Synthesis and Effective Thermal Conductivity Measurements of Hollow Mesoporous SiO$_2$ Spheres for Heat-Insulating Applications

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Mesoporous silica hollow spheres are an excellent model system to investigate the thermal conductivity for an efficient heat-insulating material with respect to its geometry. Four different monodisperse silica hollow spheres are synthesized via a three-step synthesis consisting of emulsifier-free emulsion polymerization, modified Stöber condensation process, and subsequent calcination. In this approach, cetyltrimethylammonium bromide (CTAB) is used as a structure directing component to produce a highly porous silica shell. The systematic investigations of the effective thermal conductivity (ETC) allow distinguishing the respective conduction pathways, such as solid conduction and gas conduction. The carried out thermal conductivity measurements reveal for all four samples promising low ETC values of $\approx 36$ mW m$^{-1}$ K$^{-1}$ at 1013 mbar and 35 °C. In vacuum (0.03 mbar) all four samples showed, independent of shell thickness and inner diameter, a comparable reduced ETC of about 10 mW m$^{-1}$ K$^{-1}$. The comparison with previous studies on unstructured silica hollow spheres indicates that the solid state conductivity within the bulk is more dependent on the contact strengths and the number of contacts than on the thermal conductivity within the silica material, as the scattering probability of phonons is not influenced by an increased density of defects.

1. Introduction

In many industrial and private sectors, energy management is a fundamental approach to increasing process efficiency and reducing thermal energy losses. Thermal insulation materials are of great interest, as they are supposed to counteract the natural diffusivity of thermal energy. These materials’ efficiency is based on their structural properties and can be described by their effective thermal conductivity (ETC). The ETC is a measure to describe the conductivity of heat within a system. For insulation materials, the lowest possible thermal conductivity values aim to keep the heat exchange between two heat reservoirs as low as possible. The ETC represents a superposition of the respective conduction pathways and results from individual contributions, such as heat transport over the solid, through the gas phase, as well as through radiation and convection.[1] Considering this, increasing efficiently insulation materials has been developed by reducing the structural dimensions in such materials and taking advantage of the Knudsen effect.[2] This considers, that the thermal conductivity of a gas decreases when the size of the surrounding solid material is in the order of the mean free path of the gas molecules.[2] Accordingly, the thermal conductivity is determined by the gas pressure and the interfaces within the material, so that nanostructured materials represent an alternative with potential as insulation materials. The spectrum of nanostructured materials extends over a wide range, from various aerogels (0.012 mW m$^{-1}$ K$^{-1}$) to conventional polymer materials (30 and 50 mW m$^{-1}$ K$^{-1}$).[2,3] Among these materials, silica hollow spheres are a promising material due to their structural similarity to aerogels. Unlike silica aerogels, they do not have to be dried with supercritical carbon dioxide and exhibit high mechanical stability.[3b,4] Furthermore, silica is a fascinating material system, as it is inert to most aggressive chemicals, has high thermal stability and is available in abundance. The interest in silica hollow spheres for use as thermal insulation material grew steadily.[1,5] In previous works, low effective thermal conductivities were measured in a range from 20 up to 141 mW m$^{-1}$ K$^{-1}$.[1,5a–e,6] The reason for reduced thermal conductivity is assumed to be a significantly lower density associated with a reduced number of solid–solid interfaces and a reduced thermal conductivity via gas phase.[1] It has been shown that if a self-assembled crystal made of hollow spheres deviates from the local order, the thermal conductivity is significantly reduced with decreasing pressure.[1] In such a crystal with close-packed spheres, each hollow sphere has 12 nearest neighbors and 12 contact points through which heat can be conducted. If this packing is disturbed and the number of contact points is reduced, fewer conduction pathways are made possible and consequently the bulk density as the solid thermal conductivity is reduced.[1] In addition, a
2. Results and Discussion

2.1. Synthesis and Structural Features of SiO$_2$-Hollow Spheres

Mesoporous silica hollow spheres can be synthesized using different approaches, such as soft-template-assisted methods or by dissolution-regrowth processes. In this work, a template-based method was used, which comprises three process steps: the synthesis of polystyrene (PS) core particles, the coating via modified Stöber condensation process and the subsequent calcination to remove the organic components. The stepwise synthesis has the advantage that the inner diameter can be adjusted independently of the shell properties and allows the control of structural features separately. Considering that the inner diameter of hollow spheres directly influences the ETC, two different-sized PS particles were synthesized. The emulsifier-free emulsion polymerization with [2-(methacyloyloxy)ethyl]trimethyloxonium-chlorid (MTC) as comonomer resulted in smaller PS particles with an average diameter of 120 ± 6 nm. The absence of MTC resulted in a diameter of 355 ± 10 nm determined by transmission electron microscopy (TEM) analysis. The subsequent combination of condensation process with the lyotropic liquid-crystal phase of hexadecyltrimethylammonium bromide, which acted as a soft template, allows fine adjustment of the deposited layer properties. The shell thickness, i.e., can be adjusted by the amount of precursor concentration and morphology such as the pore width by varying chain length of the carbon backbone.

The particles were examined by TEM investigations before and after calcination and are shown in Figure 1 as an example of sample IS-115/21. The red arrow indicates the transition from the organic core to the inorganic silica shell. After removing the organic compounds, the contrast increased significantly, and the inner core can be more easily distinguished from the shell. Micrographs show a homogeneous texture of the shell and indicate a high porosity. At this point, it is important to note that for all samples, a similar texture was recorded. The TEM images shown in Figure 2 depict that the shell thickness can be varied over the precursor volume. As desired, 6, 8, and 10 mL tetraethylorthosilicate (TEOS) usage resulted in shell thicknesses of 16 ± 2, 21 ± 2, and 25 ± 2 nm, respectively.

The average inner diameters varied from 115 to 306 nm, and the corresponding layer thicknesses are summarized with the respective data from the nitrogen physisorption experiments in Table 1; corresponding statistical data are shown in Figure S2, Supporting Information. Remarkable is the high monodispersity and the narrow distribution of the shell thickness.

To exclude the influence of organic residues on the thermal conductivity, thermogravimetric analysis (TGA) was carried out on the core–shell particles to determine an appropriate calcination temperature. The corresponding thermogram is shown in Figure 3. The weight plotted against the sample temperature shows three significant weight losses.

At temperatures up to 150 °C, a mass loss of about 5% is observed. This loss is caused by desorption of residual solvents or water molecules. In the range from 200 to 300 °C, a larger step with a mass loss of about 24% was recorded, but assigning is difficult due to the several residuals as polyvinylpyrrolidin (PVP), CTAB, and PS. The last and most significant mass loss of about 37.5% occurs at 400 °C. This can be assigned to the decomposition of carbonaceous polymers, which is in good agreement with the literature, where the decomposition temperatures of polymers were investigated. At 550 °C the weight loss stagnates, so we consider this temperature as an appropriate calcination temperature to exclude the organic components’ influence. This is also supported by elemental analysis using energy dispersive X-ray (EDX) spectroscopy before and after calcination. The

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![Figure 1](https://example.com/figure1.png)

**Figure 1.** TEM images (magnification: 150,000), a) core–shell particles and b) hollow silica particles.
corresponding spectra are shown in Figure S3, Supporting Information. Accordingly, higher carbon content is observed for the sample before calcination, but a reliable quantification is not provided due to the preparation on copper grids with carbonaceous films. Independent of the carbon content, the presence of silica is verified via the atomic ratio of oxygen ($61.9 \pm 2.5\%$) to silicon ($32.5 \pm 0.9\%$), which corresponds to the chemical composition of silica ($\text{SiO}_2$). Further examination of the microstructure by X-ray diffraction showed an amorphous structure for all samples (see Figure S4, Supporting Information), which is typical for silica particles synthesized via TEOS hydrolysis using a modified Stöber condensation process. Structural features as pore size distribution, specific surface area, and total pore volume are accessed via nitrogen physisorption measurement, by applying Brunauer–Emmett–Teller (BET) and nonlocal density functional theory (NLDFT) calculations on recorded data. The physisorption isotherms for the respective samples are shown in Figure 4a and have three significant features. First, a step at 0.03 relative pressure indicates the presence of micropores ($<2 \text{ nm}$), which have not been further investigated. A characteristic bend was recorded at $0.3$ relative pressure and related to filling the mesopores ($2–50 \text{ nm}$), which are also reflected by the pore size distribution shown in Figure 4b. A rapid increase at relative pressure $0.95$ can be assigned to filling the particle bed’s interstitial voids and is not relevant for further investigations. Concerning the controlled introduction of porosity via the soft-templating method, the pore size distribution obtained by NLDFT shows a narrow distribution with a mean pore size of $35 \text{ Å}$ for each sample. This is consistent with previous work where silica with mesopores in $38 \text{ Å}$ was determined using CTAB as structure directing agent. The obtained BET surface areas are comparable within the sample series and range from $1056$ to $1163 \text{ m}^2 \text{ g}^{-1}$ and the total pore volumes which range from $0.772$ to $1.068 \text{ cm}^3 \text{ g}^{-1}$ gained by NLDFT. The slight differences in total pore volume could be an indication of closed mesopores. These could occur due to the formation of hard agglomerated particles, which are also shown in the TEM image (Figure 2b). An increased local TEOS concentration with accompanying hydrolysis may have led to the merging of some particles. In summary, the applied synthesis protocol results in highly monodisperse silica hollow spheres with a distinct mesostructure.

### 2.2. ETC of SiO$_2$-Hollow Spheres

The ETC can be theoretically described by the superposition principle, according to Equation (1).
For this reason, the following discussion deals with the respective contributions, considering the different pathways.

### 2.2.1. ETC Measurements of SiO₂-Hollow Spheres Depending on Temperature and Pressure

The ETC values of different silica hollow spheres are depicted in Figure 5. The results are shown for each specimen at pressures from 0.03 to 1013 mbar and 35, 50, and 70 °C, respectively. All samples show a pronounced pressure dependency of the ETC in the range from 10 to 40 mW m⁻¹ K⁻¹. In contrast the influence of the temperature is much smaller. Increasing the temperature from 35 to 70 °C, the ETC is enhanced by about 2 mW m⁻¹ K⁻¹ at the respective pressure, which is caused by an increase in heat capacity with temperature.\(^{[19]}\) Regarding the pressure dependency, samples IS-115/X (X = 16/21/25) show a thermal conductivity of about 36 to 9.6 mW m⁻¹ K⁻¹ with decreasing pressure and sample IS-306/27 from 35.5 to 9.3 mW m⁻¹ K⁻¹. As can be seen, reduced pressure led to lowered thermal conductivity which is due to the previously introduced Knudsen effect. The mean free path increased with decreasing pressure and as result the gas phase contribution to the ETC is diminished. It is also noticeable at this point, that the thermal conductivity decreases more rapidly with decreasing pressure for smaller particles than for the larger ones, which is discussed in more detail in Section 2.2.2. First of all, it can be stated that under vacuum conditions, all samples show a strongly reduced thermal conductivity in comparison with bulk silica (1.4 W m⁻¹ K⁻¹) and can therefore be assigned to the super insulation range \((λ_{\text{eff}} ≤ 0.03)\).\(^{[19b,20]}\) In contrast, the thermal conductivity values under normal conditions (1013 mbar) are comparable with those of conventional insulation materials.\(^{[19b]}\) This correlation is considered for using hollow spheres in vacuum panels. If the vacuum in such a panel breaks, the pressure increase only leads to a small increase in thermal conductivity of about 27 mW m⁻¹ K⁻¹. Consequently, the insulation properties of such panels will not completely collapse even if residual pressure is increased.\(^{[9e]}\)

In addition, the data can be used to conclude whether the Knudsen effect can be exploited under standard conditions.
(1013 mbar). To do this, Equation (2) is used, and it is assumed that under standard conditions, gas and solid conductivity are involved simultaneously ($\lambda_{\text{eff, 1013}}$), whereas in vacuum (0.03 mbar), the solid conductivity dominates the ETC. If the difference is formed from these two values, it should represent the contribution of the gas component to the ETC. By comparing the difference with the thermal conductivity of dry air, it is possible to determine whether the thermal conduction in the gas phase is reduced following the Knudsen effect. This is shown in Figure 6.

As can be seen, the difference for all samples at 35 °C and the thermal conductivity of dry air are in the same order of magnitude of ≈27 mW m⁻¹ K⁻¹.[21] This indicates that at 1013 mbar the Knudsen effect does not occur for any of the samples. This is also supported when the mean free path is considered as a function of temperature. The mean free path increases from about 76 to 84 nm and remains below the specific lengths of the hollow spheres. It becomes clear that the structural parameters such as inner diameter and interparticle spaces are still too large for a significant reduction of the ETC according to the Knudsen effect, both under normal conditions and at elevated temperature. Under vacuum conditions (0.03 mbar), a low thermal conductivity of 8 mW m⁻¹ K⁻¹ has been reported only once in the literature and is comparable with 9.3 mW m⁻¹ K⁻¹.[3] This result was measured on silica hollow spheres with an inner diameter of 469 nm and a shell thickness of 17 nm at 25 °C, whereby no structuring of the shell was carried out. The applied nitrogen physiosorption experiments revealed a total pore volume of 0.3 cm³ g⁻¹ compared with this study (0.772 to 1.068 cm³ g⁻¹) so that the bulk densities are lowered by a factor of two. The comparison of these studies regarding the thermal conductivity values indicates that a structured shell did not lead to a measurable

![Figure 5](image_url) Energy Technol. 2021, 9, 2001048 2001048 (5 of 9) © 2021 The Authors. Energy Technology published by Wiley-VCH GmbH
reduction of the thermal conductivity, although the structuring of the shell is in a magnitude order of the scattering lengths of phonons. In amorphous silica materials, the mean free path length of the phonons is about 1–10 nm.\(^{[2,7]}\) Considering this, it is assumed that the wall thickness within the shell is not thin enough to generate an increased scattering probability of phonons, which would reduce the thermal conductivity via solid phase. However, as in previous work, the particles’ contact area and the conditions under which they were synthesized must also be considered, as these influence the bond strength and, consequently, the ETC.\(^{[5c]}\) Thus, based on the studies’ similarity, it is assumed that both the bond strength and the number of contact points have a dominant influence on the thermal conductivity.

### 2.2.2. ETC Measurements of SiO\(_2\)-Hollow Spheres Depending on Inner Diameter and Shell Thickness

Figure 7a shows ETC values of SiO\(_2\) hollow spheres with an inner diameter of 115 nm, but varying shell thicknesses of 16, 21, and 25 nm. Figure 7b shows the results of hollow spheres with inner diameters of 115 and 306 nm, but a similar shell thickness of 25 and 27 nm.

In the pressure range from 100 to 1013 mbar, sample IS-115/25 shows the highest thermal conductivity (Figure 7a). For example, at 1013 mbar, the ETC value of IS-115/25 is 37.8 mW m\(^{-1}\) K\(^{-1}\) compared with IS-306/25 which has 36.7 mW m\(^{-1}\) K\(^{-1}\). The lowest ETC value of 35.7 mW m\(^{-1}\) K\(^{-1}\) (IS-115/21). However, as the pressure is further reduced from 100 mbar, sample IS-115/21 shows even higher ETC values, with the ETC values converging from 0.1 mbar. As the results show, there is no direct trend in the comparison of the shell thicknesses. It is assumed that the random arrangement of the hollow spheres has led to a statistical distribution of the interstitial cavities between the particles and thus the Knudsen effect is differently pronounced when the pressure is reduced. However, it is noteworthy that regardless of the shell thickness, the ETC values at 0.03 mbar are comparable (IS-115/16 9.6 mW m\(^{-1}\) K\(^{-1}\); IS-115/21/25 9.9 mW m\(^{-1}\) K\(^{-1}\)). This supports that the solid conduction is dominated by the phonon contribution or thermal contact conductance. Therefore, the solid conduction is independent of the thickness of the coating, as the scattering lengths have not been influenced. As all samples have been prepared under the same conditions, it is assumed that the bond strength is comparable within the shown sample series.

The measured ETC values for hollow spheres with different diameters (Figure 7b) show a different behaviors depending on the pressure. Starting from a pressure of 1013 mbar, sample IS-115/25 shows a slightly higher ETC value of 37.8 mW m\(^{-1}\) K\(^{-1}\) compared with IS-306/27 with 35.5 mW m\(^{-1}\) K\(^{-1}\). This may be due to the increased bulk density, which is 0.097 g cm\(^{-3}\) for the larger particles and 0.136 g cm\(^{-3}\) for the smaller ones (Table 2). It is assumed that increased solids content has led to the slightly higher thermal conductivity. With further reduction from a pressure of 300 mbar, this behavior reverses and the larger particles exhibit a higher thermal conductivity. This is also due to the Knudsen effect already mentioned. If Equation 3 and a constant pressure of 100 mbar are considered then the mean free path \(l_m\) is the same for both samples. But, due to the smaller diameter, sample IS-115/25 has smaller characteristic free spaces \(d_b\) compared with IS-306/27 and thus, at the same pressure, the thermal conductivity according to Equation 3 is reduced to a greater extent. As with the comparison of the shell thickness, the ETC values converge from a pressure of 0.1 mbar. This reinforces the statement, that the solid-state conductivity in such a powder bed is dominated by the particle–particle contact conductance as the number of contact points. Independent of this, it is evident that a size reduction of the nanoparticles is necessary to reach the super isolation range at standard pressure of 1013 mbar.

### Table 2. Sample overview with used masses, calculated bulk density, and bulk porosity.

| Sample     | Weighed mass [g] | Bulk density [g cm\(^{-3}\)] | Bulk porosity [%] |
|------------|------------------|------------------------------|------------------|
| IS-115/16  | 0.8092           | 0.133                        | 93.91            |
| IS-115/21  | 0.7982           | 0.131                        | 93.99            |
| IS-115/25  | 0.8295           | 0.136                        | 93.77            |
| IS-306/27  | 0.5183           | 0.097                        | 95.59            |

Figure 7. ETC values of a) SiO\(_2\) hollow spheres with the same inner diameter, but different shell thicknesses, b) SiO\(_2\) hollow spheres with different inner diameters, but similar shell thickness in dependence of pressure.
3. Conclusion

In this study, a precise synthesis protocol for the preparation of very monodisperse silica hollow spheres was described. CTAB as a soft template led to the successful structuring of the silica shell, so that very uniform meso pores were incorporated. The ETC measurements revealed that all samples have a reduced ETC in the range of 35.7–37.8 mW m⁻¹ K⁻¹ 1013 mbar and 35 °C. Furthermore, pressure reduction provided a better understanding of the heat conductivity of these materials. It was found that thermal conductivity occurs mainly via the gas phase at 1013 mbar and dominates in the solid at lowered pressure of 0.03 mbar, where all samples showed strongly reduced thermal conductivity to 9 mW m⁻¹ K⁻¹. The comparison with previous studies on unstructured silica hollow spheres shows that the solid-state conductivity within the bulk is more dependent on the contact strengths and the number of contacts than on the thermal conductivity within the silica material. For further work, the variation of density by varying soft templates would be interesting, as well as a reduction of the inner diameter to take advantage of the Knudsen effect already at standard pressure (1013 mbar). A shell matrix with walls less than 1 nm thickness would also be attractive, but as with aerogels, this would certainly lead to a significant reduction in the samples’ mechanical stability.

4. Experimental Section

Materials and Chemicals: Styrene (≥99%, Sigma-Aldrich) was pre-treated with sodium hydroxide solution (1 m) for inhibitor removal (three times with NaOH solution and two times with distilled water), 2- (2-Azobis(2-methylpropion-aminid)dihydronitroso) (AIBA; ≥98%, Acros Organics), 2-(methacryloyloxy)ethyltrimethylammonium chloride (MTC, 75 wt% in water, Sigma-Aldrich), PVP (M₆ = 55 kg mol⁻¹, Sigma-Aldrich), ammonium hydroxide solution (NH₄OH, 25% in water, VWR Chemicals), hexadecyltrimethylammoniobromid (CTAB, ≥99%, Sigma-Aldrich), absolute ethanol (≥99.8%, Sigma-Aldrich), and TEOS (≥99%, Sigma-Aldrich) were used as-received. Water was taken from an in-house Milli-Q-system (16.3 MΩ).

Synthesis of PS Particles: Monodisperse PS particles with 120 ± 6 nm diameter were synthesized via emulsifier-free emulsion polymerization reported by Yao et al. by adding 300 μL MTC from as-received. Water was taken from an in-house Milli-Q-system (16.3 MΩ).

Synthesis of Hollow Mesoporous Silica Spheres: Synthesis of hollow mesoporous silica spheres. The previously synthesized PS particles served as sacrificial particles for the hollow sphere synthesis. For this purpose, PS particles were coated with mesoporous silica via a modified Stober condensation process and subsequently calcined to obtain hollow spheres. The coating was carried out in a single-neck flask filled with a suspension consisting of 150 mL water, 204 mL ethanol, and 7.6653 g CTAB, which corresponds ideally to PS mass concentration of 6.01 g L⁻¹. For homogenization (a homogeneous distribution of the ligands), the solutions were first stirred at 500 rpm for 1 h. Subsequently, the pH value was adjusted to 11.2 with ammonium hydroxide solution. Afterward, different TEOS volumes were slowly dripped into the solution (respectively, assigned: 6, 8, and 10 mL for IS-115/16 115/(21), 115/25, and 2.59 mL for sample IS-306/27). After all TEOS volumes had been added, the reaction was stirred for 112 h at 500 rpm and room temperature. Before calcination, the core-shell particles were washed with water by first sedimenting the particles using centrifugation (10 min at 6000 rpm). The supernatant liquid was then removed, and sediment particles were redispersed in water via sonication. This process was repeated three times, and subsequently, the particles were dried for 12 h at 60 °C. The calcination was carried out in a sintering oven, where particles were heat-treated for 2 h in the air at 550 °C applying a heating rate of 1 K min⁻¹. As the particles formed soft agglomerates after calcination, all samples were carefully ground using a pestle and mortar. The shells’ integrity was checked by TEM analysis afterward, and exemplary images are shown in Figure S1, Supporting Information.

Characterization Methods: TEM images were recorded with a Hitachi HF-2000 microscope equipped with a cold field emission gun and a Thermo Fisher Scientific NORAN energy dispersive X-ray (EDX) System SIX at an acceleration voltage of 200 kV. Images were taken to examine the morphology such as the diameter and shell thickness of the hollow spheres.

Nitrogen adsorption–desorption measurements were recorded using the NOVA 3000e from Quantachrome Instruments. Before the measurements, the as-synthesized silica hollow spheres were evacuated at 150 °C for at least 10 h. The isotherms were recorded at 77 K using the static volumetric method and nitrogen as process gas. BET surface areas were calculated from the data in a relative pressure range from 0.04 to 0.2. Pore volumes and pore size distribution were determined by applying nonlocal density functional theory (NLDFT) with silica adsorption branch kernel at 77 K based on a cylindrical pore model. The kernel was provided by NOAVin software.

To determine a suitable calcination temperature, TGA was carried out with thermal analyzer TGA/DSC 1 from Mettler Toledo equipped with a gas controller GC 200 (synthetic air: 50 mL min⁻¹). Samples were placed in a crucible, and thermograms were collected with a heating rate of 1 K min⁻¹.

Powder X-ray diffraction patterns were recorded in a Debye–Scherrer geometry on a STOE Stadi P diffractometer using Mo radiation (0.0709 Å), equipped with a primary Ge (111) monochromator (Mo Kα) and a position-sensitive Mythen1K detector. Data were collected in the range between 2° and 60° 2θ with a step width of 0.03° 2θ. Measuring time per step was 40 s. For each sample, eight scans were collected and summed after data collection. To exclude the influence of the quartz glass capillaries (Ø 0.5 mm), blank measurements were first carried out, which were later deducted from sample data.

Measurement of ETC: To determine the ETC, the Transient Plane Source (Hot Disk TPS 2500S) method was applied. For this, a modified measuring setup was developed, as shown in Figure 8.

The disk-type Kapton sensor with radius 3.189 mm was installed in a sample chamber (volume of 6.09 cm³) to be embedded in loose powder beds. The depicted chamber was placed in an autoclave which allows the heat conductivity determination under different pressures. Using needle valves, the pressure could be precisely adjusted between the 0.03 and 1013 mbar, whereby the pressure increase was carried out with synthetic air with a moisture content of 0.5 ppm mol⁻¹. In addition, the measuring chamber is equipped with a heater and a thermocouple to ensure an isothermal environment. The measurements were then carried out at three different temperatures at 35, 50, and 70 °C. For each measuring point, three individual measurements were carried out and the arithmetic mean was given in this work. Further, for each data point, the evaluation was divided into three parts. First, a temperature drift was recorded for 40 s, whereby the temperature stability was assessed to ensure a precise determination of the thermophysical properties afterward. During the second part, a heat pulse was generated, and a transient reading of the temperature change was recorded. Here, the temperature increase was directly correlated by the surrounding environment, and the
Measuring chamber a) with an open lid and b) closed lid with gas inlet.

thermophysical properties were calculated and evaluated by the device software. A detailed description can be found in the literature.[17,23]

Sample Preparation: The mesoporous silica hollow spheres were loosely filled as powder beds into the measuring chamber. To achieve comparable measurements, bulk densities and bulk porosity were calculated by weighting the sample amount divided by the filled chamber volume and shown in Table 2. Prior to measurements, all samples were evacuated overnight for at least 12 h to remove residual moisture, which may tremendously influence the overall heat conductivity.[24]

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
F.W. is grateful for basic financial support of the Max-Planck-Society. Moreover, acknowledgements go to Dr. Claudia Weidenthaler and J. Ternieden for conduction of X-ray diffraction experiments at MPI KOFO in Mulheim an der Ruhr, to Bernd Sliethoff, Microscopy Department, for TEM investigations and to Dr. Hartmut Wiggers from the University of Duisburg-Essen for fruitful discussion as well supervision during the master thesis. F.W. thanks Wolfgang Kersten’s Team, the Finemechanics, for their help in the technical implementation of the measuring equipment.

Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest
The authors declare no conflict of interest.

Authors Contribution
F.W.: conceptualization, investigation, data acquisition, writing original draft and visualization; M.F.: supervision, conceptualization and review editing; M.F.: supervision, writing, review editing, project administration.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
effective thermal conductivity, mesoporous hollow spheres, packed beads, silica nanoparticles, soft templating, transient plane source methods

Received: December 1, 2020
Revised: February 21, 2021
Published online: March 22, 2021

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