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# THERMAL CONDUCTIVITY AND DIFFUSIVITY OF NATURAL GRAPHITE SHEET

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## ABSTRACT

The in–plane and through–plane thermal conductivity and diffusivity of natural graphite sheet (NGS) at densities ranging from $0.5\,\mathrm{g\,cm^{-1}}$ to $1.7\,\mathrm{g\,cm^{-1}}$ are measured using the transient plane source method. In the in–plane direction, the thermal conductivity and diffusivity are high in the range from $100\,\mathrm{Wm^{-1}K^{-1}}$ to $350\,\mathrm{Wm^{-1}K^{-1}}$ and from $230\,\mathrm{mm^2s^{-1}}$ to $270\,\mathrm{mm^2s^{-1}}$, respectively, and they increase with density. In the through–plane direction, the thermal conductivity and diffusivity are low in the range from $5\,\mathrm{Wm^{-1}K^{-1}}$ to $2\,\mathrm{Wm^{-1}K^{-1}}$ and from $12\,\mathrm{mm^2s^{-1}}$ to $1.5\,\mathrm{mm^2s^{-1}}$, respectively, and they decrease with density. The in–plane properties are independent of the compression pressure but the through–plane properties increase with pressure especially at higher densities. The sheet-to-sheet thermal contact resistance of stacked NGS is found to be negligible.

**KEYWORDS:** Natural graphite sheet, flexible graphite, compressed exfoliated natural graphite, thermal conductivity, thermal diffusivity, transient plane source method

## 1. INTRODUCTION

Natural graphite sheet (NGS), which is also known as flexible graphite, graphite foil, or compressed exfoliated (expanded) natural graphite, is a highly anisotropic material made by compressing exfoliated natural graphite flakes into thin sheets whose thickness ranges from fractions of a millimeter to several millimeters and the density reaches up to $1.9\,\mathrm{g\,cm^{-1}}$. Its high in–plane thermal conductivity of up to $500\,\mathrm{Wm^{-1}K^{-1}}$ [1, 2], makes it an attractive material for heat transfer devices; however, the low through–plane thermal conductivity, which is typically lower than $10\,\mathrm{Wm^{-1}K^{-1}}$, can become a challenge for practical applications such as bipolar plates for fuel cells [3], heat exchangers [4], or heat sinks [5]. In our previous research [6], we performed a feasibility study of NGS heat sinks for power electronics and discovered gaps in the available material properties data.

The available literature sources report the thermal conductivity and diffusivity to increase with density in the in–plane direction and decrease in the through–plane direction. The explanation of the change of the thermal properties with density is consistent across the sources and states that both the orientation of the highly anisotropic graphite crystallites and the contact resistance at their boundaries change with density and alter the overall bulk properties of NGS. Most of the studies [1, 2, 7, 8] were performed using the laser flash method, which is inherently not capable of measuring the dependence of the thermal properties on the uniaxial compression pressure. Two relevant studies used the guarded hot plate method [3, 9], which requires the samples to be compressed to allow the heat to flow from the fluxmeters into the samples, but the pressure was

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not varied and in one case its magnitude was not reported. Only one published article [10] focused on the thermal contact resistance (TCR) between the sheets, and the authors concluded that it is negligible at 100 kPa

The goals of this study are to: i) measure the in–plane and through–plane thermal conductivity and diffusivity of NGS, ii) investigate the dependence of the measured properties on the compression pressure in the range 100 kPa to 1080 kPa, and iii) determine whether the sheet-to-sheet TCR significantly affects the overall effective thermal properties of stacked NGS.

## 2. EXPERIMENTAL METHOD

The measurements in this study were performed using the Hot Disk TPS 2500S machine, which is a commercial implementation of the transient plane source method [11, 12] whose principle is based on a sensor in the form of a nickel spiral through which an electric current is passed. During the measurement, the sensor is sandwiched between the samples and the generated heat penetrates the samples. The temperature increase of the sensor is recorded by measuring the current and voltage and correlating it to the known temperature dependence of the resistivity of nickel. Based on the shape of the temperature-versus-time curve, the thermal properties are subsequently determined.

The samples were prepared in two steps. First, the low-density semi-finished sheets (0.2 $\mathrm{g\,cm^{-3}}$, fixed carbon content 99.27 %, exfoliation volume 250 $\mathrm{ml\,g^{-1}}$, flake size composition of 81 % larger than 300 µm, supplier Nano Carbon Technology CO., LTD, Qingdao, China) at three surface densities of 70 $\mathrm{mg\,cm^{-2}}$, 140 $\mathrm{mg\,cm^{-2}}$, and 210 $\mathrm{mg\,cm^{-2}}$ were calendered between two 93.5 mm-diameter cylinders at 5 $\mathrm{cm\,s^{-1}}$ speed into sets of sheets 100 mm wide, 300 mm long, with the densities and thicknesses in the range 0.5 $\mathrm{g\,cm^{-3}}$ to 1.7 $\mathrm{g\,cm^{-3}}$ and 0.4 mm to 4.2 mm, respectively. The second step varied for the in–plane and through–plane measurements.

For the in–plane direction, the slab mode was utilized (Fig. 1a), for which 7 cm by 7 cm square samples were cut from the calendered sheet. To measure the through–plane thermal properties, the one-dimensional mode was used (Fig. 1b), which requires the samples to match the sensor size and therefore circular samples 32 mm in diameter were cut from the calendered sheet using a hole punch. Stacks of sheets with the total height ranging from 6.7 mm to 27 mm were measured at pressures of 100 kPa, 200 kPa, 400 kPa, 800 kPa, and 1080 kPa. Measurements of the through–plane properties of a single sheet were not possible as the required measurement times are much lower than the low limit of the machine.

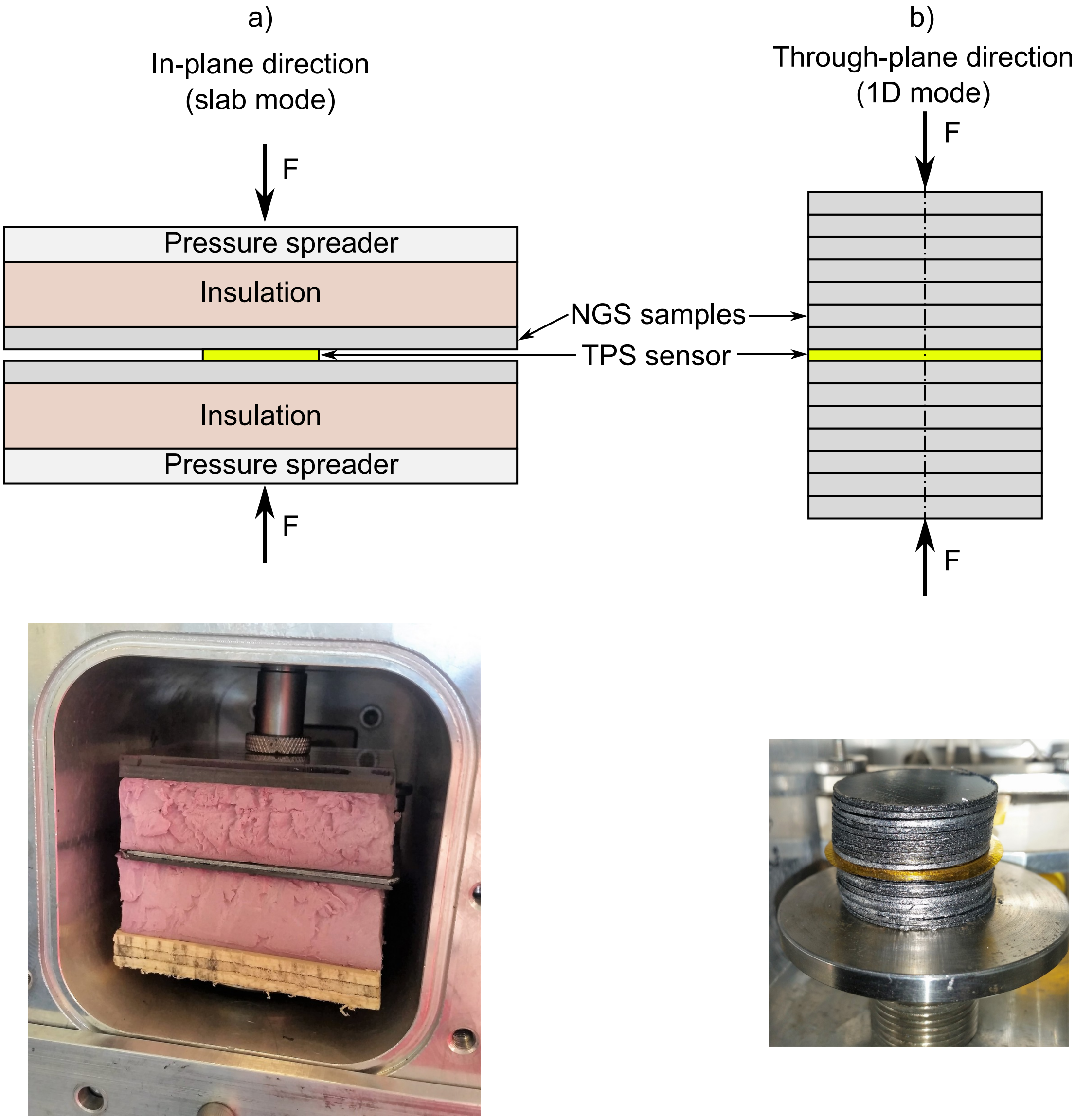

**Fig. 1** A schematic (top) and a photography of the measurement of a) the in–plane and b) through–plane thermal properties using the TPS method.

## 3. RESULTS

The results are summarized in Fig. 2. While the in–plane thermal conductivity and diffusivity increase with the density, the through–plane properties decrease with density, which is in agreement with the previously reported data. The in–plane thermal conductivity increases linearly with density and can be approximated by the relationship $k_{in} = 212d - 22$. The surface density of the sheet, which is directly proportional to its thickness, does not affect the in–plane thermal properties. The data points for the in–plane thermal diffusivity in Fig. 2b are scattered and do not follow the expected trend $\alpha_{in} = 0.29 - 0.0301d^{-1}$ outlined by the dashed line that was calculated from the thermal conductivity fit using the relationship between the thermal diffusivity and conductivity $\alpha = \frac{k}{dc_p}$. Since the values of the thermal conductivity and diffusivity were calculated using the proprietary software designed by the manufacturer of the measurement machine, further investigation of the scatter of the diffusivity data was not possible. Over the measured range of density, the in–plane thermal conductivity increases by approximately 250 % while the diffusivity increases only marginally by approximately 20 %. No dependence of the in–plane thermal properties on pressure was seen.

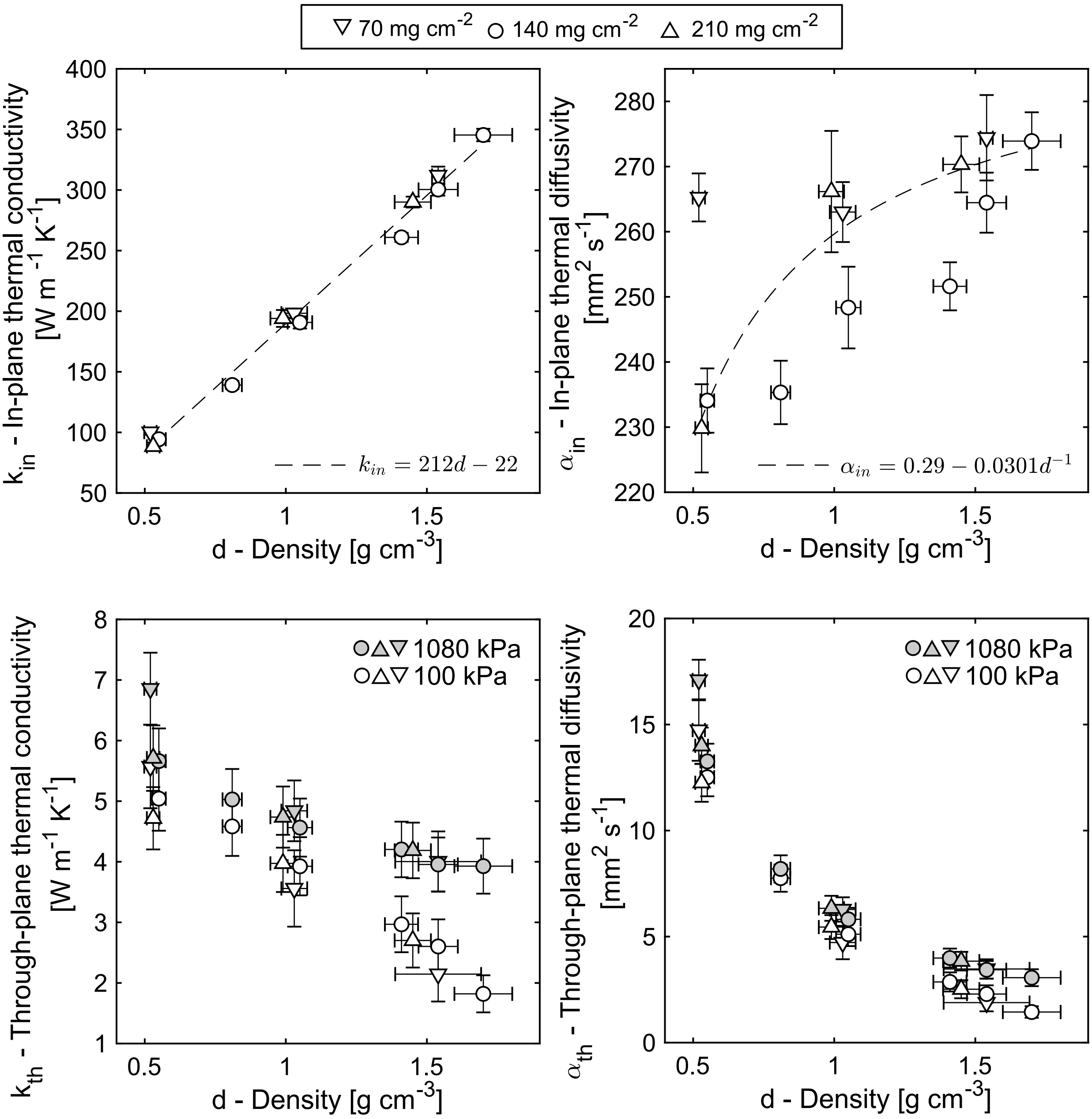

**Fig. 2** The results of the in–plane thermal conductivity (top left), in–plane thermal diffusivity (top right), through–plane thermal conductivity (bottom left), and through–plane thermal diffusivity (bottom right)

The through–plane properties are a function of not only the density but also of the pressure. With increasing density, the through–plane properties decrease non-linearly with the exception of the thermal conductivity at 100 kPa, which shows a linear decrease. Increasing the compression pressure results in increased thermal conductivity and diffusivity for all the measured densities, but the increase is more pronounced at higher densities as can be seen in Fig. 3. After increasing the compression pressure from 100 kPa to 1080 kPa, the thermal conductivity and diffusivity of the 1.7 $\mathrm{gcm^{-3}}$ sheet increased by 116 % and 112 %, respectively, while that of the 0.55 $\mathrm{gcm^{-3}}$ sheet increased only by 12 % and 6 %, respectively. The repeatability of the measurements was better at higher pressures but this trend cannot be seen in Fig.s 2 and 3 as the overall uncertainty of the measurement is dominated by the 5 % accuracy of the machine given by the manufacturer [13].

The values of the through–plane conductivity and diffusivity and the in–plane diffusivity of the 0.5 $\mathrm{gcm^{-3}}$ 70 $\mathrm{mgcm^{-2}}$ sample deviate from the rest of the measurements. For the through–plane direction the deviation is

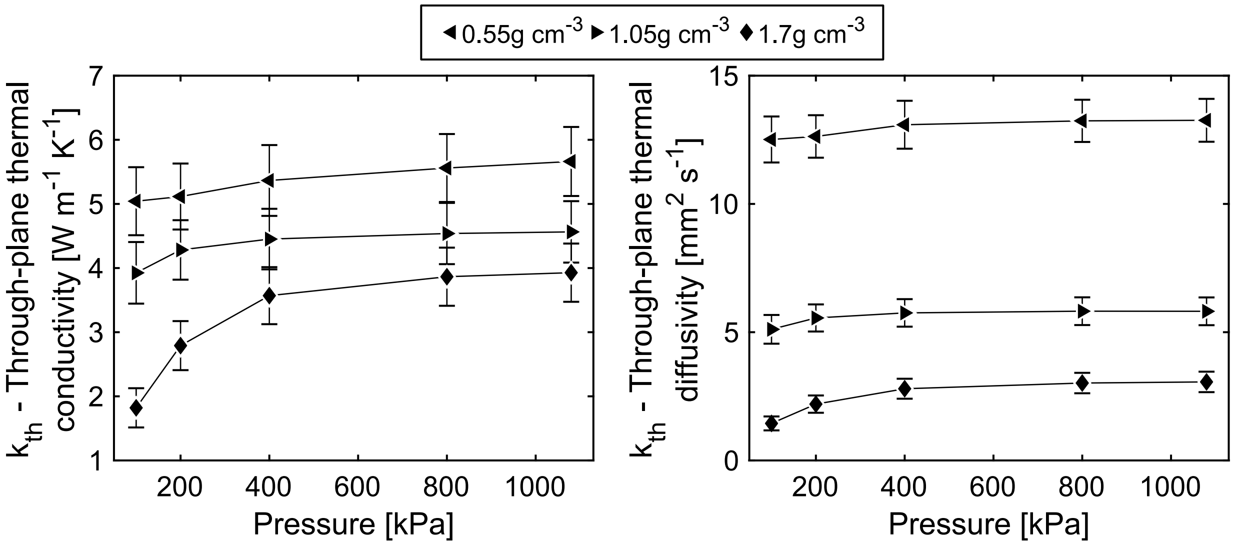

**Fig. 3** The pressure dependence of the thermal conductivity (left) and diffusivity (right) for the 140 $\mathrm{mgcm^{-2}}$ sheet

within the uncertainty and for the in–plane diffusivity the deviation might be caused by the above mentioned scatter in the data; however, a similar trend was seen in the data by [1] and therefore the possibility of the low-density and low-thickness samples showing increased thermal properties cannot be rejected.

Since stacks of sheets were measured, the sheet-to-sheet TCR is inherently included in the value of the through–plane thermal conductivity and diffusivity. Deconvolution of the sheet-to-sheet TCR is not possible neither using the TPS nor the guarded heat flux meter method. The importance of the sheet-to-sheet TCR can be estimated by comparing the results for stacks of sheets at different thicknesses. Fig. 4 illustrates the theory employed to judge the importance of the sheet-to-sheet TCR. The surface density of the sheet dictates its thickness at the given density. By stacking the low and high surface density sheets with the same volumetric density, varying number of interfaces per stack height can be achieved. If the sheet-to-sheet TCR is significant, the stack with more interfaces will show lower thermal conductivity. In practice, it was not possible to manufacture sheets at the same density as the calendering process did not allow a fine control of the distance between the calendering cylinders and therefore the sheets prepared for this measurement had comparable but not identical density as can be seen by the horizontal distance between the circular and triangular symbols in Fig. 2. The results for the through–plane thermal conductivity and diffusivity in Fig. 2 do not show the vertical stacking order outlined on the left side of Fig. 4 and therefore the contribution of the sheet-to-sheet TCR was deemed to be negligible.

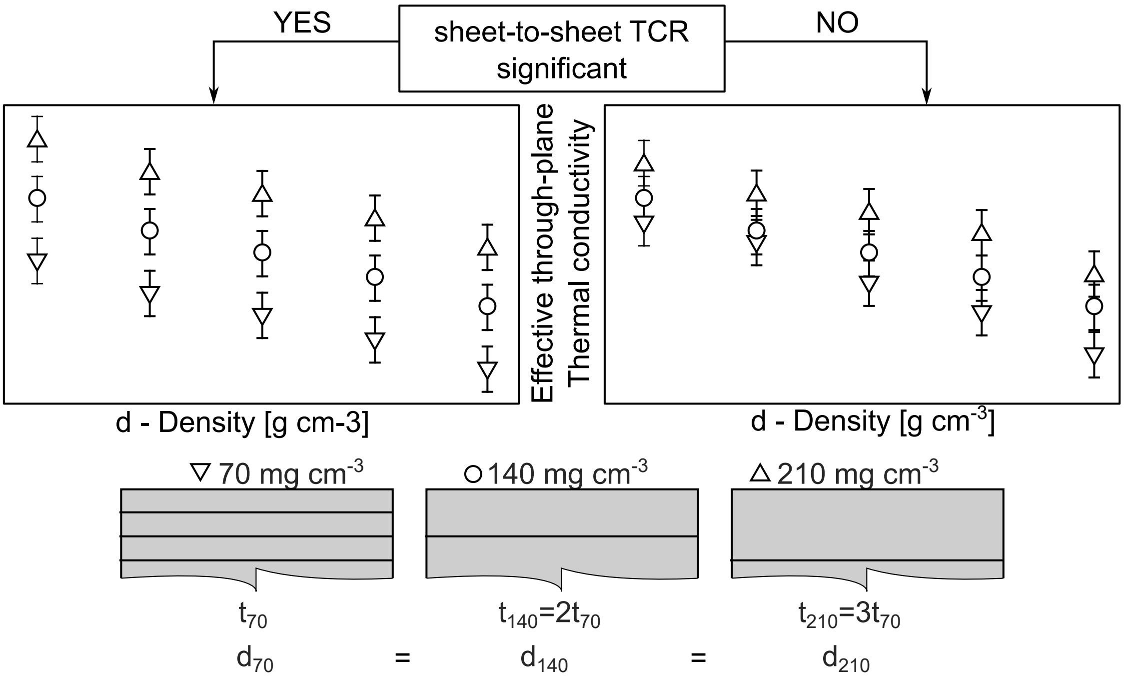

**Fig. 4** Illustration of the theory used for evaluating the importance of the sheet-to-sheet TCR

## 4. CONCLUSIONS

The in–plane and through–plane thermal conductivity and diffusivity of NGS were measured at densities ranging $0.5\,\mathrm{g\,cm^{-1}}$ to $1.7\,\mathrm{g\,cm^{-1}}$ and pressures 100 kPa to 1080 kPa. The in–plane thermal conductivity increases linearly with density from $100\,\mathrm{Wm^{-1}K^{-1}}$ at $0.5\,\mathrm{g\,cm^{-1}}$ to $350\,\mathrm{Wm^{-1}K^{-1}}$ at $1.7\,\mathrm{g\,cm^{-1}}$ and the in–plane thermal diffusivity increases non-linearly from $230\,\mathrm{mm^2s^{-1}}$ at $0.5\,\mathrm{g\,cm^{-1}}$ to $270\,\mathrm{mm^2s^{-1}}$ at $1.7\,\mathrm{g\,cm^{-1}}$. The in–plane thermal properties are not a function of the compression pressure.

The through–plane thermal conductivity at 100 kPa decreases linearly from $5\,\mathrm{Wm^{-1}K^{-1}}$ at $0.5\,\mathrm{g\,cm^{-1}}$ to $2\,\mathrm{Wm^{-1}K^{-1}}$ at $1.7\,\mathrm{g\,cm^{-1}}$ and the through–plane thermal diffusivity at 100 kPa decreased non-linearly from $12\,\mathrm{mm^2s^{-1}}$ at $0.5\,\mathrm{g\,cm^{-1}}$ to $1.5\,\mathrm{mm^2s^{-1}}$ at $1.7\,\mathrm{g\,cm^{-1}}$. The through–plane properties increase with compression pressure; the increase is within the uncertainty limit for low densities and becomes significant at higher densities. The pressure dependence shows an exponential profile with a steeper increase at lower pressures. At the highest measured pressure of 1080 kPa the through–plane thermal conductivity and diffusivity increased by 116 % and 112 % with respect to the 100 kPa values.

The effect of the sheet-to-sheet TCR was considered negligible based on the measurements of sheets at varying surface densities.

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# NOMENCLATURE

*Roman letters*

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|---|---|
| $k_{in}$ | In-plane thermal conductivity [$\mathrm{W m^{-1} K^{-1}}$] |
| $k_{th}$ | Through-plane thermal conductivity [$\mathrm{W m^{-1} K^{-1}}$] |
| $d$ | Density [$\mathrm{g\,cm^{-1}}$] |
| $t$ | Sheet thickness [mm] |

*Greek letters*

| | |
|---|---|
| $\alpha_{in}$ | In-plane thermal diffusivity [$\mathrm{mm^2 s^{-1}}$] |
| $\alpha_{th}$ | Through-plane thermal diffusivity [$\mathrm{mm^2 s^{-1}}$] |

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