Supporting Information for

Cloud Point Driven Dynamics in Aqueous Solutions of Thermoresponsive Copolymers: Are They Akin to Criticality Driven Solution Dynamics?

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Table S1. Results from the synthesis of copolymers, **P1-P3** at 70 °C in DMF for 4.5 h and their aqueous solutions properties.

Polymer	MMA content in feed	Conv[a] (%)	MMA content in copolymer[b]	$M_{n,SEC}$[c] (g/mol)	$Đ$[c]	$M_{n,NMR}$[d] (g/mol)	$M_{n,theo}$[e] (g/mol)	Cloud point (°C)	
								T_{cp}[f]	T_{cp}[g]
P1	40	58	51	12300	1.12	12400	6660	50	50
P2	50	63	55	11150	1.14	10400	6580	43	35
P3	60	58	63	10160	1.15	11600	5500	28	25

[a]Determined gravimetrically. [b]Determined from ^{1}H NMR analysis. [c]Measured by SEC in THF. [d] Calculated by ^{1}H NMR from the integration ratio of the repeating unit protons to that of the polymer chain end protons. [e]$M_{n,theo}$ = (([monomer]/[CTP] × average molecular weight (MW) of monomer × conversion (Conv)) + MW of CTP). [f]Measured from UV-vis spectrometer. [g]Determined using DLS.

Table S2: Temperature dependent density (ρ), viscosity coefficient (η) of aqueous solutions (2 mg/mL) of three copolymers **P1-P3**.

T/K	**P1**		**P2**		**P3**		Water	
	ρ/gcm^{-3}	η/mPas	ρ/gcm^{-3}	η/mPas	ρ/gcm^{-3}	η/mPas	ρ/gcm^{-3}	η/mPas
293	0.9986	1.0564	0.9985	1.0457	0.9982	1.0507	0.9982	1.0016
298	0.9974	0.9412	0.9974	0.9326	0.9966	0.9416	0.9970	0.8900
303	0.9960	0.8473	0.9960	0.8394	0.9951	0.8598	0.9956	0.7972
308	0.9944	0.7680	0.9943	0.7622	-	-	0.9940	0.7191
313	0.9925	0.7020	0.9924	0.7032	-	-	0.9922	0.6527
318	0.9905	0.6468	-	-	-	-	0.9902	0.5958
323	0.9883	0.6071	-	-	-	-	0.9881	0.5470

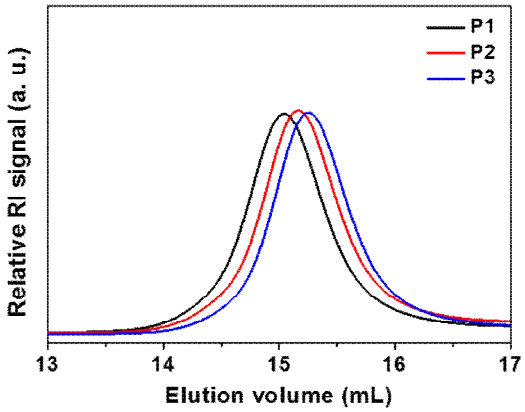

Figure S1. SEC traces of the copolymers, **P1-P3**.

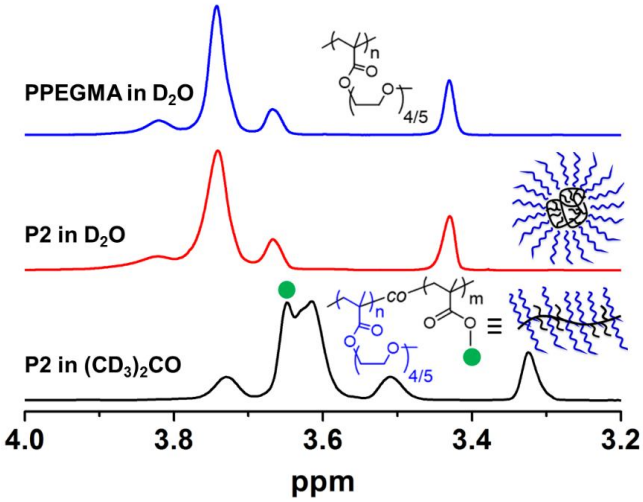

Figure S2. ^{1}H NMR spectra of P2 in acetone-d_6 (black line), P2 in D_2O (red line) and PPEGMA in D_2O (blue line).

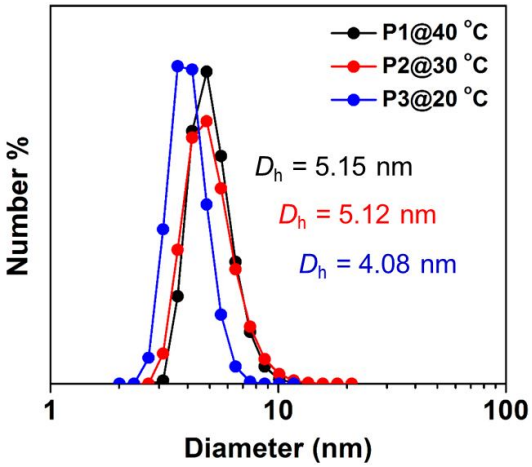

Figure S3. DLS graph of **P1**, **P2** and **P3** below their respective T_{cp}.

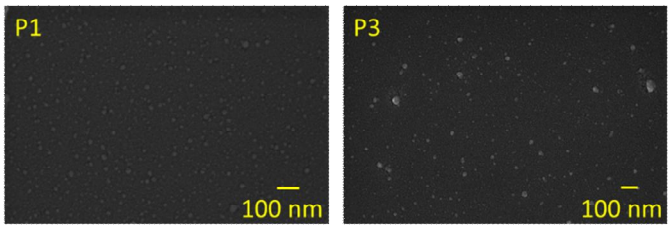

Figure S4. FE-SEM images of **P1** and **P3**.

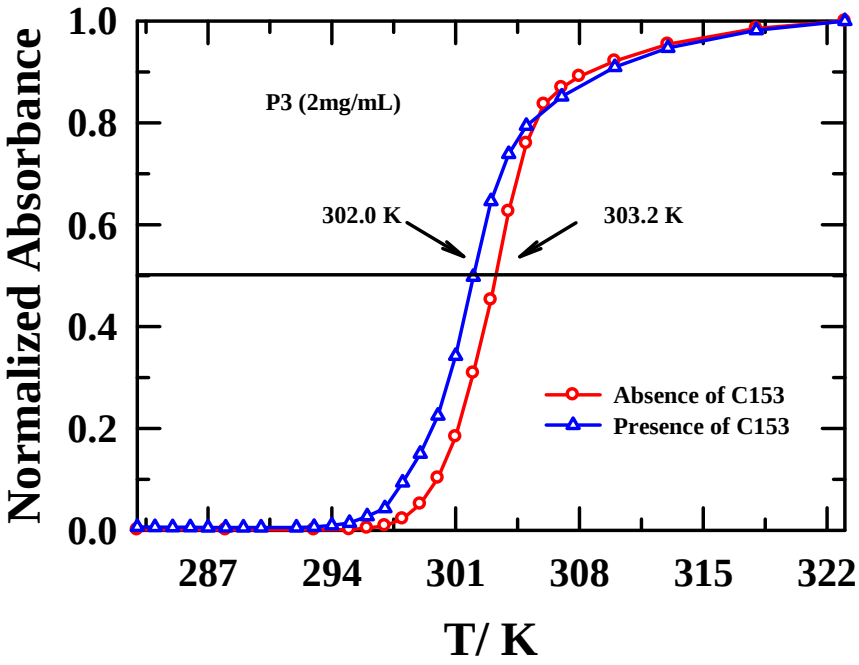

Figure S5. Impact of third component (C153) in T_{cp} determination: temperature dependent absorbance of the aqueous solution (2.0 mg/mL) of the random copolymer, **P3** in presence and absence of C153. Respective T_{cp} for each case are indicated with arrows.

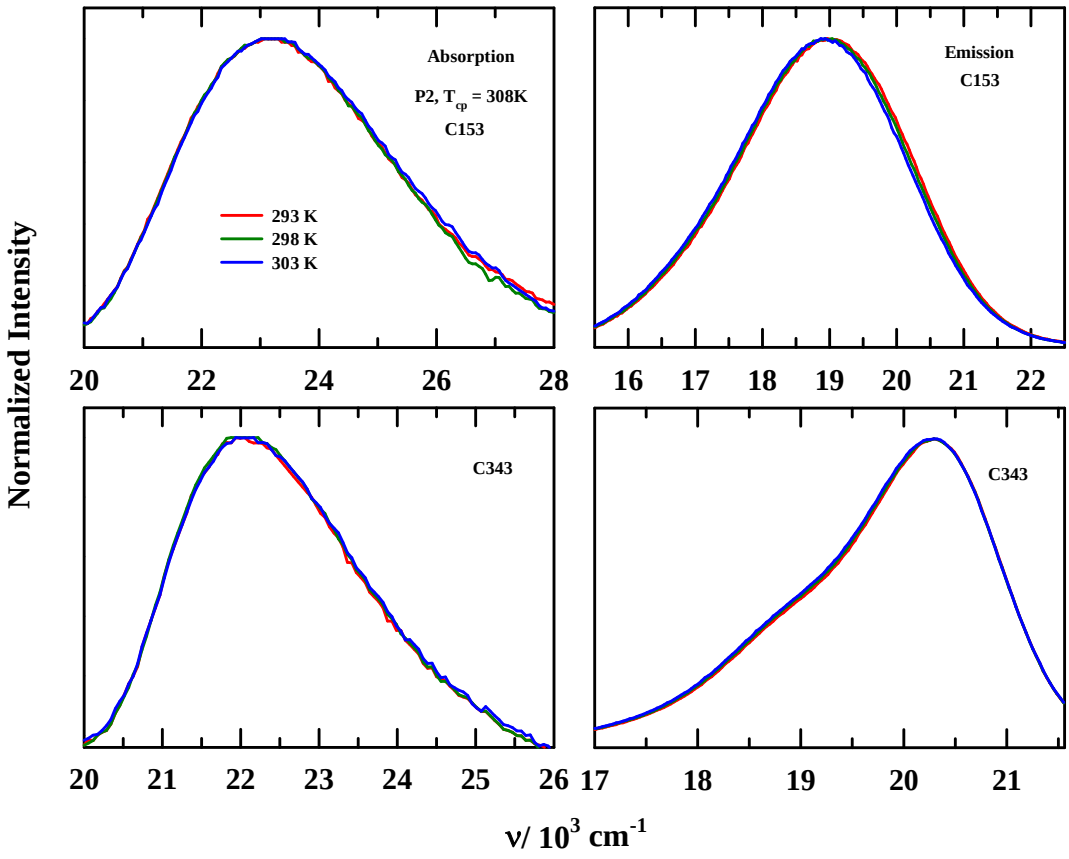

Figure S6. Representative temperature dependent UV-Vis absorption (left panels), and steady state fluorescence emission spectra (right panels) of C153 (upper panels), and C343 (lower panels) in aqueous solutions of **P2**. All representations are colour-coded.

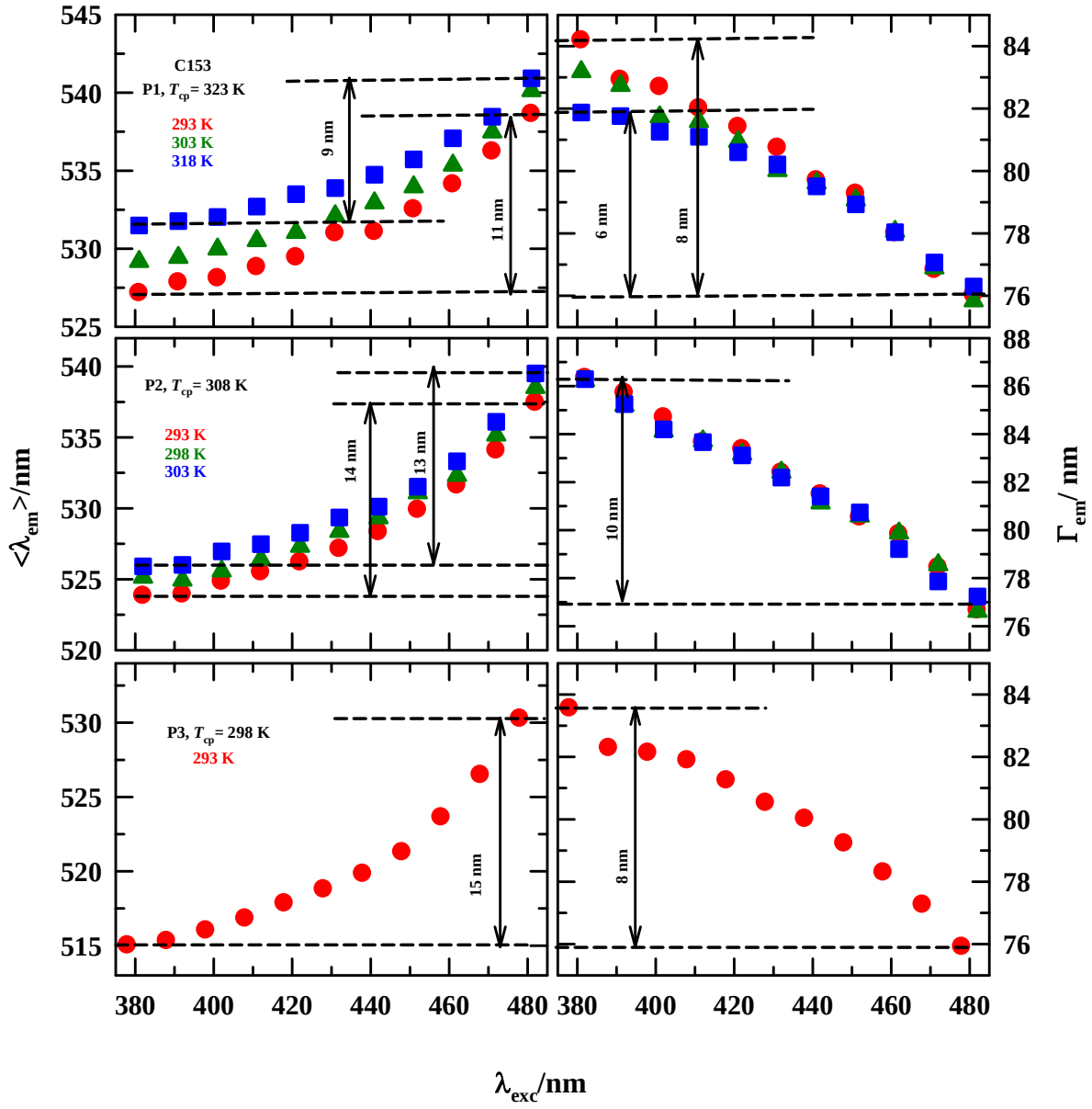

Figure S7. Excitation wavelength dependence (λ_{exc}) of steady state average peak emission wavelength ($<\lambda_{em}>$) (left panels) and spectral widths (FWHM, Γ_{em}) (right panels) of C153 in aqueous **P1**, **P2** and **P3** solutions at different temperatures. All representations are colour coded. Note the information contained in this figure is equivalent to that displayed by Fig. 7 in the main text because $\nu = 1/\lambda$.

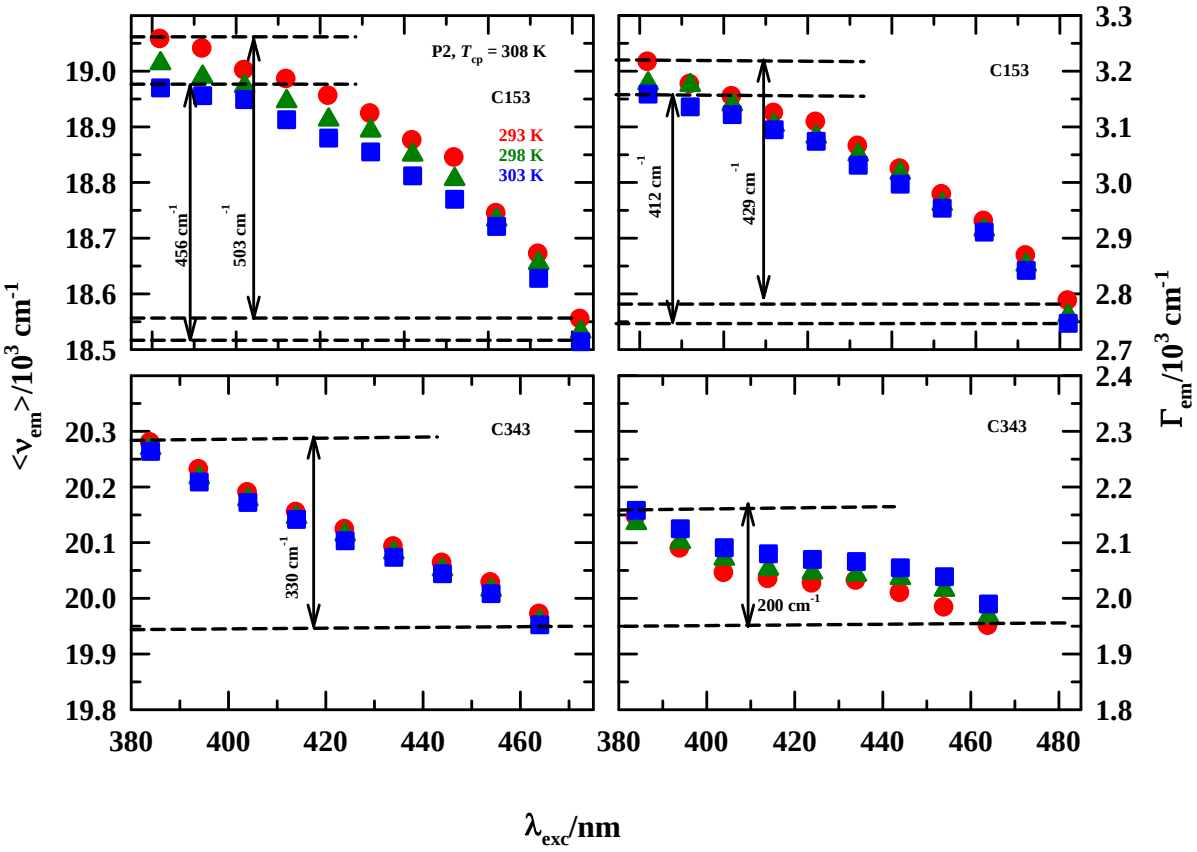

Figure S8. Excitation wavelength dependence (λ_{exc}) of steady state average peak emission frequencies ($<\nu_{em}>$) (left panels) and spectral widths (FWHM, Γ_{em}) (right panels) of C153 (upper panels) and C343 (lower panels) in aqueous **P2** solutions at different temperatures. All representations are colour-coded.

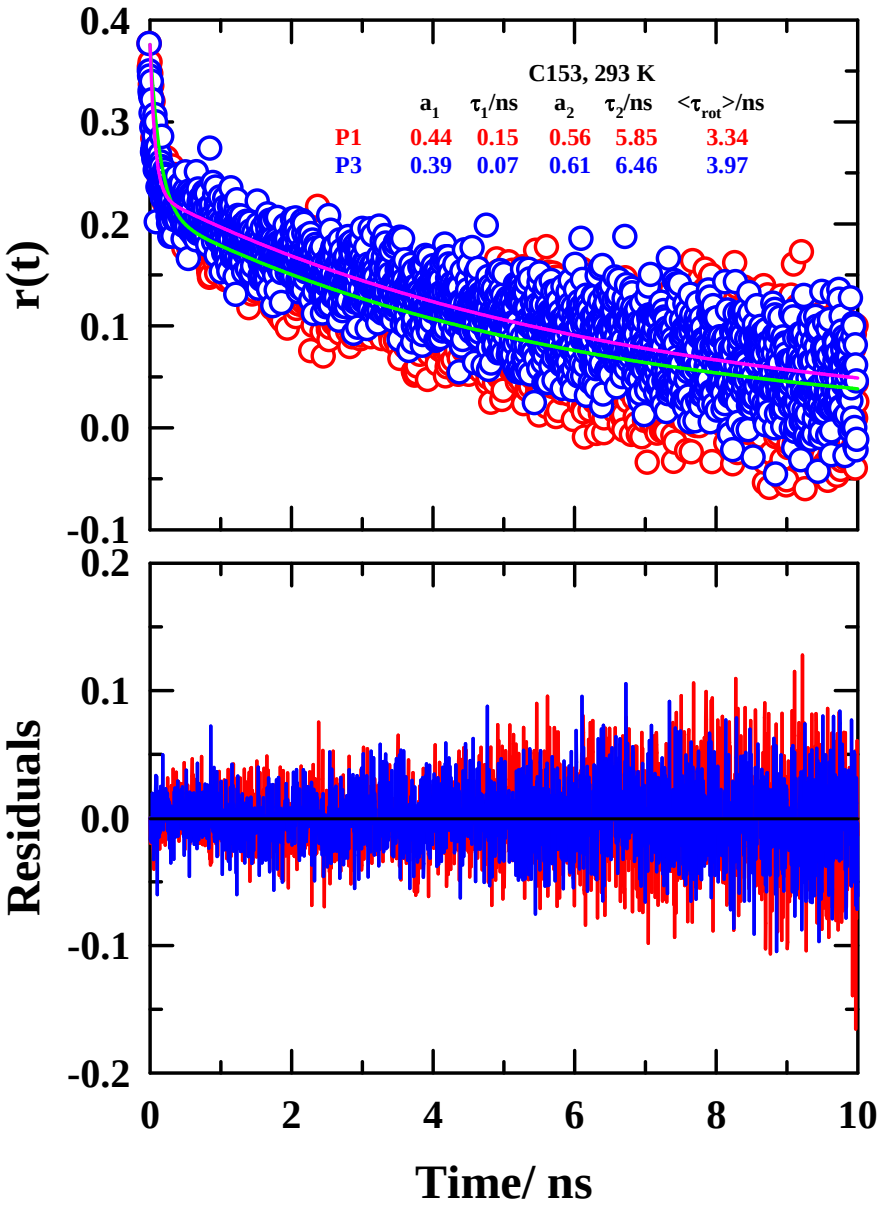

Figure S9. Representative rotational anisotropy, r(t), decays of C153 in aqueous **P1** and **P3** solutions (upper panel) and corresponding residuals (lower panel) at 293 K. Experimental data are shown by circles; solid lines going through data represent bi-exponential fits . Fit parameters are shown in the inset. All representations are colour-coded.

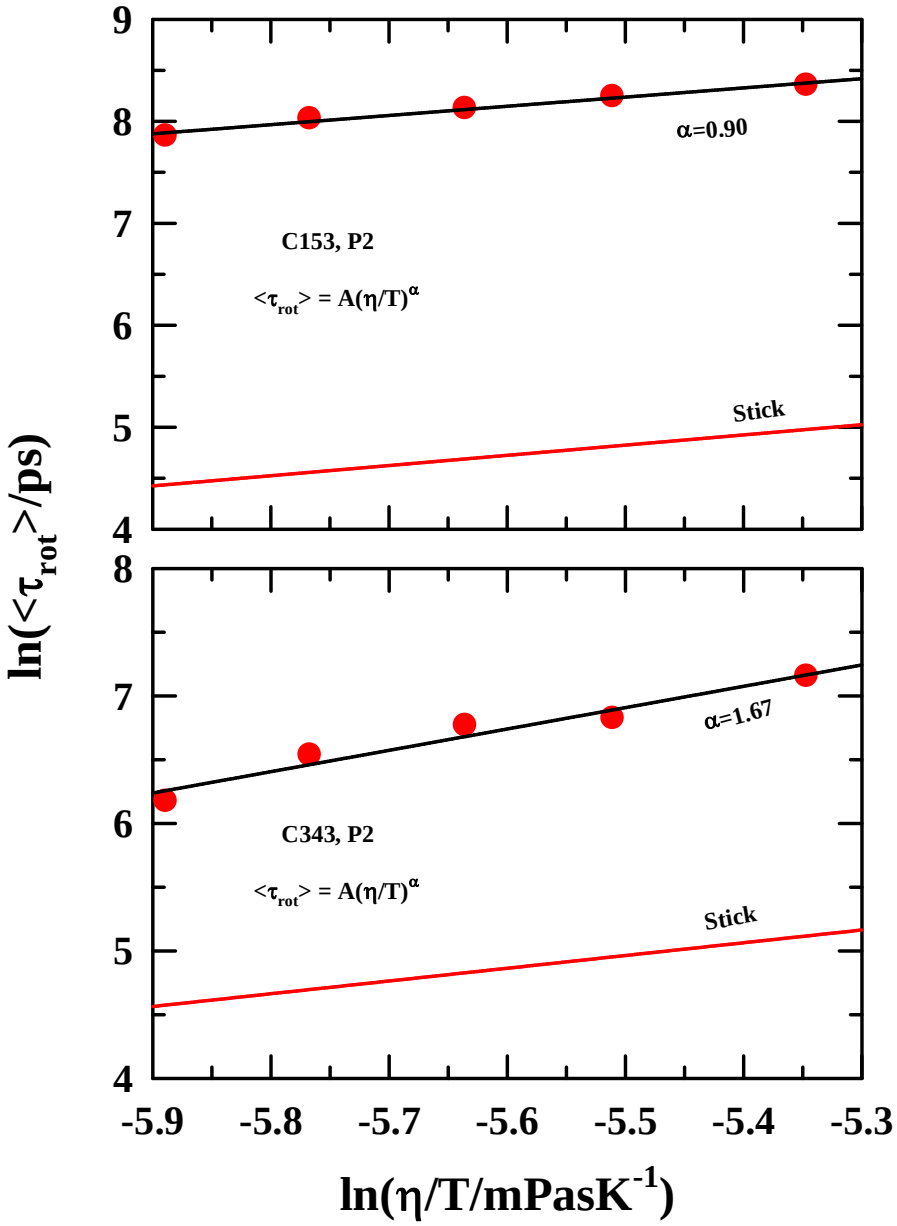

Figure S10. Viscosity coupling of solute rotation in aqueous solution of **P2** as observed from temperature dependent measurements, and a comparison with the hydrodynamic prediction.