Temperature controlled photoacoustic device for thermal diffusivity measurements of liquids and nanofluids

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In this paper, the design and development of a device for thermal diffusivity measurement of fluids exploiting the photoacoustic effect is presented. Compared to similar devices working only at room temperature, the version of the apparatus we present allows precise measurements of thermal diffusivity of fluids from room temperature to 70 °C. The reliability of the measurements is shown by the estimation of thermal diffusivity of well-known test liquids. The possibility to detect slight thermal diffusivity variations (around 1%) is verified by measurements on alumina based water nanofluids showing remarkable advantages in respect to alternative techniques.

Keywords: photoacoustic effect, photothermal, thermal diffusivity, liquids, nanofluid.

1. Introduction

The sufficiently accurate and precise measurement of thermal diffusivity has been a major concern for several years in the past and estimations on different fluids have been reported by authors with larger spreads than reported experimental errors [1]. With stationary methods, the precise measurement of temperature differences requires thick layers of material that in fluids are affected by convection, which is cause of overestimation of thermal diffusivity. Therefore, non-stationary methods have been used for this purpose, as in the case of the “Hot Wire”, the “Hot Disk” and the “Laser Flash” methods [2]. In recent years, after the advancing of the concept of nanofluids by Choi [3], namely suspensions of nanoparticles in base fluids showing or claiming enhanced heat transport characteristics, much effort has been made trying to obtain more precise determination of slight changes of thermal diffusivity, especially for basic investigation purposes.

We built a device based on, as is conventionally called, the photoacoustic effect for the estimation of the thermal diffusivity of fluids at different temperatures. The mathematical basis of photothermal techniques has been developed at the end of the seventies [4]. These techniques, just to cite some [5], have been employed for the characterization of several physical properties of matter, such as absorption spectrum, detection of gas traces, imaging and depth profiling. The proposed technique has been already successfully used for the measurement of diffusivity and thermal effusivity of fluids [6, 7] and, more recently, of nanofluids [8-10]. In particular, the photoacustic technique demonstrated a potential for a more accurate estimates of thermal diffusivity of fluids compared to more conventional techniques such as the "Hot Disk" and "Laser Flash" methods that seems more suitable for the characterization of solids and powders. As an example, Zhang et al. [11] reported an errors of 5% for thermal diffusivity estimations of nanonofluids by using the transient short-hot-wire technique.

Moreover, the preparation and implementation of measurement are much simpler compared to those of other methods. For instance, the Hot Disk method requires a very accurate stability of temperature, which is time consuming and also is often adversely affected by natural convection [12]; the Laser Flash method requires especially designed cells for liquids that generally do not allow accurate measurements above room temperature and necessitates a liquid N_2 cooled CCD sensors.

The device presented in this paper is inspired to the works of Balderas Lopez [7], that we improved by an accurate sample temperature control (from room temperature up to 70 °C), and a fully automation of measurements obtained with a National Instruments Labview® interface in order to limit possible error sources.

2. Experimental setup and theory

Figure 1 shows a schematic representation of our experimental setup. The instrument is equipped with a COMPACT-200G laser diode provided by World Star Tech emitting radiation at 808 nm. The laser diode is controlled by a TTL input to produce pulses in the range of a few Hz. The emitted IR pulse is transported by an optical fiber in order to impinge on 0.38 mm thick silicon window. The purpose of the monocrystalline Silicon slab is to absorb the laser radiation to convert light pulses in homogeneous heat waves traveling in the sample fluid. Silicon has been chosen for its high thermal diffusivity (88 mm^2/s) that make it highly responsive at the modulation frequency. The slab is placed on a stand that can be moved with micrometer precision using a computer controlled Thorlabs Z825B motorized actuator. During measurements, the Si slab is immersed in the liquid that is subject to periodic heat waves as effect of the laser pulses impinging the slab. Heat waves propagate into the liquid sample until they reach a 0.1 mm thick glass window, which is parallel to the Si slab. The glass window separates the liquid sample from a photoacoustic cavity. The cavity is a sealed chamber of a few cubic millimeters containing air, the temperature and pressure of which follows the periodic oscillations due to the thermal waves reaching the glass window. A stainless steel capillary of 0.5 mm inner diameter connects the cavity to a high sensitivity Vansonic VM-4530-2P electret condenser microphone that transduces the pressure oscillations in an electric signal, that is appropriately pre-amplified and processed by an EG&G Princeton Applied Research 5210 lock-in amplifier. The lock-in amplifier provides the amplitude of the microphone signal and the phase shift with respect to the laser pulses as a function of the thickness of the liquid, which is varied during the measurement by means of the motorized stage. The sample container is suitably heated by a Ni-Cr wire placed around the outer wall by means of a Kapton tape strip. In order to avoid possible negative interferences with the measurement, the wire is placed in the way that only a negligible magnetic field is produced when a current passes through it. During the experiments, temperature is controlled by a software PID controller that, reading a K thermocouple, settles the heater by a programmable current generator, assuring a temperature stability within 0.1 °C. The aluminum sample vessel guarantees temperature uniformity for the entire sample environment, while suitable nylon supports provide a sufficient thermal insulation for both emitter and sample vessel. Since temperature variations could negatively affect the signal to noise figure of microphone, the thermoacoustic cavity is connected to it by means of a stainless steel capillary, in order to keep the microphone in equilibrium with room temperature and afar from heated sample.

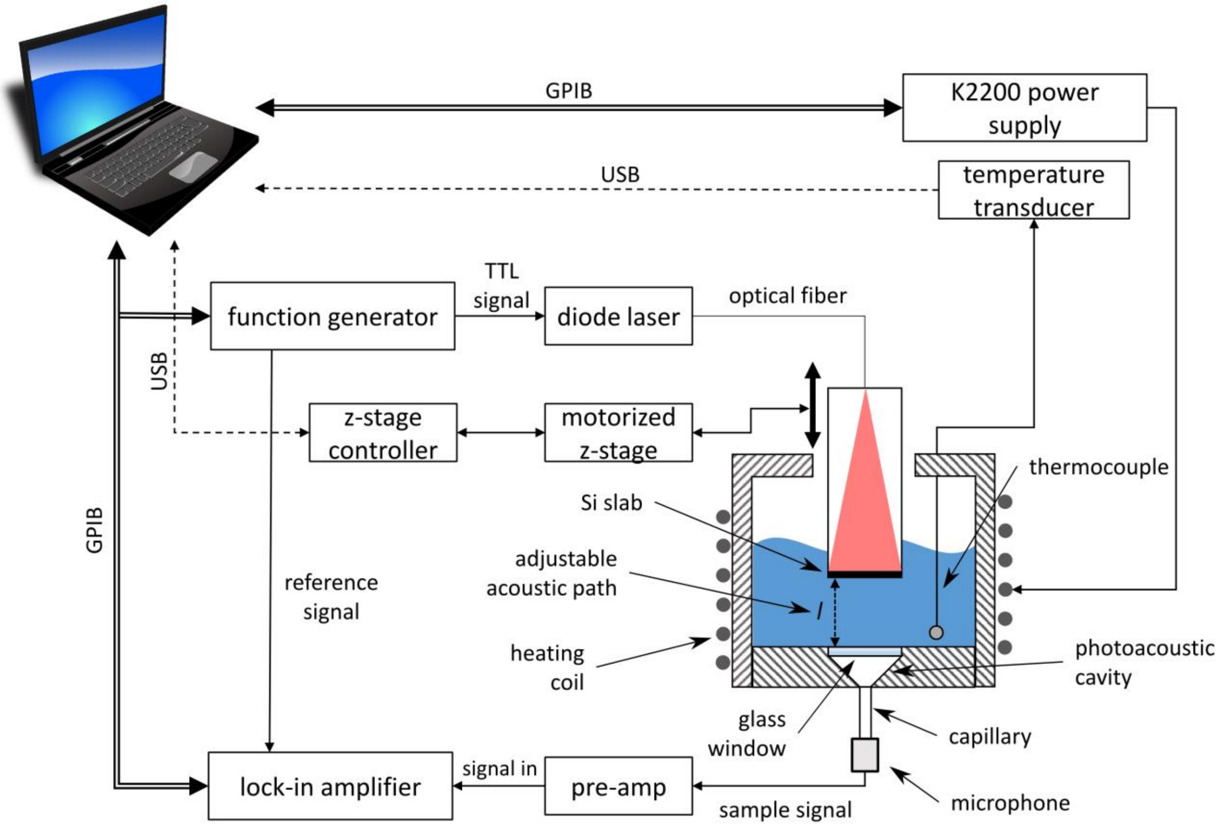

Fig. 1. Schematic view of instrumental setup.

As derived from the theory of the photoacoustic applied to liquids described by Balderas-Lopez [6], when liquid thickness *l*, i.e. the travel path of the acoustic wave front in the liquid, satisfies the following condition

$$l > \sqrt{\frac{\alpha}{f}} = l_m \, , \tag{1}$$

where α is the thermal diffusivity of the fluid and f is the operating pulse frequency of the laser, the sample under investigation can be considered as "thermally thick" and the following two relations become valid:

$$\ln A = \ln|C| - \sqrt{\frac{\pi f}{\alpha}}\, l$$

(2)

$$\Delta\varphi = \Delta\varphi_0 - \sqrt{\frac{\pi f}{\alpha}}\, l$$

(3)

where A is the microphone signal amplitude; C is a constant depending on thermal properties and geometry of the system, which is independent from l and is therefore constant during the measurement; $\Delta\varphi$ is the phase difference between the laser pulse and the signal detected by the microphone at a given l and $\Delta\varphi_0$ is the phase difference when the thickness of the liquid is null.

Therefore, by appropriately varying the fluid thickness l during the measurement, namely moving the emitter with the micrometric motorised stage in the range of few tenths of millimetre, recording both the amplitude and the phase shift of the microphone signal in respect to laser pulse, it is possible to construct two straight lines, whose slopes are inversely proportional to the square root of the thermal diffusivity of the liquid.

Hence, during the same measurement is possible to doubly measure the thermal diffusivity of a fluid from two independent quantities, such as the phase shift and the signal amplitude. Moreover, these measurement conditions avoid any adverse contribution of natural convection, which is suppressed by the thin measurement layer of fluid and the related low Reynold's numbers.

The complete measurement process, including the setting of desired parameters, data acquisition and processing of results, is fully automated by a virtual instrument (vi) developed using the Labview 2011 platform.

In order to validate the proposed experimental setup, thermal diffusivity measurements at different temperatures have been performed on deionized water (Millipore, Billerica MA, USA, 18.2 MΩ) and ethylene glycol (99%, provided by Alpha Aesar).

To show the capability of experimental setup to discriminate among slight variations of thermal diffusivity, measurements on nanofluids obtained by dispersion of an Al_2O_3 nanopowder (99.5%, 40-50 nm, provided by Alpha Aesar) in deionized water have also been performed on two different alumina dispersions, at 1 and 2 wt% respectively, and treated by sonication with a Sonics & Materials VCX130 ultrasonic processor, to assure their uniformity and stability.

Photoacoustic measurements have been performed at a Laser pulse frequency of 2.5 Hz. Data points (microphone signal amplitude change and phase shifting vs. thickness of liquid "*l*") have been acquired starting from the minimum thickness of liquid that can be considered as "thermally thick" according to condition expressed in (1), with steps of 0.02 mm.

3. Results and discussion

The instrument has been designed on the base of Finite Element (FE) analysis using the software Comsol Multiphysics 4.4. The FE analysis has been exploited mainly to evaluate the temperature distribution in the fluid in two different conditions. In order to ensure that no convective phenomena may adversely contribute to the thermal diffusivity measurements, an analysis has been performed to study the steady-state behaviour without laser pulse, as shown in figure 2. Insulating nylon elements have been considered in the model, both on the top, around the emitter, and on the bottom, under the device.

Boundary conditions have been defined to simulate convective heat transfer with air at room temperature on the external surfaces and to fix the temperature on the bottom (insulating nylon layer) at the value Tc=25°C; the temperature on the heating coil was forced to be 75 °C. The analysis has been performed with 0.38 mm water thickness between the Si slab and the glass window. In this first stage of modelling, the temperature difference between the Si slab and the window, with no laser pulse considered, was found to be $\Delta T \approx 0.2$ °C (71.9 °C on the Si slab and 72.1 °C on the glass). This slight temperature difference between the two surfaces ensures that convection cannot occur during measurement. The analysis yielded a temperature T_c =73.3 °C in the point where the thermocouple is located for the measurements.

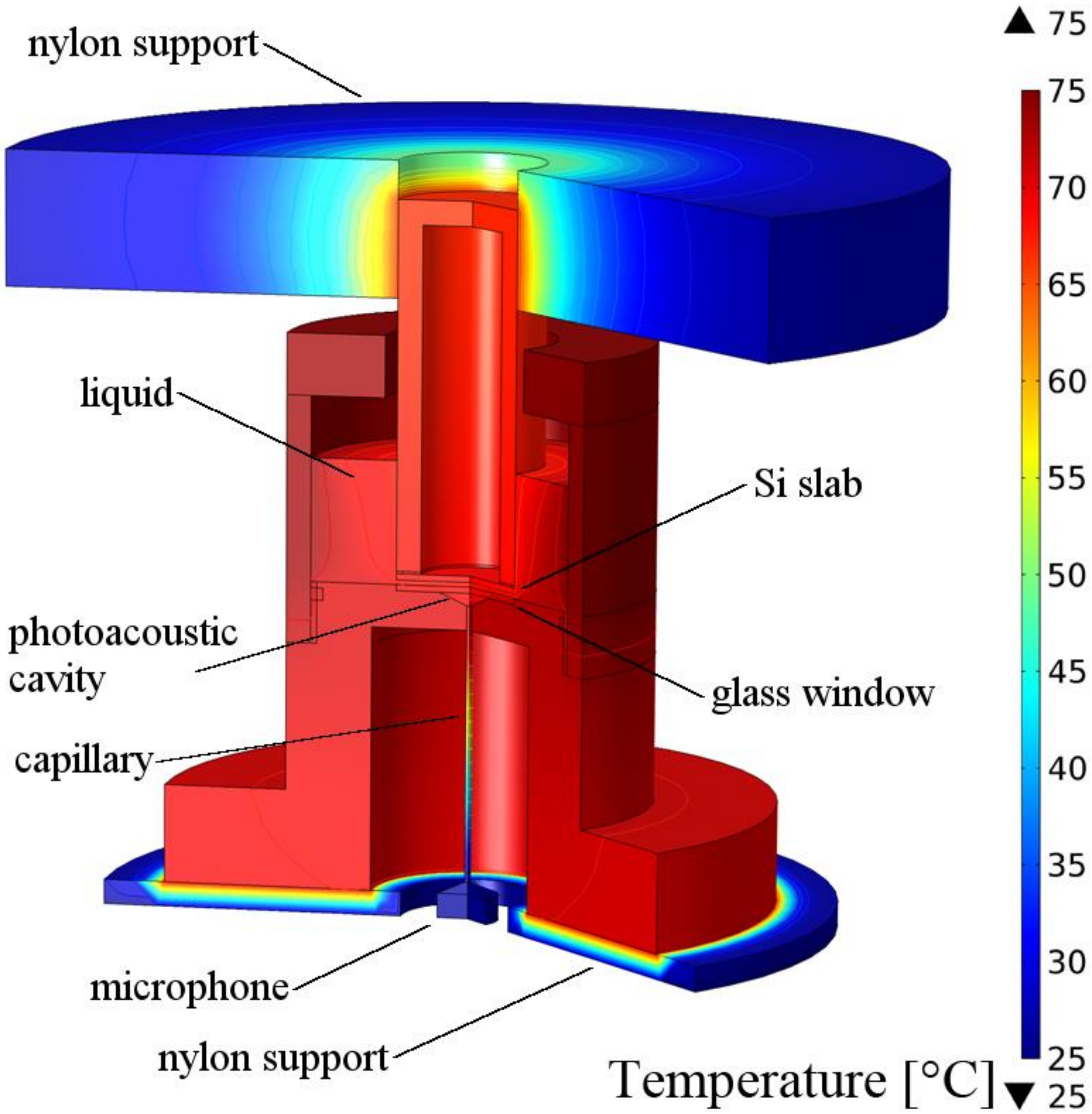

Fig. 2. Finite Element Analysis: surface and contour plot of temperature distribution in the device, the fluid and the insulating element with no Laser Pulse (steady-state analysis).

Moreover, in order to show that the laser pulses do not lead to an excessive overheating of the sample measurement volume, a transient FE analysis has also been performed. The analysis has been carried out at room temperature, considering laser pulse heating with 2.5 Hz modulation frequency; the thermal power transferred to the Si slab has been estimated as 200 mW with 50% duty cycle. This study showed that a single pulse causes an average temperature increase of about 0.25 °C in the fluid layer, leading to steady state increase of about 0.8 °C in a few seconds as shown in figure 3.

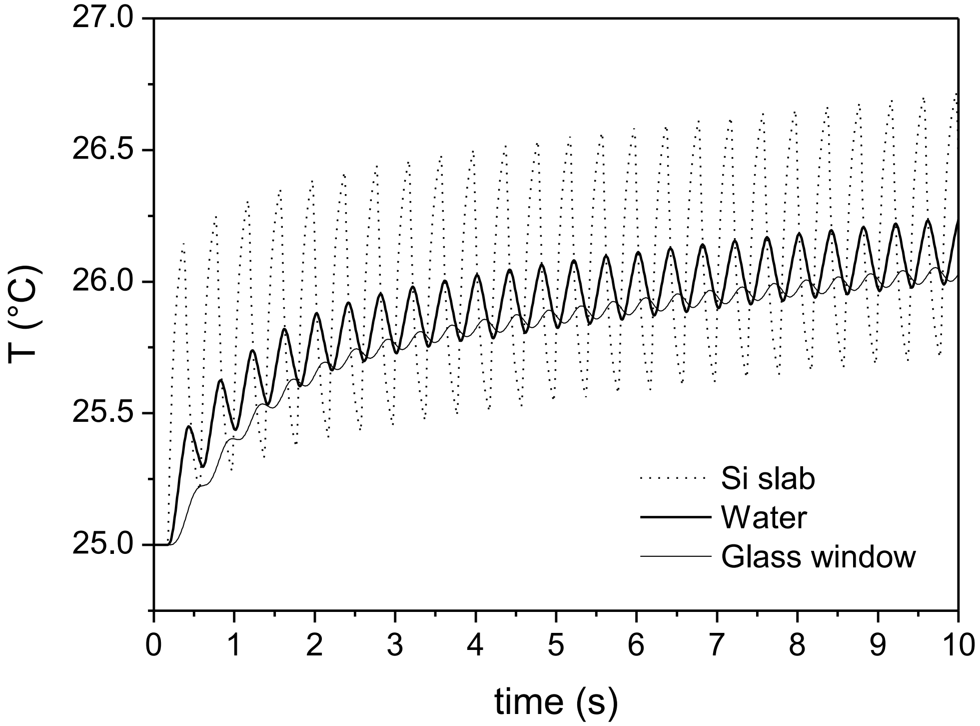

Fig. 3. Finite Element Analysis: Temperature of the Si slab, water and the glass window as a function of time.

Figures 4(a) and 4(b) show phase difference and amplitude change as a function of deionized water and ethylene glycol thickness at different temperatures in the range 25-65 °C. Thermal diffusivity has been calculated using equations (2) and (3) from the slope of linear fits shown in the same figures, errors have been evaluated from linear fit errors on the slopes. These results are summarized in figures 5(a) and (b) and compared with reference data [13, 14].

Figure 4(b) also shows as the signal amplitude is almost independent on sample temperature, confirming the effectiveness of using a capillary to thermally uncouple the microphone from the system.

The microphone signal would be deeply influenced by sample temperature, leading to unacceptably measurement uncertainties.

As shown, the average of results obtained from phase and amplitude are in good agreement with reference data of both water and glycol.

Nevertheless, a slight positive systematic component, especially in the phase data, can be noticed. This probably ~~could~~ arise from the fact that the signal coming from the microphone is not ideal, namely sinusoidal. Therefore, when the amplitude of the signal changes by effect of sample thickness variation, also the signal shape slightly changes. Since Lock-in amplifiers are meant to work better with sinusoidal signals or shape-invariant signals, some little erroneous additional phase change could be introduced. These errors tend to be higher by increasing sample temperature due to the adverse influence on microphone performances. The thermal insulation of microphone from sample vessel obtained by the capillary duct greatly reduces the performance loss due to temperature increase, even though it is not completely avoided.

The average error is estimated in the range of 2% for measurements on water and 3% on glycol. These estimates are derived from linear fits and therefore affected both by random and by systematic components deriving from intrinsic non-linearity, such as the non-ideality of signal shape above discussed.

In order to obtain an estimation of random and systematic components of error, 10 thermal diffusivity measurements have been carried out at 45 °C for both water and glycol. With water, the standard deviation of thermal diffusivity estimated from phase is 1.4% and those estimated from amplitude is 1.7%; with glycol, they are 1.7% and 1.3% respectively.

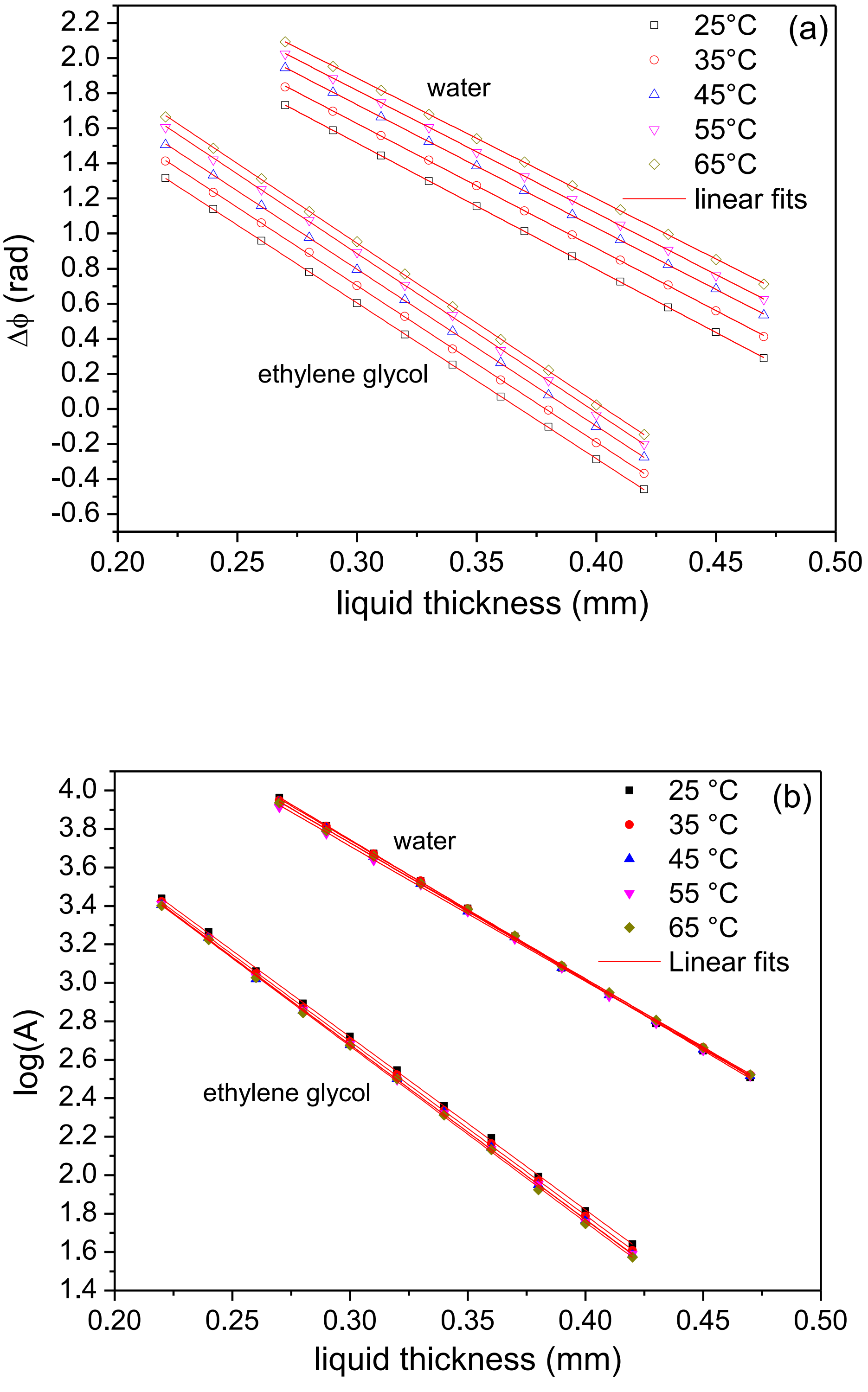

Fig. 4. Photoacoustic phase change (a) and signal amplitude (b) as a function of liquid thickness for deionised water and ethylene glycol at different temperatures.

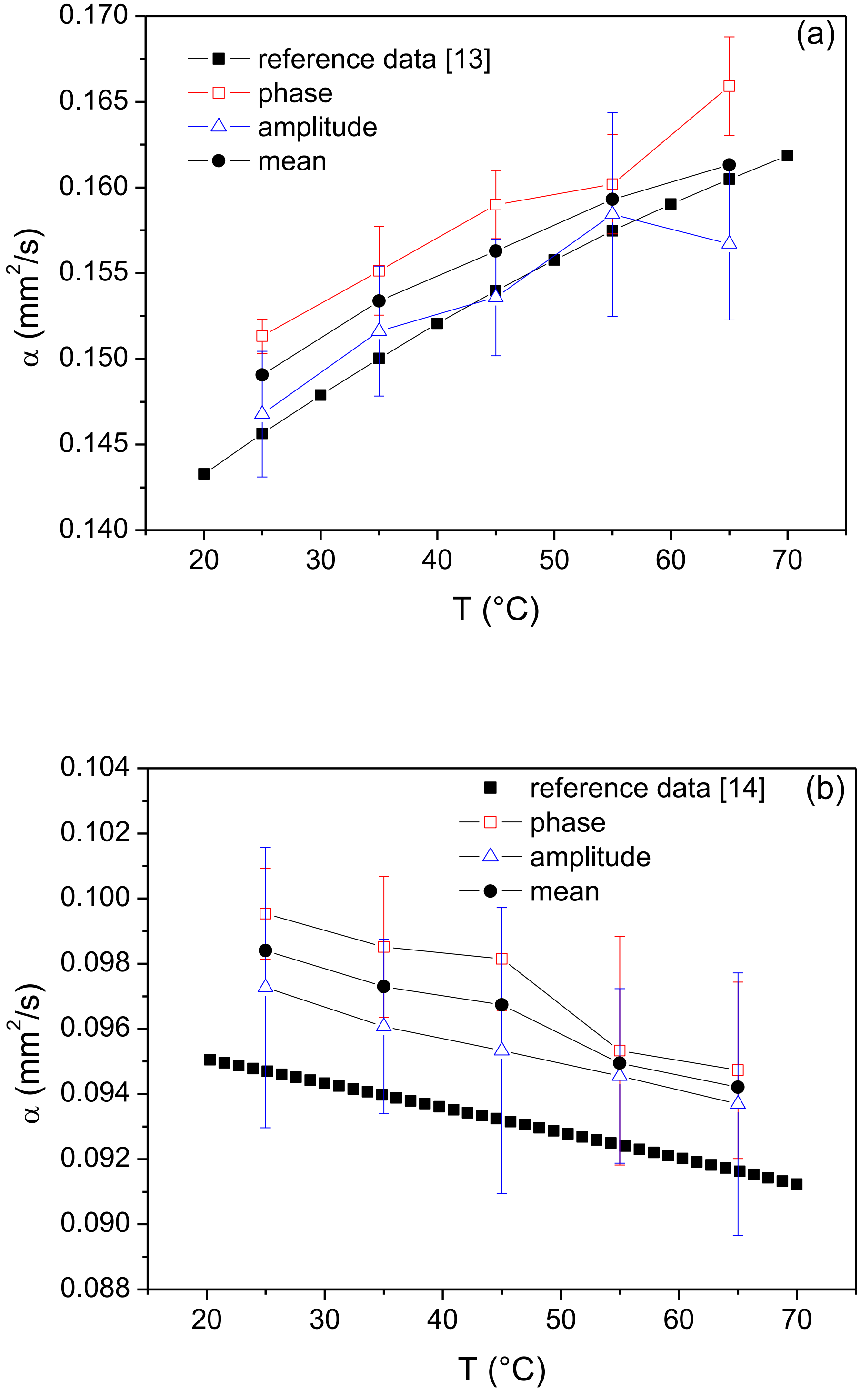

Fig. 5. Results of linear fits compared with literature data for water [13] (a) and ethylene glycol [14] (b).

On the other hand, even if a little systematic error is present in the valuation of absolute thermal diffusivity, the precision of the proposed technique is definitively advantageous in comparing thermal diffusivities of nanofluids, where the case of too low improvements to be detected using other techniques is frequent. As an example, figure 6 shows the thermal diffusivity measurements at different temperatures on Al_2O_3 based nanofluids compared to pure water. As shown, it is possible to clearly distinguish thermal diffusivity changes around 1%.

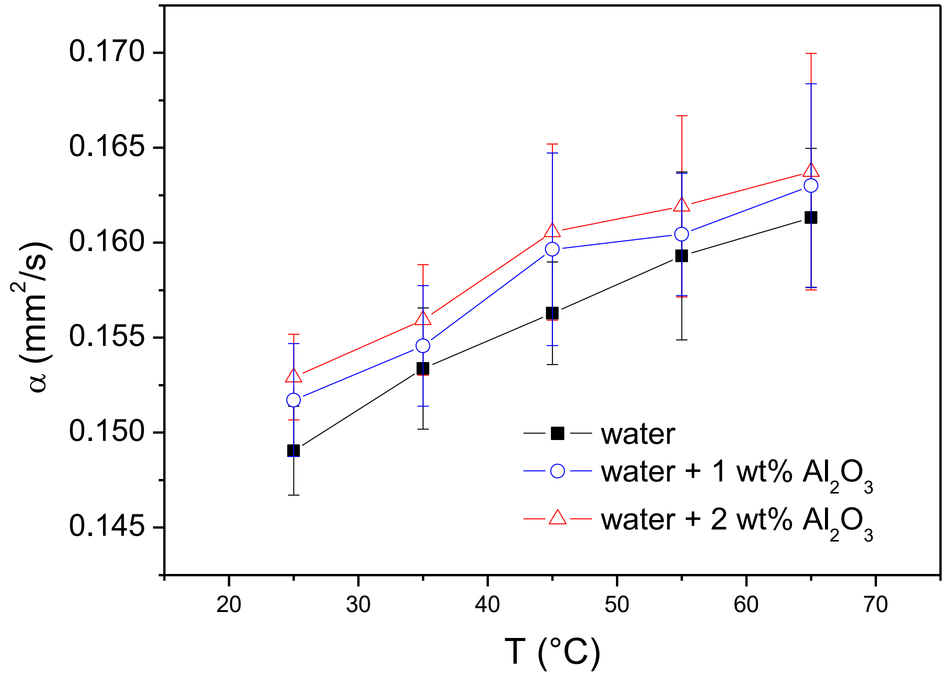

Fig. 6. Thermal diffusivity of Al_2O_3 nanofluids compared to pure water (mean values).

4. Conclusions

The proposed measurement setup demonstrates remarkable improvements compared to alternative techniques in the evaluation of thermal diffusivity of liquids and nanofluids at different temperatures. Particularly, it allows to discriminate even minimal variations of thermal diffusivity and to avoid overestimations caused by natural convection. The automation of the measurement and the handiness of the apparatus improves its use in the lab. FE analysis showed that no convective phenomena may occur in the sample measurement volume and that the laser pulses do not lead to important overheating of the sample with respect to the imposed measurement temperature.

A version of the apparatus with the implementation of a software version of the Lock-in Amplifier will be object of next studies.

Moreover the instrument will be upgraded with the possibility of adding external stimulus (like magnetic fields) in order to better characterize particular kinds of nanofluids.

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