APPLICATION AND TESTING OF A NEW SIMPLE EXPERIMENTAL SETUP FOR THERMAL CONDUCTIVITY MEASUREMENTS OF LIQUIDS

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Transient hot wire method is considered a reliable and precise technique for measuring the thermal conductivity of liquids. The present paper describes a new transient hot wire experimental setup and its initial testing. The new apparatus was tested by performing thermal conductivity measurements on substances whose reference thermophysical properties data existed in literature, namely on pure toluene and double distilled deionized water. The values of thermal conductivity measured in the temperature range 25 to 45 °C deviated +2.2% to +3% from the literature data, while the expanded measurement uncertainty was estimated to be ±4%.

Keywords: *thermal conductivity, liquid, transient hot wire method, toluene, distilled water.*

1. Introduction

Thermal conductivity values for substances in the solid, liquid and gaseous state are necessary for the design of devices and installations in which heat is exchanged. However, the thermal conductivity of liquids and gases is among the thermophysical properties that are difficult to measure with high accuracy. This is due to the fact that in fluids, in addition to conduction, both of the two other heat transfer mechanisms - convection and radiation - take place.

A number of stationary and non-stationary experimental methods have been employed in measuring the thermal conductivity of liquids: transient hot wire technique, thermal constants

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analyzer technique, steady-state parallel-plate method, cylindrical cell method, temperature oscillation technique, 3ω method and other techniques [1,2]. The transient hot wire method is considered a reliable and precise technique for determining the thermal conductivity of liquids. The measurement yields absolute value of thermal conductivity and does not necessitate calibration or comparison of the sample under test with a standard [3]. Input parameters for the calculation of thermal conductivity are the change in the electrical resistance of the wire, energy generated by the electric pulse that heats the wire and the length of the wire. The measurement uncertainty of the hot wire technique has been estimated to about 1% for the most developed experimental setups [4,5].

The present paper describes a new transient hot wire experimental setup. The apparatus was tested by measuring the thermal conductivity of toluene and distilled water whose thermophysical properties have been already accurately measured and published [6,7]. Measurements were conducted at 25, 35 and 45 °C and at atmospheric pressure.

The setup will be applied in studying thermal conductivity of various organic solvents (in pure form and in binary mixtures) used in the process of purification of flue gases from coal-fired thermal power plants of harmful compounds (CO_2 and SO_x). Some of those solvents are dimethylaniline (DMA) and tetraethylene dimethyl ether (TEGDME), mono ethanol amine (MEA), polyethylene glycols of various molecular weight (PEG), as well as ionic liquids. Considering the need of measurements on a large number of binary systems, as well as the relatively high purchase price of some of the substances to be tested in analytical quality, one important input requirement for the design of the setup was to reduce the sample volume keeping the level of measurement uncertainty comparable with the measurements published in relevant literature [3-5]. Additional requirement for the apparatus was to accommodate samples of electrically conducting liquids. Hot wire, therefore, has to be electrically insulated from the surrounding liquid, to prevent current flow through the liquid which would make heat flux measurement impossible.

2. Method

Thermal conductivity measurements of liquids by using the transient hot wire method are based on the model of a long thin conductor surrounded by a liquid of infinite volume. When electricity is pulsed through the conductor it is heated by the Joule effect and a part of the generated heat is transferred to the surrounding liquid. The rate of conductor heating is inversely proportional to the intensity of the heat transfer to the surrounding fluid, which depends on the thermal conductivity of the liquid. Thus, knowing the rate of energy flow into the conductor and monitoring the rate of a conductor temperature rise, the thermal conductivity of the surrounding liquid may be computed. As the temperature of the conductor changes, so does its electrical resistance. The temperature of the conductor is monitored via its electrical resistance – analogous to resistance thermometer. Thus, the long thin conductor in this experiment - hot wire - simultaneously plays the roles of the heat source and temperature sensor.

Therefore, the long thin straight-line conductor generates heat flux per unit length, q, heats by conduction itself and the surrounding liquid of thermal conductivity k and thermal diffusivity a. The conductor is assumed to be at a uniform temperature which is variable in time [5, 8]. Differential equation governing the spatial-temporal temperature change, T, of the liquid is the Fourier equation in cylindrical coordinates

$$\frac{1}{a}\frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) \tag{1}$$

where $T(r,t)=T_0+\Delta T(r,t)$, where r and t denote radial distance and time, respectively. The boundary conditions are (i) a constant amount of heat generated per unit length of the thin conductor,

$$\lim_{r\to 0}\left\{r\frac{\partial T}{\partial r}\right\} = -\frac{q}{2\pi k}, \tag{2}$$

where $q=I_w*V_w/L_w$ is the heat flux per unit length, with I_w, V_W *and* L_w being hot wire current, voltage, and length, respectively, and (ii) a negligible temperature change far from the conductor:

$$\lim_{r\to\infty}\{\Delta T(r,t)\} = 0. \tag{3}$$

The solution of eq. (1) in the form of an infinite series, given in [8], can after a short time be approximated as

$$\Delta T = t(r,t) - T_0 = \frac{q}{4\pi k}\left\{-\gamma + \ln\left(\frac{4at}{r^2}\right)\right\} \tag{4}$$

Differentiating this equation with respect to time for an arbitrary value of radius r the following expression can be obtained:

$$k = \frac{q}{4\pi}\frac{d\ln(t)}{d(\Delta T)}. \tag{5}$$

According to this equation, the thermal conductivity of the liquid is proportional to the heat flux per unit length of the conductor and inversely proportional to the logarithmic derivative of temperature in function of time. Equation (5) is valid as long as the predominant form of heat transfer is conduction. The occurrence of convection – the largest source of measurement error - can easily be detected when ΔT, as a function of $\ln(t)$, deviates from a straight line. Other deviations from the theoretical model - the finite length and diameter of the hot wire, non-uniform temperature distribution of the wire, the finite thermal conductivity and heat capacity of the hot wire, heat transfer by radiation, heat conduction through the wire – have significantly less influence on the measurement error [4].

3. Experimental

3.1 Materials

Two readily obtainable liquids with known thermal conductivities, toluene and double distilled deionized water, were used to test and validate the setup. Toluene (methylbenzene) is an aromatic hydrocarbon available in a high-purity state. Toluene used in this work was manufactured by MERCK and its declared purity was better than 99.9%. Double-distilled deionized water was produced locally, using equipment manufactured by MILLIPORE Corp.

3.2 Experimental Setup

A key part of the new transient hot wire experimental setup is the cell, shown in fig. 1. It consists of a sample reservoir, which is a glass test-tube epoxy-glued to a brass fitting element and a hot wire holder, which has the shape of the hollow half-cylinder and fits into the sample reservoir. When the hot wire holder is introduced into the sample reservoir partially filled with the liquid under test, its level rises until the hot wire is completely immersed.

Figure 1. Photo of the hot wire cell used in this work

The hot wire holder was made of soft soldered copper tube and brass connecting elements. An important requirement for the cell design was to make the measurement setup portable and reduce sample volume as much as possible, considering the relatively high price of the chemicals to be tested in the future: ionic liquids and nanofluids. Another criterion was to make cell assembly and disassembly relatively easy for thorough cleaning during the sample or hot wire replacements. The tension of the hot wire is provided with a helical spring. Resulting mechanical construction ensures easy disassembly, cleaning, and positioning and provides electrical connections to measuring instruments, partial electrical shielding and minimization of electrical noise. The construction of the cell is given in fig. 2.

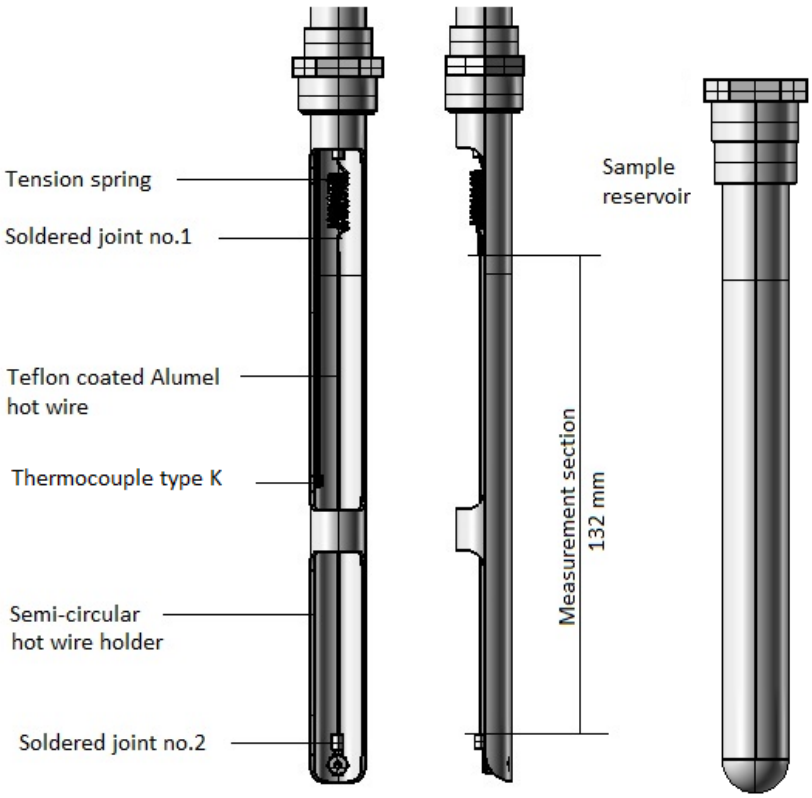


Figure 2. Construction of the hot wire cell

The hot wire material was chosen to be Alumel alloy (composition: Ni 95%, Al2% Mn2% Si1%). The selection of Alumel was guided by its chemical stability and relatively high value of its temperature coefficient of electrical resistance (rate of change in electrical resistivity with temperature), nominally $23.9 \cdot 10^{-4}$ 1/°C. Otherwise, this value of platinum, the most commonly used hot wire material, is $39.2 \cdot 10^{-4}$ 1/°C. Another consideration was the relatively high price of platinum wire (both bare and teflon-coated), which could become a limiting factor in the maintenance of the measurement apparatus in good working condition. Thus, in the present work, a commercially available Alumel conductor manufactured by OMEGA® and designated as TFAL-003-50, with 70 µm diameter and 80 µm Teflon® coating (overall diameter 230 µm) is used. Teflon® has well-known dielectric properties, it is chemically inert, corrosion resistant and tolerant to low and high temperatures without cracks.

The length of the hot wire is 132 mm, the internal diameter of the hot wire holder (i.e. the approximate diameter of the sample) is 13 mm, while the total length of the sample is 160 mm. The inner diameter of the sample reservoir is 16 mm and the sample volume approximately 27 ml.

The sample cell is immersed in a thermostatic water bath consisting of a large 3 l glass beaker, immersible 600 W heater, temperature homogenization mixer and digital temperature controller with a K-type thermocouple sensor. Bath temperature can be regulated to within ±0.1 °C.

The sample temperature is monitored by another K-type thermocouple mounted inside sample cell, on hot wire holder close to the middle of the hot wire. The hot wire holder is mechanically attached to a box made of aluminum which houses the measurement electronics – standard series resistor, current pulse generator, and data acquisition system.

The hot wire resistance is measured by the standard 4-wire method. The current pulse runs through a series connection of the standard resistor and the hot wire. The voltage across the standard resistor and the hot wire, V_s and V_w, respectively, are measured by computer-controlled data acquisition system NI 6009 manufactured by National Instruments®. Thermocouple signal is monitored between the pulses to ensure that steady state is reestablished before the next pulse is applied.

The resistance value of the standard resistor used is 9.60 Ω at room temperature and that of the hot wire is approximately 10 Ω.

The program for the collection, initial processing, and storage of collected data was developed for this experimental setup in LabView® programming environment. The operation of the setup is controlled via NI 6009 digital outputs.

4. Results and Discussion

4.1 Results

At each temperature, multiple measurements were made with the measurement duration of 50 seconds. Each measurement implied 2500 samples at the frequency of 50 Hz. There was also a 500-second interval between two current pulses to enable reduction of the sample temperature to that of the surrounding thermostatic bath.

Figure 3 depicts a hot wire temperature change for double distilled water. The linear part of the temperature change begins after approximately 1 second from the onset of the current pulse and ends after about 12 seconds. The linear part of the graph ends at the beginning of heat transfer by convection or when other deviations from ideal conditions emerge.

Equation (5) was used to calculate thermal conductivity from the obtained experimental data. To calculate $d\Delta T/d(\ln(t))$, vectors $\ln(t)$ and ΔT were formed. After establishing the interval of linearity, the slope of a linear portion of the curve $\Delta T = f[\ln(t)]$ was determined by linear interpolation. The heat flux was calculated using the formula $q = I_w V_w / L_w = V_w V_s / (R_s L_w)$. The final value of thermal conductivity for each temperature was obtained as the average of multiple measurements.

The slope of ΔT as the function of the natural logarithm of time, as seen in fig. 3 in one of the experiments with distilled water at 25 °C, was calculated by fitting a straight line to the linear part of the experimental curve. The heating current was typically 110 mA. At short and long times the function was nonlinear, however, between approximately $\ln(t) = 0.5$ and 2.5, i.e. between 1.7 s and 12.1 s, the function was close to linearity indicating that no significant convection occurred. The total increase of the wire temperature was several degrees, depending on the current intensity applied.

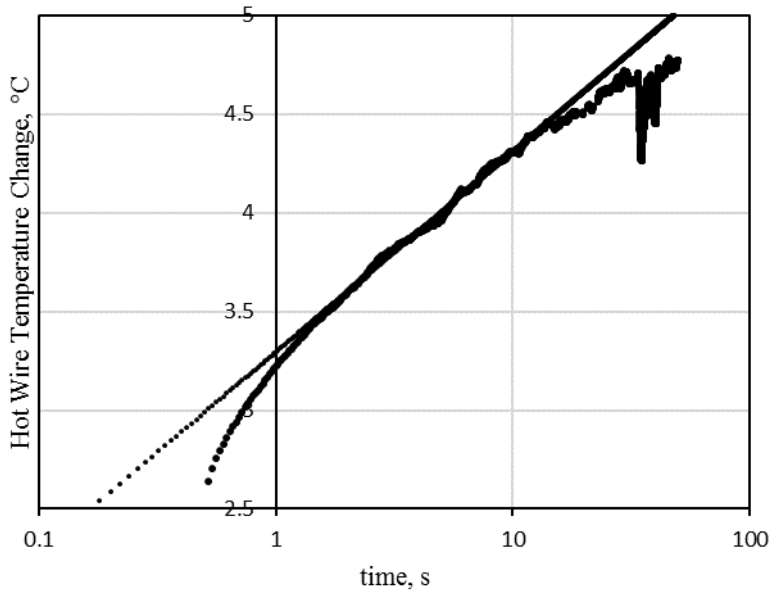
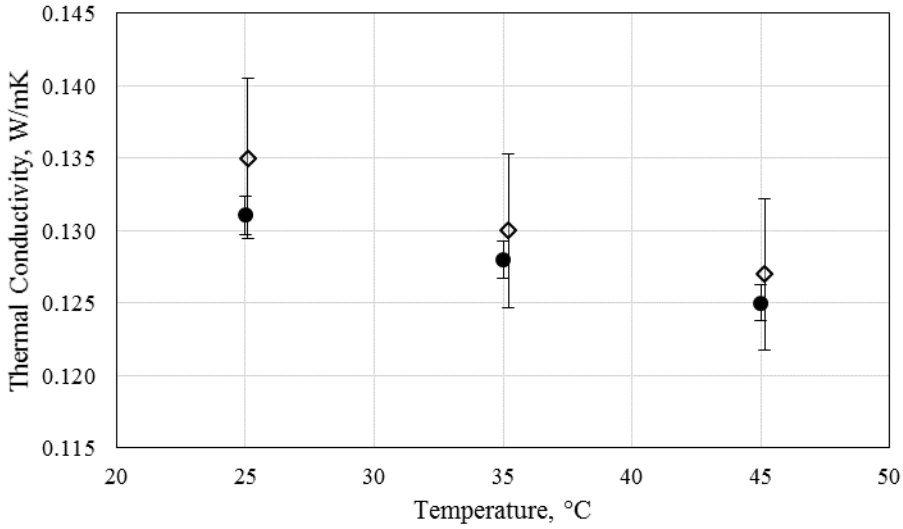


Figure 3. Hot wire temperature increase [°C] vs. time [s] (in logarithmic scale), actual curve and linear interpolation

Figure 4 represents values of thermal conductivity of toluene and double distilled deionized water, obtained experimentally at temperatures 25, 35 and 45 °C, as well as literature data for comparison. The thermal conductivity of both liquids changes only slightly in this temperature range, values are nearly constant. Deviation of experimental values from literature data is less than or equal to 3%.



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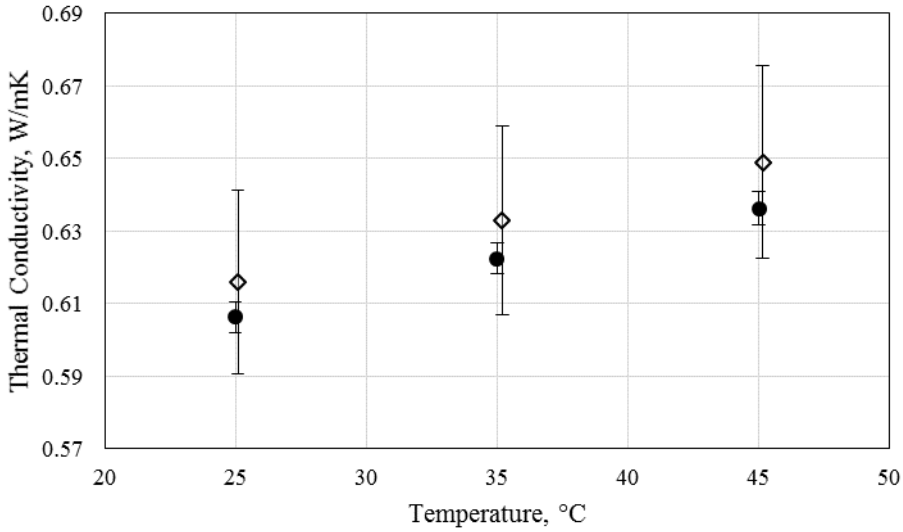

b)

Figure 4. Thermal conductivity [$Wm^{-1}K^{-1}$] vs. temperature [°C] for a) Toluene and b) distilled water. Filled circles represent literature data [6,7], squares represent values obtained in this work.

In tab. 1 all data are listed – literature and measured thermal conductivity values. Measurement uncertainty for literature data is 1% for toluene and 0.7% for distilled water [6, 7]. Percentage deviations of measured data from literature values were calculated using the formula

$$PD(k) = 100 * \frac{1}{m} \sum_{i=1}^{m} \left(\frac{k_{exp} - k_{lit}}{k_{exp}} \right)^2 \ [\%] \tag{6}$$

The number of experimental data points in the present case was *m*=30 for toluene and *m*=10 for distilled water.

Table 1. Thermal conductivity measurement results and deviations

Liquid	Temperature [°C]	Literature value [$Wm^{-1}K^{-1}$]	Measured value [$Wm^{-1}K^{-1}$]	Percentage deviation [%]
Toluene	25	0.1310 [7]	0.135	3.0
	35	0.1280 [7]	0.130	2.4
	45	0.1250 [7]	0.127	2.9
Distilled Water	25	0.6064 [6]	0.616	2.8
	35	0.6225 [6]	0.633	3.0
	45	0.6363 [6]	0.649	2.2

4.2 Uncertainties

Uncertainty in the measurement of thermal conductivity was evaluated in accordance with the Guide to the expression of uncertainty in measurement (GUM) [9]. The obtained uncertainty components and the total expanded uncertainty of performed thermal conductivity measurement are given in tab. 2. The uncertainty components listed in tab. 2 were calculated starting from eq. (5). Since the definition of the temperature coefficient of resistivity can be written in the form $\Delta R = \sigma R_{w0} \Delta T$, eq. (5) can be rearranged as

$$k = \sigma R_{w_0} \frac{q}{4\pi} \frac{1}{Z_R} \tag{7}$$

The uncertainty of k depends, therefore, on uncertainties of the temperature coefficient of resistivity σ, *the* initial value of hot-wire resistance R_{w0}, heat flux per unit length q and the slope of the change of hot-wire resistance vs. logarithm of time – $d\Delta R/d(\ln(t))$ or Z_R.

Uncertainty of k is calculated by error propagation, given by the variance formula

$$u_k = \sqrt{\left(\frac{\partial k}{\partial q} u_q\right)^2 + \left(\frac{\partial k}{\partial \sigma} u_\sigma\right)^2 + \left(\frac{\partial k}{\partial R_{w_0}} u_{R_{w_0}}\right)^2 + \left(\frac{\partial k}{\partial Z_R} u_{Z_R}\right)^2} \tag{8}$$

or the square root of the sum of squares of eq. (7) factors' uncertainties (i.e. in q, σ, R_{w0} and Z_R). In turn, uncertainties of components depend on measurement uncertainties of quantities used to calculate their values. The reported uncertainty is based on a standard uncertainty multiplied by a coverage factor 2, providing a level of confidence of approximately 95%.

Table 2. Uncertainty components and the total expanded uncertainty of thermal conductivity measurement

Uncertainty	[%]
u_q/q	2.5
u_σ/σ	2.5
u_{Rw0}/R_{w0}	0.2
u_{ZR}/Z_R	1.9
u_k/k	4.2

5. Conclusion

New experimental setup for measuring the thermal conductivity of electrically conducting liquids based on the transient hot wire method was designed, constructed and tested. The apparatus was tested by measuring the thermal conductivity of toluene and distilled water, readily available liquids with well known thermal conductivities. The setup was developed for thermal conductivity measurements of binary mixtures of organic solvents intended for the purification of flue gases of coal-fired thermal power plants.

A requirement for the sample volume minimization, maintaining reasonable measurement accuracy, was met in this work. Values obtained by test measurements deviate from the published data by about 3% with a sample volume of about 27 ml.

Future experimental work with the apparatus will concentrate on measurements of thermal conductivity of various organic solvents used for flue gas purification.

Acknowledgment

The authors gratefully acknowledge the financial support received from the Research Fund of Ministry of Science and Environmental Protection, Serbia and the Faculty of Technology and Metallurgy, University of Belgrade (project No. 172063).

Nomenclature

I	current, [A]
k	thermal conductivity, [$Wm^{-1}K^{-1}$]
L	length, [m]
m	number of experimental data points
D	percentage deviation
q	heat flux per unit length, [Wm^{-1}]
R	resistance, [Ω]

Greek letters

γ	Euler constant, [0.5772]
σ	temp. coeff. of resistivity, [$°C^{-1}$]

Subscript

exp	value obtained experimentally

T	temperature, [°C]	lit	value found in literature
u	uncertainty	w	hot wire
V	voltage drop, [V]	s	standard resistor
Z_R	slope of ΔR vs. ln(t), $d\Delta R/d(ln(t))$, [Ω°C^{-1}]	0	initial value
$T(r,t)$	temperature of liquid at radial distance r at time t, [°C]		
$\Delta R(t)$	difference between hot wire resistance at time t and initial resistance, [Ω]		
$\Delta T(r,t)$	difference between temperature of liquid at time t and initial temperature, [°C]		

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