SIZE EFFECTS AND BEYOND-FOURIER HEAT CONDUCTION IN ROOM-TEMPERATURE EXPERIMENTS

A. FEHÉR¹, N. LUKÁCS¹, L. SOMLAI¹,², T. FODOR¹, M. SZÜCS¹³, T. FÜLÖP¹³, P. VÁN¹³, R. KOVÁCS¹²³

Abstract. It is a long-lasting task to understand heat conduction phenomena beyond Fourier. Besides the low-temperature experiments on extremely pure crystals, it has turned out recently that heterogeneous materials with macro-scale size can also show thermal effects that cannot be modelled by the Fourier equation. This is called over-diffusive propagation, different from low-temperature observations, and is found in numerous samples made from metal foam, rocks, and composites. The measured temperature history is indeed similar to what Fourier’s law predicts but the usual evaluation cannot provide reliable thermal parameters. This paper is a report on our experiments on several rock types, each type having multiple samples with different thicknesses. We show that size-dependent thermal behaviour can occur for both Fourier and non-Fourier situations. Moreover, based on the present experimental data, we find an empirical relation between the Fourier and non-Fourier parameters, which may be helpful in later experiments to develop a more robust and reliable evaluation procedure.

1. Introduction

Fourier’s approach to heat conduction is one of the most widely used, and indeed successful, model in continuum physics. It expresses a relationship between the temperature gradient \( \nabla T \) and the heat flux \( q \),

\[
q = -\lambda \nabla T,
\]

in which \( \lambda \) is the thermal conductivity, a scalar for isotropic materials. It provides a reliable way of explaining the vast majority of thermal problems encountered in engineering practice. Despite its success, various extensions might be necessary depending on the physical situation. For instance, notable inertial (memory) effects appear in a low-temperature (< 20 K) case, and spatial nonlocalities become observable in both heterogeneous materials and nano-systems [1–6], including boundary effects as well [7].

According to the preceding flash experiments on heterogeneous materials [8,9], the next reasonable candidate is the Guyer–Krumhansl (GK) equation (presented in one spatial dimension),

\[
\tau \partial_t q + q = -\lambda \partial_x T + \kappa^2 \partial_{xx} q,
\]

in which \( \tau \) is the relaxation time, and \( \kappa^2 \) is a kind of ‘dissipation parameter’. The Maxwell–Cattaneo–Vernotte (MCV) equation is the special case with \( \kappa^2 = 0 \), however, no successful observation of second sound (damped wave propagation of heat) has been established under room-temperature conditions at macroscale in heterogeneous solids. Therefore, despite the parabolic property of the GK equation, the nonlocal \( \partial_{xx} q \) term becomes necessary for proper modelization of the observed phenomenon. Including \( \partial_{xx} q \) in the constitutive equation allows to properly characterize the so-called over-diffusive propagation, depicted in Fig. 1. As it is visible, in such a situation, the measured temperature signal is faster at the beginning than what Fourier’s law predicts. At the top, usually around 80 % of the asymptotic value, the deviation is the most significant. Apparently, in that region, Fourier’s law predicts a faster temperature rise. The asymptotic values are the same in both theories.

In our previous series of experiments [8,10], we made the following observations when the GK model was necessary. First, we found different thermal diffusivity than one would find using Fourier’s law, which appeared to be always smaller. Second, we observed over-diffusive propagation exclusively, i.e., we always found \( \kappa^2/\tau > \alpha \) in every case \( (\alpha = \lambda/(\rho c)) \) being the thermal diffusivity, with density \( \rho \) and specific heat \( c \) when the Fourier’s law was not applicable, and the reverse case \( (\kappa^2/\tau < \alpha) \) has not yet appeared. When the equality \( \kappa^2/\tau = \alpha \) holds, we call it Fourier resonance condition as that setting recovers the solutions of Fourier equation [9,11]. On this basis, it is reasonable to introduce a parameter \( B = \kappa^2/(\tau \alpha) \), which typifies the ‘non-Fourierness’ of the heat conduction process.

Later on, it turned out that both the thermal diffusivity \( \alpha \) and the parameter \( B \) can be size-dependent with respect to the thickness, observed on basalt rock samples [10]. Therefore, here, we focus on investigating that size dependence more closely. To this end, we have performed multiple measurements on various rock samples, each of the specimens having at least three different thicknesses with the same diameter.

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This research is challenging from multiple aspects. First, on contrary to [12, 13], the exact microstructure together with the constituents are all unknown, therefore the calculation of effective ('averaged') thermal conductivity on theoretical basis is not possible. Although size effects of thermal properties are observed in superlattices on nanoscale due to the parallel existence of various heat conduction mechanisms [14–17], its macroscale appearance is surprising: as Fig. 3 shows later, the pores are much smaller than the sample thickness, hence their effect is expected to be averaged at the end of the sample, at least to say that one measures the bulk thermal properties. Seemingly, this is not the case and that requires further understanding and experimental work. Third, the reliable evaluation of non-Fourier temperature history needs a robust algorithm. While it is quick and simple in the Fourier case without any complex optimization procedure, this is not that straightforward for the Guyer-Krumhansl equation. Recently, a novel evaluation procedure is developed [18], and the present series of experiments are helpful to test and improve this algorithm.

2. Settings and evaluation of the experiments

In the present series of experiments, we utilize the flash (or heat pulse) method due to its relatively simple arrangement, and wide measuring range for thermal diffusivity [19–21]. In order to keep the heat conduction process in one spatial dimension as much as possible, the samples are thin relative to their diameter (25 mm, fixed for every sample) and, additionally, the entire front side is excited by the heat pulse.

The heat pulse is produced by a flash lamp and lasts 0.01 second ($t_p = 0.01 \text{s}$). That flash also serves as a trigger signal for temperature detection, captured by a photovoltaic sensor. The temperature history is measured with a K-type thermocouple, the proper surface contact ensured by a thin silver layer on the rear side. The front of each sample is coated with black graphite paint to achieve high absorption on the surface. The measured temperature history is recorded with a PC oscilloscope during the measurements, and the received data is processed in Matlab environment. Measurements are conducted multiple times on each sample without taking them out from the equipment to assure the same environment and let the temperature relax to a steady state. That relaxation period lasts one hour for each measurement.

The evaluation procedure follows [18], in which an analytical solution for the GK equation is presented together with a reasonable simplification for this arrangement. It includes temperature-dependent convective heat transport on the rear side as well. The simplified rear-side temperature history is expressed as the first term of an infinite series,

$$
\hat{T}(\hat{x} = 1, \hat{t} > 30) = Y_0 \exp(-\hat{h}\hat{t}) - Z_1 \exp(x_1\hat{t}) - Z_2 \exp(x_2\hat{t}), \quad x_2 < x_1 < 0,
$$

(3)
in which all quantities are dimensionless, based on the following definitions:

\[
\begin{align*}
\hat{t} &= \frac{t}{t_p} \quad \text{and} \quad \hat{x} = \frac{x}{L} \quad \text{with sample thickness } L; \\
\hat{\alpha} &= \frac{\alpha t_p}{L^2} \quad \text{with} \quad \alpha = \frac{\lambda}{\rho c}; \\
\hat{q} &= \frac{q}{\bar{q}_p} \quad \text{with} \quad \bar{q}_p = \frac{1}{t_p} \int_0^{t_p} q_p(t) dt \quad \text{(average pulse heat flux)}; \quad (4) \\
\hat{T} &= \frac{T - T_0}{T_{\text{end}} - T_0} \quad \text{with initial temperature } T_0 \text{ and } T_{\text{end}} = T_0 + \frac{\bar{q}_p t_p}{\rho c L}; \\
\hat{h} &= \frac{h}{\rho c} \quad \text{with heat transfer coefficient } h.
\end{align*}
\]

Additionally, \(x_{1,2}\) are characteristic exponents of the GK equation, representing two different time scales. Interestingly, the characteristic exponent of the Fourier prediction, \(x_F = -\pi^2 \hat{\alpha} < 0\) in the solution \(\hat{T}(\hat{x} = 1, \hat{t} > 30) = -2 \exp(x_F \hat{t})\), proves to be always between them, i.e., \(|x_1| < |x_F|\) and \(|x_F| < |x_2|\) \[18\]. Consequently, \(x_1\) influences the deviation at the top the most, and can be best determined using data from this region. On the other hand, \(x_2\) is responsible for the initial part of the temperature history. That recognition is useful in the separation of various time scales, enabling the reliable determination of the GK parameters.

Overall, the evaluation procedure consists of the following steps:

1. We determine the heat transfer coefficient using
\[
\hat{h} = -\frac{\ln(\hat{T}_2/\hat{T}_1)}{t_2 - t_1}, \tag{5}
\]
taking two instants \(t_1, t_2\) (and corresponding temperatures \(T_1, T_2\)) where cooling (as a third time scale) is apparently significant and already dominates the process. Starting with the Fourier equation, the thermal diffusivity can be immediately determined with
\[
\hat{\alpha}_F = \frac{\ln 4}{\pi^2 t_{1/2}}, \quad \alpha_F = \frac{\ln 4}{\pi^2 t_{1/2}}, \tag{6}
\]
in which \(t_{1/2}\) is the time needed to reach the half of the adiabatic asymptotics (i.e., where \(\hat{T} = 0.5\)).

2. Having \(h\) and \(\alpha_F\) allows for the Fourier solution to be constructed and compared to the measured data. Fine tuning of \(h\) and \(\alpha_F\) is often necessary.
(3) In case Fourier’s law is proved insufficient, the GK parameters are to be determined. It is advantageous to start with $x_1$, expressed as

$$x_1 = x_F \frac{t_{F1} - t_{F2}}{t_{m1} - t_{m2}} \quad \text{with} \quad x_F = -\pi^2 \hat{\alpha}_F,$$

where the time instants $t_{m1}, t_{m2}$ are taken from the vicinity of the largest deviation, $T_{m1}, T_{m2}$ are the corresponding measured temperatures, and $t_{F1}, t_{F2}$ are the instants to which the Fourier solution assigns $T_{m1}$ and $T_{m2}$, respectively. In other words, this expression modifies $x_F$ as a function of the deviation occurring between the Fourier and the measured curves and determines the primary time scale.

(4) As a final step, $x_2$, and consequently the remaining GK parameters, can be determined; we refer to [18] for the details.

3. Rock samples and experiment outcome

We investigated seven different rock types and, for each type, samples with different thickness to detect the size dependence of thermophysical properties. All have the same constant diameter of 25 mm. All samples have their origin in Hungary, coming mostly from Máriagyűd stone pit and well-boring at the middle part of the country. The samples have been produced by ROCKSTUDY Ltd. (Kömérő Kft), Hungary. We note that there are two different Szászvár formation-type samples, having a little bit different structures: the I. is more likely coarse-grained than the type II. The differences are visible in Figure 3. Table 1 summarizes the thermal parameters we found for all rock samples. In that table, the sample ‘ID’ is equivalent to the numbering in Fig. 3. Let us recall the parameter $B = \kappa^2/(\tau \alpha)$, which characterizes the deviation from Fourier’s law. For over-diffusive propagation, $B > 1$ holds.

Figure 3. The prepared samples, all having the same diameter of 25 mm. In order: Szársonlyó limestone formation (1); Szászvár formation I. (2); Szászvár formation II. (3); Tisza metamorph komplex (4); Boda Claystone formation (5); Dark grey basalt (6); Mátra andesite formation (7).

It is visible that thermal diffusivity varies with thickness, even when all samples for the same type behave as Fourier’s law predicts. Therefore, this size effect is not the consequence of the non-Fourier behaviour, it occurs independently, although it could be small (e.g., ID-7/(a)-(c)). Interestingly, size dependence is not necessarily monotonous with respect to the thickness: samples 1, 2 and 4 show remarkable changes with the thickness for both
heat conduction models. Regarding the GK parameters—when this model was necessary to apply—size dependence also appears but in a non-monotonous way; samples 1, 2 and 3 are good examples for this behaviour. Interestingly, the magnitude of $\tau$ and of $\kappa^2$ falls into the same order for all samples, and it is in accordance with our previous measurements on Villány limestone \[9\]. The ratio $\kappa^2/\tau$ is also always at the order of the thermal diffusivity. Moreover, it is not simply at the same order but the following expression also holds:

$$\alpha_F \approx \frac{1}{2} \left( \alpha_{\text{GK}} + \frac{\kappa^2}{\tau} \right).$$

(8)

In other words, the best achievable thermal diffusivity with the Fourier theory appears to be the average of the GK parameters, demonstrated for all samples in Fig. 4. Since the Guyer-Krumhansl model always predicts lower thermal diffusivity, it restricts the ratio of $\kappa^2/\tau$, which could be either a new constraint or a checkpoint in the evaluation procedure, despite its empirical nature. With two exceptions, Eq. (8) holds for all samples within ±5\% error. We show two examples of over-diffusive propagation. The first one (Fig. 5) is related to the Szársonlyó limestone sample (ID-1/b), presenting a stronger deviation than the second one (Fig. 6) on Szászvár formation I (ID-2/a). These figures are helpful in the interpretation of parameter $B$: while the first has $B = 1.294$, it looks significantly more substantial than the second one ($B = 1.21$).

That outcome reflects how difficult it is to prepare a standard sample from a strongly heterogeneous material and to measure its thermal properties. The large variety of heterogeneity is one reason behind the diverse results: the porosity, structural defects and material composition can be different, even for the same type. Consequently, we plan further research on size dependence since it would have a serious impact on how we understand the role of thermal conductivity, the primary factor in thermal diffusivity.

Nevertheless, the non-Fourier behaviour remains apparent. Despite that the source of heterogeneity could differ in each sample, it still ensures the existence of parallel time and spatial scales, as it is discussed in detail in \[22\]. It seems natural to observe size dependence of such effects, and when there is enough space between the points of excitation and measurement, the non-Fourier effects can either be extinct or appear, depending on both the material properties and the heterogeneity present.
Table 1. The measured thermal parameters for rock samples.

| ID | Sample thickness | Fourier Thermal diffusivity ($10^{-6}$ m$^2$/s) | Thermal diffusivity ($10^{-6}$ m$^2$/s) | Guyer-Krumhansl | Dissipation parameter ($10^{-6}$ m$^2$) | B |
|----|-----------------|---------------------------------|---------------------------------|-----------------|---------------------------------|---|
| 1/a | 2 mm            | 0.678                           | 0.581                           | 0.588           | 0.481                           | 1.407 |
| 1/b | 2.15 mm         | 1.259                           | 1.025                           | 0.547           | 0.726                           | 1.294 |
| 1/c | 2.85 mm         | 0.919                           | 0.766                           | 0.503           | 0.643                           | 1.68  |
| 1/d | 3.85 mm         | 1.074                           | 1.018                           | 0.612           | 0.735                           | 1.189 |
| 2/a | 3.05 mm         | 1.544                           | 1.434                           | 0.370           | 0.643                           | 1.210 |
| 2/b | 3.8 mm          | 0.978                           | 0.922                           | 0.648           | 0.715                           | 1.203 |
| 2/c | 3.9 mm          | 1.115                           | 1.057                           | 0.597           | 0.685                           | 1.099 |
| 3/a | 1.9 mm          | 0.956                           | —                               | —               | —                               | 1     |
| 3/b | 2.7 mm          | 1.441                           | 1.317                           | 0.351           | 0.551                           | 1.192 |
| 3/c | 3.7 mm          | 1.422                           | —                               | —               | —                               | 1     |
| 4/a | 1.9 mm          | 0.798                           | 0.762                           | 0.331           | 0.257                           | 1.02  |
| 4/b | 2.7 mm          | 1.023                           | —                               | —               | —                               | 1     |
| 4/c | 3.8 mm          | 0.558                           | —                               | —               | —                               | 1     |
| 5/a | 1.9 mm          | 0.708                           | 0.680                           | 0.400           | 0.301                           | 1.106 |
| 5/b | 2.3 mm          | 0.895                           | —                               | —               | —                               | 1     |
| 5/c | 3.7 mm          | 0.862                           | —                               | —               | —                               | 1     |
| 6/a | 1.86 mm         | 0.632                           | 0.598                           | 0.352           | 0.239                           | 1.135 |
| 6/b | 2.75 mm         | 0.687                           | —                               | —               | —                               | 1     |
| 6/c | 3.84 mm         | 0.778                           | —                               | —               | —                               | 1     |
| 7/a | 1.9 mm          | 0.504                           | —                               | —               | —                               | 1     |
| 7/b | 2.74 mm         | 0.553                           | —                               | —               | —                               | 1     |
| 7/c | 3.82 mm         | 0.570                           | —                               | —               | —                               | 1     |

Figure 6. The measured rear side temperature history for Szászvár formation I. with 3.05 mm thickness (ID-2/a).
4. Discussion

Although models describing non-Fourier heat conduction behaviour have enjoyed scientific interest since many decades, it is clear that the importance of comprehensive descriptions—possibly based on irreversible thermodynamics—will increase in the future. In addition to the need for a deeper understanding of the physical reality, there is a practical reason for this: these models might offer an affordable and suitable solution in areas where today’s and tomorrow’s engineers face at a series of challenges. Examples include but are not limited to scientific and technical tasks related to the thermal conductivity of heterogeneous materials (e.g., composites, metal foams, biological tissues, various kinds of rocks) present in the electronics industry, advanced material solutions, medical applications and transport. It should be noted that many of the possible applications of the non-Fourier effects are rapidly emerging technologies as well. One of which is additive layer manufacturing (also referred to as 3D printing), especially in Powder Bed Fusion (PBF) technologies (e.g., Selective Laser Melting). Notably, parts created by PBF generally have a certain level of porosity, thus they can be perceived as heterogeneous materials. From manufacturing point of view, this stands as an outstanding example: large local temperature gradients occur in the powder bed, significantly influencing the overall outcome, the mechanical and the thermal properties of the parts.

Gaining over the control of porosity has received on-growing attention among the researchers recently. Several of the relevant processes and material parameters have already identified, however, it is still considered to be a locus of interest for researchers of diverse fields. Furthermore, as [12] presents, designing a specific microstructure, it becomes possible to determine the effective properties of the material.

Based on the results presented in this paper, the research focuses on cases where heat conduction beyond the Fourier model is expected to be observed. Although ‘only’ half of the prepared samples behave accordingly (especially samples 1 and 2), it is a huge step forward in the understanding of modelling over-diffusive phenomenon, finding relations among the Fourier and GK parameters, and it motivates further research on size dependence behaviour of heterogeneous materials. On the one hand, the evaluation procedure is proved to be efficient to characterize the observed non-Fourier behaviour, and provides a reliable and consistent theoretical background for the GK equation. On the other hand, however, both heat conduction theories (Fourier and GK) cannot explain the observed size dependence of thermal parameters. It is surely challenging and requires further research to understand its origin, not purely from theoretical point of view but it might need a detailed investigation on the material structure for each sample in order to determine the constituents and obtain a more accurate image on the heterogeneities and pore size. Nevertheless, size effects can be apparent and significant and one should take that into account when measuring even the simplest thermal parameter, the thermal conductivity. Also, as mentioned, finding the proper extensions of Fourier’s law would influence the most fundamental perspectives of how the engineers think about the world.

![Figure 5. The measured rear-side temperature history for Szársomlyó limestone with 2.15 mm thickness (ID-1/b).](image)
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References

[1] L. Tisza. Transport phenomena in Helium II. Nature, 141:913, 1938.
[2] L. Landau. Two-fluid model of liquid Helium II. J. Phys. USSR, 5:71, 1941.
[3] T. F. McNelly. Second Sound and Anharmonic Processes in Isotopically Pure Alkali-Halides. 1974. Ph.D. Thesis, Cornell University.
[4] D. D. Joseph and L. Preziosi. Heat waves. Reviews of Modern Physics, 61(1):41, 1989.
[5] P Rogolino and VA Cimmelli. Differential consequences of balance laws in extended irreversible thermodynamics of rigid heat conductors. Proceedings of the Royal Society A, 475(2227):20180482, 2019.
[6] A. Sellitto and V. A. Cimmelli. Heat-pulse propagation in thermoelastic systems: application to graphene. Acta Mechanica, 230(1):121–136, 2019.
[7] F. X. Alvarez, V. A. Cimmelli, D. Jou, and A. Sellitto. Mesoscopic description of boundary effects in nanoscale heat transport. Nanoscale Systems: Mathematical Modeling, Theory and Applications, 1:112–142, 2012.
[8] S. Both, B. Czél, T. Fülöp, Gy. Gróf, Á. Gyenis, R. Kovács, P. Ván, and J. Verhás. Deviation from the Fourier law in room-temperature heat pulse experiments. Journal of Non-Equilibrium Thermodynamics, 41(1):41–48, 2016.
[9] P. Ván, A. Berezovskii, T. Fülöp, Gy. Gróf, R. Kovács, Á. Lovas, and J. Verhás. Guyer-Krumhansl-type heat conduction at room temperature. EPL, 118(5):50005, 2017. arXiv:1704.00341v1.
[10] T. Fülöp, R. Kovács, Á. Lovas, A. Rieti, T. Fodor, M. Szűcs, P. Ván, and Gy. Gróf. Emergence of non-Fourier hierarchies. Entropy, 20(11):832, 2018. ArXiv: 1808.06858.
[11] T. Fülöp, R. Kovács, and P. Ván. Thermodynamic hierarchies of evolution equations. Proceedings of the Estonian Academy of Sciences, 64(3):389–395, 2015.
[12] A. Seppälä. Efficient method for predicting the effective thermal conductivity of various types of two-component heterogeneous materials. International Journal of Thermal Sciences, 134:282–297, 2018.
[13] H. Haddad, W. Leclerc, G. Alhajj Hassan, A. Ammar, C. Pélegris, M. Guessasma, and E. Bellenger. Numerical investigation of heat conduction in heterogeneous media with a discrete element method approach. International Journal of Thermal Sciences, 164:106799, 2021.
[14] G. Chen. Ballistic-diffusive heat-conduction equations. Physical Review Letters, 86(11):2297–2300, 2001.
[15] F. X. Alvarez and D. Jou. Memory and nonlocal effects in heat transport: from diffusive to ballistic regimes. Applied Physics Letters, 90(8):083109, 2007.
[16] V. A. Cimmelli, A. Sellitto, and D. Jou. Nonlocal effects and second sound in a non-equilibrium steady state. Physical Review B, 79(1):014303, 2009.
[17] M. Wang and Z.-Y. Guo. Understanding of temperature and size dependences of effective thermal conductivity of nanotubes. Physics Letters A, 374(42):4312–4315, 2010.
[18] A. Fehérs and R. Kovács. Novel evaluation method for non-Fourier effects in heat pulse experiments. 2021. arXiv:2101.01123.
[19] W. J. Parker, R. J. Jenkins, C. P. Butler, and G. L. Abbott. Flash method of determining thermal diffusivity, heat capacity, and thermal conductivity. Journal of Applied Physics, 32(9):1679–1684, 1961.
[20] H. M. James. Some extensions of the flash method of measuring thermal diffusivity. Journal of Applied Physics, 51(9):4666–4672, 1980.
[21] Gy. I. Gróf. Homogén és kétrétegű minták hőmérsékletvezetési mértékeinek meghatározása flash módszerrel (In Hungarian). 2002. Ph.D. Thesis, BME, Hungary.
[22] V. Jóssa and R. Kovács. Solving Problems in Thermal Engineering: A Toolbox for Engineers. Springer, 2020.

1Department of Energy Engineering, Faculty of Mechanical Engineering, BME, Budapest, Hungary 2Department of Theoretical Physics, Wigner Research Centre for Physics, Institute for Particle and Nuclear Physics, Budapest, Hungary 3Montavid Thermodynamic Research Group