cm}]$  for EB100 (recall Table 1). A similar trend is observed for the transverse parameters,  
 307 *i.e.*,  $[v_{ct}, \alpha_{ct}]$ , with optimized values ranging from  $[1134.2 \pm 0.4 \text{ m/s}, 40.1 \pm 0.1 \text{ dB/cm}]$  to  $[740.6 \pm 0.8$   
 308  $\text{m/s}, 86.3 \pm 1.5 \text{ dB/cm}]$ . Overall, it can be observed that the transverse phase velocity  $v_t$  is around two to  
 309 three times lower than  $v_\ell$  depending on the volume fraction of VW.

### 310 5.3. Dispersive properties of photopolymer materials

311 As displayed in Fig. 5, the longitudinal (respectively transverse) phase velocity  $v_\ell$  (respectively  $v_t$ )  
 312 increases of nearly 50 m/s (respectively 40 m/s) over the useful frequency bandwidth (*i.e.*, 0.75–3.75 MHz).  
 313 Likewise, the longitudinal (respectively transverse) attenuation  $\alpha_\ell$  (respectively  $\alpha_t$ ) increases from 3 dB/cm  
 314 to nearly 15 dB/cm (respectively from 12 dB/cm to 70 dB/cm). Interestingly, the dispersion of both the  
 315 longitudinal and transverse characteristics, which depends on  $\alpha_{cr}$  and  $y_r$ , is similar for all samples, no matter  
 316 whether they consist of pure or composite materials (see Fig. 6).

## 317 6. Discussion

318 The objective of the present work was to provide a methodological framework to identify both the  
319 frequency-dependent viscoelastic properties in the MHz regime and the thickness of highly attenuating sam-  
320 ples. Towards this goal, angular measurements achieved in double through-transmission were conducted on  
321 different photopolymer samples, which were obtained using multi-material additive manufacturing. A direct  
322 modeling of the double trough-transmission, based on a global matrix formalism, was proposed, along with  
323 an inverse procedure, which altogether allowed inferring frequency-dependent ultrasound characteristics.

324 The proposed framework presents several advantages. First, in contrast to the approach used in [16], the  
325 experimental input consisted of the raw temporal signals (*i.e.*, angular measurements) achieved in double  
326 through-transmission. Thereby, no signal processing steps were required to isolate the echoes of interest to  
327 compute their respective complex spectra, such as signal windowing or phase unwrapping. In addition, the  
328 use of a reference signal (*i.e.*, a double through-transmitted signal through water only) allowed weighting  
329 the measurements by the input signal bandwidth, thereby preventing any frequency range to be manually  
330 selected. A further advantage of using this reference signal was that the distances between the probe, the  
331 sample and the mirror could be considered as unknown. Second, the use of global matrix formalism for the  
332 modeling of the double through-transmitted signals allowed retrieving temporal  $\theta$ -scans, which could be sys-  
333 tematically compared to the measurements in order to identify the ultrasound characteristics. In addition,  
334 the use of a rheological model such as the Szabo's model allowed identifying frequency-dependent properties  
335 with a reduced set of model parameters. Third, all the investigated parameters, including both ultrasound  
336 characteristics (phase velocity and attenuation of longitudinal and transverse waves) and the sample's thick-  
337 ness were simultaneously optimized. In particular, an original initialization strategy using Prony's series  
338 decomposition was proposed, which is based on the measurement achieved in normal incidence. Altogether,  
339 this study provides a deeper insight on the ultrasound characteristics of state-of-the-art photopolymers  
340 commonly used for polyjet printing purposes, and in particular their transverse properties, which character-  
341 ization remained challenging for such attenuating materials. To the best of our knowledge, the transverse  
342 properties of the soft elastomer (EB) investigated herein have not been reported in the ultrasonic regime so  
343 far.

344 The longitudinal bulk properties of the glassy polymer (VW) and of the soft elastomer (EB) were in  
345 good qualitative agreement with those reported in previous studies for photopolymer materials achieved  
346 with similar additive manufacturing technologies [9, 16, 10]. Nonetheless, it is not straightforward to quan-  
347 titatively compare the results of the present work with the literature because (i) the photopolymers may

348 slightly differ across studies (their commercial names change and their compositions are unknown) and (ii)  
 349 the material properties are not necessarily evaluated in the same frequency range, whereas it has been  
 350 shown that such photopolymer materials exhibit a frequency-dependent behavior. On the one hand, the  
 351 longitudinal phase velocity of VW-like samples reported in the literature range from 2464 to 2495 m/s at a  
 352 central frequency of about 2.25 MHz [10, 15, 16]. Likewise, the values of the longitudinal attenuation of VW  
 353 reported herein (recall Table 1) is also in close agreement with those of the literature, which range from 7.6  
 354 to 8.2 dB/cm [15, 16]. On the other hand, Bakaric et al. [10] reported longitudinal bulk properties close to  
 355 2040 m/s and 18.4 dB/cm for Agilus30, which is a soft elastomer similar to ElasticoBlack (EB) used for 3D  
 356 printing purposes with the Polyjet technology.

357 In the same way, the transverse bulk properties reported by Gattin et al. [16] for a VW-like sample with a  
 358 phase velocity of  $1112.9 \pm 4.5$  m/s and attenuation of  $37.9 \pm 0.5$  dB/cm are in very good agreement with those  
 359 obtained herein (recall Table 1). While the transverse properties of EB have not been reported so far, Gattin  
 360 et al. [16] investigated a composite material made of VW and an intermediate volume fraction up to 18%  
 361 of a soft elastomer (TangoBlack™), a photopolymer also used for 3D printing with Polyjet technology. The  
 362 properties obtained for this material are consistent with our VW75 sample (intermediate volume fraction  
 363 equal to 25% of EB) with a transverse phase velocity equal to  $1066.2 \pm 3.7$  m/s (VW75:  $1069.1 \pm 0.2$ ) and an  
 364 attenuation of  $48.8 \pm 0.6$  dB/cm (VW75:  $47.2 \pm 0.2$  dB/cm).

365 Overall, the reported properties are all in very good agreement with those of the literature, thereby con-  
 366 firming the robustness of the proposed inverse identification procedure based on ultrasonic angular measure-  
 367 ments achieved in double through-transmission. Moreover, the values of the sample thickness concurrently  
 368 obtained from the optimization are very close to the reference values with less than 5% of relative error,  
 369 which is consistent with the resolution of the caliper ( $\pm 0.02$ mm). Eventually, it is worth mentioning the  
 370 possible effects of the water -and thus the sample- temperature, which could explain the small discrepancies  
 371 observed [35]. Despite these promising results, our approach suffers from some limitations. First, some  
 372 discrepancies were observed over the three measurement repetitions performed on the VW100 samples, as  
 373 well as between the two VW100 samples with different thicknesses (recall Fig. 5). The inter and intra-sample  
 374 reproducibilities, which are of the same order of magnitude and lower than 5% for both the phase velocities  
 375  $v_{cr}$  and attenuations  $\alpha_{cr}$ , were significantly higher than the uncertainty associated with the inverse proce-  
 376 dure. Such variations may be related to a misalignment of the probe/mirror, or to the parallelism/curvature  
 377 of the sample, and could be minimized by improving the experimental setup and manufacturing process.  
 378 Second, the number of investigated samples is relatively low and further composite samples with intermedi-

379 ate volume fractions should be incorporated in future studies, with the aim of establishing mixing rules for  
380 these materials in the ultrasonic regime.

381 Altogether, this study provides an original framework to retrieve the frequency-dependent ultrasound  
382 characteristics of dissimilar viscoelastic materials, ranging from glassy polymer to soft elastomer. The  
383 properties reported here are of significant interest towards applications requiring a precise knowledge of  
384 both the longitudinal and transverse bulk properties of such materials in the MHz regime, as for instance  
385 the modeling of the wave interactions with multi-material architected media [14]. A next step will be to  
386 generalize the identification procedure to other kind of polymers and to anisotropic media. In this respect,  
387 the use of a phenomenological model to describe the dispersion and attenuation relation will be avoided.  
388 To this end, the measurements will be carried out on a larger frequency-range and the inversion will be  
389 performed for each frequency. Likewise, the versatility of design at different scales (from micro to macro)  
390 offered by multi-material additive manufacturing strengthens the need to account for anisotropy in further  
391 works.

## 392 **7. Author declaration**

393 There are no known conflicts of interest associated with this publication and there has been no significant  
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## 396 **8. Data availability**

397 The experimental data as well as the MATLAB script necessary to reproduce the presented method and  
398 results are available as an open access repository [36].

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## 477 Appendix A. Global matrix formalism

In the two fluid subdomains ( $|x| > h/2$ ), the Helmholtz equation governing the pressure field accepts two plane waves solutions corresponding to the forward *incident*  $a_{\text{I}}$  and *transmitted*  $a_{\text{T}} \equiv a_0^+$  waves with  $\xi_{\text{I}} = \xi_{\text{T}} = \chi_0$  and the backward *reflected* wave  $a_{\text{R}} \equiv a_0^-$  with  $\xi_{\text{R}} = -\chi_0$  (see Eq. (7)). In the isotropic solid subdomain ( $|x| < h/2$ ), the 2D Navier-Cauchy equations apply which accept 2 solution pairs of backward and forward waves defined by  $\xi_r = \pm\chi_r$  and corresponding to *longitudinal* ( $r = \ell$ ) and *transverse* ( $r = t$ )

motion associated, respectively, with:

$$k_\ell^2 = \chi_\ell^2 + \kappa^2 = \omega^2 \frac{\rho}{\lambda + 2\mu} \quad , \quad \mathbf{u}^{(\ell)} = \frac{1}{\omega^2} \begin{bmatrix} \xi_r \\ \kappa \end{bmatrix} \quad (\text{A.1a})$$

$$k_t^2 = \chi_t^2 + \kappa^2 = \omega^2 \frac{\rho}{\mu} \quad , \quad \mathbf{u}^{(t)} = \frac{1}{\omega^2} \begin{bmatrix} -\kappa \\ \xi_r \end{bmatrix} \quad (\text{A.1b})$$

where  $(\lambda, \mu)$  are the Lamé coefficients and the scaling of the wave polarization vectors  $\mathbf{u}^{(r)}$  with  $\omega$  has been introduced to simplify the derivations. Using Hooke's law to express the local stress vector as a function of the elementary waves in the solid, the 6 continuity equations in Eq. (5) (3 equations on 2 interfaces) can be recast as the following system:

$$\sum_{r=\{0,\ell,t\}} \left( \begin{bmatrix} z_r \mathbf{B}^{(r^+)} & z_r^{-1} \mathbf{B}^{(r^-)} \\ z_r^{-1} \mathbf{B}^{(r^+)} & z_r \mathbf{B}^{(r^-)} \end{bmatrix} \begin{bmatrix} a_r^+ \\ a_r^- \end{bmatrix} \right) = -a_{\text{I}} \begin{bmatrix} z_0 \mathbf{B}^{(0^+)} \\ \mathbf{0} \end{bmatrix} \quad (\text{A.2})$$

$$\mathbf{B}^{(r)} = \begin{bmatrix} \rho \omega^2 \left[ \xi_r k_t^2 \mathcal{U}_1^{(r)} + \kappa (k_t^2 - 2k_\ell^2) \mathcal{U}_2^{(r)} \right] + i k_\ell^2 k_t^2 \mathcal{P}^{(r)} \\ \rho \omega^2 k_\ell^2 \left( \kappa \mathcal{U}_1^{(r)} + \xi_r \mathcal{U}_2^{(r)} \right) \\ \rho_0 \omega^2 \mathcal{U}_1^{(r)} + i \xi_r \mathcal{P}^{(r)} \end{bmatrix}$$

where  $z_r = e^{i\frac{h}{2}\chi_r}$ . Notice that  $a_{\text{I}}$ , as the source term, has been placed in the right-hand side. The symmetry of the forward-backward waves (given by  $\xi_r = \pm\chi_r$ ) and the symmetry of the solid-fluid interfaces (located at  $x = \pm h/2$ ) are taken into account by the change of variables  $a_r^\pm = a_r^S \pm a_r^A$ , the use of  $c_r = \cos \frac{h}{2}\chi_r$  and  $s_r = \sin \frac{h}{2}\chi_r$  and the vectors  $\mathcal{S}^{(r)} = \mathbf{B}^{(r^+)} + \mathbf{B}^{(r^-)}$  and  $\mathcal{A}^{(r)} = \mathbf{B}^{(r^+)} - \mathbf{B}^{(r^-)}$  with few non-zero coefficients:

$$\begin{aligned} \mathcal{S}_1^{(0)} &= 2i(\chi_t^2 + \kappa^2) \quad , \quad \mathcal{A}_3^{(0)} = 2i\chi_0 \\ \mathcal{S}_1^{(\ell)} &= \mathcal{S}_2^{(t)} = 2\rho(\chi_t^2 - \kappa^2) \quad , \quad \mathcal{S}_3^{(t)} = -2\rho_0\kappa \\ \mathcal{A}_1^{(t)} &= -4\rho\kappa\chi_t \quad , \quad \mathcal{A}_2^{(\ell)} = 4\rho\kappa\chi_\ell \quad , \quad \mathcal{A}_3^{(\ell)} = 2\rho_0\chi_\ell \end{aligned}$$

Altogether, this allows rewriting Eq. (A.2) as two smaller systems of uncoupled equations:

$$\begin{bmatrix} z_0^{-1} \mathcal{S}_1^{(0)} & 2c_\ell \mathcal{S}_1^{(\ell)} & 2c_t \mathcal{A}_1^{(t)} \\ 0 & 2s_\ell \mathcal{A}_2^{(\ell)} & 2s_t \mathcal{S}_2^{(t)} \\ iz_0^{-1} \mathcal{A}_3^{(0)} & 2s_\ell \mathcal{A}_3^{(\ell)} & 2s_t \mathcal{S}_3^{(t)} \end{bmatrix} \begin{bmatrix} a_0^S \\ a_\ell^S \\ a_t^A \end{bmatrix} = \begin{bmatrix} -z_0 \mathcal{B}_1^{(0+)} \\ 0 \\ iz_0 \mathcal{B}_3^{(0+)} \end{bmatrix} a_{\mathbf{I}} \quad (\text{A.3})$$

$$\begin{bmatrix} -iz_0^{-1} \mathcal{S}_1^{(0)} & 2s_\ell \mathcal{S}_1^{(\ell)} & 2s_t \mathcal{A}_1^{(t)} \\ 0 & 2c_\ell \mathcal{A}_2^{(\ell)} & 2c_t \mathcal{S}_2^{(t)} \\ z_0^{-1} \mathcal{A}_3^{(0)} & 2c_\ell \mathcal{A}_3^{(\ell)} & 2c_t \mathcal{S}_3^{(t)} \end{bmatrix} \begin{bmatrix} a_0^A \\ a_\ell^A \\ a_t^S \end{bmatrix} = \begin{bmatrix} -iz_0 \mathcal{B}_1^{(0+)} \\ 0 \\ -z_0 \mathcal{B}_3^{(0+)} \end{bmatrix} a_{\mathbf{I}} \quad (\text{A.4})$$

that can be inverted in order to finally obtain:

$$a_0^S = \frac{z_0^2}{2} a_{\mathbf{I}} \frac{(\alpha c_\ell s_t + \beta s_\ell c_t) - i\gamma s_\ell s_t}{(\alpha c_\ell s_t + \beta s_\ell c_t) + i\gamma s_\ell s_t} \quad (\text{A.5})$$

$$a_0^A = \frac{z_0^2}{2} a_{\mathbf{I}} \frac{\gamma c_\ell c_t - i(\alpha s_\ell c_t + \beta c_\ell s_t)}{\gamma c_\ell c_t + i(\alpha s_\ell c_t + \beta c_\ell s_t)} \quad (\text{A.6})$$

478 where  $\alpha$ ,  $\beta$  and  $\gamma$  are given in Eq. (9).

## 479 Appendix B. Initialization

The expression of the inverse double-transmitted wave transfer given in Eq. (14) is an harmonic polynomial which poles  $(\psi_\ell^{-\omega}, 1, \psi_\ell^\omega)$  can be estimated after compensation of the water-related phase  $\psi_0^\omega$ . More particularly, it is well-known from Prony's works that the data samples  $h_n = \psi_0^{-\omega} H_{2T}^{-1}(n\Delta\omega)$  are related by the following recurrence relation:

$$0 = h_{n+3} + \sum_{m=0}^2 p_m h_{n+m} \quad (\text{B.1})$$

and that the roots of the polynomial  $P(\pi)$  formed by the coefficients  $p_m$  are the signal poles:

$$P(\pi) = \pi^3 + \sum_{m=0}^2 p_m \pi^m = (\pi - \psi_\ell^{-\Delta\omega})(\pi - 1)(\pi - \psi_\ell^{\Delta\omega}) \quad (\text{B.2})$$

Hence the recurrence relation takes the following form:

$$(h_{n+3} - h_n) - p_2(h_{n+2} - h_{n+1}) = 0 \quad (\text{B.3})$$

$$p_2 = (\psi_\ell^{\Delta\omega} + \psi_\ell^{-\Delta\omega} + 1) = 2 \cos \frac{2h\Delta\omega}{v_\ell} + 1 \quad (\text{B.4})$$

480 The coefficient  $p_2$  can be estimated from the normal incidence data  $\mathbf{h} = [h_1 \dots h_N]$  using weighted  
481 least-squares, the chosen weight taking into account the input signal bandwidth. At the end, the initial  
482 frequency-independent longitudinal wave speed is obtained as:

$$v_\ell = \frac{2h\Delta\omega}{\text{acos}\left(\frac{p_2 - 1}{2}\right)} \quad (\text{B.5})$$