Title: *Thermal Diffusivity and Volumetric Specific Heat Measurements Using Heat Flow Meter Instruments* for Thermal Conductivity 29 / Thermal Expansion 17 Conference *

Authors: Akhan Tleoubaev
Andrzej Brzezinski

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ABSTRACT

Heat Flow Meter Method (ASTM C518, ISO 8301, EN 1946-3, etc.) instruments are the most widely used steady-state type of devices for thermal conductivity measurements. In computerized systems the heat flow meters' (transducers') signals can be recorded versus time. The recorded signals can give additional information about other important thermal properties of the samples – volumetric specific heat and thermal diffusivity.

Volumetric specific heat can be calculated simply from amount of the heat flow per square area absorbed by sample after switching instrument's plates' temperature set points from one (after reaching thermal equilibrium condition) to another (until reaching new thermal equilibrium condition) .

For thermal diffusivity calculations two unsteady state thermal problems solutions should be used: 1) with boundary conditions of 1st kind (so-called ideal thermal contact, i.e. when thermal contact resistance is negligible) - for samples of low thermal conductivity, and 2) with boundary conditions of 3rd kind (in presence of thermal contact resistance) - for samples of moderate thermal conductivity. After reaching so-called "regular regime" thermal diffusivity of the specimen can be calculated using slopes of the graphs of logarithms of the heat-flow meters' signals versus time (slopes of their linear part) during the process of the system's exponential relaxation toward the final thermal equilibrium.

Experimental checks using several materials - Expanded Polystyrene, 1450b and 1450c NIST Standard Reference Materials, Pyrex 7740, Vespel SP1, Perspex, Pyroceram 9606 were done. Probable causes of thermal diffusivity disagreement for materials of higher thermal conductivity are discussed.

LaserComp, Inc., 20 Spring Street, Saugus, Massachusetts 01906 USA

INTRODUCTION

Heat Flow Meter (HFM) instruments (ASTM C518, ISO 8301, EN 1946-3, etc.) are the most popular steady-state thermal instruments. They are routinely used for thermal conductivity λ (W m^{-1} K^{-1}) measurements only, although their heat flow meters' signals recorded versus time contain information about other important thermal properties of the samples – thermal diffusivity $a=\lambda/C_p\rho$ (m^2s^{-1}) (ρ is density in kg m^{-3}), volumetric specific heat $C_p\rho$ (J m^{-3} K^{-1}), and thermal effusivity $\varepsilon=\lambda/\sqrt{a}$. Some efforts in this direction were already undertaken (see e.g. Nicolau, et. al. [1], Bae [2]). If any two of the mentioned four thermal properties - λ, a, $C_p\rho$, and ε - are known, then other two can be calculated, i.e. full set of the four thermal properties can be determined. We developed and tried new procedures to get two of these additional thermal properties (which are described below), using our LaserComp's FOX Heat Flow Meter instruments and their modified software algorithms.

THERMAL DIFFUSIVITY MEASUREMENTS PROCEDURE USING HFM INSTRUMENTS

Theoretical consideration of the transient heat conduction in finite bodies for large time t shows that series of the thermal problem general solution [3]

$$T(x,t)=\sum_{n=1}^{\infty} C_n v_n(x)\exp\{-a\alpha_n^2 t\}$$

(where α_n are eigenvalues, v_n are eigenfunctions of the problem, and C_n are coefficients determined by the boundary and initial conditions) *"rapidly converges, and, starting at certain time, the first term different from zero predominates over the sum of the remaining terms. This corresponds to the physical fact that, independently of the initial distribution, starting at some time, a* ***"regular regime"*** *of a temperature field evolution is established in the body which has a temperature "profile" invariant with time and the amplitude decreasing exponentially with time"* [3]:

$$T(x,t)\approx C_1 v_1(x)\exp\{-a\alpha_1^2 t\} \qquad (1)$$

This can be used in practice for thermal diffusivity calculations using the recorded heat flow meters' signals. The single-exponent relaxation of the temperature field in the "regular regime" dramatically simplifies the mathematical formulas, and is useful for practice. As a result, both heat flow meters' signals calculated using Eq.(1) relax to its final equilibrium value exponentially and thermal diffusivity a can be calculated using the recorded HFMs' signals.

Analytical mathematical solution of our one-dimensional thermal problem (flat sample sandwiched between two isothermal parallel plates - see similar

problems in [4] and [5]) gave us a simple formula for thermal diffusivity using the late stage of the transient process, after the "regular regime" has been reached, when the only, the slowest one, exponent of the thermal relaxation process still exists:

$$a = L^2 \, \pi^{-2} \, (-\Delta \ln Q_i / \Delta t) \qquad [\mathrm{m}^2/\mathrm{s}] \qquad (2)$$

where L is thickness of the sample (in meters), π is 3.14159…, and $(\Delta \ln Q_i / \Delta t)$ is a slope of the graph of natural logarithms of the HFMs' signals Q_i vs. time (in seconds). The graph should look like a straight line after the "regular regime" is established (see Fig.1).

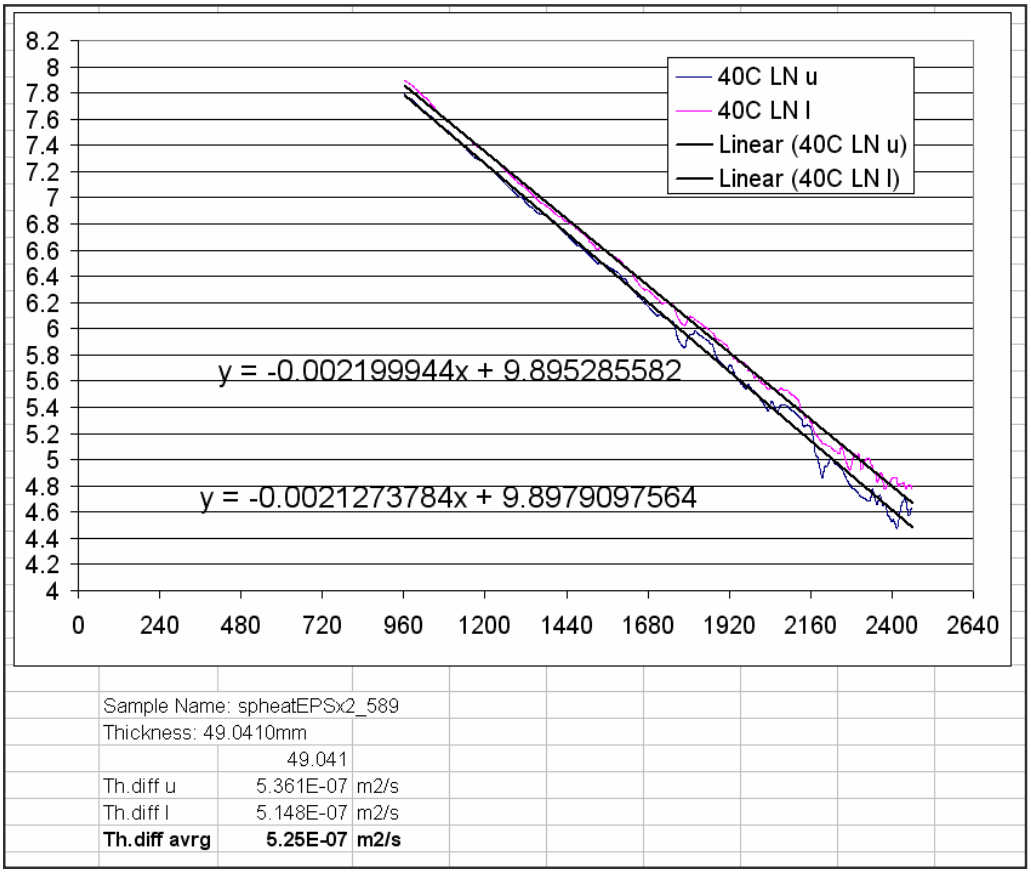

Figure 1. Typical graph of logarithm of the heat flow meters' signals vs. time in seconds. Thermal diffusivity values were calculated using Eq.( ) (FOX600, 49 mm-thick Expanded Polystyrene of 44.6 kg/m^3 or ~2.8 lbs per cubic foot density)

This simple formula was derived using boundary conditions of 1st kind (i.e. in assumption of ideal thermal contact between sample's surfaces and instrument's plates) and works satisfactorily for thermal insulation materials where thermal contact resistance is negligible in comparison with the sample's thermal resistance (which equals its thickness divide by its thermal conductivity).

No publications containing values of thermal diffusivity of Expanded Polystyrene were found in literature. Only specific heat value of 1500 J/(kg K) was found [6] which can be re-calculated into volumetric specific heat using easily measurable density of the samples (44.6 kg/m^3 on Fig.1 example), and into thermal diffusivity using thermal conductivity value (0.034 W/mK) to get an approximate reference value of 5.1×10^{-7} m^2/s for comparison with Fig.1 example result of 5.25×10^{-7} m^2/s.

In case of materials of intermediate thermal conductivity presence of the thermal contact resistance should be taken into account. This means that boundary conditions of the 3rd kind one should be used for solution of the thermal problem. Eigenvalues α_i can be found as solutions of the transcendent characteristic equation [7]:

$$2\cot\xi = \xi R/R_s - (\xi R/R_s)^{-1}$$

where $\xi = \alpha l$, l is half-thickness of the sample, R is thermal contact resistance, and R_s is sample's thermal resistance. Equation can be solved using Newton's iteration method. Then thermal diffusivity equals $1/(\alpha_1^2 \tau)$ where τ is reciprocal of the slope $-\Delta \ln Q_i / \Delta t$. Experimental checks of this equation didn't show good agreement with literature data of materials with known thermal diffusivity like Vespel, Pyrex, and especially with materials of higher thermal conductivity and diffusivity – Pyroceram and Stainless Steel 304.

As another one attempt to eliminate this disagreement A. Tleoubaev has found (also using boundary conditions of 3rd kind and separation of variables) a closer to real conditions of experiment new analytical solution for this transient thermal problem for temperature sensor buried inside the heat flow meter body. But this theory also didn't help to get agreement between calculated experimental results and literature data. Similar disagreement between thermal diffusivity data obtained by the Laser Flash method using thermocouples and by infra-red photo sensors already were discussed in [8] and [9], and where, as it was believed, thermal contact resistance on thermocouples' surfaces and their heat capacities were the causes of the disagreements.

Significantly more accurate and reliable way to obtain full set of the 4 thermal properties is measurements of the volumetric specific heat by calculating heat absorbed by the sample from instrument's plates.

VOLUMETRIC SPECIFIC HEAT MEASUREMENTS PROCEDURE USING HFM INSTRUMENTS

Volumetric specific heat $C_p\rho$ can be obtained from the total amount of the heat flow per square area absorbed by the sample sandwiched between the instruments' two plates after switching instrument's plates' temperature set points from one (after reaching thermal equilibrium condition) to another (until reaching new thermal equilibrium condition).

The heat flow meters' readings QU_i (upper plate) and QL_i (lower plate) are proportional (and direction-sensitive) to the heat flow's densities (W/m^2) in or out of the two sides of the sample, multiplied by the time interval τ between the readings, and then summed give us the total amount of heat absorbed by the sample (per unit of square area). I.e. the heat flow meter instruments can work like calorimeters. HFM signals at the final equilibrium condition QU_{equil} and QL_{equil} should be subtracted from each of the readings otherwise the total sum will never reach its plateau, and will keep slowly increasing because of the practically inevitable small edge heat losses (or, in general, gains):

$$H = \sum_{i=1}^{N} [SU_{cal}(QU_i - QU_{equil}) + SL_{cal}(QL_i - QL_{equil})]\tau$$

where SU_{cal} and SL_{cal} are the two HFMs' (upper and lower) calibration factors.

This sum H also contains amount of heat absorbed by the two heat flow meters themselves, which, as was proven by our experiments, can be quite significant, and should be excluded to get accurate values of the $C_p\rho$. Heat capacity of the heat flow meters $C_p`\rho`2\delta x$ can be found from the energy conservation equation:

$$C_p\rho x + C_p`\rho`2\delta x` = H/\Delta T$$

where ΔT is temperature change, $C_p`\rho`$ and $2\delta x`$ is specific heat and thickness of the two heat flow meters, using different ways: 1) simply running specific heat test with no sample (plates should have same temperatures simultaneously increased or decreased) – all the heat is absorbed by the heat flow meters only; 2) running specific heat tests using thin sample of material of known specific heat; 3) running two (or more) the same material samples of two (or more) different thicknesses – thin x_1 and thick x_2 – for whom we have system of 2 equations with 2 unknowns:

$$C_p\rho x_1 + C_p`\rho`2\delta x` = H_1/\Delta T$$

$$C_p\rho x_2 + C_p`\rho`2\delta x` = H_2/\Delta T$$

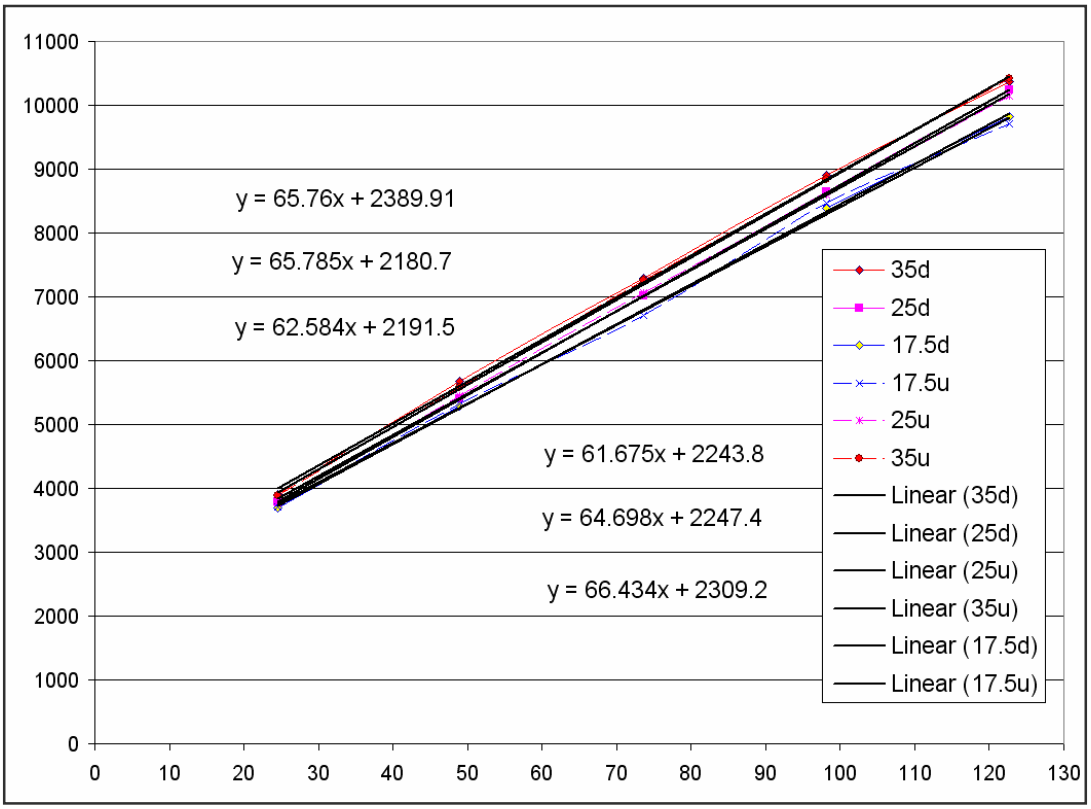

Figure 2. Graphs of the H sums divided by temperature jump vs. thickness in millimeters for Expanded Polystyrene samples (44.6 kg/m^3 or ~2.8 lbs per cubic foot density) for different temperatures. Slopes equal volumetric specific heat (divided by 1000). Extrapolations down to zero thickness are heat capacities of the HFMs.

Solving this system of 2 equations we can find both the volumetric specific heat of the samples:

$$C_p\rho = (H_2\text{-}H_1) / [(x_2\text{-}x_1)\ \Delta T]$$

and heat capacity of the two heat flow meters (per their square area):

$$C_p`\rho`\ 2\delta x` = (H_1x_2 - H_2x_1) / [(x_2\text{-}x_1)\ \Delta T]$$

then to be excluded as the apparatus constant (which is temperature dependent) to get correct specific heat of a single sample test:

$$C_p\rho = (H/\Delta T - C_p`\rho`\ 2\delta x`) / x$$

As experimental checks proved, the most accurate way to determine the HFMs' heat capacity $C_p`\rho` 2\delta x`$ is the first one - direct measurements with run of temperature change with closed instrument's plates and with no sample. After that the specific heat can be accurately determined using the latter formula.

Special versions of the WinTherm32 (for thermal insulation samples) and WinTherm50 (for intermediate thermal conductivity samples) software used by the LaserComp's FOX Heat Flow Meter instruments have been developed for the volumetric specific heat $C_p\rho$ (J/m^{3}K) measurements in addition to the routinely measured thermal conductivity. Specific heat measurements can be done simultaneously with the regular thermal conductivity tests. Temperature jump in this case is difference between mean temperatures of the two consecutive set points. For example, if plates' temperatures (set points) are 10^0C and -10^0C (mean 0^0C), 35^0C and 15 (mean 25^0), then temperature jump is 25^0C and the resulting specific heat value should be referred to the mean temperature of the two mean temperatures – i.e. to 12.5^0C.

Tests of several materials with known volumetric specific heat values were done to prove that this new method gives correct and reliable results. Our results agree within 3-5% with literature data for Expanded Polystyrene [6], for 1450b [10] - Standard Reference Material for thermal conductivity, for Vespel [11], for Pyrex [12, 13], for Pyroceram [12, 13], and for Stainless Steel 304 [13].

CONCLUSIONS

Possibilities of the traditional Heat Flow Meter method have been significantly extended using recorded signals of the heat flow meters versus time and presented algorithms to calculate additional important thermophysical properties – thermal diffusivity (for thermal insulations only) and volumetric specific heat. Comparison of results obtained using these new procedures with literature data showed good agreement and is a proof of their reliability and accuracy. Another one thermophysical property – thermal effusivity can be calculated from thermal conductivity and volumetric specific heat, so full set of four thermal properties now can be obtained using LaserComp's FOX Heat Flow Meter instruments with new updated versions of the WinTherm32 and WinTherm50 software.

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