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

Influence of the interfacial interaction strength on the viscoelasticity of hard-soft block copolymers based nanocomposite: a molecular dynamics simulation study

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The glass transition temperature

Usually, multiblock copolymers locate in the rubber state at room temperature. Moreover, the room temperature is above T_g of SSs but below T_g of HSs. So T_g of each model studied here was measured. Additionally, T_g of the system with 400 pure polymer chains in which each chain contained 96 H beads or S beads was determined. For nanocomposites, the systems were cooled from $1.5T^*$ to $0.3T^*$ under NPT ensemble and the corresponding density at each T was recorded. Two distinct slopes were observed in Fig. S1(a), which could be fitted in a linear fashion. T_g was estimated from the cross point of the two fitting lines and T_g of each model was shown in Fig. S1(b). An increasing trend of T_g along with increasing ε_{SF} was found. However, little change in model 4, 5 and 6 with higher ε_{HF} was observed owing to the small amount of H beads. As shown in Fig. S2, the T_gs of pure SSs and HSs were $0.43T^*$ and $0.92T^*$, respectively. Apart from model 3, T_gs of other models were below $0.8T^*$. Therefore the sinusoidal shear strain was imposed at $T = 0.8T^*$.

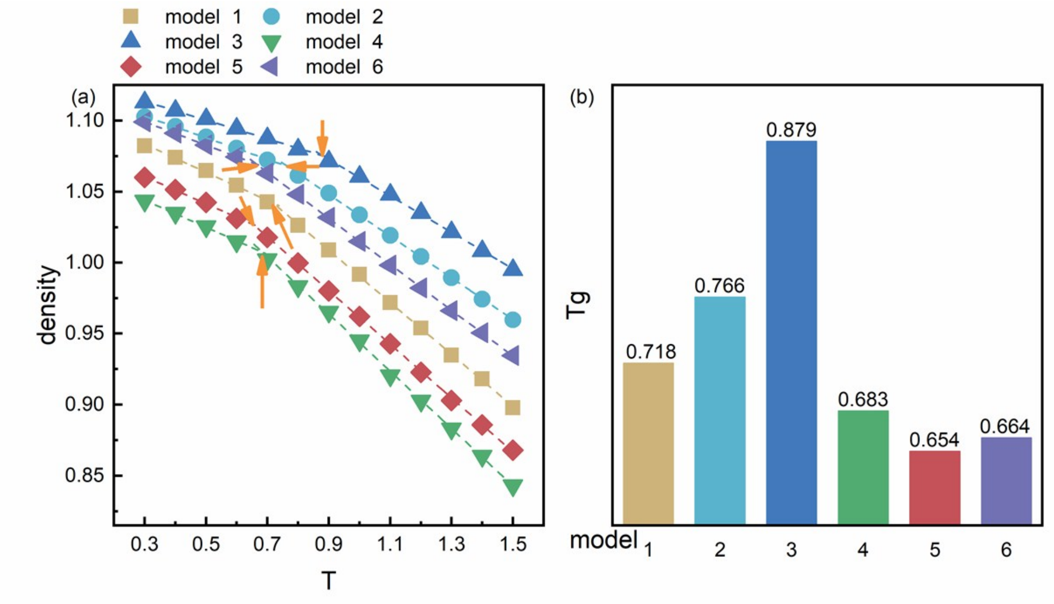

Fig. S1 (a) Density as a function of temperature for various models. The points are the data obtained from the simulation and they are fitted by lines. The cross points of the fitting lines are denoted by the orange arrows. (b) T_g for various models.

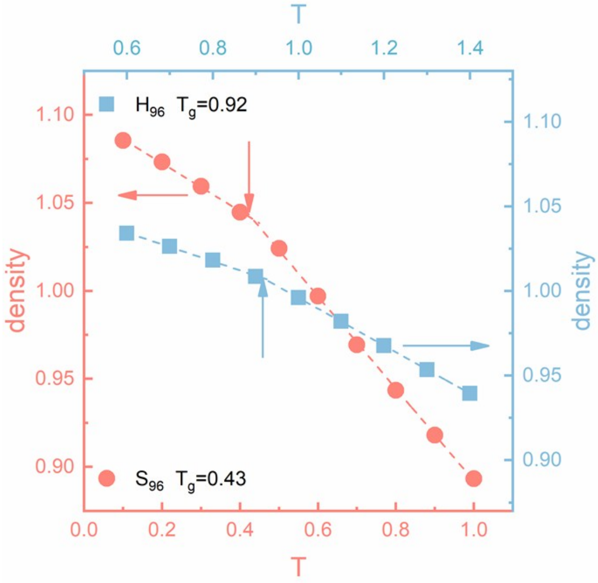

Fig. S2 Density as a function of temperature for H_{96} (blue squares) and S_{96} (pink circles). The points are the data obtained from the simulation and they are fitted by the lines. The cross points of the fitting lines are denoted by the perpendicular arrows.

Morphology

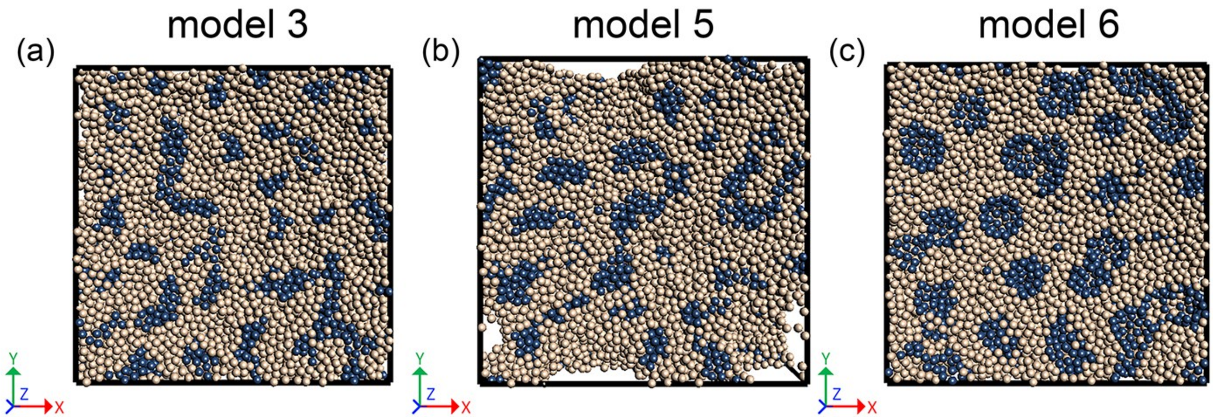

Fig. S3 Snapshots of the polymer matrix for (a) model 3, (b) model 5, and (c) model 6. The blue and peachpuff spheres denote H, and S beads, respectively.

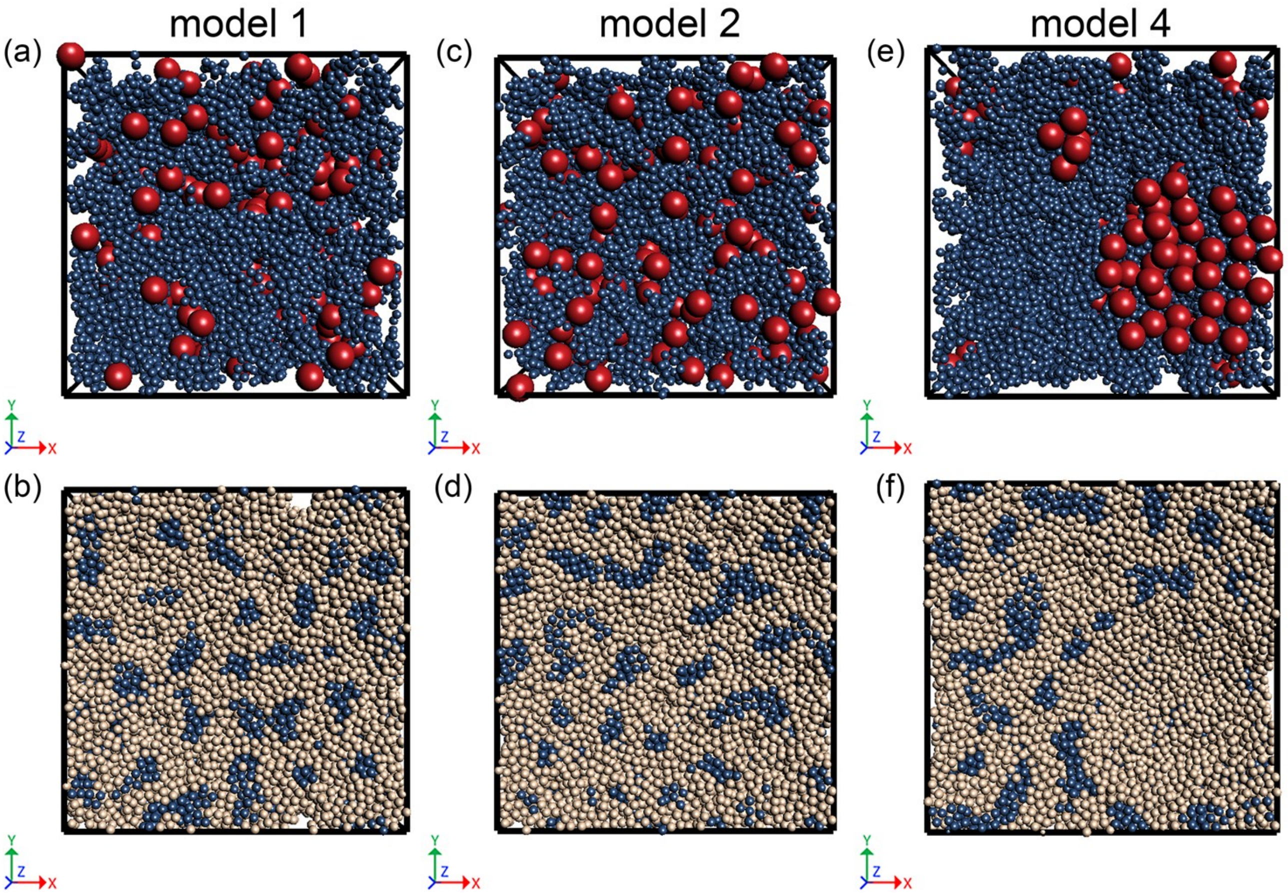

Fig. S4 Snapshots for (a, b) model 1, (c, d) model 2, and (e, f) model 4. The first line (a, c, e) only draws H beads and NFs, and the second line draws the H beads and S beads. The blue, peachpuff and red spheres denote H, S, and NF beads, respectively.

Strain dependent rheological properties

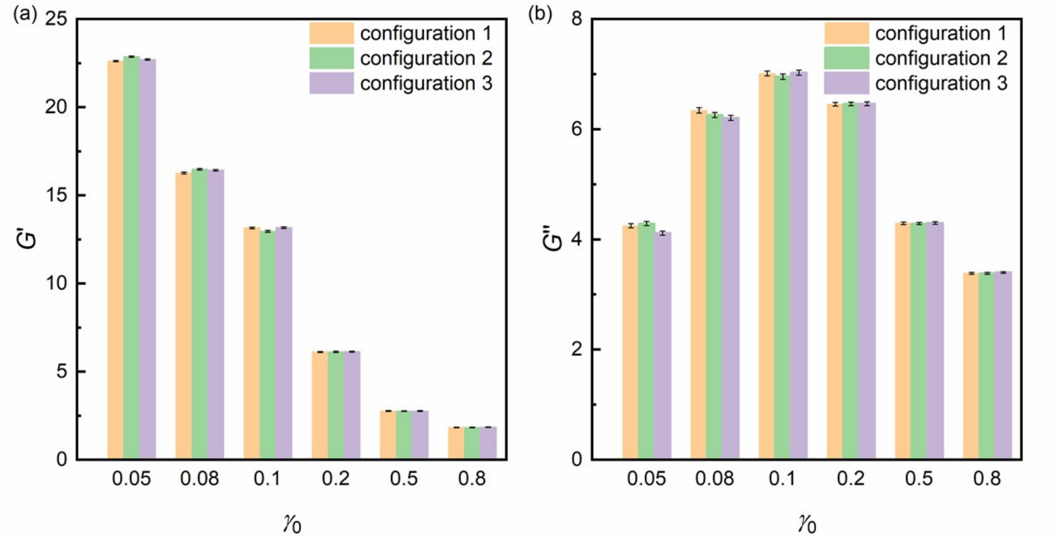

Fig. S5 (a) Storage moduli G' and (b) loss moduli G'' of model 3, which were obtained from 3 different simulations starting from distinct equilibrium configurations, as a function of the shear strain amplitude γ_0. Each column shown in this figure was fitting $\sigma_{xy}^{batch}(t)$ during 10 cycles with Eq. (8) and the error bars were added.

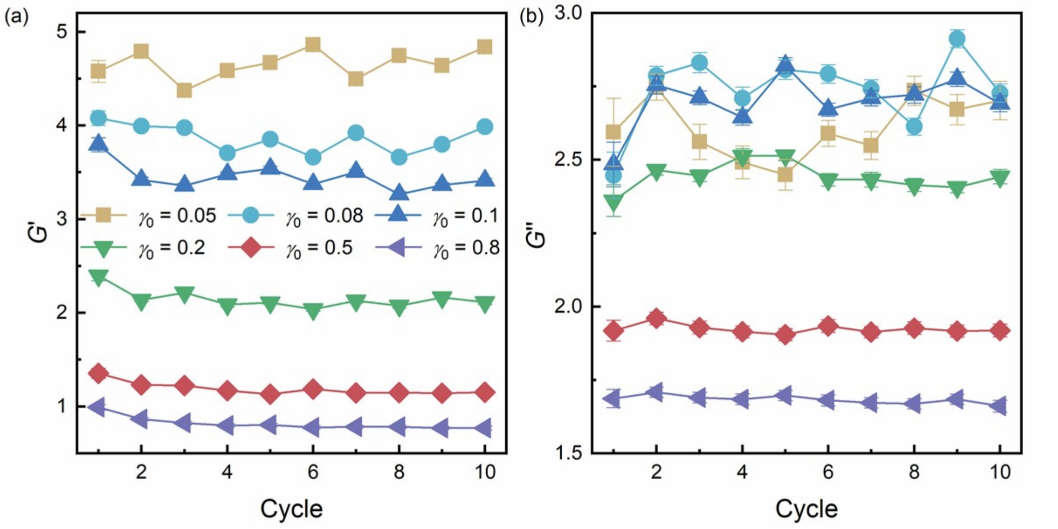

Fig. S6 (a) Storage moduli G' and (b) loss moduli G'' versus the cycle number for various γ_0 in model 4. Each data point shown in this figure was fitting $\sigma_{xy}^{batch}(t)$ during a variable number of cycles with Eq. (8) and the error bars were added.

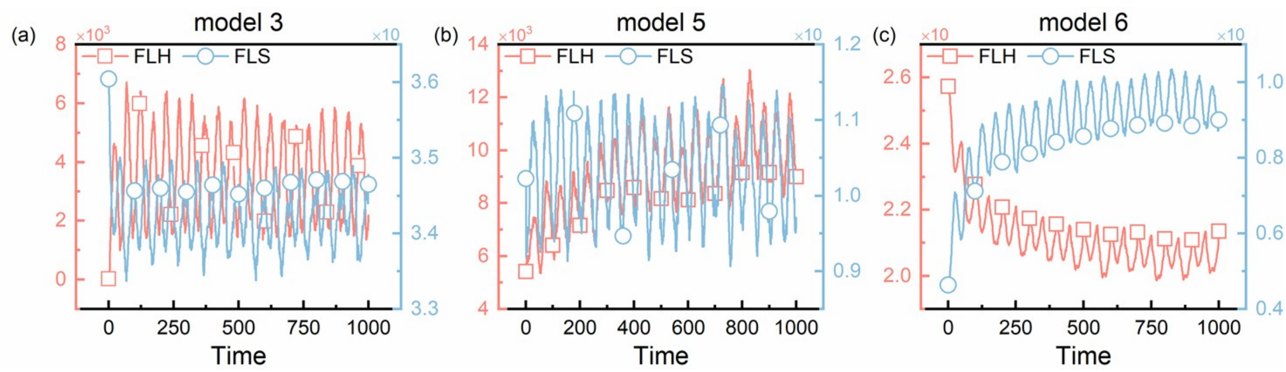

Fig. S7 The FLH (left axis) and FLS (right axis) as a function of deformation time for (a) model 3, (b) model 5, and (c) model 6 at $\gamma_0 = 0.8$.

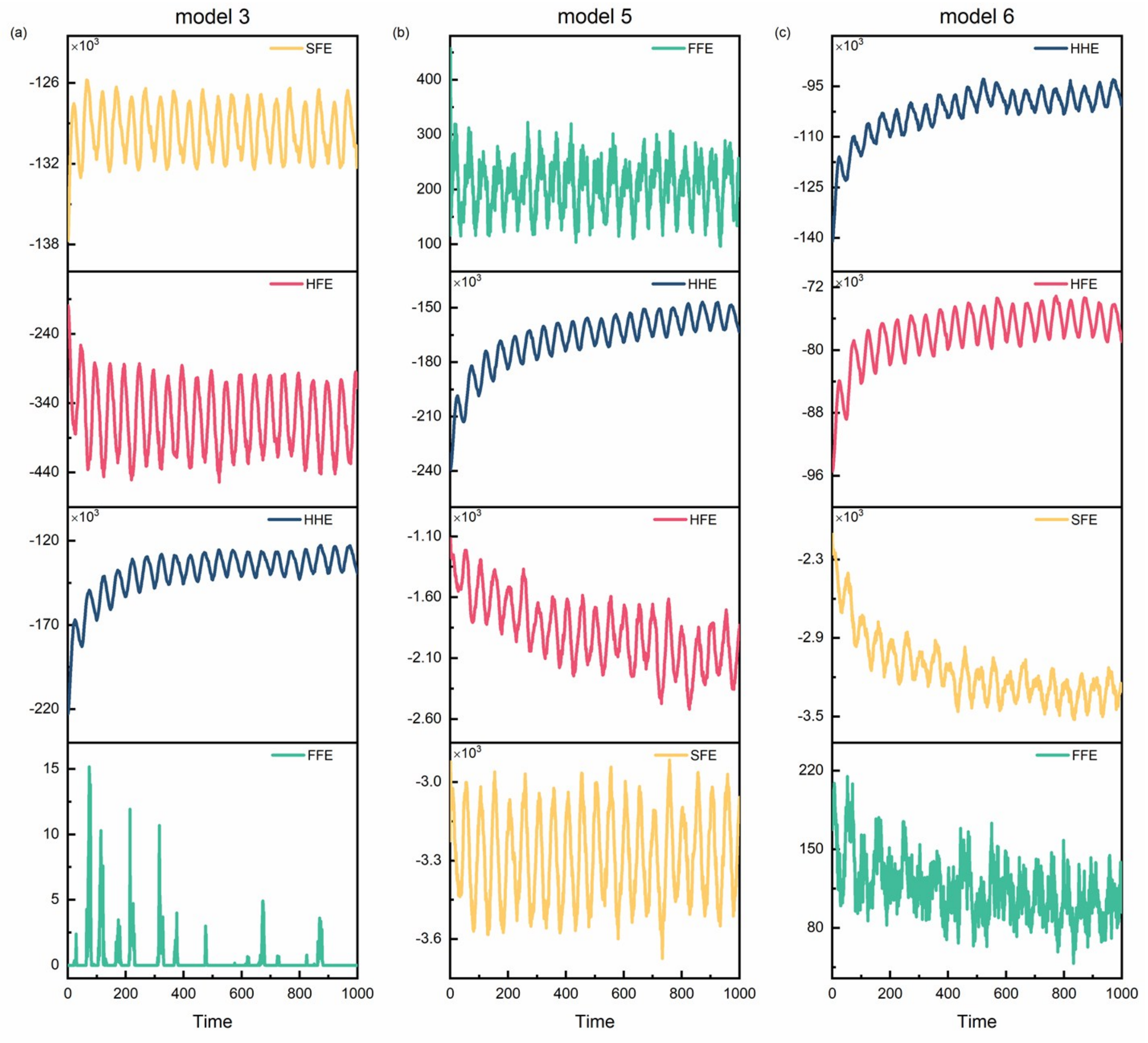

Fig. S8 (a) The SFE, HFE, HHE, and FFE as a function of deformation time for model 3 at $\gamma_0 = 0.8$. (b) The FFE, HHE, HFE, and SFE as a function of deformation time for model 5 at

$\gamma_0 = 0.8$. (a) The HHE, HFE, SFE, and FFE as a function of deformation time for model 6 at $\gamma_0 = 0.8$.

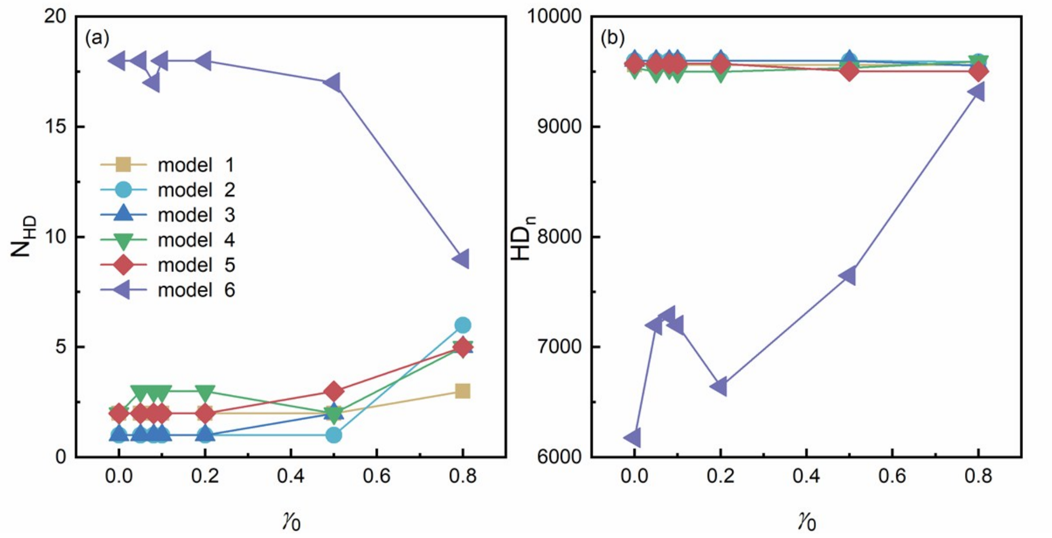

Fig. S9 (a) The number of hard domains N_{HD}, and (b) the number of hard beads contained in the maximum hard domain7 HD_n at the equilibrium state and each γ_0 in the first cycle for various models.

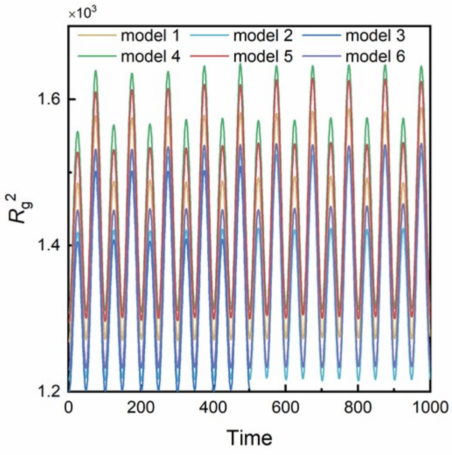

Fig. S10 R_g^2 of all H beads for various models as a function of deformation time.

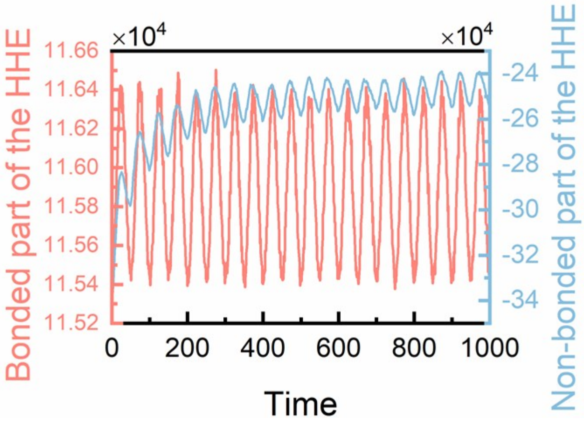

Fig. S11 The bonded and non-bonded part of the HHE as a function of deformation time for model 3.