Supplementary Information

Adhesive, biocompatible, and conductive reduced graphene oxide hydrogel-based bioelectrodes for epidermal electrophysiological signal monitoring

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KEYWORDS: Wearable bioelectronics, epidermal bioelectrode, reduced graphene oxide hydrogel, electrophysiological signal, adhesive, biocompatibility

Supplementary Information.

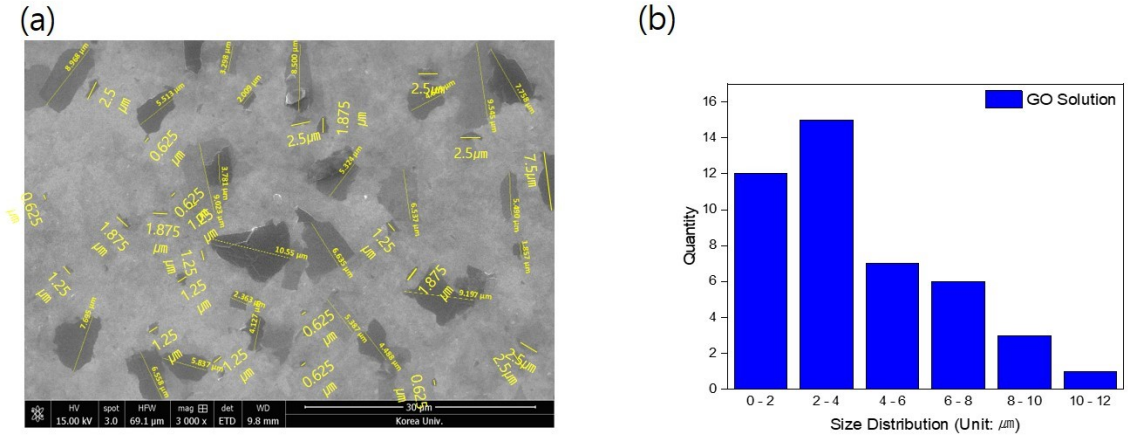

Figure S1. Characterization of GO flakes. (a) FESEM image of GO flakes on silicon substrates. (b) Size distribution of GO flakes.

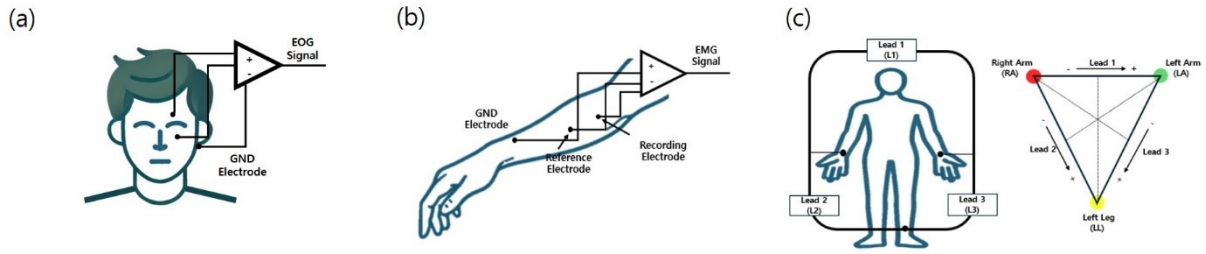

Figure S2. Schematic illustration of epidermal electrode placement for (a) EOG, (b) EMG, and (c) ECG measurements.

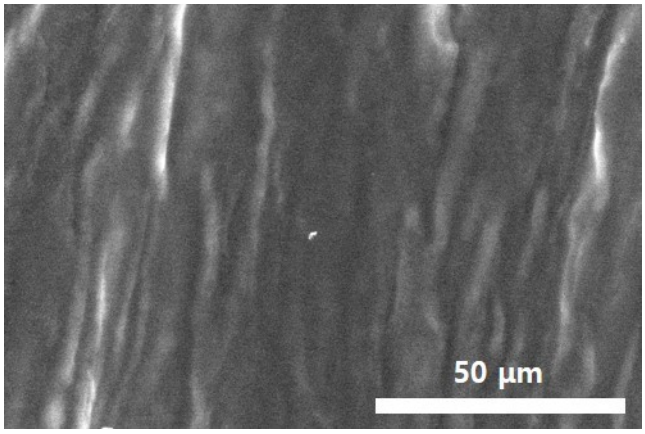

Figure S3. ESEM image of the RGO hydrogel surface without PAA adhesive infiltration.

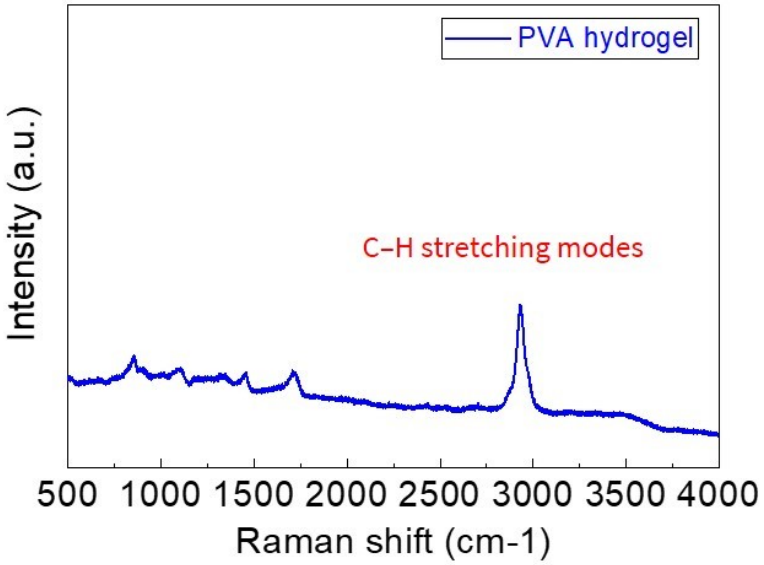

Figure S4. Raman spectrum of the PVA hydrogel.

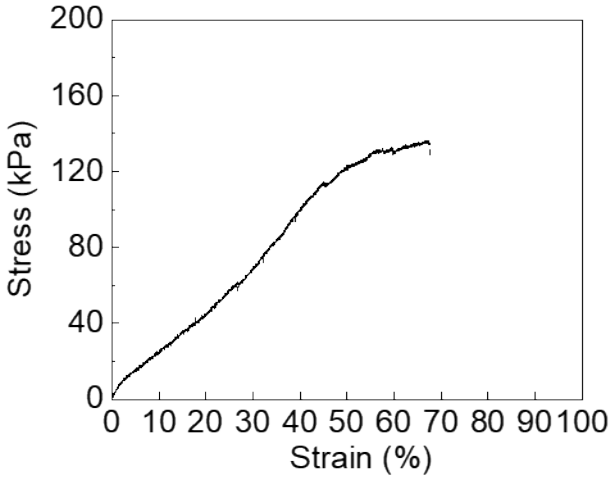

Figure S5. Tensile stress-strain curve of the RGO hydrogel.

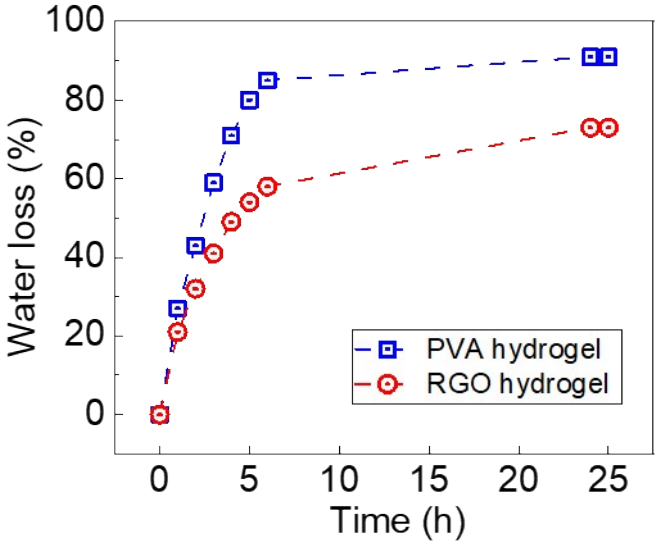

Figure S6. Water retention capacity of the RGO hydrogel.

(a)

(b)

Adheison Area	Back of the hand		Forehead		Forearm		Wrist	
	Adhesive force (N)	Adhesive strength (kPa)	Adhesive force (N)	Adhesive strength (kPa)	Adhesive force (N)	Adhesive strength (kPa)	Adhesive force (N)	Adhesive strength (kPa)
1	1.53	4.870	0.45	1.432	2.3	7.321	1.79	5.698
2	1.29	4.106	0.57	1.814	1.92	6.112	1.86	5.921
3	1.57	4.997	0.53	1.687	2.32	7.385	1.62	5.157
4	0.99	3.151	0.37	1.178	1.5	4.775	2.13	6.780
5	1.44	4.584	0.31	0.987	2.55	8.117	1.34	4.265
6	1.42	4.520	0.24	0.764	1.79	5.698	1.36	4.329
7	1.22	3.883	0.23	0.732	2.33	7.417	2.23	7.098
8	1.44	4.584	0.23	0.732	2.24	7.130	1.6	5.093
9	1.59	5.061	0.32	1.019	2.07	6.589	1.47	4.679
10	1.04	3.310	0.27	0.859	1.42	4.520	1.45	4.615
Average	1.353	4.307	0.352	1.120	2.044	6.506	1.685	5.364

Figure S7. (a) Photoimages of normal adhesion strength measurement and (b) Adhesive force and strength values of the RGO hydrogel to different skins.

Table S1. Detailed information about the synthesis of different GO hydrogels.

Hydrogels	PVA (mg)	GO (mg)	DI water (mL)
GO 0-PVA	1000	0	40
GO 10-PVA	1000	50	40
GO 15-PVA	1000	66.7	40
GO 20-PVA	1000	100	40

Table S2. Summary of perfo ce comparison with previously reported GO/RGO hydrogel EP bioelectrodes.

Bioelectrode	Backbone material	Bioadhesive material	Reduction	Adhesion strength	Electrical conductivity	Impedance	Biocompatibility	Target tissues	EP signals	Ref
Adhesive RGO hydrogel	Polyvinyl alcohol (PVA)	Polyacrylic acid (PAA)	Chemical reduction ($Na_2S_2O_4$)	max. 6.506 kPa at forearm min. 1.12 kPa at forehead	~ 0.11 S/m (in dry condition)	~ 127.5 Ω at 10^2 Hz ~ 89.7 Ω at 10^3 Hz	> 90% In vitro cell viability	Back of the hand, forehead, forearm, wrist	EOG, EMG, and ECG under talking and sweating	This wo
GO hydrogel	Polyvinyl alcohol/Polyethylene glycol (PVA/PEG)	-	-	-	~ 0.02 S/m (in dry condition)	-	No skin allergy	Chest	ECG under static and walking states	27
Thermally annealed graphene-channeled agarose hydrogel (TAGAH)	Agarose	-	Thermal reduction (70°C for 60 min)	-	~ 20.7 S/m (in PBS solution) ~ 2.5 S/m (in DI water)	~ 29.2 Ω at 10^4 Hz	> 90% In vitro cell viability	Leg	Implantable EMG in a rat model	28
GO hydrogel	Agarose	-	-	-	~ 12.0 S/m (in PBS solution) ~ 0.5 S/m (in DI water)	~ 370.0 Ω at 10^4 Hz	> 80% In vitro cell viability	Leg	Implantable EMG in a rat model	28
Gelatin/ polypyrrole/ RGO organohydrogel	Gelatin	-	Chemical reduction (Ascorbic acid)	-	~ 5.25 S/m (in dry condition)	-	-	Forearm, wrist	ECG under static state	29
Polydopamine/ RGO/ polyacrylamide hydrogel	Polyacrylamide (PAM)	Polydopamine (PDA)	Chemical reduction (PDA)	max. 30 kPa at porcine skin	~ 10.0 S/m	72 kΩ (first) 125 kΩ (tenth)	-	Forearm	ECG under static state and implantable EMG in a rat model	30
Electrical bioadhesive	PVA	PAA	Chemical reduction ($Na_2S_2O_4$)	-	~ 2.6 S/m (in PBS buffer)	~ 50 Ω at 10^2~10^5Hz	> 90% In vitro cell viability	Heart, skin, muscle, sciatic nerve	Implantable ECG in a rat model	31