Supporting Information

Effects of the Filler Property, Electron-Phonon Coupling on Thermal Conductivity and Strain Rate on the Mechanical Property of Polyethylene Nanocomposites

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1. Physical model of PE nanocomposites

The main view and the lateral view of equilibrated Cu-PE are shown in Figure S1. The size of Cu-PE after NPT equilibration is 3.25 nm×4.2 nm×9.8 nm. The number of atoms in Cu-PE is 13000, which is large enough to overcome size effects.
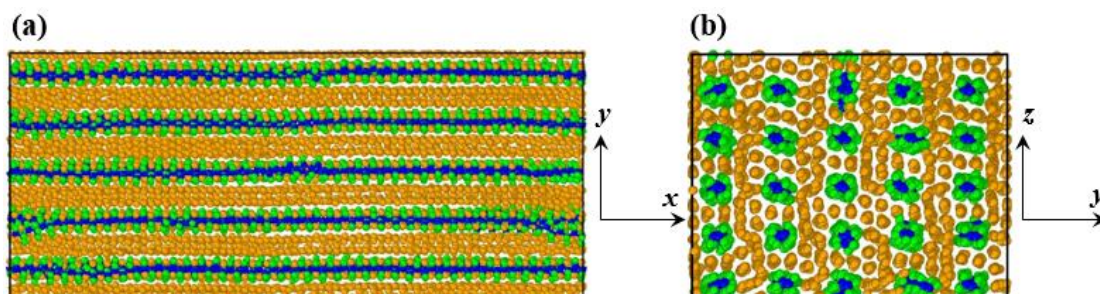

Figure S1. (a) The main view of Cu-PE. (b) The lateral view of Cu-PE.

The schematic diagram of building the initial structure of Cu-PE system is shown in Figure S2. The bigger initial structure of Cu-PE and other similar initial structures are built in the same way. Firstly, a metal Lattice is created and replicated 2 times along the y and z direction, respectively. Secondly, metal atoms inside the lattice are removed to form a hole, whose size is around 3.615×3.615×3.615 $Å^3$. Thirdly, an ethylene unit is inserted into the hole to form a unit. Then the unit is replicated 5 times along the y and z direction to form a bigger unit. After that, the bigger unit is replicated along the x direction to produce the initial structure. The bigger Cu-PE initial structure is produced by replicating the unit 300 times along the x direction.
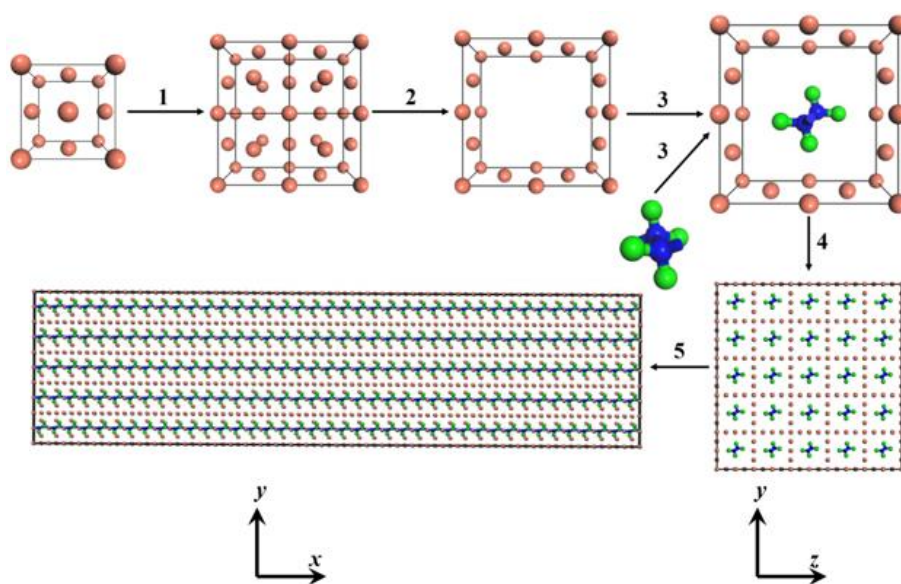

Figure S2. Schematic diagram of Cu-PE initial structure.

The initial Cu-PE system firstly experiences energy minimization, then equilibrates in the isothermal-isobaric (NPT) ensemble for 10 ns to reach the equilibrium state. The potential energy, kinetic energy, volume, and density evolution are monitored in the equilibrium process and the results are depicted in Figure S3. The potential energy is associated with the position of atoms and the kinetic energy is associated with the vibration of the atoms. The volume and density can be used to observe the overall state of the system. The results in Figure S3 clearly show that these four terms all stabilize at their own equilibrium value, which indicates the Cu-PE system reaches the equilibrium state[1].
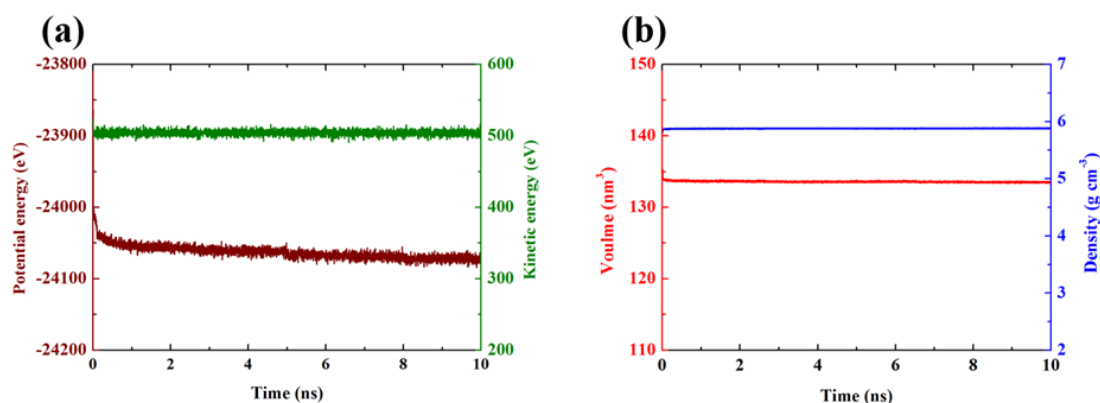

Figure S3. (a) The main view of Cu-PE. (b) The lateral view of Cu-PE.

The structural parameters of equilibrium PE nanocomposites are listed in Table S1. For Cu-PE, Ni-PE, Ag-PE and Au-PE system, each PE chain consists of 40 ethylene units. For bigger Cu-PE with length of 69.83 nm, each PE chain consists of 300 ethylene units. The periodic boundary conditions are applied to eliminate size effects. The lateral dimension is larger than 2 times of the cutoff and thus the impacts of the image atoms can be avoided.

Table S1. Structural parameters of equilibrium PE nanocomposites.

Equilibrium structure	Length (nm)	Width (nm)	Height (nm)	Number of atoms
Cu-PE	9.8	3.25	4.2	13000
Bigger Cu-PE	69.83	3.91	3.68	97500
Ni-PE	5.71	4.82	4,62	13000
Ag-PE	9.35	4.73	3.84	13000
Au-PE	9.55	4.47	3.79	13000

2. Detailed parameters of potential energy function

Based on density-functional ideas, Daw et al.[2] developed the embedded-atom method (EAM), which is suitable for the face-centered cubic (FCC) metals like Ni, Cu, Ag, Au, Pd, and Pt. In the EAM, the energy of each atom is computed from the energy needed to embed the atom in the local-electron density. The site potential energy U_i can be written as

$$U_i = \frac{1}{2}\sum_{j \neq i} \phi\left(r_{ij}\right) + F_i(\rho_{h,i}) \tag{1}$$

where $\phi(r_{ij})$ is the core-core pair repulsion energy between atoms i and j separated by the distance r_{ij}, $F_i(\rho_{h,i})$ is the energy to embed atom i into the background electron density $\rho_{h,i}$, and $\rho_{h,i}$ is the host electron density at atom i owing to the remaining atoms of the system. The electron density can be approximated by the superposition of atomic densities,

$$\rho_{h,i} = \sum_{j(\neq i)} \rho_j^a\left(r_{ij}\right) \tag{2}$$

where $\rho_j^a(r)$ is the electron density contributed by atom j. The atomic electron density is computed from the Hartree-Fock wave functions by

$$\rho^a(r) = n_s\rho_s(r) + n_d\rho_d(r) \tag{3}$$

where n_s, n_d, ρ_s, ρ_d are the number of outer s electrons, d electrons, and the densities related to the s and d wave functions, respectively. The pair interaction between atoms of types A and B can be written as

$$\phi_{AB}(r) = Z_A(r)Z_B(r)/r \tag{4}$$

where Z(r) is the pair-repulsion term,

$$Z(r) = Z_0(1 + \beta r^v)e^{-ar} \tag{5}$$

The accurate PCFF[3] potential is used to describe the atomic interactions of PE, which is based on quantum mechanical calculations. The PCFF includes bond, angle, dihedral, improper and nonbonding terms, which can be written as

$$E = \sum E^b + \sum E^a + \sum E^d + \sum E^{mbt} + \sum E^{ebt} + \sum E^{at} + \sum E^{aat} + \sum E^{bb13} +$$
$$E^{aa} + \sum E^{elec} + \sum E^{VDW} \tag{6}$$

where

$$E^b = \sum_{i=2}^{4} k_i^b\left(b - b_0\right)^i \tag{7}$$

$$E^a = \sum_{i=2}^{4} k_i^a\left(\theta - \theta_0\right)^i \tag{8}$$

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$$E^d = \sum_{i=1}^{3} k_i^d \left(1 - \cos(i\phi - \phi_i)\right) \tag{9}$$

$$E^{mbt} = (r_{jk} - r_2) \sum_{i=1}^{3} A_i \cos(i\phi) \tag{10}$$

$$E^{ebt} = \left(r_{ij} - r_1\right) \sum_{i=1}^{3} B_i \cos(i\phi) + (r_{kl} - r_3) \sum_{i=1}^{3} C_i \cos(i\phi) \tag{11}$$

$$E^{at} = \left(\theta_{ijk} - \theta_1\right) \sum_{i=1}^{3} D_i \cos(i\phi) + \left(\theta_{jkl} - \theta_2\right) \sum_{i=1}^{3} E_i \cos(i\phi) \tag{12}$$

$$E^{aat} = M(\theta_{ijk} - \theta_1)\left(\theta_{jkl} - \theta_2\right)\cos(\phi) \tag{13}$$

$$E^{bb13} = N\left(r_{ij} - r_1\right)(r_{kl} - r_3) \tag{14}$$

$$E^{aa} = M_1\left(\theta_{ijk} - \theta_1\right)\left(\theta_{kjl} - \theta_3\right) + M_2\left(\theta_{ijk} - \theta_1\right)\left(\theta_{ijl} - \theta_2\right) + M_3\left(\theta_{ijl} - \theta_2\right)\left(\theta_{kjl} - \theta_3\right) \tag{15}$$

$$E^{elec} = \sum_{i,j} q_i\, q_j / r_{ij} \tag{16}$$

$$E^{\mathrm{VDW}} = \sum_{i,j} \varepsilon_{i,j} \left[2\left(\frac{r_{i,j}^o}{r_{i,j}}\right)^9 - 3\left(\frac{r_{i,j}^o}{r_{i,j}}\right)^6\right] \tag{17}$$

Table S2. Parameters of PCFF for PE.

Style	Bond style class 2
	Angle style class 2
	Dihedral style class 2
	Improper style class 2
	Pair style lj/class 2/coul/cut 10.0 Å 10.0 Å
Parameters (units: metal)	All parameters are from PCFF[3].
Bond coefficients	1.5300 13.0172 -21.7962 29.5300 (C-C)
	1.1010 14.9863 -30.0547 36.6882 (H-C)
Angle coefficients	110.7700 1.8007 -0.4606 0.2228 (H-C-C)
	112.6700 1.7165 -0.3233 -0.4152 (C-C-C)
	107.6600 1.7219 -0.5613 -0.1056 (H-C-H)
Bond-bond term	0.1469 1.5300 1.1010 (H-C/C-C)
	0.0000 1.5300 1.5300 (C-C/C-C)
	0.2312 1.1010 1.1010 (H-C/H-C)
Bond-angle term	0.9000 0.4953 1.5300 1.1010 (H-C, C-C/H-C-C)
Bond-angle term	0.3476 0.3476 1.5300 1.5300 (C-C, C-C/C-C-C)
	0.7850 0.7850 1.1010 1.1010 (H-C, H-C/H-C-H)
	-0.0062 0.0000 0.0027 0.0000 -0.0047 0.0000

Dihedral term	(H-C-C-H) 0.0000 0.0000 0.0014 0.0000 -0.0073 0.0000 (H-C-C-C) 0.0000 0.0000 0.0022 0.0000 -0.0062 0.0000 (C-C-C-C)
Middle bond torsion term	-0.6184 -0.0231 -0.0211 1.5300 (C-C/H-C-C-H) -0.6452 -0.1586 -0.0136 1.5300 (C-C/H-C-C-C) -0.7713 -0.3117 0.0000 1.5300 (C-C/C-C-C-C)
End bond torsion term	0.0092 0.0135 0.0034 0.0092 0.0135 0.0034 1.1010 1.1010 (H-C/H-C-C-H) 0.0108 0.0105 -0.0040 0.0035 0.0026 0.0096 1.5300 1.1010 (H-C, C-C/H-C-C-C) -0.0032 0.0000 0.0000 -0.0032 0.0000 0.0000 1.5300 1.5300 (C-C/C-C-C-C)
Angle torsion term	-0.0351 0.0241 -0.0107 -0.0351 0.0241 -0.0107 110.7700 110.7700 (H-C-C/H-C-C-H) 0.0135 0.0196 -0.0086 -0.0106 0.0000 -0.0049 110.7700 112.6700 (H-C-C/H-C-C-C) 0.0169 -0.0136 0.0060 0.0169 -0.0136 0.0060 112.6700 112.6700 (C-C-C/C-C-C-C)
Angle angle torsion term	-0.5448 110.7700 110.7700 (H-C-C, H-C-C/H-C-C-H) -0.7009 112.6700 110.7700 (H-C-C, C-C-C/H-C-C-C) -0.9559 112.6700 112.6700 (C-C-C,C-C-C/C-C-C-C)
Angle angle improper term	0.0119 0.0119 -0.0209 107.6600 110.7700 110.7700

	-0.0572 0.0051 -0.0572 110.7700 112.6700
	110.7700
Lj/class2/coul/cut	0.0023 4.010 (C-C)
	0.001 2.995 (H-H)

Table S3. Parameters of LJ potential between Cu and PE.

Parameters (units: metal)	The parameters are from universal force field[4].
LJ/cut	0.051609 2.947025 (Cu-C)
	0.026152 2.636 (Cu-H)

3. Velocity autocorrelation function (VACF) of Cu and PE

Figure S4 shows the VACF of Cu and PE versus the electron-phonon coupling factor. The VACF decays to zero within the correlation time of 2.5 ps, which demonstrates the correlation time is enough. The VACF of Cu is more affected than the VACF of PE by the electron-phonon coupling factor.
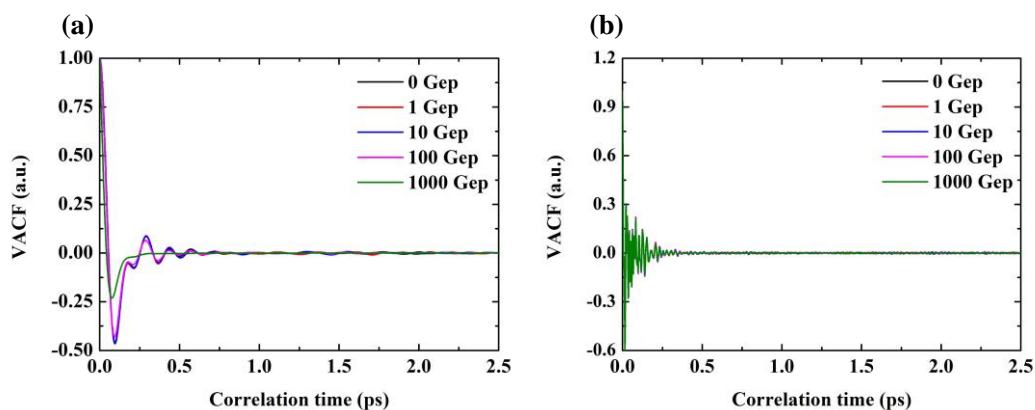

Figure S4. (a) The VACF of Cu as a function of electron-phonon coupling factor.

(b) The VACF of PE as a function of electron-phonon coupling factor.

References

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