Self-powered, ultrasensitive, and high-resolution visualized flexible pressure sensor based on color-tunable triboelectrification-induced electroluminescence

Li Su\textsuperscript{a,\textcopyright,c,\textcopyright}, Zhiye Jiang\textsuperscript{a}, Zhen Tian\textsuperscript{a}, Hailu Wang\textsuperscript{b}, Haojie Wang\textsuperscript{a}, Yunlong Zi\textsuperscript{c,\textcopyright,\textasteriskcentered}

\textsuperscript{a} National-Local Joint Engineering Laboratory of New Energy Photoelectric Devices, State Key Laboratory of Photovoltaic Materials & Technology, Yingli Solar, College of Physics Science and Technology, Institute of Life Science and Green Development, Hebei University, Baoding 071002, China

\textsuperscript{b} CAS Center for Excellence in Nanoscience, Beijing Key Laboratory of Micro-Nano Energy and Sensor, Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 100083, China

\textsuperscript{c} Department of Mechanical and Automation Engineering, The Chinese University of Hong Kong, Shatin, Hong Kong SAR, China

** Corresponding author.

E-mail addresses: suli@hbu.edu.cn (L. Su), ylzi@cuhk.edu.hk (Y. Zi).

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ABSTRACT

With the capability of mapping local pressure information and converting it into human-readable visual optical signals simultaneously, self-powered visualized flexible pressure sensor (SP-VFPS) has aroused a new surge of interest because of its good solution to the wireless communication of human stress/strain sensing. Herein, a new class of SP-VFPS based on micro compression-dependent porous architecture was reported to be capable of enabling color-tunable triboelectrification-induced electroluminescence (TIEL) in response to vertical pressure in real time, exhibiting a rather low detection limit of tribo-pressure at \( \approx 10 \) kPa, the high sensitivity (S > 190 kPa\textsuperscript{-1}) within a wide pressure range as well as a rapid response time of below 10 ms. Through the integration of SP-VFPS with a single-electrode triboelectric nanogenerator (TENG) matrix, the hybrid sensing system was further developed with multi-functional pressure sensitivity for multi-touch sensing, motion monitoring, and high-resolution (500 \( \mu \)m) eye-detectable spatial pressure mapping by a combination of electrical and optical responses. This SP-VFPS is the first demonstration towards color-tunable TIEL as an all-in-one device for the instantaneous detection of pressure information which may provide new insight into its applications in robots, smart sensor networks, future real-time pressure mapping systems, and human-machine interfaces through wireless optical communication.

1. Introduction

The advent of 5G information age and the Fourth Industrial Revolution has led to the emergence of numerous intelligent terminals in advanced industries and multidisciplinary domains, including big data, cloud computing, the Internet of Things (IoT), and artificial intelligence (AI) such as smart portable/wearable equipment \([1-3]\), human-machine interactions \([4,5]\), and artificial electronic skins \([6,7]\). A flexible pressure sensor (FPS) that owns excellent sensing performance and low power consumption as a crucial component becomes particularly urgent to satisfy the needs of practical applications, emerging as a hot area \([8-10]\). In particular, a self-powered visualized flexible pressure sensor (SP-VFPS) which can wirelessly display local pressure information and simultaneously convert it into human-readable visual optical signals has aroused a new surge of interest due to its good solution to current overcrowding radio frequency-based communication. Until now, the reported SP-VFPS has mainly been based on mechanochromic (MC) or mechanoluminescence (ML) materials. Nevertheless, some significant deficiencies hinder its practical applications: (i) Both MC and ML materials require substantial pressure in the order of MPa to change luminescence and/or intrinsic colors with a high limit of detection; (ii) Most MC materials are poor in recoverability and call for harsh conditions for recovery, like long-time fumigation or heating to return to its initial state \([11,12]\); (iii) Additional data processing and display systems are still indispensable, which results in extra power consumption and structural complexity. For instance, Man et al. \([12]\) reported a sensitive
and recoverable SP-VFPS based on the J-type aggregation of MC materials capable of switching between green and orange emissions with a detection limit of 0.5 MPa at 120 °C. An SP-VFPS was introduced by Wang et al. [13] based on the ML of ZnS:Mn particles to instantaneously visualize the pressure of single-point and 2D planar distributions, whose function however was limited to a restricted range of over 0.6 MPa. Therefore, it is meaningful to develop a new sensing system based on new mechanisms to address the above-mentioned challenging issues of SP-VFPSs.

Recently, triboelectrification-induced electroluminescence (TIEL) as a new class of luminescence in which extremely gentle mechanical interactions can be converted to luminescence has drawn significant attention for its practical significance in self-powered illumination, human-machine interactive display, anti-counterfeiting, and real-time vision sensors [14–19]. It was found that TIEL is essentially different from traditional ML resulting from piezoelectric effect [20,21]. In particular, TIEL enhances pressure sensitivity in the low-pressure region by several hundred times compared with ML. As a result, SP-VFPSs based on TIEL for visualized sensing possess prominent advantages over ML. Wei et al. [14] were the first to report such novel TIEL which achieved high spatial resolution motion tracking and whose sensitivity in a low-stress region (< 20 kPa) presented a 750-fold enhancement over ML. Wang et al. [16] reported an enhanced high-resolution TIEL through the introduction of a conductive layer based on transparent silver nanowires (Ag NWs) for self-powered visual interactive sensing and successfully revealed surface texture on a nitrile glove. However, these previous works mainly focused on the variation in the luminescence intensity of TIEL. Therefore, it is desirable to realize color-tunable TIEL in response to applied pressure by constructing an all-in-one SP-VFPS where both eye-detectable spatial pressure mapping and the unique ability to rapidly change color with exteroception surroundings can be achieved without any complex signal processing instrument, and whose adaptability and portability can be further promoted.

Herein, a new class of SP-VFPS enabling color-tunable TIEL in response to mechanical pressure is reported for the first time, comprising two parts, namely a porous red photoluminescence (PL) architecture and a TIEL component composed of luminescent and electrification layers. PL phosphors are capable of absorbing green TIEL from ZnS:Cu and then transforming it into red PL through a color-conversion process [22,23]. The compression of the porous structure under pressure would lead to a significant increase in the intensity ratio of green TIEL to red PL to a large extent, which was significantly associated with a wide color-changing range. This SP-VFPS showed a relative detection limit of as low as 10 kPa, a response time of less than 10 ms, and the high sensitivity (S > 190 kPa −1) within a wide pressure range from 10 kPa to 2.4 MPa, which covered the whole pressure range of daily activities. Through integrating with a TENG matrix and combining the advantages of both components, the hybrid device enabled multi-functional pressure sensitivity for motion monitoring, real-time multi-touch sensing, and eye-detectable spatial pressure mapping. It is expected that this new high-performance SP-VFPS would contribute to the development of future large-area sensing systems for the self-powered wireless optical transmission of mechanical signals.

2. Results and discussion

Surface structures and morphologies like cracks and folds were evolved to lead to the not only constant-changing but also revocable control of modification to transparency [24], fluorescence [25], and coloration [26]. These studies were inspired by the display strategy of animals like chameleons and cephalopods making use of the superficial structure controlled by muscle to quickly change colors with outstanding reversibility for communication and camouflage. Such a change was subject to the control of the moving chromatophore where the sac involved naturally-derived coloring materials with radial muscle on their periphery. In addition, structural reflectors such as iridophores, leucophores and the likes under the chromatophore were reflective of light for improved visibility, as shown in Fig. 1a. However, these studies mainly focused on stretch-stimulated sensing rather than lateral tactile sensing. Therefore, SP-VFPS was fabricated analogously by a multilayered composite material, composed of two parts, namely a compression-dependent porous red PL architecture with red PL phosphor powders (Sr, Ca) SiAlN3:Eu2+ (denoted as R-PL in this case) in the polydymihaloxilone (PDMS) matrix and a TIEL component consisting of the PDMS matrix with green TIEL phosphor powders ZnS:Cu (denoted as G-TIEL in this case) and an electrification layer (fluorinated ethylene propylene, FEP) generating tribo-charges with an external object, as schematized in Fig. 1b. Among them, PDMS matrix was utilized by virtue of its unique flexibility, mechanical robustness, easy fabrication as well as low cost. The top TIEL component was the source of green TIEL light, while the bottom porous red PL layer acted as a color-conversion layer. Collected on the side of the porous layer, the optical emission signal was the combination of emitted red PL and leaked green TIEL. Moreover, the left part of Fig. 1b shows that the surface modification of FEP can be made to create vertically aligned polymer nanowires with an average diameter 200 nm and a mean length of 1.5 μm, which was beneficial to improving the intensity of the electric field and achieving high-sensitivity low-pressure detection [27]. The FEP film had a thickness of around 50 μm. A piece of the as-prepared SP-VFPS in a hand is exhibited in Fig. S1 (Supporting Information). Fig. 1c schematically displays the detailed fabrication process without an electrification layer, while the Experimental Section illustrated the specific experimental procedure based on the method of sacrificial template [28]. Corresponding scanning electron microscopy (SEM) images and their magnified views display the surface, cross-sectional and bottom-sectional morphologies of the double-layered material, as shown in Fig. 1d. It can be found that R-PL and G-TIEL phosphors had an average diameter of around 20 μm and 50 μm respectively, indicating their uniform dispersion in PDMS. The distinct layers of the red porous PL layer (400 μm) and green TIEL layer (500 μm) had a total thickness of around 900 μm. Moreover, the red porous PL layer contains densely distributed pores with an average diameter ranging from 200 μm to 400 μm. The energy dispersive spectrometer (EDS) measurement of elemental analysis and the X-ray diffraction (XRD) patterns of both independent phosphors are shown in Fig. 1e and f respectively. The results confirmed that the two phosphors were corresponding to wurtzite ZnS:Cu (JCPDS card NO. 1-792) and orthorhombic (Sr, Ca)SiAlN3:Eu2+ (JCPDS card NO. 39-0747). Moreover, Fig. 1g shows the absorption band of R-PL and its correlation with the emission of G-TIEL. Energy transfer was believed to occur in the case of the overlapping between the emission (400–600 nm) and absorption (350–650 nm) bands of donor and acceptor respectively [29], indicating that green TIEL in the top luminescent layer can be effectively transmitted to the bottom porous layer and converted into red PL through a color-conversion process (Fig. S2, Supporting Information). Derived from conventional thin-film and phosphors materials, the SP-VFPS was extremely low-cost and highly practical, whose structural design and fabrication process showed compatibility with potential large-scale production.

An experimental platform was constructed to collect light emission from the SP-VFPS, as shown in Fig. S3 (Supporting Information). The top electrification layer was subject to rubbing by a pen-shaped contacted object driven by a linear stepper. Placed under the bottom substrate, an optic-fiber probe and a force sensor were used to collect and direct light emission from the side of the red porous PL layer to a spectrometer and monitor vertical pressure on the contact interface simultaneously. For the purpose of illustration, different working principles of the SP-VFPS based on TIEL and ML are diagrammed in Fig. 2a. TIEL relied on the drastic change of external triboelectric potential from the dynamic interactions between two dissimilar materials able to excite the EL of underlying phosphors along the motion locus. It is worth noting that a porous structure-based SP-VFPS would not only induce charges on the contact surface, but also generate additional charges on the porous
However, ML resulted from the strain-induced piezoelectric internal potential in piezoelectric materials because of strong electron-lattice coupling. As previously reported [13], an extra grounded layer made from indium tin oxide (ITO) can shield the external electric field, exclude TIEL and then separate the two luminescence phenomena of TIEL and ML. As one of the key components of SP-VFPSs, the top TIEL component was discussed from the aspect of two parameters. Firstly, the concentration of ZnS:Cu and the thickness of the luminescent layer were determined according to the previous work for a high TIEL intensity [16]. Secondly, scanning Kelvin probe microscopy (SKPM) was used for characterizing the surface charge density of the electification layer rubbed with different polymers, as shown in Fig. 2b. As a most triboelectric negative material, FEP was inclined to produce negative charges when contacting almost any other material. In terms of “triboelectric series”, more positive materials of the external object would increase tribocharge density, resulting in the enhancement of TIEL intensity. Thus, thermoplastic polyurethane (TPU) was selected as the external object in this case. The luminescence intensity of SP-VFPSs without R-PL phosphors in the porous layer was further investigated based on TIEL and ML respectively in order to exclude the conversion between R-PL and G-TIEL. Fig. 2c and d show that ML had a corresponding pressure threshold value of 0.2 MPa, but TIEL can be observed to be as low as 10 kPa, powerfully proving the highly efficient sensitivity of TIEL in a low-pressure region. Moreover, the weight percentage concentration of R-PL phosphor in the porous layer was determined to be 35% from the aspect of the red porous PL layer so as to achieve high color conversion efficiency and ensure elasticity based on PDMS (Fig. S4, Supporting Information). In addition, the thicknesses and porosity of the porous red PL layer were studied as factors influencing the changes in the Commission Internationale de l’Eclairage (CIE) coordinates in the color space diagram under the applied pressure ranging from 0 to 2.4 MPa with an increment of 0.4 MPa. The CIE coordinate changes of the SP-VFPS with four thicknesses (300, 400, 500 and 600 µm) of the porous layer are exhibited in Fig. 2e, whereas red and green colors from pure R-PL and G-TIEL phosphors were described as (0.71, 0.32) and (0.21, 0.51) respectively. The results showed that the SP-VFPS underwent a distinct color transition between pure red PL and green TIEL with the increment
of pressure. When an increase occurred in the thickness of the porous layer, stronger red PL was obtained as a result of more excited materials. The initial color was very close to pure red PL, but an excessively thick porous layer led to the less leakage of green TIEL and the deviation of the final color away from pure green TIEL, resulting in a small range of color transition. The maximal color transition range reached an optimal value when the thickness of the phosphor layer was 400 µm. Besides, a group of SP-VFPS samples with different porosity values (0%, 11%, 21% and 32%) were subject to measurements. The surface-sectional morphologies of the porous layer and CIE coordinate changes are shown in Fig. 2 f and g respectively. The concentration of NaCl powders was adjusted to regulate the level of porosity. Porosity (θ) was obtained with the following equation [28].

$$\theta = (1 - \frac{M}{V}) \times 100\%$$

where, $M$ and $V$ represent the mass and volume of a porous red PL layer respectively, and $\rho$ refers to the density of a nonporous control sample of the same number. With the increase of porosity, the final color would be close to pure green TIEL despite more leakage. However, the initial color deviated away from the pure red PL due to the shortage of red PL materials, leading to a smaller color transition range. Thus, the SP-VFPS with 21% porosity presented the largest color transition range.

Finally, the corresponding emission spectra of the SP-VFPS (21%, 400 µm in the porous layer) under different values of applied pressure are shown in Fig. 2 h. A spectral variation was observed between the pure red PL peak (excited by a light of 510 nm) and green TIEL peak centered at 650 nm and 510 nm respectively with the increase of pressure.

To quantitatively characterize the pressure sensitivity of the SP-VFPS and explore its working mechanism, three SP-VFPSs with different structures were fabricated for the purpose of comparing and
investigating the relationship between color transition properties and applied pressure in detail. Firstly, Fig. S5 (Supporting information) shows that a Gaussian fitting curve was used to separate measured relative emission spectra so as to get red and green color parameters and similar processing for other conditions. The results were analyzed, as illustrated in Fig. 3a–c respectively, with corresponding device structures schematized in the inset and denoted as devices 1 (nonporous-TIEL), 2 (porous-ML) and 3 (porous-TIEL). Device 2 with a grounded ITO layer was designed to exclude TIEL, while device 3 was the SP-VFPS developed in this case. Notably, relative slopes showed significant difference although red and green lights were both promoted with the increase of applied pressure. Secondly, the relative G/R intensity ratios \( \Delta C/C_0 \), where \( C \) refers to G/R color intensity ratio) of the three devices under applied pressure and the magnified view in the low-pressure region are analyzed in Fig. 3d and e respectively. Compared with devices 1 and 2, device 3 had a rather low detection limit of tribo-pressure at \( \sim 10 \) kPa and gave rise to a more vivid color change (Fig. 3d). Two critical pressure values of 0.2 MPa and 1.6 MPa divided the detection area into three distinct regions, as shown in Fig. 3e. At last, Fig. 3f shows that the sensitivity (S) of devices was calculated as \( S = (\Delta C/C_0)/\Delta \varepsilon \) where \( \Delta \varepsilon \) stands for applied pressure, with a wide pressure range from 0 to 2.4 MPa covering the pressure range of daily activities (inset of Fig. 3f).

A three-section behavior of the color transition sensitivity of the porous-TIEL device (the top curve) can be analyzed based on contact pressure. In the low-pressure region of 0.01–0.2 MPa (Section I), the corresponding sensitivity of 0.267 MPa\(^{-1}\) presented a rapid increase because of the nanoscale contact area due to the nanostructure on the electrification layer. In section II (0.2–0.16 MPa), the elastic deformation of the luminescence layer would slow down the rate of color transition (0.19 MPa\(^{-1}\)) with the increase of pressure to more than 0.2 MPa, which was in line with previous research on the threshold value of materials based on ZnS for ML. In section III (1.6–2.4 MPa), however, the high pressure loaded on the porous layer became the main factor, significantly thinning the dielectric layer and resulting in the sharp enhancement of color transition sensitivity (1.06 MPa\(^{-1}\)). Moreover, it can be further found that the porous-TIEL (the top curve) exhibited much higher sensitivity than porous-ML (the middle curve) and nonporous-TIEL (the bottom curve), corresponding to different working mechanisms in low- and high-pressure regions, suggesting that the surface modification on the electrification layer and the decoration of the porous structure both had a great impact on the high sensing performance of the SP-VFPS. Our primary goal is to achieve higher sensing performance under lower detection thresholds through device remolding and upgrading in the future.

According to the results discussed above, the working principle on the high sensing performance of the SP-VFPS was discussed in detail based on the decoration of the porous structure since the effect of surface modification on the electrification layer in low-pressure detection was explored in a great amount of previous research [27,31]. Firstly, the altered transmittance of the red porous layer under compression was the major factor that can be quantitatively calculated by means of the simulation software COMSOL. Fig. 4a and b present the compressibility of the solid and porous red layer with different 3D shapes respectively. The detailed simulation process is shown in Fig. S6 (Supporting Information). The results clearly showed that the porous structure resulted in more deformation under the same pressure than the solid one. When a compressive pressure was loaded, the porous structure with air space in the whole material experienced more deformation in the pore region due to the decrease of effective Young’s modulus unlike solid structures in which deformation was homogeneously distributed throughout the matrix, as shown in Fig. 4c. This would lead to the considerable decrement of material thickness, which can be determined by the average deformation difference between upper and bottom surfaces, as illustrated in Fig. 4d and e. The results in Fig. 4f showed that the porous structure exhibited a considerable thinner equivalent thickness under increasing pressure compared with the solid one. Then, the transmittance of the porous layer related to applied pressure can be calculated based on the measured transmittance (21%, ultraviolet visible (UV–Vis) measurement) and thickness (400 \( \mu \m \)) of the control solid sample at 0 MPa. It can be seen from Fig. 4g that the transmittance of the porous structure increased from 37.0% to 60.7% with the increase of applied pressure compared with that of the solid structure increasing from 21% to 37.6% in the absence of inner air space, which would lead to a significant increase in the intensity ratio of green TIEL to red PL and

Fig. 3. Pressure sensitivity of the SP-VFPS. (a)–(c) Relative emission intensities of red and green color parameters. Inset: Structure of the corresponding device. (d) Relative G/R intensity ratio in the enlarged view of selected areas shown in (e). (e) Relative G/R intensity ratio of the three devices with the increase of pressure (\( \Delta C/C_0 \), where \( C \) refers to G/R color intensity ratio). (f) Pressure sensitivity of the three devices. Inset: Diagram of pressure regions and related applications in daily life.
result in high sensitivity in the whole pressure region. In this regard, this simulation result provided convincing evidence that the compression-depended porous structure can modify the color transition of the SP-VFPS. Secondly, the porous structure can dramatically improve the intensity of the triboelectric field \([29,32]\). On one hand, the porous structure can effectively expand the contact area under compression and contribute to the generation of more charges on the contact surface. On the other hand, compressed pores plus relevant electrostatic effects gave rise to the generation of additional charges on the porous surface, causing a larger triboelectric potential. Zheng et al. \([32]\) reported that a porous aerogel film-based TENG contributed to the enhancement of more than 11-fold power density compared with a dense polymer film-based TENG. Therefore, the largely triboelectric charge density induced by the porous structure was also of importance for the sensing mechanism of the SP-VFPS. Moreover, the response time of the SP-VFPS (1.6 MPa, monitored at 510 nm) can be predicted for consecutive measurement shown in Fig. 4h, with a rapid response time of 10 ms. In fact, the actual response time was much shorter, given that the spectrometer had a time resolution of 8 ms. Thus, this SP-VFPS had a huge advantage in the processing of the optical signals emitted with this rapid response time as the basis. Furthermore, a cyclic experiment (velocity: \(6 \text{ cm s}^{-1}\); pressure: 1.6 MPa) was performed by reciprocating sliding against the surface of the electrification layer under the condition of normal atmosphere (Fig. S7, Supporting Information). After 20,000 cycles, the luminescence intensities of both red and green peaks basically remained the same (Fig. 4i), suggesting that the color-changing ability of the SP-VFPS was excellently stable and repeatable. Bending stability is presented in Fig. S8, demonstrating that the variation of luminescence intensities was below 3% with the increase of bending curvature from 0 to 100 m\(^{-1}\). The results clearly indicated that the SP-VFPS in this study exhibited robustness in mechanical and luminescence performance after long-time operations, contributing to its reliability in practical applications. It is worth mentioning that the SP-VFPS in this case demonstrated high sensing performance due to the unique...
merits discussed above, exhibiting superiority to the mechanochromic SP-VFPSs in previous reports [11,33–35].

The potential application of this SP-VFPS was demonstrated in terms of eye-detectable spatial pressure mapping capability as a proof of concept. The SP-VFPS in this case can be easily integrated with transparent and flexible Ag NWs/PDMS electrodes to be a single-electrode TENG matrix (4 × 4 array) on the basis of the coupling effect electrostatic induction and contact electrification, thereby generating a triboelectric voltage between the electrification layer and external object in response to applied pressure, as shown in Fig. 5a. The Experimental Section illustrates the detailed fabrication procedure. As a crucial premise of practical application on the human-machine interface, both static and dynamic pressure was detected under dual modes based on multi-touch sensing and motion monitoring through the electric response from the TENG component. For instance, the real-time motion monitoring of personal writing was identified by measuring the output voltage signal of every single pixel generated. As shown in Fig. 5b, the writing content of letter “Z” can be recognized based on the order of signal generation for different pixels (A → B → C → D as the path displayed in the inset of Fig. 5b). Moreover, Fig. 5c schematically illustrates two-point and Z-shaped contact. Fig. 5d displays generated relative voltage signals according to these two-touch modes, and pressure location and magnitude can be effectively detected in the case of the contract between objects. At present, a variety of pressure sensors based on TENG have been proposed [36,37]. Inconsistent with literature, this hybrid sensing system made it possible to instantly visualize and track

![Fig. 5. Display of the SP-VFPS in the application of visualized sensing. (a) Schematized SP-VFPS integrated with a TENG matrix (4 × 4 array electrodes) for multi-touch and motion monitoring by testing electric signals. (b) Voltage signals measured for the motion monitoring of writing letter “Z” by multichannel measurement (The path: A → B → C → D) and corresponding 3D pressure distribution. (c) Schematic diagram of multi-point contact. (d) Voltage signals measured for two-point and multi-point contact with the letter-shaped objects of “Z”. (e) Collected optical photographs of spatial pressure distribution after the contact between a variety of objects (i: tip, ii: stick and iii: tube). (f) Collected optical photographs of spatial pressure distribution for tracking the motion of different external pressures (i: slight, ii: heavy and iii: sharp). Bottom section: Corresponding enlarged areas. (g) A continuous trajectory for the word “tribo”. Top section: Live color transition optical photographs. Bottom section: Corresponding luminescence intensity mapping using the software MATLAB.](image-url)
the location of applied external pressure. Fig. 5e shows the optical photographs collected after the contact of various objects of different shapes (i: tip, ii: stick and iii: tube) (Movie S1, Supporting Information). Local color distribution patterns matched the shape and wall thickness of objects, exhibiting a minimum line of 500 µm for the contact of a tip. Furthermore, letter “Z” shown in Fig. 5f was completed by a pen-shaped object (a diameter of 2 mm) under significantly distinct pressure (i: slight, ii: heavy and iii: sharp) (Movie S2, Supporting Information). It is obvious that color changes can realize the real-time detection of the motion trajectory. What is worth mentioning is that the change of increasing pressure resulted in the increasing density and intensity of green granules, as shown in the large view part of Fig. 5e, thus leading to a macroscopic view of color transition. The results confirmed that microsponges served as a “small window” to adjust the exposure area and concomitant luminescence color from SP-VFPS, which can shield green dielectric light by absorbing red PL powders in the initial state. Moreover, it is apparent that a significant variation occurred in green granules along the motion trajectory as a result of applied pressure and in velocity during the course of writing. For instance, luminescence colors located at “1” and “2” demonstrated a significant difference. Thus, this color distribution characteristic can effectively reflect the pattern of individual handwriting, contributing to its applications including anti-counterfeiting and electronic commerce. Fig. 5g shows the color transition of successive writing letters “tribo” and its corresponding intensity mapping (processed by the software MATLAB), which further confirmed its ability to obtain detailed pressure information. Unlike intensity mapping, the SP-VFPS in the present study can realize eye-detectable spatial pressure distribution without any display instrument or complicated additional data processing. Hence, the hybrid sensing system can be performed by electrical readout and optical visualization, and play a replaceable role according to different requirements. Through the SP-VFPS, the location, shape, and distribution of pressure can be provided in eye-detectable and wireless ways, and the accurate electrical signal magnitude can be achieved by the TENG part. Multi-functional pressure sensitivity was shown in the layout and fabrication of a simplified device, thus leading to an immediate application in a variety of areas, including robots, smart sensor networks, human-machine interfaces, and real-time pressure mapping systems. Moreover, the SP-VFPS is suitable for colorful display, image recognition as well as security surveillance in addition to the example of eye-detectable pressure distribution demonstrated above.

Supplementary material related to this article can be found online at doi:10.1016/j.nanoen.2020.105431.

3. Conclusion

In summary, a novel SP-VFPS with the color-changing ability of pressure-sensitive visualized sensing was developed, realizing color-tunable TIEL in an all-in-one device for instantaneously eye-detecting the pressure information from surroundings without any additional data processing and display instrument, and therefore representing a breakthrough concept of pressure sensing/imaging based on TIEL. Due to the new-type sensing mechanism and distinct physical and chemical modifications, the SP-VFPS exhibited a rather low detection limit of tribo-pressure at ~ 10 kPa, an unprecedentedly high sensitivity (S > 190 kPa⁻¹) within a wide pressure range and simultaneously a fast response time of below 10 ms. A complete sensing system was built up by integrating the SP-VFPS with a TENG matrix, whose various applications such as multi-touch, motion monitoring, and eye-detectable spatial pressure mapping were demonstrated by combining electrical and optical responses. Because of featuring other main advantages such as scalability, durability, low cost and easy implementation, the SP-VFPS exhibits bright prospects in robots, smart sensor networks, and human-machine interfaces.

4. Experimental section

4.1. Fabrication process of an SP-VFPS

ZnSn:Cu (G-TIEL) and (Sr, Ca) SiAlN₃: Eu²⁺ (R-PL) fluorescent powders were purchased from Shanghai Keyan Phosphor Technology Co., Ltd. and Hebei Ledphor Optoelectronics Technology Co., Ltd. respectively. A hollow PET frame with a central size of 50 × 50 × 0.4 mm³ was pasted along the edges on an acrylic substrate with a size of 55 × 55 × 3 mm². A certain amount of R-PL phosphors and NaCl particles (used as the sacrificial template) were dispersed uniformly in the PDMS (Sylgard 184 silicon elastomer, Dow Corning) with a curing agent (Ratio of curing agent to base was 1:10), then scraped on the PET frame and cured at 80 °C for 1 h to generate a porous red PL layer. ZnSn:Cu phosphors were uniformly mixed in PDMS and spin-coated to deposit on the cured porous red PL layer, then cured at 80 °C for 60 min and then peeled off. The as-fabricated composite film was immersed in deionized water, heated at 90 °C for 3 days and replaced every 10 h to ensure the removal of all NaCl particles. FEP with the physical modification of vertically aligned polymer nanowires by plasma dry etching was directly attached to the surface of the luminescence layer, thereby forming an all-in-one device [27,31].

4.2. Fabrication process of an SP-VFPS/Teng hybrid matrix

Obtained from Nanjing XFNANO Materials Tech Co., Ltd. Ag NWs were dispersed in ethanol and underwent 10 min sonication so as to minimize the agglomeration of nanowires. The solution obtained was dropped onto a pre-treated glass substrate (a size of 55 × 55 × 5 mm³) at room temperature. After fully dried, Ag NWs were coated by PDMS with a curing agent, and then air bubbles were removed. Subsequent curing took place at 80 °C for 60 min. After peeled off from the glass substrate, the Ag NWs/PDMS film (a thickness of ~ 0.1 mm) was fabricated into an array (10 × 10 mm²) by laser-cutting techniques. The SP-VFPS/TENG hybrid was acquired through the encapsulation of exposed Ag NWs/PDMS electrodes between the TIEL layer and the red porous layer to produce a sandwich structure in the process of fabricating the SP-VFPS. Silver paste was applied to the edges of the Ag NWs electrode to lead to the external electrical circuit.

4.3. Characterisation

Crystal structure was characterized by XRD (X’pert 3, Powder). A field emission SEM (450 FEI, Nova Nano) was adopted to characterize all EDS and SEM images. Absorption characteristics were analyzed by UV–Vis–NIR light source (UV-3600, Shimadzu). Periodic mechanical traction was performed by a linear motor (E1100, Lin Mot). A pressure sensor (Nano17, ATI) was utilized to evaluate the contact pressure. A digital temperature-humidity atmospheric pressure gauge (622, Testo) was applied to measure both temperature and humidity. Sheet resistances of Ag NWs/PDMS conductive layers were detected by a standard four-probe method (RTS-9, ProbesTech). A spectrometer with vertically arrayed optical fibers and collimating lenses (NOVA, Ideaoptics) was used to measure the optical performance of the device. A digital multimeter (PXIe 4300, National Instruments) was used for the electrical measurement of the array device. SKPM measurements were conducted with the MFP-3D atomic force microscope (AFM) (NX-10, Park Systems) at room temperature.

CRediT authorship contribution statement

Li Su: Conceptualization, Methodology, Investigation, Data curation, Writing - original draft, Supervision, Writing - reviewing & editing. Zhiye Jiang: Data curation, Visualization, Software. Zhen Tian: Visualization, Software. Hailu Wang: Data curation, Validation. Haojie Wang: Software, Validation. Yunlong Zi: Conceptualization,
Methodology, Supervision, Writing - reviewing & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Competing financial interests

The authors declare no competing interests.

Appendix A. Supporting Information

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References

Zhiye Jiang is currently pursuing her master degree in Hebei University. She received her B.Eng. degree in Photoelectric Information Science and Engineering from Industrial and Commercial College in Hebei University in 2019. Her research interests are mainly focused on triboelectrification-induced electroluminescence and sensor manufacturing.

Zhen Tian is currently pursuing his master degree in Hebei University. He received his B.Eng. degree in New Energy Materials and Devices from Hebei University in 2019. His research interests are mainly focused on stretchable nanocomposite materials and wearable devices for energy harvesting.

Hailu Wang received her B. Eng. degree in Chemistry from Shandong University. Now she is pursuing her Ph.D. degree in Beijing Institute of Nanoenergy and Nanosystem, Chinese Academy of Sciences. Her current research mainly focuses on energy harvesting and sensing device based on electret generators.

Haojie Wang is pursuing her master degree in Hebei University. Her research interests are mainly focused on the energy conversion, storage and some novel applications.

Dr. Yunlong Zi received his B.Eng. degree from Tsinghua University in 2009. He received his Ph.D. degree from Purdue University in 2014. After his graduate study, he worked as a Postdoctoral Fellow at Georgia Institute of Technology during 2014–2017. Dr. Zi joined the Chinese University of Hong Kong as an Assistant Professor in November, 2017, as the founder of the NanoEnergy and Smart System laboratory.