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Recent progress of triboelectrification-induced electroluminescence: from fundamentals to applications

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Abstract

Luminescence is the premise for humans and even all creatures in the world to survive, as well as a crucial means for humans to interact with the environment. Recently, triboelectrification-induced electroluminescence (TIEL) has been applied in anti-counterfeiting, real-time vision sensor, human/machine interactive system, as well as self-powered illumination and display, thus attracting significant attention. It can convert kinetic energy into light through the coupling effects of triboelectrification and electroluminescence. In this review, we focus on the latest advancements of TIEL, including its fundamentals, novel applications, and outlook. It is predicted that TIEL will be widely applied in daily activities and industrial productions, so as to achieve self-powered smart systems.

1. Introduction

With the rapid development of science and technology in the era of the Internet of Things, the demand for smart materials/devices is growing rapidly. Among them, mechanical stimulus-response smart materials/devices have important scientific significance and practical value. They are capable of detecting the morphology, position, trajectory, and other motion parameters of dynamic objects, appropriately process them, and then perform its own executive function, which makes it widely applicable in such fields as mechanical sensing, positioning and tracing, infrastructure safety, security monitoring, biomedicine and so on \cite{1–5}. Until now, the state-of-art detection method is mainly based on the electrical signals released from the external mechanical motion, for example, the capacitive touch screen of personal electronic devices \cite{6–10}. It is favorable to display visualized signals in real time through light luminescence, without needing complex electronic signal processing circuits.

As an existing technology, the mechanoluminescence (ML) material can convert mechanical energy into optical radiation \cite{11–15}. According to the degree of destruction suffered by the material, the ML can be categorized into fracto-ML, plasto-ML and elastico-ML \cite{16–18}. Among them, fracto-ML is a destructive luminescent phenomenon, relying on the asymmetrically charged surfaces when the material fractures. This phenomenon would result in the irreversible destruction of materials, as well as the limitations on both repeatability and stability. In comparison, plasto-ML and elastico-ML depend on the plastic and elastic deformation of materials. Although the material does not fracture during operation, there still exist several potential problems as follows. Firstly, the luminescent intensity usually decreases significantly with the increase of the external force, so that the auxiliary irradiation with additional ultraviolet light is needed to compensate for the initial intensity \cite{19, 20}. Secondly, there is a certain strain threshold for the plasto-ML and elastico-ML, which is difficult to approach due to the large elasticity modulus of inorganic solid ML materials \cite{21, 22}.
Additionally, in order to adapt the developing trends of miniaturization, functionalization, and intelligence of optoelectronic devices, researchers continue to explore the integration and coupling effects of materials for obtaining renewable functional materials/devices with special properties [23, 24]. Considering that the triboelectric field can be used to control the transport behavior of carriers, triboelectrification-induced electroluminescence (TIEL) has attracted significant attention since its discovery [25]. As a new type of motion-driven luminescence, TIEL process is not limited to the asymmetric structure of the crystal and must generate dislocations and strain fields in the material through deformation. This requires no additional light source or electrical energy supplement, thus making up the shortcomings of existing ML materials. It features low threshold of stress, high response efficiency, and nondestructive operation, so that it holds huge potential of applications as light sources and self-powered sensors under weak mechanical stimulus conditions. Existing research has demonstrated its potential of applications in such fields as anti-counterfeiting, smart sensing, human–machine interface, electronic skin, lighting and display, and wearable devices, as illustrated in figure 1. This review focuses on the basic principles, operations, applications, and perspectives of TIEL, which reveals its great potentials in various applications.

2. Fundamentals of TIEL

2.1. The mechanism and operation modes of TIEL
Triboelectrification is a charge transfer phenomenon occurring on the material surfaces when two different materials come into contact or slide against each other, thus converting mechanical energy into electrical energy [35, 36]. Meanwhile, the transferred surface static charge can generate strong local electric field inside the material to accelerate the high-energy electrons impact ionization, thus exciting the luminescence center and generate electroluminescence (EL). Therefore, the mechanism of TIEL is mainly the coupling effect between triboelectrification and EL [25, 34], as shown in figure 2(a). However, its production may also involve a highly complex process, such as temperature variation, electric breakdown, piezoelectric effect, and ML [37, 38]. These mechanisms remain in the hypothetical stage which requires further studies. Consequently, there is still no unified theoretical system at the present time for understanding the TIEL mechanism. Given the opinions put forward from various perspectives, one important direction of future fundamental research on TIEL is to identify the functions of these factors and mechanisms in the TIEL process.

The reported TIEL materials mainly focus on trap-type inorganic semiconductor electroluminescent materials doped with lanthanide luminescent elements and transition metal ions, such as ZnGa2O4:Mn2+, ZnAl2O4:Mn2+, ZnS:Cu/Mn, ZnS:Cu, Al [25, 39–41]. Given the dependency of TIEL on the alternating electric field generated during the triboelectrification process, we believe that the semiconductor materials with high-field EL characteristic can be used for TIEL in general. In particular, such merits as long service
Figure 2. The mechanism and operation modes of TIEL. (a) The mechanism of the TIEL based on triboelectric field and band diagrams of the ZnS:Cu phosphors. (b) Two-dimensional structural diagram of charge distribution along the luminescent layer of sliding-contact mode. [25] John Wiley & Sons. © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) Schematic diagram of the charge distribution between the materials in the contact state (internal TIEL) and for the separation state (external TIEL) of contact-separation mode, respectively. Reprinted with permission from [34]. Copyright (2019) American Chemical Society. (d) Illustration of the rationale and charge distribution of non-contact mode [29]. Reproduced from [29] with permission of The Royal Society of Chemistry.

life, high luminescence efficiency and different luminescence colors of red, orange, green or blue can be achieved by various transition metal ions-doped zinc sulfide (ZnS) materials [42, 43]. Thus, to develop TIEL devices, plenty of studies have been conducted on the polydimethylsiloxane (PDMS) and ecoflex-supported ZnS composite, which features low cost, good elasticity, high mechanical robustness, and facile fabrication. It is noteworthy that ordinary semiconductor refers to the luminescent material of TIEL, and the local electric field cannot be easily sustained. For this reason, the rapidly changing electric voltage can continuously generate new electric field, which is conducive to the formation of TIEL [44–46]. According to the triggering method, these TIEL devices can be classified to three types, namely, contact-sliding mode, contact-separation mode, and non-contact mode.

2.1.1. Contact-sliding mode
Tribocharges result from the gentle mechanical dynamic interaction between two dissimilar materials, which can abruptly and significantly generate the electric potential up to hundreds of volts within milliseconds. The drastically increased electric potential can be utilized to excite the EL of underlying phosphor along the trajectory of motion, as shown in figure 2(b). Within the perimeter of the sliding object, the electric potential reaches its maximum at the center, whereas it drastically drops outside of the perimeter. As the charged object slides over, the electric potential of the luminescent layer along the trajectory sharply increases and then immediately drops. As a result, the luminescence of phosphor is excited by this dynamic variation [25]. Likewise, TIEL can be driven by a single-electrode triboelectric nanogenerator (TENG) during the contact-sliding process, in which a varying electric field can be generated between the top surface of the dielectric layer and the grounded electrode. As a result, the luminescent center of the phosphor is excited in the dielectric layer [41].

2.1.2. Contact-separation mode
The contact-separation mode is clearly different from the sliding mode. As the external objects come to contact with and get separated from with each other, there would be two-luminescence phenomenon arising, as shown in figure 2(c). The first luminescence refers to the internal TIEL as the external objects come into contact, in which the pore interfaces are formed due to the elastic matrix. The second one refers to the external TIEL after the contact, in which the charged interface changes the electric potential of the phosphors [34]. However, compared with the contact-sliding mode, it takes less time to complete the separation after the contact. Moreover, the afterglow of ZnS EL lasts tens to hundreds of milliseconds. Thus, it is generally difficult to distinguish the two kinds of luminescence phenomena during the period. Moreover, TIEL device can be driven by a double-electrode TENG when agitated by mechanical vertical contact and separation (e.g. finger tapping). The luminescent layer of ZnS:Cu is sandwiched between two electrodes, with connection to the electrodes of TENG, respectively. Then, it is easy to observe the bright and durable green light emitted from the TIEL device with naked eyes [47].
2.1.3. Non-contact mode

According to the above modes, all of the signals can only be generated with two surfaces in contact, which may result in material failure, induce additional energy consumption and restrict the form of movements. Thus, non-contact mode is advantageous since it enables the objects to roam above the interface. In the meantime, plenty of motions can be perceived. As shown in figures 2(a) and (d) negatively charged electret is used to generate the electric field along the trajectory of a floating conductive object, so as to modulate the emission of ZnS:Cu, which is an effective non-contact means to excite TIEL [29].

2.2. Optimization of the TIEL

2.2.1. Improvement of TIEL intensity

In general, the TIEL excited by gentle mechanical action is relatively weak. Thus studies were conducted to improve the luminescence efficiency. Currently, there are two main ways to enhance TIEL intensity, which are surface treatment/modification and the introduction of the conductive layer. The enhancement of TIEL intensity by surface treatment/modification may be attributed to the improvement of triboelectric surface charges. In 2017, Wei et al [30] prepared an array of micro-sized PDMS pillars at the surface for triboelectrification, as shown in figure 3(a). When the micro-sized surface and the luminescent film come into close contact with each other through a relative sliding motion, intensive uniform luminescence appears due to the strong surface charge generation. Compared with the device with the pristine PDMS surface, luminescence intensity is significantly improved by twice. Besides, through just the surface treatment by plasma, luminescence intensity was also demonstrated to increase for the ZnS/ecoflex composite system [39].

A conductive layer embedded below the EL layer can modulate the electric field and confine it around the boundary of the contact object. In 2019, Wang et al [48] reported a self-powered high-resolution TIEL sensor based on conductive layer of transparent silver nanowires (Ag NWs), as illustrated in figure 3(b). It was found out that the high electric field in the boundary of the object increased luminescence intensity and the spatial resolution significantly. Compared with the TIEL sensor without the conductive layer, its luminescence intensity increased by 90%, with the spatial resolution reaching about 500 µm.

2.2.2. Color tuning ability

The color tuning ability is highly favorable for TIEL devices to broaden the applications in display, in which progress has been made to overcome this challenge. Instead of blue luminescence, the TIEL from blue ZnS:Cu will generate green luminescence on the whole. Since the weak friction energy is insufficient, the blue emission band with deep energy levels cannot be effectively excited, so that a good appearance is unlikely to be formed [29, 46, 47].

In 2019, Cheng et al [31] first reported tunable wide-spectrum TIEL, in which three independent micro-sized phosphors, including ZnS:Cu, Sr2MgSi2O7:Eu2+,Dy3+ and fluorescent dyes/Sr4Al14O25:Eu2+,Dy3+ were uniformly distributed in PDMS, as shown in figure 4(a). The green TIEL from ZnS:Cu can be absorbed by long afterglow phosphors before the conversion into a range of color of photoluminescence (PL) as affected by the function of photo-stimulated luminescence. For such a feasible technique, the tunable color expression under weak stimuli is successfully generated, thus enabling the display-related applications based on TIEL.

In 2020, Su et al [26] demonstrated the color-tunable TIEL in an all-in-one device for real-time pressure information detection for the first time, as illustrated in figure 4(b). The results showed that microspores served as a ‘small window’ to adjust the exposure area and concomitant luminescence color from the device, thus shielding the green TIEL light under the slight sliding motion. Moreover, when the sliding at a certain pressure was applied, more green light was emitted along the trajectory of motion. Through this color-tuning
ability subject to different pressures during sliding, this TIEL has massive potential for application in color display, motion recognition, eye-detectable pressure distribution sensing, as well as security surveillance.

2.2.3. Improvement of the spatial resolution
It is always expected to improve the spatial resolution of dynamic motion sensing, which can be achieved by fabricating compartment structure in the TIEL. A light-tight micron-sized square compartment array structure is constructed inside the composite material, which prevents the pressure in a single compartment from exciting the luminescence in the adjacent compartment. Wei et al. [25] filled the phosphor particles into an array of fabricated cells, as shown in figure 5(a). The opaqueness of the cell walls blocks the lateral diffusion of the emitted luminescence, thus preventing crosstalk between adjacent cells. As a result, the position and the profile of the sliding object can be precisely revealed. The resolution of the segmented structure can be further improved by the phosphor particles with smaller size, as shown in figure 5(b).

3. Applications
Compared with traditional luminescent devices, TIEL devices can capture the weak mechanical energy available in the daily life as the excitation source to generate the high-intensity light with the high response rate. Moreover, there is no external power supply required for TIEL devices. It is expected to become a new generation luminescent solution that is energy-saving, environmentally friendly and sustainable for lighting, display, imaging and smart sensing. Up to now, TIEL has been demonstrated as applicable in anti-counterfeiting, smart sensing system, self-powered light source and display, as well as wearable electronics.

3.1. Anti-counterfeiting application
Recently, researchers have paid much more attention to digital feature recognition to realize the security of information processing. Electronic signature, as a kind of intelligent data containing the identity information about the signer in electronic form, can easily and conveniently provide identity verification and physical records for users through the Internet. In figure 6, Wei et al. [25] reported TIEL-based electronic signature
anti-counterfeiting system with a high spatial resolution. The luminescent layer is comprised of polymethyl methacrylate (PMMA) matrix and ZnS:Cu phosphor particles, while the electrification layer is composed of fluorinated ethylene propylene (FEP). A string of letters are visualized through the writing pen as a sliding object, with the position and trajectory accurately displayed. The intensity of luminescence significantly varies along the trajectory due to the varying force and speed applied during writing. The characteristic distribution of luminescent intensity can reflect the unique handwriting habits, thus enabling the anti-counterfeiting function.

3.2. Intelligent sensing system

Nowadays, wind and hydraulic power is regarded as sustainable and environmentally friendly energy sources, thus attracting attention from more and more researchers. Therefore, TIEL-based wind speed sensor can be widely applied in weather forecast and other fields of environmental monitoring. A wind speed sensor was developed by Su et al [27] in 2019, as shown in figure 7(a). A perovskite-based photodetector (PD) is combined with a wind-driven TIEL component. This hybrid sensor features an enlarged electric current output. The power is generated via four typical states in a working cycle. In the original position I, when the two thermoplastic polyurethane films and the ZnS:Cu/PDMS composite film are made in full contact with each other, the opposite charges are generated on their surfaces, before reaching a new electrostatic balance. When the wind causes flutters and vibrations, the ZnS:Cu/PDMS film tends to be bent. It results in the dynamic changes into the electric potential, which reaches the peak at the maximum displacement position (states II and IV) and then drops to the minimum position rapidly (states I and III) at the equilibrium point. Thus, electric fields are produced on ZnS:Cu/PDMS film surface, which excites the TIEL. Compared with the conventional sensor based on TENG, this sensor has a low detection limit of 5 m s\(^{-1}\), and the sensitivity is three times higher than that of conventional sensors at the low bias voltage, with the extremely short response time required (<0.3 s). The research as illustrated above laid a foundation for the study and development of sensor systems based on TIEL and light sources that are environmentally friendly.

Zhao et al [28] demonstrated a novel solid–liquid interfacing triboelectrification induced electroluminescence (SL-TEL). With a flexible sandwiched configuration, the SL-TEL device consists of as few as three flexible layers, with the EL layer sandwiched between two thin FEP films. When a droplet strikes the flexible device, it scatters as a thin sheet and the impact lasts a brief period. Then, the droplet shows a
Figure 8. Human–machine interactive system. Structure of the NCIVS and image acquisition and analysis system showing the production process of the patterned barcode. Reproduced from [29] with permission of The Royal Society of Chemistry.

pancake-like shape before spreading and contracting in the form of rebound. In the meantime, there are plenty of splatters observed on the smooth surface. The triboelectrification between the droplets and the film generates surface charge as well as electric field for luminescence, so as to track the motion of droplets, as shown in figure 7(b). A visible pattern of aquatic distribution is identified when the droplet splatters, which provides an effective means to achieve the EL visualization of impact water movement.

Moreover, the self-powered visualized flexible pressure sensor (SP-VFPS) was developed to map and transform the local pressure information into human-readable visual optical signals. Such a sensor shows the potential applications in wireless human stress/strain sensing. Based on the micro compression-dependent porous architecture, Su et al [26] proposed a new class of SP-VFPS for color-tunable TIEL to respond to different vertical pressures on a real-time basis, as shown in figure 7(c). An ultra-low limit on the detection of the pressure at ∼10 kPa was achieved. Besides, a shorter response time than 10 ms was taken in the context of high sensitivity (S > 190 kPa⁻¹) and wide pressure range. As an all-in-one device for the instantaneous detection of pressure information, SP-VFPS is considered as the first demonstration towards color-tunable TIEL with great potential of extensive applications.

3.3. Human-machine interaction
Furthermore, smart electronic devices have changed our daily life in many aspects with the advent of the fourth industrial revolution and 5G information age, especially our relationship with the surrounding environment. Human–machine interactive systems have become a new focus of research. In 2019, Wang et al [29] described an innovative non-contact interactive visualization sensor (NCIVS) based on electrets. An alternating electric field is generated along the trajectory of the floating conductive object after the use of a negatively charged electret in NCIVS, so as to adjust the emission information on NCIVS, as illustrated in figure 8. Such floating objects as symbols, characters and geometric figures can be recognized. In addition, aided by the local charge injection into the electret film, an invisible charged pattern can be successfully created on the NCIVS through a non-contact scanning process, thus identifying external objects and hidden barcodes. In this case, the interactive visualization sensor with non-contact sensing mode can make the object roam over the interface while perceiving a large amount of motion information, which is more favorable in the intelligent human-machine interactive system.

Zhao et al [41] proposed a self-powered user-interactive electronic skin (SUE-skin) in 2020 based on TIEL. It exhibits a low cost and a simple structure that consists of the substrate, a shielding layer, an insulating layer, an electrode layer, and a phosphor layer. Under the trigger pressure threshold of 20 kPa, touch stimulation can be converted into electrical signals and instantaneous visible light. Moreover, a programmable touch operation platform was constructed by integrating a microcontroller unit into the SUE-skin for consumer electronics control (e.g. audio and display). As a result, various touch tracks can be recognized and over 156 types of touch interaction logic can be supported. On this basis, touch stimulus can be perceived by human eyes and monitored by electronic readings.

3.4. Self-powered light source and display
Based on the micro-sized contacts during relative sliding motion, Wei et al proposed a high-intensity TIEL device in 2017, as shown in figure 9(a) [44]. In the luminescence layer, the tiny structure can considerably improve the changing rate of the electric field. The TIEL with micro-sized stamp shows twice luminescence
intensity as that with the plain-surface stamp, thus enabling the application in self-powered illumination and display.

Subsequently, in order to achieve wide-spectrum manipulation, Cheng et al [31] proposed a series of TIEL devices capable of emitting blue, green, red, and white fluorescence, as illustrated in figure 9(b). The motions were tracked by white TIEL, with a high spatial resolution. In this paper, multi-color display is made achievable in the presence of ultra-low mechanical stimulation, which is of much significance to the display-related applications.

A stretchable hybrid luminescent composite (HLC) was proposed by Chen et al [32] in 2019 to generate TIEL under various mechanical stimuli. The HLC is composed of ZnS:Cu and PDMS matrix, as illustrated in figure 9(c). Under TIEL mode, the level of luminescence intensity increases with the polytetrafluoroethylene (PTFE) nanoparticle content, HLC thickness, contact surface stress and sliding object speed within a certain range. Under ML mode, the impact of various strain rates, strain and thickness of HLC on the luminescence intensity is studied as well. The two luminescence modes as discussed above could be superimposed to improve the overall luminescence intensity by 72%, in comparison with what is solely obtained from the ML. It is revealed that this HLC can be applied as self-powered light sources.

In 2019, Wang et al [33] demonstrated an intrinsically stretchable and transparent alternating current electroluminescent (ACEL) device driven by a TENG. The luminescence layer of ZnS:Cu/PDMS composite can be easily cast into any shape, as shown in figure 9(d). An ACEL device with ‘ZZU’ pattern in the EL layer shows the corresponding luminescent pattern as driven by TENG. ACEL devices exhibit persistently bright green EL under external mechanical stimulation, which is presumed to have the potential of applications in robotics and self-powered displays.

At the same time, Wei et al [49] proposed an MDEL structure to successfully integrate the horizontal EL and energy collector into a single device, as illustrated in figure 9(e). The MDEL structure consists of a flexible substrate, a light-emitting layer (ZnS:Cu + PMMA composite), an electrode layer and a tribo-charge layer (FEP). The electrode layer is formed by two sets of coplanar electrodes, each set of which includes a metal block and a set of metal strips as separated by a minimum interval of the same size. Capable to ensure efficient luminescence, integrated MDEL devices have with a more compact structure and a simpler manufacturing process compared with traditional EL devices. The lower friction pressure threshold of $\sim 20$ kPa and the friction speed of $\sim 10$ mm s$^{-1}$ are sufficient to provide a large electric field in the order of MV/m, and the luminescence intensity of MDEL is three times higher compared with other devices without the electrodeless design. A rapidly changing electric field can be generated by the movement of the hand on the energy collector, while custom-made luminescence can be easily achieved by optimizing the geometry of the electrode and designing the required pattern, in which a string of letters can be clearly observed.
3.5. Wearable electronics

In addition, the smart light-emitting electronic devices intended for wearable applications were developed rapidly in recent years, despite power supply is a challenging issue. Therefore, it is essential to develop self-powered luminous systems. In 2019, Park et al. [34] developed a new textile motion driven electroluminescence (TMEL) system. The structure of the system mainly consists of the light-emitting part and PTFE, as shown in figure 10. The article shows that the plain weave structure is the simplest weaving pattern, which allows it to be easily stretched diagonally. Meanwhile, the continuous surface frictions between the warp and weft yarns show the highest durability in all directions. In this system, EL layer is mechanically deformed as triggered by random movement, without needing the bulky batteries with limited lifetime. Continuous light can be emitted through various movements, which is a key step towards developing self-powered systems for wearable applications. Therefore, the self-luminous textile system can be easily demonstrated and applied in intelligent clothing.

4. Conclusions and outlooks

Since the concept of TIEL was first proposed, continuous efforts have been made to understand its mechanism and improve its performance in various applications. Its principle is usually attributed to the EL excited by the strong triboelectrification-induced electric field. TIEL has demonstrated a number of advantages in practical applications. Firstly, the TIEL material is easily prepared without the high-temperature process. Thus, it is cheap and suitable for mass production. Secondly, different from conventional ML materials, it can be characterized using damage-free methods, with ultra-low stress threshold and relatively high responsiveness. Thirdly, the major material utilized is zinc sulfide that has been widely applied in daily activities, which lays a foundation for the application of TIEL.

However, there are still some challenging issues to be addressed in this field. Even though our understanding of ZnS materials has improved significantly, the studies on other TIEL materials are still limited, which requires further exploration. Besides, there is no systematic, unified standard for the accurate evaluation of the electric field, luminescence intensity, and other parameters, which makes it difficult to quantitatively evaluate luminescence effectiveness for the comparison between different devices. Moreover, the studies on the applications of TIEL are still limited. Developing novel functional TIEL materials is still the most important method to expand the application of TIEL. For instance, the reported forms of mechanical energy excitation mainly include human motion, wind, and water, while other energy forms such as vibration, mechanical trigger, and rotation can be further investigated to develop TIEL materials. Since most TIEL materials have the special application scenarios of their own, another way to expand the...
application is to integrate TIEL material into multifunctional devices. In addition, there is a lot of room to realize the intelligent control of TIEL materials, as an emerging direction in the future.

In summary, even though the studies on TIEL are still in the initial stage, it has attracted much attention due to its unique performance and great potential of applications. It is expected that the self-powered luminescence performance can be further improved for its widespread applications in such fields as sensing, anti-counterfeiting, human-machine interaction, illumination and so on.

Data availability statement

No new data were created or analysed in this study.

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