Supporting Information for: An Enhanced Scheme for Multiscale Modeling of Thermomechanical Properties of Polymer Bulks

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The same coarse-grained (CG) molecular dynamics (MD) simulations as in the main document have been carried out on the two polymer bulk systems (64MA100 and 64ST100) with partial charges switched off. Similar conclusions can be drawn: all four types of data (ρ-T, v-T, ln(ρ)-T, ln(v)-T) can be well described by the hyperbola and tanh functions (Equations 4 and 6 in the main document, respectively) so that the T_g, α_g and α_r are obtained; The results obtained using the two functions are close, and the T_g obtained by fitting to the ρ-T data represents the lower limit, and the T_g by fitting to the v-T data represents the upper limit, and the values of T_g obtained by fitting to the ln(ρ)-T and ln(v)-T data are close; And other properties obtained by fitting to the four types of data are also close.

As typical examples, the lnρ-T data during the cooling procedure are plotted in **Figure S1**, and the instantaneous P-lnV data at 300 K are presented in **Figure S2**. As similar to Tables 9-10 in the main document, the simulated thermomechanical properties are summarized and compared in **Table S1**.Among those, the B_{300} was obtained by linearly fitting to the P-lnv data from the equilibrium MD simulation at 300 K, and the T_g, α_g and α_r were obtained by fitting to the lnρ-T or lnv-T data with the hyperbola and tanh functions, and the ρ_{300} was obtained from the equilibrium MD simulation at 300 K. These results suggest that an inclusion of electrostatic

interactions tends to obviously reduce the T_g and ρ_{300} in different degrees for the two polymers whereas all other properties remain almost unchanged within the error bars.

As shown in **Table S2**, in essence, the electrostatic interactions are repulsive and the van der Waals interactions are attractive, and the van der Waals interactions dominate over the electrostatic interactions. As shown in **Table S3**, a concrete comparison is made on the small systems (50MA9 and 50 ST9) to appreciate the computational efficiency. For the same job (NPT MD at 500 K) of the same systems, it can be seen that the required computational times for the AA simulations are about 40 times than those for the CG simulations.

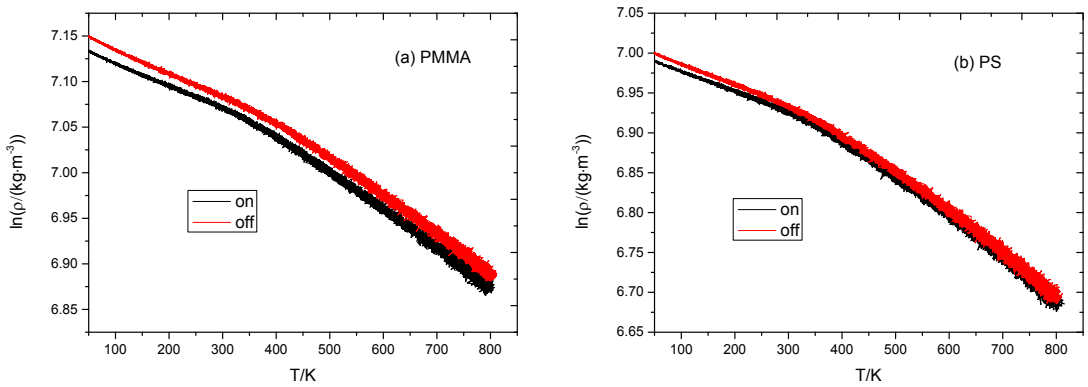

Figure S1. The simulated lnρ-T data of the two polymer systems (64MA100 and 64ST100) based on the current CG potentials with partial charges switched on and off

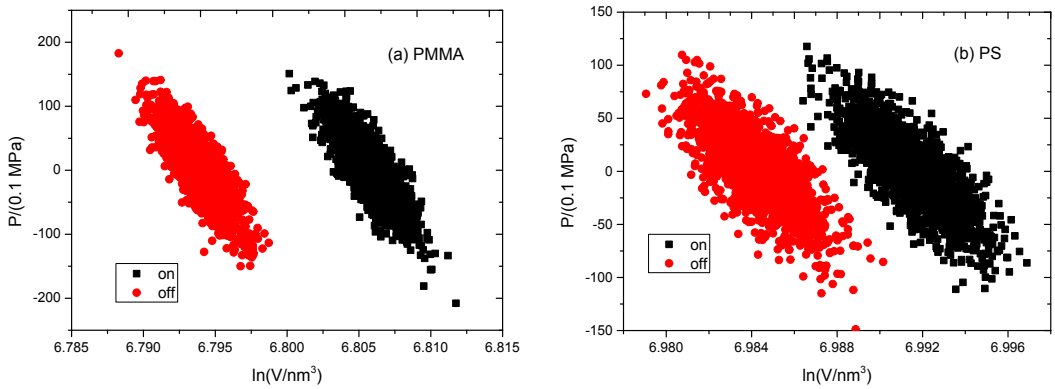

Figure S2. The simulated P-lnv data of the two polymer systems (64MA100 and 64ST100) based on the current CG potentials with partial charges switched on and off

Table S1. The simulated thermo-mechanical properties of the two polymer bulk systems (64MA100 and 64ST100) based on the current CG potentials with partial charges switched on and off, where T_g is the glass transition temperature, α_g and α_r are the volumetric expansive coefficients at the glassy and rubbery states, and ρ_{300} is the mass density, and B_{300} is the bulk modulus

Property, Unit	PMMA		PS	
	On	Off	On	Off
T_g, K	373-377	411-414	365-377	377-385
α_g, $\times 10^{-4}$ K^{-1}	2.3-2.5	2.5-2.6	1.7-2.7	1.9-2.8
α_r, $\times 10^{-4}$ K^{-1}	4.1-4.3	4.1-4.3	4.0-5.5	4.7-5.5
ρ_{300}, $g \cdot cm^{-3}$	1.178±0.002	1.192±0.002	1.018±0.002	1.026±0.002
B_{300}, GPa	2.62±0.04	2.69±0.04	1.68±0.03	1.61±0.03

Table S2. The various non-bonded interaction energies (unit: $KJ \cdot mol^{-1}$) of the two polymer bulk systems (64MA100 and 64ST100) at temperatures T=800 K and T=300 K based on the current CG potentials, where LJ-(SR) and Coulomb-(SR) denote the short-range van der Waals and electrostatic interactions, respectively, and Coul.-recip. represents the long-range electrostatic interactions in reciprocal space

Property	PMMA		PS	
	800 K	300 K	800 K	300 K
LJ-(SR)	-129041±27	-193139±48	-114571±32	-178594±14
Coulomb-(SR)	4009.8±1.1	4456±1	1400.6±0.8	1622.3±1.9
Coul.-recip.	19217±27	21714.5±3.3	9232±9	9921.9±6.1

Table S3. The computational efficiency defined by the duration times per core per day for the CG and AA simulations and the ratio between the two for the two small systems (50MA9 and 50ST9), where the data before "/" represent the duration time per day and the data after "/" represent the numbers of cores used for performing the simulations

Computational capability	PMMA	PS
Duration time (in ns) per day per core for AA simulations	47/24	45/24
Duration time (in ns) per day per core for CG simulations	635/8	532/8
Speedup ratio of CG to AA simulations	40.5	38.7