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Behavior of ultrasounds crossing perfluorocarbon liquids and random propagation times

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ABSTRACT

Random propagation times are able to model waves attenuation and velocity. It is true for electromagnetic waves (light, radar, guided propagation) and also for acoustics and ultrasounds (acoustics for high frequencies). About the latter, it can be shown that stable probability laws are well-fitted for frequencies up to dozens of megahertz in numerous cases. Nowadays, medical applications are performed using propagation through perfluorocarbon (PFC). Experiments were done to measure attenuations and phase changes. Using these results, this paper addresses the question to know if stable probability laws can be used to characterize the propagation of ultrasounds through PFC liquids.

1. Introduction

Perfluorocarbons (PFC) are molecules characterized by their high ability to transport oxygen and other gases, their biological and chemical inactivity and the lack of affinity with water and many liquids. Experiments have shown that the metabolism of rats could be sustained by inhalation of PFC, which has given great hopes for surgery and other domains outside medecine. For instance, PFC liquids are currently used in eye surgery [1]. PFC emulsions allow molecular imaging, targeted therapeutics and blood substitutes [2–4]. Detailed experiments and simulations are available about parameters of acoustic propagation for PFC liquids and emulsions [5–7].

This paper addresses a model about the propagation of ultrasounds (acoustics in high frequencies) through PFC liquids. It is based on results given in the paper [8] of Strohm and Kolios. The latter have developed photoacoustic measurements for three kinds of PFC liquids, after validation of devices by measurements holding on well-studied liquids for a long time, water, ethanol and castor oil. Measured attenuations show a power law frequency dependence with exponents between 1 and 2. Velocities are constant for water and ethanol, and are functions slowly increasing for other liquids particularly for PFC liquids. Results on water and ethanol are in good accordance with literature but differences exist about castor oil (Table 1 in [8]).

The refraction index of a homogeneous medium depends on the repartition of molecules and on the corresponding fields. Because molecules move, little and random variations of the celerity around a mean value are inescapable and random variations of propagation times follow. This allows to model propagation of acoustic and ultrasonic waves [9,10], and also propagation of electromagnetic waves in coaxial cables, power cables, radar and others [11–13]. A propagation time is the sum of the propagation times corresponding to parts of the crossed medium. In a random context, this means that a version of the central limit theorem has to be applied. When the two first statistical moments are defined, the Gaussian probability law is the solution. Otherwise, stable probability laws [14,15] provide the good tools for modeling [10,12]. The following section gives a summary of technics which we will try to apply in Section 3 to liquids studied in [8].

2. Random propagation times and stable probability laws

2.1. Random propagation times

The random propagation time $\mathbf{A} = \{A(t), t \in \mathbb{R}\}$ transforms the monochromatic wave $\exp[i\omega_0 t]$ into the random process

$$Z(t) = \exp[i\omega_0(t - A(t))]. \tag{1}$$

When the probability laws of $A(t)$ and $A(t) - A(t-\tau)$ do not depend on t, the following decomposition is available (see Appendix A)

$$Z(t) = \psi(\omega_0)e^{i\omega_0 t} + V(t) \tag{2}$$

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where

$$\begin{aligned} &\psi(\omega) = \mathrm{E}\left[e^{-i\omega A(t)}\right] \\ &\phi(\omega,\tau) = \mathrm{E}\left[e^{-i\omega(A(t)-A(t-\tau))}\right] \\ &\mathrm{E}[V(t)V^*(t-\tau)] = \left[\phi(\omega_0,\tau) - |\psi(\omega_0)|^2\right]e^{i\omega_0\tau} \end{aligned} \tag{3}$$

E[...] and the superscript * stand for the ensemble mean (the mathematical expectation) and the complex conjugate. ψ and ϕ are characteristic functions (in the probability calculus sense) and define the probability laws of the random variables (r.v.) $A(t)$ and $A(t) - A(t-\tau)$ (they are assumed independent of t). The random process **A** takes into account variations of the refractive index of the medium due to random motions of molecules or variations of electromagnetic fields. The process **V** defined by (2) summarizes losses and the power of **Z** remains equal to 1. When a monochromatic wave is transmitted at some frequency $\omega_0/2\pi$, the receiver is matched to this frequency, and is constructed to cancel noises, for instance with a bandpass filter at its input. Then devices look at the harmonic part $\psi(\omega_0)e^{i\omega_0 t}$, and neglect the second part **V** because its (continuous) power spectrum is too widened (see the Appendix A and [9,10]).

2.2. Stable probability laws

When the medium can be split into sequences of independent pieces with invariant properties, the Gaussian law is a natural model provided the existence of statistical moments. In this case, $A(t)$ is a sum of an arbitrary number of random variables and the central limit theorem can be applied [16]. Stable probability laws appear as a generalization of the central limit theorem when the second order moment does not exist. $A(t)$ follows a stable probability law when (neglecting the case $b = 1$) [14,15]

$$\psi(\omega) = \exp\left[-im\omega - a\omega^b\left(1 - i\beta\tan\frac{\pi b}{2}\right)\right] \tag{4}$$

for $\omega > 0$, $\psi(\omega) = \psi^*(-\omega)$ and with real parameters verifying

$a > 0, \quad 0 < b \leqslant 2, \quad |\beta| \leqslant 1.$

Stable probability laws are defined by a characteristic function ψ rather than a probability density, because the latter has a simple analytic form only for three values of b (2, 1, 1/2 respectively for Gaussian, Cauchy and Levy probability laws). As explained above, $\psi(\omega_0)e^{i\omega_0 t}$ is the medium answer to $e^{\omega_0 t}$ (the noises being neglected). The modulus $|\psi(\omega_0)|$ provides the wave attenuation, and the argument $\mathrm{Arg}\psi(\omega_0)$ provides the transmission delay and then the celerity. Then, the attenuation att(f) and the velocity $v(f)$ at frequency $f = \omega/2\pi$ are defined by

$$\begin{aligned} &\mathrm{att}(f) = -20\log\exp\left[-a\omega^b\right] = \alpha f^b \\ &v(f) = \left(m - a\beta(2\pi f)^{b-1}\tan\frac{\pi b}{2}\right)^{-1} \end{aligned}$$

and we have:

$$\begin{aligned} &\frac{\Delta v}{v(f_0)\,v(f_1)} = 0.0183\alpha\beta\left[f_1^{b-1} - f_0^{b-1}\right]\tan\frac{\pi b}{2} \\ &\alpha = 8.686a(2\pi)^b \end{aligned} \tag{5}$$

where Δv is the variation of the velocity between f_0 and f_1. Because $b > 1$ in [8], the velocity at low frequency will be equal to m^{-1}. When $\beta = -1$ ($\tan\frac{\pi b}{2} < 0$), we retrieve the Szabo's model based on the Kramers–Krönig relations [18,19].

In the signal theory context, the medium appears to devices as a Linear Invariant Filter (LIF) with complex gain $\psi(\omega)$ [16] (when the "noisy part" **V** is neglected). The LIF impulse response is the Fourier transform $g(t)$ of $\psi(\omega)$. The LIF is "causal" when $g(t) = 0$ for $t < 0$. This means that the answer to an impulse at the time t_0 begins after t_0. For $b > 1$ (it is the case in this paper), this condition is never fulfilled [15], but the best possible value is obtained for $\beta = -1$, where $g(t)$ is the most asymmetric possible. Actually, we can consider that the propagation time is close to m (its mean value), which is large enough to verify the condition $Pr[A(t) < 0] \simeq 0$. The value $\beta = -1$ appears the most often, particularly in ultrasonics [10,12].

3. Application to perfluorocarbon liquids

3.1. Used devices

The apparatus used by authors of [8] were calibrated using experiments in water, ethanol and castor oil. For water and ethanol, $b = 2$ and then $\tan\frac{\pi b}{2} = 0$ (the velocity v does not depend on f). Usual tables give results in good accordance with experiments. In both cases, the value of β is indifferent (because multiplying by $\tan\frac{\pi b}{2} = 0$). For castor oil, the measured attenuation is

$$\mathrm{att}(f) = 0.553f^{1.59} \tag{6}$$

From (5), and with $\alpha = 0.553,\ b = 1.59$:

$$\frac{\Delta v}{v(f_0)\,v(f_1)} = 0.01\alpha\beta\left[f_1^{0.59} - f_0^{0.59}\right]\tan 0.795\pi. \tag{7}$$

From Fig. 2A of [8] at $f_0 = 200$ MHz and $f_1 = 700$ MHz, $\Delta v = 45$ is an admissible value which implies $\beta = -1$. But $\beta = -1.18$ for $\Delta v = 53$ (Table 1 in [8]), value which is a little too large in the framework of stable laws ($|\beta| \leqslant 1$). β and Δv are proportional (with coefficient of proportionality in the order of 1), and the accuracy of measurements of Δv seems weak.

Fig. 5a and b of [17] give

$$\mathrm{att}(f) = 0.42f^{1.69} \tag{8}$$

in a lower frequency band and at 25 °C, which leads to $\beta = -1$ when $\Delta v = 60$.

To summarize, devices used in [8] provide results rather close to previous measurements of well-studied liquids, water, ethanol, and castor oil, in the frequency band (200 MHz, 600 MHz). Nevertheless, Fig. 2B in [8] shows inaccurate values of attenuation outside this interval.

3.2. Global study

Authors of [8] study three liquids denoted for simplicity

P_1 = perfluorohexane
P_2 = perfluoroheptane
P_3 = FC-77.

The authors adopt the same attenuation formula for the three liquids fitted on the whole cloud of data

$$\mathrm{att}(f) = 0.352f^{1.56}. \tag{9}$$

Consequently, (5) becomes ($\alpha = 0.352, b = 1.56$)

$$\frac{\Delta v}{v(f_0)\,v(f_1)} = 0.0064\beta\left[f_1^{0.56} - f_0^{0.56}\right]\tan 0.78\pi. \tag{10}$$

Table 1 in [8] gives following values of $v(200)$, $v(700)$ and Δv the value in m/s (f in MHz)

	$v(200)$	$v(700)$	Δv
P_1	480	484	4
P_2	516	521	5
P_3	557	566	9

(11)