Title: Fast Measurements of Absolute Thermal Conductivity Excluding Thermal Contact Resistance Errors

Thermal Conductivity 28 / Thermal Expansion 16 Conference

Authors: Akhan Tleoubaev
Andrzej Brzezinski

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

ABSTRACT

Two-Thickness and Multi-Thickness procedures [1] used in LaserComp's FOX50 Heat-Flow-Meter instruments enable the total exclusion of thermal contact resistance. Measurements without taking thermal contact resistance into account may result in very large errors (as much as hundreds percent in case of thin specimens of high thermal conductivity).

Recently, a new combined Guarded Hot Plate and Heat-Flow-Meter Method [2] was developed. This combination of the two widely used traditional steady-state methods (i.e. ASTM C177 / ISO 8302, and ASTM C518 / ISO 8301) provides very accurate *absolute* (i.e. not related to a calibration standard) values of thermal conductivity of many important materials such as ceramics, glasses, plastics, rocks, polymers, composites, fireproof materials, etc.

A thin flat guarded heater of known area is placed between two flat-parallel specimens of the same material and of different thicknesses. The stack is clamped between two isothermal plates each having a heat flow meter and a temperature sensor. After applying a constant electric power to the heater the heat fluxes through each of the two specimens (and, consequently, the two heat flow meters' signals) eventually reach some final thermal equilibrium values. Each of the equilibrium heat fluxes is inversely proportional to the respective total thermal resistance – sum of each specimen's thermal resistance (thickness divided by thermal conductivity) and two surface contact resistances (which are assumed to be the same for the two specimens). The absolute thermal conductivity of the specimens then can be calculated using the measured electric power of the center of the heater, temperatures of the heater and of the plates, thicknesses of the two specimens, and the two heat-flow meters signals (Eq.1).

The Combined Guarded Hot Plate and Heat-Flow-Meter Method now has been modified and tested for fast thermal conductivity measurements. A few minutes after applying the electric power (i.e. long before reaching the final thermal equilibrium, after reaching so-called "regular regime") the absolute thermal conductivity of the specimens can be calculated using both heat-flow meters' readings arrays and a special mathematical "prediction" procedure. Experimental checks (using reference materials – Pyrex® 7740, Vespel® SP1) of the new procedures were completed.

Mathematical algorithms and Mathcad programs were developed using Prony's method to determine if the system has reached the "regular regime", and using the Least Squares methods to calculate parameters of the system's exponential relaxation toward the final thermal equilibrium.

INTRODUCTION

The equation for thermal conductivity, λ, based on our new combined method [2] is:

$$\lambda =[(L_2 - L_1) /(T_h - T_p)] \ (W/A) / [(Q_1 /Q_2)/(Q_{1c} /Q_{2c}) - (Q_{1c} /Q_{2c})/(Q_1 /Q_2)] \quad (1)$$

where L_1 and L_2 are the two specimens thicknesses, T_h is temperature of the heater , T_p is temperature of the plates, W/A is electric power of the heater, W, divided by its area, A, Q_1 and Q_2 are the heat flow meter signals, $Q_{1c}/Q_{2c}\approx 1$ is the ratio of the heat-flow meter signals during the same specimens calibration procedure used to eliminate the effect of the two heat-flow meters difference [2].

In all the steady-state methods the thermal conductivity can be calculated only after reaching the full thermal equilibrium – i.e. only after the Fourier number $Fo = kt/L^2 >>1$, where k is the thermal diffusivity, t is time, and L is thickness of the specimen. The thermal diffusivity, k, for materials like Pyrex 7740, Pyroceram 9606, and Vespel 1, is about 10^{-6} m^2/s [3, 4], so a 20 mm-thick specimen theoretically needs at least an hour. In practice, usually longer times are necessary for the system to achieve steady state.

By recording the temperature of the heater and the two heat flow meter signals after turning on the electric power, it is theoretically possible then to calculate both thermal conductivity and thermal diffusivity of the specimens, to exclude both thermal contact resistances (between heater and specimens, and between specimens and plates). A.Tleoubaev (using boundary conditions of 3rd kind and separation of variables) recently has found the analytical solution for this transient thermal problem. The solution is not presented here because it is cumbersome, and most probably, is hardly to be used in practice. Shirtliffe [5], and

Flynn and Gorthala [6] considered similar transient thermal problems for flat specimens without contact resistance.

Theoretical consideration of the transient heat conduction in finite bodies for large *t* shows that series of the thermal problem general solution [7]

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

(where Λ_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"* [7]:

$$T(x,t) \approx C_1\, v_1(x)\, exp\,\{-\, k\, \Lambda_1\, t\} \tag{3}$$

This can be used in practice for fast tests of thermal conductivity (and, in future, probably, for thermal diffusivity calculations or, at least, estimations) using the new combined Guarded Hot Plate and Heat-Flow-Meter method - i.e. result can be "predicted" long before the system reaches the steady state. In our one-dimensional case the general solution is [8]:

$$T(x,t) = \sum_{n=1}^{\infty} \exp\{-k\Lambda_n t\}[A_n \cos(\Lambda_n^{1/2} x) + B_n \sin(\Lambda_n^{1/2} x)] \tag{4}$$

where A_n and B_n are coefficients, and Λ_n are eigenvalues, which should be determined from the problem's boundary and initial conditions (two separate sets - for each of the specimens by solving transcendent equations [8], [9]). So, after a certain moment of time only a single (number 1) exponent "survives", and the temperature distribution's evolution inside the flat specimen can be described as:

$$T(x,t) \approx \exp\{-k\Lambda_1 t\}[A_1 \cos(\Lambda_1^{1/2} x) + B_1 \sin(\Lambda_1^{1/2} x)] \tag{4a}$$

The single-exponent relaxation of the temperature field in the "regular regime" dramatically simplifies the mathematical formulas, and is very useful for practice. As a result, the "current" thermal conductivity calculated using Eq.(1) relaxes to its final equilibrium value exponentially, and quite fast (because ratio of the signals Q_1/Q_2 relaxes faster than the individual signals Q_1 and Q_2), and it can be "predicted" using its approximation by a single exponent.

“PREDICTION” PROCEDURES

It is important to determine the time required for a system to reach the “regular regime”, i.e. when the evolution of the calculated (using Eq. (1)) “current” thermal conductivity at large time *t* can be described by a single exponent. Prony’s method [10] using five equidistant (with, say, ~1 minute intervals) values of the calculated “current” thermal conductivity seems to be the most appropriate for the task. The *Mathcad 2000 Professional* procedure (shown in Appendix 1) was used to calculate the parameters of the two exponents (in the case of Pyrex specimens 6.60 mm and 19.08 mm thick). The procedure predicted the Pyrex’s thermal conductivity value of 1.094 W/mK very accurately long before reaching steady state. The value of one of the exponents turned out to be negligibly small (2.084×10^{-9}), which means that the system reached the “regular regime”.

Fig. 1 and Fig.2 show examples how the calculated exponents perfectly match the experimental relaxation curves in case of Pyrex and Vespel specimens (standard calibration materials routinely used to calibrate LaserComp’s FOX50 Heat Flow Meter instruments).

To get the best possible accuracy the array of experimental points should be used for calculations using the Least Squares Method to find three parameters of the single exponent:

- some initial value λ_0 (at time zero);
- final value λ_∞ at infinite time; and
- characteristic number of the exponent i_e , related with rate of the relaxation, i.e. number of the point when the difference $\lambda_0 - \lambda_\infty$ diminishes *e* times ($e = 2.71828\ldots$).

Analytical formula describing the array of λ_i ($i = 1 \ldots N$) is:

$$\lambda(i) = (\lambda_0 - \lambda_\infty) \exp\{- i / i_e\} + \lambda_\infty \qquad (5)$$

These three parameters can be calculated using the Method of Least Squares to “predict” the final value λ_∞ of thermal conductivity at infinite time. The calculated i_e parameter should be related with the value of the specimens’ thermal diffusivity $k = \lambda / C_p \rho$. We used the Mathcad’s *“genfit”* generalized regression

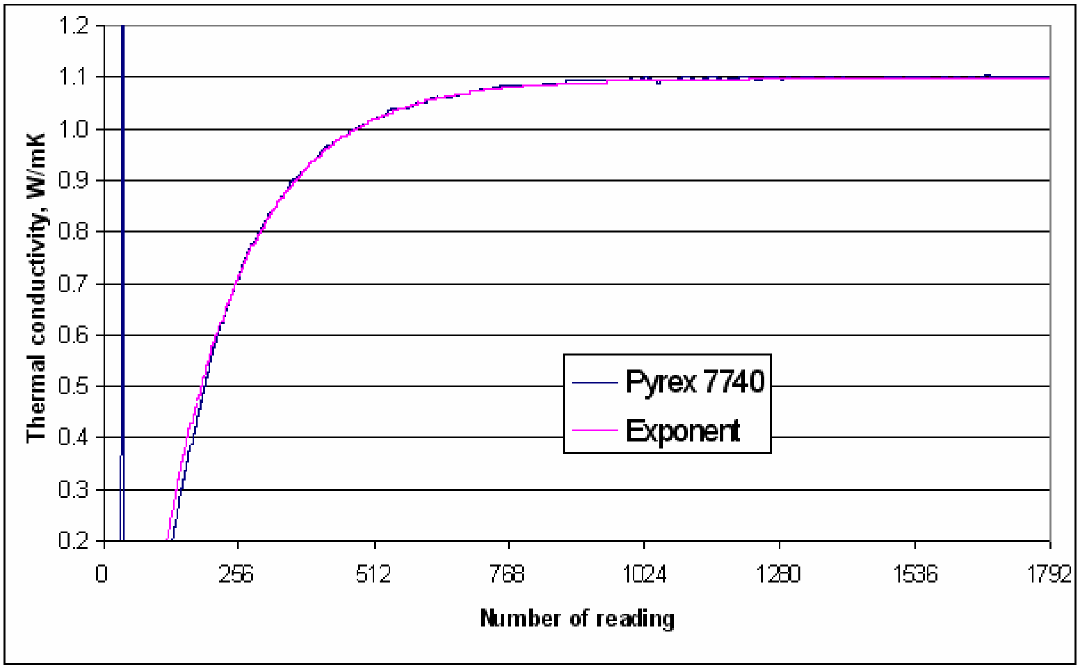


Figure 1. Comparison of the calculated exponent and experimental relaxation of the calculated "current" thermal conductivity of the Pyrex 7740 specimens (each reading was taken every 0.64 seconds).

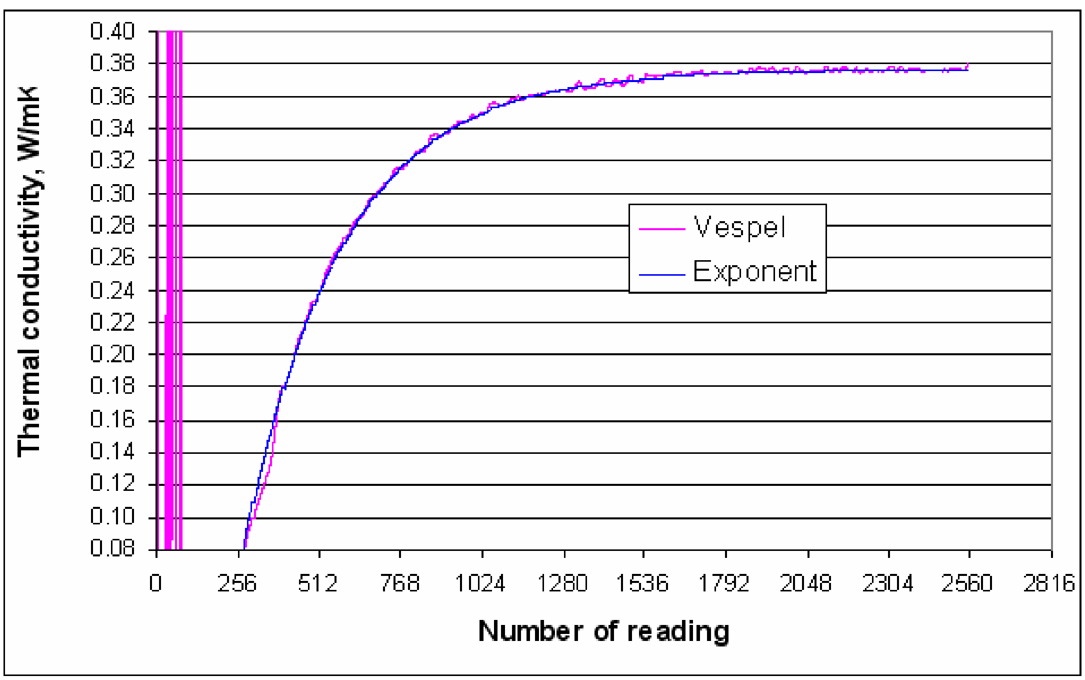


Figure 2. Comparison of the calculated exponent and experimental relaxation of the calculated "current" thermal conductivity of the Vespel specimens (each reading was taken every 0.64 seconds).

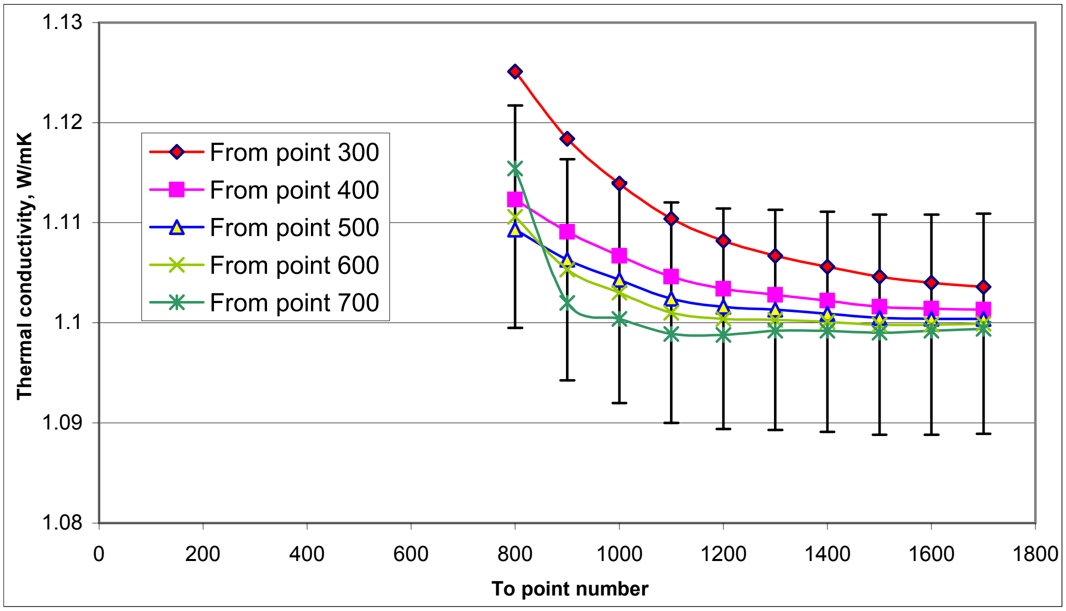


Figure 3. Thermal conductivity calculated using Least Squares Fit for various experimental points windows (Pyrex λ=1.10 W/mK, error bars +/- 1%, point number 1000 corresponds ~ 10 minutes)

function [11] to calculate the parameters. The "*genfit*" function uses three partial derivatives with respect to the three parameters, and their initial guess values (see Appendix 2).

Sum of squares of differences between analytical, $\lambda(i)$, and experimental, λ_i values is so-called residue function, $F(\lambda_0, \lambda_\infty, i_e)$, to be made as small as possible:

$$F(\lambda_0, \lambda_\infty, i_e) = \sum_{i=1}^{N} [\lambda(i) - \lambda_i]^2 = \sum_{i=1}^{N} [(\lambda_0 - \lambda_\infty)\exp\{-i/i_e\} + \lambda_\infty - \lambda_i]^2 \Rightarrow \min$$

The three parameters, λ_0 , λ_∞ and i_e are being calculated by the Mathcad *genfit* function in iterations (using Levenberg-Marquardt algorithm) to get better and better accuracy until the residue function $F(\lambda_0, \lambda_\infty, i_e)$, becomes small enough to guarantee that the calculated set of the parameters adequately describes the experimental exponent. Figure 3 demonstrates how the described algorithm calculates the "predicted" values of thermal conductivity of Pyrex for various sets (or windows) of experimental points.

CONCLUSIONS

It was shown and experimentally checked that fast and accurate measurements of absolute thermal conductivity excluding thermal contact resistance errors are possible using new combined Guarded Hot Plate and Heat-Flow-Meter method long before reaching the steady state - when the system is in “regular regime”.

Prony’s method was proposed and used to determine the point when the “regular regime” begins, i.e. when a single exponent can be used to describe the thermal relaxation of the system. The Least Squares method can be used to calculate accurately three parameters of the exponential relaxation to calculate thermal conductivity (and in future, probably, thermal diffusivity as well).

REFERENCES

1. Brzezinski, A., and A.Tleoubaev. 2002. "Effects of Interface Resistance on Measurements of Thermal Conductivity of Composites and Polymers," - in: *Proceedings of the 30th Annual Conference on Thermal Analysis and Applications (NATAS),* Pittsburgh, Pennsylvania. Ed.Keith J.Kociba, The Lubrizoil Corp., B&K Publishing, pp.512-517.
2. Tleoubaev, A., and A. Brzezinski. 2003. "Combined Guarded Hot Plate and Heat Flow Meter Method Excluding Thermal Contact Resistance," – in: *Thermal Conductivity 27*, DES*tech* Publications, Inc., pp.502-510.
3. Assael, M.J., K.Gialou, K.Kakosimos, and I.Metaxa. 2003. “Thermal Conductivity of Reference Solid Materials,” presented at the 15th Symposium on Thermophysical Properties, June 22-27, 2003.
4. Jacobs-Fedore, R.A., and D.E. Stroe. 2003. “Thermophysical Properties of Vespel™ SP1,” – in: *Thermal Conductivity 27*, DES*tech* Publications, Inc., pp.231-238.
5. Shirtliffe, C.J. 1974. “Establishing Steady-State Thermal Conditions in Flat Specimens,” – in: *Heat Transmission Measurements in Thermal Insulations, ASTM STP 544,* American Society for Testing and Materials, pp.13-33.
6. Flynn, D.R., and R. Gorthala. 1997. “Early-Time Estimation of the Thermal Resistance of Flat Specimens – Theoretical Analysis for Pure Conduction in a Homogeneous Material,” – in: *Insulation Materials: Testing and Applications: Third Volume, ASTM STP 1320,* R.S.Graves and R.R. Zarr, Eds., American Society for Testing and Materials, pp.381-400.
7. Tikhonov, A.N., and A.A.Samarskii. 1963 *Equations of Mathematical Physics*, Dover Publications, Inc., pp. 515-516.
8. Carslaw, H.S., and J.C.Jaeger, 1959. *Conduction of Heat in Solids,* 2nd edition, Oxford at the Clarendon Press, pp. 114-120.
9. Polyanin, A.D., 2001. *Handbook of Linear Partial Differential Equations for Engineers and Scientists,* Chapman & Hall/CRC, p.48
10. Hamming, R.W. 1986. *Numerical Methods for Scientists and Engineers,* 2nd edition, Dover Publications, Inc., pp. 620-624.
11. Mathcad 2000 *User’s Guide,* MathSoft, Inc., 1999, pp. 171-172.

Express Absolute Two-Thickness Procedure (using Prony method) by Akhan Tleoubaev

datafile := "S:\Documents\PAPERS\ThermalCond 28 Conference\Pyrex.txt"   Q := READPRN(datafile)

$n0 := 256 \qquad n := 128$

$f0 := Q_{n0,4} \qquad f1 := Q_{n0+n,4} \qquad f2 := Q_{n0+2\cdot n,4} \qquad f3 := Q_{n0+3\cdot n,4} \qquad f4 := Q_{n0+4\cdot n,4}$

$f0 = 0.709 \qquad f1 = 0.923 \qquad f2 = 1.018 \qquad f3 = 1.06 \qquad f4 = 1.086$

System to find the characteristic equation coefficients:

$$Sys := \begin{pmatrix} 1 & 1 & 1 \\ f0 & f1 & f2 \\ f1 & f2 & f3 \end{pmatrix} \qquad \text{Vector of the right sides:} \quad v := \begin{pmatrix} -1 \\ -f3 \\ -f4 \end{pmatrix}$$

$C := \text{lsolve}(Sys, v)$

$$\text{Solution} \quad C = \begin{pmatrix} 19.081 \\ -61.622 \\ 41.541 \end{pmatrix} \qquad C_1 = 19.081 \qquad C_2 = -61.622 \qquad C_3 = 41.541$$

Characteristic equation:
$ro^3+C_2ro^2+C_1ro+C_0=0$
(indexes are shifted by 1)

$$CharEq := \begin{pmatrix} C_1 \\ C_2 \\ C_3 \\ 1 \end{pmatrix} \qquad CharEq = \begin{pmatrix} 19.081 \\ -61.622 \\ 41.541 \\ 1 \end{pmatrix}$$

Roots of the characteristic equation:

$roroots := \text{polyroots}(CharEq)$

$$roroots = \begin{pmatrix} -42.984 \\ 0.444 \\ 1 \end{pmatrix} \qquad roroots_1 = -42.984 \qquad roroots_2 = 0.444 \qquad roroots_3 = 1$$

$$System := \begin{bmatrix} 1 & 1 & 1 \\ 1 & roroots_1 & roroots_2 \\ 1 & (roroots_1)^4 & (roroots_2)^4 \end{bmatrix} \qquad \text{Vector of the right sides:} \quad vv := \begin{pmatrix} f0 \\ f1 \\ f4 \end{pmatrix}$$

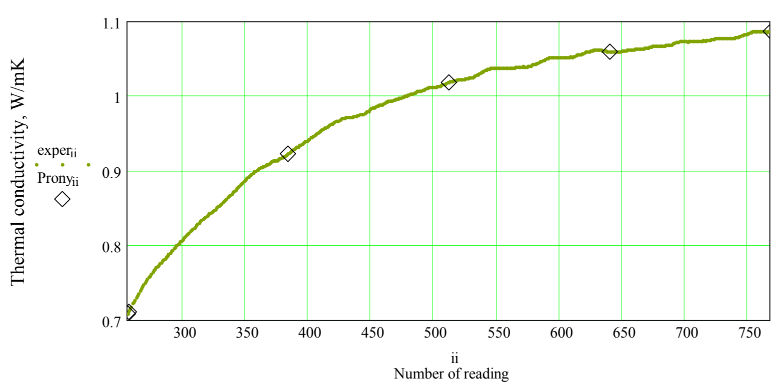
$AAA := \text{lsolve}(System, vv)$

$$AAA = \begin{pmatrix} 1.094 \\ 2.084 \times 10^{-9} \\ -0.385 \end{pmatrix} \qquad AAA_1 = 1.094 \qquad AAA_2 = 2.084 \times 10^{-9} \qquad AAA_3 = -0.385$$

$ii := n0 .. n0 + 4 \cdot n \qquad exper_{ii} := Q_{ii,4} \qquad exper_{n0+4\cdot n} = 1.086$

$$Prony_{ii} := AAA_1 + AAA_2 \cdot (roroots_1)^{\frac{ii-n0}{n}} + AAA_3 \cdot (roroots_2)^{\frac{ii-n0}{n}}$$

$Prony_{n0} = 0.709 \qquad Prony_{n0+4\cdot n} = 1.086$



Appendix 1. Mathcad procedure for calculations of parameters of two unknown exponents using Prony's method (Pyrex specimens of 6.60 and 19.08 mm thick).

MATHCAD LEAST-SQUARE FIT OF EXPONENT

Calculates 3 parameters of exponential relaxation:
- initial value(u0),
- infinite time value (u1) - is most important for us, and
- number of the reading (u2), when the difference (u-u1) becomes e tim
(e=2.71828...) smaller than initial difference (u0-u1).

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The fit function of 3 parameters and its 3 partial derivatives:

$$F(z,u) := \begin{bmatrix} (u_0 - u_1)\exp\left(\frac{-z}{u_2}\right) + u_1 \\ \exp\left(\frac{-z}{u_2}\right) \\ -\exp\left(\frac{-z}{u_2}\right) + 1 \\ (u_0 - u_1)\exp\left(\frac{-z}{u_2}\right)\cdot\frac{z}{(u_2)^2} \end{bmatrix}$$

$Q := \text{READPRN}(\text{datafile})$

$\text{First} := 400$ $\text{Last} := 1200$

$j := \text{First}..\text{Last}$

$vx_j := Q_{j,0}$ $vy_j := Q_{j,3}$

Initial guess values: $vg := \begin{pmatrix} Q_{First,3} \\ Q_{Last,3} \\ 300 \end{pmatrix}$

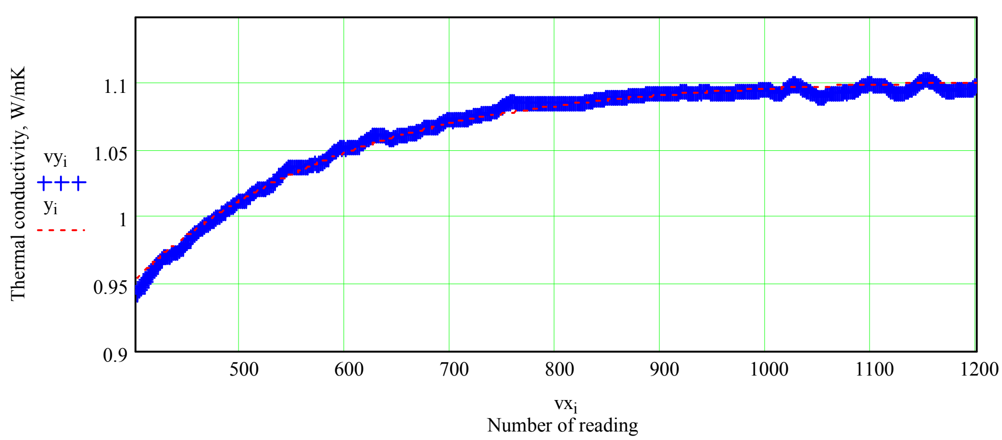
$Q_{First,3} = 0.942$

$Q_{Last,3} = 1.099$

$P := \text{genfit}(vx, vy, vg, F)$

$$P = \begin{pmatrix} -3.2816\times 10^{-5} \\ 1.1034 \\ 200.4182 \end{pmatrix}$$

$i := \text{First}..\text{Last}$ $y_i := (P_0 - P_1)\exp\left[\frac{(-vx)_i}{P_2}\right] + P_1$



Appendix 2. Mathcad procedure of the exponential Least Square fit applied to the "current" thermal conductivity array relaxing to the final steady-state value (Pyrex specimens of 6.60 and 19.08 mm thick).