Assessment of modulated hot wire method for thermophysical characterization of fluid and solid matrices charged with (nano)particle inclusions

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Assessment of modulated hot wire method for thermophysical characterization of fluid and solid matrices charged with (nano)particle inclusions

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Abstract. Recently we reported on simultaneous thermal conductivity k and thermal diffusivity a measurement of liquids and in particular of nanofluids in a configuration using an ac excited hot wire combined with lock-in detection of the third harmonic (3ω method) [1]. The conductive wire is used as both heater and sensor. The requirements for the asymptotic validity of the line heat source model are fulfilled at low modulation frequencies below a few Hz. The study of the relative sensitivity of signal amplitude and phase to changes in k and a indicates that there is an optimum frequency range for accurate and stable results. We extend by up to two decades the feasible frequency range for 3ω measurements by considering various more elaborate models for the heat transfer between the wire and the fluid. Finally we show that the same ac hot wire method can be applied to soft solid, composite materials. We measured the k enhancement of a poly(ethylene vinyl acetate) EVA polymer matrix charged with various fractions of graphite.

1. Introduction

The effect of (nano)particle inclusions on the effective thermal conductivity of liquids and solids has attracted a great interest experimentally and theoretically. Despite a significant number of experimental studies, there still exist disagreements between data from different research groups [2]. Besides our work cited in the Abstract, another recent work reports on thermal conductivity k and heat capacity measurement of fluids by 3ω technique [3].

2. Theoretical background

In an inhomogeneous material one can define locally the specific thermal impedance as the ratio between the temperature increase θ generated by the heat flow and the heat flux density ϕ at that location, $z=\theta/\phi$ [m^{2}K/W]. For a semi-infinite material, the (plane) surface impedance writes $z=(1-i)/[(4\pi f)^{1/2}e]$ and it depends exclusively on the effusivity $e=(Ck)^{1/2}$, with C the volume specific heat and k the thermal conductivity. A periodic heat flux of frequency f was assumed. In cylindrical

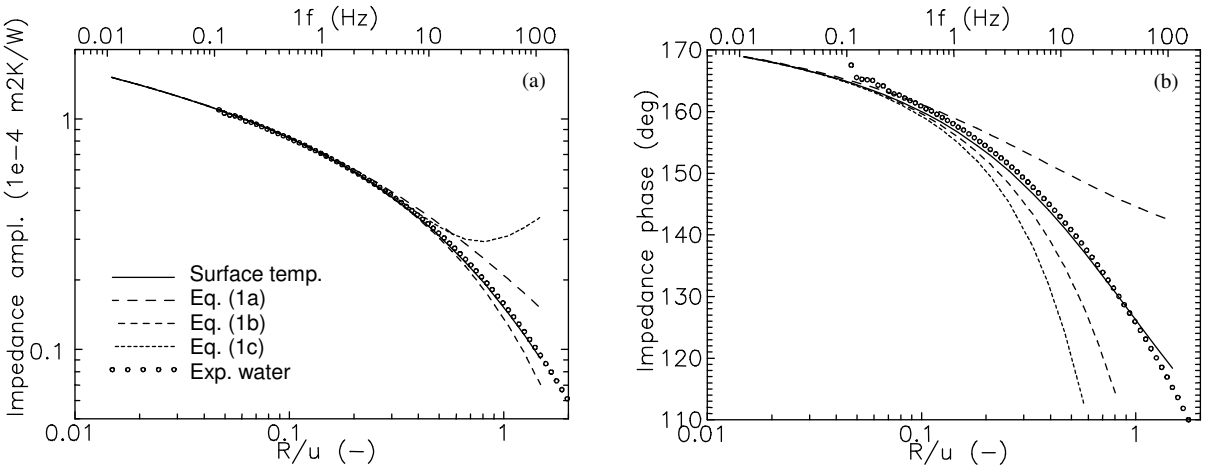

Figure 1. Amplitude (a) and phase (b) of the thermal impedance in cylindrical geometry calculated with Eqs. (1a-c) (dashed curves) and with the exact surface temperature model (solid curve) (see further below). The points are experimental results for water. The lock-in has a mixing phase shift of 180^0 which explains the shifted phase origin in (b).

geometry, the specific impedance has the expression [4]:

$$z = \frac{R}{k}\frac{K_0(\sigma R)}{\sigma R\, K_1(\sigma R)} \approx \frac{R}{k} K_0(\sigma R) \approx \frac{R}{k}\left(\log_e \frac{\sqrt{a/\pi f}}{1.2594\, R} - i\frac{\pi}{4}\right) \qquad (1a\text{-}c)$$

where K_0 and K_1 are the modified Bessel functions, $\sigma=(1+i)/\mu$ with $\mu=(a/\pi f)^{1/2}$ the thermal diffusion length and $a=k/C$ the thermal diffusivity, and R is the radius of the cylindrical surface beyond which the material extends to infinite. The successive approximations (Eqs. 1b,c) are valid at low frequency such that $R/\mu<<1$ and they correspond to the model of a line heat source embedded in an infinite medium [5, 6]. Amplitude and phase simulations of Eqs. (1a-c) are plotted in Fig. 1 for a Ni wire with a diameter of 45 μm immersed in distilled water. Assumption is made for integral dissipation of electrical power in the medium in the form of heat and for a sufficiently long wire ($2L$=16.2 mm).

Eq. (1c) reveals that in cylindrical geometry the primary thermophysical parameters are k and a. The imaginary part Im(z) and the phase $\varphi(z)$ of Eq. (1c) allow for simultaneous and independent determination sample's k and a relative to a reference medium (index r) having similar properties [1]:

$$\frac{k}{k_r} = \frac{\mathrm{Im}(z_r)}{\mathrm{Im}(z)} \qquad \frac{\alpha}{\alpha_r} = 1 + \frac{\pi(\varphi-\varphi_r)}{2\sin^2\varphi_r} \qquad (2a,b)$$

Eqs. (2a,b) are evaluated at a single frequency. Alternatively, a can be determined from the slope of Re(z) vs. $\log_e(f)$ [6].

The relative sensitivity S represents the ratio of relative function variation to relative argument variation. The Eq. (1c) indicates that the modulus |z| is proportional to $1/k$, and so do the real and imaginary parts Re(z), Im(z). Therefore, their sensitivity to k is S_k=-1. Since the phase is independent on k, $S_{k,\varphi}$=0. By contrast, due to the $\log_e$ function, the amplitude and phase sensitivities to a are weak (Fig. 2). However, $S_{a,A}\neq 0$ and it would interfere significantly with the determination of k from the signal amplitude. With increasing frequency $S_{a,\varphi}$ increases rapidly, but eventually the approximation of Eq. (1c) is not valid any more and it gives large errors (especially in the phase) as seen in Fig.1. At 1 Hz, $S_{a,\varphi}$=0.2 implying a relative variation $\Delta a/a$=8.7% for 1^0 phase change. The same conversion factor applies for the error on a caused by the signal phase difference between model and experiment. It turns

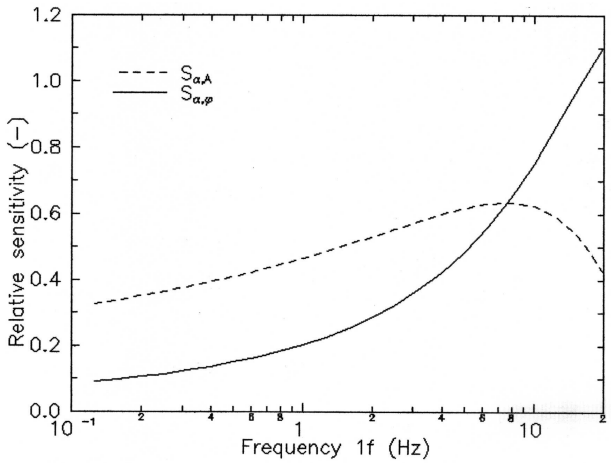

Figure 2. Relative sensitivity of 3ω amplitude and phase signal of Eq. (1c) to thermal diffusivity a of sample (water). $S_{a,\varphi}$ is expressed in rad/100% change in a.

out that there is an optimum frequency range such that $R/\mu<1$ in which Eqs. (2a,b) yield accurate and stable results.

In reality, the (modulated) heat flux dissipated in the medium is smaller than the electrical input power. The difference is stored in the heat capacity of the wire. The solid curves in Fig. 1 were calculated with a model for the wire surface temperature that accounts for this correction. Thus the frequency range for agreement between theory and experiment is extended by two decades up to about 100 Hz.

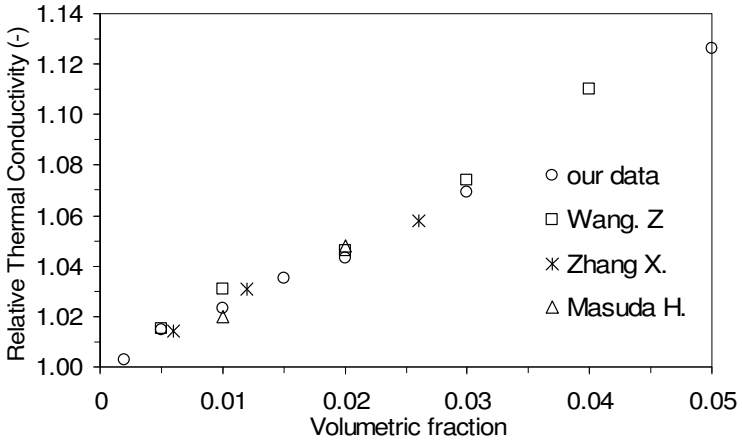

Figure 3. Experimental results of thermal conductivity enhancement of TiO_2 nanofluids compared to literature data [10-12] as indicated.

3. Experimental results and discussion

The principle and procedures of the 3ω hot wire method have been described elsewhere [7, 8]. The 3ω signal is proportional to the specific thermal impedance z introduced in Section 1. The points in Fig. 1 show a typical measurement in water. Below 0.5 Hz the agreement with all three forms of Eq. (1) is good and justifies the imposed condition $R/\mu<<1$. It appears that Eq. (1b) is a better approximation than Eq. (1c) and its subsequent Eqs. (2a,b). The deviations at higher frequency are more pronounced in the signal phase. We determined previously k and a enhancements of about 3% and 4.5% respectively for 12% wt Aerosil 200V silica nanoparticles (12 nm in diameter) in water, using Eqs. (2a,b) [1]. In Ref. [9] we measured k and the viscosity of silica and alumina nanofluids. In this study

we present experimental results on k of water based TiO_2 nanofluids prepared without any surfactant. TiO_2 nanoparticles having 21 nm in diameter and specific surface area (BET) of 50±15 m^2/g, manufactured by Degussa Co. were dispersed in deionized water by ultrasound. In Fig. 3, our results demonstrate excellent agreement with selected literature data. The k values are slightly (<1%) lower than the predictions of Hamilton–Crosser model [13]. Thus other works reporting higher (anomalous) k on the same compound have not been confirmed.

Finally we show that the same hot wire method can be applied to soft solid, composite materials. We measured the k enhancement of a poly(ethylene vinyl acetate) EVA matrix charged with 8-50 % wt graphite. In practice, the wire is squeezed between two flat sample slabs. Initially, the 3ω signal amplitude is abnormally high due to poor thermal contact with the sample. Above a pressure threshold, the signal amplitude stabilizes, indicating a good thermal contact. We observed an up to threefold k enhancement relative to pure EVA at the fundamental frequency of 0.5 Hz. The phase signal could not be exploited and improvement of sample holder should solve this problem.

Concerning the k homogenization model of the samples, we developed a generalized model of the thermoelastic tensor for composite materials having arbitrary texture and containing anisotropic inclusions of variable shapes [14, 15]. The model includes a thermal interface resistance between the particle and the matrix and explains better than the Maxwell model certain experimental data.

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References

[1] A. Turgut, C. Sauter, M. Chirtoc, J.F. Henry, S. Tavman, I. Tavman and J. Pelzl, *Europ. Phys. J. Spec. Top.*, **153**, 349 (2008).
[2] S.M.S. Murshed, K.C. Leong and C. Yang, *Appl. Thermal Engn*. **28**, 2109 (2008).
[3] S.R. Choi, J. Kim and D. Kim, *Rev. Sci. Instrum*. **78**, 084902 (2007).
[4] D. Maillet, S. André, J.C. Batsale, A. Degiovanni and C. Moyne, *Thermal Quadrupoles*, John Wiley and Sons, Chichester, 2000.
[5] H.W. Carslaw and J.C. Jaeger, *Conduction of Heat in Solids*, Second ed., Oxford Univ. Press, London, UK, 1959.
[6] D.G. Cahill, *Rev. Sci. Instrum*. **61**, 802 (1990).
[7] C. Dames and G. Chen, *Rev. Sci. Instrum*. **76**, 124902 (2005).
[8] M. Chirtoc and J.F. Henry, *Europ. Phys. J. Spec. Top*. **153**, 343 (2008).
[9] I. Tavman, A. Turgut, M. Chirtoc, H.P. Schuchmann and S. Tavman, *Archives of Mat. Sci. Engin.* **34**, 99-104 (2008).
[10] Z.L. Wang, D.W. Tang, S. Liu, X.H. Zheng and N. Araki, *Int. J. Thermophysics,* **28**, 1255 (2007).
[11] X. Zhang, H. Gu and M. Fujii, *Int. J. Thermophysics*, **27**, 569 (2006).
[12] H. Masuda, A. Ebata, K. Teramae and N. Hishinuma, *Netsu Bussei (Japan)* **4**, 227 (1993).
[13] R.L. Hamilton and O.K. Crosser, *Ind. Engn. Chem. Fundamentals* **1**, 187 (1962).
[14] K. Hadjov, *Europ. J. Mech. Environ. Engn*. **46**, 171 (2001).
[15] O. Fudym, B. Ladevie and J.C. Batsale, *Numerical Heat Transfer, Part B: Fundamentals*, **42**, 325 (2002).