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Electronic Supplementary Information (ESI)

Effects of Amino Acid Sequence on Thermal Conduction through β-Sheet Crystals of Natural Silk Protein

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S1. Structure: The atomistic structures of the β-sheets were build based on previous work simulating the mechanical response of β-sheets [1]. Our models were verified by visualization with the Visual Molecular Dynamics (VMD) package [2]. A snapshot from VMD [Figure S1(a)] shows that hydrogen bonds (red lines) are spontaneously formed in the equilibrated β-sheet structure.

To demonstrate the stability of hydrogen bonds in our models, Figure S1(b) plots the *percentage of hydrogen bond formation* versus simulation time. The percentage of hydrogen bond formation was calculated by taking a ratio between the number of hydrogen bonds at any time instant and the number of hydrogen bonds that could be formed in the ideal condition. Results show that the percentage of hydrogen bond formation remains ~70% during the equilibration and velocity swapping procedures. VMD snapshots in Figure S1(c) show that the structure does not change much in velocity swapping.

The insensitivity of protein structure to velocity swapping is due to the very careful procedure we followed when performing the simulations. The frequency of temperature swap was fine-tuned for all jobs such that the temperature difference between the two ends of the protein is about 30±2 K. The temperature difference is small enough to avoid protein denaturation at high temperatures [3]. It is also large enough to overcome thermal perturbation such that smooth temperature profiles could be obtained.

S2. Identification of Hydrogen Bonds

Molecular dynamics simulation does not require the identification of hydrogen bonds. However, we need to identify hydrogen bonds in the visualization and analysis processes, e.g. Figure S1(a,b). Although there are several different geometric definitions of hydrogen bonds available [4,5], we employed the default criteria in VMD [6]:

"A hydrogen bond is formed between an atom with a hydrogen bonded to it (the donor, D) and another atom (the acceptor, A) provided that the distance D-A is less than the cut-off distance (default 3.0 Angstroms) and the angle D-H-A is less than the cut-off angle (default 20 degrees)."

We note that, using different criteria could give slightly different numbers of hydrogen bonds. However, this would not affect any simulation result because molecular dynamics simulation does not involve the identification of hydrogen bonds. This would not significantly affect visualization and analysis [e.g. Figure S1(a,b)] either because the physics is not changed.

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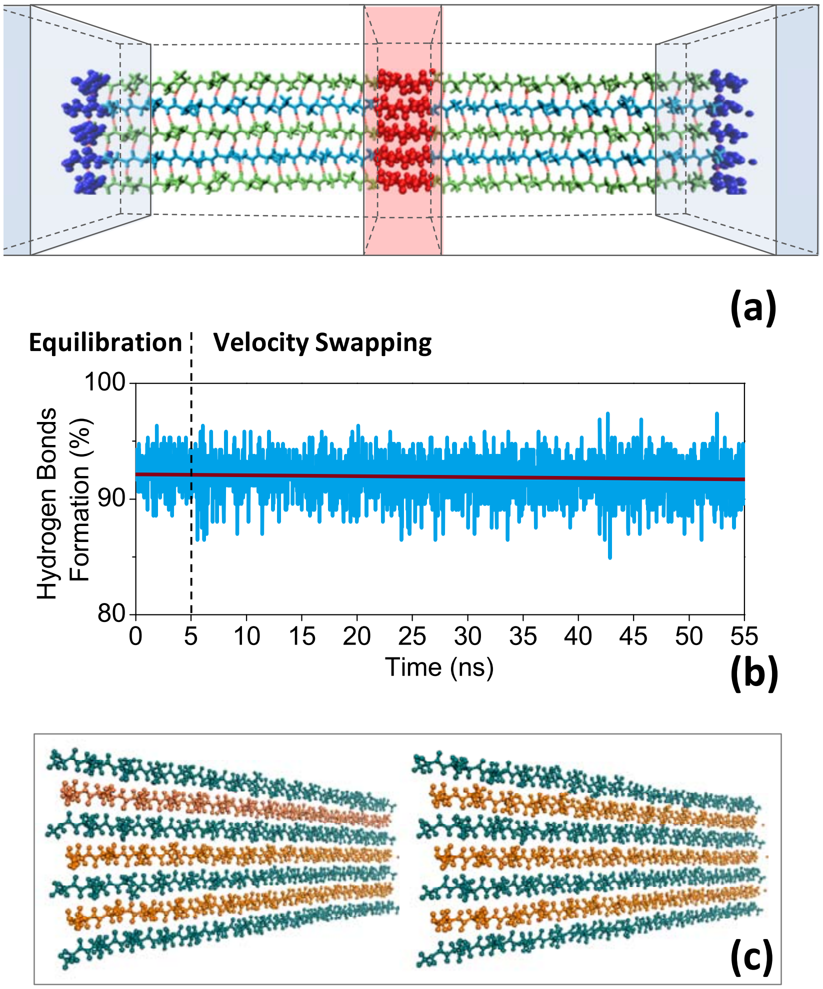

Figure S1. (a) A snapshot of the simulation model from VMD. (b) Percentage of hydrogen bond formation versus simulation time. (c) Structural comparison for a β-sheet before and after velocity swapping.

S3. Calculation of Thermal Conductivity

To calculate thermal conductivity, we adopted the reverse non-equilibrium molecular dynamics method developed by Muller-Plathe [7,8] which generates a heat flux by a velocity swapping algorithm. Each non-equilibrium molecular dynamics simulation led to a temperature profile similar to that shown in Figure S2. The temperature profile is nonlinear due to the perturbation caused by the artificial velocity swapping. An average was taken between the temperature profiles to the left and right of the heat source. Line regression of the linear portion of the temperature profile was then calculated to obtain the temperature gradient. On the other hand, the induced heat flux was evaluated by $J = \Delta E / 2tA$, where t denotes the time interval between two swaps, ΔE is the averaged kinetic energy transferred per swap, A is the cross-sectional area of the material, and the coefficient “2” reflects the two symmetric heat transport paths from the hot slab to the cold slab. The value of A was calculated based on the chainaveraged cross-sectional area in the multilayer nanocrystals. Finally by Fourier's law, thermal conductivity was calculated by $\kappa = -J / \frac{dT}{dx}$, where $\frac{dT}{dx}$ is the temperature gradient along the *x*-direction.

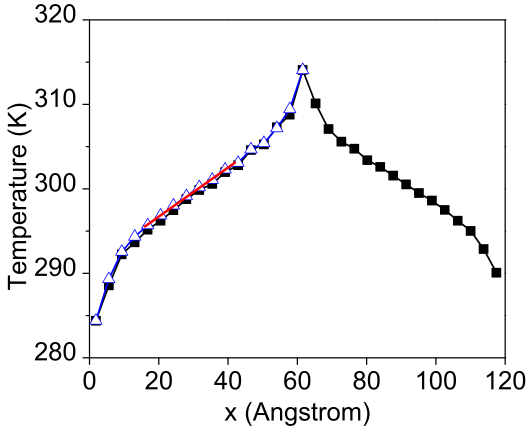

Figure S2. Temperature profile along the length of a β-sheet

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