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Review

Conversion Electrode and Drive Capacitance for Connecting Microfluidic Devices and Triboelectric Nanogenerator

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Abstract: Microfluidics is a technique that uses channels of tiny sizes to process small amounts of fluid, which can be used in biochemical detection, information technology, and other fields. In the process of microfluidic development, there are many problems that need to be solved urgently. Many microfluidic systems require the support of external devices, which increases the construction cost, and the electronic interface technology is not mature. A triboelectric nanogenerator (TENG) can harvest mechanical energy and turn it into electrical energy. It has been greatly developed now and is widely used in various fields. Nowadays, many studies are committed to the study of TENGs and microfluidic systems. The microfluidics device can be combined with a TENG to convert fluid mechanical signals into electrical signals for transmission. Meanwhile, TENGs can also act as a high-voltage source to drive microfluidic motion. In this paper, we reviewed the development of microfluidics and related technologies of microfluidic systems in conjunction with TENGs and discussed the form of electronic interface between microfluidic systems and TENG devices.

Keywords: microfluidics; triboelectric nanogenerator; electronic interface; conversion electrode; drive capacitance; liquid sensor

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1. Introduction

Since 1990, people are beginning to create structures and patterns on a micron scale or smaller, including the development of many devices for manipulating fluids (microfluidic devices) [1]. Microfluidics refers to the use of extremely small channels to process small volumes of fluids [2]. Since microfluidics technology was proposed, it has shown great advantages in various industries. It can reduce the number of expensive reagents used in experiments and the footprint of experimental equipment to reduce the costs. In addition, it exhibits high accuracy and sensitivity in molecular analysis and detection [3]. However, the operation of microfluidic devices requires the support of external devices, which reduces their portability and increases the cost of infrastructure. At present, the electronic interface technology between devices is not mature enough, which hinders the development of microfluidics technology [2,3]. In 2012, Wang came up with triboelectric nanogenerator (TENG) technology, which can harvest mechanical energy and turn it into electrical energy [4,5]. When triboelectric pairs are in contact, two thin films with a large difference in electronegativity rub against each other. When the films separate, a potential difference generates because the two films carry opposite charges. By connecting the back electrodes of the two films, the potential difference will cause electrons to flow between the two electrodes. When films coincide again, the potential difference disappears, causing reverse flow of electrons. With such continuous contact and separation, the TENG will output pulse signals, thereby producing electric energy to the outside. So far, the TENG already has four working modes: contact separation, freestanding triboelectric layer, lateral sliding, and single electrode. The simple structure and fabrication process of the TENG is beneficial for integration with microfluidics. Further, using the TENG as an external support device for the microfluidic system can increase the portability of the whole system. At the same time, the manufacturing cost of the system is also reduced. The motion of the target droplet or particle can be precisely controlled under the action of Coulomb force. On the other hand, using microfluidic devices as conversion electrodes to construct TENG can also realize microfluidic sensing and biochemical detection. The combination of the TENG and microfluidics can open up various potential applications for microfluidics.

A micro total analysis system (μ-TAS) is very crucial for biochemical and cell analysis, and has been developed for more than 20 years. Microfluidics is an important component of μ-TAS, which can monitor and control the flow rate and detect the composition and biochemical properties of the fluid [6,7]. However, the complex structure of traditional analysis equipment limits the development of the system. Although some new devices based on acoustic electricity have been proposed for miniaturization applications, most of them are expensive and have complex working mechanisms, which are greatly limited in practical applications [8]. The simple structure of the TENG makes it suitable to miniaturize. People design various fluidic channel-based TENGs based on charge transfer at the solid–liquid interface [9]. The mechanical signal of the target droplet is collected and converted into an electrical signal output through the conversion electrode, and the quantity and nature of the target droplet are judged according to the output signal of the electrode. These studies all indicate that there is great potential for constructing a self-powered micro-total analysis system.

Microfluidics technology requires precise control and manipulation of a droplet, which is the basis for biochemical analysis and drug delivery [10]. This makes the investigation of locally driven fluid methods very important. Electrowetting technology can manipulate droplets by applying an electric field with low energy consumption and high sensitivity [11]. However, in this technique, a dielectric layer separating the fluid from the electrodes is essential, resulting in very high driving voltages [12]. People usually use a power supply as a high-voltage source for microfluidics, which requires the establishment of complex control circuits. This increases the complexity of the entire microfluidic devices and hinders the development of technical applications. TENG's open-circuit voltage can reach thousands of volts [13]. Therefore, a drive capacitor can be constructed. The output of the TENG is exerted on both ends of the capacitor to realize the manipulation of the target droplet under the action of Coulomb force. The ability of rapid response of the TENG can ensure the efficient operation of the microfluidics system and simplify the complexity of the system. This technology has broad application prospects in drug delivery, micromechanics, and other fields.

In this paper, we reviewed the application of microfluidics technology, and summarized the research on the combination of the TENG and microfluidics technology. On this basis, the electronic interface between the TENG system and the microfluidic device is discussed.

2. Microfluidics: A Rapidly Emerging Technology

2.1. The Development of Microfluidics

Microfluidics begins with microanalytical methods. The introduction of gas chromatography and capillary electrophoresis revolutionized chemical analysis under capillary conditions [2]. An early study of on-chip electrophoresis was carried out by Manz et al. in 1994. They pointed out that the integration of capillary electrophoresis on planar microstructures is a step toward μ-TAS [14]. In 1995, Manz further extended the previous research and proposed silicon and glass microstructures for capillary liquid chromatography and capillary electrophoresis to optimize the system performance [15]. Whtesizes et al., in 1998, designed and fabricated a microfluidics system using an elastomeric material poly(dimethylsiloxane) [16]. In 2000, Fair et al. proposed the concept of microelectric fluids. Meanwhile, Quake et al. fabricated an active microfluidics system, including switching valves and pumps using soft lithography [17]. In 2004, scientists proposed organ-on-chip

(OOC) microfluidics. The technique provides new 3D cell culture models, which can better mimic the microscopic features of living organs [18]. With the continuous research of scientists, various innovative microfluidic technologies have been developed, such as acoustic microfluidics [19], optical microfluidics [20], and flexible microfluidics [21]. Nowadays, microfluidics has become an indispensable technology.

2.2. Microfluidics Are Used for Biochemical Analysis

Between 2000 and 2010, microfluidics developed rapidly, especially in the field of chemical/biochemical analysis. Microfluidics has shown great advantages in chemical analysis, which can reduce the number of expensive reagents and precisely manipulate tiny droplets. In 2002, Choi et al. designed a microfluidics biochemical detection device using magnetic beads for rapid and low-volume immunoassays. The system can be used as an immobilization surface and biomolecule carrier The determination takes no more than 20 min, and the required sample volume does not exceed 50 µL [22]. In 2003, McLean et al. established microfluidics integrating cell disposal, electrophoretic separation, rapid cell split, and fluorescent cytosolic dyes. The results showed that the cells were completely lysed within 33 milliseconds and the analysis rate reached 12 cells/min, which was more than 100 times the traditional analysis rate [23]. Techniques that handle small volumes of fluid can also be used to manipulate cell lysis and reduce the degree of sample dilution following lysis. Microfluidics technology provides a rapid and accurate test method for single-cell biology [24]. In 2005, Sudarsan et al. fabricated microfluidic networks based on thermoplastic elastomer gels. The system can be processed at 100 °C and fabricated within 5 min [25]. Lee et al. 2007 fabricated a microfluidics chip with matrix-assisted laser desorption. The chip can analyze biochemical reactions and the required sample volume is reduced to one percent [26]. In 2010, Lien et al. designed a new 3D microfluidics platform that can rapidly isolate and detect cancer cells using magnetic bead-based technology extracted from large sample sizes (e.g., ~1 mL) [27]. Early diagnosis of cancer is very important, and aptamers have shown usefulness as cancer probes. Lin et al. established an integrated microfluidics system to automatically optimize aptamer selection, and successfully screened two aptamers with high affinity for ovarian cancer cells [28]. In 2012, Ota et al. proposed a simple microfluidics approach to generate high-density arrays of Femotoliter-sized microreactors in microfluidic channels. The resulting reactor is small and stable enough for single-molecule biochemical analysis [29]. The birth of microfluidics technology has greatly promoted the development of the field of biochemistry, and up to now, a lot of meaningful results have been achieved.

2.3. Microfluidics Drive Material Delivery

Efficient intracellular drug delivery is crucial for the development of biomedicine [30]. Traditionally, viral vectors and electroporation have been used to deliver external biomolecules into cells. However, they are difficult to maintain cell viability and transport efficiently. It is worth noting that microfluidics has shown great potential in this field [31]. In 2017, Li et al. proposed a novel nanocarrier synthesized with a microfluidic focus. This is a safe and effective VEGF siRNA carrier, which has the potential therapeutic application value of siRNA [32]. Uguz et al. proposed a microfluidics ion pump capable of electrophoretic drug delivery in the absence of solvents. It can transport GABA to target solution in vitro. The introduction of microfluidic ion pumps represents an important advance in the study of implantable drug delivery systems [33]. Mulholland et al. proposed a microfluidics system for drug screening of cancer cell spheroids from tumor biopsies to screen multiple anticancer compounds prior to treatment [34]. Moarefian et al. presented an iontophoresis that accurately detected the anticancer effects of drug delivery at various voltages. They used a hydrogel microfluidic device to simulate the system's mode of operation. Model predictions were then validated by comparing predicted concentrations of fluorescent cationic dyes with actual concentrations in the microfluidic device. The results show that iontophoresis is an advanced drug delivery modality [35]. Grisanti et al. proposed a multi-step protocol for coupling optical and acoustic setups that, together with the versatility of the microfluidics platform, increases the reliability of drug delivery [36]. They assembled a microfluidics chip directly onto the tips of two hydrophobically coated capillaries. The liquid droplets generated by the nano pipettor pass through the capillary to the microfluidics chip to achieve gas–liquid separation [37]. Du et al. fabricated a microfluidics contact lens that uses a stressor to trigger drug release. The microfluidics chip is prepared by photolithography and cast into a curved surface by secondary thermal curing. It was found that microfluidics contact lenses can be loaded with different types of drugs in different regions [38]. Compared with traditional drug delivery methods, microfluidics systems have advantages in terms of sample consumption, reaction time, and running costs [39]. Microfluidics hold great promise for micro-based drug delivery. Figure 1 shows the principle of a microfluidic device for drug delivery [40].

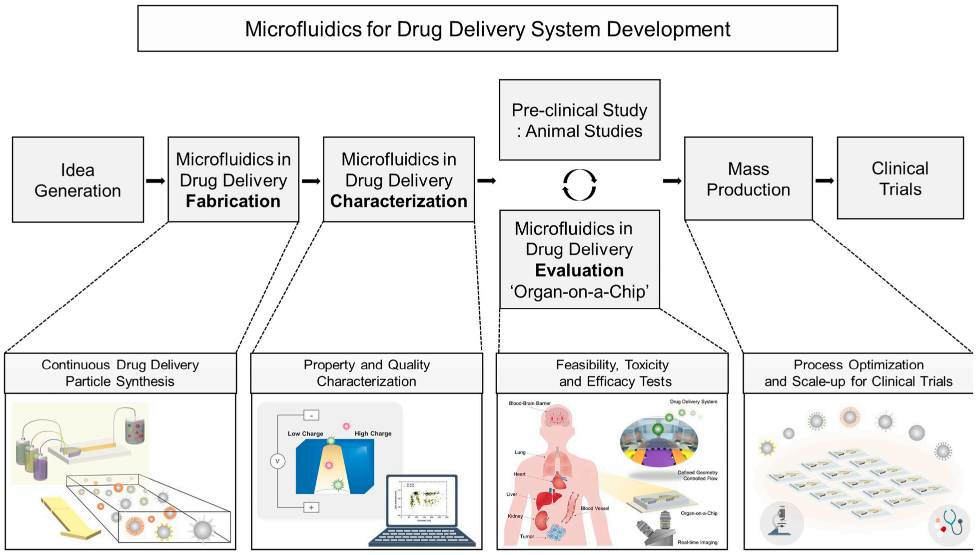

Figure 1. The pipeline of microfluidics for drug delivery [40].

2.4. Challenges Facing Microfluidics Technology

In the study of microfluidics, there are many problems that have to be solved. Examples include material selection for the fabrication of microfluidic amplifiers, operation of microfluidic devices, sample handling, and studies of electronic interfaces between microfluidic devices and external devices. Many microfluidics require external support to function, which reduces portability and increases cost. In addition, the electronic interface technology between microfluidic devices and various external devices is not mature enough, which also limits the development of microfluidics technology.

3. Triboelectric Nanogenerators and Microfluidics: Infinite Possibilities

3.1. Conversion Electrode Based on Microfluidics Device

The miniaturization of microfluidic devices has brought many advantages such as reducing the demand for sample liquid, increasing the test rate, and realizing multichannel testing. However, this also makes the manipulation of the sample liquid more complicated and difficult. However, with the advancement of technology, people have proposed various self-powered sensor-driven droplet microfluidic devices [34]. Microfluidics devices can be combined with the TENG through a conversion electrode. Based on the principle of triboelectrification and electrostatic induction, the conversion electrode can convert the motion signal of the droplet sliding across the friction layer into an electrical output signal. We can judge the property of the fluid according to the output signal of the system; for example, combined with flow sensors to determine the number of droplets.

In 2016, Chen et al. designed a self-powered microfluidic sensor to detect the flow of liquid droplets through the output signal generated by a conversion electrode. A demonstration of patient infusion process monitoring shows that as the infusion rate increases from 3.14 to 62.8 μL/s, the frequency of the output signal increases from 0.2 to 3.0 Hz. This system has shown great advantages in creating self-powered microfluidic devices [39]. In 2017, Chen et al. designed a capillary TENG (ct-TENG) capable of biochemical detection of microfluidics. They used PTFE as the inner wall of the capillary and double-helix aluminum as the electrodes. The model and operating principle of the system are shown in Figure 2a. Microliter sampling can be achieved with high flexibility. The system uses the conversion electrode to convert the motion signal of the droplet into an electrical signal. The study found that the output signal of ct-TENG can judge the categories of water due to the current generated by different water being different. The ct-TENG also can sense trace liquids of different volumes. In addition, the electrical output results obtained by using the system to detect ten standard ion solutions, such as Na^{+}, Mg^{2+}, and Zn^{2+}, are consistent with the conductivity curve measured by the conductivity meter (Figure 2b). In terms of biological monitoring, using the system to detect the total oxygen demand of drinking water can determine whether the drinking water is suitable for drinking. This provides new opportunities for multifunctional sensing and microfluidic biochemical detection technologies [41]. Pan et al. designed a U-TENG in solid–liquid mode and used various liquids to detect the effect of liquid type on TENG's output. The experimental results show that the V_{oc} generated by pure water is 81.7 V and generated by sandwich water is 350 V. They use conversion electrodes to collect the mechanical energy of the liquid and turn it into electrical energy, and successfully lit 60 LEDs and powered a thermohygrometer [42]. Kim et al. prepared PDMS-based microfluidic channels and injected different liquids. The results demonstrate the possibility of detecting liquid species and impurities in liquids by transmitting signals through a conversion electrode. This microfluidic system, as a multi-liquid sensor, is expected to provide a convenient method for the biomedical and chemical industries to detect the type, and physical and chemical properties of the droplet [43].

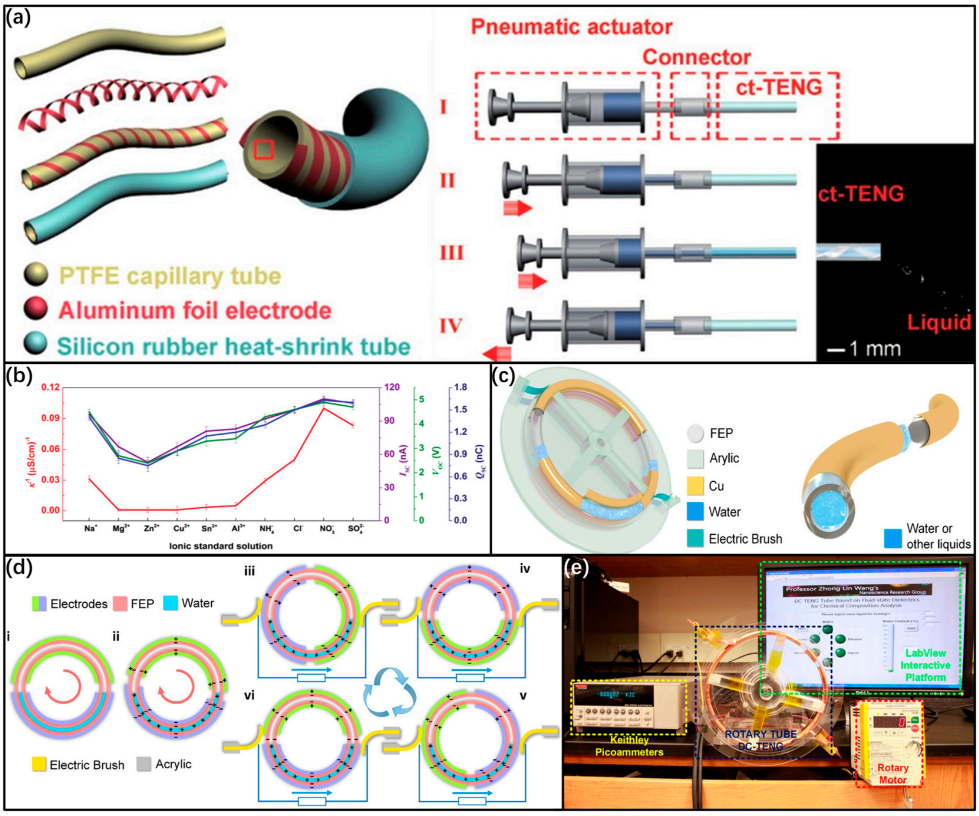

Figure 2. (**a**) The structure, working steps, (**b**) electrical output results, and conductivity curves of the system [41]. (**c**) Model and (**d**) working mechanism of the direct-current TENG [44]. (**e**) Water quality detection and chemical composition analysis platform [44].

Yuan et al. designed a tubular TENG based on solid–liquid contact charging. For detecting subtle differences in touch, press, and stretch pressures [45]. Shahriar et al. designed a mechano-sensing element based on flexible fluids and a conversion electrode in which droplets flow in micro-hydrophobic channels of the polymer, which can generate an output voltage of 20 mV when a pressure of 40 mbar is applied to the electrodes [46]. Wang et al. designed a DC output TENG using the liquid medium interface. It consists of FEP tubes and copper electrodes, and its structure and working mechanism are shown in Figure 2c,d. The contact between the liquid and the copper particles as the fluid dielectric and the FEP tube generates triboelectric charges, which are transported by the conversion electrode. Sandwiching three such structures yielded open-circuit voltages as high as 550 V. This TENG possesses a more powerful energy harvesting capability, which greatly facilitates its development in DC self-powered systems. At the same time, combined with microfluidic devices, it can also be used for biochemical analysis and water quality testing (Figure 2e) [44]. Zhong et al. designed a droplet TENG, which can sense the droplet's movement through the conversion electrode and collect the generated mechanical energy. When the electrode plate length is 0.2 m, 0.4 m, and 0.6 m, the open circuit voltages are 0.89, 5.48, and 4.87 V. The mechanism is shown in Figure 3a,b [47]. Subsequently, Karthikeyan et al. and Zhang et al. proposed a tubular TENG for harvesting water wave energy, which can be collected and converted into electrical energy through corresponding conversion electrodes. These works are of great significance for the development of microfluidic-based TENG [48,49]. Xu et al. proposed an optimized TENG structure with a top conversion electrode to harvest the mechanical energy of droplet motion (Figure 3c,d), which can generate I_{sc} of 0.48 μA. The improvement of detection speed is about 1000 times compared with the application of microfluidic sensors for droplet detection, which provides a new idea for the design of triboelectric microfluidics. The system and its external circuit diagram are shown in Figure 3e [50].

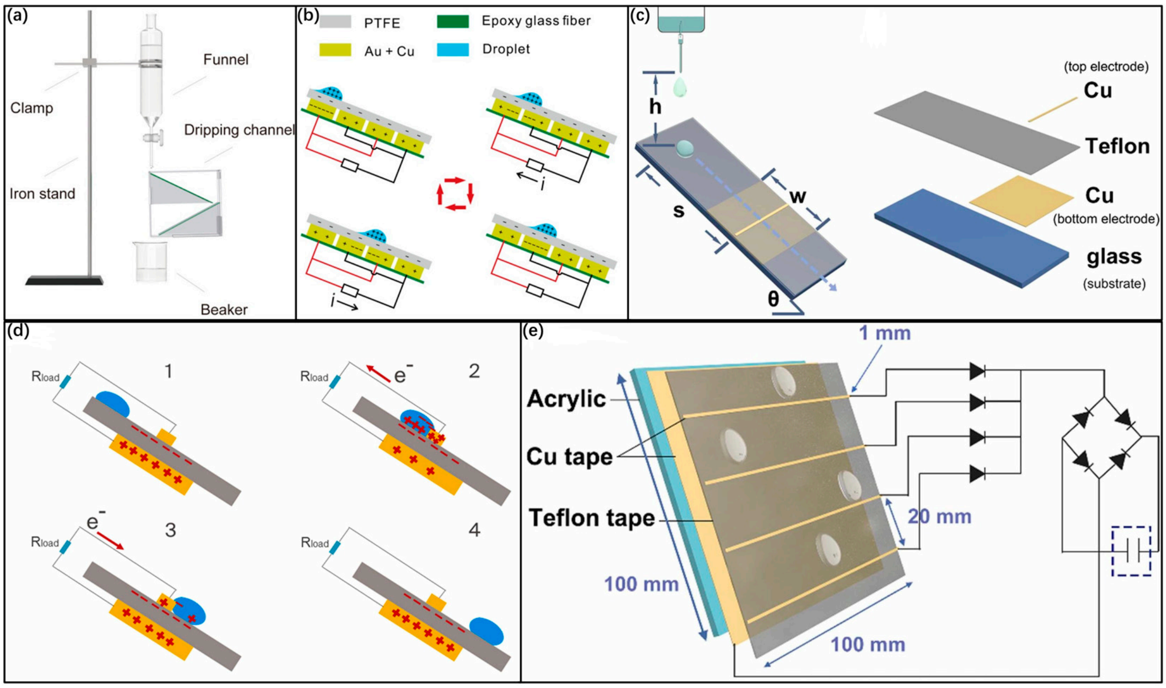

Figure 3. (**a**) The model and (**b**) working mechanism of L-TENG [47]. (**c**) The model and (**d**) working mechanism of D-TENG [50]. (**e**) D-TENG-based droplet energy harvester model and charging circuit diagram [50].

TENG can not only supply microfluidic systems, but also serve as sensing devices for microfluidic systems. Various microfluidic devices combined with TENG also provide more models to harvest energy from droplets. Meanwhile, the micromechanical system drives the sample liquid through the microfluidics device and generates electrical signals in the TENG through the conversion electrode. The TENG's output depends on the characteristics of the sample liquid passing through the microfluidic channel. Therefore, microfluidic sensing can be applied to trace biochemical detection. When the TENG is used as an active sensor, it has a good particle detection capability. Based on the special chemical properties of the polymer material, the acid and alkali resistance of the conversion electrode can be enhanced. The integration of microfluidics and TENG provides more development opportunities for microfluidics biochemical detection technology.

3.2. Drive Capacitance of a Dynamo-Controlled Microfluidics Device Based on Triboelectric Nanogenerators

The TENG's output voltage is very high, but the short-circuit current is very small, which makes it very difficult to directly power commercial electronics. However, these properties also offer opportunities for high-voltage applications. The TENG can not only drive solid dielectric materials, but also liquid droplet motion. Microfluidic systems require the precise control of small amounts of fluid and conventional high-voltage sources, and control circuits add to the complexity of microfluidic systems. Utilizing the high voltage of the TENG as a high-voltage source can simplify the control circuit of the microfluidics system and control the movement of droplets through the drive capacitance. The output of the TENG is exerted on both ends of the drive capacitor, and the TENG studio generates a potential difference across the capacitor. Droplets can move between capacitors under the action of Coulomb force [51].

Nie et al. conducted a series of research in this field. Firstly, they constructed a physical model of a self-powered system. This model can accurately simulate the process of the TENG using drive capacitance to drive droplets. This can help them better study the important parameters that affect the function of microfluidics. Then, based on the TENG and photo-controlled adhesive surface, an intelligent microfluidics system is designed to realize long-distance micro/nano droplet transport. Figure 4a,b show the system's working principle and structure. TENG's output can promote droplets to a maximum distance of 640 mm through the drive capacitor. Furthermore, they came up with the idea of engineering the technique to localized chemical reactions (Figure 4c). The combination of a photo-controlled adhesive surface and the TENG can realize an effective self-powered microfluidics mode system. In addition, they designed a self-powered microfluidics delivery system (Figure 4d). In this system, four liquid droplets are used to carry a tray to form a trolley, and the output of the TENG is exerted on the drive capacitor to allow the trolley to transport some tiny objects on the electrode track. The trolley can withstand a maximum weight of 500 mg and the maximum running speed is 1 m/s. The minimum volume of the droplet carrying the tray is 70–80 nL. Utilizing the high output of the TENG as the driving source of microfluidics has great feasibility and reduces the complicated control circuit. This technology has shown great potential in fields such as drug delivery and biochemical detection [52–54].

Combined with the TENG, Zhou et al. came up with a self-powered microfluidics sensor. This system drives the target solution to flow through the microfluidics channel through the drive capacitance and detects the ion concentration of the target solution according to the generated electrical signal. They also used this system to monitor the degradation of 4-nitrophenol. The model of the systems is shown in Figure 5a,b [55]. Yu et al. built a self-powered droplet processing device. This device consists of brushes, the TENG, and microfluidic devices. As a high-voltage source, the TENG, combined with different brushes and microfluidic devices, can realize different operations on droplets such as moving, separating, mixing, transporting, etc., through the drive capacitance. The results show that the system can drive droplets to complete a long-distance linear motion, and can also drive droplets to move in the circular track composed of 40 electrodes simultaneously. Additionally, it can break up droplets with volumes up to 400 µL. Compared with the traditional mixed droplet method, the system can increase the mixing speed of droplets by

6.3 times. It can be seen that this self-powered microfluidics system has high applicability in the development of drug delivery and biochemical analysis [56]. To address the limitations of current droplet control techniques, Sun et al. assembled a novel self-powered electrostatic operating system with the TENG based on novel polyaniline and polyvinylidene fluoride nanowires. Figure 5c shows the manufacturing process. TENG's output signal is exerted on both ends of the drive capacitance and the driving target is also low to complete different motions (Figure 5d,e). This driving method is 3.63 s shorter than the conventional DC electric field. This new method of controlling the movement of target droplets has shown great advantages in the biochemical detection and directional delivery of drugs [57]. In addition, Zhou et al. also conducted a series of research on this technology. They came up with a self-powered microfluidics system. The TENG acts as a high-voltage source, applying a signal across the drive capacitance. Combined with the microfluidics system, the AC electroosmotic flow and the induced charged electroosmotic flow in the microfluidics channel are successfully realized and the two fluids are mixed. They also used this system to realize the aggregation and separation of particles [58]. Wang et al. designed a strategy for the TENG to power microfluidics systems. When TENG's output voltage is exerted on the drive capacitance, it can drive the movement of particles. On this basis, they further designed a self-powered capillary filling and micro-forming method, which made it easier to fabricate microstructures [59]. Although the high-voltage output of the TENG is difficult to use directly as a power source, applying it to the drive capacitance and combining it with a microfluidics system can precisely control the target droplet. This technology is expected to be used in drug delivery, biochemical detection, etc., and other fields have achieved rapid development [60].

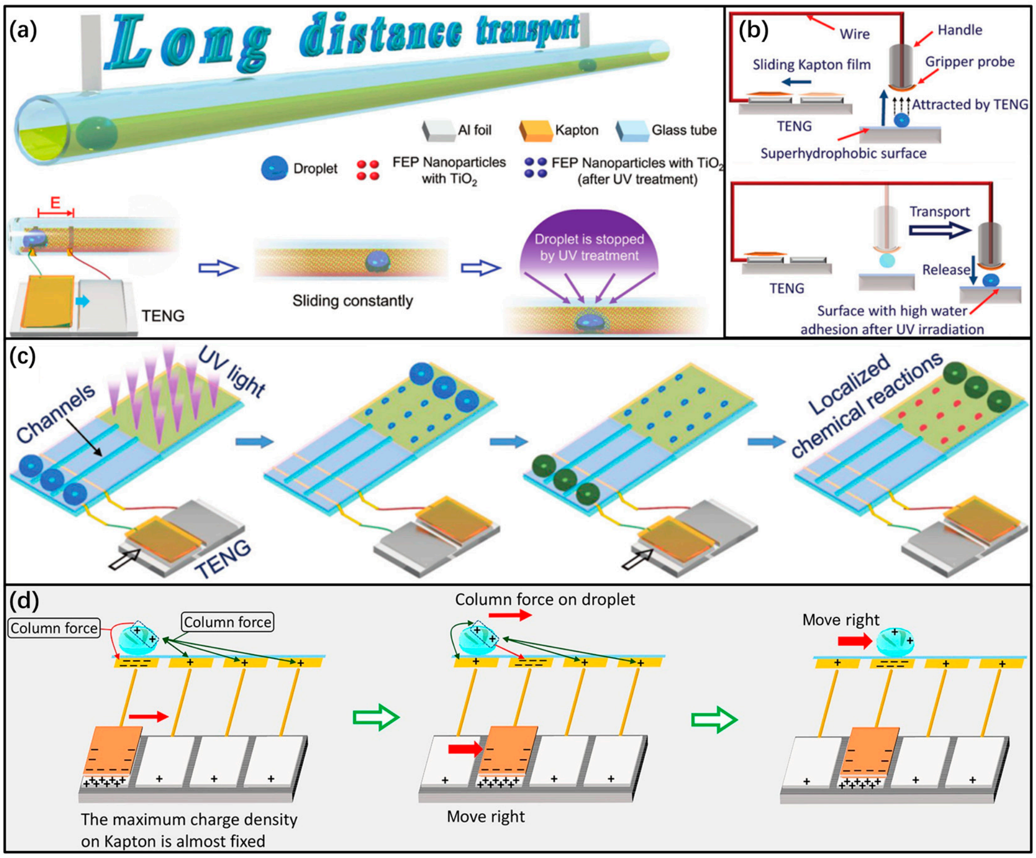

Figure 4. (**a**) The structure of the droplet driving system and (**b**) the motion process of the droplet [53]. (**c**) A model of the system is applied to chemical reactions [53]. (**d**) The working principle of the two-electrode system [54].

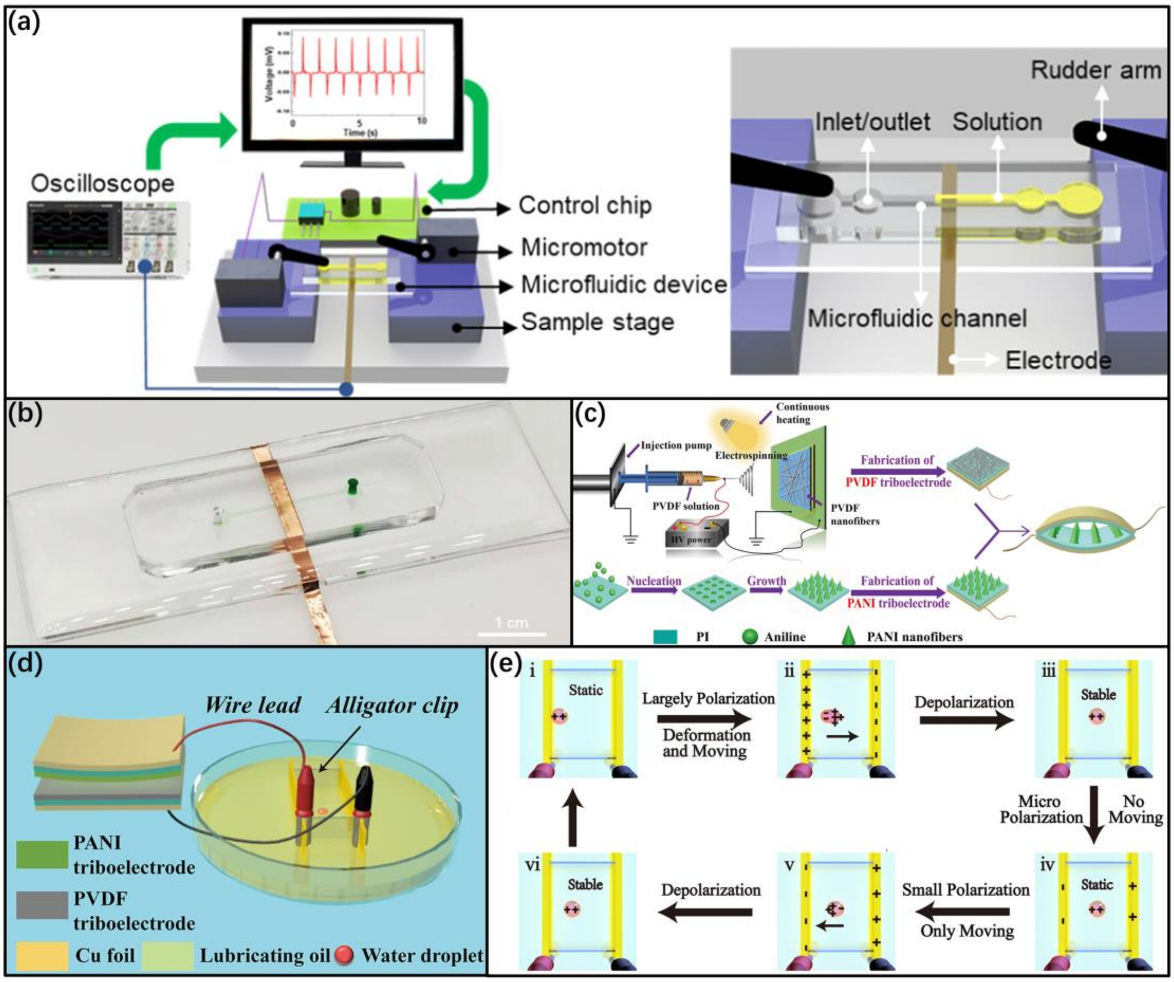

Figure 5. (**a**) Model of the system structure and (**b**) model of the microfluidics device [55]. (**c**–**e**) The NW-TENG and EMS system [57]. (**c**) Assembly process of NW-TENG. (**d**) The dual-motor EMS model. (**e**) The working step of the droplet-driven system.

The TENG can power microfluidic devices by drive capacitance. Under the condition of not needing an external power supply and complex control circuit, the precise control of the sample liquid is realized. The combination of the TENG and microfluidics can realize technological advancement in multiple fields, including inkjet printing, biochemical analysis, drug delivery, etc. Meanwhile, the self-powered technology of the TENG may also enable this smart microfluidic to play an important role in the human–robot interaction of liquid robotic systems.

4. Discussion

The miniaturization of microfluidic devices has brought many advantages such as small sample demand, fast test speed, and multichannel tests. However, the small amount of sample liquid accumulation makes it more difficult to handle the sample liquid. Further, the dependence of microfluidic devices on external supporting equipment also hinders its development [1–3]. In 2012, Wang proposed the TENG, which can turn motion energy into electrical energy [4,61–63]. Nowadays, with the development of technology, scientists have proposed a variety of droplet microfluidic devices driven by self-powered sensors. They can combine with the TENG through a conversion electrode. The mechanical signal of the droplet passing through the friction layer is turned into an electrical output signal through the conversion electrode, and the quantity and chemical properties of the droplet can be judged according to the output signal. The technology is expected to provide biomedical and chemical engineering with an easy way to detect the type and composition of liquids [39,41–50]. In addition, TENG's output voltage is very high but the current is tiny, which makes it very difficult to directly use the TENG to power commercial electronics. However, it also offers opportunities for high-voltage applications. The goal of microfluidics is to precisely control and manipulate small volumes of fluid but using high-voltage sources and control circuits to drive droplets adds complexity to microfluidic systems. The TENG can simplify the control circuit of the microfluidic system and provide a high-voltage source to drive the microfluidic system through the drive capacitance. TENG's output signal is exerted on the drive capacitance, and drives the droplet motion through the Coulomb force generated by capacitance. Therefore, the motion of the TENG can accurately control the movement of liquid droplets [52–60]. The combination of the TENG with microfluidics, electrophoresis, and iontophoresis has broad application prospects in microsolid/liquid manipulators, microrobots, drug delivery systems, and human–computer interactions. However, although the combination of microfluidic technology and the TENG has great development prospects, there are still problems to be solved. A mature microfluidic product usually includes a photoelectric detection module, a signal processing module, a core microfluidics chip, a chip driver platform, human–computer interaction software, and other components. However, the production technology of components is not mature enough at present, and it is difficult to realize the generalization of components. Therefore, the process integration of microfluidic triboelectric systems is still relatively difficult, and there is no fixed specification or standard. These are difficulties that must be broken in order to promote the rapid development of microfluidic triboelectric systems.

5. Conclusions

In previous studies, microfluidic devices can be combined with TENG devices through conversion electrodes. Thus, a variety of self-powered sensor-driven droplet microfluidic devices based on energy harvesting have been proposed, showing great advantages in biochemical detection. TENG's output characteristics make it impossible to directly power electronics. However, its high-pressure nature offers opportunities for integration into microfluidic devices. The TENG can simplify the control circuit of the microfluidic system and provide a high-voltage source for driving microfluidics through the drive capacitance. The combination of the TENG and microfluidic technology has brought new impetus to the fields of biochemical detection and drug delivery.

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