## **Supporting Information**

## How Does Microstructural Design Affect the Dynamics and

## **Rheology of Segmented Polyurethanes?**

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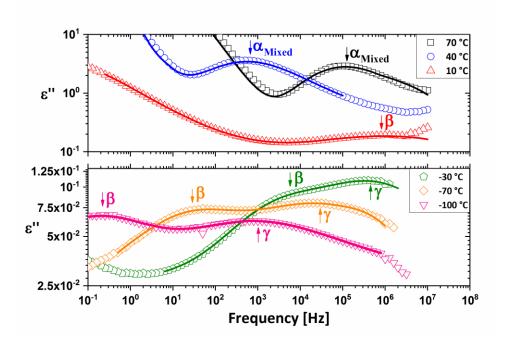


Figure S1. Dielectric loss spectrum of PU-425-H40 in different temperatures. Solid lines stand for the fits to the Havriliak–Negami equation.

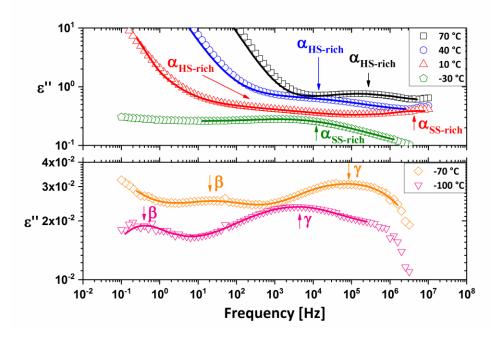


Figure S2. Dielectric loss spectrum of PU-2000-H40 in different temperatures. Solid lines stand for the fits to the Havriliak–Negami equation.

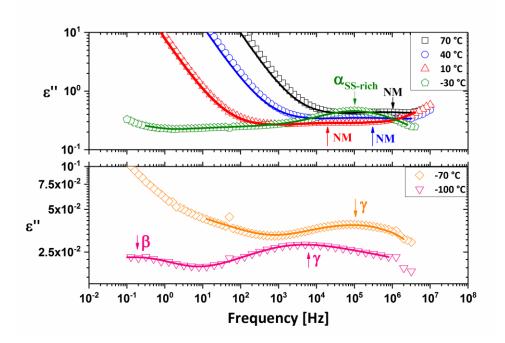


Figure S3. Dielectric loss spectrum of PU-4000-H40 in different temperatures. Solid lines stand for the fits to the Havriliak–Negami equation.

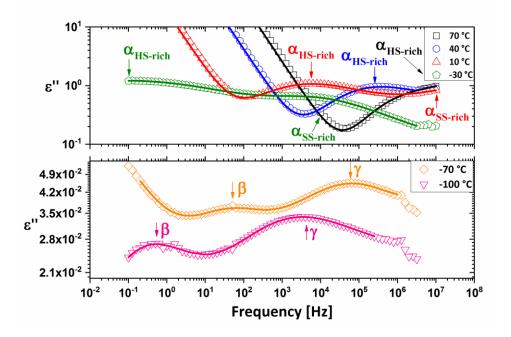


Figure S4. Dielectric loss spectrum of PU-2000-H20 in different temperatures. Solid lines stand for the fits to the Havriliak–Negami equatio

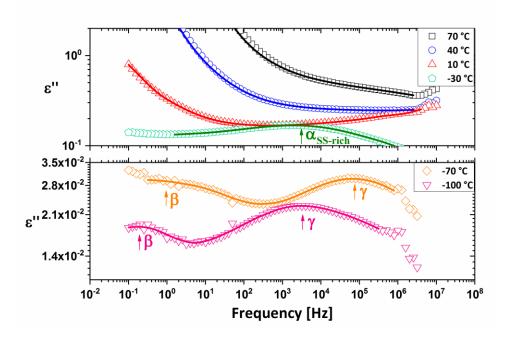


Figure S5. Dielectric loss spectrum of PU-2000-H60 in different temperatures. Solid lines stand for the fits to the Havriliak–Negami equation.

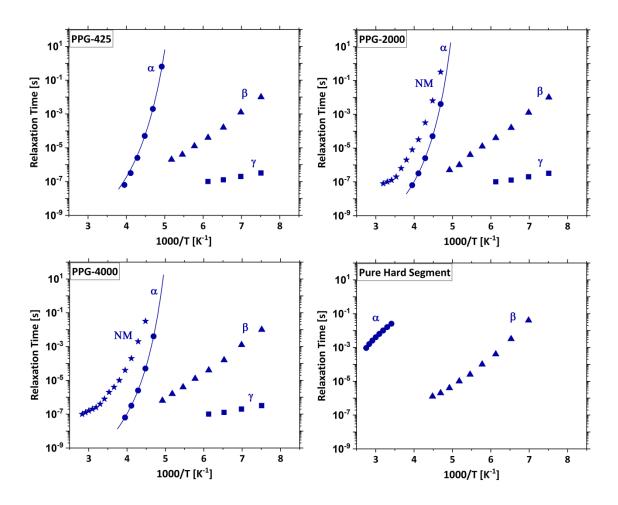


Figure S6. Characteristic relaxation times versus inverse temperature for the pure soft and hard segments. The fitting based on VFT equation was shown with solid lines.

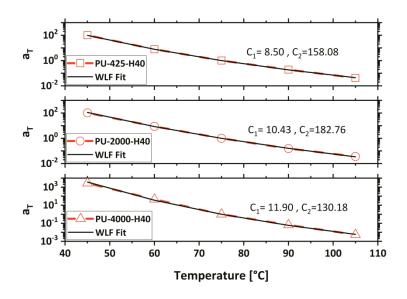
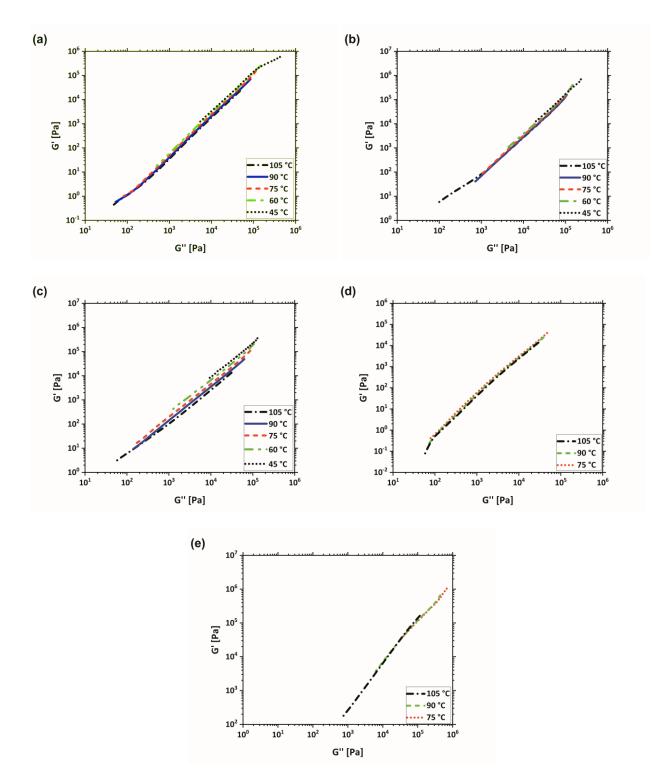
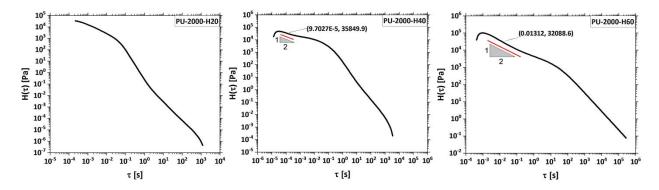


Figure S7. The used Shift factors of time-temperature superposition in figure 10.



**Figure S8.** Rheological Cole-Cole plot for: (a) PU-425-H40; (b) PU-2000-H40; (c) PU-4000-H40; (d) PU-2000-H20; and (d) PU-2000-H60 at different temperature



**Figure S9.** Relaxation spectra for the model PUs with similar soft segments at 90 °C. The selected points were used in Bueche–Ferry law.

Table S1 Segmental Rouse times in sec and friction coefficients in kg/s resulted from segmental
relaxation processes of pure segments and PPG-2000-based PUs at 90 °C.

Sample	$\alpha_{\rm SS}$		$\alpha_{\rm HS}$		$lpha_{ m SS-rich}$		a <sub>HS-rich</sub>	
	$ au_S$	ξss	$ au_S$	$\xi_{HS}$	$ au_S$	$\xi_{SS-rich}$	$ au_S$	$reve{\xi}_{HS ext{-rich}}$
PPG-425	1.81E-10	1.70E-11						
PPG-2000	1.08E-10	1.02E-11						
PPG-4000	1.08E-10	1.02E-11						
Pure HS			9.33E-4	3.32E-5				
PU-2000-H20					1.09E-9	1.50E-11	1.58E-8	2.16E-10
PU-2000-H40					1.42E-10	1.94E-12	4.98E-7	6.79E-9
PU-2000-H60					3.99E-10	5.44E-12	1.01E-3	1.38E-5