Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Effect of Na-ion intercalation on the thermal conductivity of carbon honeycomb nanostructure

Supplementary Information

Table S1. Charges of the Na ions at different ion concentrations based on the DFT-D2 model (unit: e)

Concentration *x* / **Species**	0.11	0.33	0.50	0.67	0.83	1.0
Na ions	0.873	0.612	0.515	0.452	0.420	0.360
C atoms	0.031	0.066	0.083	0.097	0.113	0.116

Table S2. Charges of the Na ions at different ion concentrations based on the DFT-D3 model (unit: e)

Concentration *x* / **Species**	0.11	0.33	0.50	0.67	0.83	1.0
Na ions	0.895	0.610	0.422	0.379	0.332	0.295
C atoms	0.032	0.065	0.068	0.081	0.089	0.095

Table S3. Elastic constants C44, C55, C66 at different ion concentrations

Concentration *x* / **Elastic constants**	0.11	0.33	0.50	0.67	0.83	1.0
C44	105.1	102.8	102.4	104.3	110.8	104.6
C55	101.7	102.5	101.9	105.1	96.4	104.6
C66	9.7	8.1	7.9	8.2	6.8	7.4

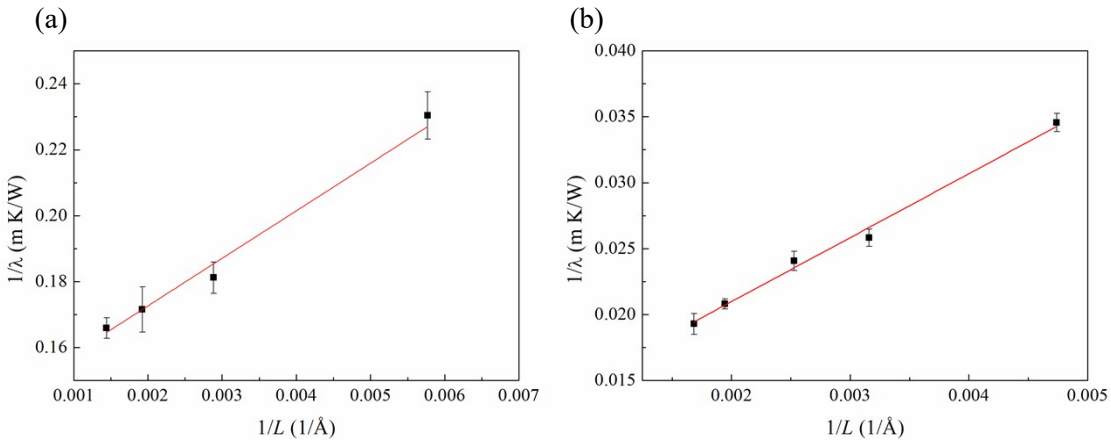

Fig. S1. Relationship between 1/λ and 1/L for the Na ion intercalated CHC with an ion concentration of x=0.67 along the (a) armchair and (b) axial direction.

Theoretically, the reciprocal of the thermal conductivity ($1/\lambda$) of the CHC nanostructure is related to the reciprocal of its length ($1/L$) via a linear relationship $1/\lambda = A + B/L$. Hence, the thermal conductivity of an infinitely long system can be obtained by extrapolating this linear relationship to $L \to \infty$. To address the size effect of the anisotropic ratio of the thermal conductivity, additional simulations are conducted for the ion-intercalated CHC with the concentration of x=0.67 at different lengths. Since the values of thermal conductivity in the armchair and zigzag directions are close to each other, the additional simulations are conducted only for the armchair and axial directions. The linear fittings of the simulation results presented in Fig. S1 show that the thermal conductivity of the Na-ion intercalated CHC in the armchair and axial direction increases to 7.05 and 88.65 W/mK at the infinite length, respectively, which corresponds to an increase in the anisotropic ratio from ~7 to ~11.

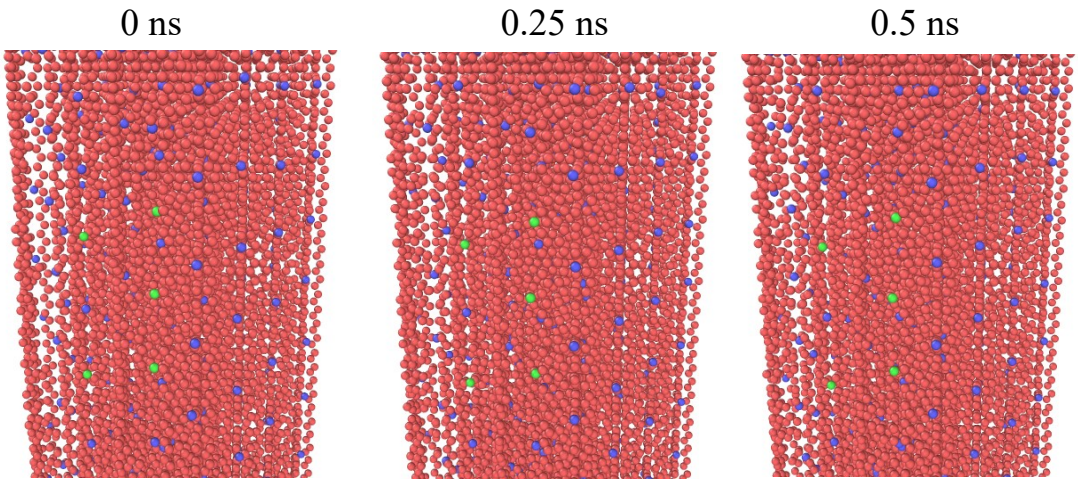

Fig. S2. Snapshots of the simulation system at the time of 0, 0.25, and 0.5 ns after a steady temperature gradient is established along the heat flux direction. The red dots represent the C atoms while the blue and green dots represent the Na ions. The Na ions colored in green are selected as markers to show their slow diffusion.

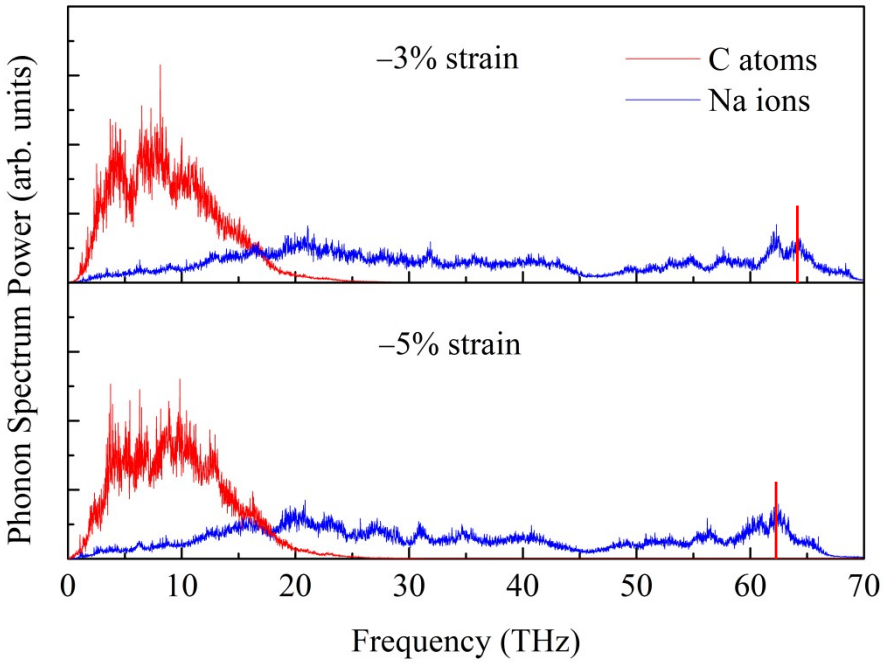

Fig. S3. Comparison of the projected phonon spectra of the C atoms and Na ions at the compressive strain of -3% and -5%. The red dashed lines indicate the positions of the spectra peaks and show their red-shift induced by the strain.