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Model calculations for ultrasonic plate–liquid–plate resonators: peak frequency shift by liquid density and velocity variations

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Received 26 September 1996, in final form 3 February 1997, accepted for publication 24 March 1997

Abstract. Ultrasonic resonators—consisting of the liquid sample, enclosed by two planar piezoelectric transducer plates—permit a direct and accurate determination of liquid sound velocities \( c_L \) at kilohertz and megahertz frequencies. Such plate–liquid–plate (PLP) resonators offer a resolution \( \frac{1}{c_L} < 10^{-3} \), which is important for analytical work in chemistry, bio- and physico-chemistry and for some technical applications.

A simple one-dimensional resonator model is derived which permits calculation of the spectrum of the longitudinal (nonharmonic) eigenfrequencies \( f_n \) \((n = 1, 2, 3, 4 \ldots)\) as a function of liquid and resonator parameters. This model obtains the peak \( f_n \) shift by variation of liquid velocity \( c_L \) and density \( \rho_L \) as well for plane wave propagation; the effect from variations in \( \rho_L \) has been neglected in most ultrasonic studies so far.

Caused by a frequency-dependent phase shift for sound reflection at both liquid/transducer interfaces, the differential quotients \( \frac{df_n}{dc_L} \) and \( \frac{df_n}{d\rho_L} \) of the \('real'\) resonator deviate from those of an \('ideal'\) PLP resonator with perfect, \('hard'\) reflection (reflection factor \( \equiv 1 \)) and harmonic overtones \( nf_L \) (\( f_L \) is the liquid fundamental frequency; one half liquid wavelength \( \lambda_L/2 \) equals transducer separation \( x \)).

The figures show typical acoustic impedance and admittance spectra, which are calculated for a model configuration; they illustrate features of the harmonic numbers in the liquid cavity and longitudinal mode counting. Equations are given for the dimensionless, normalized differential quotients \( \frac{df_n}{dc_L} \cdot c_L/f_n \) and \( \frac{df_n}{d\rho_L} \cdot \rho_L/f_n \). Plots demonstrate systematic aberrations from \('ideal'\) resonator behaviour, which can affect high-precision sound velocity measurements.

1. Introduction

Ultrasonic PLP resonators, consisting of two air-backed, planar piezoelectric transducers and the enclosed liquid sample (volume \( \sim 0.1–10 \text{ ml} \)), are a valuable tool for the determination of liquid sound absorption and velocity in the 0.1 to 100 MHz range (Eggers 1967/68, 1992, 1994, Eggers and Funck 1973, Eggers et al 1994, Eggers and Kaatze 1996, Kaatze et al 1987, Labhardt and Schwarz 1976, Sarvazyan 1982, Sarvazyan and Chalikian 1991). Resonator eigenfrequencies \( f_n \) can be obtained with high resolution, particularly in low-loss liquids, by means of amplitude, phase, phase slope or group delay time \( t_g = -\delta \phi/\delta \omega \) (\( \phi \) is the phase, \( \omega \) the angular frequency) measurements. From a series of \( f_n \) values the liquid column fundamental frequency \( f_L = c_L/2x \) and—with the liquid pathlength or transducer separation \( x \) known—the sound velocity \( c_L \) can be determined by nonlinear regression, which also yields the specific acoustic impedance ratio \( z_L/z_T = (\rho_L c_L)/(\rho_T c_T) \) (\( \rho \) is the density) for longitudinal, plane wave propagation in the liquid (L) and the transducer medium (T; \( X \)-cut quartz or \( Y \)-cut lithium niobate in most devices).

If the sound beam diameter \( D \) exceeds the wavelength \( \lambda_L / (D > 50\lambda_L) \), one-dimensional, plane wave equations describe the performance of such PLP resonators to a very good approximation (Eggers 1992). Piezoelectric coupling in the transducer material, given by the electromechanical coupling coefficient \( k_T \) (Kino 1987), affects the eigenfrequencies, including the overtones, of the transducer plates (Onoe et al 1963). The piezoelectric effect can be taken into account by introducing effective eigenfrequencies \( f_{Tm} \), but it causes loading of the cavity due to electromechanical energy coupling, particularly at the transducer fundamental and overtones.
For applications in ultrasonic velocimetry (Sarvazyan 1982, 1991) it is desirable to know the influence of liquid velocity $c_L$ and density $\rho_L$ and of their changes on the eigenfrequencies $f_n$ ($n = 1, 2, 3, \ldots$) quantitatively. This dependence can be obtained from the derivatives $df_n/dc_L$ and $df_n/d\rho_L$ of a characteristic trigonometric equation

$$F(f_n, f_T, x, c_L, \rho_L, c_T, \rho_T) = 0,$$

which permits determination of the fundamental frequency of the liquid column $f_L$ and of the $f_n$ spectrum (Eggers 1967/68, 1992, Eggers and Richmann 1993).

2. Acoustic resonator eigenfrequencies—one-dimensional model

The characteristic acoustical impedance equations for PLP resonators relate $f_n$ to the sample parameters $c_L$, $\rho_L$, $f_L$ and to the corresponding piezotransducer parameters $c_T$, $\rho_T$ (fundamental frequency; plate thickness equals $\lambda_T/2$). Usually the compressional waves in the liquid cavity undergo nearly ‘hard’ reflection due to the higher specific impedance $Z_T > Z_L$ of the transducers, except at the fundamental $f_T$ and the overtones $f_n$, where the wave ‘sees’ the lower backing (air?) impedance. Usually the region near $f_T$ is not employed for velocity and absorption measurements, because enhanced electromechanical energy coupling in the transducers—stronger in LiNbO$_3$ than in quartz—decreases the cavity $Q$, in addition to liquid absorption. It is advisable to refer the nonharmonic $f_n$ series of the PLP resonator and its harmonic numbers $n$ to the harmonic spectrum $n f_L$ of an ‘ideal’ resonator with perfect reflection ($r = 1$). The harmonic number $n = 1, 2, 3, 4\ldots$ of such resonators is approximately the number of half-wavelengths $\lambda_L/2$ in the liquid standing wave field: $n \approx 2x/\lambda_L$. The impulse balance of the total vibrating system is established by the mass of the cavity vessel and frame plus the affixed transducers, being quite large compared to the liquid mass.

In consequence of resonator symmetry, oscillational modes of the compound PLP system have either a particle velocity extremum and a pressure node ($n$ odd) or a particle velocity node and a pressure extremum ($n$ even) in the centre plane of the resonator. Therefore it is sufficient to consider one half of the resonator, consisting of one transducer and half of the liquid column (assumed to be lossless), terminated in the centre plane either by impedance zero ($n$ odd) or by infinite impedance ($n$ even).

The acoustic impedance $Z$ (per unit area) is the complex ratio of sound pressure to particle velocity at a point (or plane) in the sound field (Gooberman 1968). Resonance occurs for reactance compensation at the liquid–transducer interface or the equality and opposite sign for the imaginary parts of transducer impedance $Z_T$ and liquid impedance $Z_L$ (pathlength $x/2$), i.e. $\text{Im}(Z_T) = -\text{Im}(Z_L)$ (Eggers 1967/68). For a plane wave field:

$$Z_T = iz_T \tan \left( \frac{\pi f_n}{f_T} \right)$$

$$Z_L = \left\{ \begin{array}{ll}
+ & iz_L \tan \left( \frac{\pi f_n}{2 f_L} \right) \\
- & iz_L \cot \left( \frac{\pi f_n}{2 f_L} \right)
\end{array} \right\} \left\{ \begin{array}{ll}
n \text{odd} \\
n \text{even}
\end{array} \right\} \quad (2a)$$

It should be noted that the assignment of odd and even $n$ in equations (2a) and (2b) is different from that in some earlier publications, apparently erroneous. Both equations can be combined in one equation, valid for odd and even $n$ as well

$$Z_L = -iz_L \cot \left( \frac{\pi f_n}{f_T} \right) - n \right]. \quad (2c)$$

Equations (1) and (2c) lead to a characteristic equation $Z_T = -Z_L$, which determines the series of $f_n$ for a one-dimensional plane wave PLP resonator

$$Z_T \tan \left( \frac{\pi f_n}{f_T} \right) = -Z_L \cot \left( \frac{\pi f_n}{2 f_L} - n \right). \quad (3)$$

For certain applications peak frequencies close to $f_T/2$, $3f_T/2$, $5f_T/2$ etc. with the transducer thickness being an odd multiple of $\lambda_T/4$, are of particular interest, because the low air backing impedance of the transducer is inverted by a ‘quarter-wavelength-transformation’, resulting in ‘infinite’ impedance behaviour and hard reflection between liquid and transducers.

Equations (1) and (2c) yield the corresponding acoustic admittances

$$A_T = -\frac{i}{Z_T} \cot \left( \frac{\pi f_n}{f_T} \right)$$

$$A_L = \frac{i}{Z_L} \tan \left( \frac{\pi f_n}{2 f_T} - n \right)$$

resulting in a characteristic equation, which is equivalent to equation (3)

$$\frac{1}{Z_T} \cot \left( \frac{\pi f_n}{f_T} \right) = \frac{1}{Z_L} \tan \left[ \frac{\pi f_n}{2 f_L} - n \right]. \quad (6)$$

Here the cases $n = 1$ and $n = 2$ cover all odd and even longitudinal modes. The anharmonicity of the $f_n$ spectrum in the frequency range 2–10 MHz is illustrated in figure 1 for impedance and in figure 2 for admittance, depicting the terms of equations (1) and (2c) and (4) and (5) respectively. The intersections of $-Z_L$ (conjugate complex) with $Z_T$ and $-A_L$ (conjugate complex) with $A_T$ determine the $f_n$ spectrum. It is obvious that impedance relations are preferable at and near the odd transducer harmonics $f_T$, $3f_T$, $5f_T$ etc.—the even harmonics of $f_T$ are sonically inactive—while admittance is advantageous near $f_T/2$, $3f_T/2$, $5f_T/2$ etc. In figure 2 the intersections of $-A_T$ with the abscissa define the harmonics of $f_T$, i.e. the spectrum of an ‘ideal’ resonator with infinitely hard ($z_T = \infty$) reflectors. It appears reasonable to refer the resonances of a ‘real’ resonator to the nearby harmonics $n f_L$. In order to illustrate mode numbering around $f_T$, figure 3 shows an expanded plot of $-A_L$ and $A_T$ between 4 and 6 MHz for water and quartz transducers, now with $f_T = 5100$ kHz assumed for clarity. It reveals, that the $-A_L$ branch ($n = 10$) has two intersections with $A_T$; the same is true for the neighbouring branch ($n = 11$). In this case four consequent peaks $f_{10-1}$ (~4793 kHz), $f_{11-1}$ (~5001 kHz), $f_{10+1}$ (~5208 kHz) and $f_{11+1}$ (~5582 kHz) are observed. Peak $f_{11-1}$ originates from a ‘soft’ reflection with low $Z_T$ at transducer $\lambda_T/2$ resonance. On the other hand, if
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Figure 1. Calculated acoustic impedance $Z$ (per unit area; imaginary part) versus frequency $f$: $Z_T$ for X-cut quartz ($Z_T^{\text{quartz}} = 15 \times 10^6$ kg m$^{-2}$ s$^{-1}$) and for Y-cut lithium niobate ($Z_T^{\text{LiNbO}_3} = 34 \times 10^6$ kg m$^{-2}$ s$^{-1}$) transducers; $f_T = 5000$ kHz. $-Z_T$ for odd (- - -) and even (---) modes in water at pathlength $x/2$; $c_L = 1.5$ km s$^{-1}$, $\rho_L = 10^3$ kg m$^{-3}$, $x = 2.5$ mm, $L = 500$ kHz. Intersections between $Z_T$ and $-Z_T$ branches determine the resonant frequencies $f_n$.

Figure 2. Calculated acoustic admittance $A$ (per unit area, imaginary part): $A_T$ (quartz and lithium niobate) and $-A_L$ for odd and even modes in water. Curve types and parameters as in figure 1. Intersections of $A_T$ and $-A_L$ branches determine $f_n$ values. Harmonic numbers $n$, given at the top of the plot, are characteristic by abscissa crossings points $A_{L,n} = 0$ and refer $f_n$ series to harmonic spectrum $n f_L$ of an ‘ideal’ resonator with ‘hard’ reflection ($Z_T = \infty$).

$\quad f_T < 10 f_L$ is assumed, the sequence around $f_T$ changes to $f_\ldots f_{10} f_{9} f_{8} f_7 f_6 f_5 f_4 f_3 f_2 f_1 f_0$. Model parameters used are listed in the captions. Liquid loss is always neglected and one-dimensional, plane wave propagation is assumed.

From the implicit admittance equation

$$F = \frac{1}{\rho c_T} \cot \left( \frac{\pi f_n}{f_T} \right) \frac{d f_T}{c_L} + \frac{1}{\rho_L c_L} \tan \left( \frac{\pi}{2} \frac{f_n}{c_L/2x} - n \right) = 0 \quad (7)$$

the differential quotients $d f_n/d c_L$ and $d f_n/d \rho_L$ are obtained, using common mathematical rules for the differentiation of implicit functions (e.g. Korn and Korn 1968)

$$\frac{d f_n}{d c_L} = \frac{\partial F}{\partial c_L} / \partial f_n \quad \frac{d f_n}{d \rho_L} = - \frac{\partial F}{\partial \rho_L} / \partial f_n \quad (8), (9)$$

leading—after some manipulation—to

$$\frac{d f_n}{d c_L} = \frac{f_n}{c_L} \left[ 1 + \frac{2 z_L f_T}{f_n} \right] \left[ \frac{\pi}{2} \left( \frac{f_n}{f_L} - n \right) \right]^2 \times \left[ \sin \left( \frac{\pi f_n}{f_T} \right) \right]^{-2} \quad (10)$$

the second factor being dimensionless, and to

$$\frac{d f_n}{d \rho_L} = \frac{f_n}{\rho_L} \frac{1}{\pi} \left[ \frac{\pi}{2} \left( \frac{f_n}{f_L} - n \right) \right]^2 \times \left[ \sin \left( \frac{\pi f_n}{f_T} \right) \right]^{-2} \quad (11)$$

For an ‘ideal’ resonator ($r = 1$) the spectrum $f_n^{\text{ideal}} = n f_L = n c_L/2x$ is harmonic; in this case is $d f_n^{\text{ideal}}/d c_L = n / 2x = f_n/c_L$, a commonly used approximation for equation (10); on the other hand $d f_n^{\text{ideal}}/d \rho_L \equiv 0$ is in clear contrast to equation (11).

For a plot of equations (10) and (11) numerical $f_n$ values ($n = 4 \ldots 20$) have been calculated directly from equation (7). Another proven way to obtain numerical values of the $f_n$ series is by iteration, starting from $f_n^{(0)} = n f_L$ and using the relation, which results from equation (7)

$$f_n^{(m+1)} = f_L \left\{ n + \frac{2}{\pi} \tan^{-1} \left[ \frac{z_L}{z_T} \cot \left( \frac{\pi f_n^{(m)}}{f_T} \right) \right] \right\} \quad (12)$$

However, in pursuing this way convergence might fail near $f_T$ and its harmonics.

Figure 4 shows a plot of the dimensionless $(d f_n/d c_L) \times (c_L/f_n)$, equivalent to $d (\ln f_n)/d (\ln c_L)$, for the calculated $f_n$ values between 2 and 10 MHz. The $f_n$ points ($n = 4 \ldots 20$) represent even and odd acoustic modes in the water-filled cavity ($f_L = 500$ kHz), enclosed by transducers from X-cut quartz (specific impedance value $Z_{X-\text{quartz}} = 15 \times 10^6$ kg m$^{-2}$ s$^{-1}$) and Y-cut LiNbO$3$ ($Z_{\text{LiNbO}_3} = 34 \times 10^6$ kg m$^{-2}$ s$^{-1}$). In a similar way the differential
first sight it might be surprising, that even at transducer ‘λT/4 frequencies’ \(f_T/2, 3f_T/2, 5f_T/2\ldots\), the maximum of \(\frac{df_n}{dc_n}\times c_L/f_n\) is slightly below 1. This is quantified by an approximation of equation (10) at those frequencies

\[
\frac{df_n}{dc_L} \approx 1 - 2z_L f_n/
\]

yielding 0.98 at 7.5 MHz for a water-filled quartz resonator (\(z_L/z_T = 0.1; f_L = 0.5\) MHz; \(f_T = 5\) MHz). This is reasonable because the liquid velocity \(c_L\) affects both \(f_L\) and \(Z_L\), shifting \(n f_L\) and the reflection phase in an opposing way. On the other hand the maximum value of \(\frac{df_n}{dc_L} \times c_L/f_n\) becomes closer to 1 for resonators with increased pathlength \(x\) and in consequence lower \(f_L\), because fewer reflections will then occur in the cavity. Please note that near \(f_T\) and \(2f_T\) the values \(\frac{df_n}{dc_L} \times c_L/f_n\) fall below 0.6 and are not shown in figure 4 with its expanded ordinate scale.

The effect from density variations is minimized near the ‘λT/4 frequencies’ \(f_T/2, 3f_T/2, 5f_T/2, \ldots\), but is zero only exactly at those. Figure 5 shows that the density influence increases in regions below and above \(f_T\) and its overtones; here liquid density can be determined by ultrasonic measurements via the specific impedance ratio \(z_L/z_T\). High-resolution measurements of velocity (\(|\delta f_n|/f < 10^{-6}\)) aiming at adiabatic compressibility values (Sarvazyan 1991), have to take density into account, since both liquid velocity and density affect the \(f_n\) spectrum.

A reliable, proven way for velocity evaluation, including density effects, is to measure a series of 3 to 11 (and possibly more) subsequent \(f_n\) values around the ‘λT/4 frequencies’ \(f_T/2, 3f_T/2, 5f_T/2, \ldots\). Nonlinear regression, applying equation (6) and a LEVENBERG–MARQUARDT algorithm (command ‘minerr’ in MATHCAD®), helps to identify the harmonic numbers \(n\) and obtains both \(f_L\) and \(z_L/z_T\). Assuming plane wave propagation at sufficiently high frequencies, an extra check of measured \(f_n\) values and \(n\) is to apply equation (6) again and calculate a series of individual liquid fundamental \(f_{L,n}\) values:

\[
f_{L,n} = f_n \left\{ n + \frac{2}{\pi} \tan^{-1} \left[ \frac{z_L}{z_T} \cot \left( \frac{\pi f_n}{f_T} \right) \right] \right\}^{-1}
\]

which should be constant and close (within a few Hz and less for 3 to 5 peaks) to the \(f_L\) value obtained by regression. This has been verified in many experiments near 7.5 and 12.5 MHz. For the evaluation of resonant peaks with accompanying spurious ‘satellites’ further precautions are advisable: a regression procedure should be preceded by an algebraic separation of superposing acoustic modes and of eventual electromagnetic crosstalk (Eggers 1992). Peak frequencies can be ‘pinned’ by means of the: (a) amplitude maximum; (b) algebraic fit of sampled amplitude values of a total peak; (c) algebraic fit of sampled phase values of a total peak; (d) maximum of group delay time (phase slope); (e) algebraic fit of sampled group delay time values of a total peak. The method to be preferred depends on the available equipment. The evaluation of a total peak as in (b) or in (c) appears to be more accurate than the search for a rather ‘flat’ amplitude top. Methods (d) and (e)

Figure 4. Calculated normalized differential quotient \(\frac{df_n}{dc_L} × \frac{c_L}{f_n}\) displayed in figure 5 for quartz and for LiNbO_3 transducers. Other parameters as in figure 1.

Figure 5. Calculated normalized differential quotient \(\frac{df_n}{dc_L} × \frac{\rho_L}{f_n}\) displayed in figure 5 for quartz and for LiNbO_3 transducers. Other parameters as in figure 1.

3. Discussion and conclusions

The plots demonstrate that liquid density as well as velocity shifts cause typical resonant peak shifts in ultrasonic PLP resonators, differing from ‘ideal’ resonator behaviour and depending on frequency and on transducer parameters. At

\[
\text{df}_{\text{L,n}} = f_n \left\{ n + \frac{2}{\pi} \tan^{-1} \left[ \frac{z_L}{z_T} \cot \left( \frac{\pi f_n}{f_T} \right) \right] \right\}^{-1}
\]
gain from the elimination of a base line phase shift caused by electrical connections, but require more sophisticated electronic equipment.

In general ultrasonic resonator measurements are affected by several sources of error. One has to realize the practical limits of accuracy, which are given primarily by sound field diffraction, by liquid absorption and, last but not least, by temperature fluctuations in the liquid cavity. For practical applications these model calculations should be seen in the frame of the total accuracy obtainable today for high-precision acoustic measurements in liquids.

Acknowledgments

Thanks go to Professor Leo De Maeyer for stimulating discussions and to Dr Udo Kaatze and Mr Kurt-Helmut Richmann for valuable advice.

References

Eggers F 1967/68 Eine Resonatormethode zur Bestimmung von Schallgeschwindigkeit und Dämpfung an geringen Flüssigkeitsmengen Acustica 19 323–9
——1992 Ultrasonic velocity and attenuation measurements in liquids with resonators, extending the MHz frequency range Acustica 76 231–40
——1994 Analysis of phase slope or group delay time in ultrasonic resonators and its application for liquid absorption and velocity measurements Acustica 80 397–405
Eggers F and Funck Th 1973 Ultrasonic measurements with milliliter liquid samples in the 0.5–100 MHz range Rev. Sci. Instrum. 44 969–77
Eggers F and Richmann K-H 1993 Ultrasonic absorption measurements in liquids above 100 MHz with continuous waves, employing algebraic crosstalk elimination Acustica 78 27–35
Gooberman G L 1968 Ultrasonic (London: English University Press)
Labhardt A and Schwarz G 1976 A high resolution and low volume ultrasonic resonator method for fast chemical relaxation measurements Ber. Bunsenges. 80 83–92
Sarvazyan A P 1982 Development of methods of precise ultrasonic measurements in small volumes of liquids Ultrasonics 20 151–4
Sarvazyan A P and Chalikian T V 1991 Theoretical analysis of ultrasonic interferometer for precise measurements at high pressures Ultrasonics 29 119–24