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Self-normalized front photopyroelectric technique for thermal effusivity measurements in liquids

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ABSTRACT

In this paper a measurement methodology is proposed to avoid some limitations of conventional front photopyroelectric technique for thermal effusivity measurements, related to the necessity of calibration using a reference sample. The same pyroelectric sensor heated at high frequencies acts as a reference sample. The proposed method is tested with measurements of thermal effusivity in liquid samples.

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1. Introduction

Thermal effusivity, e, determines the thermal behavior of materials at surfaces and interfaces when time varying heat sources are present. It also governs the thermal impedance of a sample. Thus its knowledge is very important for the design and characterization of heat transfer processes. During the last decades the photopyroelectric (PPE) technique, in the so-called front configuration (FPPE), has been established as a reliable method for thermal effusivity measurements [1,2]. In this technique a sample is located in good thermal contact with a pyroelectric (PE) temperature sensor, which is periodically heated using an intensity modulated light beam at the surface opposite to that in contact with the sample. The illuminated side of the sensor is usually black painted to increase optical absorption. In this way the PE sensor acts as both a source of thermal waves propagating through the PE-sample sensor and as the detector of them. Usually a thermally thin PVDF (polyvinylidene difluoride) PE sensor is used and due to the good thermal contact that can be achieved the method is preferred for the characterization of liquids. If the measurements are performed in voltage mode, the amplitude V_{max} of the PE signal V_p can be represented as [3] (see Section 2 for details):

$$V_{max} = A \times |\Gamma| \quad (1)$$

where A is an instrumental transfer function independent on light modulation frequency, f, and of sample's properties, while $|G|$ is a modulus of Γ, which is a function of both f and thermal properties, in particular of e (see later). In a typical experiment measurements of V_{max} as a function of f are performed for both the investigated and a reference sample of known thermal properties. From the amplitudes quotient the parameter A is eliminated and e is obtained from a fitting procedure using the theoretical expression for Γ. This method has some handicaps that can limit its application in a general way: it becomes difficult to maintain the same experimental conditions when measurements are performed for the investigated sample and for the reference sample. For example, the actual value of A can change because the light energy absorbed by the sensor can be altered due to non-uniformities in the paint layer and sensor positioning due to samples loading, removal and subsequent cleaning. If air is used as a reference, care must be taken to avoid burning of the PE sensor, in particular when PVDF pyroelectric materials are used as sensors together with very high excitation light power densities. On the other hand the experiment is time consuming because it involves measurements in two different samples. In this paper we propose a method that avoids these limitations. It involves only one

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frequency scanning step for the investigated sample and uses the PE sensor itself as a reference to account for the instrumental function. The validity of the method is demonstrated with measurements of thermal effusivity performed in test liquid thermally thick samples.

2. Theory

Fig. 1 shows schematically the experimental configuration used in the PPPE technique, where *g* represents air, *p* the PE sensor and *s* the sample.

Assuming uniform sinusoidal heating of the sensor's surface, the pyroelectric voltage signal, V_p, can be written as [3]:

$$V_p(t) = \frac{pP_0(1-R)}{4C_p}\Gamma e^{i\omega t} = A\Gamma\epsilon^{i\omega t} = V_{max}(f,e)\times e^{i[\omega t+\varphi(f,e)]} \quad (2)$$

where P_0 is the laser power, R is the PE sensor optical reflection coefficient, p and C_p are the pyroelectric coefficient and electrical capacity respectively, $V_{max}(f,e)$ and $\varphi(f,e)$ are the signal amplitude and signal phase, respectively, and

$$\Gamma = \frac{(1-e^{-\sigma_p l_p})(1+R_{gp})}{\sigma_p l_p \quad k_p\sigma_p}$$
$$\left[\frac{(R_{sp}+R_{gs}e^{-2\sigma_s l_s})e^{-\sigma_p l_p}+(1+R_{sp}R_{gs}e^{-2\sigma_s l_s})}{R_{gp}e^{-2\sigma_p l_p}(R_{sp}+R_{gs}e^{-2\sigma_s l_s})-(1+R_{sp}R_{gs}e^{-2\sigma_s l_s})}\right]$$

In Eq. (3) the following notation has been used:

$$R_{ij} = \frac{1-b_{ij}}{1+b_{ij}},\ b_{ij} = \frac{e_i}{e_j},\ \sigma_j = (1+i)a_j,\ a_j^{-1} = \mu_j = \sqrt{\frac{\alpha_j}{\pi f}}$$

where $\omega = 2\pi f$ with f as modulation frequency, α_i,e_i and k_i are the thermal diffusivity, effusivity and conductivity of the i-th region of Fig. 1 (i = g,s,p), respectively, μ_i is the so-called thermal diffusion length and R_{ij} is the thermal wave reflection coefficient at the interface between media i and j.

Note that as sample is thermally thick even for lower frequencies, then $\sigma_s l_s \gg 1$ and Eq. (3) can be rewritten as:

$$\Gamma = \frac{(1-e^{-\sigma_p l_p})(1+R_{gp})}{\sigma_p l_p \quad k_p\sigma_p}\ \frac{R_{sp}e^{-\sigma_p l_p}+1}{R_{gp}R_{sp}e^{-2\sigma_p l_p}-1}, \quad (4)$$

that only depends on the sample's thermal effusivity.

The PE chosen for the experiments was a PVDF ($\alpha_p = 0.58\times10^{-7}\,m^2/s$ and $e_p = 559.4\,W\,s^{1/2}\,m^{-2}\,K^{-1}$ [4]) with a thickness of 110 μm, for which the sensor becomes thermally thick at relatively low frequencies without the appearance of thermoelastic effects [5]. The backing material was air ($\alpha_g = 22.260\times10^{-6}\,m^2\,s^{-1}$, $e_g = 5.510\,W\,s^{1/2}\,m^{-2}\,K^{-1}$ [6]).

The amplitude of the PE sensor signal V_{max} has been calculated using Eqs. (2) and (4) and it is showed as a function of the light modulation frequency in Fig. 2 for different liquids: distilled water ($\alpha_s = 1.44\times10^{-7}\,m^2/s$, $e_s = 1588\,W\,s^{1/2}\,m^{-2}\,K^{-1}$), ethanol ($\alpha_s = 0.88\times10^{-7}\,m^2/s$ and $e_s = 602\,W\,s^{1/2}\,m^{-2}\,K^{-1}$), methanol ($\alpha_s = 0.99\times10^{-7}\,m^2/s$, $e_s = 625\,W\,s^{1/2}\,m^{-2}\,K^{-1}$), ethylene glycol ($\alpha_s = 0.92\times10^{-7}\,m^2/s$, $e_s = 848.5\,W\,s^{1/2}\,m^{-2}\,K^{-1}$), glycerin ($\alpha_s = 0.9\times10^{-7}\,m^2/s$, $e_s = 943\,W\,s^{1/2}\,m^{-2}\,K^{-1}$), tetrachloromethane ($\alpha_s = 0.73\times10^{-7}\,m^2/s$, $e_s = 366\,W\,s^{1/2}\,m^{-2}\,K^{-1}$) [7,8] and propylene glycol ($\alpha_s = 0.92\times10^{-7}\,m^2/s$, $e_s = 721\,W\,s^{1/2}\,m^{-2}\,K^{-1}$ [9]).

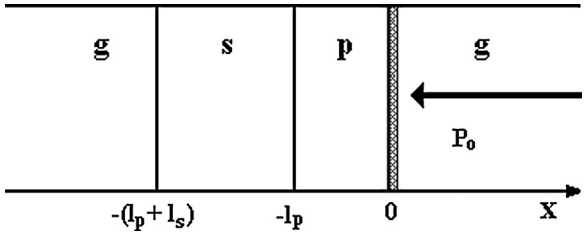

Fig. 1. Configuration used in PPPE measurements.

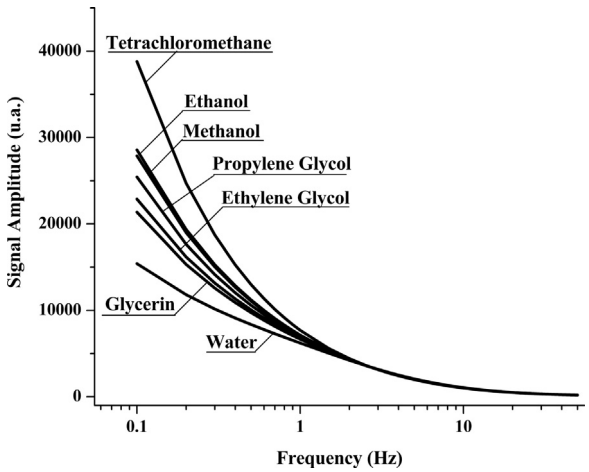

Fig. 2. Amplitude of the PE signal as a function of the modulation frequency calculated using Eqs. (2) and (4) for different liquids.

It can be seen from Fig. 2 that for modulation frequencies higher than 20 Hz the curves corresponding to the different liquids are overlapped. In order to account for the relative difference between the curves corresponding to the samples with the higher (water) and the lower (tetrachloromethane) thermal effusivity value, a parameter D has been introduced which is given by [10]:

$$D(f) = \frac{2[V_{max}(f,e_{tetrachloromethane}) - V_{max}(f,e_{water})]}{V_{max}(f,e_{tetrachloromethane} + V_{max}(f,e_{water})} \times 100\% \quad (5)$$

It is represented in Fig. 3 as a function of frequency.

It can be seen that for frequencies above 28 Hz the value of D becomes lower than 0.5%. This is not surprising, because for these frequencies the thermal diffusion length in the PVDF material is about 26 μm. The PVDF thickness is about four times larger than this value so that when arriving to the sample the thermal wave has an amplitude equal to approximately 1.8% of its original value [11]. The thermal wave is practically fully damped within the PVDF material and therefore the PVDF voltage becomes independent on sample's thermal properties. As a consequence, the parameter Γ in Eq. (2) has the same behavior for all liquids at higher frequencies. This is the foundation of the here proposed method. It consists in performing measurements of the amplitude as a function of the

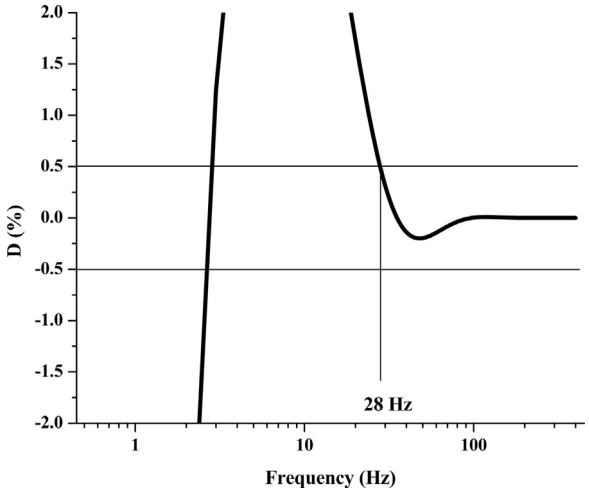

Fig. 3. Relative difference D between the signal amplitudes for water and tetrachloromethane.

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