STUDY, DEVELOPMENT, AND IMPLEMENTATION OF ANALYSIS TECHNIQUE OF BIPHASIC ATTENUATION SYSTEMS USING ULTRASOUND

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Abstract − These papers describe the first set of experiment conducted to validate a measurement system for physical-chemistry analysis of fluids using ultrasound as principle. Frequencies of 2.25, 3.00, and 3.50 MHz were used, with the emission-reception technique with a pair of transducers. Experimental results showed excellent agreement with the theory for attenuation of glycerol at 99.5% of concentration. The system is able to be used in other liquids, being biofuels particularly of interest for this project.

Keywords: ultrasound, biphasic systems, measurement uncertainty.

1. INTRODUCTION

Ultrasound is a mechanical wave way of energy propagation, with frequencies above 20 kHz. In general, sonochemistry applications use low frequency ultrasound, up to about 100 kHz. Industrial applications, such as acoustic emission and other related to non-destructive testing (NDT) may use ultrasound up to 1 MHz. In health care (treatment and diagnosis) typically uses ultrasound from 1 to 5 MHz, and even up to 100 Hz for biomicroscopy.

Ultrasound has been used frequently and many different steps in a chemical process, for instance, reaction velocity [1], components separation [2], and even in its identification and analysis [3]. Other related activities, such as flow measurement, had also been undertaken though ultrasound as physical principle. However, considering a metrological point of view, there is a huge field of development and improvement, what is fundamental to fulfill in order of stablishing a base to prove cientifcly advantages of using ultrasound in sonochemistry and control of chemistry processes.

There some papers been published, such as [1], [2], and [3], but attenuation coeficient and soud velocity are usually expressed as na average value, and their "uncertainty" are expressed as simply as a standard deviation. Uncertainty, as widely considered consensus after [4], has not been evaluated as proposed herein for the measurand under investigation. For other measurand, uncertainty is already considered for rules to decide if a measurement assures conformity or nonconformity of items, for instance as [5].

This work aims to express uncertainty of measurement of attenuation coefficient as first step of a development of biphasic systems to be applied, for instance, in biofuel analysis.

2. MATERIAL AND METHODS

2.1. Experimental setup

Two ultrasound transducers pairs were used in the experiments, both with 25.4 mm of nominal diameter. Their nominal center frequencies were 2.25 MHz and 3.50 MHz.

For the 2.25 Hz transducer pair, samples of materials used to measure attenuation were inserted in a cylinder with 40 mm of axial dimension and with 64 mm of internal diameter. It was excited with a 25 cycles burst signal in 2.25 Hz.

For the 3.50 Hz transducer pair, the cylinder used has 22 mm of length and with 64 mm of internal diameter. It was excited with 25 cycles burst signals of both 3.50 MHz and of 3.00 MHz.

In both case, the containers (cylinders) were sealed with a PVC membrane in one of their end and positioned with their symmetry axis vertically.

As reference, it was used fresh water as sample, and the testing sample was glycerol with nominal concentration of 99.5%. This testing material was chosen to calibrate the system because their physical-chemical properties are well known and present in the technical literature.

Measurements were undertaken in two steps for each different system configuration, using emission-reception technique. In the fist step, water was used as reference, and it was used to fill the cylinder positioned between each pair of transducer, one of them used to emit (output) the

ultrasound, and the other used to receive the signal after transmission through the media. In the second step, water was replaced by glycerol (99.5%), which sample was positioned in between the same pair of transducers and at the same distance, using the same signal as excitation. Figure 1 discloses the experimental setup illustratively (the cylinder is positioned horizontally in this figure, despite it was actually positioned vertically).

All measurements (water and glycerol in sequence) were repeated 5 times, and the system was dismounted and settled again in between repetitions.
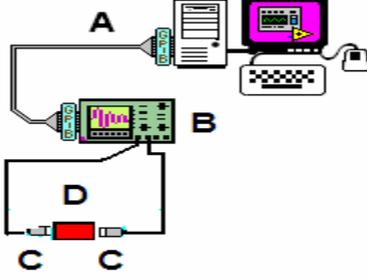
Fig. 1. Experimental setup for attenuation measurement:
A → Computer with automation software; B → Digital scope;
C → Ultrasound transducers (emission and reception);
D → Material sample to measure attenuation.

2.2. Theoretical formulation

Attenuation was assessed in two ways: using a "theoretical approach" and an "experimental approach". The system validation was done comparing, in a statistical base, both approaches. Some of the parameters used to both approaches were obtained in the literature, whilst others were determined empirically.

Equation 1 was used in the "theoretical approach".

$$AT_T = \frac{20.\log[\exp(x \cdot \alpha_T)]}{x} \text{ [dB/cm]} \tag{1}$$

where x [cm] is the distance between the pair of transducer and

$$\alpha_T = \frac{(2\pi \cdot f)^2}{2 \cdot \rho_A c_A^3}\left(\frac{4\eta}{3} + \eta_B\right) \text{ [cm}^{-1}\text{]} \tag{2}$$

In eq. 2, f [MHz] is the output frequency, η [mPa.s] is the dynamic viscosity and η_B [mPa.s] is the bulk viscosity, ρ_A [kg/m^3] is the density of the sample and c_A [m/s] is the sound velocity for the analysed material.

Equation 3 was used in the "experimental approach", were V_0 [V] is the amplitude of the measured singal after transmission through the reference sample (water), V [V] is the amplitude measured after transmission through the measured sample (glycerol), and x is as in eq. 1.

$$AT_E = \frac{20\log\left(\frac{V_0}{V}\right)}{x} \text{ [dB/cm]} \tag{3}$$

2.3. Statistical analyses

First of all, test F (Fisher) was applied to verify homogeny between the variances. Accordingly to test F, two samples (set of measurements) can be consider consistent if

$$F_{cal} < F_{tab} \tag{4}$$

where $F_{cal} = s_1^2 / s_2^2$ (s_1 and s_2 are standard deviations of each sample of measurements) and F_{tab} is the tabled value for the F distribution, considering a determined coverage probability (typically 95%). The number of degrees of freedom shall be considered for both sets of measurements.

If the sets of measurements are considered consistent accordingly to their variance, one can combine their standard deviation as following:

$$s_c = \sqrt{\frac{\sum\limits_{i=1}^{N}(\upsilon_i s_i^2)}{\sum\limits_{i=1}^{N}\upsilon_i}} \tag{5}$$

where s_c is the combined standard deviation of N sets of measurements, υ_i is number of degrees of freedom for the i-th measurement set, and s_i is the standard deviation for the i-th measurement set.

Considering a pair of measurements set that can be combined accordingly to eq. 5, i.e, if they fulfill the requirement of eq. 4, so their means ($\overline{x_1}$ and $\overline{x_2}$) can be considered statistically identical if superior and inferior limits of the interval assessed for test $t_{\upsilon,p}$ (eq. 6) includes the value 0, considering a determined coverage probability p (typically 95%).

$$\left|\overline{x_1} - \overline{x_2}\right| \pm t_{\upsilon,p} \cdot \sqrt{\frac{n_i + n_2}{n_i \cdot n_2}} \cdot s_c \tag{6}$$

where $\upsilon = n_i + n_2 - 2$, n_i and n_2 are the number of measurements for both sets, and s_c is assessed by eq. 5.

2.4. Uncertainty assessment

To calculate the overall uncertainty for AT_T, eq. 1 and 2 were combined and simplified. Firstly, accordingly to general theory, $\eta_B = 1.03.\eta$. So, an equivalent viscosity η_E was defined as

$$\eta_E = \frac{4}{3}\eta + \eta_B \text{ [mPa.s]} \tag{7}$$

what leads to $\eta_E = \frac{4}{3}\eta + 1.03.\eta = \frac{7.09\eta}{3}$. Substituting this value into eq. 2, and then in eq. 1, after some simplifications, it could be rewritten as:

$$AT_T = \frac{20 \cdot 4\pi^2 \cdot 7.09 \cdot \log e}{2 \cdot 3} \cdot \frac{f^2 \cdot \eta}{\rho_A c_A^3} \quad (8)$$

Defining the constant $c_0 = \dfrac{40\pi^2 \cdot 7.09 \cdot \log e}{3}$, eq. 8 becames

$$AT_T = c_0 \cdot \frac{f^2 \cdot \eta}{\rho_A c_A^3} \quad (9)$$

The partial derivatives of each parameter in eq. 9 result in $\dfrac{\partial AT_T}{\partial c_0} = C_{c_0} = \dfrac{f^2 \cdot \eta}{\rho_A c_A^3}$, $\dfrac{\partial AT_T}{\partial f} = C_f = 2 \cdot c_0 \dfrac{f^3 \cdot \eta}{\rho_A c_A^3}$,

$\dfrac{\partial AT_T}{\partial \eta} = C_\eta = c_0 \dfrac{f^2}{\rho_A c_A^3}$, $\dfrac{\partial AT_T}{\partial \rho_A} = C_{\rho_A} = -c_0 \dfrac{f^2 \cdot \eta}{\rho_A^2 c_A^3}$,

and $\dfrac{\partial AT_T}{\partial c_A} = C_{c_A} = -3 \cdot c_0 \dfrac{f^2 \cdot \eta}{\rho_A c_A^4}$. As the uncertainty for c_0 equals 0 ($\mu_0 = 0$), the combined uncertainty for AT_T is

$$\mu_{\alpha_T}^2 = C_f^2 \cdot \mu_f^2 + C_\eta^2 \cdot \mu_\eta^2 + C_{\rho_A}^2 \cdot \mu_{\rho_A}^2 + C_{c_A}^2 \cdot \mu_{c_A}^2 \quad (10)$$

In eq. 10, uncertainty for each parameter is expressed as μ with an index corresponding to the quantity considered.

For the "experimental approach" (eq. 3), the combined uncertainty for AT_E is

$$\mu_{AT_E}^2 = C_x^2 \cdot \mu_x^2 + C_{V_0}^2 \mu_{V_0}^2 + C_V^2 \cdot \mu_V^2 \quad (10)$$

where $C_x = \dfrac{\partial AT_E}{\partial x} = -1 \cdot \dfrac{20 \log\left(\frac{V_0}{V}\right)}{x^2}$,

$C_{V_0} = \dfrac{\partial AT_E}{\partial V_0} = \dfrac{20}{x \cdot V_0 \cdot \ln(10)}$, and $C_V = \dfrac{20}{x \cdot V \cdot \ln(10)}$.

3. RESULTS

Using parameters as defined in [6], results for the "theoretical approach" are presented in table 1. The sample was glycerol in concentration of 99.5%. Viscosity depends on the bath temperature, and is corrected for it.

For "experimental approach", in table 2 are disclosed the results for all measurements for each frequency. Temperatures varied from 19ºC and 23º throughout all measurements for both water and glycerol.

Regarding the homogeneity of measurements variances, experimental and theoretical approaches were tested accordingly to eq. 4, and results are presented in table 3 (95% of coverage probability). Applying a coverage interval of 95% of probability, the means were tested accordingly to eq. 6, and results are presented in table 4.

In tables 5 and 6, the combined uncertainties for both approaches are disclosed for each tested frequency.

Table 1. Attenuation measurement for the "theoretical approach".

Freq. [MHz]	x [cm]	ρ_A [kg/m³]	c_A [m/s]	η [mPa.s]	α_T [m⁻¹]	AT_T [dB/cm]
2.25	3.9	1261	1884	1.235	34.18	2.97
	4.0	1261	1899	1.235	33.38	2.90
	4.1	1261	1889	1.200	32.93	2.86
	4.0	1261	1894	1.259	34.29	2.98
	4.0	1261	1893	1.235	33.69	2.93
					Mean [dB/cm]	**2.93**
3.00	2.13	1261	1896	1.112	53.69	4.66
	2.13	1261	1896	1.112	53.61	4.66
	2.12	1261	1894	1.080	52.27	4.54
	2.14	1261	1895	1.060	51.20	4.45
	2.14	1261	1896	1.070	51.63	4.48
					Mean [dB/cm]	**4.56**
3.50	2.12	1261	1893	1.144	75.55	6.56
	2.12	1261	1897	1.177	72.22	6.71
	2.12	1261	1896	1.177	77.37	6.72
	2.14	1261	1896	1.133	74.44	6.47
	2.14	1261	1899	1.122	73.40	6.38
					Mean [dB/cm]	**6.57**

Table 2. Attenuation measurement for the "experimental approach".

Frequency [MHz]	x [cm]	V_0 [V]	V [V]	AT_E [dB/cm]
2.25	0.039	4.60	1.26	2.88
	0.040	4.78	1.29	2.81
	0.041	4.75	1.16	3.01
	0.040	4.87	1.27	2.89
	0.040	4.68	1.27	2.87
			Mean [dB/cm]	**2.89**
3.00	0.0212	0.814	0.262	4.59
	0.0212	0.802	0.2772	4.31
	0.0212	0.801	0.277	4.30
	0.0214	0.812	0.271	4.44
	0.0214	0.812	0.27	4.45
			Mean [dB/cm]	**4.42**
3.50	0.0213	1.12	0.2401	6.30
	0.0213	1.13	0.2372	6.38
	0.0214	1.13	0.243	6.29
	0.0214	1.12	0.249	6.10
	0.0214	1.15	0.252	6.15
			Mean [dB/cm]	**6.24**

Table 3. Test F for the homogeneity of variances for the "experimental" and the "theoretical" approaches (coverage probability of 95%).

Frequency [MHz]	F_{cal}	F_{tab} (p = 0.95; $\upsilon_1 = \upsilon_2 = 4$)
2.25	2.079	
3.00	1.599	5.05
3.50	1.478	

Table 4. Test *t* to compare the means of the "experimental" and the "theoretical" approaches (coverage probability of 95%).

Frequency [MHz]	Superior limit	Inferior limit
2.25	0.171	-0.240
3.00	0.492	-0.212
3.50	0.689	-0.044

Table 5. Combined (not expanded) attenuation uncertainties assessed for the tested sample, i.e., glycerol at 99.5% of concentration for both "experimental" and the "theoretical" approaches.

Frequency [MHz]	Experimental [dB/cm]	Theoretical [dB/cm]
2.25	0.22	0.099
3.00	0.47	0.050
3.50	0.40	0.083

4. DISCUSSION

Table 1 and 2 depict all 5 measurements (repetitions) performed for each pair of transducers and different frequencies. The final result is the attenuation, in dB/cm for the testing sample, i.e., glycerol at 99.5% of nominal concentration. Reference sample was water for the "experimental approach", as disclosed in table 2. In a quick analysis, attenuation increases with frequency, what is expected. There are not enough data to perform a curve fit relating frequency and attenuation, and the range of frequency evaluated is not large enough either. Further testes will be performed in a wider frequency range to establish a mathematical empirical relation between frequency and attenuation in glycerol.

Statistical analyses were performed to validate the results. Table 3 and 4 show the results, witch were really good. The variances were considered homogeneous enough (under 95% coverage probability) and could be combined. The means were statistically identical (p = 95%) as evident after table 4.

Finally, uncertainties for both approaches are disclosed in table 5. Despite they are shown in dB, it is not difficult to convert it in percentage, and one can find out combined uncertainties less than 5% for the experimental approach, and less than 1% for the theoretical one. Detailed analyses are not shown in this paper, but great contribution for those uncertainties arises from the scope uncertainty (about 2%) for voltage measurements. And substantial improvement could be done if a more accurate system to measure voltage was used.

Another remark regarding uncertainties in table 5, attenuation grows with the power of 2 (or more) of frequency. That is way it was necessary to use a small cylinder for 3.00 and 3.50 MHz. Even so, the measured voltage was about hundred of mV, what increased even more the combined overall uncertainty (see table 2). It is not the case in the theoretical approach as voltage is not one of the computed quantities in eq. 1 (or 9).

3. CONCLUSION

The measurement system developed for this experiment can be considered validated, accordingly to the existing theory. A natural improvement and further work is to extend the frequency range using glycerol as testing material, and use the so far valildated method with other materials, as biofuel, for instance.

This work is part of a large project of research and development at Inmetro, and its main goal is to evaluate the use of ultrasound as a metrological tool to physical-chemistry analyses of fluids.

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