Supporting Information

Exploring the Structure-Property Relations of Three-Dimensional Hexagonal Boron Nitride Aerogels with Gyroid Surface

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Note 1. Mathematical formulation

The dimensionless Swift–Hohenberg (SH) free energy functional was adopted to model the atomic structures of hBNAGs,[1]

$$F = \int dr\left[-\frac{1}{2}\varepsilon_B n_B^2 + \frac{1}{2}n_B(1+\nabla^2)^2 n_B - \frac{1}{3}g_B n_B^3 + \frac{1}{4}n_B^4 - \frac{1}{2}\varepsilon_N n_N^2 + \frac{1}{2}n_N(1+\nabla^2)^2 n_N - \frac{1}{3}g \right. \quad \text{(S-1)}$$

To model the evolution process of the density field $n_B(r,t)$ and $n_N(r,t)$, dimensionless dynamics equations can be deviated according to overdamped conservative dynamics as,

$$\frac{\partial n_B}{\partial t} = \nabla^2\frac{\delta F}{\delta n_B} = \nabla^2\Big[-\varepsilon_B n_B + (1+\nabla^2)^2 n_B - g_B n_B^2 + n_B^3 + \alpha_{BN} n_N + \beta_{BN}(1+\nabla^2)^2 n_N + wn \quad \text{(S-2)}$$

$$\frac{\partial n_N}{\partial t} = \nabla^2 \frac{\delta F}{\delta n_N} = \nabla^2 \Big[-\varepsilon_N n_N + \big(1+\nabla^2\big)^2 n_N - g_N n_N^2 + n_N^3 + \alpha_{BN} n_B + \beta_{BN} \big(1+\nabla^2\big)^2 n_B + \frac{1}{2} w$$

(S-3)

Four new variables ($\vartheta_B, \mu_B, \vartheta_N, \mu_N$) are introduced to reduce the sixth order of spatial derivatives of Eq. S2 and S3 to a set of second order partial differential equations as,

$$\frac{\partial n_B}{\partial t} = \nabla^2 \mu_B \quad \text{(S-4)}$$

$$\frac{\partial n_N}{\partial t} = \nabla^2 \mu_N \quad \text{(S-5)}$$

$$\vartheta_B = \nabla^2 n_B \quad \text{(S-6)}$$

$$\vartheta_N = \nabla^2 n_N \quad \text{(S-7)}$$

$$\mu_B = \big(1-\varepsilon_B\big) n_B + 2\vartheta_B + \nabla^2 \vartheta_B - g_B n_B^2 + n_B^3 + \big(\beta_{BN} + \alpha_{BN}\big) n_N + 2\beta_{BN} \vartheta_N + \beta_{BN} \nabla^2 \vartheta_N + w n_B n_N +$$

(S-8)

$$\mu_N = \big(1-\varepsilon_N\big) n_N + 2\vartheta_N + \nabla^2 \vartheta_N - g_N n_N^2 + n_N^3 + \big(\beta_{BN} + \alpha_{BN}\big) n_B + 2\beta_{BN} \vartheta_B + \beta_{BN} \nabla^2 \vartheta_B + \frac{1}{2} w n_B^2 +$$

(S-9)

Due to the periodic boundary condition of the function domain Ω, the surface (edge) integral terms will vanish. Therefore, the variational problem of the PFC modeling associated with the Eq. S4-S9 can be stated as: *find* ($n_B, \vartheta_B, \mu_B, n_N, \vartheta_N, \mu_N$) $\in \Omega$ *such that for all test functions* ($p_B, l_B, m_B, p_N, l_N, m_N$) $\in \Omega$, *the following equations are satisfied,*

$$\int_\Omega \frac{\partial n_B}{\partial t} p_B dr + \int_\Omega \nabla \mu_B \cdot \nabla p_B dr = 0 \quad \text{(S-10)}$$

$$\int_\Omega \frac{\partial n_N}{\partial t} p_N dr + \int_\Omega \nabla \mu_N \cdot \nabla p_N dr = 0 \quad \text{(S-11)}$$

$$\int_\Omega \vartheta_B l_B dr + \int_\Omega \nabla n_B \cdot \nabla l_B dr = 0 \quad \text{(S-12)}$$

$$\int_{\Omega} \vartheta_N l_N dr + \int_{\Omega} \nabla n_N \cdot \nabla l_N dr = 0 \tag{S-13}$$

$$\int_{\Omega} \mu_B m_B dr - ב_B + \int_{\Omega} \nabla \vartheta_B \cdot \nabla m_B dr + \beta_{BN} \int_{\Omega} \nabla \vartheta_N \cdot \nabla m_B dr = 0 \tag{S-14}$$

$$\int_{\Omega} \mu_N m_N dr - ב_N + \int_{\Omega} \nabla \vartheta_N \cdot \nabla m_N dr + \beta_{BN} \int_{\Omega} \nabla \vartheta_B \cdot \nabla m_N dr = 0 \tag{S-15}$$

where,

$$ב_B = \int_{\Omega} \Big[(1 - \varepsilon_B) n_B + 2\vartheta_B - g_B n_B^2 + n_B^3 + (\beta_{BN} + \alpha_{BN}) n_N + 2\beta_{BN} \vartheta_N + + w n_B n_N + \frac{1}{2} u n \tag{S-16}$$

$$ב_N = \int_{\Omega} \Big[(1 - \varepsilon_N) n_N + 2\vartheta_N - g_N n_N^2 + v n_N^3 + (\beta_{BN} + \alpha_{BN}) n_B + 2\beta_{BN} \vartheta_B + \frac{1}{2} w n_B^2 + u n_B n \tag{S-17}$$

For time integration, Eq. S10 - S17 are discretized as,

$$\int_{\Omega} \frac{n_B^{n+1} - n_B^n}{\Delta t} p_B dr + \int_{\Omega} \nabla \frac{\mu_B^{n+1} + \mu_B^n}{2} \cdot \nabla p_B dr = 0 \tag{S-18}$$

$$\int_{\Omega} \frac{n_N^{n+1} - n_N^n}{\Delta t} p_N dr + \int_{\Omega} \nabla \frac{\mu_N^{n+1} + \mu_N^n}{2} \cdot \nabla p_N dr = 0 \tag{S-19}$$

$$\int_{\Omega} \vartheta_B^{n+1} l_B dr + \int_{\Omega} \nabla n_B^{n+1} \cdot \nabla l_B dr = 0 \tag{S-20}$$

$$\int_{\Omega} \vartheta_N^{n+1} l_N dr + \int_{\Omega} \nabla n_N^{n+1} \cdot \nabla l_N dr = 0 \tag{S-21}$$

$$\int_{\Omega} \mu_B^{n+1} m_B dr - ב_B^{n+1} + \int_{\Omega} \nabla \vartheta_B^{n+1} \cdot \nabla m_B dr + \beta_{BN} \int_{\Omega} \nabla \vartheta_N^{n+1} \cdot \nabla m_B dr = 0 \tag{S-22}$$

$$\int_{\Omega} \mu_N^{n+1} m_N dr - ב_N^{n+1} + \int_{\Omega} \nabla \vartheta_N^{n+1} \cdot \nabla m_N dr + \beta_{BN} \int_{\Omega} \nabla \vartheta_B^{n+1} \cdot \nabla m_N dr = 0 \tag{S-23}$$

and,

$$\text{ב}_B^{n+1} = \int_\Omega \Big[(1-\varepsilon_B) n_B^{n+1} + 2\vartheta_B^{n+1} - g_B\left(n_B^{n+1}\right)^2 + \left(n_B^{n+1}\right)^3 + (\beta_{BN} + \alpha_{BN}) n_N^{n+1} + 2\beta_{BN}$$

(S-24)

$$\text{ב}_N^{n+1} = \int_\Omega \Big[(1-\varepsilon_N) n_N^{n+1} + 2\vartheta_N^{n+1} - g_N\left(n_N^{n+1}\right)^2 + \left(n_N^{n+1}\right)^3 + (\beta_{BN} + \alpha_{BN}) n_B^{n+1} + 2\beta_{BN}$$

(S-25)

where $\Delta t = t^{n+1} - t^n$, $()^n$ and $()^{n+1}$ represent the values of variables (n_B, ϑ_B, μ_B, n_N, ϑ_N, μ_N) at time t^n and t^{n+1}, respectively. The above discretized variational formulations (Eq. S18 - S25) can be paraphrased through the Unified Form Language (UFL) implemented within the FEniCS platform directly.[2]

Note 2. Modification and validation of the interatomic potential

1. Mechanical properties

In this study, the interatomic interactions in hBNAGs are described by the extended Tersoff potential for boron nitride (BN-ExTeP),[3] which was recently developed to accurately reproduce the basic material properties of pristine and defective *h*-BN structures. Note that, unphysical stiffening behavior was observed with the original Tersoff potential under high deformation (see the dot lines in Figure S1).[4] After testing different values of the cutoff parameters, we choose R = 1.75 and D = 0.0 to eliminate unphysical behavior and obtain accurate behavior of fracture properties (see solid lines in Figure S1). The tensile strength of

pristine *h*-BN in the armchair and zigzag direction are obtained as 90.6 GPa and 102.8 GPa, respectively, close to the DFT results of 88 GPa and 102 GPa, respectively.[5] The Young's Modulus is about 800.0 GPa, consistent with DFT and experimental results.[5, 6] Therefore, the modified BN-ExTeP can give consistent fracture behaviors, retaining an accurate description of basic equilibrium and elastic properties of pristine and defective *h*-BN structures. In this study, the reference mass density, tensile strength and Young's Modulus of pristine *h*-BN are set as $\rho_R = 2.24\ g/cm^3$, $\sigma_R =$ 96.7 GPa and $E_R =$ 800.0 GPa, respectively.

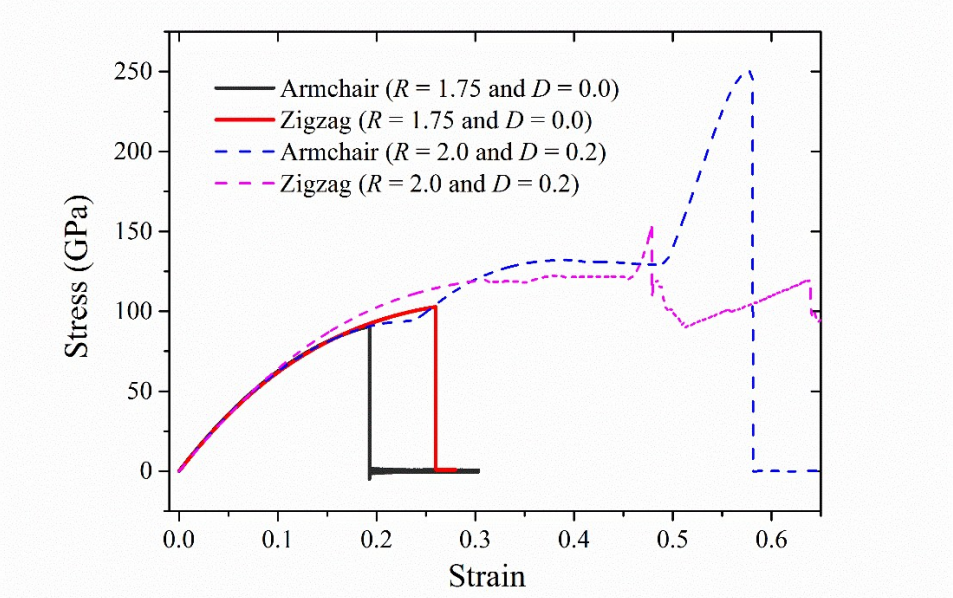

Figure S1 The tensile stress-strain curves for pristine *h*-BN under armchair and zigzag deformation using the Tersoff potential with modified parameters: R = 1.75 and D = 0.0 (The original value: R = 2.0 and D = 0.2).

2. Thermal properties

To verify the effectiveness and accuracy of the BN-ExTeP potential for thermal conductivity calculation, we have calculated the thermal conductivity of *h*-BN using this potential. We

construct a series of *h*-BN models with sample length ranging from 60 to 200 nm. The thermal conductivity shows a strong size-dependence. The calculated thermal conductivities of *h*-BN are 400.2 $Wm^{-1}K^{-1}$ and 482.1 $Wm^{-1}K^{-1}$ for the length of 60 nm and 200 nm, as shown in Figure S2(a). Besides, the relationship between the inverse thermal conductivity and the inverse sample length is shown in Fig. S2(b), thus the thermal conductivity of an infinite sample length κ_∞ is calculated to be 529.1 $Wm^{-1}K^{-1}$, in reasonable agreement with the experimental values where the thermal conductivity is found to be around 751 $Wm^{-1}K^{-1}$ for monolayer h-BN and around 400 $Wm^{-1}K^{-1}$ for bulk h-BN at room temperature.[7]

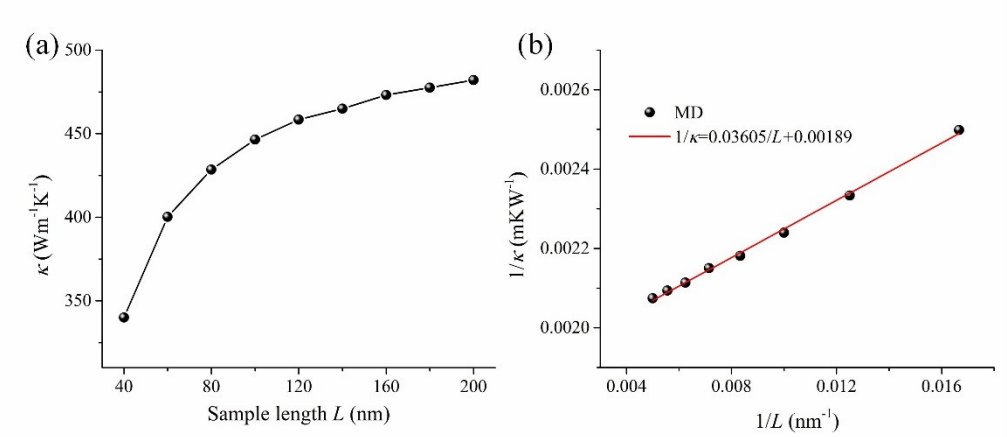

Figure S2 Variation of the thermal conductivity of *h*-BN with sample length and (b) the relationships between the inverse thermal and the inverse sample length.

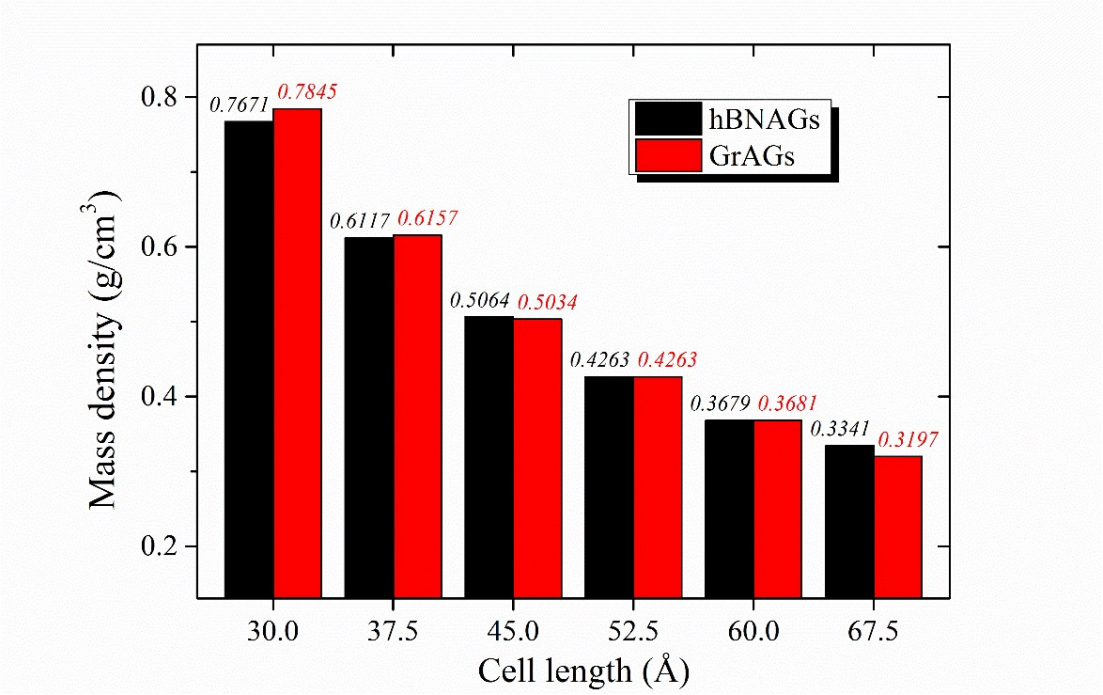

Figure S3 The mass density of hBNAGs and GrAGs with different cell lengths. It can be seen clearly that the density decreases with the increase of cell length due to the increase of space.

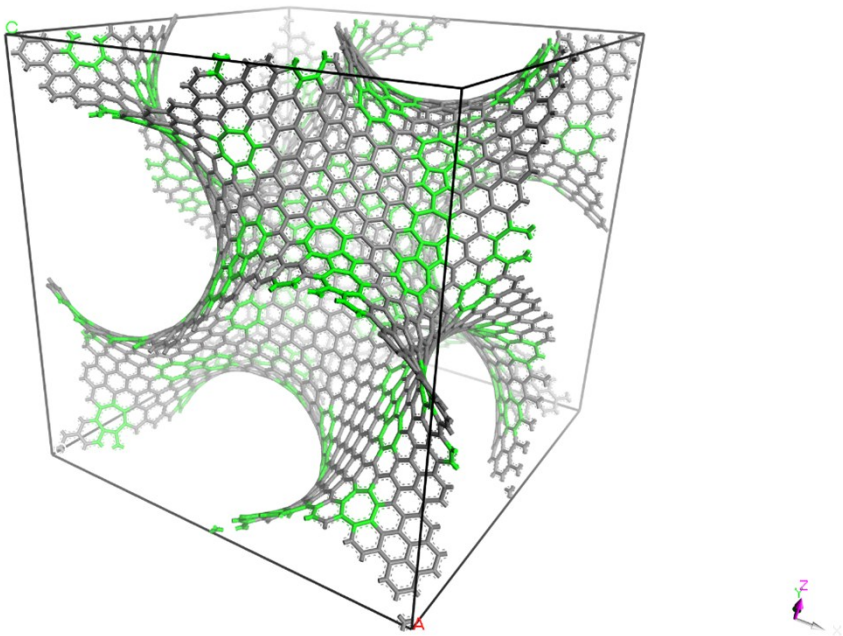

Figure S4 The spatial distribution of topological defects for graphene aerogel will cell length of 45 Å.

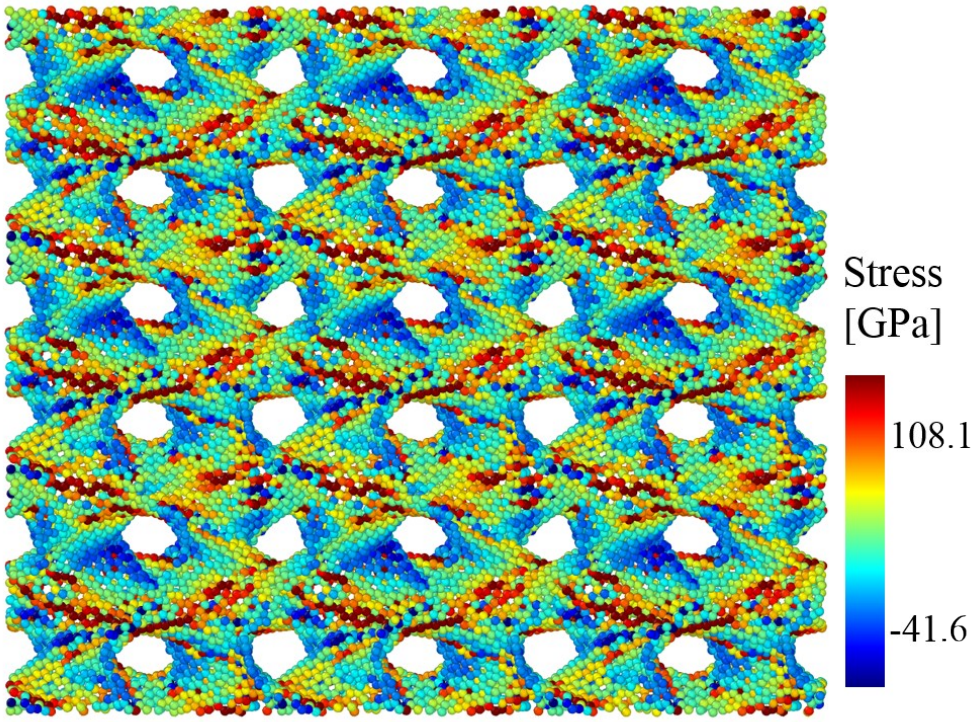

Figure S5 Atomic configuration under tensile strain ε_x=0.154 colored with atomic stress in the *x* direction.

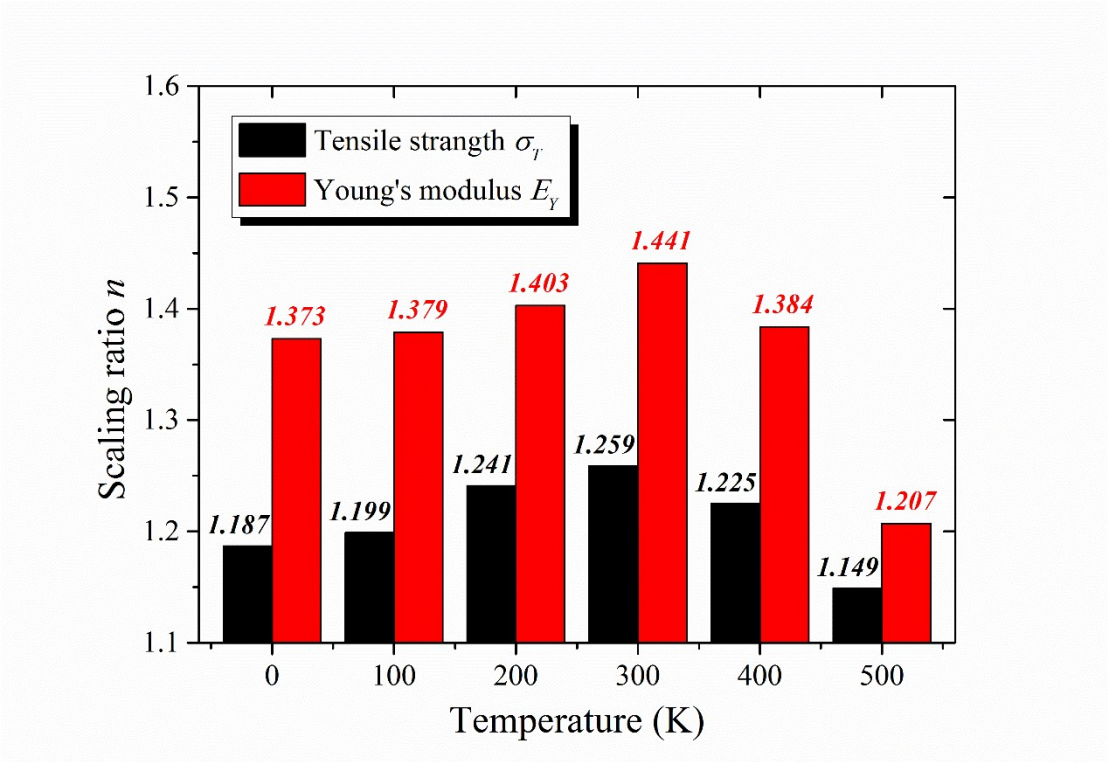

Figure S6 Scaling ratios n for tensile strength σ_T and Young's modulus E_Y of nanoporous hBNAGs at different temperatures.

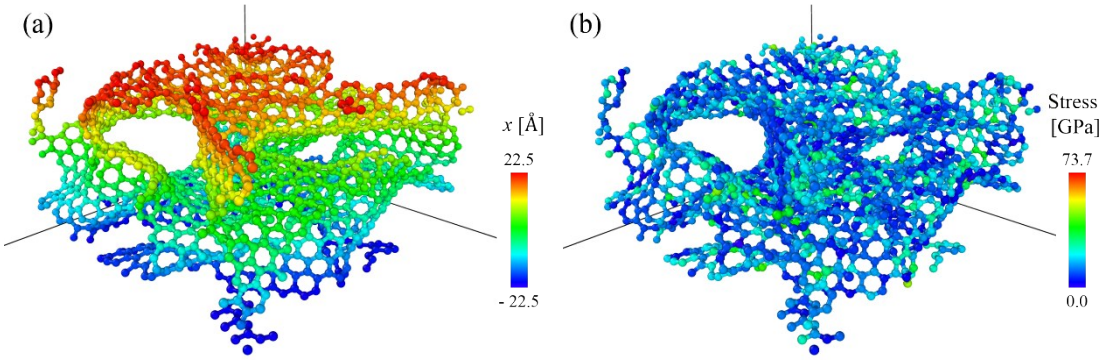

Figure S7 Atomic configuration under compressive strain ε_x=-0.5 colored with relative coordinate in the x direction (a) and atomic Von Mises stress (b).

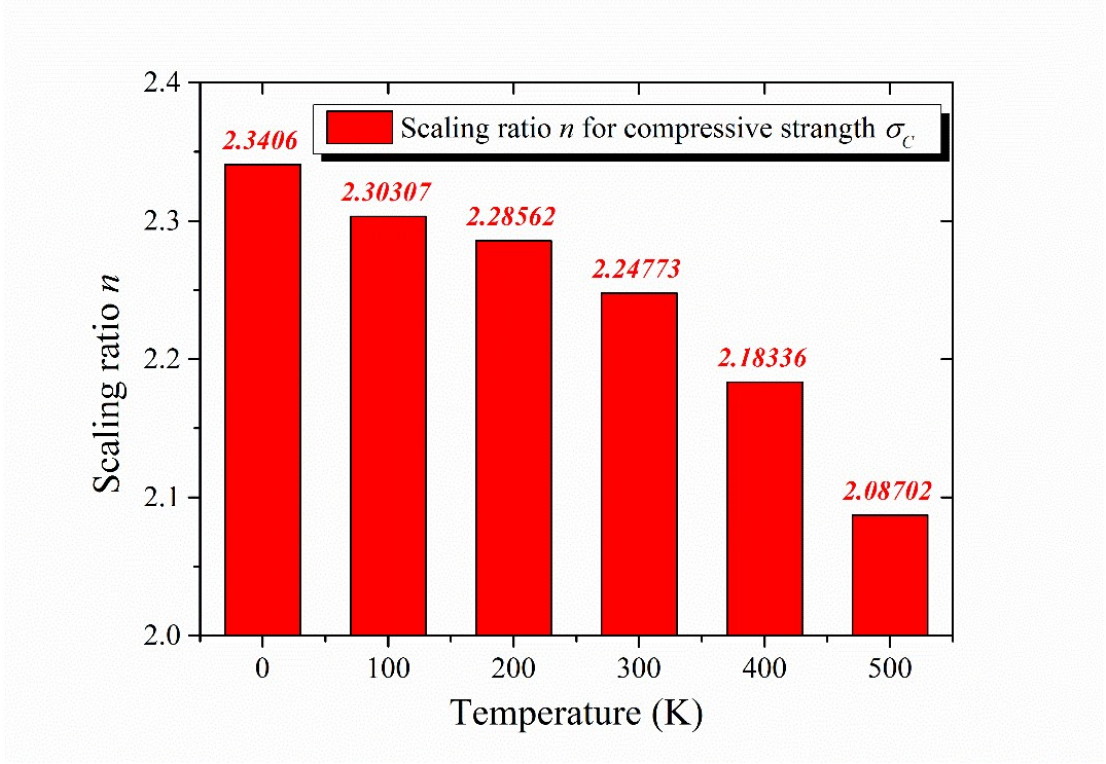

Figure S8 Scaling ratios n for compressive strength σ_C of nanoporous hBNAGs at different temperatures.

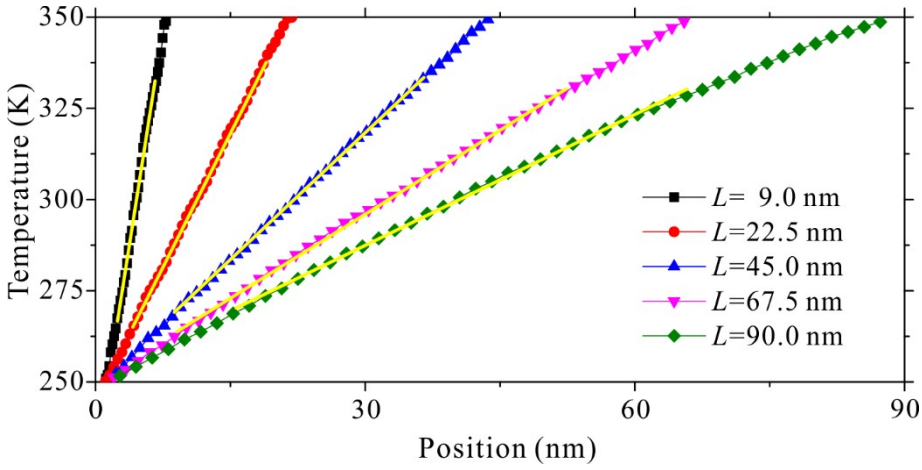

Figure S9 Temperature distribution of hBNAGs with cell length 45.0 Å with various lengths.

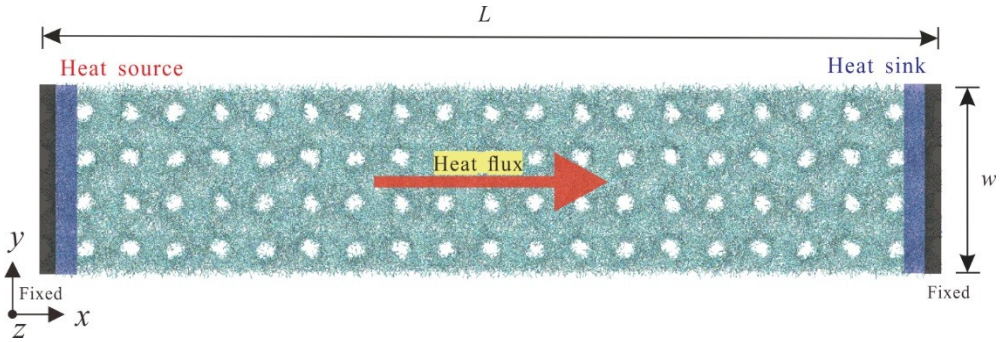

Figure S10 Atomic heat flux vectors of hBNAGs with cell length 45.0 Å, L=45 nm, w=9 nm, and h=9 nm.

Table S1 The scaling ratios $\boldsymbol{n}$ of tensile strength σ_T, compressive strength σ_C and Young's modulus E_Y for nanoporous hBNAGs

Temperature (K)		0.0	100.0	200.0	300.0	400.0	500.0
Scaling ratio $\boldsymbol{n}$	for σ_T	1.1867	1.1996	1.2411	1.2587	1.2252	1.1496
	for σ_C	2.3406	2.3031	2.2856	2.2477	2.1834	2.0870
	for E_Y	1.3734	1.3794	1.4034	1.4412	1.3835	1.2065

Table S2 Detailed NEMD configurations for all simulation models

l (nm)	3	3.75	4.5	5.25	6	6.75
L (nm)	9,15,30, 60,90	11.25,18.75,37.5, 75,112.5	9,22.5,45, 67.5,90	10.5,26.25,52.5, 78.75,105	12,30,60, 72,90	13.5,33.75,67.5, 81,101.25
w (nm)	6.0	7.5	9.0	10.5	12	13.5
h (nm)	6.0	7.5	9.0	10.5	12	13.5

The modified BN-ExTeP potential file

 # Original file: CONTRIBUTOR: J.H. Los, J.M.H. Kroes CITATION: Los et al. Phys. Rev. B 96, 184108 (2017)

 # The modified ExTeP potential file for exploring mechanical and thermal properties of the h-BN system

 # we choose R = 1.75 and D = 0.0 to eliminate unphysical behavior and obtain accurate behavior of fracture properties

 # ExTeP parameters for various elements and mixtures

 # multiple entries can be added to this file, LAMMPS reads the ones it needs

 # these entries are in LAMMPS "metal" units:

 # A,B = eV; lambda1,lambda2,lambda3 = 1/Angstroms; R,D = Angstroms

 # other quantities are unitless

 # format of a single entry (one or more lines):

#I J K m, gamma*, lambda3, c, d, h, n, gamma, lambda2, B, R, D, lambda1, A

B B B 3 1.0 0.0 26617.3000 141.2000 -0.1300 1.1422470 0.01498959 2.5211820 2768.7363631 1.75 0.0 2.6857244 3376.3350735

N N N 3 1.0 0.0 23.5000 3.7500 -0.4000 0.6650000 0.01925100 2.6272721 2563.5603417 1.75 0.0 2.8293093 2978.9527928

B B N 3 1.0 0.0 26617.3000 141.2000 -0.1300 1.1422470 0.01498959 2.5211820 2768.7363631 1.75 0.0 2.6857244 3376.3350735

N N B 3 1.0 0.0 23.5000 3.7500 -0.4000 0.6650000 0.01925100 2.6272721 2563.5603417 1.75 0.0 2.8293093 2978.9527928

B N B 3 1.0 0.0d0 306.586555205d0 10.d0 -0.7218d0 0.6576543657d0 0.0027024851d0 2.69335d0 2595.6860833266d0 1.75d0 0.0d0 2.95d0 3330.0655849887d0

B N N 3 1.0 0.0d0 306.586555205d0 10.d0 -0.7218d0 0.6576543657d0 0.0027024851d0 2.69335d0 2595.6860833266d0 1.75d0 0.0d0 2.95d0 3330.0655849887d0

N B B 3 1.0 0.0d0 306.586555205d0 10.d0 -0.7218d0 0.6576543657d0 0.0027024851d0 2.69335d0 2595.6860833266d0 1.75d0 0.0d0 2.95d0 3330.0655849887d0

N B N 3 1.0 0.0d0 306.586555205d0 10.d0 -0.7218d0 0.6576543657d0 0.0027024851d0 2.69335d0 2595.6860833266d0 1.75d0 0.0d0 2.95d0 3330.0655849887d0

#

1.9925 Bicubic Splines Parameters

#

F_corr [B, B]

#

#t1	t2	i	j	val	dx	dy	dxy
B	B	0	0	0.0000	0.0000	0.0000	0.0000
B	B	0	1	0.0054	0.0000	0.0000	0.0000
B	B	0	2	0.0182	0.0000	0.0000	0.0000
B	B	0	3	-0.0034	0.0000	0.0000	0.0000
B	B	0	4	-0.0034	0.0000	0.0000	0.0000
B	B	1	0	0.0054	0.0000	0.0000	0.0000
B	B	1	1	0.0100	0.0000	0.0000	0.0000
B	B	1	2	0.0062	0.0000	0.0000	0.0000
B	B	1	3	0.0154	0.0000	0.0000	0.0000
B	B	1	4	0.0154	0.0000	0.0000	0.0000
B	B	2	0	0.0182	0.0000	0.0000	0.0000
B	B	2	1	0.0062	0.0000	0.0000	0.0000
B	B	2	2	0.0154	0.0000	0.0000	0.0000
B	B	2	3	-0.0390	0.0000	-0.0727	0.0000
B	B	2	4	-0.0390	0.0000	-0.0727	0.0000
B	B	3	0	-0.0034	0.0000	0.0000	0.0000
B	B	3	1	0.0154	0.0000	0.0000	0.0000
B	B	3	2	-0.0390	-0.0727	0.0000	0.0000
B	B	3	3	-0.1300	0.0000	0.0000	0.0000
B	B	3	4	-0.1300	0.0000	0.0000	0.0000
B	B	4	0	-0.0034	0.0000	0.0000	0.0000
B	B	4	1	0.0154	0.0000	0.0000	0.0000
B	B	4	2	-0.0390	-0.0727	0.0000	0.0000
B	B	4	3	-0.1300	0.0000	0.0000	0.0000
B	B	4	4	-0.1300	0.0000	0.0000	0.0000

#

F_corr [B, N]

#

#t1	t2	i	j	val	dx	dy	dxy
B	N	0	0	0.0170	0.0000	0.0000	0.0000
B	N	0	1	0.0078	0.0000	0.0000	0.0000
B	N	0	2	0.0000	0.0000	0.0000	0.0000
B	N	0	3	-0.0860	0.0000	0.0000	0.0000
B	N	0	4	-0.0860	0.0000	0.0000	0.0000

```
  B    N      1     0    -0.0090      0.0000      0.0000      0.0000
  B    N      1     1     0.0090      0.0000      0.0000      0.0000
  B    N      1     2    -0.0068      0.0000     -0.0214      0.0000
  B    N      1     3    -0.0338      0.0000      0.0388      0.0000
  B    N      1     4    -0.0338      0.0000      0.0388      0.0000
  B    N      2     0     0.0000      0.0000      0.0000      0.0000
  B    N      2     1    -0.0198      0.0000      0.0000      0.0000
  B    N      2     2     0.0000      0.0000      0.0000      0.0000
  B    N      2     3    -0.0084      0.0000      0.0169      0.0000
  B    N      2     4    -0.0084      0.0000      0.0169      0.0000
  B    N      3     0    -0.0750      0.0000      0.0000      0.0000
  B    N      3     1    -0.0168      0.0306      0.0000      0.0000
  B    N      3     2    -0.0138      0.0084      0.0000      0.0000
  B    N      3     3     0.0000      0.0000      0.0000      0.0000
  B    N      3     4     0.0000      0.0000      0.0000      0.0000
  B    N      4     0    -0.0750      0.0000      0.0000      0.0000
  B    N      4     1    -0.0168      0.0306      0.0000      0.0000
  B    N      4     2    -0.0138      0.0084      0.0000      0.0000
  B    N      4     3     0.0000      0.0000      0.0000      0.0000
  B    N      4     4     0.0000      0.0000      0.0000      0.0000
#
#    F_corr    [ N,   N]
#
#t1 t2   i     j            val           dx           dy           dxy
  N    N      0     0     0.0000      0.0000      0.0000      0.0000
  N    N      0     1    -0.0282      0.0000      0.0000      0.0000
  N    N      0     2    -0.0018      0.0000      0.0000      0.0000
  N    N      0     3    -0.0004      0.0000      0.0000      0.0000
  N    N      0     4    -0.0004      0.0000      0.0000      0.0000
  N    N      1     0    -0.0282      0.0000      0.0000      0.0000
  N    N      1     1     0.0200      0.0000      0.0000      0.0000
  N    N      1     2     0.0180      0.0162     -0.0027      0.0000
  N    N      1     3     0.0146      0.0000      0.0000      0.0000
  N    N      1     4     0.0146      0.0000      0.0000      0.0000
  N    N      2     0    -0.0018      0.0000      0.0000      0.0000
  N    N      2     1     0.0180     -0.0027      0.0162      0.0000
  N    N      2     2     0.0306      0.0000      0.0000      0.0000
  N    N      2     3     0.0060      0.0000     -0.0073      0.0000
  N    N      2     4     0.0060      0.0000     -0.0073      0.0000
  N    N      3     0    -0.0004      0.0000      0.0000      0.0000
  N    N      3     1     0.0146      0.0000      0.0000      0.0000
  N    N      3     2     0.0060     -0.0073      0.0000      0.0000
  N    N      3     3     0.0000      0.0000      0.0000      0.0000
  N    N      3     4     0.0000      0.0000      0.0000      0.0000
```

N	**N**	**4**	**0**	**-0.0004**	**0.0000**	**0.0000**	**0.0000**
N	**N**	**4**	**1**	**0.0146**	**0.0000**	**0.0000**	**0.0000**
N	**N**	**4**	**2**	**0.0060**	**-0.0073**	**0.0000**	**0.0000**
N	**N**	**4**	**3**	**0.0000**	**0.0000**	**0.0000**	**0.0000**
N	**N**	**4**	**4**	**0.0000**	**0.0000**	**0.0000**	**0.0000**

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