Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Supplementary Information for:

Durable and Thermally Switchable Polysilsesquioxane Adhesive via Dynamic Covalent Bond: Effect of Crosslinker on Reversible Chemistry

Seung Hyun Kang [a,b,1], Jin Young Seo [a,1], Hyeon Ji Oh [a], Jung-Hyun Lee [b], Albert S. Lee [a], and Kyung-Youl Baek [a,c,d,*]

[a] Materials Architecturing Research Center, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea.
[b] Department of Chemical and Biological Engineering, Korea University, Seoul 02481, Republic of Korea.
[c] Division of Nano & Information Technology, KIST School, Korea University of Science and Technology, Seoul 02792, Republic of Korea.
[d] KHU-KIST Department of Converging Science and Technology, Kyung Hee University, Seoul 02447, Republic of Korea.

[1] These authors contributed equally to this work.

Corresponding Author's e-mail: baek@kist.re.kr (K.-Y. Baek)

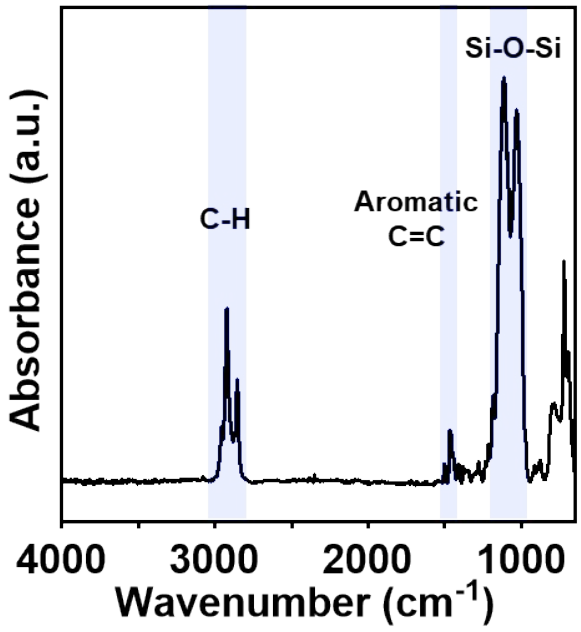

Figure S1. FT-IR spectrum of ladder-like poly(silsesquioxane) with octyl and pyrrole functional groups.

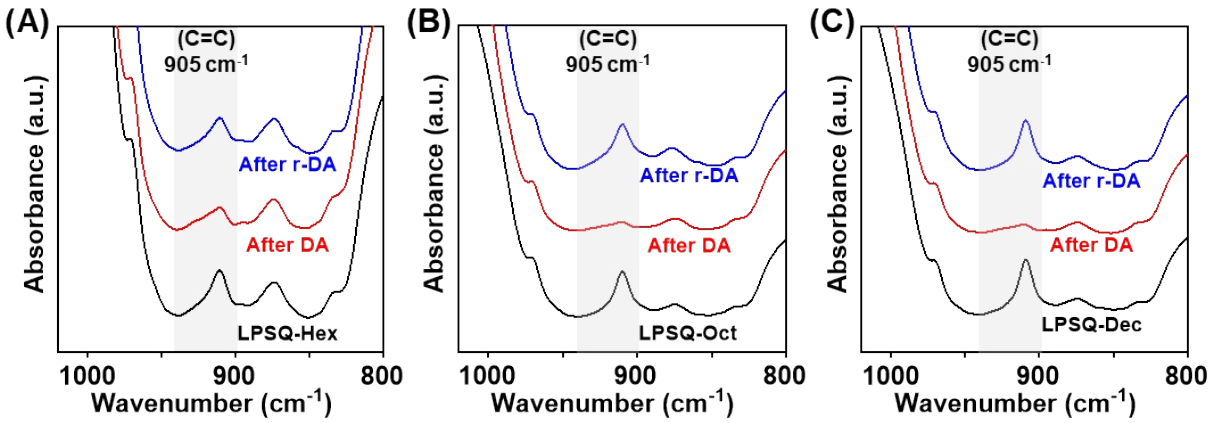

Figure S2. FT-IR spectra of (A) LPSQ-Hex, (B) LPSQ-Oct, (C) LPSQ-Dec before and after thermal annealing. (DA reaction and rDA reaction were performed at 90 ˚C and 135 ˚C respectively)

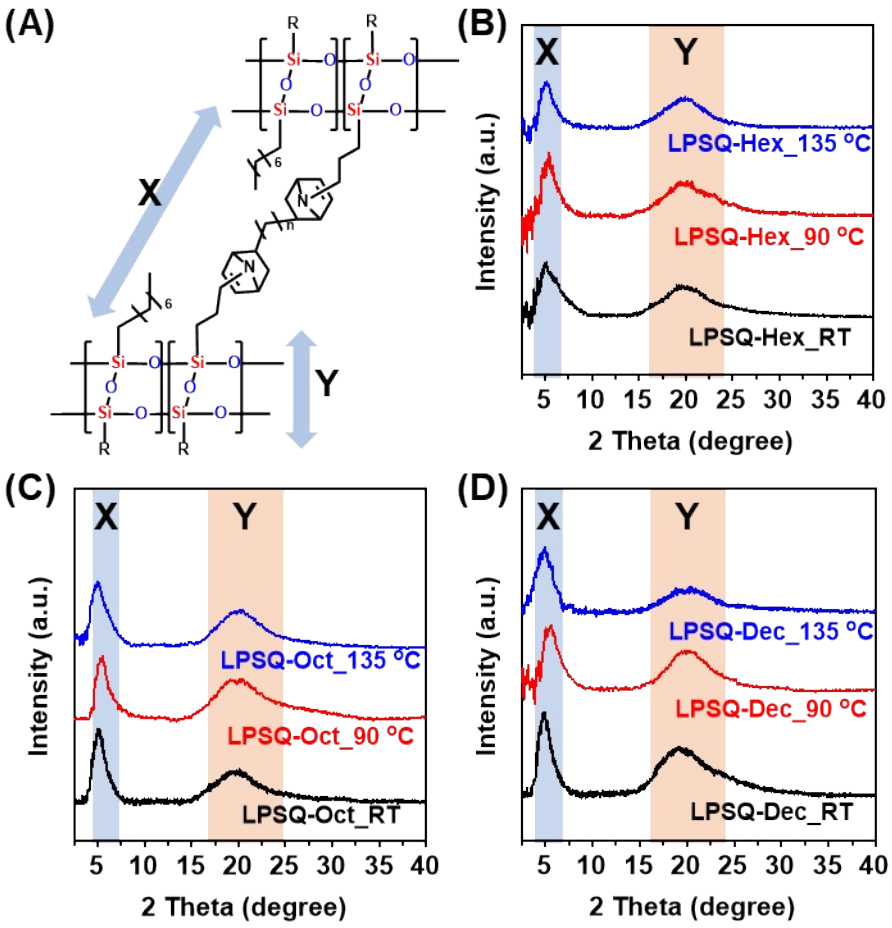

Figure S3. (A) Chemical structure of crosslinked LPSQ adhesives, XRD patterns of (B) LPSQ-Hex, (C) LPSQ-Oct, (D) LPSQ-Dec before and after thermal annealing (DA reaction and rDA reaction were performed at 90 °C and 135 °C respectively.)

Table S1. Molar volume calculation of LPSQ adhesives by Bondi's group contribution method.

	Group	V_w (cm^3/mol)	Frequency			Total V_w (cm^3/mol)		
Octyl group	Si (Tetravalent)	16.6	2			33.2		
	-O-	5.5	3			16.5		
	$-CH_2-$	10.23	14			143.22		
	$-CH_3$	13.67	2			27.34		
Pyrrole group	Si (Tetravalent)	16.6	2			33.2		
	-O-	5.5	3			16.5		
	$-CH_2-$	10.23	8	10	12	81.84	102.3	122.76
	-N< (Trivalent)	4.3	2			8.6		
	Cyclohexene	53.36	2			106.72		

Table S2. Polymer-solvent interaction parameter calculation of LPSQ adhesives using Hansen solubility parameter.

Entry	Sample	Solubility parameter ($MPa^{0.5}$) [a]			Distance ($MPa^{0.5}$) [b]	χ
		δ_d	δ_p	δ_h		
1	LPSQ-Hex	17.706	1.425	5.084	3.14	0.322
2	LPSQ-Oct	17.712	1.399	5.038	3.092	0.319
3	LPSQ-Dec	17.725	1.375	4.993	3.043	0.313

[a] Hansen solubility parameter was calculated by Hoftyzer-Van Krevelen method (solubility parameter components could be predicted from group contributions); [b] Distance between points in Hansen space

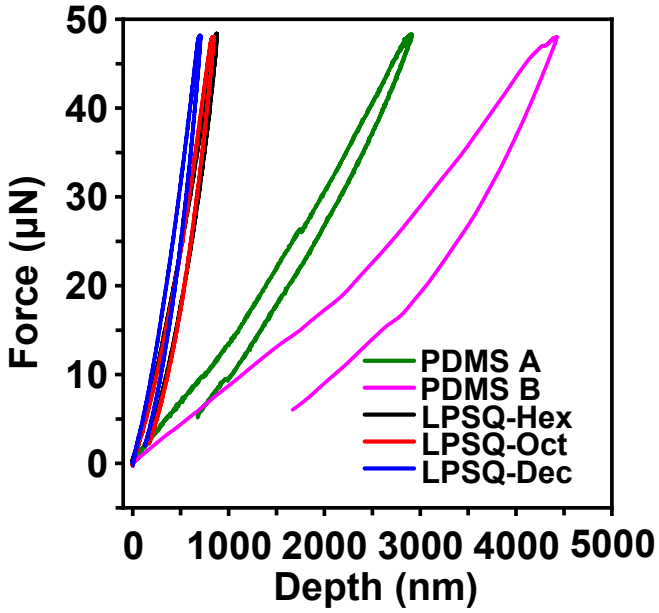

Figure S4. Force and depth profiles of PDMS and LPSQ adhesives measured by nano-indentation.