*Supplementary Information*

# Next Generation of Thermal Insulators for High-Temperature and Humid Environments thru Aerogel Carbonization

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## Calculation of the thermal conductivity of aerogels

The thermal conductivity of aerogel can be calculated from the contributions from thermal radiation and conduction, as follows:

$$\lambda_{total} = \lambda_{solid} + \lambda_{gas} + \lambda_{rad} \quad \text{(S1)}$$

where $\lambda_{solid}$, $\lambda_{gas}$ and $\lambda_{rad}$ are the solid conductivity, gas conductivity, and radiative conductivity, respectively. The contributions from heat conduction through solid and gas are considered as the conductive thermal conductivity, $\lambda_{cond} = \lambda_{solid} + \lambda_{gas}$.

The solid thermal conductivity indicates the contribution from heat conduction through the solid, which can be calculated using an empirical correlation, as follows [1,2]:

$$\lambda_{solid} = \lambda_{s0}\left(\frac{\rho}{\rho_0}\right)^{\alpha} \tag{S2}$$

where $\rho$ is the density of aerogel and $\rho_0$ is the density of solid backbone (1,560 kg m$^{-3}$ for RF aerogel and 1,950 kg m$^{-3}$ for carbon aerogel). $\lambda_{s0}$ is the thermal conductivity of solid backbone, which was taken as 0.18 W m$^{-2}$ K$^{-1}$ for RF aerogel [3] and as 0.7 W m$^{-2}$ K$^{-1}$ for carbon aerogel (adopted from the thermal conductivity of nano-sized graphite [4,5], which is in the same range of the thermal conductivity of activated carbon [5,6]). The semi-empirical constant $\alpha$, dependent on the random and complex pore structure, was taken as 1.2 for RF aerogel [7] and as 1.5 for carbon aerogel [8].

Based on the kinetic theory, the thermal conductivity of gaseous molecules in the porous structure can be calculated by the Knudsen model, as follows [9]:

$$\lambda_{gas} = \frac{1}{1 + 2C_1\Lambda_g/d}\lambda_{g0} \tag{S3}$$

Where $\Lambda_g$ and $\lambda_{g0}$ are, respectively, the mean free path and the thermal conductivity of gas in the bulk conduction (67 nm and 0.026 W m$^{-1}$ K$^{-1}$, respectively, for air at 300 K and 1 bar). $d$ is the mean pore size. Modified from the value around 2 for thermal transport of gas molecules confined by two parallel walls [9,10], the dimensionless coefficient $C_1$, was taken as 1.0, based on semi-empirical fitting for the complex aerogel structures from experimental observation and those generated by the Direct Simulation Monte Carlo (DSMC) simulation [11].

The radiative thermal conductivity can be expressed as [3]:

$$\lambda_{rad} = \frac{16n^2\sigma T^3}{3\rho K_s/\rho_0} \tag{S4}$$

where $\sigma$ is the Stefan-Boltzmann constant ($5.67037\times10^{-8}$ W m$^{-2}$ K$^{-4}$). $T$ is the mean absolute temperature. $n$ is the refractive index of aerogel, around 1.1, calculated by $n = 1 + (n_0 - 1)\rho/\rho_0$, with $n_0$ being the refractive index of solid backbone [12]. $K_s$ is the mean Rosseland extinction coefficient of aerogel. The specific extinction coefficient, i.e. the ratio of the mean extinction coefficient to the density of solid backbone, $K_s/\rho_0$, was taken as 50.1 m$^2$ kg$^{-1}$ for RF aerogel [1] and 1,000 m$^2$ kg$^{-1}$ for carbon aerogel [3].

**Figure S1** shows the calculated values of the total thermal conductivity and the contributions from radiation and conduction through gas and solid for the RF aerogel and carbon aerogel samples. **Figure S2** shows the calculated thermal conductivity of RF aerogel and carbon aerogels as a function of the relative density.

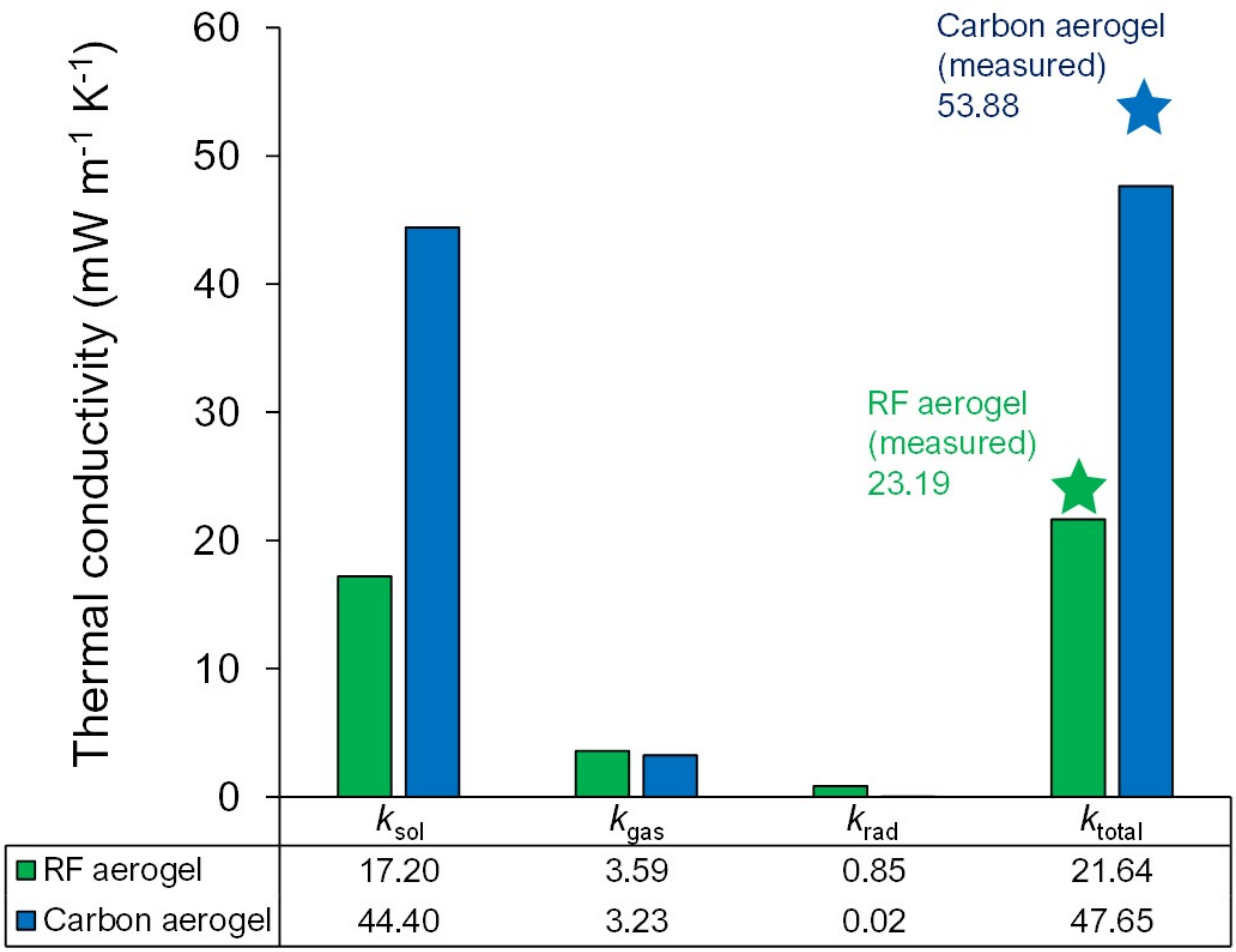


**Figure S1.** The calculated and measured thermal conductivities of RF aerogel and carbon aerogel samples.

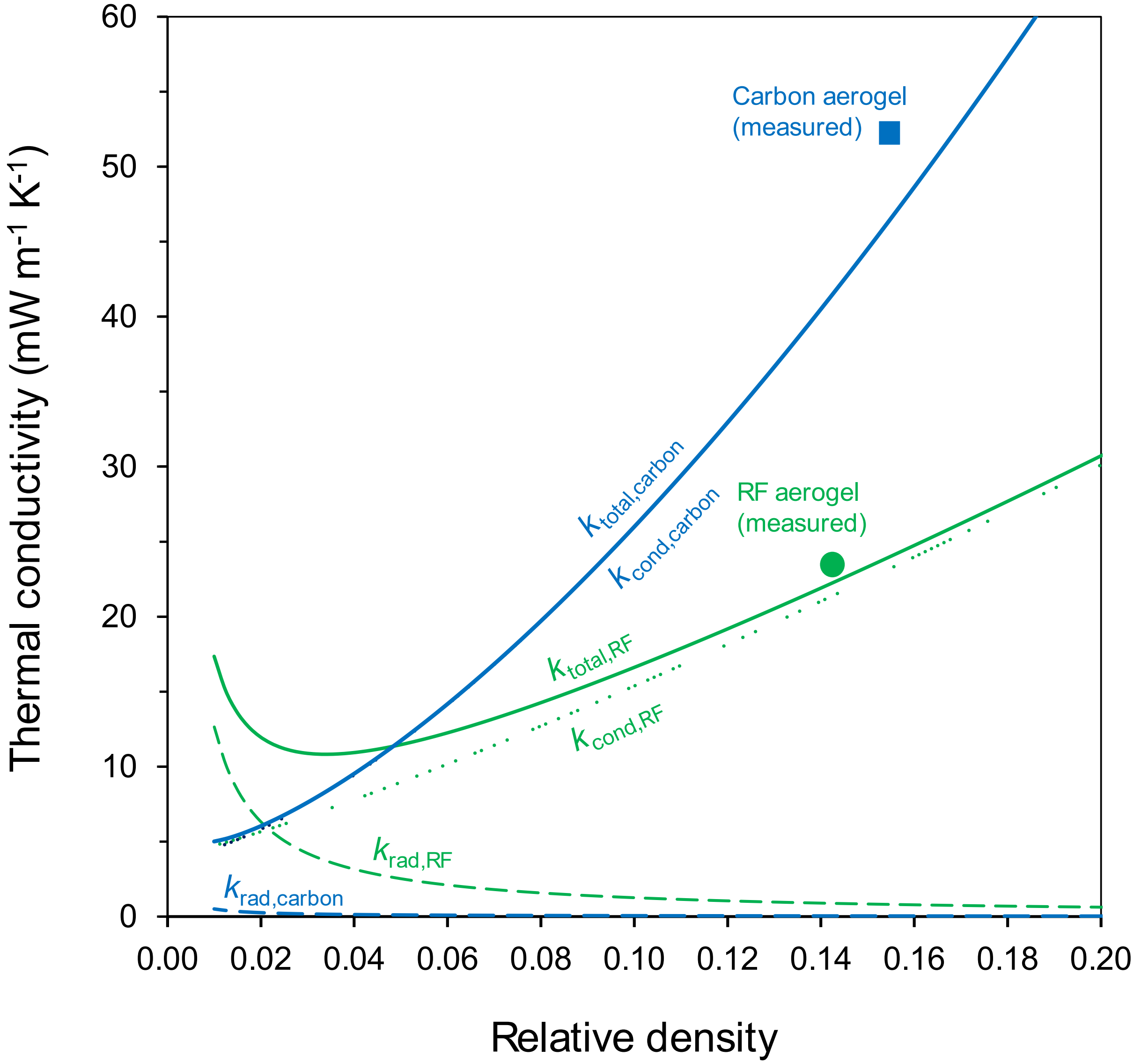


**Figure S2.** The calculated thermal conductivity of RF aerogel and carbon aerogel as a function of the relative density.

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