Supporting Information

A humidity-resistant bio-inspired microfibrillar adhesive fabricated by a phenyl-rich polysiloxane elastomer for reliable skin patches

Shuang Xia, Yukun Chen, Wenxin Fu, Jinfeng Tian, Yilin Zhou, Yini Sun, Ruoxuan Cao, Huawei Zou, Mei Liang*

Shuang Xia, Yukun Chen, Jinfegn Tian, Yilin Zhou, Yini Sun, Ruoxuan Cao, Huawei Zou, Mei Liang

The State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China

Wenxin Fu

Key Laboratory of Science and Technology on High-tech Polymer Materials, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China

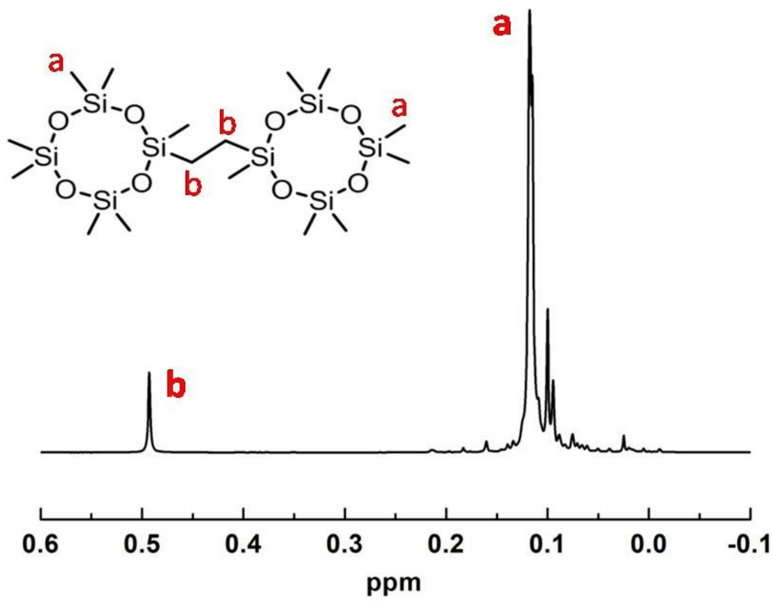

Figure S1. 1H-NMR of Bis-D4. 600 MHz, $CDCl_3$, δ = 0.49 ppm (4H, 2 $-CH_2-$), 0.13-0.08 ppm (42H, 14 $-CH_3$).

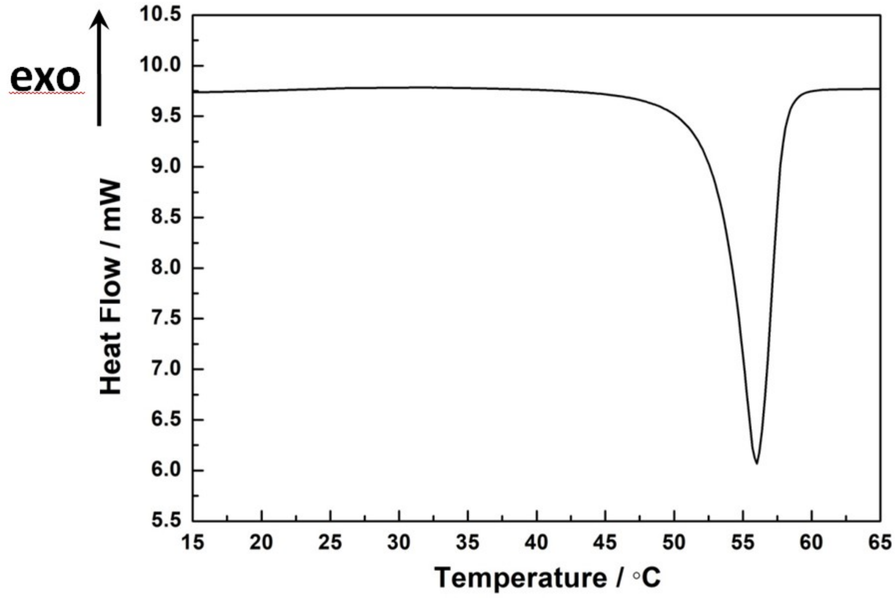

Figure S2. DSC curves of Bis-D4.

Table S1. Formulations of the synthetic polysiloxane with various phenyl contents.

Elastomers	P2 / mol [a)] (544.9 g/mol)	D4 / mol (296.6 g/mol)	TMAH / mol (181.0g/mol)	Bis-D4 / mol (619.2 g/mol)
PDMS	-	0.0337	0.00019	0.00067
PDMS-co-PPMS-30%	0.0081	0.0189	0.00027	0.00081
PDMS-co-PPMS-60%	0.0135	0.0090	0.00022	0.00089
PDMS-co-PPMS-90%	0.0170	0.0019	0.00019	0.00113

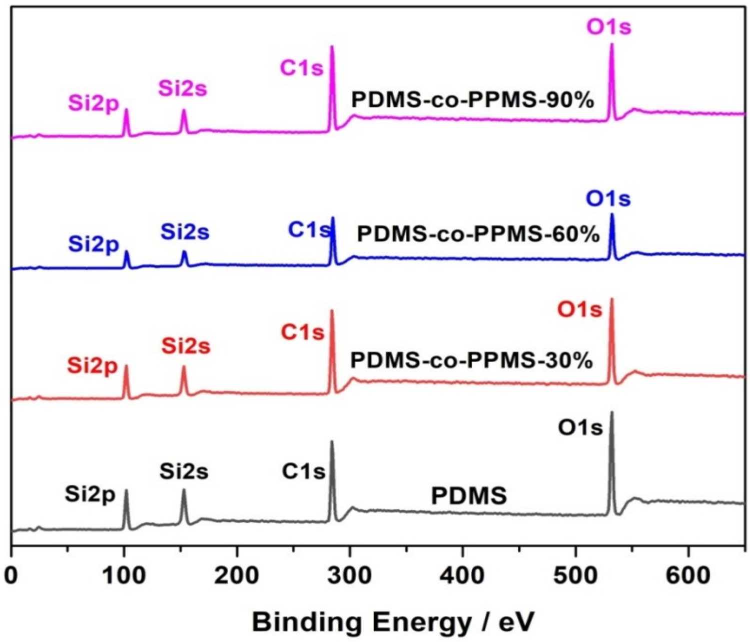

Figure S3. XPS spectra of bulk elastomers with various phenyl contents. The binding energy of N1s is around 403 eV, which was not labeled due to the low relative contents.

Table S2. The calculation results of O/Si, C/Si and N/Si based on the XPS results.

Atomic composition / %	PDMS	PDMS-co-PPMS-30%	PDMS-co-PPMS-60%	PDMS-co-PPMS-90%
O/Si	1.04/1	1.13/1	1.12/1	1.14/1
C/Si	2.15/1	2.77/1	2.83/1	3.40/1
N/Si	0.06/1	0.10/1	0.06/1	0.08/1

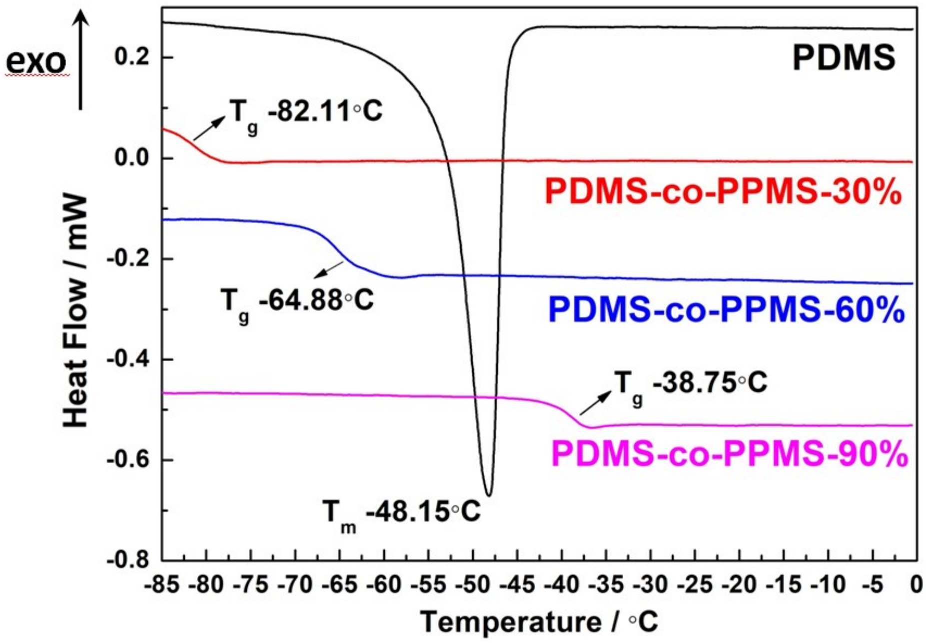

Figure S4. DSC curves of polysiloxane elastomers with various phenyl contents.

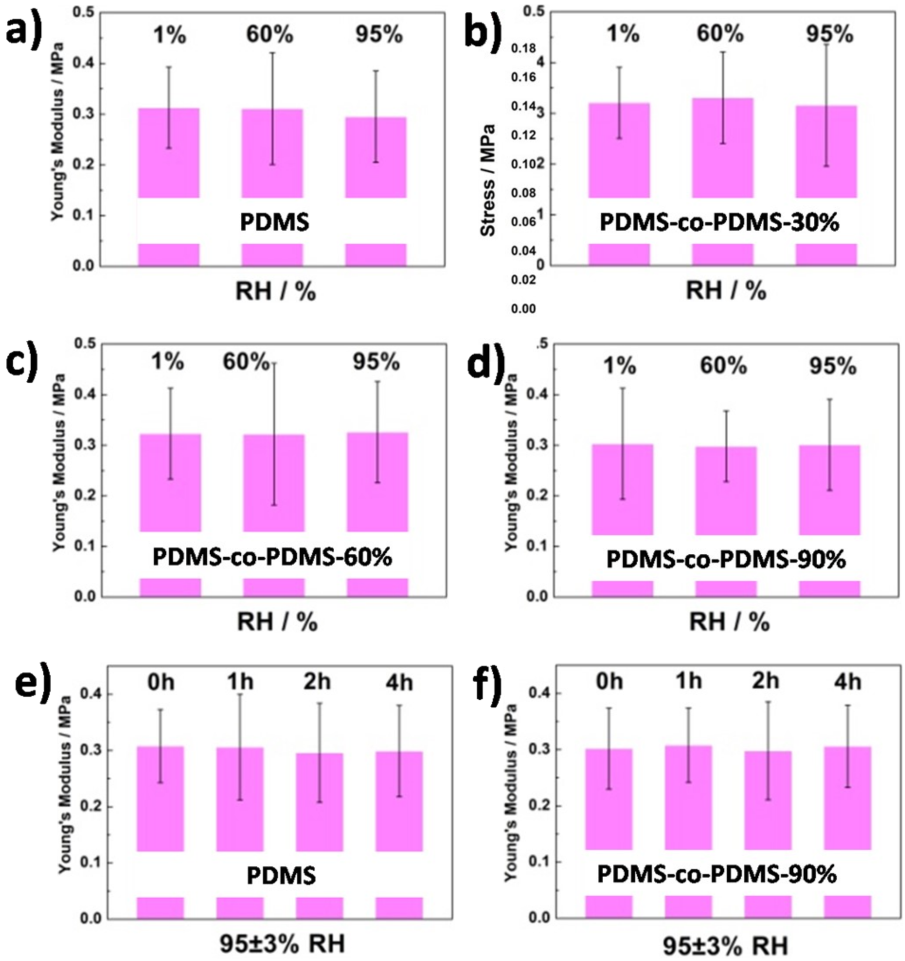

Figure S5. Results of humidity tensile tests results. Young's moduli of PDMS (a), PDMS-co-PPMS-30% (b), PDMS-co-PPMS-60% (c), and PDMS-co-PPMS-90% (d) at RH of 1±0.5%, 60±3%, and 95±3%, respectively, and each humidity set point was equilibrated for 1h. Young's moduli of two specimens, PDMS (e) and PDMS-co-PPMS-90% (f), equilibrated for 1h, 2h, 3h and 4h at 95±3% RH, respectively.

Table S3. The tensile properties of four elastomers at different RH (equilibration for 1h at testing RH).

Relative humidity / %	Young's modulus / Stress at 100% strain [MPa] a)							
	PDMS		PDMS-co-PPMS -30%		PDMS-co-PPMS -60%		PDMS-co-PPMS -90%	
1±0.5%	0.313 ±0.081	0.148 ±0.091	0.321 ±0.073	0.157± 0.078	0.323 ±0.091	0.162 ±0.098	0.303 ±0.112	0.163 ±0.105
60±3.0%	0.311 ±0.112	0.151 ±0.109	0.331 ±0.091	0.159 ±0.098	0.322 ±0.142	0.162 ±0.136	0.298 ±0.076	0.163 ±0.086
95±3.0%	0.295 ±0.092	0.150 ±0.087	0.316 ±0.122	0.159 ±0.118	0.326 ±0.103	0.163 ±0.112	0.301 ±0.092	0.164 ±0.101

Table S4. The tensile properties of PDMS and PDMS-co-PPMS-90% at 95±3.0% RH under various equilibration time, respectively.

Equilibration time		0 h	1 h	2 h	4 h
Young's modulus / Stress at 100% strain [MPa] a)	PDMS	0.308 ±0.065	0.306± 0.094	0.296± 0.088	0.299± 0.081
		0.140 ±0.076	0.143±0.089	0.143±0.0 98	0.142±0.0 89
	PDMS-co-PPMS-90%	0.302± 0.072	0.308± 0.066	0.298± 0.087	0.306± 0.073
		0.151 ±0.087	0.152±0.076	0.152±0.0 83	0.153±0.0 80

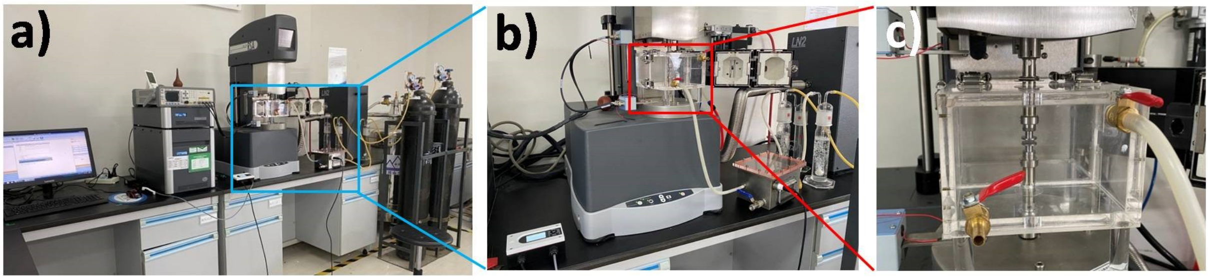

Figure S6. The photograph of humidity macroadhesion measurement device. It consists of RSA G2 solid rheometer (**a**) and humidity regulating setup. By aerating the dry and wet nitrogen with different flow rates through the tailored mixing chamber (**b** and **c**), RH levels can be varied from 1±0.5%% to 95±3%.

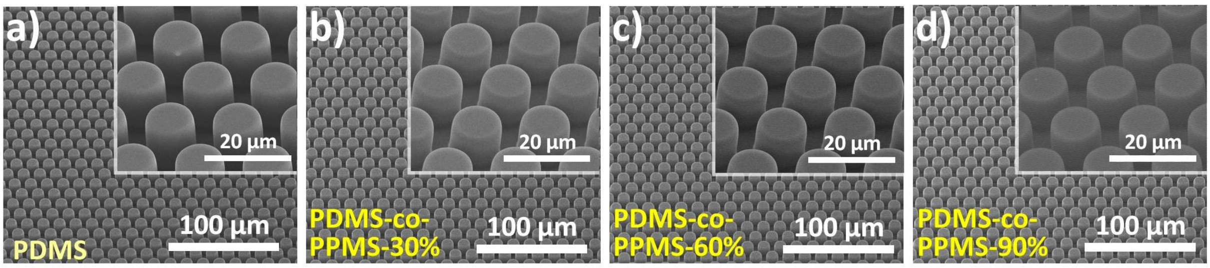

Figure S7. Morphologies of microfibrillar adhesives fabricated by PDMS (**a**), PDMS-co-PPMS-30% (**b**), PDMS-co-PPMS-60% (**c**), and PDMS-co-PPMS-90% (**d**), with pillar diameter of 10 μm, pillar height of 10 μm and spacing of 15 μm, respectively. Insets showed the magnified morphologies of four microfibrillar adhesives.

Table S5. The average pull-off strength of four microfibrillar adhesives at various RH (silicon wafer substrate, preload 0.05N, 25±2°C).

Relative humidity / %	Pull-off strength / $N \cdot cm^{-2}$			
	PDMS	PDMS-co-PPMS -30%	PDMS-co-PPMS -60%	PDMS-co-PPMS -90%
1±0.5%	0.404±0.031	0.571±0.037	0.762±0.034	1.159±0.119
30±3.0%	0.323±0.032	0.483±0.038	0.599±0.036	1.032±0.091
60±3.0%	0.248±0.035	0.398±0.049	0.589±0.039	0.969±0.085
80±3.0%	0.232±0.031	0.382±0.043	0.519±0.048	0.970±0.090
95±3.0%	0.153±0.046	0.328±0.055	0.425±0.063	0.919±0.127

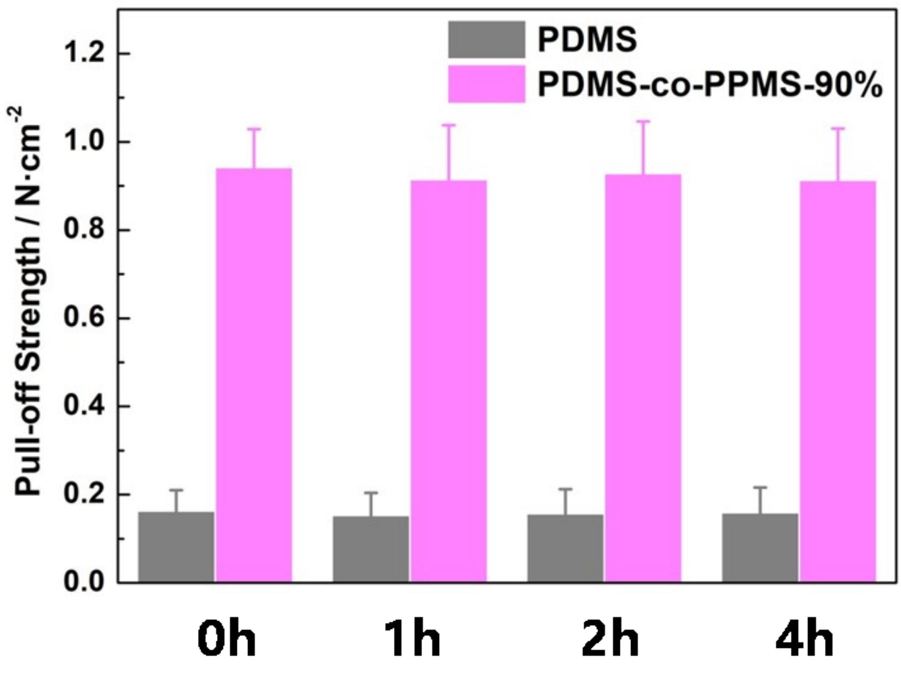

Figure S8. The comparison of average pull-off strength of PDMS and PDMS-co-PPMS-90% at 95±3.0% RH under various equilibration time.

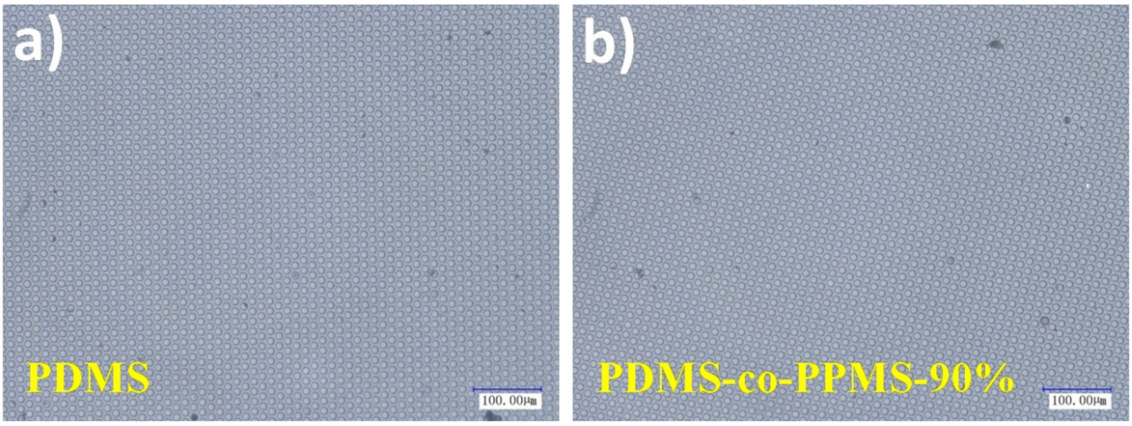

Figure S9. Optical microstructures of PDMS (**a**) and PDMS-co-PPMS-90% (**b**) microfibrillar adhesives after humidity macroadhesion tests.

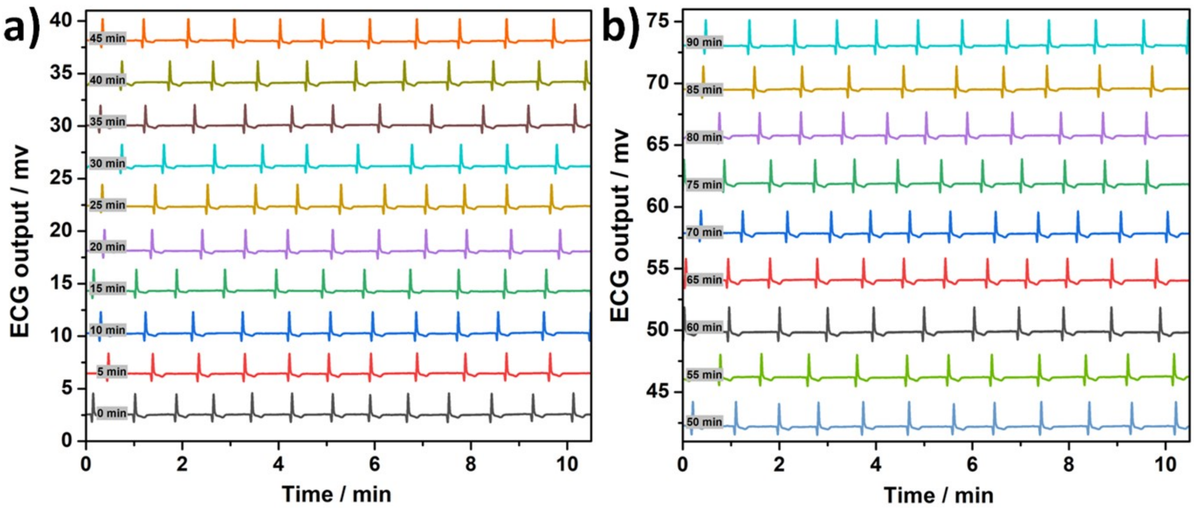

Figure S10. ECG data taken on a seated subject based on electrodes without gel. (a) 0-45min; (b) 50-90min.

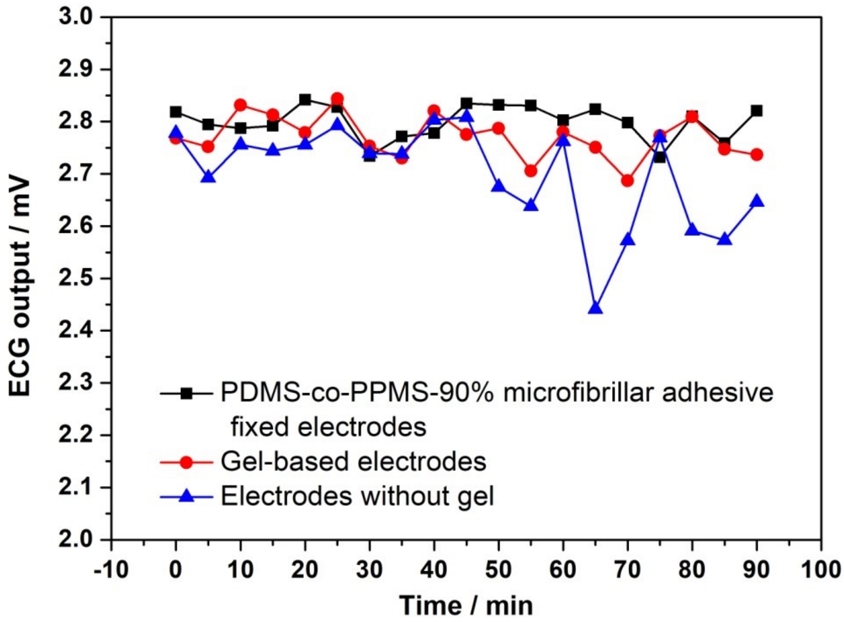

Figure S11. The ECG output voltage (R-wave) of three kinds of electrodes as a function of monitoring time.

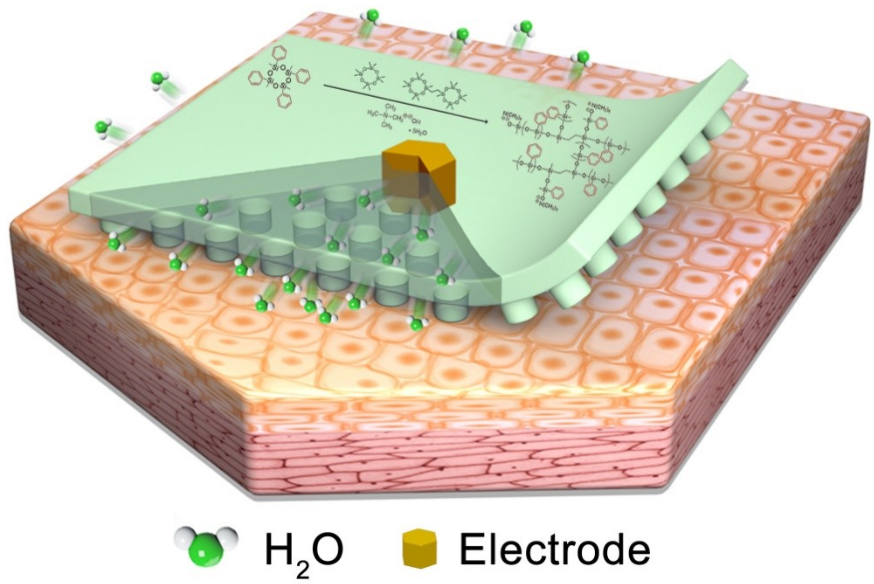

Figure S12. Conceptual illustration of the phenyl-rich microfibrillar adhesive used as skin patches showing strong hydrophobic effect.

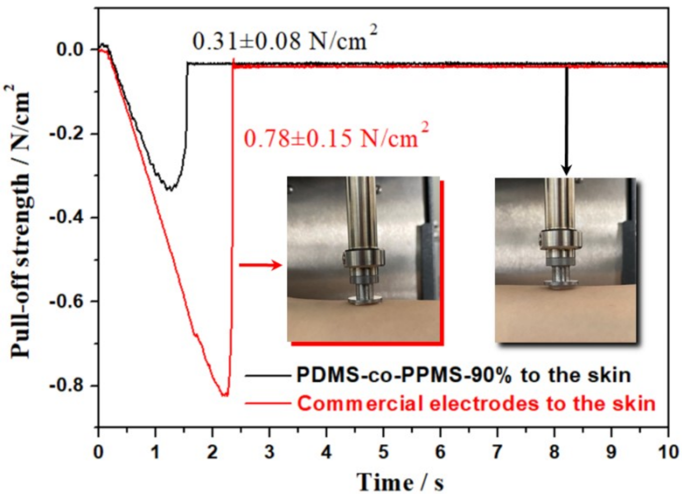

Figure S13. Typical pull-off strength-time curves of PDMS-co-PPMS-90% and conventional electrode on a volunteer's forearm skin, respectively.

Video S1. The panorama of humidity macroadhesion tests.