

Supporting Information for:

Investigating Hydrogen Bonding in Poly(Vinyl Butyral) Copolymers near Glass-transition Temperature Under Uniaxial Stress: A Coarse-grained Molecular Dynamics Study

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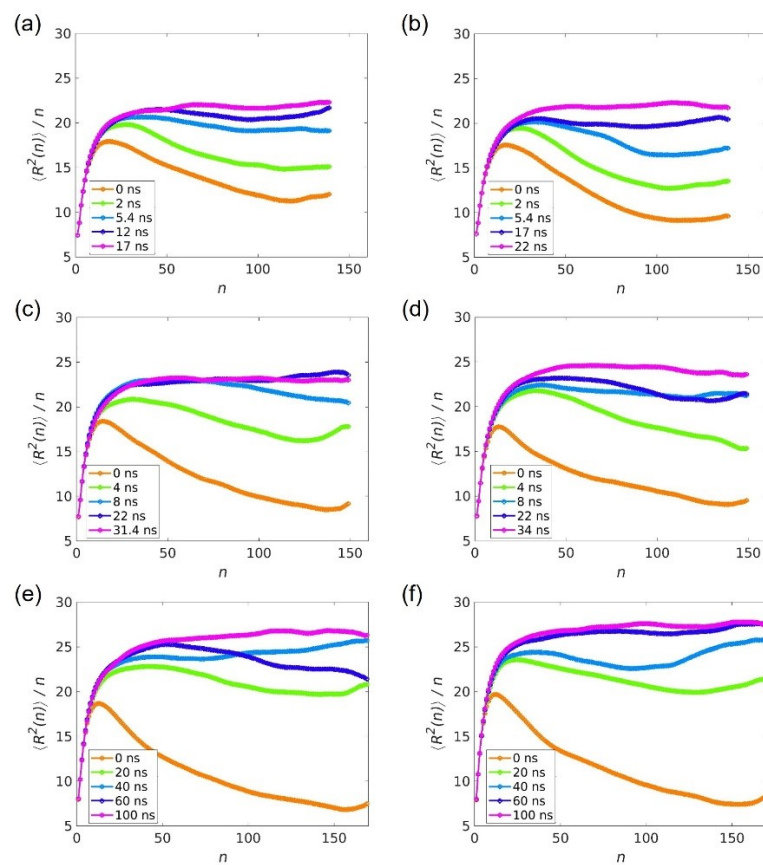


Figure S1. The mean square internal distances of the molecular chain, $\langle R^2(n) \rangle / n$, for the random PVB copolymers containing 25 mol% VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol% VA, $b=0.00$ (c) and $b=0.50$ (d), 75 mol% VA, $b=0.00$ (e) and $b=0.50$ (f), relaxed for different times.

mol% VA	b	Number of entanglements
25	0.00	0.94
25	0.50	1.02
50	0.00	1.80
50	0.50	1.64
75	0.00	3.13
75	0.50	3.44

Table S1. The number of entanglements in the equilibrated systems.

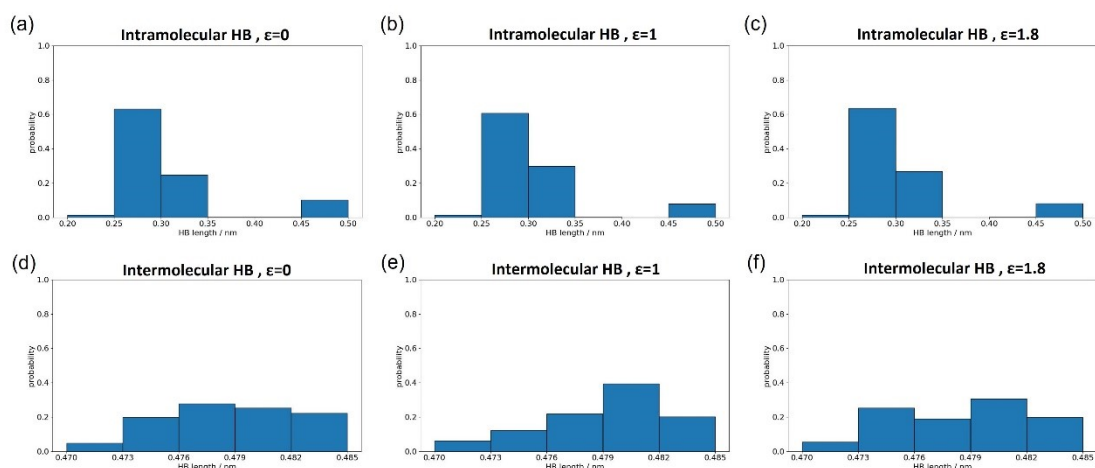


Figure S2. Probability distributions of intramolecular/intermolecular hydrogen bond length based on the $r \leq 0.4840$ nm criterion under various strain conditions in the 50 mol% VA ($b = 0.00$) system.

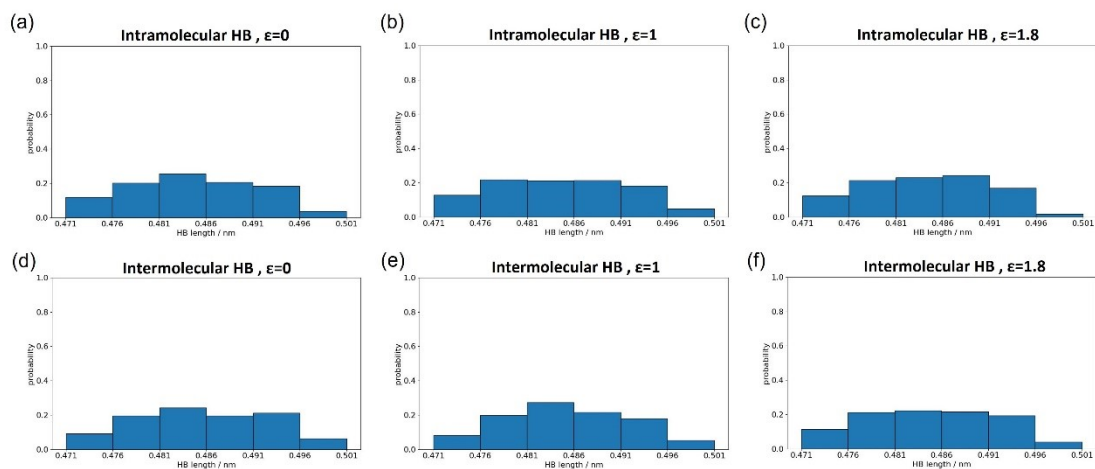


Figure S3. Probability distributions of intramolecular/intermolecular hydrogen bond length based on the $0.4712 \text{ nm} \leq r \leq 0.4914 \text{ nm}$ criterion under various strain conditions in the 50 mol% VA ($b = 0.00$) system.

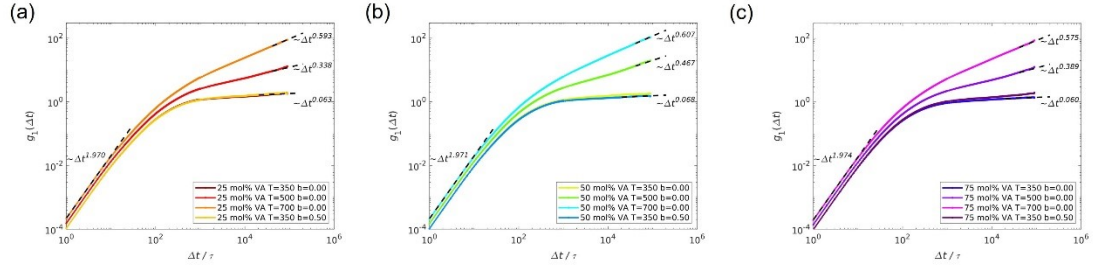


Figure S4. The plot of $g_1(\Delta t)$, where $g_1(\Delta t)$ represents the simulated MSD of the PVB copolymer chains containing various amount of VA and degrees of blockiness.

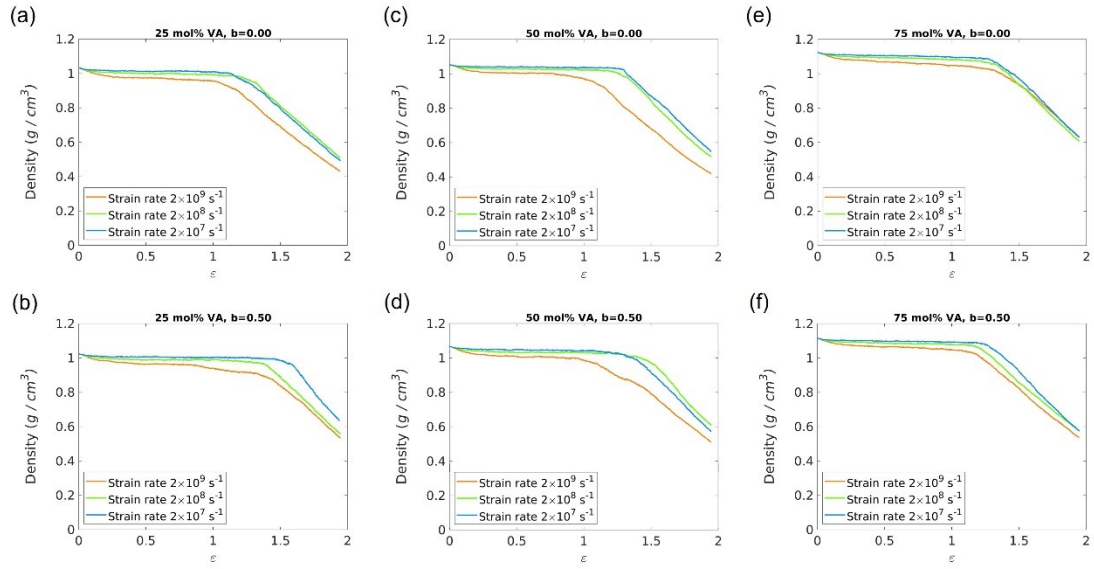


Figure S5. Density-strain curves of PVB copolymers system containing 25 mol % VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol % VA, $b=0.00$ (c) and $b=0.50$ (d); 75 mol % VA, $b=0.00$ (e) and $b=0.50$ (f) under different uniaxial strain rates at $T=350$ K.

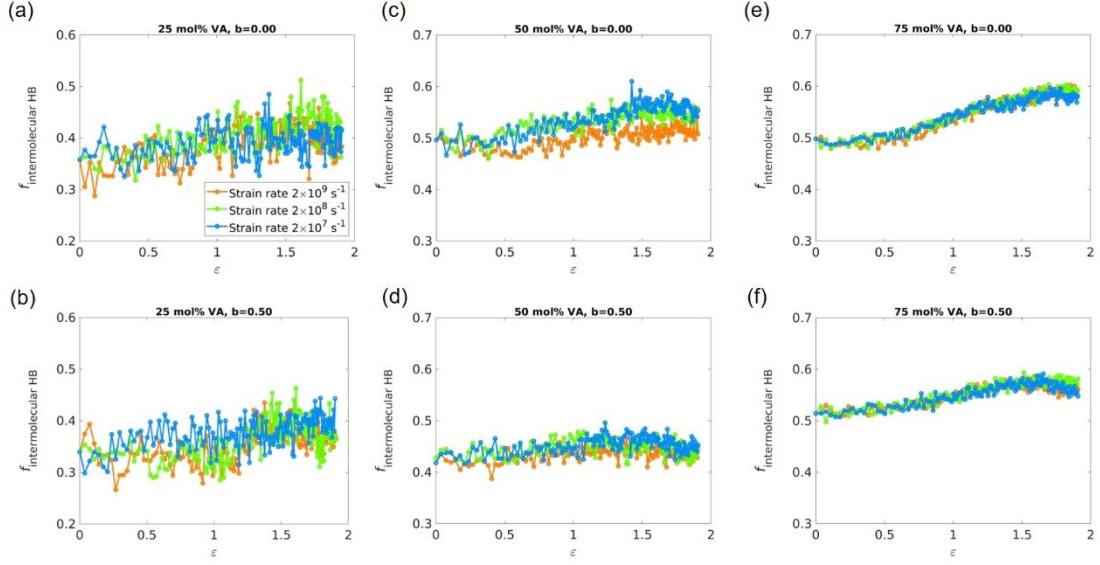


Figure S6. The fraction of intermolecular hydrogen bonds versus applied strain for PVB copolymers system with 25 mol% VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol% VA, $b=0.00$ (c) and $b=0.50$ (d); 75 mol% VA, $b=0.00$ (e) and $b=0.50$ (f) under different uniaxial strain rates at $T=350$ K.

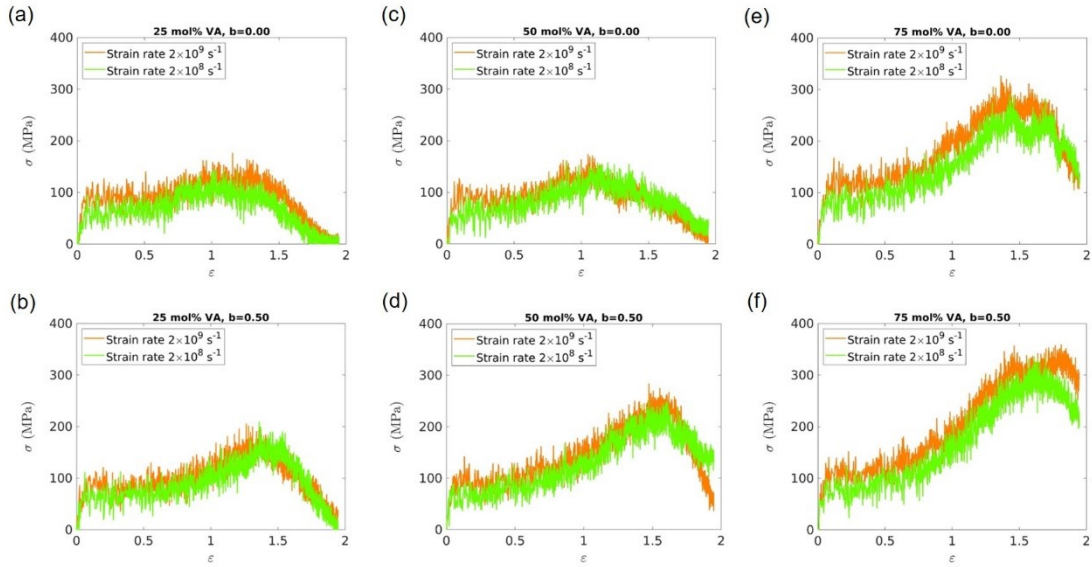


Figure S7. Stress-strain behavior of PVB copolymer replica systems containing 25 mol% VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol% VA, $b=0.00$ (c) and $b=0.50$ (d); 75 mol% VA, $b=0.00$ (e) and $b=0.50$ (f) under different uniaxial strain rates at $T=350$ K.

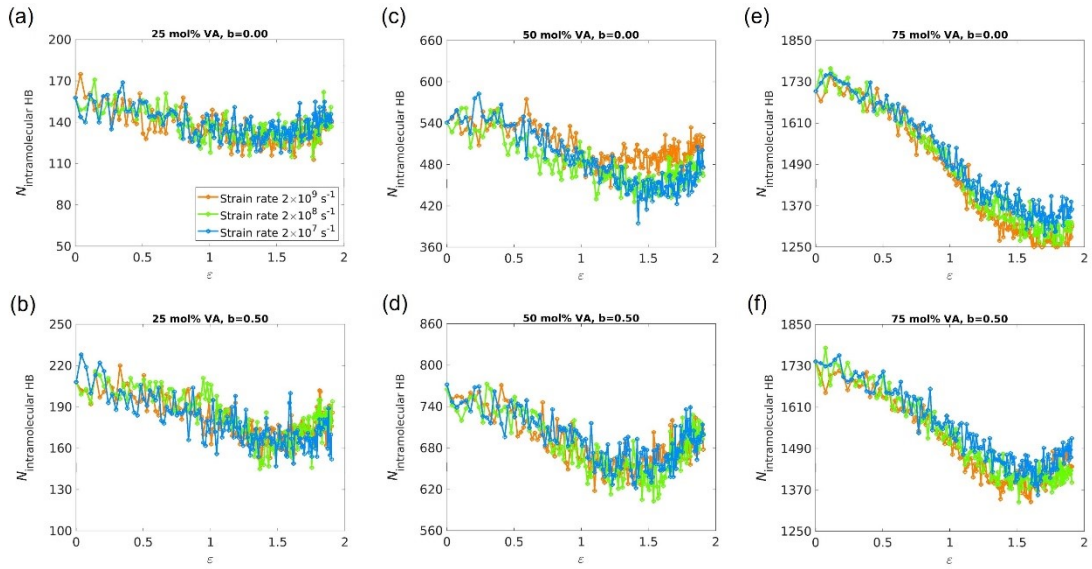


Figure S8. The number of intramolecular hydrogen bonds versus applied strain for PVB copolymers system with 25 mol% VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol% VA, $b=0.00$ (c) and $b=0.50$ (d); 75 mol% VA, $b=0.00$ (e) and $b=0.50$ (f) under different uniaxial strain rates at $T=350$ K.

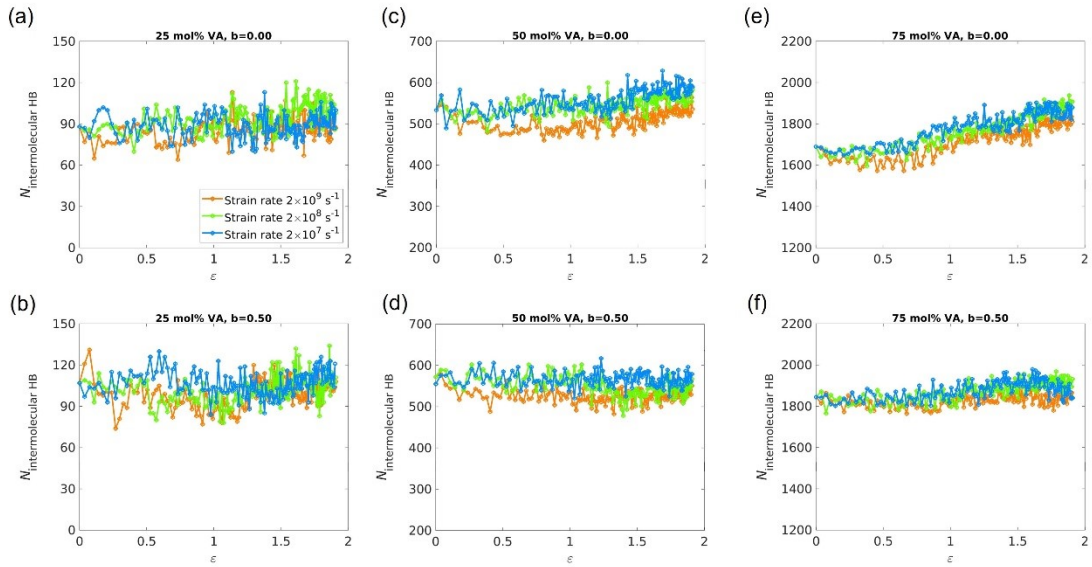
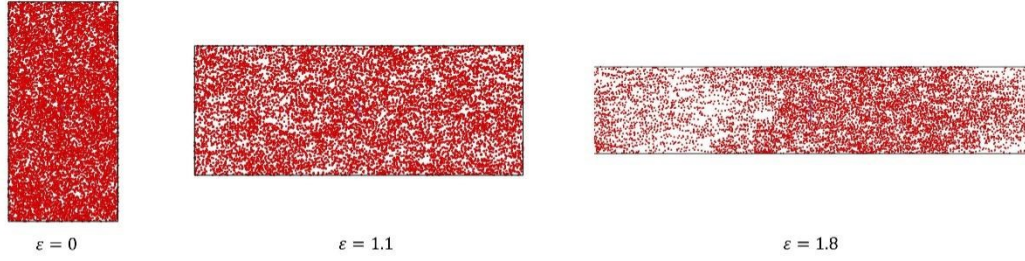


Figure S9. The number of intermolecular hydrogen bonds versus applied strain for PVB copolymers system with 25 mol% VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol% VA, $b=0.00$ (c) and $b=0.50$ (d); 75 mol% VA, $b=0.00$ (e) and $b=0.50$ (f) under different uniaxial strain rates at $T=350$ K.

(a) 75 mol % VA $b=0.00$ Strain rate $2 \times 10^9 \text{ s}^{-1}$



(b) 75 mol % VA $b=0.00$ Strain rate $2 \times 10^7 \text{ s}^{-1}$

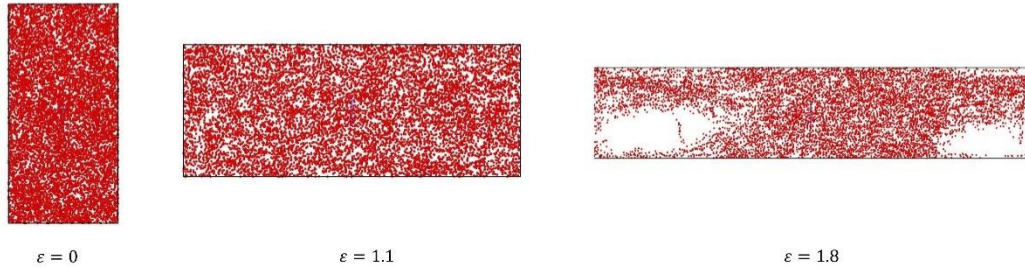
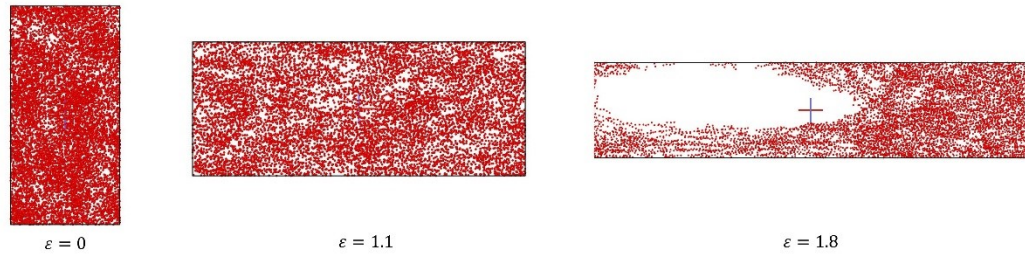


Figure S10. Snapshots of the VA beads at composition 75 mol % VA with $b=0.00$ under strain rate $2 \times 10^9 \text{ s}^{-1}$ (a) and $2 \times 10^7 \text{ s}^{-1}$ (b), respectively.

(a) 75 mol % VA $b=0.50$ Strain rate $2 \times 10^9 \text{ s}^{-1}$



(b) 75 mol % VA $b=0.50$ Strain rate $2 \times 10^7 \text{ s}^{-1}$

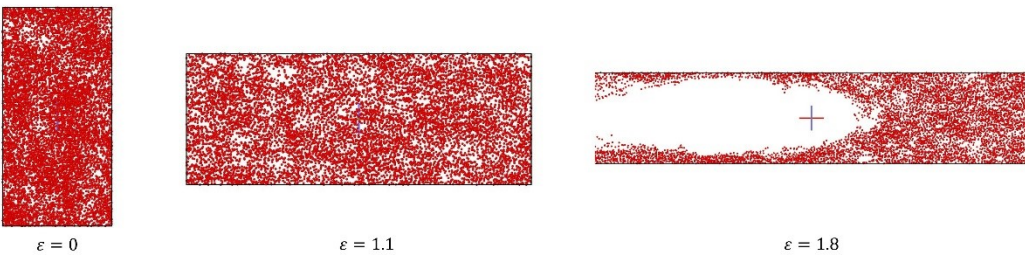


Figure S11. Snapshots of the VA beads at composition 75 mol % VA with $b=0.50$ under strain rate $2 \times 10^9 \text{ s}^{-1}$ (a) and $2 \times 10^7 \text{ s}^{-1}$ (b), respectively.

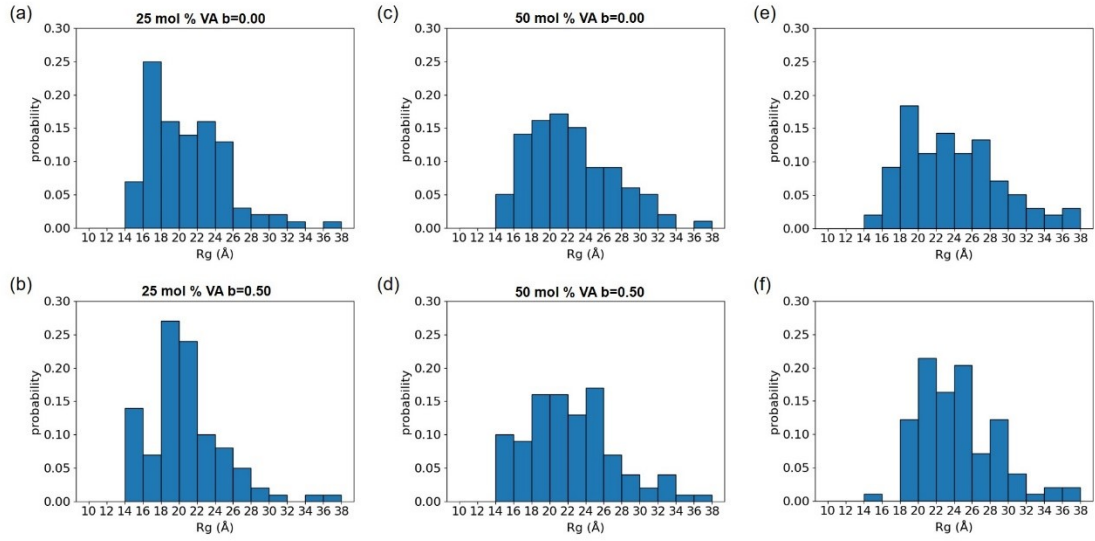


Figure S12. Probability distribution of radius of gyration of PVB copolymer replica systems in equilibrium states with 25 mol% VA, $b=0.00$ (a) and $b=0.50$ (b); 50 mol% VA, $b=0.00$ (c) and $b=0.50$ (d); 75 mol% VA, $b=0.00$ (e) and $b=0.50$ (f) at $T=350$ K.

mol% VA	b	$\bar{R}_g$ (Å)
25	0.00	20.95
25	0.50	20.80
50	0.00	22.51
50	0.50	22.48
75	0.00	24.19
75	0.50	24.61

Table S2. Mean radius of gyration ($\bar{R}_g$) of PVB copolymer replica systems at 350 K.