Supporting Information

Environmentally-friendly Polylactic Acid-based Thermal Insulation Foams Blown with Supercritical CO_2

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The IR transmittance and reflectance of the PLA film were measured using FTIR and the data are shown in Figure S1. These data were used to calculate the absorption index of the PLA matrix.

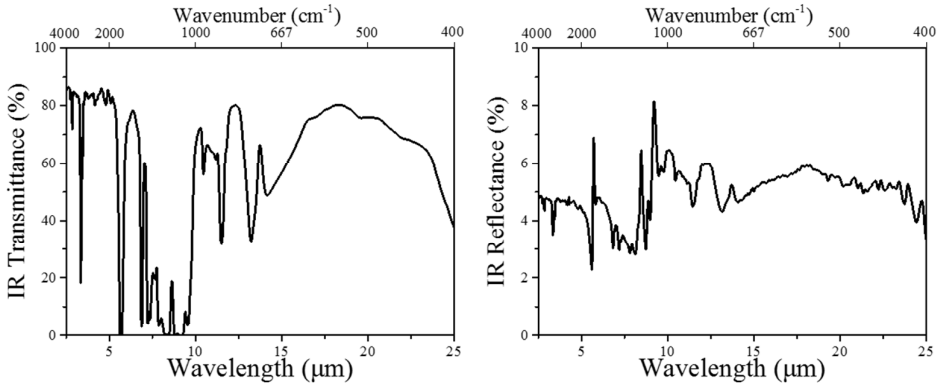

Figure S1 IR transmittance and reflectance of PLA film

Five representative wavelengths were selected as examples to show how $K_{e,\lambda}$ was calculated. By the linear regression of the $ln(\tau_{n,\lambda})$ at each wavelength against the foam thickness, L, the slope of the straight line is $K_{e,\lambda}$, as shown in Figure S2.

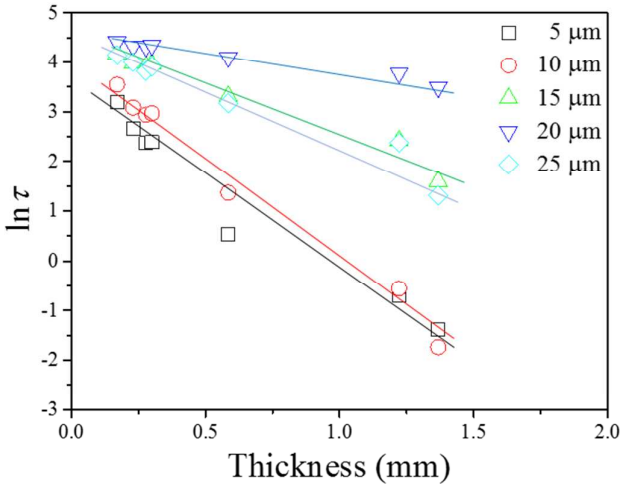

Figure S2 Transmittance of PLA foams as a function of the foam thickness.

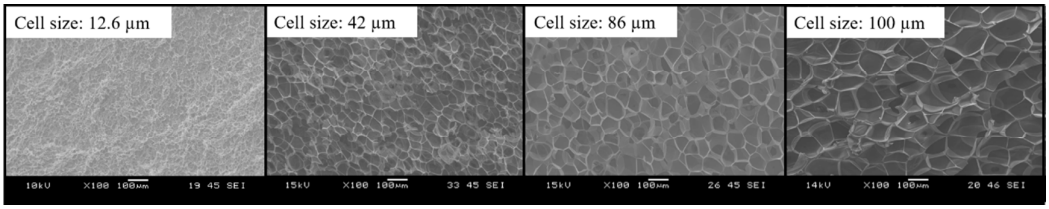

Figure S3 SEM micrographs of PLA foams

When the cell size in PLA foams is less than 3 mm, the convection in the small cells is negligible,[1] and the PLA foams' total thermal conductivity (k_{total}) can be expressed as follows:

$$k_{total} = k_{gas} + k_{solid} + k_{rad} \qquad \text{(S1)}$$

where k_{gas} and k_{solid} are the thermal conductivities via gas and solid conduction, respectively. k_{rad} is the thermal conductivity by radiation.

The PLA foams' gas conductivity is as follows:[2]

$$k_{gas} = \varepsilon_{VF} k^0_{gas} \quad \text{(S2)}$$

where ε_{VF} is the void fraction of the PLA foam and k^0_{gas} is the bulk gas conductivity, 26 mW/m-K for air.

Along with cell growth, the original spherical cells after nucleation will become polygonal when the PLA foam has a large void fraction, typically the ε_{VF} is over 94%. In this case, the heat transfer through the cells can be simplified by using Glicksman's model.[1, 3, 4] This model assumes that the cells are cubic and that the heat flux only passes through 4 faces and 4 struts of the cubic cell. Therefore, the solid conductivity can be expressed as follows:[4]

$$k_{solid} = (1-\varepsilon_{VF})(\frac{2-f_s}{3})k^0_{solid} \quad \text{(S3)}$$

where f_s is the strut fraction. The k^0_{solid} is the bulk solid conductivity.

The transmitted radiative energy can be obtained from the transmittance of IR radiation through the polymeric foams. In polymeric foams, the reflected radiation will be multi-reflected inside the porous structure. Therefore, the radiative thermal conductivity can be calculated using the Rosseland equation, which is based on a diffusion approximation.[2, 3, 5] The uniform PLA foams can be regarded as homogeneous samples from a macroscopic perspective. Then, the spectral extinction coefficient ($K_{e,\lambda}$) is independent of the foam thickness. The transmittance ($\tau_{n,\lambda}$) can be expressed as follows:[6, 7]

$$\tau_{n,\lambda} = ce^{(-\int_0^L K_{e,\lambda} dx)} \quad \text{(S4)}$$

where L is the sample thickness, and c is constant. According to the Planck distribution of IR radiation over the wavelength, the Rosseland extinction coefficient ($K_{e,R}$) has an average of $K_{e,\lambda}$ as follows:[8, 9]

$$\frac{1}{K_{e,R}} = \frac{\int_0^\infty \frac{1}{K_{e,\lambda}} \frac{\partial e_{b,\lambda}}{\partial T} d\lambda}{\int_0^\infty \frac{\partial e_{b,\lambda}}{\partial T} d\lambda} = \int_0^\infty \frac{1}{K_{e,\lambda}} \frac{\partial e_{b,\lambda}}{\partial e_b} d\lambda \quad \text{(S5)}$$

$$\frac{\partial e_{b,\lambda}}{\partial e_b} = \frac{\pi}{2} \frac{C_1 C_2}{\lambda^6} \frac{\sigma^{1/4}}{e_b^{5/4}} \frac{\exp(C_2/\lambda T)}{[\exp(C_2/\lambda T)-1]^2} \quad \text{(S6)}$$

$$e_b = \sigma T^4 \quad \text{(S7)}$$

where C_1 and C_2 are constant, 5.96×10^7 W-μm^4/m^2 and 1.44×10^4 μm-K, respectively. The σ is Stefan-Boltzmann's constant, 5.67×10^{-8} W/m^2-K^4 and $e_{b,\lambda}$ is the local spectral energy.

Based on the Rosseland extinction coefficient, the k_{rad} through the polymeric foams can be expressed as follows:[10, 11]

$$k_{rad} = \frac{16 n_{ref}^2 \sigma T^3}{3 K_{e,R}} \quad \text{(S8)}$$

where n_{ref} is the effective index of refraction. Due to the high void fraction in PLA foams, the n_{ref} in this study is 1, which has the same value as air.

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