Supporting Information

Single Molecule Studies Reveal Temperature Independence of Lifetime of Dynamic Heterogeneity in Polystyrene

Alyssa S. Manz[1], Keewook Paeng[1,2], Laura J. Kaufman[1,*]

[1]Department of Chemistry, Columbia University, New York, NY 10027

[2]Department of Chemistry, Sungkyunkwan University, Suwon 16419, Republic of Korea

Temperature (K)	**373.0**	**378.1**	**379.6**	**383.6**	**Combined**
number of molecules	1164	1054	1095	1443	4756
median τ_c (s)	312.24	7.70	2.98	0.32	1.00
median τ_{fit} (s)	146.56	3.66	1.39	0.17	1.00
median β	0.51	0.51	0.51	0.54	0.51
median trajectory length ($\tau_{fit,med}$)	912	872	932	935	913
median frame rate (frames/τ_{fit})	20.93	18.27	20.72	20.60	20.13
FWHM (log (τ_c) distribution)	0.50	0.45	0.49	0.51	0.49
FWHM (log (τ_{fit}) distribution)	0.54	0.44	0.48	0.44	0.48
FWHM (β distribution)	0.33	0.32	0.29	0.31	0.30

Table S1. Characteristics of data obtained from longest trajectory pPDI measurements in polystyrene and presented graphically in Figure 1. The combined data is normalized by median τ_c or τ_{fit} before further analysis.

Temperature (K)	trajectory length (τ_{fit})	number of molecules	median β	τ_{fit} distribution FWHM
373.0	18	1105	1.04	1.17
	23	1092	0.97	1.04
	46	1059	0.82	1.08
	91	1101	0.70	0.91
	228	1019	0.58	0.73
	456	1298	0.53	0.54
	912	1164	0.51	0.54
378.1	18	1101	1.01	0.94
	20	1174	0.98	0.95
	41	1034	0.83	0.86
	87	1174	0.71	0.78
	205	1090	0.59	0.67
	461	993	0.54	0.57
	872	1054	0.51	0.44
379.6	19	1216	1.05	1.03
	23	1344	0.98	1.03
	47	1343	0.79	0.95
	93	1501	0.69	0.81
	233	1652	0.58	0.66
	466	1129	0.55	0.56
	932	1095	0.51	0.48
383.6	19	1007	1.07	1.08
	23	1016	1.04	1.00
	47	1020	0.87	0.91
	94	1011	0.76	0.77
	230	1212	0.60	0.65
	460	1301	0.56	0.53
	935	1443	0.54	0.44

Table S2. Characteristics of data obtained from trajectory length-dependent pPDI measurements in polystyrene, presented graphically in Figure 2.

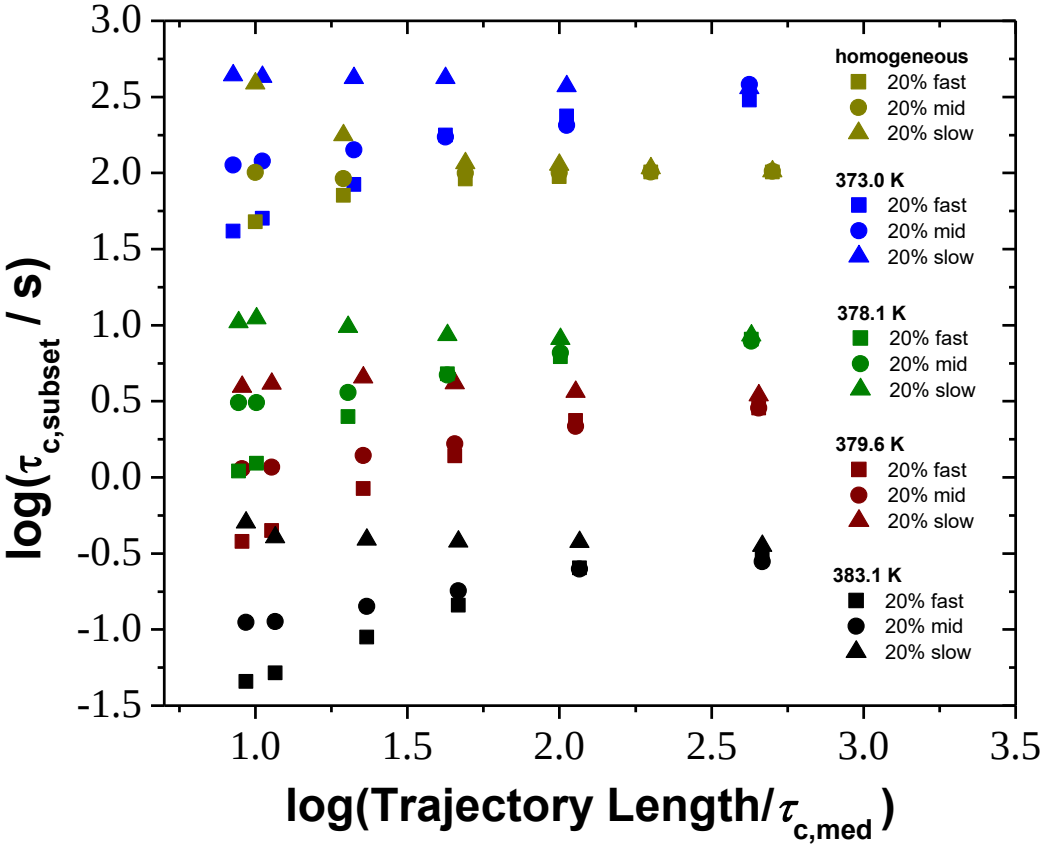

Figure S1. Median τ_c values as a function of trajectory length for the 20% initially fastest (squares), initially slowest (triangles), and middle (circles) of the initial relaxation time sub-ensembles at each temperature studied, as well as for a homogeneous simulation.

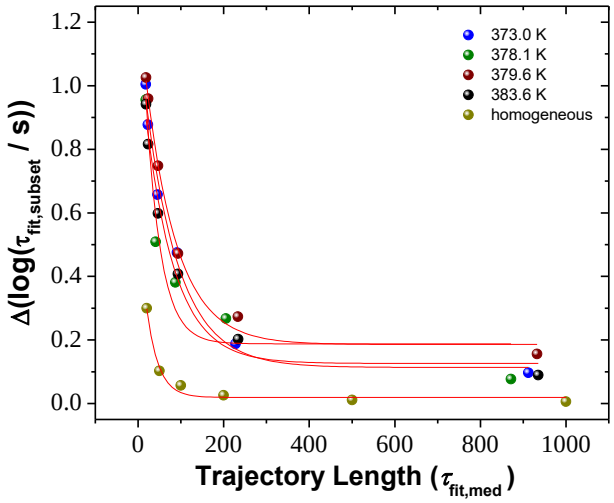

Figure S2. Exponential fits to the convergence of fast and slow sub-ensembles of molecules as a function of trajectory length, as presented in Figure 3(b) in the main text.

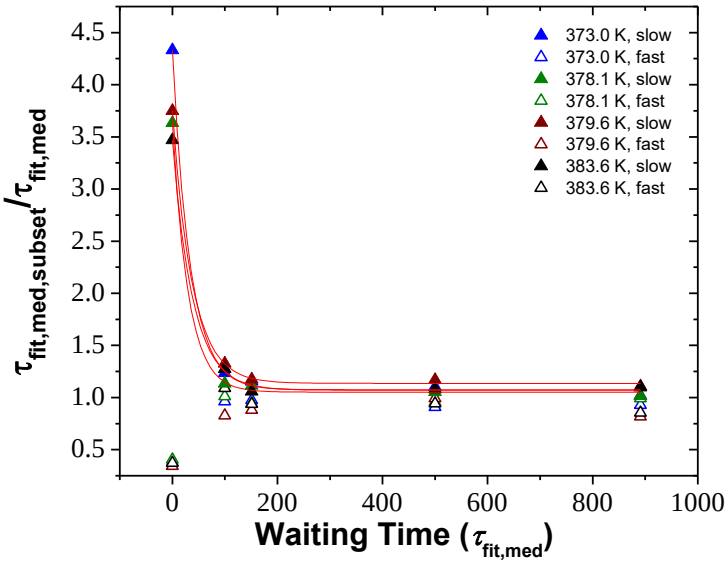

Figure S3. Exponential fits to the slow subsets of molecules analyzed via a waiting time approach, as shown in Figure 4 of the main text.

Videos S1-S4. Four hundred frame movies, each played back at 7 Hz, collected at (**S1**) 373.0 K, (**S2**) 378.1 K, (**S3**) 379.6 K, and (**S4**) 383.6K. Frame rates of data collection were 0.143 Hz (373.0 K), 5.0 Hz (378.1 K), 14.8 Hz (379.6 K), and 124.8 Hz (383.6K).