Title: *Errors of the Heat Flow Meter Method Caused by Thermal Contact Resistance* for Thermal Conductivity 29 / Thermal Expansion 17 Conference

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ABSTRACT

Calculation formulas used in the Heat Flow Meter Method (ASTM C518, ISO 8301, EN 1946-3, etc.) are simple, but they are accurate only in the case of samples of low thermal conductivity, i.e. when thermal contact resistance is negligible in comparison with the sample's thermal resistance. For samples of intermediate thermal conductivity, those regular formulas become inaccurate both for calibrations of the Heat Flow Meter instruments and for tests, and the errors depend on the ratio of the contact and the sample's thermal resistances.

Two-thickness and Multi-thickness procedures of calibrations and tests [1] effectively eliminate thermal contact resistance errors. These procedures are used in LaserComp's FOX50 Heat Flow Meter instrument and WinTherm50 software both designed to obtain the best possible accuracy for thermal conductivity measurements of such samples. But in practice, very often, users of the FOX50 instruments have only single-thickness samples available for testing.

We have analyzed errors associated with thermal contact resistance for: a) single-thickness calibrations; b) single-thickness tests using single-thickness calibrations; c) single-thickness tests using two-thickness calibrations.

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INTRODUCTION

The Heat Flow Meter Method (ASTM C518, ISO 8301, EN 1946-3, etc.) is the most widely used traditional comparative steady-state method to measure thermal conductivity of thermal insulation materials. For low conductivity materials thermal contact resistance $2R$ (of two surfaces) is negligible in comparison with the samples' thermal resistances x/λ (thickness x divided by thermal conductivity λ):

$$2R << x/\lambda$$

and the simple formulas normally used for calibration factor S_{cal} and thermal conductivity λ_{test} calculations are valid:

$$S_{cal}=\lambda_{cal}\,\Delta T/(x_{cal}\,Q_{cal}) \qquad (1)$$

$$\lambda_{test} = S_{cal}\,x_{test}\,Q_{test}\,/\,\Delta T \qquad (2)$$

$$\lambda_{test} = \lambda_{cal}\,(x_{test}\,/x_{cal}\,)(Q_{test}\,)/\,Q_{cal}) \qquad (2a)$$

where ΔT is temperature difference between the plates (or more strictly – between the temperature sensors) – same for the calibrations and tests, Q are signals of the heat flow meters (transducers). These formulas are the result of the relationship between the steady-state heat flow density q (or heat flux, measured in Watts/meters^2) and all other parameters:

$$q = S_{cal}\,Q = \Delta T/(x/\lambda) \qquad (3)$$

This Fourier law expression looks like the well-known Ohm's law, where q is analogous to an electric current, ΔT – to voltage difference, and x/λ - to electric resistance. Physically, calibration factor S_{cal} is a heat flow density necessary to create 1 microvolt (or sometimes 1 millivolt) electric voltage (signal) on the heat flow transducer's output.

When testing materials of intermediate thermal conductivity ($\sim 0.1 < \lambda < \sim 20$ W/mK), and, a fortiori, in the case of higher conductivity materials, the thermal contact resistance $2R$ cannot be neglected. It must be excluded, otherwise significant errors may result – especially in the case of thin samples and/or of higher thermal conductivity when the thermal contact resistance $2R$ may even exceed the sample's thermal resistance x/λ. For example ¼" (6.35mm) thick Pyroceram has a thermal resistance x/λ=1.6x10^{-3} m^{2}K/W, whereas the thermal contact resistance $2R$ can be about 3x10^{-3} m^{2}K/W – or almost 2 times larger.

The total thermal resistance $R_{total} = x/\lambda + 2R$ should be used in the denominator of the Eq.(3). The corrected relation between the heat flow density q and all other parameters now is:

$$q = S_{cal}\, Q = \Delta T / (x/\lambda + 2R) \tag{3a}$$

The electric signal Q of the heat flow meter is proportional to the heat flow density q, which in a steady-state condition is equal to the temperature difference ΔT divided by the total thermal resistance - sum of thermal resistance of the sample x/λ and two thermal contact resistances $2R$, which, in general, includes not just contact resistance between adjoined surfaces, but all thermal resistance between temperature sensors and samples' surfaces. A crude and very labor-consuming way to exclude thermal contact resistance is using thermocouples placed directly into grooves machined on the sample to measure temperatures of the sample's surfaces. Use of thermo conductive grease will only diminish thermal contact resistance but not eliminate it. Depending on the amount of the applied grease and its thickness, its effect can vary from one case to another, and is not very repeatable.

TWO-THICKNESS METHOD

An accurate and effective way of excluding the thermal contact resistance is the two-thickness Method [1]. By using at least two samples of the same material with different thicknesses x_1 and x_2 a system of two equations containing two unknown values can be solved:

$$S_{cal}\, Q_1 = \Delta T / (x_1/\lambda + 2R) \tag{3b}$$

$$S_{cal}\, Q_2 = \Delta T / (x_2/\lambda + 2R) \tag{3c}$$

where Q_1 and Q_2 are signals from the heat flow transducers, x_1 and x_2 are the thicknesses of the thin and thick samples. We assume that the thermal contact resistances for both samples are the same. The solution of the system of the two Eqs. (3b) and (3c) for calibrations is:

$$S_{cal} = \Delta T\, \lambda_{cal}\, (Q_1 - Q_2)/[Q_1 Q_2 (x_2 - x_1)] \tag{4}$$

$$2R_{cal} = (x_2 Q_2 - x_1 Q_1)/[\lambda_{cal}\, (Q_1 - Q_2)] \tag{5}$$

For calibrations of LaserComp's FOX50 Heat Flow Meter instruments, four materials with known thermal conductivity [5-8] – Pyrex® 7740, Pyroceram® 9606, Vespel® DuPont™, and Perspex® are used (accuracy of the values is believed to be about 2-3%; ~5% for Pyroceram®):

TABLE I. THERMAL CONDUCTIVITY OF CALIBRATION MATERIALS, (W/mK)

T, ^{0}C	Perspex® [8]	Vespel®	Pyrex® 7740 [5]	Pyroceram® 9606, TPRC
0	0.1860	0.365	1.063	4.15
20	0.1885	0.371	1.086	4.04
40	0.1909	0.377	1.115	3.94
60	0.1933	0.386	1.145	3.85
80	-	0.389	1.175	3.78
100	-	0.396	1.203	3.71
For high temperature versions of the FOX50 HFM Instrument:				
150	-	0.411	1.270	3.58
200	-	0.426	1.330	3.49
250	-	0.441	1.391	3.42
300	-	0.457	1.452	3.34

Ideally, all the calibration runs should give the same values of the calibration factor no matter which method or what reference material was used for calibration, because the calibration factor is a physical property of the heat flow meter.

Let us analyze sensitivity function of the calibration factor and thermal contact resistance to the heat flow meter's signals:

$$(\partial S_{cal} / \partial Q)(Q/S_{cal}) = +/- \, (1 + Q_2^2 / Q_1^2) \, / \, (1^2 - Q_2^2 / Q_1^2) =$$

$$= +/- \, (1 + R^2_{1\ total} / R^2_{2\ total}) \, / \, (1 - R^2_{1\ total} / R^2_{2\ total});$$

$$(\partial 2R_{cal} / \partial Q_1)(Q_1 / 2R_{cal}) = - \, (R_{1\ total} / 2R_{cal}) \, [R_{2\ total} / (R_{2\ total} - R_{1\ total})]$$

To get the most accurate measurements the sensitivity functions should be neither big, nor small, i.e. ideally about either 1 or -1, thus the total thermal resistances and signals of the two calibration samples (thin - 1, and thick - 2) should be as different as possible:

$$Q_1 >> Q_2 \quad \text{or} \quad R_{1\ total} << R_{2\ total}$$

$$R_{1\ total} \sim 2R_{cal} \quad \text{and/or} \quad R_{2\ total} \sim (R_{2\ total} - R_{1\ total})$$

In other words, a thick sample should be as thick as possible (within the instruments capability), and a thin one – as thin as possible, but, of course, in case of a very thick or very thin samples, other limitations and sources of errors and distortions appear.

The solution of the system of the two Eqs. (3b) and (3c) for tests is:

$$\lambda_{test} = S_{cal}\, Q_1 Q_2 (x_2 - x_1)/[\, \Delta T\, (Q_1 - Q_2)] \tag{6}$$

$$2R_{test} = (x_2 Q_2 - x_1 Q_1)\, \Delta T/[(Q_1 Q_2 S_{cal}(x_2 - x_1)] \tag{7}$$

$$\lambda_{test} = \lambda_{cal}\, [(Q_{1\,cal} - Q_{2\,cal})/(Q_{1\,test} - Q_{2\,test})]\, [(Q_{1\,test} Q_{2\,test})/(Q_{1\,cal} Q_{2\,cal})]$$

$$[(x_{2\,test} - x_{1\,test})/(x_{2\,cal} - x_{1\,cal})] \tag{6a}$$

Eq.6a shows that in this comparative method thermal conductivity λ_{test} is calculated from the ratios and differences of values measured during the test and calibration, so practically all experimental bias errors are eliminated. Due to the good accuracy of the x and Q measurements, the accuracy of thermal conductivity results mostly depends on accuracy of the calibration sample's thermal conductivity λ_{cal}.

Thermal conductivity also can be calculated from the slope (its reciprocal value) of the graph of the total thermal resistance against the thickness of the samples of different thickness. This procedure is used to get the average results for tests of several samples of different thicknesses.

Sensitivity functions of thermal conductivity and thermal contact resistance to the heat flow meter signals are:

$$(\partial \lambda / \partial Q_2)(Q_2/\lambda) = 1 / (1 - R_{1\,total} / R_{2\,total})$$

$$(\partial 2R / \partial Q_1)(Q_1/2R) = -\, [1 + (x_1/\lambda)/(2R)] \,/\, (1 - x_1/x_2)$$

This means the same as in the previous case of calibrations. The difference of the samples' thermal resistances should be as big as possible to get better accuracy of the measurements. We may say that a thick sample test gives information mostly about thermal conductivity, and a thin sample test – gives information mostly about thermal contact resistance to be excluded in the calculation. The two-thickness procedure of calibrations and tests significantly improves accuracy of thermal conductivity measurements.

Let us analyze now errors associated with presence of thermal contact resistance when using regular single-thickness Eqs. (1) and (2) instead of the accurate two-thickness formulas - first for calibrations, and then for tests results.

SINGLE THICKNESS CALIBRATION

First we will get the formula for the relation between the single-thickness calibration factor $S_{1\text{-}thickness}$ and the correct calibration factor S_{cal} by substituting Q expressed from the accurate Eq. (3a):

$$Q = \Delta T / [(x/\lambda + 2R)\, S_{cal}] \tag{8}$$

into the regular, and not so accurate single-thickness Eq. (1):

$$S_{1\ thickness} = \lambda_{cal}\, \Delta T / \{x_{cal}\, \Delta T / [(x_{cal}/\lambda_{cal} + 2R_{cal})\, S_{cal}]\}$$

i.e. $$S_{1\ thickness} = 1 / \{(x_{cal}/\lambda_{cal}) / [(x_{cal}/\lambda_{cal} + 2R_{cal})\, S_{cal}]\} =$$

$$= [1 + 2R_{cal}/(x_{cal}/\lambda_{cal})]\, S_{cal} \tag{9}$$

The calibration factors become equal if thermal contact resistance is negligible in comparison with the sample's thermal resistance x_{cal}/λ_{cal}. Otherwise the single-thickness calibration factor is higher. The larger the $2R_{cal}/(x_{cal}/\lambda_{cal})$ ratio – the larger and less accurate is the single-thickness calibration factor.

Next we will analyze single-thickness tests done vs. single- and two-thickness calibrations.

SINGLE-THICKNESS TESTS vs. SINGLE-THICKNESS CALIBRATION

To get the formula for a single-thickness test we should substitute expression (8) for the single-thickness calibration factor, and expression (9) for the HFM signal Q during the test into the non-accurate Eq.(2):

$$\lambda_{1\text{-}thickness\ test} = S_{1\ thickness}\, (x_{test}/\Delta T)\, Q_{test} =$$

$$= [1+2R_{cal}/(x_{cal}/\lambda_{cal})]\, S_{cal}\, (x_{test}/\Delta T)\, \Delta T / [(x_{test}/\lambda_{test\ correct} + 2R_{test})\, S_{cal}] =$$

$$= [1+2R_{cal}/(x_{cal}/\lambda_{cal})]\, (x_{test}) / [(x_{test}/\lambda_{test\ correct} + 2R_{test})]$$

Thus, the value of the thermal conductivity $\lambda_{1\text{-}thickness\ test}$, calculated using the regular single-thickness formula (used in regular Heat Flow Meter instruments) is related to the correct thermal conductivity $\lambda_{test\ correct}$ as:

$$\lambda_{1\text{-thickness test}} = \lambda_{\text{test correct}} [1+2R_{\text{cal}}/(x_{\text{cal}}/\lambda_{\text{cal}})]/[1+2R_{\text{test}}/(x_{\text{test}}/\lambda_{\text{test correct}})]$$

where x_{cal}/λ_{cal} is the calibration standard's thermal resistance, and $x_{test}/\lambda_{test\ correct}$ is the sample's thermal resistance. This formula also can give a correct result when the ratios $2R/(x/\lambda)$ are the same for the calibration and for test by coincidence. In all other cases the thermal conductivity test results can be either higher or lower depending on the ratios of thermal resistances.

TABLE II. SINGLE-THICKNESS TESTS VS. SINGLE-THICKNESS CALIBRATIONS

	Tests		Pyroceram®	Pyrex®	Vespel®	Perspex®	
			6.35	6.60	6.18	5.00	mm
		UP	63031	31618	14123	9505	μV
		LP	-61795	-31012	-13797	-9242	μV
Ref.values	**Calibrations**						
(W/mK)	6.35	mm	X	2.091	0.875	0.476	W/mK
	63031	μV	X	2.092	0.872	0.472	W/mK
	-61795	μV	X	2.092	0.873	0.474	W/mK
4.011	Pyroceram®		**X**	**91.2**	**134.1**	**150.8**	%
	6.45	mm	2.15	X	0.468	0.255	W/mK
	31618	μV	2.15	X	0.466	0.253	W/mK
	-31012	μV	2.15	X	0.467	0.254	W/mK
1.094	Pyrex®		**-46.5**	**X**	**25.3**	**34.2**	%
	6.18	mm	1.71	0.892	X	0.203	W/mK
	14123	μV	1.72	0.895	X	0.202	W/mK
	-13797	μV	1.71	0.894	X	0.203	W/mK
0.373	Vespel®		**-57.3**	**-18.3**	**X**	**7.2**	%
	5	mm	1.59	0.830	0.347	X	W/mK
	9505	μV	1.61	0.838	0.349	X	W/mK
	-9242	μV	1.60	0.834	0.348	X	W/mK
0.1891	Perspex®		**-60.1**	**-23.8**	**-6.7**	**X**	%

Table II presents results of single-thickness tests at 25^{0}C vs. single-thickness calibrations using Eq.2a (i.e. without taking into account thermal contact resistances) of 4 standard materials (about ¼" thick) routinely used at LaserComp for calibrations of the FOX50 heat flow meter instruments – Pyroceram® vs. Pyrex® calibration, vs. Vespel®, vs. Perspex®, and vice versa, etc. – all 12 possible combinations. We can see that errors can be very significant – sometimes more than one hundred percent. This means that regular single thickness formulas and regular heat flow meter instruments can be unacceptable for materials in this range of thermal conductivities.

Next, we will analyze single-thickness test results obtained using thermal contact resistances from two-thickness calibration of the FOX50 instrument which is subtracted from the total thermal resistance of the single-thickness tests.

SINGLE-THICKNESS TESTS vs. TWO-THICKNESS CALIBRATION

In practice, very often, users of the FOX50 instruments have only single-thickness samples available for testing. In this case we would have to solve one equation with two unknowns which is impossible. The only more or less acceptable way is to use the second unknown, thermal contact resistance - one from the two-thickness calibration. To get an expression for a single-thickness test, thermal conductivity is calculated using a two-thickness calibration, where we have to substitute expression (4) for the HFM signal Q during the test, into the non-accurate Eq.(2):

$$\lambda_{1\text{-thickness test}} = S_{cal}\, x_{test}\, Q_{test} / \Delta T = S_{cal}\, x_{test} / [(x_{test} / \lambda_{test\ correct} + 2R_{test})\, S_{cal}] =$$

$$= x_{test} / [x_{test} / \lambda_{test\ correct} + 2R_{test}] = \lambda_{test\ correct} / [1 + 2R_{test} / (x_{test} / \lambda_{test\ \mathrm{correct}})$$

But this formula (which always gives lower thermal conductivity) is not used in the FOX50's "WinTherm50" software. When the single-thickness test is running on the FOX50 instrument using a two-thickness calibration, the "WinTherm50" software <u>subtracts</u> the thermal contact resistance $2R_{cal}$ obtained during the Two-Thickness calibration from the total thermal resistance calculated from the single-thickness test:

$$R_{total} = \Delta T / (S_{cal}\, Q_{test}) = x_{test} / \lambda_{test\ correct} + 2R_{test} =$$

$$= x_{test} / \lambda_{1\text{-thickness}} + 2R_{cal}$$

i.e.

$$x_{test} / \lambda_{1\text{-thickness}} = x_{test} / \lambda_{test\ correct} + 2R_{test} - 2R_{cal}$$

so the calculated single-thickness thermal conductivity is related to the correct thermal conductivity $\lambda_{test\ correct}$ as:

$$\lambda_{1\text{-thickness test}} = \lambda_{test\ correct} / [1 + (2R_{test} - 2R_{cal}) / (x_{test} / \lambda_{test\ correct})]$$

We can see from this formula that the thermal contact resistances may cause some error of the single-thickness tests especially in the case of samples with small thermal resistance (i.e. small thickness and/or high thermal conductivity), and when the contact resistance of the tested sample differs significantly from the calibration's contact thermal resistance.

TABLE III. SINGLE-THICKNESS TESTS VS. TWO-THICKNESS CALIBRATIONS

			Tests:	**Pyroceram®**	**Pyrex®**	**Vespel®**	**Perspex®**	
				6.35	6.60	6.18	5.00	mm
			Upper plate	63031	31618	14123	9505	µV
			Lower plate	-61795	-31012	-13797	-9242	µV
Two thickness calibrations:								
Pyroceram®		4.011	W/mK	X	1.149	0.385	0.199	W/mK
S cal u		0.0761	W/m^2µV	X	1.165	0.388	0.199	W/mK
S cal l		0.0783	W/m^2µV	X	1.157	0.386	0.199	W/mK
2R		0.00257	m^2K/W	**X**	**5.8**	**3.6**	**5.5**	%
Pyrex®		1.094	W/mK	3.696	X	0.373	0.193	W/mK
S cal u		0.0740	W/m^2µV	3.579	X	0.366	0.189	W/mK
S cal l		0.0745	W/m^2µV	3.638	X	0.369	0.191	W/mK
2R		0.00257	m^2K/W	**-9.3**	**X**	**-0.9**	**1.0**	%
Vespel®		0.373	W/mK	3.699	1.094	X	0.191	W/mK
S cal u		0.073	W/m^2µV	3.940	1.134	X	0.194	W/mK
S cal l		0.0763	W/m^2µV	3.819	1.114	X	0.193	W/mK
2R		0.00263	m^2K/W	**-4.8**	**1.8**	**X**	**1.8**	%
Perspex®		0.1891	W/mK	3.239	1.064	0.369	X	W/mK
S cal u		0.0743	W/m^2µV	3.249	1.067	0.368	X	W/mK
S cal l		0.0759	W/m^2µV	3.244	1.065	0.369	X	W/mK
2R		0.00231	m^2K/W	**-19.1**	**-2.6**	**-1.2**	**X**	%

We can see that errors of the single-thickness tests now are significantly smaller in comparison with Table 2 results. This means that single-thickness tests give much more accurate, reasonable results when two-thickness calibrations are used, especially when test samples and calibration materials have similar thermal conductivities.

CONCLUSIONS

Two-thickness procedures of calibrations and tests used in the LaserComp's FOX50 Heat Flow Meter instrument and its "WinTherm50" software provide excellent accuracy of thermal conductivity tests of materials like glasses, ceramics, plastics, polymers, etc. It was shown that old, simple formulas used in regular heat flow meter instruments give incorrect results for such materials because of the presence of thermal contact resistance that is not accounted for. It also was shown that single-thickness tests using two-thickness calibrations give reasonable results when the unknown sample's thermal contact resistance is replaced by the contact resistance of the two-thickness calibration. Formulas for relations between single-thickness and correct two-thickness parameters were developed to help to understand the influence of the thermal contact resistance.

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