Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Surface-control enhanced crater-like electrode in gelatin/polyvinyl alcohol/carbon composite for biodegradable multi-modal sensing systems with human-affinity†

Jingzhe Sun [‡a], Dahye Ahn [‡a], Junseo Kim [‡a], Seongcheol Ahn [a], Jum Soo Hwang [b], Joo Youl Kwon [b], Jae Soo Lee [c], Jung Moon Oh [b*], Kiyoung Nam [c*], Jong-Jin Park [a*]

a. Department of Science and Polymer Engineering, Chonnam National University, Gwangju 61186, South Korea

E-mail: jjpark@jnu.ac.kr

b. DEERS i, 76 Cheongbuksandan-ro, Cheongbuk-eup, Pyeongtaeksi, Gyeonggi-do 17792, South Korea

E-mail: jmoh@deersi.co.kr

c. Tara Distribution, 9F, 34, Sangamsanro, Mapogu, Seoul 03939, South Korea

E-mail: kynam@tarapaper.co.kr

‡ These authors contributed to this work equally.

† Electronic Supplementary Information

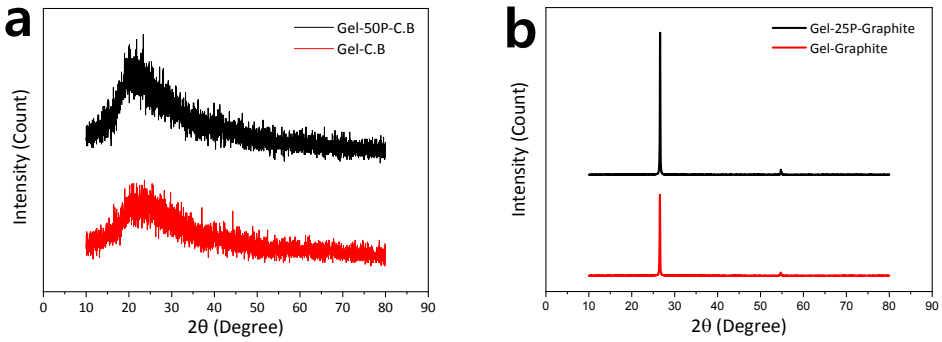

Fig. S1 XRD patterns of gelatin and gelatin-nPVA according to carbon material. a Carbon Black. **b** Graphite.

When carbon black was used as a conductive additive, it was confirmed that the intensity of the XRD peak increased when PVA was added than when there were only gelatin and carbon black. Likewise, when graphite was used, it was confirmed that the intensity of the XRD peak increased when PVA was added. The crystallinity was also increased as a result. As described in the text (Fig. 1d), gelatin and PVA could form a self-assembled structure with crystallinity. Thus, both mixtures showed crystallinity. In all three cases, they showed a crystalline structure.

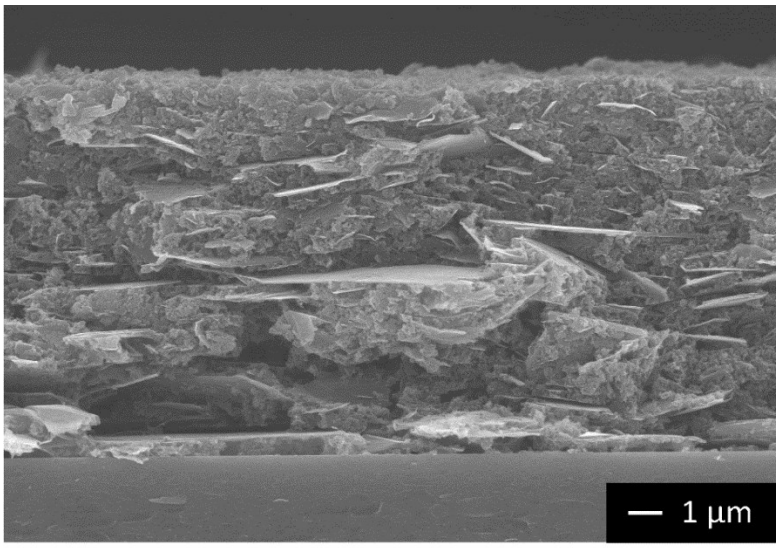

Fig. S2 SEM image of layer by layer well-packed graphite flakes through gelatin self-assembly effect.

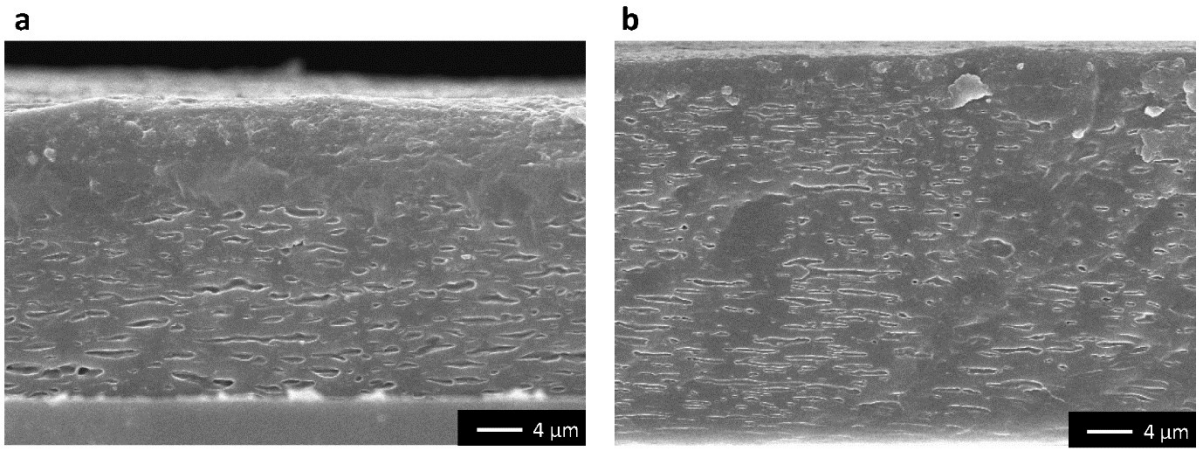

Fig. S3 SEM image of gelatin-PVA film cross-section which shows the isolated pores. a Gel-14P free standing film. **b** Gel-25P free standing film.

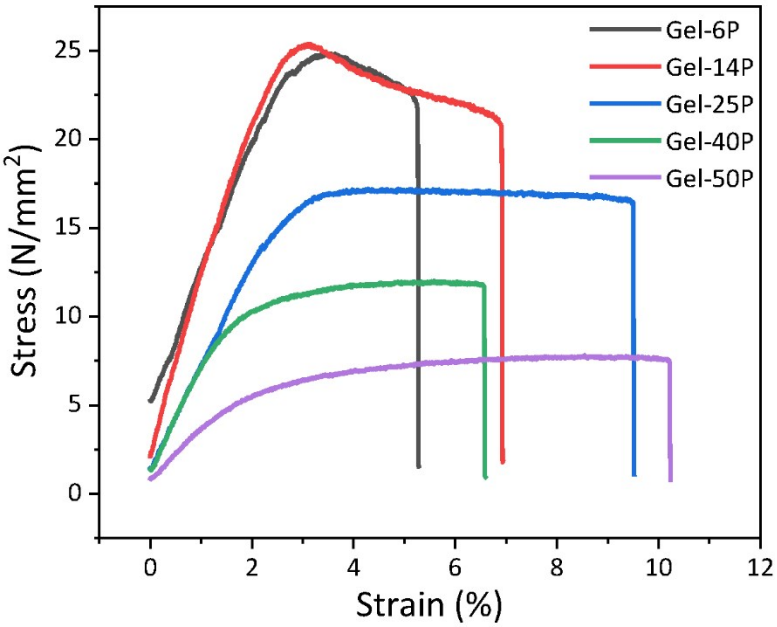

Fig. S4 Strain-Stress curve for different PVA content Gel-PVA blending free standing film.

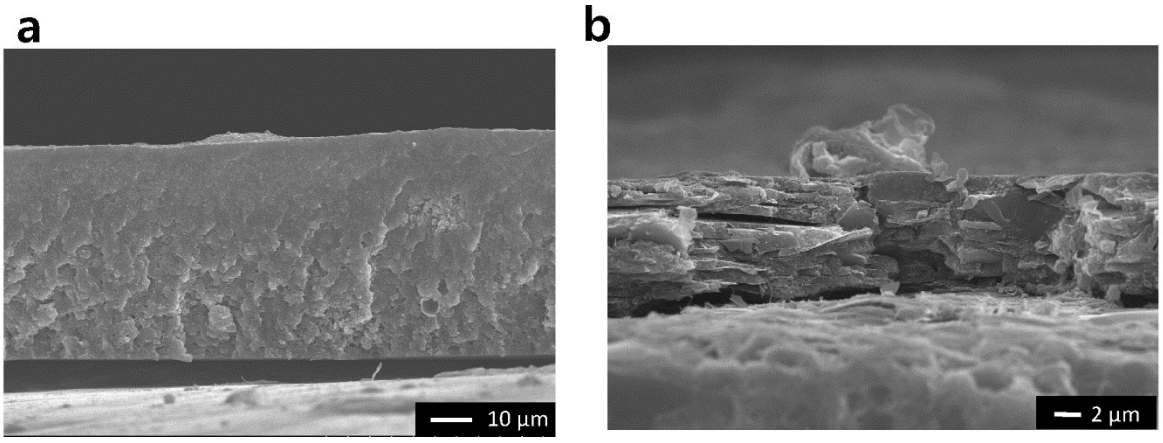

Figure S5. Cross section SEM images of film made with different methods. **a** Film casting method (Thickness : 51 µm). **b** Direct skin coating method (Thickness : 9 µm).

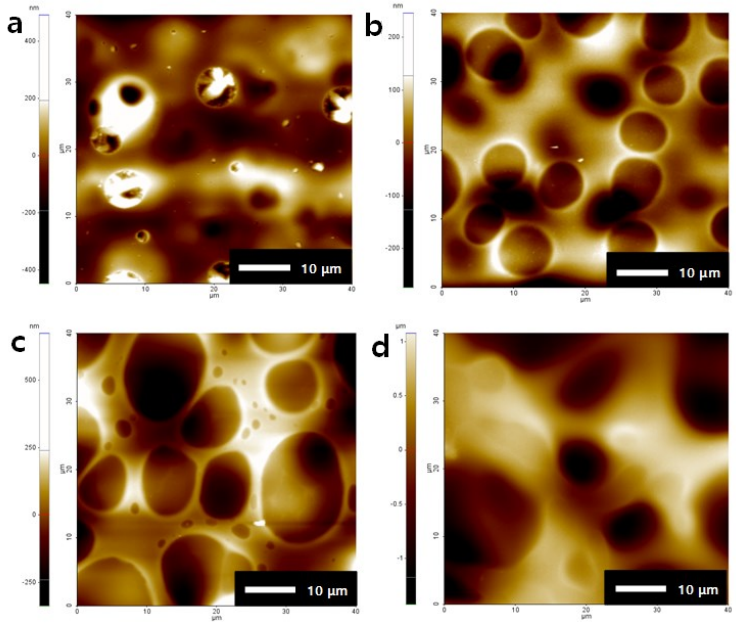

Fig. S6 The AFM image of different PVA content gelatin coating layer. a 6wt% PVA content. **b** 14wt% PVA content. **c** 25wt% PVA content. **d** 50wt% PVA content.

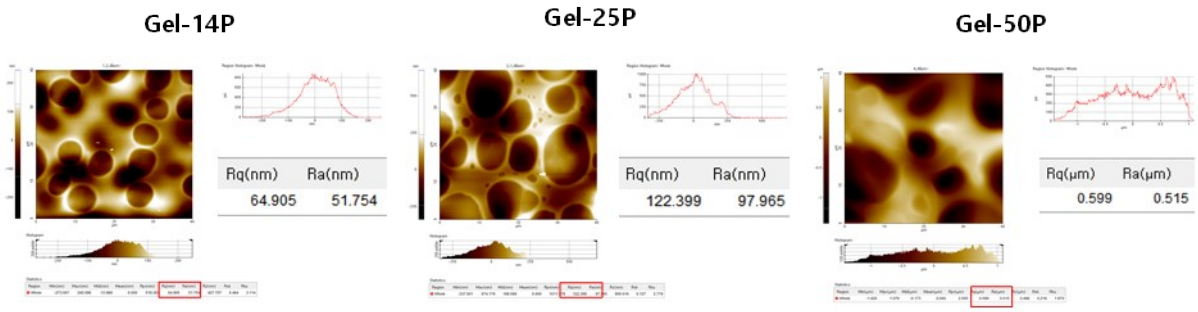

Fig. S7 Surface roughness values of the gelatin-nPVA coating layer.

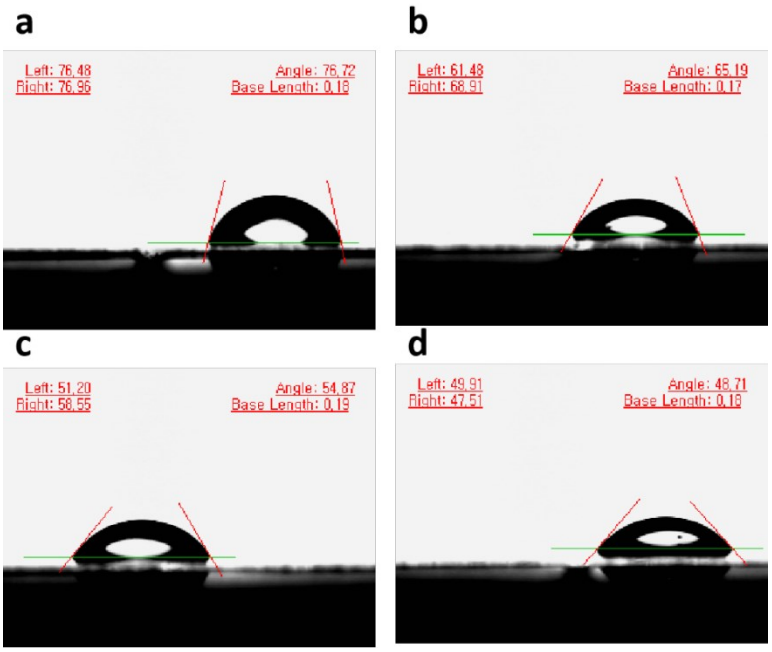

Fig. S8. The contact angle for different PVA percentage content GP solution. a Pure gelatin solution. **b** Gel-25P solution. **c** Gel-40P solution. **d** Gel-50P solution.

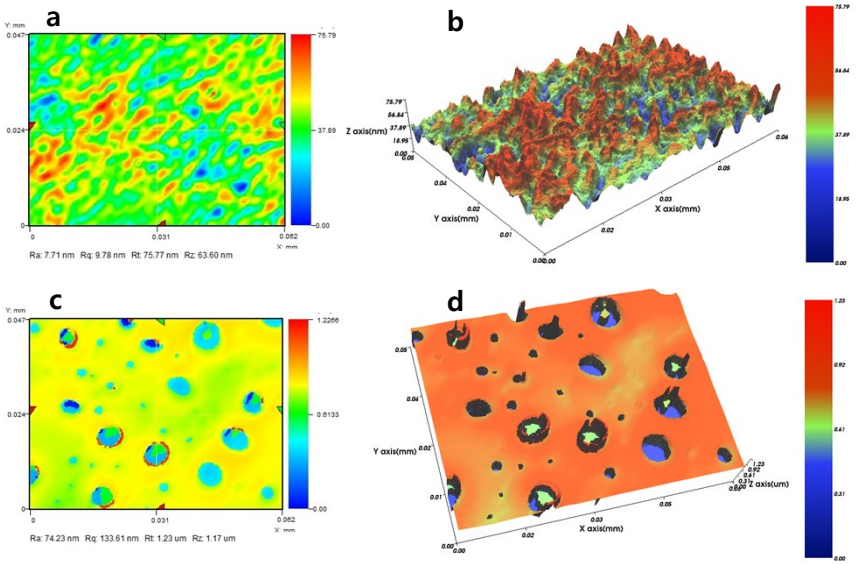

Fig. S9 The 3D optical profiler image of GP casting layer under different pH value. a GP casting layer (pH=2) 2D picture. **b** GP casting layer (pH=2) 3D picture. **c** GP casting layer (pH=6) 2D picture. **d** GP casting layer (pH=6) 3D picture.

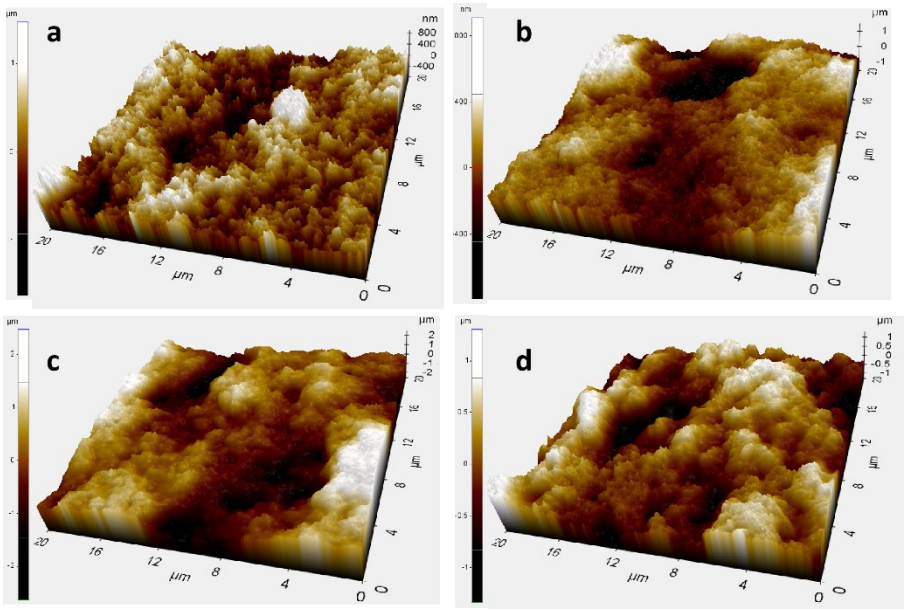

Fig .S10 The AFM image of different PVA percentage content Gel-PVA-Carbon black film. a Gel-0P –Carbon black sample. **b** Gel-14P-Carbon black sample. **c** Gel-40P-Carbon black sample. **d** Gel-50P-Carbon black sample.

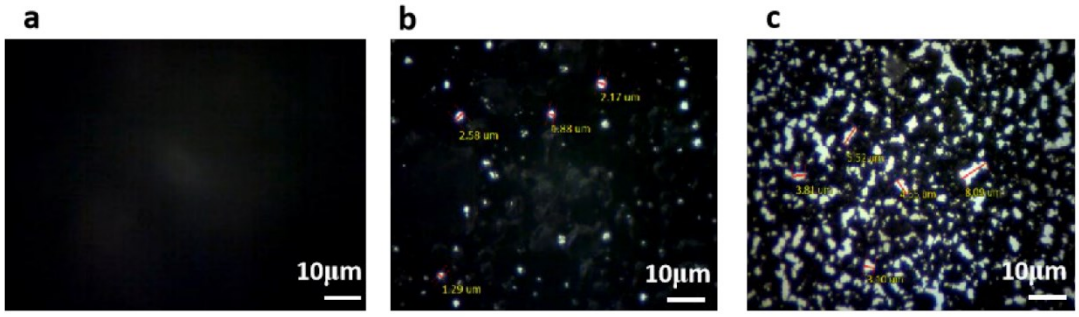

Fig. S11 The optical microscope image for different PVA percentage content in the GPC casting layer. a Gel-0P. **b** Gel-25P. **c** Gel-50P.

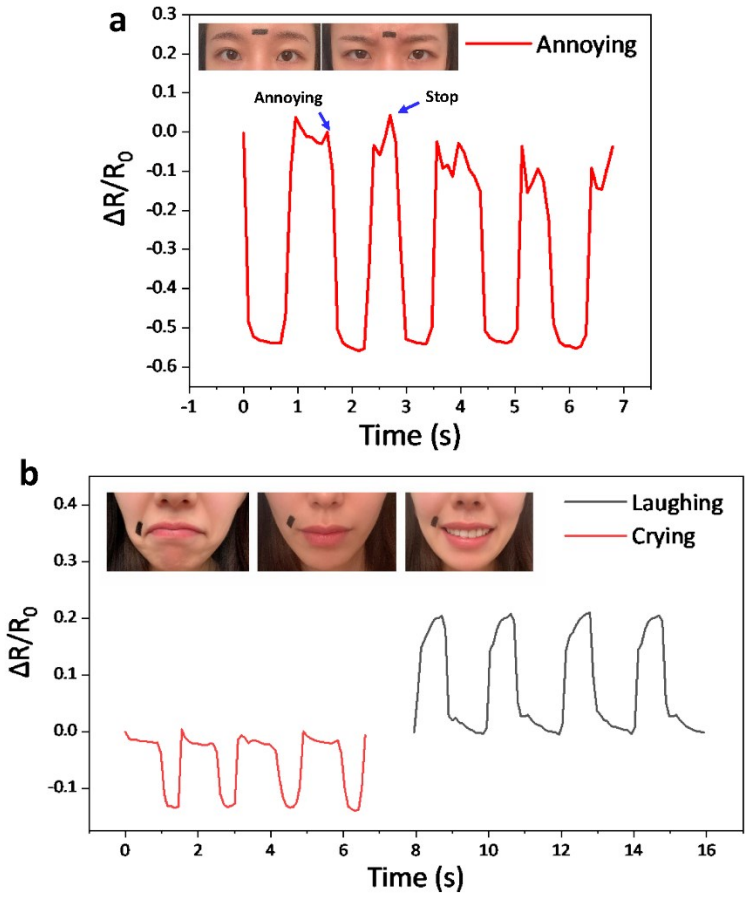

Fig. S12 The output signal of a human emotion sensing unit. a The forehead emotion sensing unit for detecting the annoying signal. **b** The near mouth sensing unit for detecting laughing and crying signals.

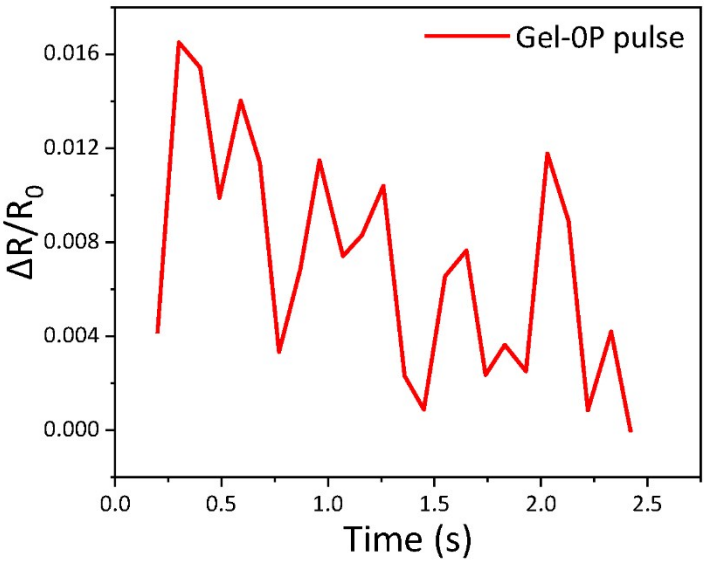

Fig. S13 The pulse measure signal of pure gelatin-Carbon black free standing film, which shows the irregular output with low sensitivity.

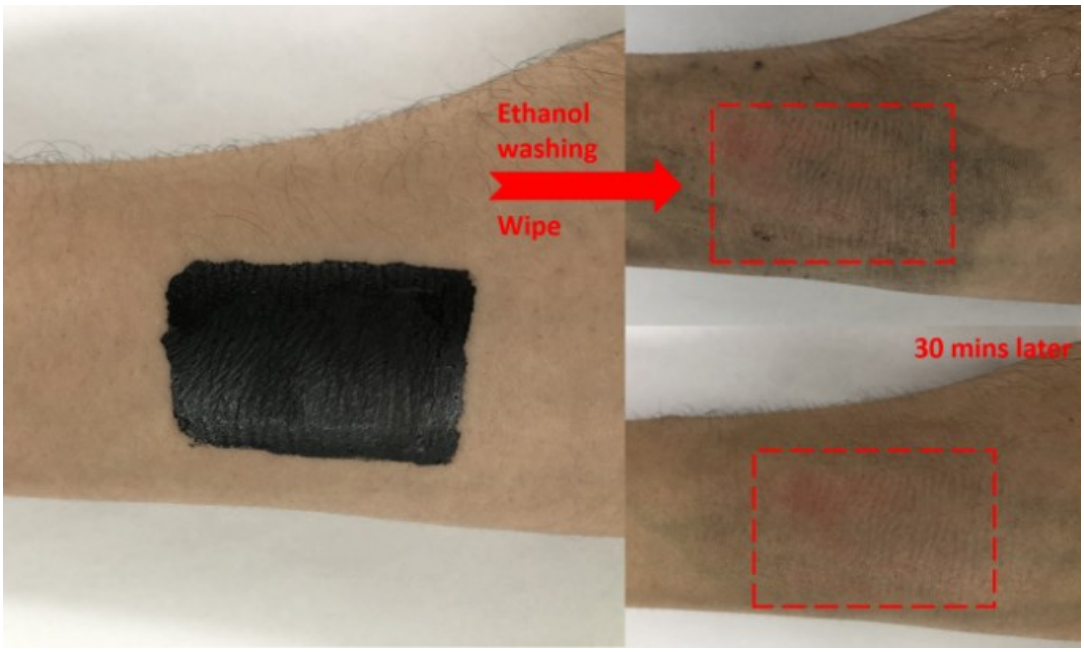

Fig. S14 The irradiation (red swollen) appears when using PVB (binder)-Carbon black solution coating on human skin.

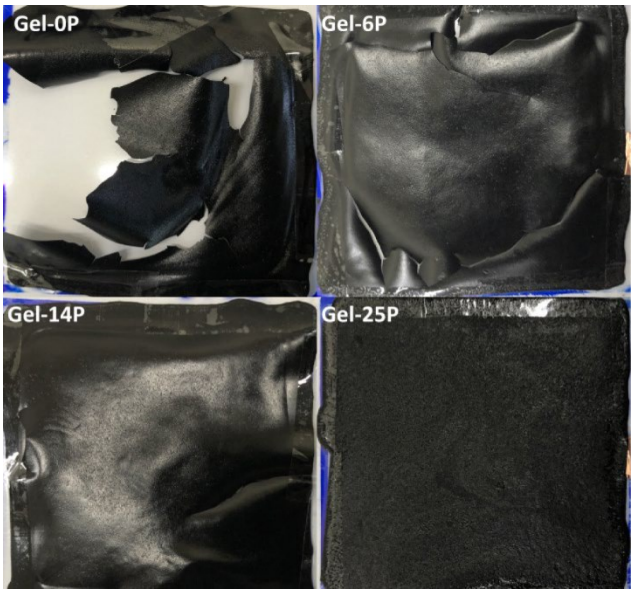

Fig. S15 The crack occur at low PVA percentage content GPC free standing film. Cracks gradually disappeared as PVA content increase.

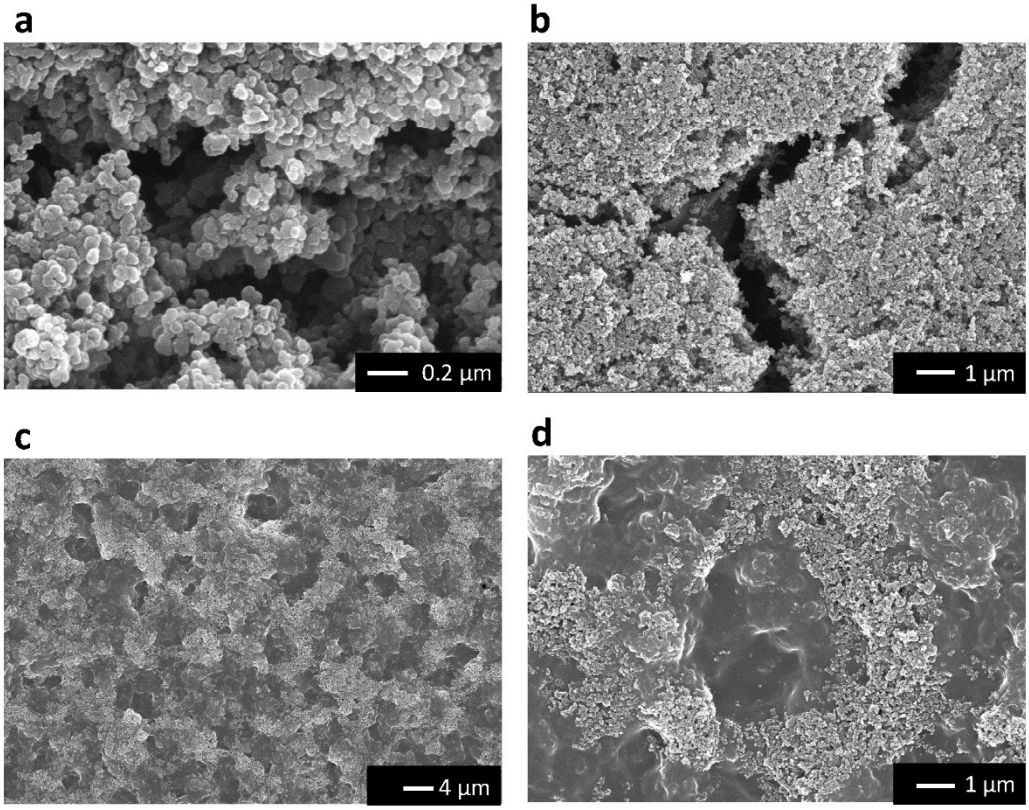

Fig. S16 The SEM image of PVA anti-crack mechanism. a 50k times magnified gel-Carbon black picture. **b** 10k times magnified gel-Carbon black picture. **c** 2k times magnified gel-PVA-Carbon black picture. **d** 10k times magnified gel-PVA-Carbon black picture.

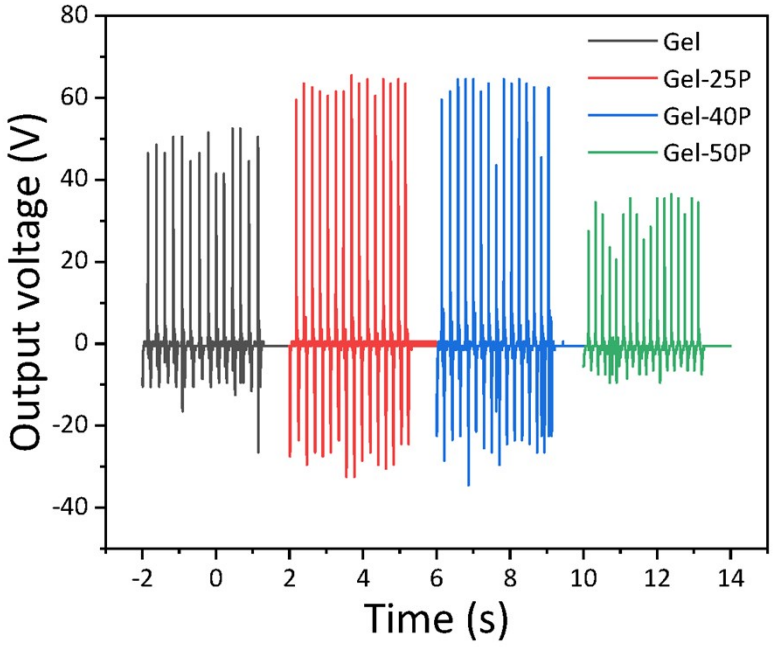

Fig. S17 The triboelectric output voltage for different PVA percentage content of GPC film.

	Water	Gelatin	PVA	$NaHCO_3$	CB*	Graphite:CB=1:1*
PVA 6 wt% 10 mg ml^{-1} conductive filler	10 g	0.3 g	0.02 g	0.02 g	0.1 g	0.1 g
PVA 6 wt% 60 mg ml^{-1} conductive filler	10 g	0.3 g	0.02 g	0.02 g	0.6 g	0.6 g
PVA 14 wt% 10 mg ml^{-1} conductive filler	10 g	0.3 g	0.05 g	0.02 g	0.1 g	0.1 g
PVA 14 wt% 60 mg ml^{-1} conductive filler	10 g	0.3 g	0.05 g	0.02 g	0.6 g	0.6 g
PVA 25 wt% 10 mg ml^{-1} conductive filler	10 g	0.3 g	0.1 g	0.02 g	0.1 g	0.1 g
PVA 25 wt% 60 mg ml^{-1} conductive filler	10 g	0.3 g	0.1 g	0.02 g	0.6 g	0.6 g
PVA 40 wt% 10 mg ml^{-1} conductive filler	10 g	0.3 g	0.2 g	0.02 g	0.1 g	0.1 g
PVA 40 wt% 60 mg ml^{-1} conductive filler	10 g	0.3 g	0.2 g	0.02 g	0.6 g	0.6 g
PVA 50 wt% 10 mg ml^{-1} conductive filler	10 g	0.3 g	0.3 g	0.02 g	0.1 g	0.1 g
PVA 50 wt% 60 mg ml^{-1} conductive filler	10 g	0.3 g	0.3 g	0.02 g	0.6 g	0.6 g

*CB or graphite-CB mixture is used for different experiments, not both used at the same time.

Table S1. Chemical compositions of GPC samples with different PVA and carbon amounts

Sensor fabrication

The amount of PVA varied according to the sensor usage. In order to make a breathable electrode, PVA content was 40 wt% or more. When tapping or pressure was applied to the film such as a touch pad and a triboelectric generator, 50 wt% PVA was used. In addition, 50 wt% of PVA was used for knee pads that must be bent at various angles.

Direct skin coating should be well coated on curved skin. Thus, physical properties of PVA should not be hard. However, resistance decreased as the amount of PVA increased, confirming that the optimal amount of PVA that could secure the resistance without making the film brittle was 40 wt%.