Supplementary Materials for

Self-powered user-interactive electronic skin for programmable touch operation platform

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Note S1. Synthesis and Characterization of ZnS particles

In the amorphous ZnS (luminescent grade), 0.05 wt.% CuSO$_4$, 0.1 wt.% Al$_2$(SO$_4$)$_3$, 1 wt.% NaI and 1 wt.% NaBr were added. The mixed powder was calcined in a tube furnace at 930 °C for 1 hour. Among them, the quartz tube of the tube furnace is wrapped with activated carbon at both ends to prevent the powder from being oxidized during the calcination process. The reaction atmosphere is close to atmospheric pressure due to the evaporation of sulfur at the calcination temperature. The furnace was then naturally cooled to room temperature. After the firing process was completed, the powder was washed with water and HCl. The powder was immersed in deionized water at 80 °C and stirred for 10 minutes and then allowed to stand. Repeat this step several times. After the powder was precipitated, water was removed and washed with a 2 wt % dilute HCl solution. The powder was then washed several times with deionized water to remove residual HCl solution. After ball milling for 12 hours, the obtained powder was dried at 120 °C in a vacuum atmosphere to obtain desired luminescent powder. The as-synthesized ZnS: Cu, Al powder is granular particulates with a diameter ranging about 5-20 µm, as shown in Fig. S1B, C. The XRD pattern in Fig. S1D indicates that the ZnS: Cu, Al powder has a sphalerite structure. ZnS as the solid solution matrix for ZnS: Cu, Al has two crystal structures of sphalerite (cubic structure-β phase) and wurtzite (hexagonal structure-α phase), which mainly depends on the calcination temperature (1020 °C for pure ZnS) during the preparation. Past studies have shown that sphalerite-ZnS has higher electroluminescence intensity than wurtzite-ZnS.

Note S2. The triboelectric-optical model in contact-separation mode

When the human skin is in full contact with the SUE-skin (Fig. S2A), the human skin and the dielectric adhesive acquire net positive and net negative triboelectric charges, respectively. Once
the human skin with the positively charged surface starts to slide outward (Fig. S2B), the in-plane charge separation is initiated, and an induced current is generated in the external circuit. During this process, there is a varying electric field between the top surface of the dielectric binder and the electrode. The electrons produced within the lattice by impact ionization move toward the bottom of the ZnS phosphor particle under the induction of the above varying electric field. During the movement, some of the electrons impact the luminescence centers, thereby exciting the luminescence centers and causing electroluminescence. When the human skin fully slides out of the SUE-skin (Fig. S2C), no current is generated in the external circuit. At the same time, the electrons have been concentrated at the bottom of the ZnS phosphor particle under the electric field induction and will not impact the luminescence centers, so the device does not emit light. When the human skin slides backward (Fig. S2D), an induced current is generated again in the external circuit. During this process, the electric field between the upper surface of the dielectric binder and the electrode gradually disappears. The electrons inside the ZnS that lose the effect of the electric field move in reverse and impact with the luminescence centers again to excite it and cause electroluminescence again.

**Note S3. The concept of pre-stress**

The “pre-stress” is the pressure applied to the SUE-skin through the glass for close contact between the glass and the SUE-skin before measuring the output signal of the SUE-skin. The purpose of applying the pre-stress is to determine whether the output signal of the SUE-skin originates from the triboelectric effect or the piezoelectric effect. In detail, if the glass and the SUE-skin are not in close contact, triboelectricity will occur during the pressing process. However, if the two are in close contact, triboelectricity will not occur when pressed. As shown in Fig. S3, when the glass and the SUE-skin are completely separated, the pressure between the
glass and the SUE-skin is zero. The greater the pre-stress applied, the closer the contact between the glass and the SUE-skin. During the measurement, adding stress is added to the pre-stress so that the final pressure is equal to 300kPa. As Fig. 2C, when the pre-stress is higher than 240kPa and adding stress (60 kPa) is applied, the SUE-skin does not generate an output signal. If the touch luminescence of SUE-skin is caused by the piezoelectric effect, there will be emitting light as long as adding stress is applied. For the triboelectric effect, if the two friction surfaces are in close contact, no output signal will be generated when adding stress is applied. The pre-stress of 240 kPa makes the two friction surfaces in close contact, so no output signal is generated when additional stress is applied in Fig. 2C. This result indicates that the output signals of the SUE-skin come from the triboelectric effect instead of the piezoelectric effect.

**Note S4. Description of the TL /EL /PL spectrum**

In these tests, to ensure the controllability and comparability of the test, glass was used as a substitute for human skin for contact-separation friction with a contact pressure of 100 kPa. Figure 2D is the emission spectrum when the SUE-skin is touched. Since this luminescence phenomenon is caused by friction, it is conventionally referred to as triboluminescence (TL). The center of the TL spectrum is at 543 nm, which is attributed to the electron transition from the shallowly electronic trap to the t2 or e levels level of Cu$^{2+}$. Then, we fabricated a device, as shown in Fig. S4, to obtain the electroluminescence (EL) spectrum of the synthesized ZnS powder. The ZnS and PDMS were mixed at 1:1, and two ITO glasses were bonded to the upper and lower surfaces. By applying an alternating current of 50 Hz between the two ITO electrodes, an EL spectrum of ZnS, as shown in Fig. 2E, can be obtained. The center of the EL spectrum is at 537 nm, which is also attributed to the electron transition from the shallowly electronic trap to the t2 or e levels level of Cu$^{2+}$ ($40$). The characteristics of the TL and EL spectra are very close.
The small difference between the centers of the two emission spectra is mainly because when the frequency of the alternating electric field applied to ZnS is different, it will cause the electrons to transition from the shallow electronic trap to the t2 level or e level of Cu$^{2+}$ in different relative probability. Besides, we can see from the above figure that when the alternating current voltage applied between the two ITO electrodes is continuously increased, the EL intensity of ZnS is also gradually improved, and the spectral center remains unchanged. The law of change is the same as the variation of the touch luminescence intensity of the SUE-skin with the magnitude of the applied stress. Our proposed triboelectric-optical model indicates that the magnitude of the applied touch stress directly affects the strength of the triboelectrification. Under the same conditions, the higher the triboelectrification intensity, the greater the intensity of the EL induced by it. Figure 2F is the photoluminescence (PL) spectrum of ZnS. The PL spectrum can be deconvolved into two luminescence peaks centered at 451 nm and 543 nm, respectively. The emission at 451 nm is caused by the recombination of natural defect states such as sulfur vacancies (40). This result indicates that the touch luminescence of the SUE-skin is not PL induced by ultraviolet rays or x-rays generated by friction. The characterization of ZnS EL and PL performance confirms the correctness of the triboelectric-optical model.

**Note S5. The liquid flows through the surface of the SUE-skin**

In the study, we found that not only the visible light is generated when the solid contacts with the SUE-skin surface, but also when the liquid slides over the SUE-skin surface, it also produces instantaneous visible light on the trajectory of the liquid slip. This is a phenomenon that has not been reported yet. The core of the triboelectric-optical model is the induction of electrostatic induction and electroluminescence by triboelectrification. Based on a large number of research results in recent years, the former almost no need to prove, but the strict argument of the latter is
challenging. From actual results, we may call the phenomenon of triboelectrically induced electroluminescence directly as triboluminescence. Similar to triboelectrification, triboluminescence is also an ancient phenomenon (Fig. S5A). As early as 1605, Francis Bacon reported the phenomenon of frictional luminescence produced when grinding sugar cubes. So far, it has been found that more than 50% of the crystals have the property of triboluminescence. Although the mechanism of triboluminescence appears to be very complicated, it mainly includes friction-induced electroluminescence, photoluminescence, electroluminescence (piezoelectric effect, or lattice distortion), and thermoluminescence. Therefore, when we prove the triboelectric-optical model, it is not enough to compare the emission spectrum of SUE-skin with the electroluminescence spectrum of ZnS. We also need sufficient evidence to verify that the luminescence produced by the touch SUE-skin is not caused by friction-induced photoluminescence, electroluminescence, and thermoluminescence. Above, we confirmed that the luminescence phenomenon generated by the touch SUE-skin is not caused by the photoluminescence caused by friction by comparing the emission spectrum of SUE-skin with the photoluminescence spectrum of ZnS. However, since the touch process is inevitably accompanied by the application of stress and frictional heat generation, it is tough to prove by traditional methods that the luminescence phenomenon generated by the touch SUE-skin is not caused by friction-induced electroluminescence and thermoluminescence. However, because the liquid is different from many properties of solids, the characterization of this phenomenon is an excellent proof of our proposed triboelectric-optical model. Taking deionized water as an example, Fig. S5B shows the luminescence mechanism of a single water droplet and a continuous plurality of water droplets sliding across the SUE-skin’s surface. The only thing to note is that direct contact of ZnS with liquids such as water significantly reduces the intensity of
the luminescence, so the SUE-skin surface here requires an insulating layer (also as triboelectrification layer). Figure S5C compares the emission spectrum of the touch SUE-skin and the emission spectrum of the droplet as it slides across the SUE-skin surface. The consistency shows that the luminescence mechanism of the two is the same. The luminescence intensity of different liquids as they slide across the SUE-skin surface is compared in Fig. S5D. Viscous liquids such as cooking oil have a significantly weaker luminescence than water since viscous liquids adhere to the surface after contact with the SUE-skin and are difficult to separate. The volume of a single water droplet used in the test was about 10 μL, and the instantaneous contact area when the SUE-skin surface slipped was greater than 10 mm². Through a rough estimation, we can know that the pressure generated by a single water droplet on the surface of the SUE-skin is about 0.01 to 0.1 kPa. Compared to the pressure range described in Fig. 2B, the pressure applied by a single water droplet on the SUE-skin surface is negligible. Under such a low pressure, the luminescence phenomenon obviously does not come from mechanoluminescence (the luminous stress threshold is at the MPa level). Besides, when a liquid such as water slides over the SUE-skin surface, since the contact stress is very small and the specific heat capacity of the droplet is large, frictional heat is hardly generated, which is a suitable proof that the luminescence generated by SUE-skin is not thermoluminescence caused by friction.

**Note S6. Dual-signal superposition for highly robust touch track monitoring**

Touch track sensing and visualization have broad application prospects in interactive input/control devices, robotics, and medical/health monitoring. The traditional approach of track monitoring is to use mechanical-to-electrical sensing or mechanical-to-optical sensing, respectively. However, mechanical-to-electrical sensing could be affected by the integrity of the
electrode and circuit. Moreover, the surrounding magnetic fields and electrical equipment could also interfere with the sensing process. Besides, mechanical-to-optical sensing is very susceptible to ambient light, viewing angles, and obstacles. Therefore, the use of mechanical-to-electrical or mechanical-to-optical sensing alone could result in poor monitoring accuracy or even errors due to insufficient sensing robustness. If the mechanical-to-electrical and mechanical-to-optical energy conversion can be used simultaneously to realize the monitoring of the touch track, the robustness of the monitoring system can be significantly improved by the superposition of the electrical and the optical signals. Here, an SUE-skin with 36 electrodes (6 * 6 matrix) was fabricated. The matrix design is achieved by patterning the electrodes, each electrode corresponding to a channel of electrical signal outputs. Under the stimuli of sliding touch, the optical photograph of instantaneous optical output obtained by the time-lapse photography corresponds to the touch tracks, as shown in Fig. S10A. The picture in Fig. S10B is the electric readout mapping plotted at the moment when human skin arrives at the center of particular electrodes intuitively display the path that the human skin is sliding over. To make the output voltage mapping more accurately reflects the touch tracks and more conveniently compares with the optical photograph of optical output, we reconstructed the mapping by algebraic reconstruction technique (ART) in MATLAB, as shown in Fig. S10C. In this way, every time a sliding touch occurs, a pair of pictures (from the optical signal and the electrical signal, respectively) would be getting, and then the two signals can be superimposed by the logical operation of the pair of pictures in LabView, which is embedded with NI Vision development module. To illustrate the effect of dual signals superposition on improving the robustness of touch track sensing, a set of cases is demonstrated. The results of the corresponding single signals and binary signals obtained by superposition are shown in Fig. S10D. The six images (α,
β, γ, 1, 2, 3) are gathered with the same shape (N shape) of the sliding touch tracks. Image α is the optical photograph of the visual output collected when there is no obstruction. Image β and γ are the optical photographs in which parts of the tracks are blocked. Image 1 is the reconstructed mapping when the circuit is complete. Image 2, 3 are the reconstructed mappings in which some of the electrodes are broken. The nine orange images (α-1, α-2, α-3, β-1, β-2, β-3, γ-1, γ-2, γ-3) are corresponding superimposed images by OR logical operation. In this set of cases, when using a single-signal source (whether optical or electrical) for touch track monitoring, the accuracy is only 1/3. While using dual-signal sources (by superimposing the optical and electrical signals), the accuracy rate has been dramatically improved to 8 / 9. Dual-signal superposition is more robust than any single signal, which is essential for improving the accuracy and usefulness of touch track monitoring and visualization. Moreover, the weight of the SUE-skin for the programmable touch operation platform is 4.2 g and the weight of the SUE-skin for the dual-signal superposition is 3.8 g.
Fig. S1. Characterization of luminous properties, structure and mechanical stability of the SUE-skin. (A) The optical photograph of the visual output during a sliding touch in ambient condition. (B, C) SEM characterization of ZnS: Cu, Al. (D) XRD pattern of ZnS: Cu, Al. (E) Comparison of luminous brightness between ZnS: Cu and the synthesized ZnS: Cu, Al. (F) SUE-skin luminous intensities (every 40 cycles), (G) time-lapse photos (every 400 cycles) and (H) surface SEM images (every 400 cycles) during and after 1200 cycles of bending. (I) The SUE-skin’s luminous intensities (every 40 cycles), (J) time-lapse photos (every 400 cycles) and (K) surface SEM images (every 400 cycles) during and after 1200 cycles of rolling. [Photo credit for A, G, J: Xuan Zhao, University of Science and Technology Beijing.]
Fig. S2. Schematic diagram of the physical model of the triboelectric-optical effect in a sliding cycle.
Fig. S3. Schematic diagram of the pre-stress. (A) Exaggerated sketch of the pressure loading process. (B) Corresponding stress loading curve.
Fig. S4. Characterization of ZnS: Cu, Al electroluminescence.
Fig. S5. Research history of triboluminescence and the working principle and output characterization of the SUE-skin as the liquid flows through the surface.
Fig. S6. Schematic of the testing equipment.
**Fig. S7.** Characterization of Ecoflex surface morphology before and after RIE treatment and the composite film surface morphology with different mass ratios of ZnS to Ecoflex.
Fig. S8. Signal processing flowchart of the programmable touch operation platform.
Fig. S9. 156 touch interaction logics supported by the programmable touch operation platform.
Fig. S10. Dual signals superposition for highly robust touch track sensing. (A) Time-lapse optical photograph of optical output under the sliding touch. (B) Mapping of output voltage under a sliding touch. (C) Reconstructed mapping of the output voltage by algebraic reconstruction technique (ART). (D) Original photograph/mapping and superimposed images by the logical operation, green: optical photograph of optical output, blue: reconstructed mapping of the output voltage by ART, orange: superimposed images by the OR logical operation. [Photo credit for A, D: Xuan Zhao, University of Science and Technology Beijing.]
Movie S1.
Sliding touch with an N-shaped track.

Movie S2.
Sliding touch in ambient condition.

Movie S3.
Two luminescence phenomena in a contact-separation cycle.

Movie S4.
Flowing-liquid-driven luminescence.

Movie S5.
Luminescence of the ZnS-Ecoflex composite film rubbed by hand.

Movie S6.
Interactive operation demonstration of the control of the external audio module.

Movie S7.
Interactive operation demonstration of the control of the external display module (text).

Movie S8.
Interactive operation demonstration of the control of the external display module (picture).
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