

## Thermal Conductivity of Filled Sol-Gel-Derived Hybrid Materials

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**ABSTRACT:** A novel sol-gel-based hybrid material has been synthesized from organofunctional silanes and aluminates via hydrolysis and condensation reactions. The hybrid material was—in comparison to a silicone rubber—filled with equal amounts of metal filler particles in order to investigate the effective thermal conductivity. The data obtained were compared with theoretical models available in literature to gain understanding of mechanisms responsible for the measured conductivities. Samples were prepared via casting and spin-coating techniques on Teflon® and silicon substrates and characterized using laser flash analysis (LFA) and scanning electron microscopy (SEM). It was demonstrated, that the hybrid material with an aluminum content of 40 vol % offers the possibility to achieve an up to five times higher thermal conductivity than filled silicone references. The influence of thermal transition between matrix material and filler was evaluated by a sandwich-layer setup. The results suggest that increased thermal conductivity is also caused by the lower thermal resistance and improved interphase connections within the hybrid material. © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 41037.

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### INTRODUCTION

Silicone materials are used in a broad range of technical products, e.g. for corrosion protection of electronic devices, as glues and sealants in optical products or as potting materials.<sup>1–7</sup> In order to improve thermal stability of these materials, various amounts and types of inorganic fillers are added, sometimes also to tune optical properties such as refractive index.<sup>8–13</sup> The amount of filler used is often limited by changes of properties relevant for the corresponding application such as increased viscosity or mechanical properties (brittleness) when operating at high particle concentrations.<sup>10–14</sup> At filler grades below the percolation threshold the thermal conductivity of filled silicones are mostly unchanged in accordance to their unfilled values ( $0.1\text{--}0.3\text{ W m}^{-1}\text{ K}^{-1}$ ).<sup>8,12,13</sup> This can be a problem, when silicone coated or potted devices generate excess heat thus leading to a bottleneck in heat transfer and finally to a reduced lifetime of the device.

Although it has been known for many years that silicones lack in thermal conductivity they are still being used in thermally critical applications due to their outstanding optical (transparency, UV-stability) and ageing properties (thermal stability, elastic properties). For gluing and sealing applications where the silicone-typical elasticity is not required, a material alternative is commercially available based on silica water glass.<sup>15,16</sup> After

application of this liquid material and a subsequent thermal treatment, a glass like compound is generated which can be used as a high temperature stable (up to 500°C) glue with high thermal conductivity.<sup>17,18</sup> Due to their glass like nature, these materials also show some disadvantages regarding elastic properties, hydrolytic stability or applicability for layers and components with higher thickness.<sup>18</sup>

The aim of this work was to synthesize a new material class for gluing and potting applications, which can provide the main advantages of the silicone-material class while overcoming the problems of thermal barrier properties. Furthermore the investigation for the improved thermal conductivity of hybrid material related to different filler grades of conductive particles were compared with diverse theoretical equations in order to acquire an interpretation about thermal conduction mechanisms and thermal resistance performance.

In previous studies it has been shown, that the thermal conductivity of filled silicone rubber is limited by the silicon material properties and can only moderately be increased by high filling amounts of about 60 vol %.<sup>8,10,12,13</sup> As thermal conductivity does not correlate with the filler grade in a linear way and strongly depends on the type, size, and shape of the filler used, different models have been developed in order to predict and optimize this parameter. Agari et al. postulate two different

systems with highly conductive filler:<sup>19</sup> a system with low particle filler content, where no thermal percolation is possible is called as a dispersed system. This system can be described by the Maxwell model.<sup>20</sup> The basic thinking behind this model is a two phase mixture of binder (silicone) and filler. The filler is homogeneously distributed in the binder but does not form an interpenetrating network—i.e. filler particles are isolated from each other:

$$\lambda_c = \lambda_m \frac{\lambda_p + 2 \lambda_m + 2 \phi_p (\lambda_m - \lambda_p)}{\lambda_p + 2 \lambda_m - \phi_p (\lambda_m - \lambda_p)} \quad (1)$$

where in this equation and in the following eqs. (2–6)  $\lambda_c$ ,  $\lambda_m$ , and  $\lambda_p$  are the thermal conductivities of composite, matrix and the particles, respectively, and  $\phi_p$  is the volume fraction of the filler. For low filler contents this model predicts the thermal conductivity of composites accurately. At higher concentrations, when filler percolation occurs, this model becomes inaccurate.

An implicit relationship between the thermal conductivities of the composite, the filler and the matrix for dilute suspension of spheres in a dispersed system was developed by Bruggeman:<sup>21</sup>

$$1 - \phi_p = \frac{\lambda_p - \lambda_c}{\lambda_p - \lambda_m} \sqrt[3]{\frac{\lambda_m}{\lambda_c}} \quad (2)$$

The second system with a higher content of particles is referred as “attached system”, in which conductive chains are formed by the filler and contribute to a large increase in thermal conductivity of the composite. The typical theoretical models were proposed by Agari and Uno:<sup>19,22,23</sup>

$$\log \lambda_c = \phi_p C_2 \log \lambda_p + \phi_m \log (C_1 \lambda_m) \quad (3)$$

According to this equation, the overall thermal conductivity depends on the thermal percolation through the conductive filler network ( $C_2$ ) and the crystallinity/secondary structure of the matrix material ( $C_1$ ). Both constants are determined experimentally. According to Agari, values for  $C_1$  and  $C_2$  are in the range of 0–1. The closer  $C_2$  is to 1, the more thermally conductive particle chains are formed in the composite and therefore the constant  $C_2$  gives information about particle interconnection.

As it is a major issue of this work to develop and understand highly conductive material systems below the thermal percolation threshold, the Agari equation is used for low filler grades as it is the only way to calculate the density of heat transfer paths.

Taking geometric aspects of particles and the maximum packing fraction into account, Lewis–Nielsen developed another model for highly filled composites:<sup>24–26</sup>

$$\lambda_c = \lambda_m \left( \frac{1 + A B \phi_p}{1 - B \phi_p \psi} \right) \quad (4)$$

$$\text{with } B = \frac{(\lambda_p / \lambda_m) - 1}{(\lambda_p / \lambda_m) + A}; A = 2 \left( \frac{L}{D} \right) \quad (5)$$

$$\text{and } \psi = 1 + \left( \frac{1 - \phi_m}{\phi_m^2} \right) \phi_p \quad (6)$$

where  $\phi_m$  is the maximum packing fraction (in volume) of dispersed fillers in the system. For high aspect ratio fillers,  $A$  is related to the aspect ratio  $2 (L/D)$  of the filler, but is also

dependent on the extent of orientation of the fillers. It is important to note that this model assumes perfect interfaces between the filler and matrix and does not explicitly account for contact resistances between the matrix and filler particles. The Lewis–Nielsen model predicts thermal conductivity over a wide range from  $\Phi_p = 0$  to  $\Phi_p = \Phi_m$ .<sup>27</sup>

All models described above can be used for a calculative prediction of thermal conductivity but cannot be a replacement for a real measurement. In fact, these calculations are used in this work to identify the special mechanism of conductivity as each model takes different factors influencing conductivity into account. That means e.g., if a measurement series for one sample fits the Agari calculations better than the Lewis–Nielsen values, that the filler amount is in a sub-percolative concentration, where the particles are not connected to each other.

## EXPERIMENTAL

### Materials

Precursor materials used to prepare the gels were 3-glycidioxypropyltrimethoxysilane (GPTMS, 98% ABCR), 3-aminopropyltrimethoxysilane (APTMS, 98% ABCR), and aluminum-tri-*sec*-butoxide (ASB, Al-(O<sup>*sec*</sup>Bu)<sub>3</sub>, 98% ABCR). Ethylacetate (EAA, 99% Sigma Aldrich) was used as a chelating agent in order to control the rate of hydrolysis of Al-(O<sup>*sec*</sup>Bu)<sub>3</sub>. The chemicals were used as received. Diluted hydrochloric acid (0.1M HCl) was used as catalyst and solvent.

Aluminum powder (Ecka Granules, Germany) was selected as thermally conductive filler with an average particle ( $d_{50}$ ) size of 25  $\mu\text{m}$  ( $d_{10} = 14 \mu\text{m}$ ;  $d_{90} = 55 \mu\text{m}$ ). The thermal conductivity of aluminum is 220  $\text{W m}^{-1} \text{K}^{-1}$ .

The silicone rubber used as a reference material was an organopolysiloxane mixture (type C-5547S-1) manufactured by Shin-Etsu Chemical Co Ltd.

### Sol Synthesis

The sol was prepared by sol gel processing with a starting mixture of GPTMS and APTMS. This mixture was kept under continuous stirring and while cooling the solution. ASB and EAA were mixed as a separate solution and added drop wise to the mixture of GPTMS/APTMS. After a few minutes of reaction time hydrochloric acid (0.1M) was slowly added and the mixture was kept under continuous stirring for 2 h under ambient condition. The molar ratio of GPTMS, APTMS, EAA, and HCl to Al-(O<sup>*sec*</sup>Bu)<sub>3</sub> was 12 : 1 : 1.6 : 22, respectively.

### Sample Preparation

Thermal conductivity measurements of the aluminum filled composite materials were performed on casted sample plates. These were prepared by mixing the filler with the base material (silicone or sol-gel-material) and pouring an adequate amount into a PTFE mould. The molds were kept for one day under ambient conditions in order to allow the solvent to evaporate. Sedimentation of the particles was prevented by storing the samples on a vibrating plate. After reaching its gel-point the sample was cured for 1 h at 140°C (silicone-based samples were cured according to the specifications in the data sheet for 4 h at 150°C). Afterwards the samples were removed from the mold

and cut into 10 X 10 mm plates to meet the requirements for thermal conductivity analysis.

In order to better understand the differences in thermal conductivity when using new sol-gel-based binder materials, an additional sample setup was chosen. The idea of this sandwich setup was to compare the thermal transition rates between high conductive materials (silicon or aluminum filler) and the less conductive binder material. As samples previously prepared via casting only allow the measurement of an overall thermal conductivity and do not specifically display information about the thermal transition from matrix material to filler particles. It was necessary to prepare a sample set-up made of the characterized materials with two defined material transitions (silicon-binder and binder-silicon). A silicon wafer was coated via spin coating process with the particle-free matrix material resulting in a thickness of 20–30  $\mu\text{m}$ . Another silicon wafer was attached on top using a pressure of 5 kPa. The sandwich-sample was also characterized by LFA.

### Characterization

The thermal conductivity was calculated from the thermal diffusivity with the following equation:

$$\lambda = \alpha \times \rho \times c_p$$

Where  $\lambda$ ,  $\alpha$ ,  $\rho$ , and  $c_p$  are the thermal conductivity, thermal diffusivity, specific density, and specific heat capacity of the compound under constant pressure and at corresponding temperature. The measurement of thermal diffusivity was conducted on a Netzsch LFA 447 system (Selb, Germany) in the temperature range 25–150°C.

Specific heat capacity measurements were performed with a differential scanning calorimeter (DSC Q1000, TA Instruments; New Castle, DE). The temperature was increased from 5 to 170°C at a heating rate of 10 K  $\text{min}^{-1}$ . Each sample was preheated for 3 min at 150°C to dehydrate the sample. The differential scanning calorimeter had been calibrated in the same temperature region before each experiment by a sapphire sample as an internal standard with a well known specific heat capacity.<sup>28</sup> Data for the heat capacity was collected in the temperature range of 25 to 150°C.

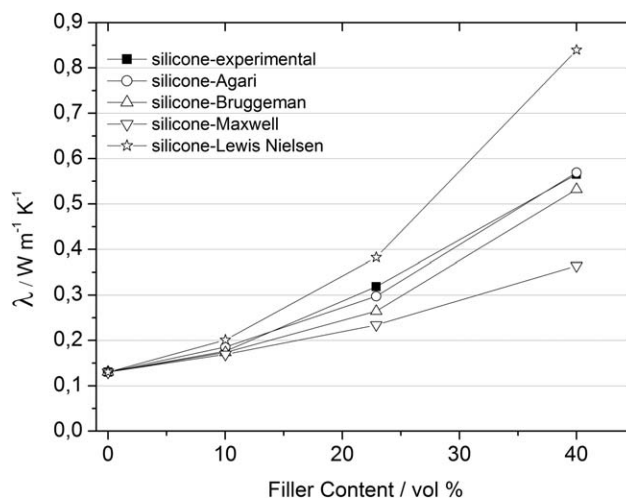
Specific density values of the sol-gel composite material were determined with the Archimedeian method. The density was measured by the displacement of water. The weight measurement was performed with an electronic balance (Mettler Toledo) at ambient temperature.

### Morphology Observations

Morphological investigations of the different composites were performed using a scanning electron microscope (Hitachi S-4800 II). Cross sections of the samples (broken in liquid nitrogen) were investigated in order to study particle distribution and morphology affecting the thermal conductivity of the different systems. The major goal was to check, whether a homogenous filler distribution over the sample was given and to visually prove for the samples with low to medium filler contents, that no thermal percolation over the filler network was present.

## RESULTS AND DISCUSSION

Thermal conductivities of silicone rubber as a function of aluminum filler loading are indicated in Figure 1 measured at a



**Figure 1.** Comparison of thermal conductivity of aluminum filled silicone rubber with theoretical predictions (A = 10 for Lewis–Nielsen calculation; T = 25°C).

temperature of 25°C. Samples with a thermal conductivity of up to 0.3  $\text{W m}^{-1} \text{K}^{-1}$  were measured for non-percolative filler content (23 vol %) and up to 0.55  $\text{W m}^{-1} \text{K}^{-1}$  in a percolative system (40 vol %). Compared to the initial thermal conductivity of an unfilled silicone (0.1–0.2  $\text{W m}^{-1} \text{K}^{-1}$ ) this is a moderate increase.

In the dispersion system with low volume content (10–15%) of aluminum, a heat transfer via formation of conductive filler particle chains is not possible, as the filler is homogeneously distributed in the polymer matrix and the filler content is too low. The moderate increase in thermal conductivity is due to the fact, that the distances between the highly conductive particles are too long—for that reason, the thermal conductivity of the silicone is the determining parameter.

In the range of high volume content (20–40%) the thermal conductivity increases, which is attributed to formation of conductive chains by the aluminum particles. Figure 1 show also the comparison between theoretical models (Eqs. (1–4)) and experimental data for the thermal conductivity of aluminum filled silicone rubber systems. The predictions of Nielsen and Maxwell deviate significantly from the experimental data in the range above 20 vol % of aluminum filler content while fitting well the calculated data according to Agari and Bruggeman (see above).

The Agari model is a semi-theoretical method to predict the thermal conductivity of composites, and hence this model represents the data better than other models. The constants  $C_1$  and  $C_2$  are determined by curve fitting of the experimental data. Through the determination of constant  $C_2$  the ease for the formation of conductive chains was taken into account.

After a calculation  $C_2$  (information about particle interconnection) was obtained with a value of 0.351. Constant  $C_1$  gives information about the influence on the secondary structure of the matrix. This value of about 0.995 is very close to 1, which indicates that aluminum particles have a negligible effect on the secondary structure of the silicone rubber. But the determined  $C_2$  value proved that it is not very easy to form conductive

**Table I.** Experimental Data of Material Properties of Silicone Rubber, Hybrid-Material and Filler

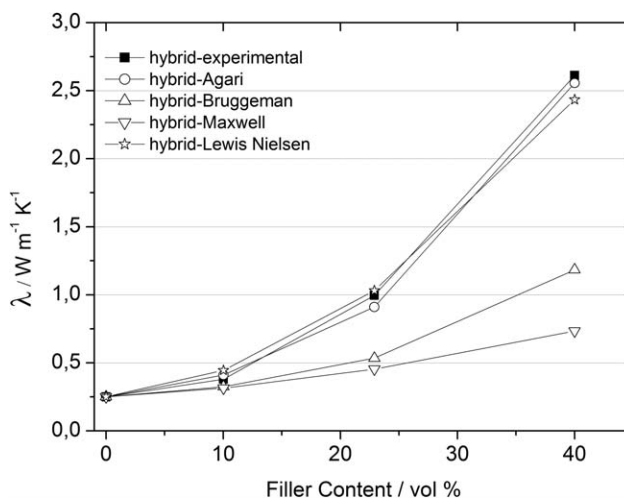
Material	Silicone	Hybrid	Aluminum
Thermal conductivity ( $\text{W m}^{-1} \text{K}^{-1}$ )	0.1–0.15	0.25	220
Density ( $\text{g cm}^{-3}$ )	1.1	1.35	2.74
Average Particle size ( $\mu\text{m}$ )	–	–	24–26
Viscosity ( $\text{Pa s}$ ) (@25°C)	4.5	0.150	–

chains in the filled silicone rubber system at the given filler concentration. In the Lewis–Nielsen model this content was just not taken into account. The average deviations between predicted and experimental conductivities are shown in Table II.

The values in Table II indicate that Agari's predictions meet the measured thermal conductivities quite well and Bruggeman's deviates significantly only at a filler content of 23 vol %. However Maxwell and Lewis–Nielsen deviate in the whole range of filler concentrations. Maxwell's assumption, that filler particles are geometrically isolated from each other in the matrix becomes inaccurate for certain filler grades (in this series starting at filler contents somewhere between 10 and 23 vol %). Taking most parameters and factors into account, the model from Lewis–Nielsen predicts a higher thermal conductivity than measured. An explanation for this observation is that the packing behavior of the particles, based on particle size distribution measurements and ideal packing density, cannot be reached in the samples prepared. Another aspect is the orientation of the particles—as the Lewis–Nielsen calculation is based on perfectly orientated particles in a matrix and perfect interfaces between filler and matrix.

The above analyses clearly indicate that a possibility to push aluminum filled silicone materials over a thermal conductivity higher than  $0.6 \text{ W m}^{-1} \text{K}^{-1}$  by filler contents of 40 vol % cannot be reached. The reason for this is the insulating impact of the silicone rubber, and—in accordance with the value of  $C_2$ —the fact that the formation of conductive chains is obviously not very likely in the considered standard silicone composites.

The new hybrid material was examined regarding its thermal conductivity using the same volume content of aluminum particles, respectively as was used for the standard silicone rubber. In Figure 2 the results of a comparison of thermal conductivity of aluminum filled hybrid material with theoretical predictions are indicated. The thermal conductivity is significantly higher in

**Figure 2.** Comparison of the thermal conductivity of aluminum filled sol-gel-derived hybrid material with theoretical predictions ( $A = 10$  for Lewis–Nielsen calculation;  $T = 25^\circ\text{C}$ ).

the hybrid material composite. All thermal conductivity models fit the measurement results very well for low volume contents (0–10 vol %).

In the range above 10 vol % only the Lewis–Nielsen and the semi-theoretical Agari model agree with the experimental data. The  $C_1$  and  $C_2$  constants were also determined through curve fitting of the experimental data. Like for the silicone rubber discussed above, the  $C_1$  value of 0.988 is also very close to 1. The  $C_2$  value of 0.797 indicates a better thermal conductivity compared to the silicone samples. This allows the assumption, that conductive pathways are formed more easily in the synthesized hybrid materials. The high thermal conductivity of particles contributes to the change of the conductivity of the sol-gel composite material effectively.

By filling the hybrid material with particle contents of 10 vol % a higher thermal conductivity compared to silicone rubber can be achieved while reaching almost the values of a 23 vol % filled silicone. Going up to 40 vol % filler amount in the hybrid material matrix a thermal conductivity of  $2.6 \text{ W m}^{-1} \text{K}^{-1}$  was measured. The average deviations between predicted and experimental conductivities are listed in Table III and represent the accuracy for each model.

For the Lewis–Nielsen model, there is a deviation of the calculated values from the experimental ones in the low filler content range. An explanation for this is, that the particles used, are not perfectly oriented in the binder as assumed by the Lewis–

**Table II.** Average Deviation Between Calculated and Experimental Thermal Conductivities of Silicone Material with Different Volume Fraction of Aluminum Particles

Filler content (vol %)	Experimental $\lambda$ ( $\text{W m}^{-1} \text{K}^{-1}$ )	Average deviation (%)			
		Agari	Bruggeman	Maxwell	Lewis–Nielsen
10	0.176	4.9	1.8	8.8	14.2
23	0.318	6.6	16.9	26.4	20.2
40	0.565	1.2	5.9	35.5	48.5



**Table III.** Average Deviation Between Calculated and Experimental Thermal Conductivities of Hybrid Material with Different Volume Fraction of Aluminum Particle

Filler content (vol %)	Experimental $\lambda$ ( $\text{W m}^{-1} \text{K}^{-1}$ )	Average deviation (%)			
		Agari	Bruggeman	Maxwell	Lewis-Nielsen
10	0.380	8.0	14.5	17.3	17.1
23	0.995	8.5	46.3	54.4	3.6
40	2.612	2.1	54.7	71.9	6.8

Nielsen approach. This effect is more pronounced, the lower the filler content is. As particles with an aspect ratio of 5 are used, a mismatch in the orientation will strongly influence the experimental measurements.

According to Table III, the Agari and Lewis–Nielsen prediction deviates slightly from the experimental data. This suggests, that the reduced viscosity of the sol-gel-derived hybrid material allows the filler particles to move and orientate more easily to each other after casting the sample. As a result conductive pathways are generated and the interfaces between filler and hybrid binder can be tightly formed. Preparing a hybrid material sample with an identical content of filler amount will therefore result in a more dense packing of the filler particles than in a silicone reference.

The experimentally measured thermal conductivity of the filled hybrid material and silicone rubber are compared in Figure 3. It is obviously indicated that the same filler grade precipitates a higher overall thermal conduction in the hybrid material composite. As described above a reason for this could be a more dense packing structure in the hybrid material. Morphological analysis was performed by SEM to get information about packing behavior of the aluminum particles in the different matrices.

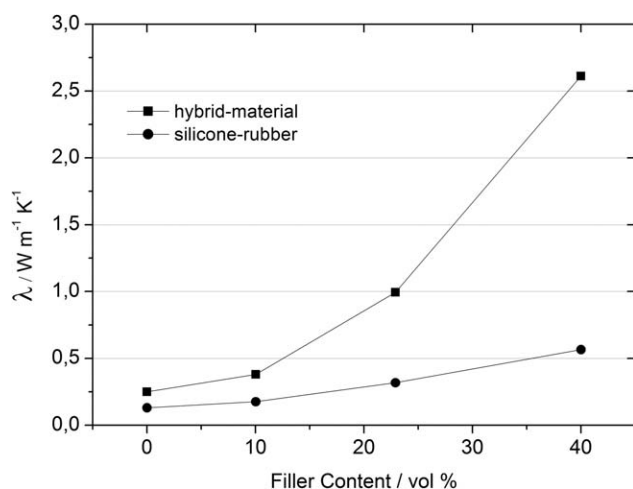
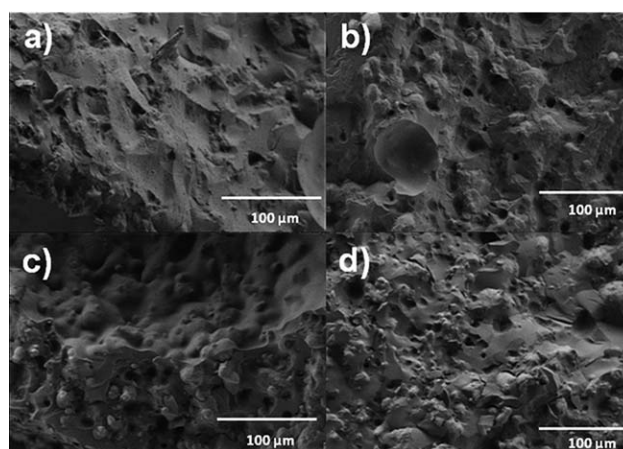
The state of filler distribution in the various aluminum-composites is shown in Figure 4 for particle contents of 23 and 40 vol %. Comparing the SEM images of filled silicones and hybrid matrices, there is no obvious structural evidence for the formation of conductive pathways, that means, the filler is homogeneously distributed in the binder without forming clearly visible domains of

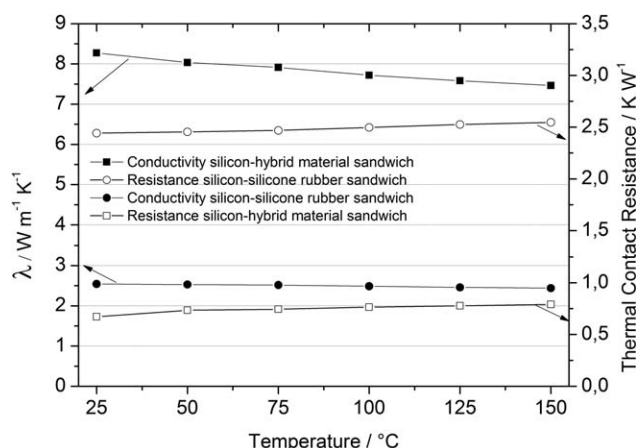
increased particle concentration. As a result, another mechanism must be responsible for the overall increased thermal conductivity of the hybrid material samples. Besides the previously described facilitated orientability of particles in the hybrid material there was the assumption, that improved thermal phase interconnections between filler and matrix play a major role.

### Sandwich Set-Up

To verify the assumption that differences in the matrix-filler interface regions in the aluminum-composites are responsible for the above described observations, a new test set-up was chosen. Therefore a sandwich-sample set-up of two silicon wafers, glued together with a silicone rubber or hybrid material with a defined thickness of about 30  $\mu\text{m}$ , was developed. Silicon wafers were chosen as an aluminum replacement material due the smooth surface properties and comparable thermal conductivity. The native oxide layer of the silicon offers the possibility to generate covalent bonds with the binder material. As all parameters of the set-up, such as conductivities of silicon and the unfilled matrix as well as the wafer and interlayer thickness were known, it was possible to calculate the thermal interface barriers. These results are displayed below in Figure 5. The thermal contact resistance of the hybrid material interface was measured to be in the range of 0.6–0.55  $\text{K W}^{-1}$  (25–150°C) while the silicone reference sandwich set-up thermally insulates four times better (ca. 2.5  $\text{K W}^{-1}$ ) in the same temperature range.

The analysis indicates that these improved thermal interlayer transitions are due to the fact, that the hybrid material is able to

**Figure 3.** Thermal conductivity of hybrid material and silicone rubber with different amounts of filler content.**Figure 4.** SEM images of silicone rubber samples filled with 23 vol % (a) and 40 vol % (b) aluminum particles and hybrid material samples filled with 23 vol % (c) and 40 vol % (d).



**Figure 5.** Comparison of overall thermal conductivity of sandwich set-up ( $y$ -left), and overall thermal contact resistance ( $y$ -right).

form covalent bonds with the filler. This can happen for example by the reaction of epoxy- or silanol-groups of the binder with hydroxyl groups of the aluminum. In contrary, silicone is much less reactive and can only form bonds via hydrogen-bridging, which are less efficient in conducting lattice vibrations.

## CONCLUSIONS

A sol-gel-derived hybrid material based on organofunctional silanes was synthesized and tested with different fractions of aluminum particles as a filler to increase the thermal conductivity. This hybrid material is compared to a silicone rubber, which was filled with aluminum particles in the same manner. The advantage of this hybrid material class is the ability to adjust the overall thermal conductivity of the material system by using different amounts of filler in a broad range. The filled hybrid material offers an up to five time's higher thermal conductivity (e.g.  $2.6 \text{ W m}^{-1} \text{ K}^{-1}$ ) than the corresponding silicone reference (e.g.  $0.6 \text{ W m}^{-1} \text{ K}^{-1}$ ). Different theoretical thermal conduction models and the measured values were compared. Besides, an additional sandwich test set-up was applied to analyze thermal conduction behavior. The reason for increasing the overall thermal conductivity with a hybrid material (unfilled state  $0.25 \text{ W m}^{-1} \text{ K}^{-1}$ ) which provides higher thermal conductivities than a thermally comparable silicone ( $0.15 \text{ W m}^{-1} \text{ K}^{-1}$ ), was investigated. It was found that the thermal connection between matrix and filler is up to four times better in the sol-gel hybrid material and as the samples prepared are mostly filled below the percolation threshold, resulting in a fourfold increased thermal conductivity of the samples.

These results reveal that the thermal interface between the aluminum filler particles with high thermal conductivity and the low conductive binder phase plays a major role for the overall thermal conductivity. It can be stated, that the improved interphase connections in the hybrid material lead to a double or even higher thermal conductivity when working at filler amounts above 10 vol %.

Finally it was demonstrated that sol-gel-based hybrid materials are potentially useful for applications as thermally conductive potting materials or glues. For further evaluation of applicability

in specific areas or products requires additional tests such as hydrolytic stability, UV-resistance or thermo mechanical analysis.

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