

## Electronic Supplementary Information (ESI†)

### Wearable gold-graphene dry electrode-based headband for effective brain-computer interface applications

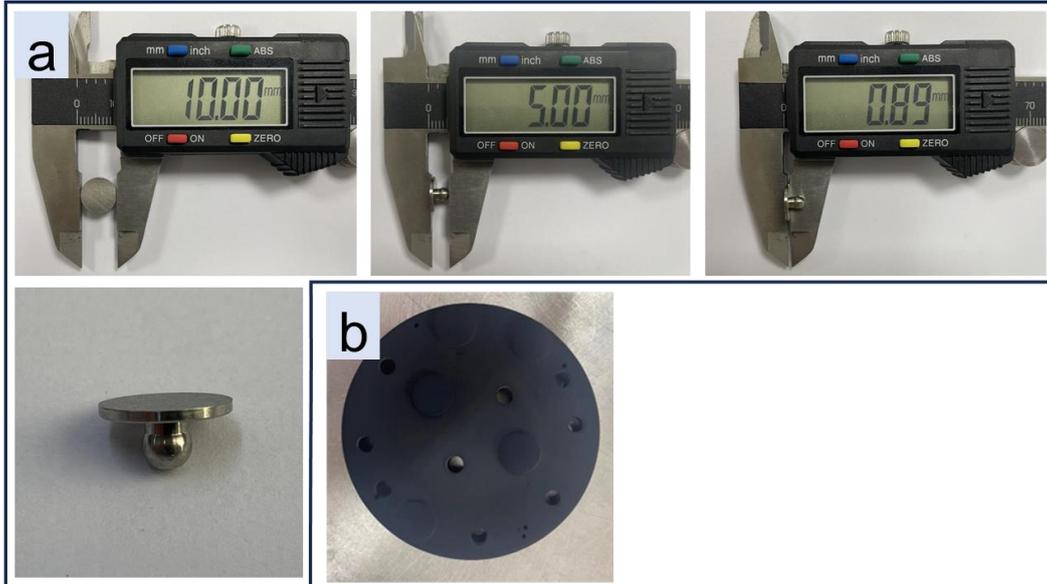
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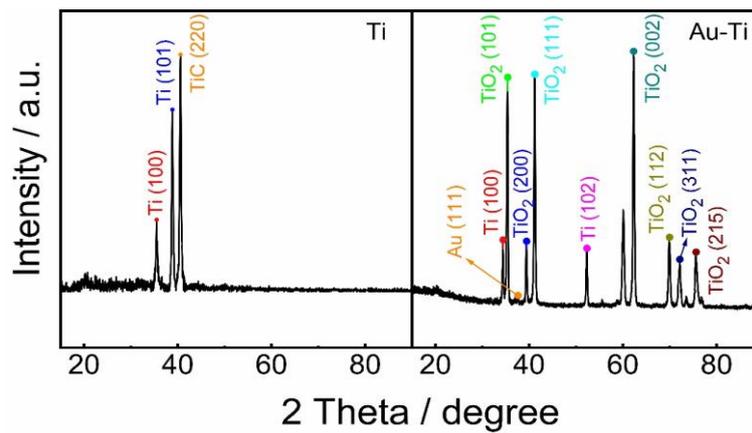
<sup>b</sup> *Tianjin Key Laboratory of Film Electronic and Communication Devices, School of Integrated Circuit Science and Engineering, Tianjin University of Technology, Tianjin 300384, PR China*

**\*Corresponding Authors.**

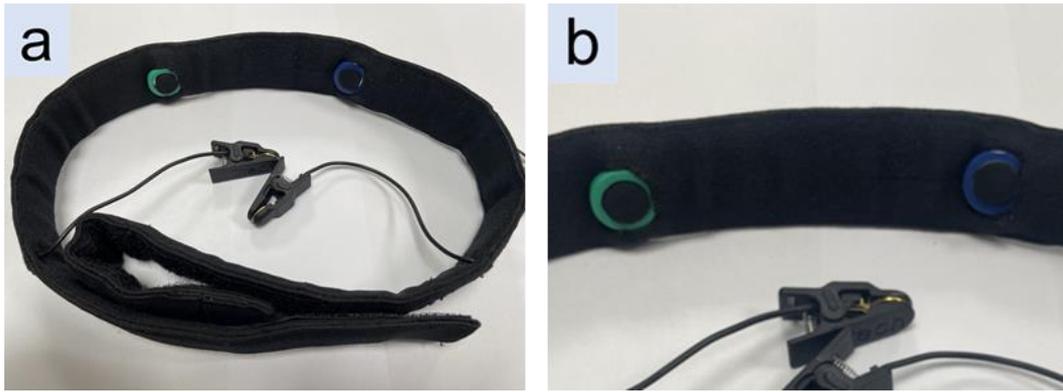
E-mail: hongjili@yeah.net (H. Li); limingji@163.com (M. Li); sunyy1977@htmail.com (Y. Sun)



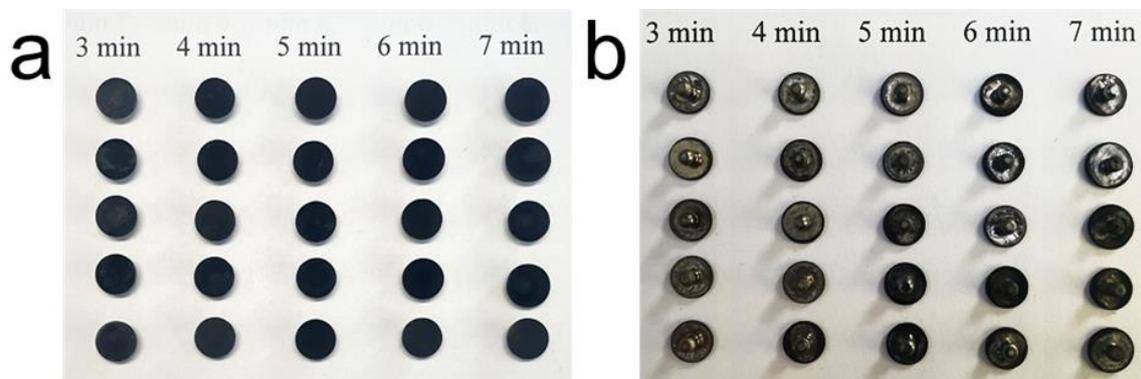
**Fig. S1.** Photographs of (a) the Ti shank button substrate and (b) two Au-FLG electrodes on the sample stage. Au-FLG, gold-doped few-layer graphene



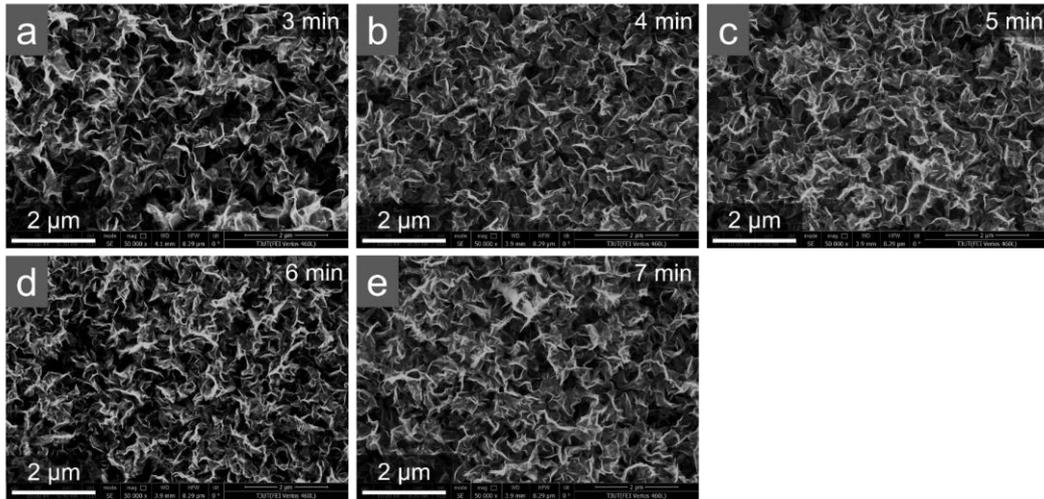
**Fig. S2.** XRD patterns of the Ti and Au-Ti ( $CA/Au^{III} = 20$ ) button electrodes. XRD, X-ray diffraction



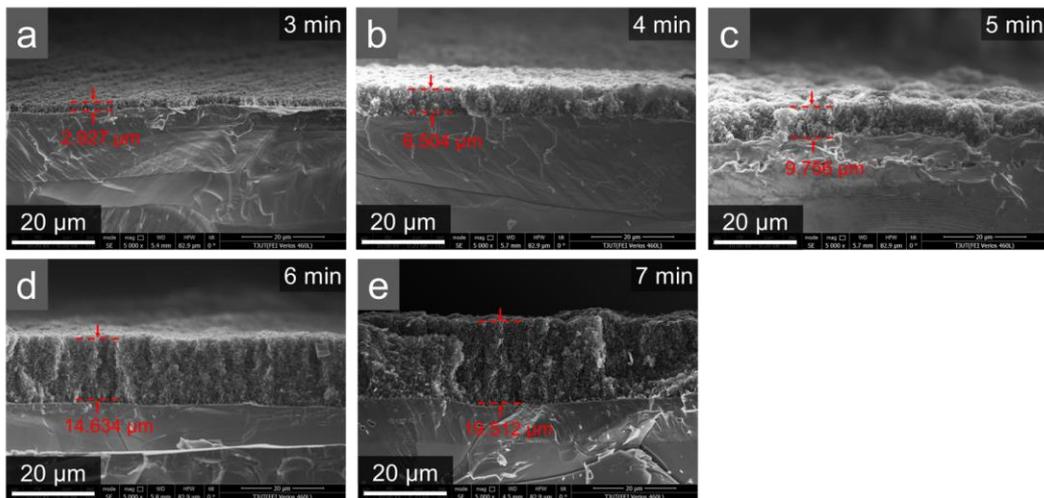
**Fig. S3.** Optical images of an EEG headband with two Au-FLG day electrodes. (a) Whole. (b) Electrode position. Au-FLG, gold-doped few-layer graphene; EEG, electroencephalogram



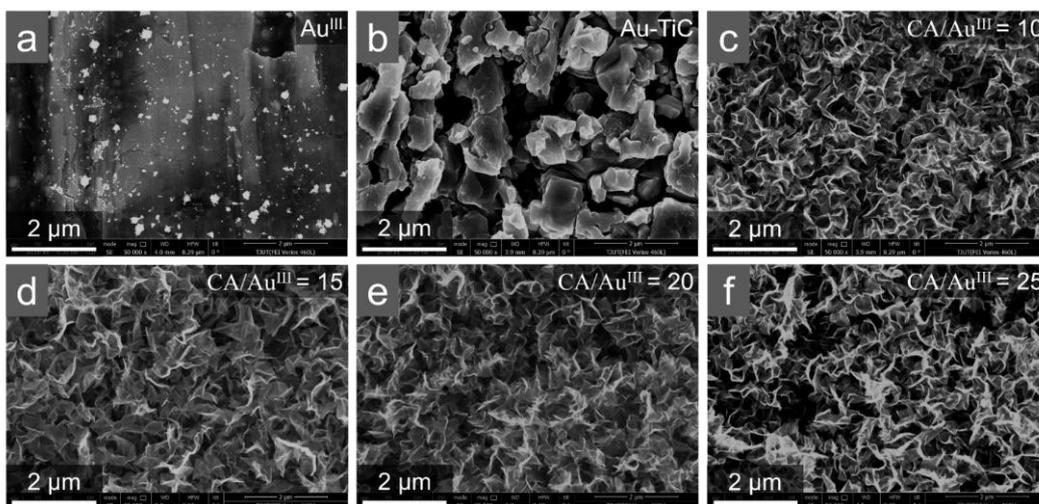
**Fig. S4.** Optical images of FLG button electrodes. (a) Top. (b) Bottom.



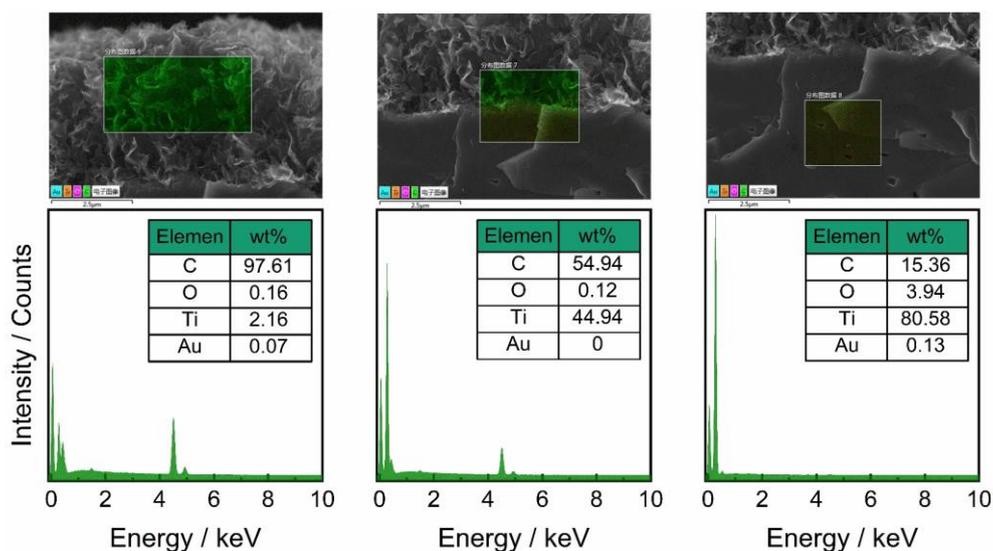
**Fig. S5.** Surface SEM images of FLG samples with growth times of 3, 4, 5, 6, and 7 min. FLG, few-layer graphene; SEM, scanning electron microscopy



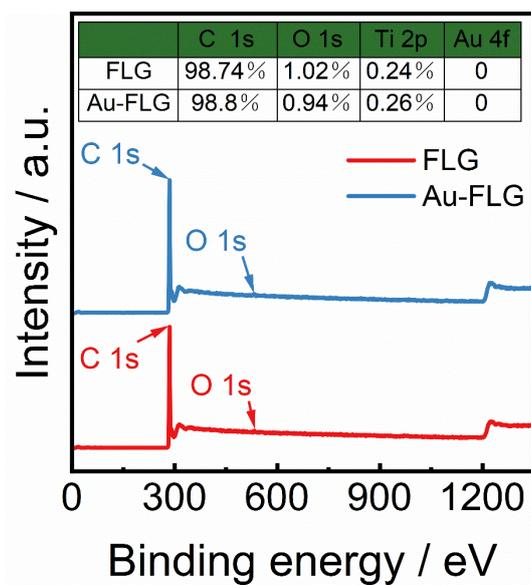
**Fig. S6.** Cross-sectional SEM images of FLG samples with growth times of (a) 3, (b) 4, (c) 5, (d) 6, and (e) 7 min. The height of graphene films was marked. FLG, few-layer graphene; SEM, scanning electron microscopy



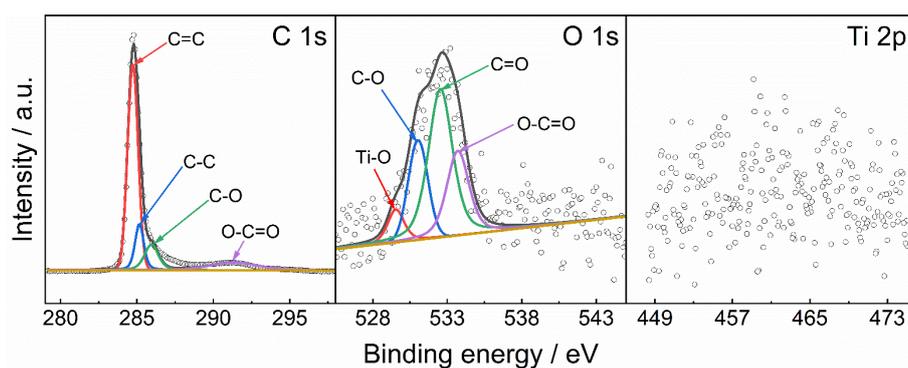
**Fig. S7.** Surface SEM images of (a) Au<sup>III</sup> at Ti substrate, (b) Au-TiC layer, and (c–f) Au-FLG samples with different citric acid/HAuCl<sub>4</sub>(CA/Au<sup>III</sup>) concentration ratios of 10, 15, 20, and 25. Au-FLG, gold-doped few-layer graphene



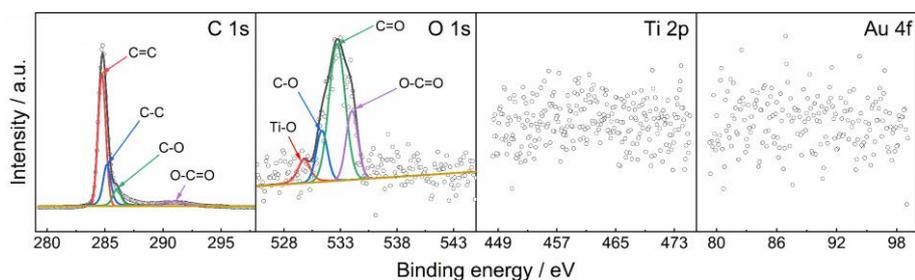
**Fig. S8.** Cross-sectional SEM images and corresponding EDS spectra of an Au-FLG/Ti electrode with a CA/Au<sup>III</sup> concentration ratio of 20. Au-FLG, gold-doped few-layer graphene; EDS, energy-dispersive X-ray spectroscopy; SEM, scanning electron microscopy



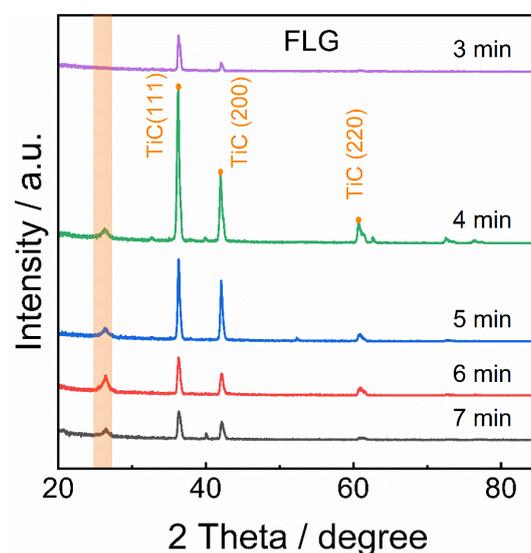
**Fig. S9.** Survey XPS profiles of FLG and Au-FLG samples. Au-FLG, gold-doped few-layer graphene; XPS, X-ray photoelectron spectrometry



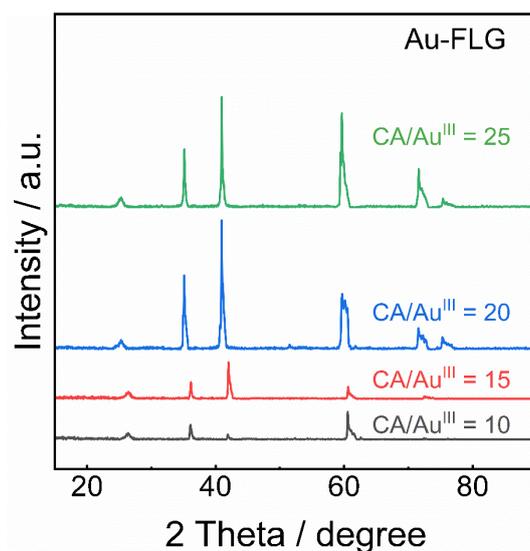
**Fig. S10.** C 1s and O 1s core-level XPS profiles of the FLG sample with a growth time of 4 min. FLG, few-layer graphene; XPS, X-ray photoelectron spectrometry



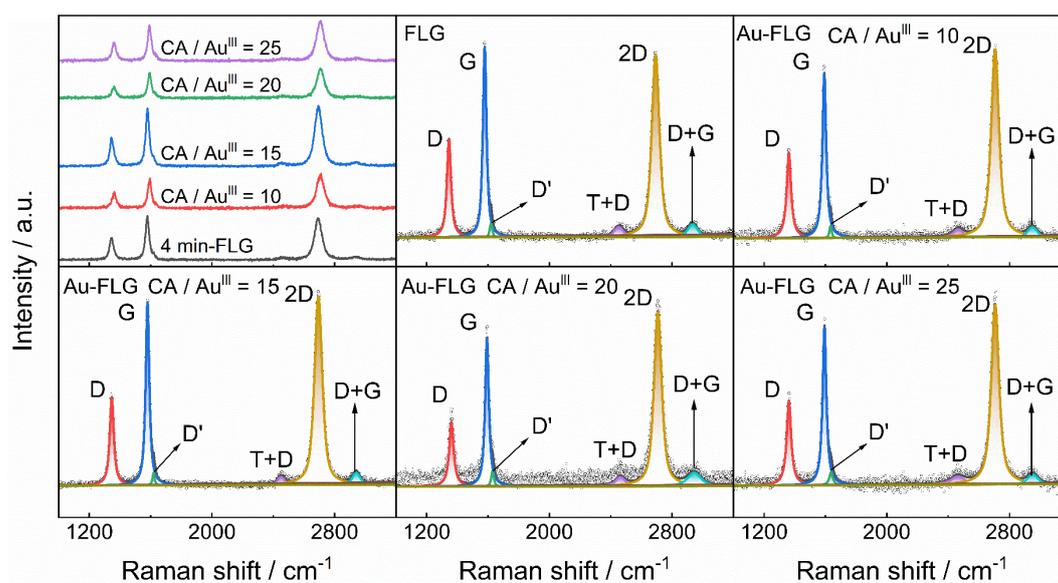
**Fig. S11.** C 1s, O 1s, Ti 2p, and Au 4f core-level XPS profiles of an Au-FLG sample with a CA/Au<sup>III</sup> volume ratio of 20 and a growth time of 4 min. Au-FLG, gold-doped few-layer graphene; XPS, X-ray photoelectron spectrometry



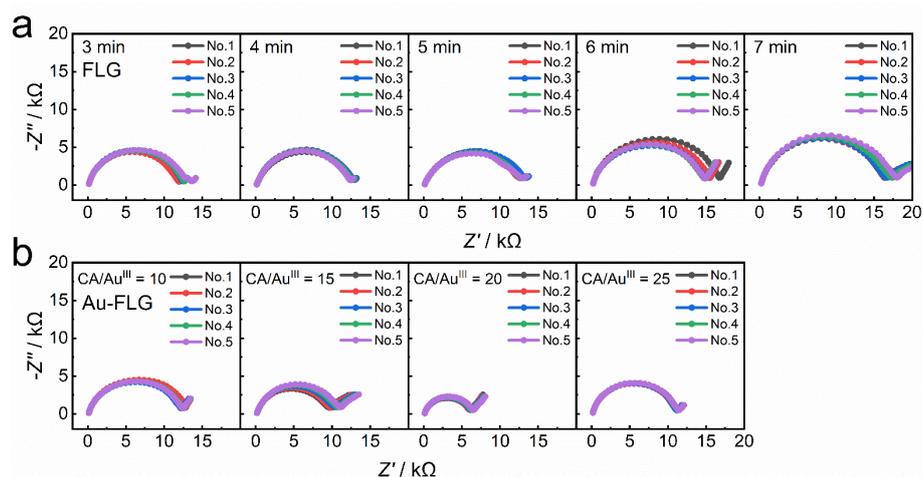
**Fig. S12.** XRD patterns of the five FLG samples with growth times of 3–7 min. FLG, few-layer graphene; XRD, X-ray diffraction



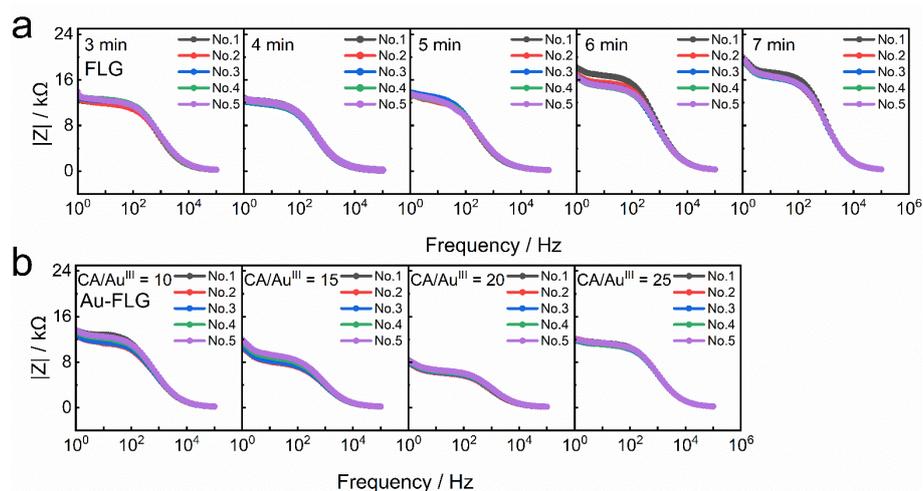
**Fig. S13.** XRD patterns of the Au-FLG samples prepared with CA/Au<sup>III</sup> concentration ratios of 10–25 and a growth time of 4 min. Au-FLG, gold-doped few-layer graphene; XRD, X-ray diffraction



**Fig. S14.** Raman spectra of the 4 min-FLG and four Au-FLG samples with different CA/Au<sup>III</sup> volume ratios and a growth time of 4 min. Au-FLG, gold-doped few-layer graphene



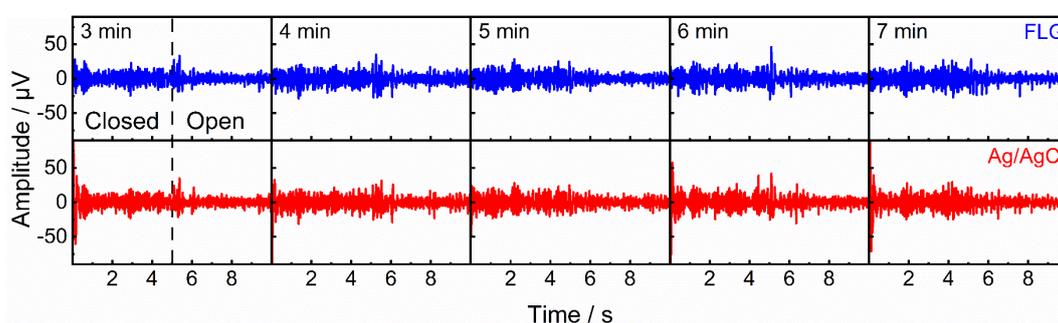
**Fig. S15.** Nyquist plots obtained through EIS using different FLG and Au-FLG electrodes on the forearm skin: (a) with FLG growth times of 3–7 min, and (b) Au-FLG/Ti prepared with CA/Au<sup>III</sup> ratios of 10–25 and a growth time of 4 min. Au-FLG, gold-doped few-layer graphene; EIS, electrochemical impedance spectroscopy



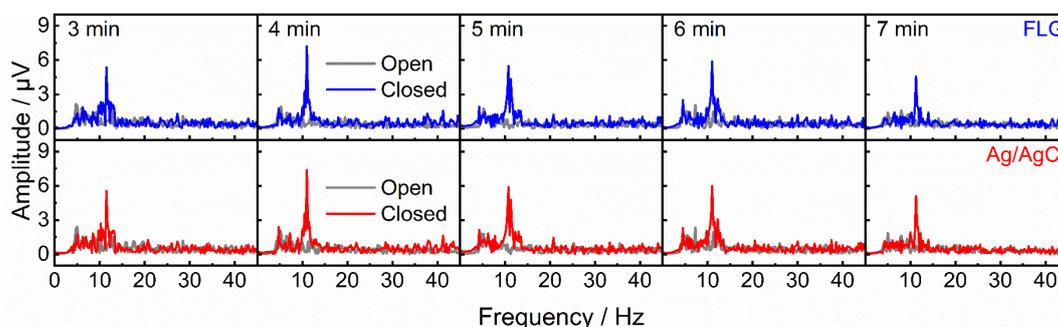
**Fig. S16.** Frequency-impedance plots obtained through EIS using different FLG and Au-FLG electrodes on the forearm skin: (a) with FLG growth times of 3–7 min, and (b) Au-FLG/Ti prepared with CA/Au<sup>III</sup> concentration ratios of 10–25 and growth times of 4 min. Au-FLG, gold-doped few-layer graphene; EIS, electrochemical impedance spectroscopy



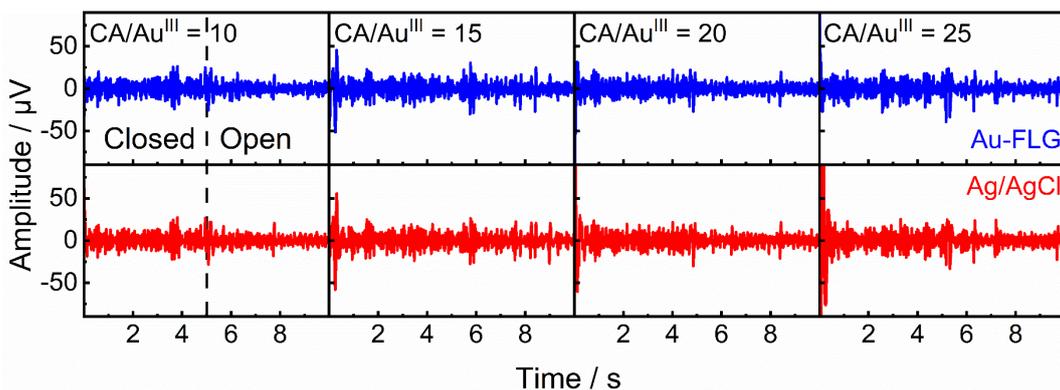
**Fig. S17.** Position of Au-FLG and Ag/AgCl electrodes on the scalp (Fp1).



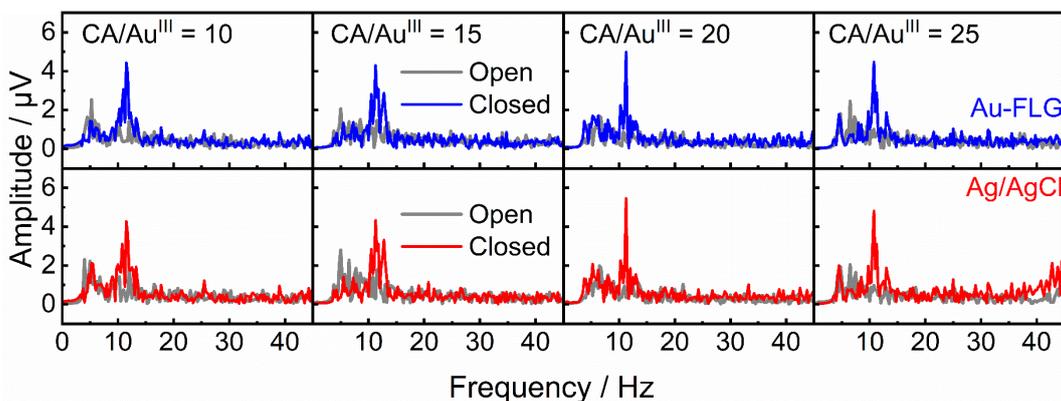
**Fig. S18.** EEG signals collected from the forehead Fp1 using FLG electrodes with different growth times of 3–7 min and gel-Ag/AgCl electrodes for the closed/open eye paradigm. Each cycle is 10 s, with eyes closed and opened each for 5 s. FLG, few-layer graphene; EEG, electroencephalogram



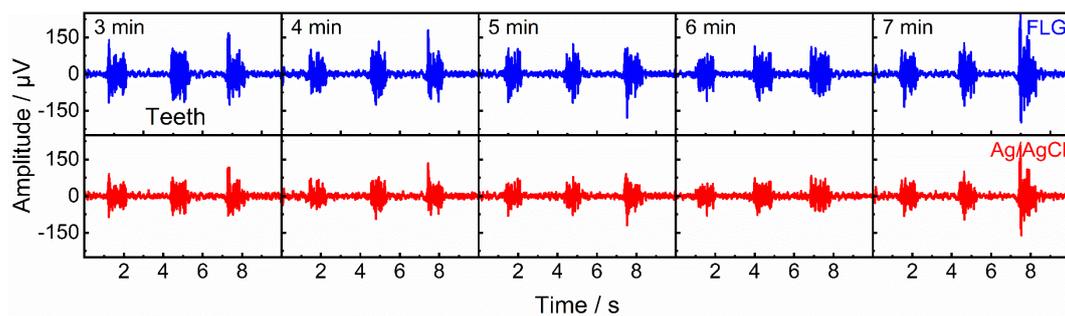
**Fig. S19.** Amplitude spectra of the EEG signals for the closed/open eyes paradigm collected from the forehead Fp1 using five FLG electrodes with different growth times of 3–7 min and gel-Ag/AgCl electrodes. FLG, few-layer graphene; EEG, electroencephalogram



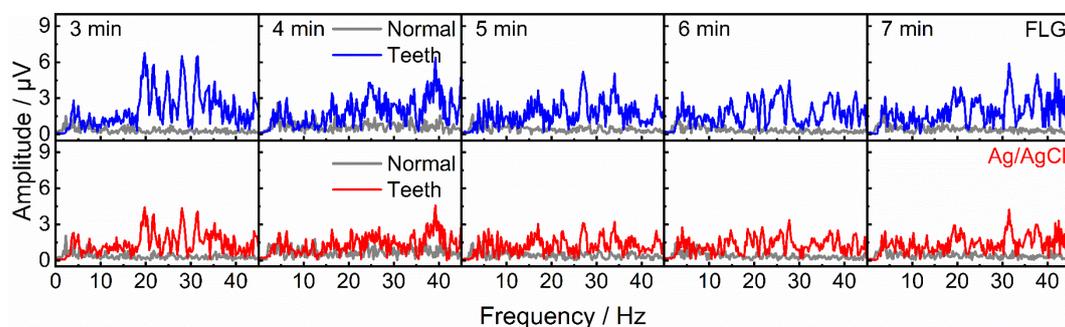
**Fig. S20.** EEG signals collected from the forehead Fp1 using four Au-FLG electrodes with CA/Au<sup>III</sup> ratios of 10–25 and gel-Ag/AgCl wet electrodes for the closed/open eye paradigm. Each cycle is 10 s, with eyes closed and opened each for 5 s. Au-FLG, gold-doped few-layer graphene; EEG, electroencephalogram



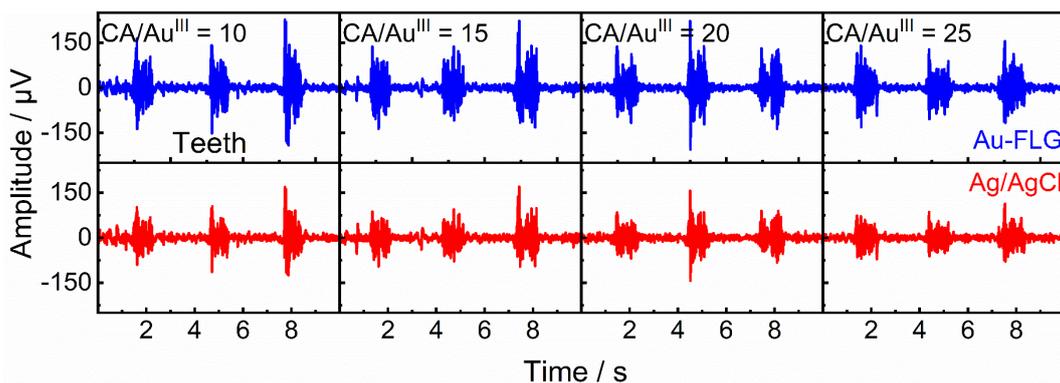
**Fig. S21.** Amplitude spectra of the EEG signals for the closed/open eyes paradigm collected from the forehead Fp1 using four Au-FLG electrodes with CA/Au<sup>III</sup> ratios of 10–25 and a gel-Ag/AgCl wet electrode. EEG, electroencephalogram; Au-FLG, gold-doped few-layer graphene



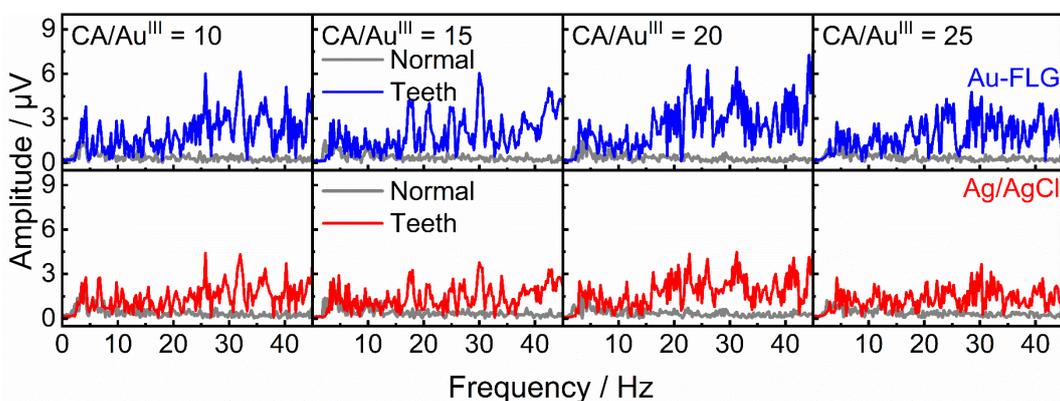
**Fig. S22.** EMG signals collected from the forehead Fp1 using FLG electrodes with different growth times of 3–7 min and gel-Ag/AgCl electrodes for the teeth-grinding paradigm. Teeth are rubbed every 3 s. EMG, electromyogram; FLG, few-layer graphene



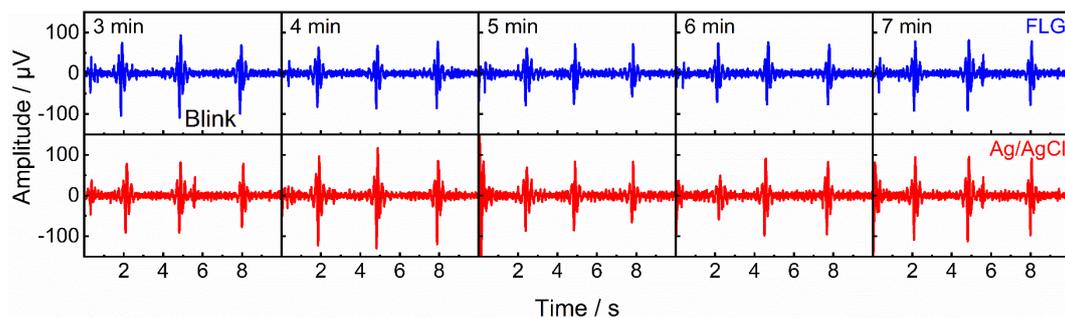
**Fig. S23.** Amplitude spectra of the EMG signals for the teeth-grinding paradigm collected from the forehead Fp1 using five FLG electrodes with different growth times of 3–7 min and gel-Ag/AgCl electrodes. EMG, electromyogram; FLG, few-layer graphene



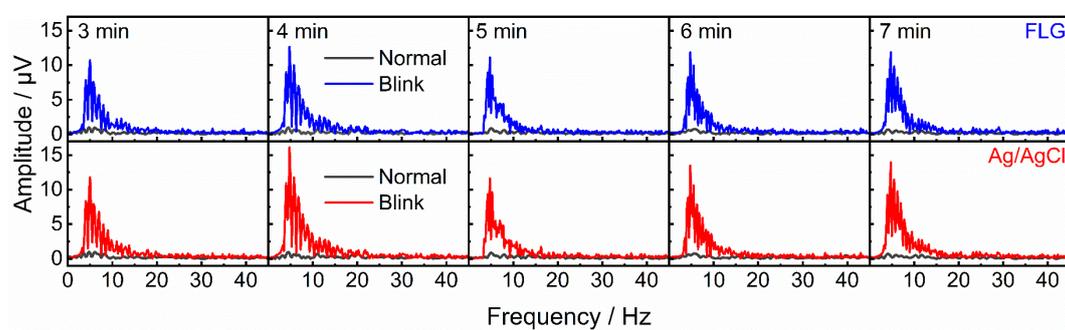
**Fig. S24.** EMG signals collected from the forehead Fp1 using four Au-FLG electrodes with different  $CA/Au^{III}$  ratios of 10–25 (grown for 4 min) and gel-Ag/AgCl electrodes, with the teeth-grinding paradigm. Teeth are rubbed every 3 s. Au-FLG, gold-doped few-layer graphene; EMG, electromyogram



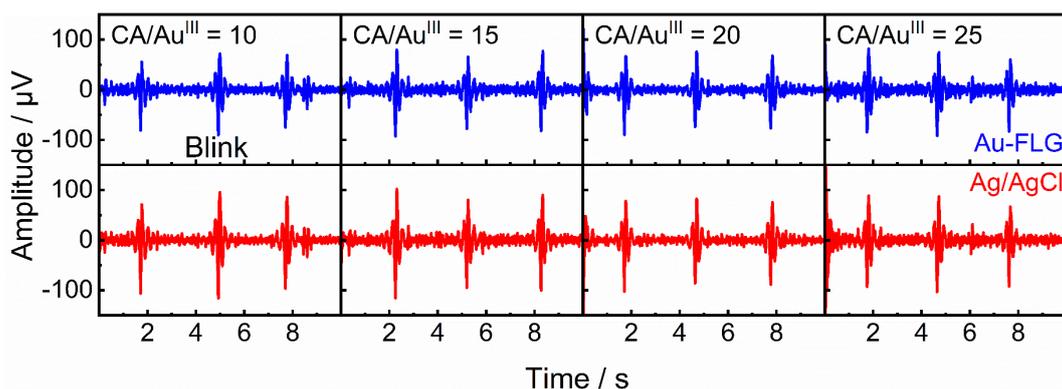
**Fig. S25.** Amplitude spectra of the EMG signals for the teeth-grinding paradigm collected from the forehead Fp1 using four Au-FLG electrodes with different  $CA/Au^{III}$  ratios of 10–25 (growth time: 4 min) and gel-Ag/AgCl electrodes. Au-FLG, gold-doped few-layer graphene; EMG, electromyogram



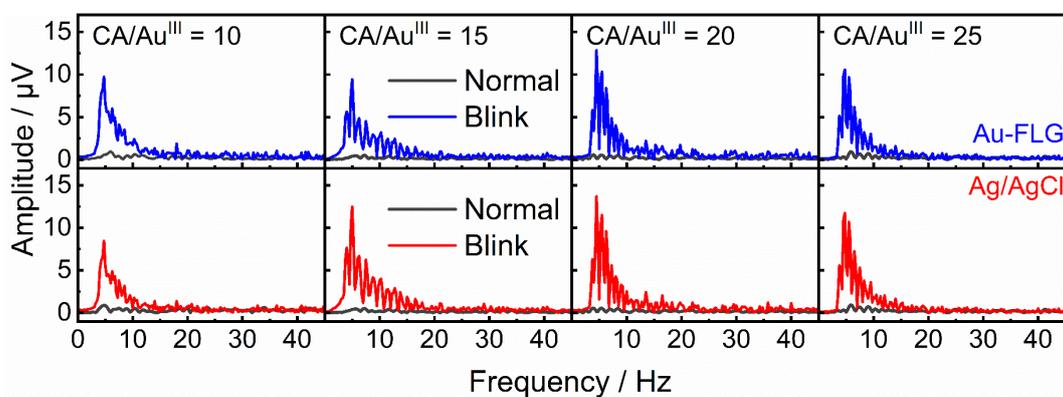
**Fig. S26.** EOG signals collected from the forehead Fp1 using FLG electrodes with different growth times of 3–7 min and gel-Ag/AgCl dry electrodes for the eye-blinking paradigm. The eyes are blinking once every 3 s. EOG, electrooculogram; FLG, few-layer graphene



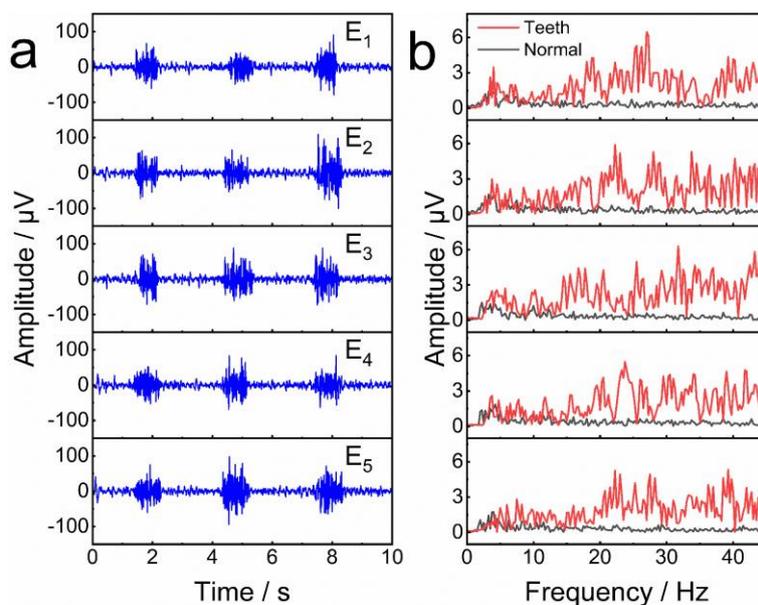
**Fig. S27.** Amplitude spectra of the EOG signals for the eye-blinking paradigm collected from the forehead Fp1 using five FLG dry electrodes with different growth times of 3–7 min and gel-Ag/AgCl electrodes. EOG, electrooculogram; FLG, few-layer graphene



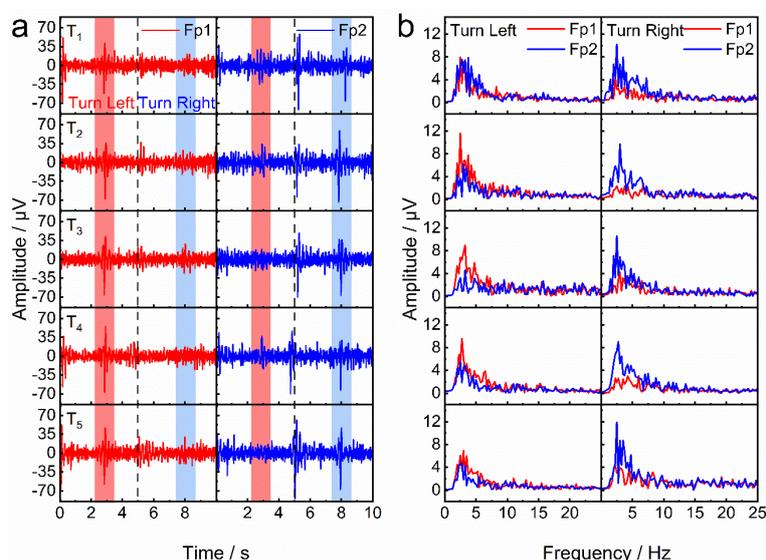
**Fig. S28.** EOG signals for the eye-blinking paradigm collected from the forehead Fp1 using four Au-FLG dry electrodes with different CA/Au<sup>III</sup> ratios of 10–25 (growth time: 4 min) and gel-Ag/AgCl wet electrodes. Au-FLG, gold-doped few-layer graphene; EOG, electrooculogram



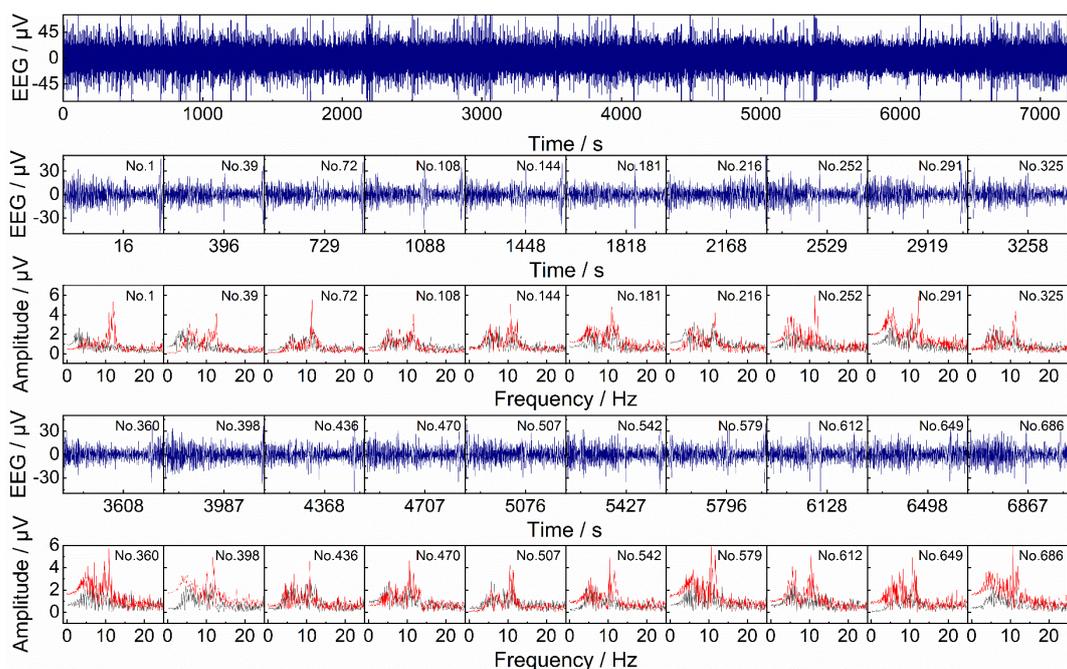
**Fig. S29.** Amplitude spectra of the EOG signals for the eye-blinking paradigm collected from the forehead Fp1 using four Au-FLG dry electrodes with different CA/Au<sup>III</sup> ratios of 10–25 (growth time: 4 min) and gel-Ag/AgCl wet electrodes. Au-FLG, gold-doped few-layer graphene; EOG, electrooculogram



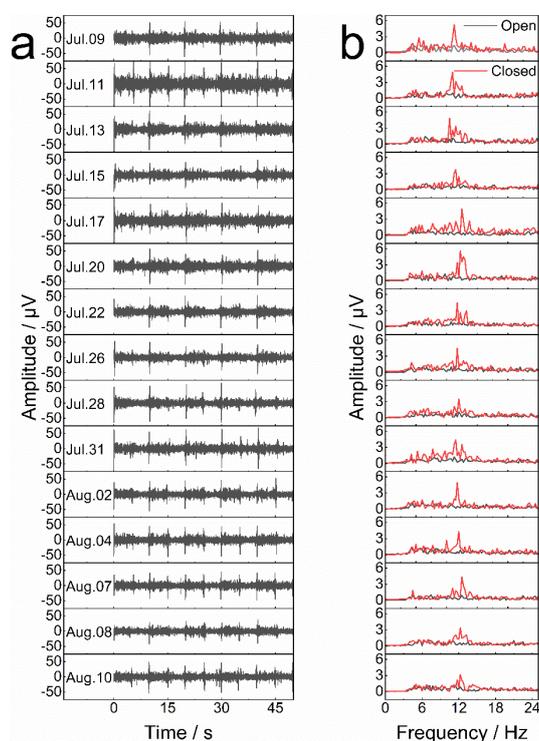
**Fig. S30.** Reproducibility. (a) Time- and (b) frequency-domain EMG spectra collected from the forehead Fp1 position using five Au-FLG electrodes with a CA/Au<sup>III</sup> volume ratio of 20 in the teeth-grinding paradigm. Teeth are rubbed every 3 s. Au-FLG, gold-doped few-layer graphene; EMG, electromyogram



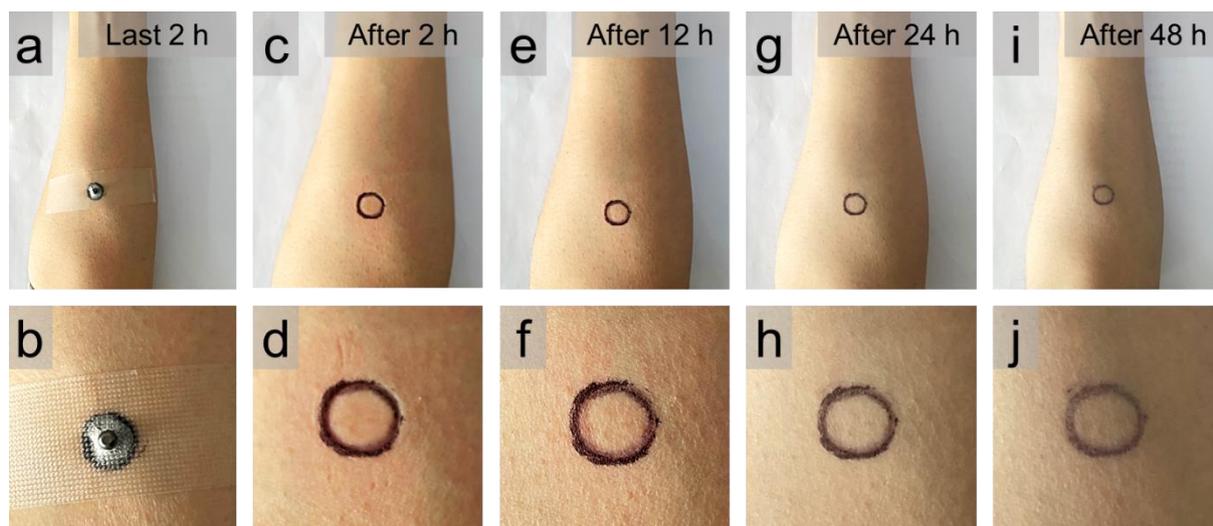
**Fig. S31.** Five repeatability experiments. (a) Time- and (b) frequency-domain spectra collected from the forehead Fp1 and Fp2 positions using one Au-FLG electrode with the CA/Au<sup>III</sup> ratio of 20. A cycle lasts for 10 s, with the eyeball turning left at 3 s and right at 8 s. Au-FLG, gold-doped few-layer graphene



**Fig. S32.** Short-term stability of an Au-FLG electrode for 2 h. The figure contains both time-domain and frequency-domain spectra. Cycle of 10 s, with the serial number indicating the order of recording for each cycle.



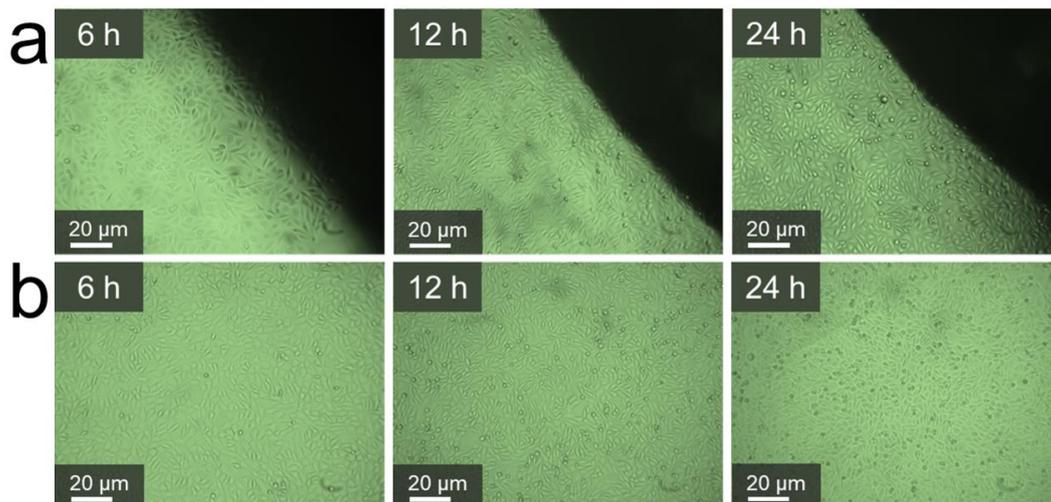
**Fig. S33.** Long-term stability of the Au-FLG electrode: (a) Time-domain diagram. (b) Frequency-domain diagram. Au-FLG, gold-doped few-layer graphene



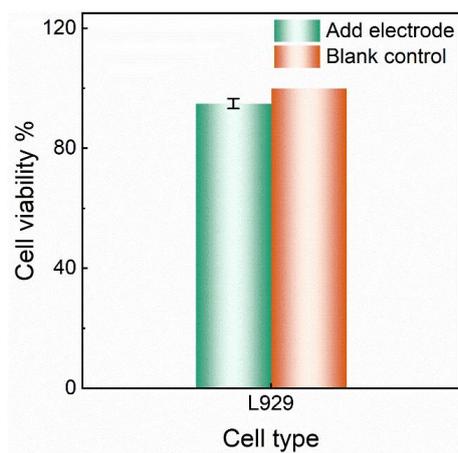
**Fig. S34.** Skin irritation. (a, b) Au-FLG button electrode attached to the forearm skin. After removing the electrode for (c, d) 2 h, (e, f) 12 h, (g, h) 24 h, and (i,j) 48 h.



**Fig. S35.** Photograph of L929 cells cultured in environments with and without Au-FLG electrode.



**Fig. S36.** Optical microscope images of L929 cells during cultivation in the (a) presence and (b) absence of Au-FLG electrode.



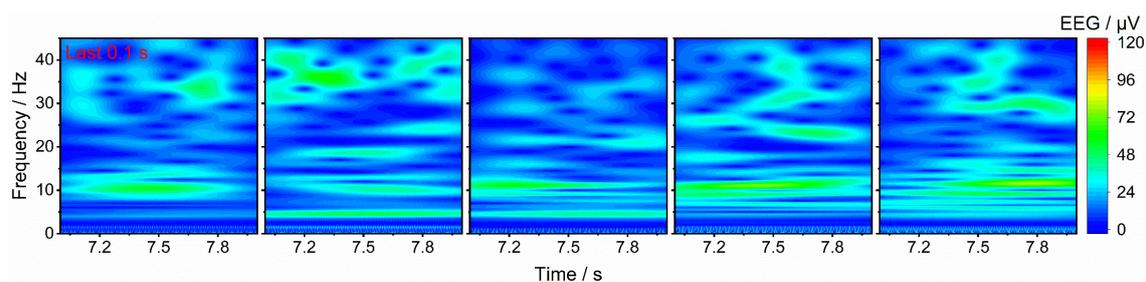
**Fig. S37.** Cell viability of L929 in culture medium with and without Au-FLG electrode.

Comfort Evaluation Form

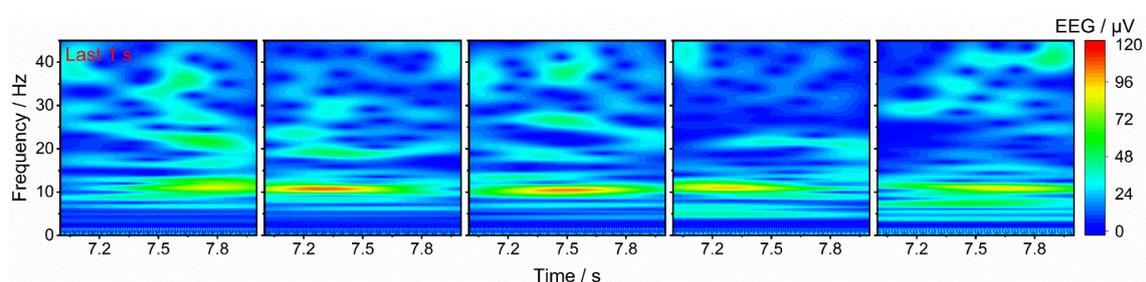
Sub jects	Wearing time			Pain				Comfort			Sign
	Long	Middle	Short	High	Middle	Low	No	High	Middle	Low	
1			✓				✓	✓			Mo kun Li
2	✓						✓	✓			Hao Bai
3			✓				✓		✓		Zhibo Zhuang
4		✓			✓				✓		Jinzhe Bao
5			✓				✓	✓			Yuhang Zheng
6			✓				✓	✓			Xinyu Zhang
7			✓				✓	✓			Zhiyu Tao
8			✓				✓	✓			Mingyu Mo
9			✓				✓	✓			Maisheng Tang
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11			✓				✓	✓			chaoyan Li
12			✓				✓	✓			Yihang Wu
13			✓				✓	✓			Junhao Jia
14			✓				✓	✓			JiaWei Liu
15			✓			✓		✓			Ruiyi Rong
16		✓					✓	✓			Bing Zhang
17			✓				✓	✓			Jiahui Tang
18			✓				✓	✓			Xiaolong Wu
19			✓				✓	✓			Jie Chen
20			✓				✓		✓		Ziteng Li
21			✓				✓	✓			shiming Chen
22			✓				✓	✓			Jiao Zu
23			✓				✓	✓			Zhiming Chen
24			✓				✓	✓			Yunfei Yang
25	✓		✓				✓	✓			Yongtan Ma
26			✓			✓				✓	Lianzheng Ren
27			✓				✓	✓			Chunxia Gao
28			✓				✓	✓			Zhanxin Shen
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Yuhang Zheng Apr. 12. 2024

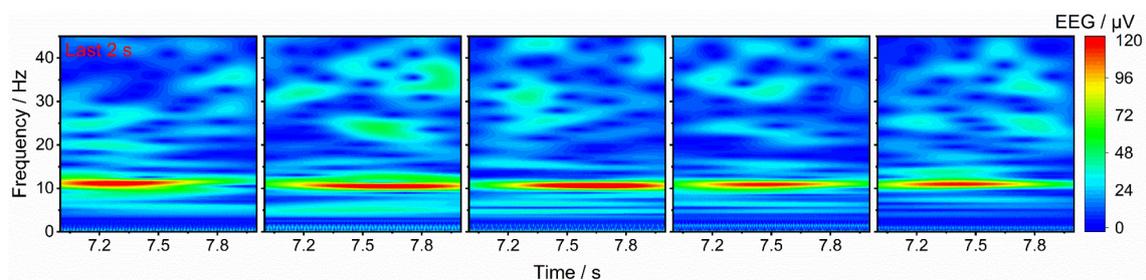
Fig. S38. Comfort evaluation form



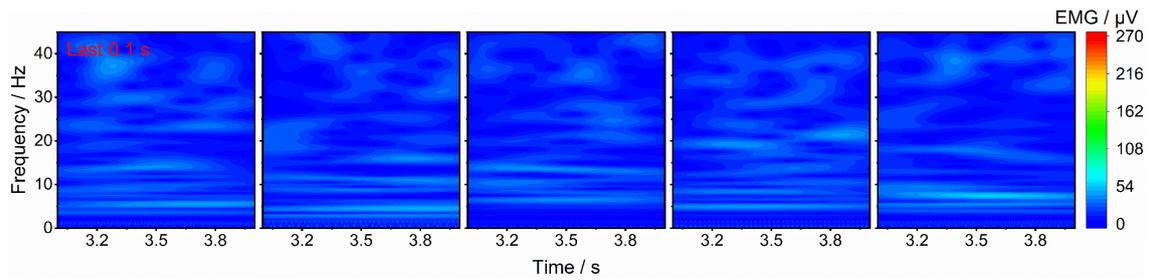
**Fig. S39.** Time-frequency spectra of EEG signals recorded using an Au-FLG button electrode in the Fp1 channel of the forehead, with eyes closed for 0.1 s. The figure provides five experimental results. Au-FLG, gold-doped few-layer graphene; EEG, electroencephalogram



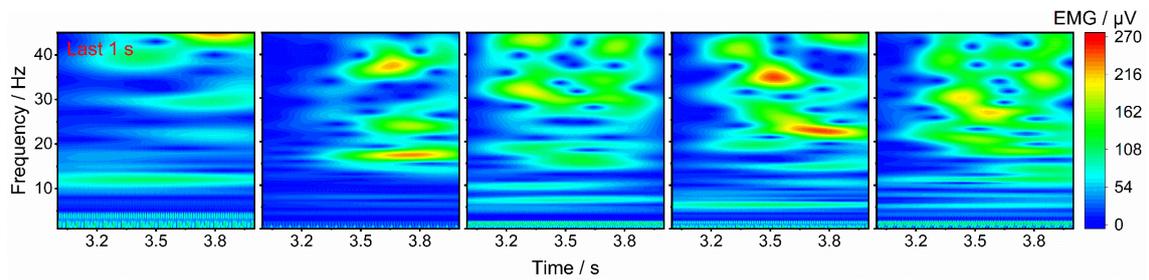
**Fig. S40.** Time-frequency spectra of EEG signals recorded using an Au-FLG button electrode in the Fp1 channel of the forehead, with eyes closed for 1 s. Au-FLG, gold-doped few-layer graphene; EEG, electroencephalogram



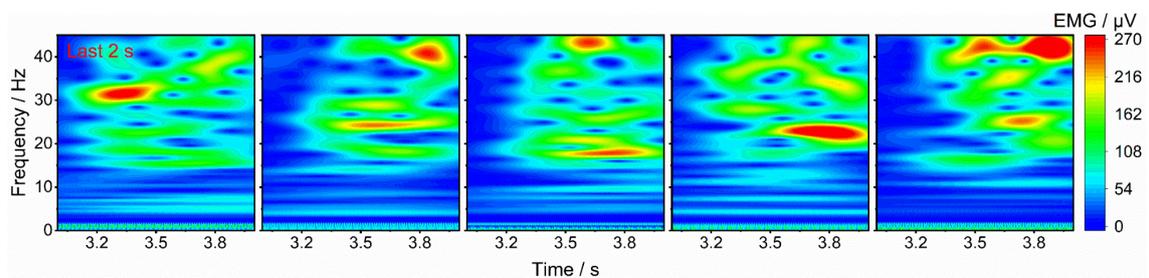
**Fig. S41.** Time-frequency spectra of EEG signals recorded using an Au-FLG button electrode in the Fp1 channel of the forehead, with eyes closed for 2 s. Au-FLG, gold-doped few-layer graphene; EEG, electroencephalogram



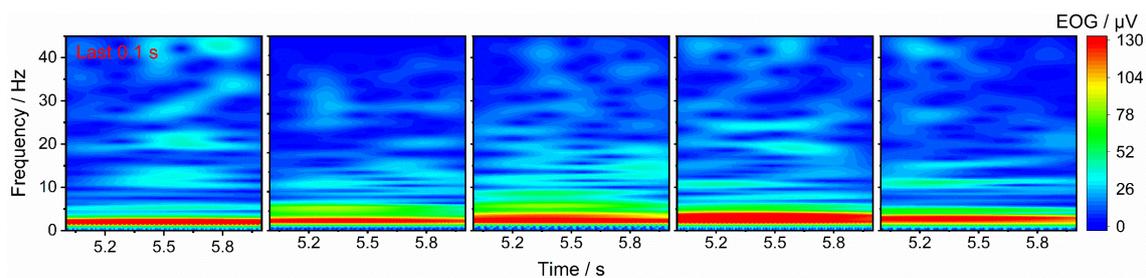
**Fig. S42.** Time-frequency spectra of EMG signals recorded on the Fp1 channel of the forehead using an Au-FLG button electrode with the grinding teeth for 0.1 s. Au-FLG, gold-doped few-layer graphene; EMG, electromyogram



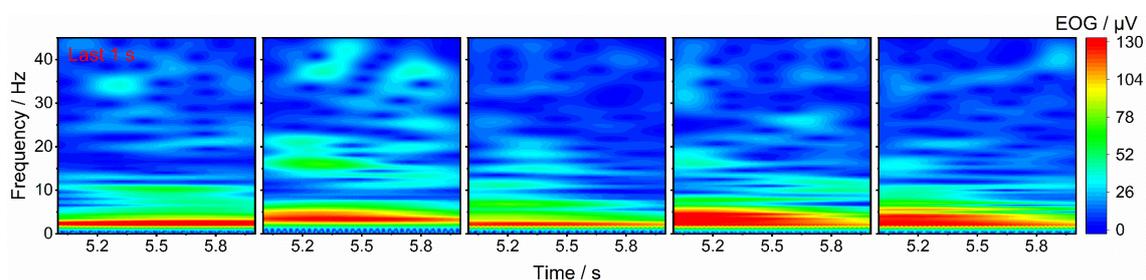
**Fig. S43.** Time-frequency spectra of EMG signals recorded on the Fp1 channel of the forehead using an Au-FLG button electrode with the grinding teeth for 1 s. Au-FLG, gold-doped few-layer graphene; EMG, electromyogram



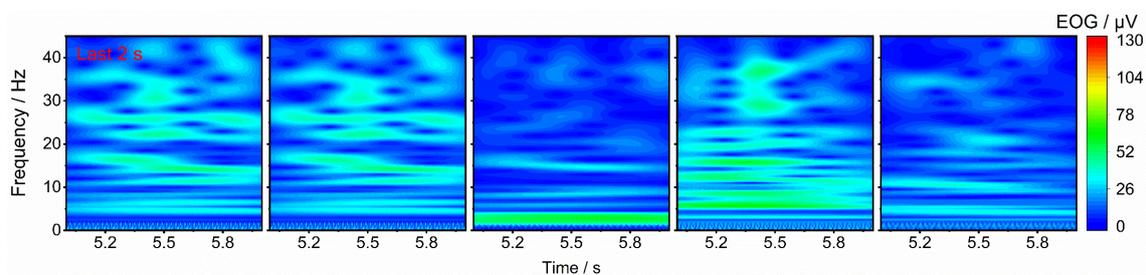
**Fig. S44.** Time-frequency spectra of EMG signals recorded on the Fp1 channel of the forehead using an Au-FLG button electrode with the grinding teeth for 2 s. Au-FLG, gold-doped few-layer graphene; EMG, electromyogram



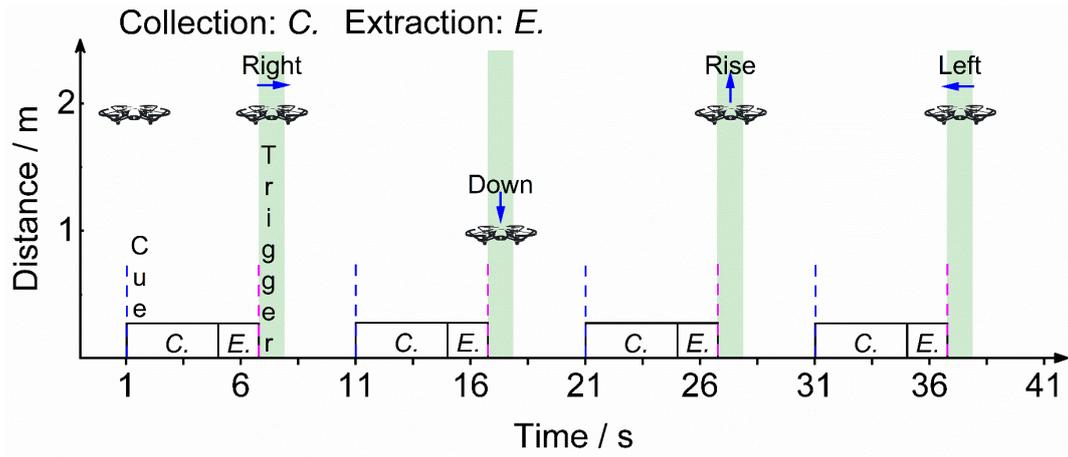
**Fig. S45.** Time-frequency spectra of EOG signals recorded on the Fp1 channel of the forehead using an Au-FLG button electrode with the eyeball turned to the left for 0.1 s. Au-FLG, gold-doped few-layer graphene; EOG, electrooculogram



**Fig. S46.** Time-frequency spectra of EOG signals recorded on the Fp1 channel of the forehead using an Au-FLG button electrode with the eyeball turned to the left for 1 s. Au-FLG, gold-doped few-layer graphene; EOG, electrooculogram



**Fig. S47.** Time-frequency spectra of EOG signals recorded on the Fp1 channel of the forehead using an Au-FLG button electrode with the eyeball turned to the left for 2 s. Au-FLG, gold-doped few-layer graphene; EOG, electrooculogram



**Fig. S48.** A brainwave game plan for controlling UAVs for image monitoring.