Supporting Information

Retardation on blending in the entangled binary blends of linear polyethylene: a molecular dynamics simulation study

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Mean squared internal distances

To judge the equilibration of our studied systems or to determine whether the conformations are well equilibrated or not, we calculate the mean squared internal distances, $\langle R^2(n) \rangle$, for our studied monodisperse and bidisperse polyethylene (PE) systems after the equilibration process. Here, the typical result for the monodisperse system with the longest chain length, m600, which has the longest $\tau_c$ is presented below. As shown in Figure S1, the rescaled mean squared internal distances $\langle R^2(n) \rangle / n$ of m600 increases monotonically with the increase of $n$ and reaches a plateau value after $n=100$ (though little fluctuations exist for large $n$), indicating that the chains are well equilibrated at the short and long length scales. Particularly, the curve is in good agreement with the linear PE system with the same force field and in the same thermodynamic conditions shown in Figure 5. of Ref 1. The characteristic ratio, $C_\infty$, estimated from the ratio of the plateau value of $\langle R^2(n) \rangle / n$ to $l_b^2$ (squared averaged bond length) is 8.43, which is in the reasonable range of 9.60~8.06. All these demonstrate that the equilibration runs of 500ns can guarantee conformational relaxation of our studied PE chains and hence the equilibration time is probably sufficient for m600.
Figure S1. Rescaled mean squared internal distance, as a function of n for m600 after the equilibration stage.

**Time-decay of ACF for the independent runs of m120 and b120**

we present the time-decay of the autocorrelation function (ACF) for the end-to-end vector for two independent runs of m120 and b120 in Figure S2. The curves of the independent runs nearly fall on top of each other, which indicates the unavoidable uncertainty is almost neglectable (Note that each of the independent run is run over 200 correlation times).

Figure S2. Time-decay of ACF for the (probe) chain end-to-end vector in both monodisperse and bidisperse systems with the chain length of 120 in semi-logarithm coordinates. Two independent runs of the monodisperse and bidisperse systems are denoted as m-I, m-II and b-I, b-II respectively.
Consistency with the earlier PE simulations

The atomistic molecular dynamics (MD) simulation trajectories of PE melts used by Stephanou et al.\(^3\) were accumulated from the work of Karayiannis and Mavrantzas\(^4\). In Table I of Ref 3 Stephanou et al showed the disentanglement time, \(\tau_d\), which is determined from the third characteristic break of the time-dependent segmental mean square displacement (MSD), corresponding to the passage from the Rouse-like diffusion to free diffusion as stated by Stephanou et al. However, in Table 4 of Karayiannis and Mavrantzas\(^4\) the correlation time, \(\tau_c\), was presented, as we did in Table 2 and Figure 2 in our manuscript. \(\tau_c\) is determined by the relaxation of the chain end-to-end vector, which describes the orientational relaxation. Therefore, it seems more reasonable and interesting to compare the relaxation times of Karayiannis and Mavrantzas\(^4\) in order to elucidate the consistency with the earlier PE simulations. As shown in Figure S3, the relaxation times of our monodisperse melts are reasonably consistent with those reported by Karayiannis et al. except for the point of \(N = 270\), which seems smaller than the estimation from other points. The consistency would be improved if this point \((N = 270)\) is excluded as indicated by Figure S4. As for comparisons with the relaxation time result of Stephanou et al., both Figure S3 and Figure S4 indicate that the disentanglement times \(\tau_d\) of monodisperse PE systems of Stephanou et al. are longer than the correlation times \(\tau_c\) reported in our manuscript and of Karayiannis et al. We stress that the MD simulation trajectories of Stephanou et al. were taken from work of Karayiannis et al., such difference hence could be attributed to the analyzing methodologies and different physical observables for determining the
relaxation time but not numerical errors. Further, as illustrated in Figure S3 and Figure S4 we can use the power law of $\tau_c \propto N^\alpha$ for $N \geq 174$ to describe the dependence of the relaxation time on the chain length. For the monodisperse melts, as reported in our manuscript the exponent is $\alpha \approx 3.3$ and it is also 3.3 for Karayiannis’ data no matter the point of $N = 270$ is excluded or not, which are all larger than that in the blends ($\alpha \approx 3.1$). However, the exponent derived from Stephanou’ data is $\alpha \approx 2.7$, which seems to deviate from the predication of reptation theory.

Figure S3. Dependence of the correlation time, $\tau_c$, on the length of the probe chain, $N$, in the monodisperse systems (m) and the binary blends (b) denoted as black squares and red circles, respectively. The relaxation time reported by Stephanou et al. and Karayiannis et al. are also included for comparison, denoted as green up and blue down triangles, respectively.
Multi-mode Maxwell functions’ fitting on the ACF curves

We replot Figure 1 (the time-decay of the autocorrelation function (ACF) for the end-to-end vector) double-logarithmically, which is shown below as Figure S5. At a first glance, clearly the longer (probe) chain the slower relax. For example, the time taken for the ACF curves to decay to a finite small value is increased with the (probe) chain length. Additionally, the probe chains in the bidisperse blends relax more slowly than those in the monodisperse systems, indicative of the retardation on blending. Actually, the same conclusions as we drawn in the manuscript are reached here.
Further, the multi-mode Maxwell function is used to fitting these ACF curves:

\[ ACF(t) = \sum_{p \geq 1} g_p \exp \left( -\frac{t}{\tau_p} \right) \text{ with } \sum_{p \geq 1} g_p = 1 \]  

(R1)

where \( g_p \) is the normalized intensity of \( p \)th relaxation mode and \( \tau_p \) is the relaxation time of this mode.\(^5\) The 5-mode Maxwell function is applied to all the ACF curves, which provides a good fit to the data as typically shown in Figure S6.
Figure S6. Decay of ACF for the chain end-to-end vector for m120 from the MD simulation (black squares). The red line is the best fit from the 5-mode Maxwell function.

The resulting relaxation times are listed in Table S1. Generally, the longest relaxation time (in both monodisperse and bidisperse systems) is larger from the Maxwell fitting than from KWW fitting except for m600, probably because the ACF of only decays to about 0.2 and m600 has not fully relaxed yet within the simulated time scale. Note that “the relaxation function of a fully relaxing system can be expressed as a superposition of exponential decaying modes (multi-mode Maxwell function)”\(^5\). Thus, the time of 591 ns in Table S1 for m600 derived from the Maxwell fitting is not the actual longest relaxation time. Longer MD simulation would be needed for a more rigorous test of the Maxwell fitting in m600. Nevertheless, similar with the result from KWW fitting, the relaxation time is longer in the blends than in the monodisperse systems, quantitatively indicating the retardation on blending as well. As for the dependence of the longest relaxation time on the chain length, as illustrated in Figure
S7 the resulting exponents $\alpha$ of the power law of $\tau_c \propto N^\alpha$ for the monodisperse and bidisperse systems are 3.3 and 3.1 respectively. Such smaller exponent and longer relaxation times in the blends indicate again that the CR suppression from the matrix in blends becomes weaker with the reducing of the ratio of the chain length of the probe to matrix. Hence the multi-mode Maxwell function fitting has no qualitative effect on our discussions and conclusions.

Table S1. The longest relaxation time ($\tau_c$) obtained from the multi-mode Maxwell function fitting of ACF curves for both monodisperse and bidisperse PE systems. For comparisons, values of the characteristic relaxation time, $\tau_{KWW}$, the stretching exponent, $\beta$, and the corresponding correlation time, $\tau_c$ derived from KWW function fitting are listed as well.

<table>
<thead>
<tr>
<th>N</th>
<th>$\tau_{KWW}$ (ms)</th>
<th>$\beta$</th>
<th>$\tau_c$ (ms) KWW</th>
<th>$\tau_c$ (ms) Maxwell</th>
<th>$\tau_{KWW}$ (ms)</th>
<th>$\beta$</th>
<th>$\tau_c$ (ms) KWW</th>
<th>$\tau_c$ (ms) Maxwell</th>
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</thead>
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<tr>
<td>120</td>
<td>4.03</td>
<td>0.72</td>
<td>4.94</td>
<td>5.72</td>
<td>5.41</td>
<td>0.71</td>
<td>6.79</td>
<td>7.86</td>
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<tr>
<td>180</td>
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<td>0.68</td>
<td>15.1</td>
<td>17.2</td>
<td>14.9</td>
<td>0.65</td>
<td>20.4</td>
<td>23.9</td>
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<td>240</td>
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<td>0.64</td>
<td>35.6</td>
<td>38.8</td>
<td>29.5</td>
<td>0.62</td>
<td>42.4</td>
<td>52.0</td>
</tr>
<tr>
<td>300</td>
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<td>0.58</td>
<td>82.9</td>
<td>98.2</td>
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<td>0.58</td>
<td>101</td>
<td>112</td>
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</table>
Figure S7. Dependence of the longest relaxation time from the Maxwell fitting, $\tau_c$, on the length of the probe chain, N, in the monodisperse systems (m) and the binary blends (b) denoted as black squares and red circles respectively.

**Fluctuations in the viscoelastic relaxation functions and results of the Orientation relaxation functions**

Although the calculation of viscoelastic function or stress is straightforward in MD simulations, a reliable estimate of the viscoelastic relaxation or stress relaxation $G(t)$ is more difficult than that of the end-to-end relaxation. As stated by Likhtman et al. the limited simulation time scale and the huge ratio of the amplitude of the stress fluctuations to the value of the plateau modulus hinder the measurement of the stress relaxation. This means that in addition to calculate the stress at every time step one should conduct the simulations with larger box sizes, longer simulation times and several independent runs to improve the signal-to-noise ratio or to ensure good statistics. For example, Likhtman et al. simulated several reptation times and averaged over 8-10 independent runs to get good enough stress relaxation data wherein the longest run took almost a year. However, for atomistic PE models, especially for the blends in our present work, it is almost impossible for computation sources we hold nowadays to conduct such great scale simulations, which could cost more than 10 million core hours. Taking the shortest system of m120 for example, as shown in Figure S8, compared to the relaxation curve of the longest simulated time (200 correlation times), large
fluctuations are obviously observed for other curves when MD times in a production run were approximately at the terminal time scale. Since extremely long simulation runs (of the order of $10^{-100}$ correlation times) are almost computationally prohibitive at the moment for atomistic models, we do not think the stress relaxation could be obtained with good enough quality to quantitatively study the CR effect especially for the systems with longest chain and blends. Therefore, as stated in the manuscript, the use of coarse-grained model could be a useful methodology to study the viscoelastic relaxation of entangled polymers in a large box size on a long time scale at reasonable computational cost, which are hard to access by current atomistic MD simulations.

Figure S8. Stress relaxation for the monodisperse system with chain length of 120, namely, m120, in the semi-logarithmic (a) and in the double logarithmic (b) plots, when MD times in a production run after equilibration were 1, 2, 4, 8, 20, and 200 correlation times. The method for calculating the stress relaxation is described by eq(4) in Ref 7 and via multiple-tau correlator algorithm with processing the stress at every time step.
On the other hand, for the relaxation function of the end-to-end vector, ACF, as typically shown in Figure S9, large fluctuations are absent at the terminal time scale even when MD times in a production run were only several correlation times, indicating that the relaxation of the end-to-end vector is relatively more robust against the simulation time after equilibration than the relaxation of the stress. To further confirm its robustness to the variation of the production run length after equilibration, we make another simple test on the relaxation of ACF. As shown in Figure S10, a dozen independent samples of m120 with MD times in a production run just over 1 correlation time can relax simultaneously with the sample with MD times in a production run over 200 correlation times and do not show large fluctuations at the terminal time region. KWW function is used to fit these curves and the average value of the resulting correlation times for the samples with the production run length of 1 correlation time is 5.65±0.76ns, which is still reasonable when compared to the correlation time (4.94ns) for the sample of 200 correlation times. Hence, due to the unavoidable large fluctuations in the viscoelastic relaxation function without the extremely long run, we only perform the analysis on the relaxation of the end-to-end vector and leave the estimate of the viscoelastic relaxation for future studies, although the calculation of the viscoelastic relaxation is straightforward, general, and useful.
Figure S9. ACF relaxation for the monodisperse system with chain length of 120, namely, m120, in the semi-logarithmic (a) and in the double logarithmic (b) plots when MD times in a production run after equilibration were 1, 2, 4, 8, 20, and 200 correlation times. The method for calculating the relaxation is the multiple-tau correlator algorithm with processing the end-to-end vector at every 4000 steps (8ps).

Figure S10. ACF relaxation for the monodisperse system with chain length of 120, namely, m120, in the semi-logarithmic (a) and in the double logarithmic (b) plots when MD times in a production run after equilibration were 1 and 200 correlation times. Note that a dozen independent samples with the production run length of 1 correlation time are denoted as the thin lines and the sample of 200 correlation times.
is denoted as the green thick line. The method for calculating the relaxation is the multiple-tau correlator algorithm\textsuperscript{8} with processing the end-to-end vector at every 4000 steps (8ps).

Despite the infeasibility of calculating the stress relaxation directly, to gain insight into the CR effects on the stress relaxation of our monodisperse and bidisperse PE systems, an indirect way forward is to resort to the use of the stress-optical law. According to the stress-optical law, the total stress relaxation function, $G(t)$, is proportional to the total relaxation function of the orientation tensor, $S(t)$:

$$G(t) = \frac{1}{\gamma} S(t) \quad \text{(R2)}$$

where $S(t) = \frac{N_b}{k_B T} \sum_{j=1}^{N_c} \left\{ O_j^{\alpha\beta}(t) O_j^{\alpha\beta}(0) \right\}$, $O_j^{\alpha\beta}(t) = \sum_{i=1}^{N-1} u_i^\alpha(t) u_i^\beta(t)$ is the orientation tensor of chain $j$, $N_c$ is the number of chains, $N$ is the number of monomers in chain $j$, $N_b = N_c \times (N-1)$ is the total number of bonds, $u_{ij}$ is the $i$th bond vector of chain $j$, $\alpha$ and $\beta$ are the Cartesian components, and $\gamma$ is the stress-optical coefficient\textsuperscript{8}. Further, the total orientation relaxation function could be decomposed into the autocorrelation within the same chain, $A(t)$, and cross-correlation between different chains, $C(t)$, namely,

$$S(t) = A(t) + C(t) \quad \text{(R3)}$$

where

$$A(t) = \frac{N_b}{k_B T} \sum_{j=1}^{N_c} \left\{ O_j^{\alpha\beta}(t) O_j^{\alpha\beta}(0) \right\}, \quad \text{(R4)}$$

$$C(t) = \frac{N_b}{k_B T} \sum_{i \neq j} \left\{ O_i^{\alpha\beta}(t) O_j^{\alpha\beta}(0) \right\}. \quad \text{(R5)}$$
For the binary blends with long and short chains, the total relaxation function of the orientation tensor, $S(t)$, is described as:

$$S(t) = \varphi_l A_l(t) + \varphi_l^2 C_{ll}(t) + \varphi_s A_s(t) + \varphi_s^2 C_{ss}(t) + 2\varphi_l\varphi_s C_{ls}(t)$$

(R6)

where the subscripts $l$ and $s$ are denoted as long and short chains in the blends, and $\varphi$ is the volume fraction.

To be consistent with the analyses above, we take the shortest system of m120 as an example. Figure S11 shows the normalized total orientation relaxation function, $S(t)/S(0)$, and its corresponding normalized autocorrelation relaxation function, $A(t)/A(0)$, and cross-correlation function, $C(t)/C(0)$ for m120 when MD times in a production run after equilibration were 2 and 200 correlation times. Similar with the behaviors of the stress relaxation shown in Figure S8, compared to the data with the production run length of 200 correlation times, the relaxations of $S(t)/S(0)$ and $C(t)/C(0)$ with the production run of 2 correlation times show large fluctuations, which would hinder the quantitative investigations of CR effects. However, the curves of $A(t)/A(0)$ seem to be more robust to the variation of production run times. As shown in Figure S12, $A(t)/A(0)$ with different production run times relaxes simultaneously and shows smaller fluctuations when compared to $S(t)/S(0)$ and $C(t)/C(0)$. The values of $A(0)/S(0)$ and $C(0)/S(0)$ are 0.73 and 0.27 for the production run of 2 correlation times, 0.63 and 0.37 with the production run of 200 correlation times respectively. We see that the values of $A(0)/S(0)$ are 2 or 3 times larger than the values of $C(0)/S(0)$, namely, $A(0)/S(0)$ and $C(0)/S(0)$ are on the same magnitude, suggesting that the fluctuations amplified or diminished by the
normalization are tolerable. In this regard, we could quantitatively (though indirectly and partially) investigate the CR effects on the viscoelastic relaxation by the use of autocorrelation orientation relaxation without performing extremely long atomistic MD simulation runs (of the order of 10~100 correlation times).

Figure S11. Normalized orientation relaxation for the monodisperse system with chain length of 120, namely, m120, in the semi-logarithmic (a) and in the double logarithmic (b) plots when MD times in a production run after equilibration were 2 and 200 correlation times. S, A, and C are denoted as the total orientation relaxation, autocorrelation relaxation, and cross-correlation relaxation, respectively. The method for calculating the orientation relaxation is the multiple-tau correlator algorithm with processing the bond vector at every 4000 steps (8ps).
Figure S12. Normalized autocorrelation relaxation for the monodisperse system with chain length of 120, namely, m120, when MD times in a production run after equilibration were 1, 2, 4, 8, 20, and 200 correlation times. The method for calculating the relaxation is the multiple-tau correlator algorithm with processing the bond vector at every 4000 steps (8ps).

Inspired by the above result, we calculate the autocorrelation functions of orientation relaxation of our studied monodisperse and bidisperse PE systems. Figure S13 presents the decay of normalized autocorrelation orientation relaxation $A(t)/A(0)$ as a function of time in both semi-logarithm and double-logarithmic scales. Similar with the relaxation of ACF for the end-to-end vector, the curves of the probe chains in the blends relax more slowly than those in the monodisperse systems, indicating that the stress relaxation is retarded on blending, which is consistent with the result from the primitive chain network simulations. To quantify the relaxation behaviors of $A(t)/A(0)$, the multi-mode Maxwell function is utilized to fit the curves, and the resulting longest relaxation time, $\tau_{p,\text{longest}}$, is shown in Figure S14.
Figure S13. Time-decay of $A(t)/A(0)$ for the bond vector along the (probe) chain in both monodisperse and bidisperse systems in semi-logarithm scales (a) and in double logarithmic scales (b).

Very similar to the relaxation of the end-to-end vector in Figure S7, the longest relaxation time is smaller in the monodisperse systems than in the bidisperse blends, which quantitatively confirms the retardation on blending as well. The exponent, $\alpha$, of the power law in the form of $\tau_{p, longest} \propto N^{\alpha}$ is 3.1 in the monodisperse systems and 3.0 in the blends. The exponent is somewhat smaller in the weak CR environment than in the monodisperse systems, which is also qualitatively consistent with results of the end-to-end vector relaxation. Nevertheless it is a pity for us that the $S(t)/S(0)$ and $C(t)/C(0)$ are inaccessible here as mentioned above.
Figure S14. Dependence of the longest relaxation time, $\tau_{p,\text{longest}}$, from the multimode Maxwell fitting on the length of the probe chain, $N$, in the monodisperse systems (m) and the binary blends (b) denoted as black squares and red circles, respectively.

**MSD for mean path**

The MSD of a monomer, $g_1(t)$, is defined as $g_1(t) = \langle (r(t + \tau) - r(\tau))^2 \rangle$, where $r(t)$ is the position of the monomer. Here we focus on the inner six monomers in a chain to eliminate the end effects and thus $g_1(t)$ are averaged over these monomers as showed in Figure S15. For $g_1(t)$ in the atomistic positions, a crossover could be observed at the time scale around the entanglement time, $\tau_e \approx 2320$ ps, after which the probe chains begin to feel the restriction from other chains. The slope in the
reptation regime is 0.40 since this system is not well entangled. For \( g_1(t) \) in the mean-path positions, the crossover is delay to about the averaging time \( \tau_{ave} \approx 2 \tau_e \). The values of \( g_1(t) \) is smaller than in the mean-path positions than in the atomistic positions in the short time scale since the mean paths represent the center line of the tube. In the long time scale, \( g_1(t) \) is the same in both positions. For the primitive chains which are constructed via the space averaging, the degree of the coarse graining affects the dynamics of the monomers in the short time scale as well\(^{10} \). The variations of the averaging time during the constructions of the mean path or the number of the atomistic beads in each united bead of the primitive chain affect the dynamics of the mean path or primitive chain in the short time scale, however, hardly affect the dynamics in the long time scale.

Figure S15. MSD, \( g_1(t) \), of the inner six monomers, in the atomistic and mean-path positions for the monodisperse system 300, denoted as m300 and m300MP respectively.
Kremer-Grest model simulation

The model we use here is the Kremer-Grest (KG) bead-spring model where the purely repulsive Lennard-Jones (LJ) beads are connected via finite extensible nonlinear elastic (FENE) potential\textsuperscript{6,10}. Additionally, a bond bending potential is introduced to increase the chain stiffness and thus decrease the entanglement length:

\[
U_{\text{bending}}(\theta) = k_\theta (1 - \cos (\theta))
\]

where \( k_\theta = 3 \) is the strength of the bending potential. This cos-form bending potential would be practically the same as the harmonic bending potential used in Ref\textsuperscript{6,11} The resulting entanglement length is \( N_e \approx 15 \) and entanglement characteristic time is \( \tau_e \approx 300 - 1000 \).\textsuperscript{12-13} Thus, a cross marking the beginning of the tube constraint would appear around \( \tau_e \) for \( g_1(t) \) of the inner monomers. In Figure S16, we show the results for this KG model with the chain length is 200 and the number of chains is 200. The crossover is observed at \( t \approx 1000\tau \) for \( g_1(t) \) of the inner six monomers. Meanwhile, a crossover occurs for \( g_\perp(i, t) \) of the inner six monomers.
Figure S16. $g_{\perp}(i, t)$ of the inner six monomers, mean-square displacement of the center of mass of the molecules, $g_3(t)$, and of the inner and outer six monomers, $g_1(t)$-inner, and $g_1(t)$-end.

**Self-looping**

As for the self-looping, we can refer to case wherein the value of $g_{\parallel}(i, t)$ would increase quickly while the spatial distance between the monomers of $i$ and $j$ keeps short at an intersection of a loop. Furthermore, the intrachain radial distribution function (RDF), $g(r)$, is used to describe the probability of the monomers in a chain to foldback and meet each other. As shown in Figure S17, $g(r)$ increases with the increasing of the chain length for $r > 1$ nm ($bond\ legth = 0.154$ nm, $\sigma_{ij} = 0.395$ nm), indicating
that the longer chain length is, the more likely monomers would foldback and form loops.

![Figure S17. Intrachain radial distribution function, $g(r)$, vs $r$ for our studied the monodisperse PE melts](image)

**Estimators of the entanglement length**

Entanglement length $N_e$ is a fundamental quantity in the tube theory and could be determined by several possible methods, for example, primitive path analysis. Here, basing on the primitive path analysis, we discuss briefly the entanglement length $N_e$ determined from different estimators proposed in Ref14, namely,

- classical S-kink (CSK): $N_e(N) = \frac{N(N-1)}{<Z>(N-1) + N}$
- modified S-kink (MSK): $N_e(N) = \frac{N}{<Z>}$
classical S-coil (CSC): \[ N_e(N) = (N - 1) \left( \frac{<R_{ee}^2>}{<L_{pp}>^2} \right) \]
modified S-coil (MSC): \[ N_e(N) = (N - 1) \left( \frac{<L_{pp}^2>}{<R_{ee}^2>} - 1 \right)^{-1} \]

The characteristic value of the entanglement length \( N_e \) given by Robert et. al.\(^{14} \) is about 84 calculated from the S-coil or 44 calculated from the S-kink which is close to the value estimated here respectively, as showed in Figure S18. Experimentally, Fetters et. al.\(^{15} \) proposed a value of \( N_e \approx 82 \) as calculated from the plateau modulus at a temperature of \( T = 443 \) K that is very close to the value estimated from S-coil. However, we should note that the value of the entanglement length relies on the methods for its calculation, and the reason of the difference is not clear yet.

Figure S18. Entanglement Length, \( N_e \), determined from different estimators as a function of the chain length, \( N \), in binary blends (denoted as b-estimator) and monodisperse systems (denoted as m-estimator).
References