Supplementary materials: Isotropic thermal expansion in anisotropic thermal management composites filled with carbon fibres and graphite.

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S1. Spark plasma sintering

Samples were produced by Spark Plasma Sintering in a *Dr. Sinter Lab 211-Lx* from *Fuji Electronic Industrial Co. Ltd.* The experimental design is shown in Fig. S1

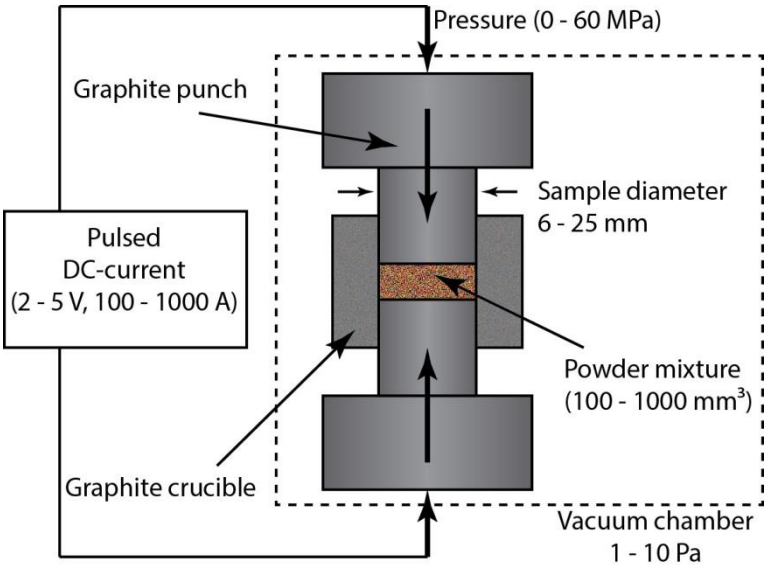

Fig. S1 Scheme of the used SPS system and typical sintering parameters.

The detailed, material dependent sinter parameters are found in Section 2.2 of the publication.

S2 Thermal diffusivity measurement

Thermal diffusivity was measured on disc-shaped, 25 mm diameter samples by the flash method with a *Netzsch LFA447 NanoFlash*. A xenon discharge lamp heats the lower side of the sample with a flash, while the temperature profile of the upper side of the sample is measured by an infrared camera. To enhance the absorption and emission of radiation, the surface of the sample was coated with a thin layer of black graphite spray. Since the filler particles are significantly smaller than the dimensions of the sample, the material is assumed to behave as a homogeneous medium [1].

A typical temperature profile is shown in figure S2. Thermal diffusivity measures the propagation speed of a heat wave. For the evaluation, the time from the flash to the peak temperature is evaluated. The absolute magnitude of the signal does not directly influence the measurement, a strong signal provides a better signal-to-noise ratio).

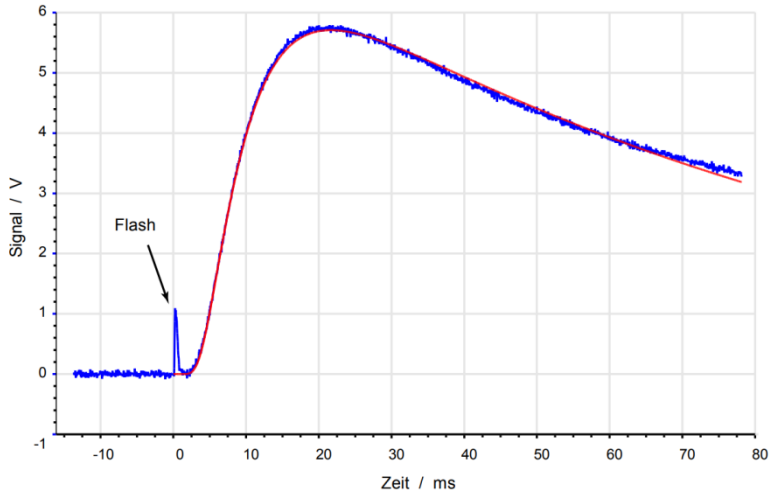

Fig. S2 Typical temperature profile for a through-plane thermal diffusivity measurement (z-direction) of a 1 mm thick sample (Al2024 matrix and 50vol% filler with 1:3 CF:Gr-ratio).

For every sample, at least five measurements were performed for the z-direction, and at least sixteen (eight per side) for the x,y-direction by the radial heat flow method (Fig. S3 left). The validity of the radial heat flow method was verified for few materials by cutting slices of the sample. These were rotated by 90° and glued to a laminate, so the x,y-direction of the material could be measured by a normal through plane measurement (Fig. S3 right).

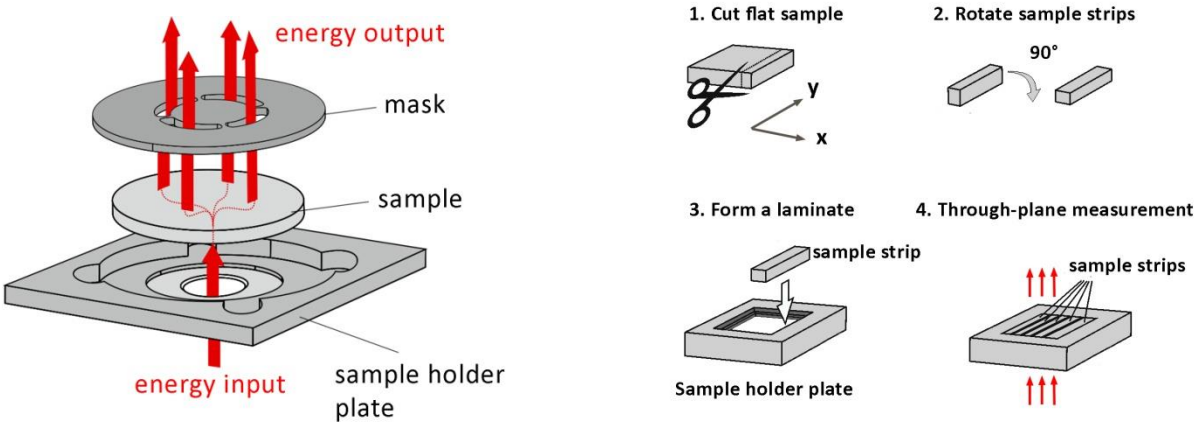

Fig. S3 In-plane (x,y) thermal diffusivity measurement by radial flow method (left) and by cutting the sample into slices for a through-plane measurement (right). LFA 447 manual; Image courtesy of *NETZSCH-Gerätebau GmbH*.

S3 Determination of the thermal conductivity

The thermal conductivity λ of the samples is obtained by multiplication of the measured thermal diffusivity *TD* by the density ρ and specific heat capacity *c* of the samples.

$$\lambda = TD \cdot \rho \cdot c$$

Given the regular cylindrical geometry of the samples the density was measured by geometrical method (mass divided by volume). In previous studies Archimedes' principle was used. We discarded it later because porous samples were found to absorb small quantities of liquid.

Starting from literature values for the specific heat capacities of matrix c_m and filler c_f (Table S1), we determined the specific heat capacity of the composite c_c by rule of mixture by mass according to

$$c_c = c_m \cdot w_m + c_f \cdot w_f$$

with the mass concentrations w_m and w_f for matrix and filler. Due to the macroscopic size of the filler and the negligible chemical reaction with the matrix, we consider the calculated specific heat capacity to be sufficiently accurate for the purpose of this publication.

Table S1. Density ρ and specific heat capacity *c* of the used metal powders and fillers for the determination of the filler mass concentration w_f and specific heat capacity of the composites c_c. The ρ and *c* for metal mixtures were calculated starting from their composition. The method correctly predicted the *c* of alloys with similar composition, i.e. Al7075 for Alumix 431 [2], Al-14Si for Alumix 231 [3], Mg for Mg-0.9Ca.

	Graphite/CF	**Al2024**	**Alumix 231**	**Alumix 431**	**Aluminium**	**Mg-0.9Ca**	**Copper**
ρ / **kg m^3**	2200-2220	2780	2790	2790	2700	1740	8930
c **/ J g^{-1} K^{-1}**	0.700-0.720	0.875 [4]	0.858	0.864	0.897	1.019	0.386

The specific heat capacities of matrix and filler are very similar for composites with aluminium alloys. The thermal diffusivity in the metal matrices is 50-100 mm² s^{-1} (isotropic), while it assumes values between 5 mm² s^{-1} and 1000 mm² s^{-1} in the fillers, depending on the direction.

S4 CTE Measurements

CTE measurements were performed by dilatometry. We measured for all composites with at least 25% CF concentration in the filler linear dilatometry curves (Fig. S4). In composites with pure graphite flake filler, the z-CTE increased at higher temperatures and the curves showed a hysteresis.

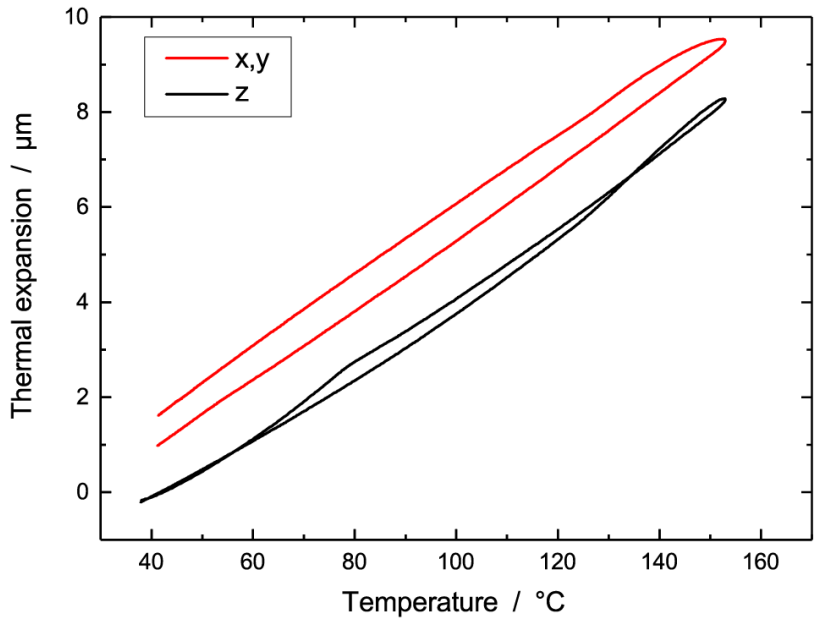

Fig. S4 Typical dilatometry curves (Al2024 matrix and 50vol% filler with 1:3 CF:Gr-ratio after T6 thermal treatment). The sample has 6.13 mm diameter (x,y) and 5.46 mm height (z).

References

1. Vozár L, Hohenauer W (2004) Flash method of measuring the thermal diffusivity. A review. High Temp Press 36:253–264.
2. Zahra a. M, Zahra CY, Jaroma-Weiland G, et al (1995) Heat capacities of aluminium alloys. J Mater Sci 30:426–436. doi: 10.1007/BF00354407
3. Angadi BM, Hiremath CR, Reddy AC, et al (2014) Studies on the Thermal Properties of Hypereutectic Al--Si Alloys by Using Transient Method. J Mech Eng 2:536–544.
4. Kammer C (1995) Aluminium-Taschenbuch: Grundlagen und Werkstoffe. Aluminium-Verlag Düsseldorf