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Response to "Comment on "High-Temperature Thermal Transport in Porous Silica Materials: Direct Observation of a Switch from Conduction to Radiation""

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In our work "High-Temperature Thermal Transport in Porous Silica Materials: Direct Observation of a Switch from Conduction to Radiation"[1] we investigated SiO_2 (main text) and TiO_2 (supporting information) particulate samples with respect to their high-temperature thermal transport properties. We focused particularly on the transition from bulk glass to solid and hollow spheres from a material point of view. Based on laser flash analysis (LFA), we observed an increasing amount of radiatively transported thermal energy with increasing temperature. Therefore, we show and discuss the time-signal evolution obtained from the LFA instrument for temperatures up to 925 °C, where two distinct processes can be observed. Furthermore, based on the variation of our material, we demonstrated how this radiative heat transport depends on the composition of the specimen, which is represented by the first maximum of the measured signal at short timescales.

Radiative heat transfer in LFA experiments is accounted for by appropriate models, as Dr. Lunev outlined in his comment (equation 1), and is implemented in the commercial software ("transparent model") that we used for fitting our reference samples. Since we intentionally wanted to study conductive and radiative heat transport processes at high temperatures, we did not use a metallic interlayer between our sample and the necessary graphite coating to ensure a good emissivity. We appreciate the critical discussion of Dr. Lunev on fitting the quartz glass data at high temperatures (figure 2e of ref. [1]), which resulted in thermal diffusivities that agree more closely with the literature reference based on his analysis. We note that measurements with a high radiative heat transport contribution are demanding for the evaluation procedure and that the transparent model we used for the quartz glass evaluation systematically overestimates the thermal diffusivity at high temperatures. We, therefore, thank Dr. Lunev for pointing this out and agree that the source of the discrepancy between the model that we used and the model termed "PULsE solution"[2] requires further investigation.

Dr. Lunev outlines in his response an additional model that directly couples conductive and radiative heat transport (equation 2) that he has published.[3] This model seems suitable for analyzing the radiation-conduction transition that we reported in our paper. Applying this model qualitatively reproduces the time-dependent features of a hollow sphere sample ($d = 882$ nm, $t = 31$ nm). This model includes effective material parameters such as the Planck number N_P and optical thickness τ_0. In essence, Dr. Lunev arrives at the same interpretation we outlined in our article: the radiative heat transport contribution grows with increasing temperature. The optical thickness decreases with increasing temperature since the spectrum of the grey body emitter continuously shifts toward the transparent part of the SiO_2 specimen (except for 8 µm beads).

While we appreciate the confirmation of our analysis by the model of Dr. Lunev, we do not agree with some comments in his response to our work that question the validity of our interpretation. Using a superposition of two processes that describe thermal conduction (weighted sum of the Cape–Lehman model) may appear unusual if one of these processes is of radiative nature. This simple approach, nevertheless, allowed us to separate the time-temperature signal in two separate processes that resemble a fast (radiative) and slow (conductive) heat transfer across the hollow sphere samples. Furthermore, the T^4 dependency of the contribution of the fast process further hints at its radiative nature. We, furthermore, included detailed optical modeling to analyze the radiative heat transfer in all three types of specimen (bulk, solid and hollow particles) based on fluctuational electrodynamics. This allowed us to identify the wavelength-dependent radiative heat transport channels and to model the conduction-radiation transition qualitatively.

Furthermore, our work did not report on an ad hoc measurement, but it is a result of a material development accompanied by many measurements. This development resulted in the presented work, which naturally focuses on the most relevant samples. We are well aware that the thickness of the specimen influences the interplay between conductive and radiative heat transport. However, a systematic thickness variation is hard to achieve experimentally due to the fragility of the particulate pellets.

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Overall, we appreciate the comment of Dr. Lunev to utilize and improve the existing models to describe coupled conductive and radiative heat transport processes in LFA experiments. We understand the evaluation of his model as an additional confirmation of our presented analysis. The interpretation of our results and conclusions we draw from there remain unaffected by Dr. Lunev's comment.

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Conflict of Interest

The authors declare no conflict of interest.

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[1] A. M. Neuhöfer, K. Herrmann, F. Lebeda, T. Lauster, C. Kathmann, S. A. Biehs, M. Retsch, *Adv. Funct. Mater.* **2021**, *32*, 2108370.

[2] A. Lunev, V. Zborovskii, T. Aliev, R. Heymer, O. Vilkhivskaya, *Softw. Impacts* **2020**, *6*, 100044.

[3] A. Lunev, V. Zborovskii, T. Aliev, *Int. J. Therm. Sci.* **2021**, *160*, 106695.