

Recent Studies on Solid–Liquid Contact Electrification

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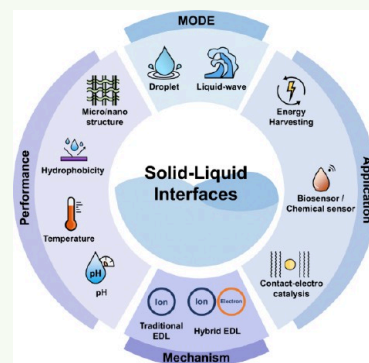
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ABSTRACT: Solid–liquid interfaces are crucial in basic sciences, such as chemistry, biology, and engineering, particularly in catalysis, electrochemistry, and energy storage technologies. Recent developments in the study of contact electrification at solid–liquid interfaces have led to a paradigm shift from traditional interpretations centered on the electric double layer (EDL) concept. These advancements highlight the critical role of electron transfer at the solid–liquid interface, suggesting that both electrons and ions contribute to form a stern layer. This statement requires a reassessment of our fundamental understanding of interfacial charge dynamics, expanding the range of mechanisms that are at work within this crucial boundary layer. This Spotlight on Applications aims to explore the principles of solid–liquid contact electrification (SLCE) and SLCE-based triboelectric nanogenerator (SLCE-TENG) applications. We also examine the effects of different solids, ranging from semiconductors to insulators, and quantify the effect of liquid triboelectric series on contact electrification. Next, we discuss how to optimize triboelectric outputs through solid and liquid material modifications. We address the applications of these findings in SLCE-TENGs, including energy harvesting, self-powered sensors, and contact electrocatalysis. Finally, we highlight the current challenges and offer perspectives on the electrification of solid–liquid interfaces, providing an outline of future research directions and potential.

KEYWORDS: solid–liquid contact electrification (SLCE), solid–liquid interfaces, hybrid electric double layer (EDL), solid–liquid contact electrification-based triboelectric nanogenerators (SLCE-TENGs), advanced solid–liquid contact electrification applications



1. INTRODUCTION

Solid–liquid interfaces have a fundamental role in various basic sciences, including chemistry, biology, and engineering, with widespread applications in catalysis, electrochemistry, and energy storage solutions.^{1–3} Investigating the mechanisms of contact electrification at these interfaces has been a topic of particular interest, leading to significant developments that challenge the conventional understanding centered on the electric double layer (EDL) model.⁴ Recently, it has been reported that not only ions but also electrons contribute to the formation of the EDL in the solid–liquid interface to form a stern layer.⁵ This discovery broadens our understanding of interfacial charge dynamics and opens a new step for technological innovation.^{6,7}

The improved understanding of solid–liquid contact electrification (SLCE) has been crucial in the advancement of SLCE-based triboelectric nanogenerators (SLCE-TENGs). In the growing field of the Internet of Things, TENGs are becoming essential supplementary power sources for low-power electronics.^{8–10} They are driving the development of self-powered sensing devices and innovative charging solutions for implantable medical technologies.¹¹ In recent developments, TENGs have been combined with self-powered sensors and developed for applications such as monitoring gas sensors, self-sensing actuators, and therapeutic applications.^{12–14}

TENGs are also useful in neurotherapeutics and neuro-modulation, pioneering regenerative treatments, electrical-force-mediated microbial control, and enhanced wound-healing protocols.^{15–19} In Figure 1, we describe the timeline of TENG development focused on SLCE-based triboelectric energy-harvesting technologies.

The first TENG with solid–solid contact electrification was invented in 2012, and it developed into SLCE-TENGs in 2013.^{20,21} Then TENGs advanced to various driving modes to utilize an effective way to harness the electrical energy during the middle to end of the 2010s.^{22,23} The focus on efficiency led to a rising demand to understand the underlying principles of the energy-harvesting process. As a result, further studies were conducted to investigate contact electrification between solid–liquid interfaces. These research efforts revealed that electrons, like ions, play a crucial role in the formation of the stern layer, which represents a significant advancement in the mechanisms behind triboelectric energy harvesting.²⁴ Recently, the

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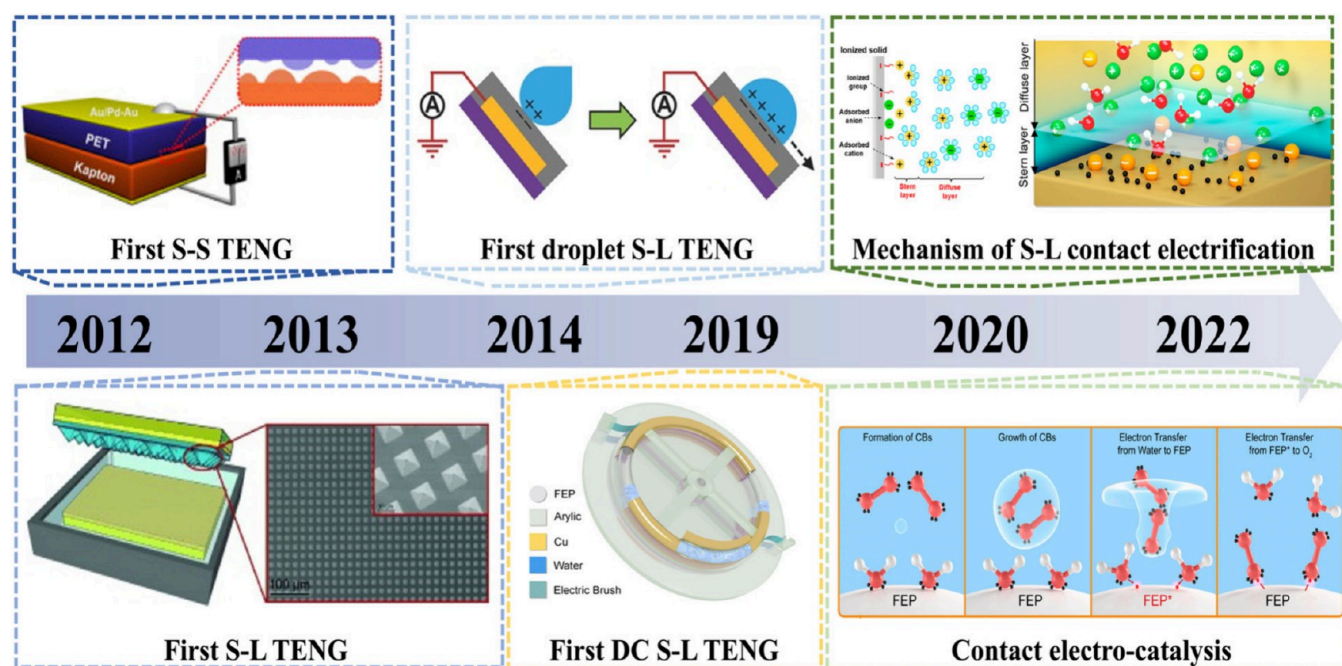


Figure 1. Development of SLCE-TENG in chronological order. Reproduced with permission from ref 20. Copyright 2012 Elsevier. Reproduced with permission from ref 21. Copyright 2013 John Wiley and Sons. Reproduced with permission from ref 22. Copyright 2014 John Wiley and Sons. Reproduced from ref 23. Copyright 2019 American Chemical Society. Reproduced from ref 24. Copyright 2022 American Chemical Society. Reproduced from ref 25. Available under a CC-BY 4.0 license. Copyright 2022 Wang, Z. et al.

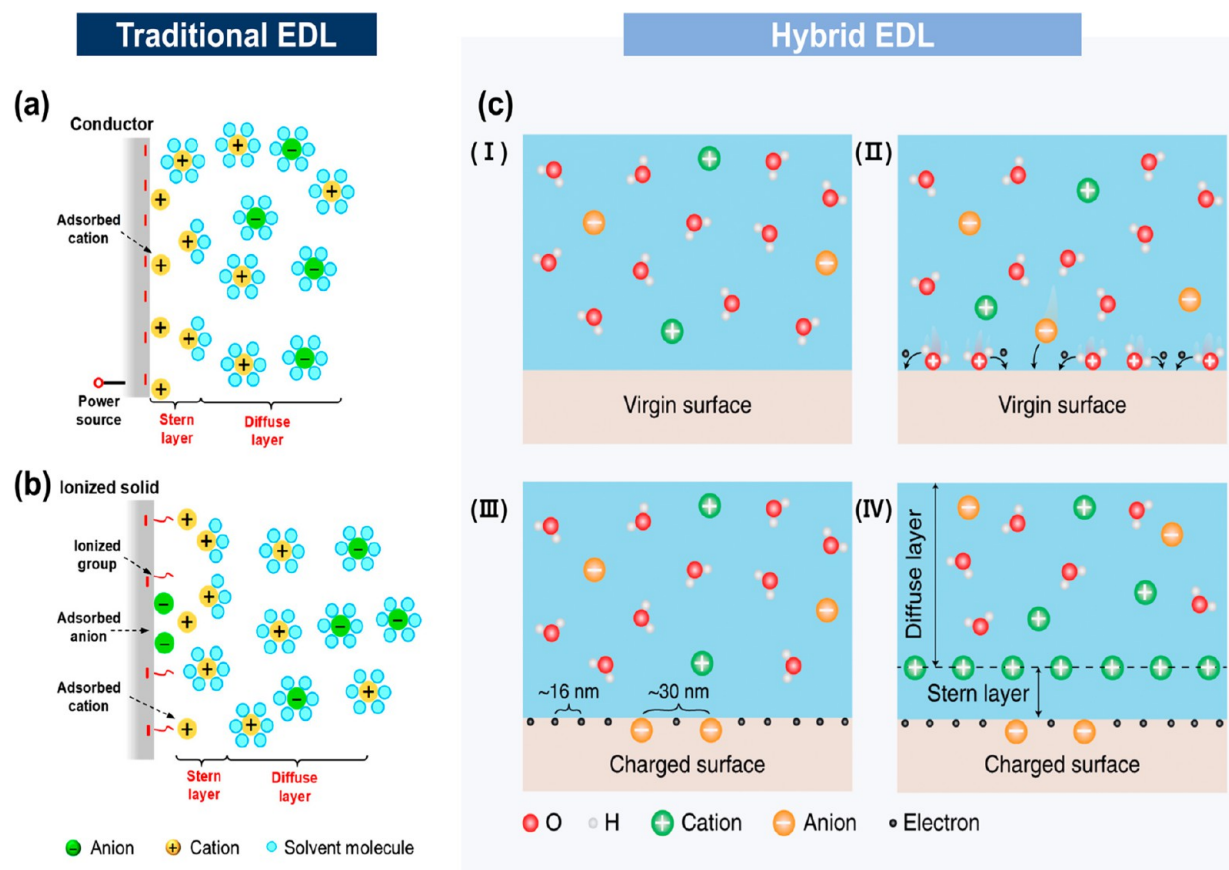


Figure 2. Comparison of the traditional and hybrid EDL. Ion distribution in the traditional EDL of (a) conductor and (b) ionized solid surfaces. Reproduced from ref 24. Copyright 2022 American Chemical Society. (c) Electron and ion distribution in the hybrid EDL. Reproduced from ref 32. Available under a CC-BY 4.0 license. Copyright 2020 Lin, S. et al.

discovery of contact electrocatalysis marked the beginning of a new era for solid–liquid interfaces, offering innovative advancements in this growing field.²⁵

In this Spotlight on Applications, we explore the effects of various solid materials, from conductors to insulators, and assess the impact of the liquid triboelectric series. We also summarize optimization of the contact electrification to enhance the TENG performance. These findings not only pave the way for improved energy-harvesting technologies but also have implications for the development of self-powered sensors and innovative approaches to contact electrocatalysis. In the end, we address the current challenges and perspectives of new directions for the solid–liquid interfaces research field, preparing for a detailed exploration of the mechanisms, applications, and potential advancements of this phenomenon, emphasizing the need for proactive research to unlock the vast potential of contact electrification across diverse practical and industrial domains.

2. STUDIES OF THE SLCE MECHANISM

Recent research on contact electrification has revealed the dynamics of charge transfer at the solid–liquid interface. This interfacial layer between solid and liquid, teeming with electrochemical activity, has been the subject of extensive study, with traditional models focusing predominantly on ion adsorption to elucidate the formation of the EDL.²⁶ Recent research in this area is expanding the focus beyond ions to include electron transfer in an integrative model.²⁷ This research has implications for practical applications such as the improvement of energy-harvesting systems and the development of new environmental cleaning techniques.^{28,29} In this section, we aim to explain SLCE by exploring the traditional EDL model and the newer hybrid EDL model. First, we classify the traditional EDL model, which has been used to understand charge distributions focused on ions at the solid–liquid interfaces. Subsequently, we move to the hybrid EDL model, which combines electron transfer and ion adsorption. This model presents a more comprehensive understanding of charge dynamics in the solid–liquid interfaces. By analysis of these two models together, our goal is to comprehend the SLCE mechanism.

2.1. Traditional EDL. The traditional EDL model epitomizes a foundational construct in electrochemistry, proving the complex electrostatic interactions at the solid–liquid interfaces.³⁰ First, the EDL model was proposed by Helmholtz. He found that, in the electrode–electrolyte interfaces, there are two layers of opposite charges constituted with a small distance. Then, Gouy and Chapman modified the model so that the ions are not closely attached on the electrode (solid), and they built a thin layer, merged these two models, and defined it as a stern layer, which is a collection of adsorbed ions firmly attached to the solid surface and diffuse layer so that ions gradually dissipate into the adjacent liquid, as depicted in Figure 2a. The contiguous liquid infuses ions into the outer layer, resulting in a net charge. This charge attracts counterions from the bulk liquid, forming the diffuse layer and completing the EDL. For the view of SLCE, the traditional EDL is assumed to be ion transfer simply because ions are often present in liquids, such as H^+ and OH^- in water.³¹

The traditional EDL divides the ion distribution into solid conduction states. For the conductors, the charge carriers are free electrons that exhibit a high mobility within the materials. When a conductor comes into contact with a liquid, it can

rapidly exchange electrons with the liquid, resulting in the formation of ions in the liquid and creation of the EDL (Figure 2a).²² The inner layer is composed of the conductor's surface charges, while the outer layer, known as the diffuse layer, is made up of liquid ions that neutralize the conductor's charge. An insulating solid can become conductive when applied to an external field (Figure 2b). In ionized solids, the charge carriers are ions, and when an ionized solid interacts with a liquid, solid ions are adsorbed at the interface and contribute to the formation of the EDL. This interaction is slower and less dynamic compared with that of conductors because these solid ions are less mobile than free electrons.

2.2. Hybrid EDL. The traditional EDL model assumes that ions act as charge carriers and form an EDL through charge transfer. Recent advances in contact electrification research, particularly at the solid–solid interface, have highlighted electron transfer as a major mechanism of charge transport. Inspired by these findings, we expanded the study of solid–liquid interfaces. Lin et al. postulated that electron transfer, in conjunction with ion transfer, occurs at the solid–liquid interface.³² This insight paved the way for the development of a hybrid electric bilayer model, which posits a dual-transfer process involving both electrons and ions. They verified electron transfer at the SLCE with several experiments using kelvin probe force microscopy (KPFM) to distinguish it from ion transfer. They measured surface charge densities on different solid materials after their interaction with deionized (DI) water and aqueous solutions at varying temperatures. The experimental data suggested that the transient charges consist primarily of electrons rather than ions, which exhibit a reluctance in detaching from the surface. Also, they found that higher temperatures increase the thermionic emission of electrons, leading to a faster decrease in the electron density on the surface. This result underlined the pivotal role of electrons in the SLCE and led to the design of a hybrid EDL.

The formation of the hybrid EDL through SLCE involves a complex interplay of electron and ion transfers, encapsulated within a “two-step” model, as shown in Figure 2c. The model begins with the interaction between a solid and liquid “virgin” surface (Figure 2c-I), where the molecules and ions within the liquid interact with the solid surface due to thermal motion and liquid pressure. The overlapping electron clouds of solid atoms and liquid molecules enable electron transfer between them, resulting in electron transfers from the liquid to the solid and potential ionization reactions on the solid surface. This process results in a charged surface (Figure 2c-II). After the initial charge generation, the surface acquires an electrical charge, mainly electrons (Figure 2c-III). Ions of the opposite polarity in the liquid are attracted to this charged surface through Coulombic attraction, which screens the initial charge and results in the formation of the EDL (Figure 2c-IV). The resulting EDL consists of a closely adherent stern layer and a more diffuse layer, which integrate both electrons and ions near the solid surface.

3. SLCE-TENG

From fundamental studies in SLCE, the development of SLCE-TENGs has emerged as an innovative approach to energy harvesting.³³ SLCE-TENGs can adapt to various environmental conditions such as rain drops or ocean waves.³⁴ The efficiency of the SLCE-TENGs is based on the interaction between solid conductive profiles and liquid physicochemical attributes.³⁵ In this section, we aim to

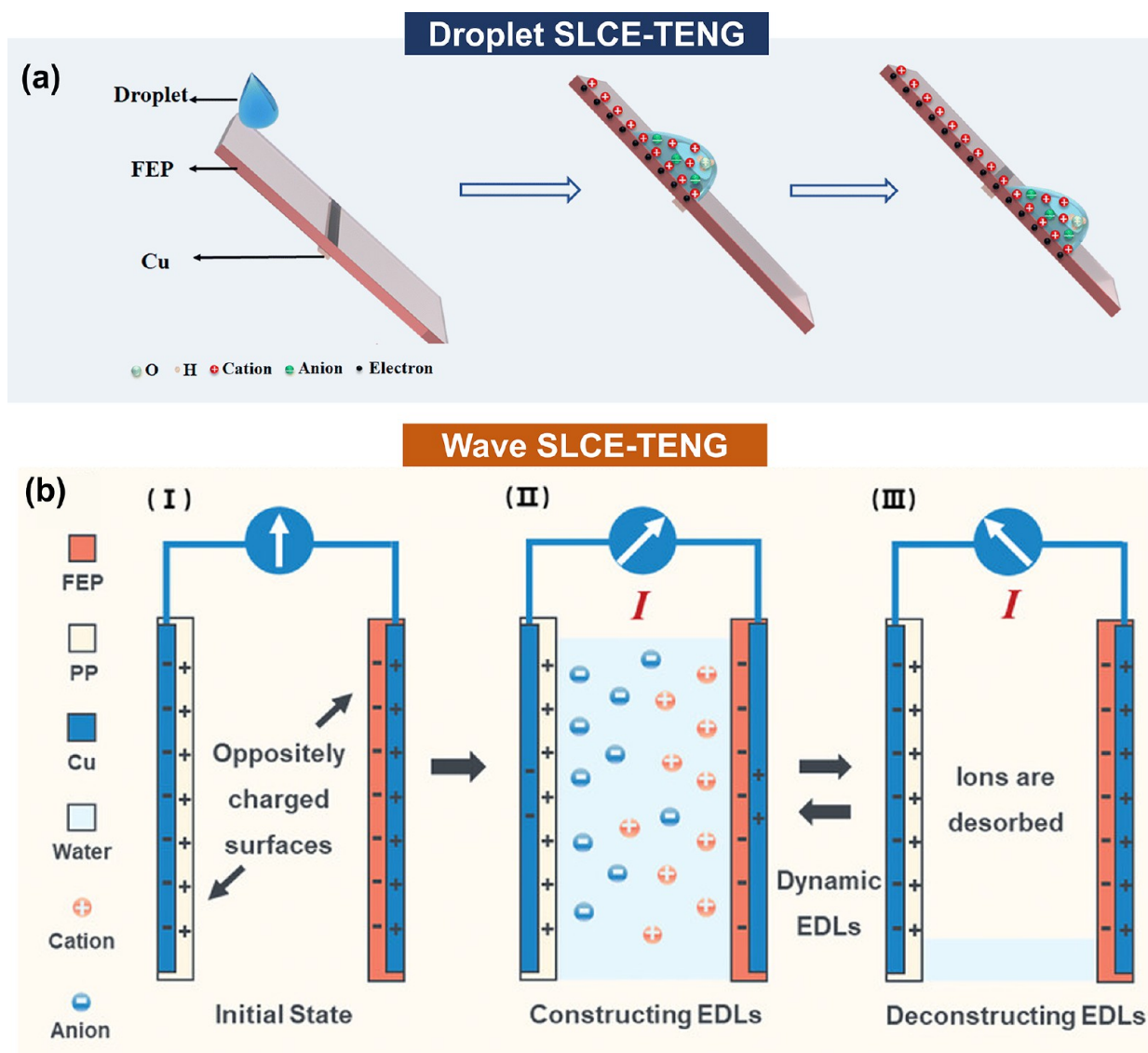


Figure 3. Representative SLCE-TENG modes. (a) Schematic of the liquid droplet SLCE-TENG when the liquid droplet is sliding on the FEP solid surface. Reproduced with permission from ref 43. Copyright 2023 Royal Society of Chemistry. (b) Working mechanism of the liquid wave SLCE-TENG when water comes in contact with the precharged PP and FEP surface. Reproduced with permission from ref 46. Copyright 2023 John Wiley and Sons.

organize the operating modes of SLCE-TENGs and define the materials of solid and liquid using outlining methods to improve the performance of SLCE-TENGs' research directions and potential.

3.1. SLCE-TENG Modes. SLCE-TENGs utilize the triboelectric effect, which occurs when materials come into contact, particularly at the interface between a solid and a liquid. During contact electrification, solid materials and liquids become charged. These triboelectric charges are often facilitated by movements such as shaking, stirring, or flowing.³⁶ The operation of SLCE-TENGs is based on repeated contact and separation between solid and liquid phases. This process results in charge accumulation at the solid interface, which generates an electric potential. An electrical current is generated when a potential difference drives electrons through an external circuit from the solid to the liquid. SLCE-TENGs continuously generate electrical output through contact separation from various mechanical forces, such as acoustic vibrations and fluid dynamics.³⁷

3.1.1. Liquid Droplet SLCE-TENG. Liquid droplet SLCE-TENG operation is based on the interaction between liquid droplets and a solid surface.³⁸ Liquid droplet SLCE-TENG converts the mechanical energy of moving liquid droplets into electrical energy through the principle of contact electrification and electrostatic induction.^{39,40} When a liquid droplet contacts a solid surface, charge transfer occurs due to contact electrification between the two materials. This charge can cause charge separation through electrostatic induction in an adjacent electrode. As the droplet moves, the distance between the droplet and the electrode changes, resulting in a variable electrical potential across the interface.^{41,42} The mechanical energy of a moving droplet is converted into electrical energy by inducing an alternating current (ac) in any connected external circuit due to dynamic fluctuations in the potential Wang et al.-developed liquid droplet SLCE-TENG, as shown in Figure 3a.⁴³ Wang and colleagues used fluorinated ethylenepropylene (FEP) as a solid surface, DI water, and 0.01 mol L⁻¹ NaCl droplet of the liquid to quantify the charge-

transfer properties. They also use the charges varying the sliding droplet speed, height of the droplet falls, and ion concentration of the droplet to understand how to achieve high performance of liquid droplet SLCE-TENGs. We summarize the liquid droplet SLCE-TENG performance in Table 1 to provide mostly used materials of solids and liquids with the electrical output.

Table 1. Performances of the Liquid Droplet SLCE-TENGs

solid	liquid	electrical output		ref
		voltage (V)	current	
FEP	oil	1	0.8 nA	5
FEP	DI water	400	15 μ A	28
PTFE	ethanol	0.09		38
PTFE	DI water	0.1		38
PTFE	DI water	23.5	553 nA	39
PTFE	DI water	62.2	86.5 μ A	40
silicon	DI water	1.1	4.26 nA	40
fluorinated SiO ₂	glycerol		2.5 pA	41
MoS ₂	1 mol L ⁻¹ NaCl	6	5 nA	42

3.1.2. Liquid Wave SLCE-TENG. The liquid wave SLCE-TENG is another mode that converts the mechanical energy of liquid wave movement into electrical energy.^{44,45} Liquid wave SLCE-TENG is immersed in liquid and focuses on the electrical potential differences in solids caused by the movement of the liquid.⁴⁶ As liquid waves interact with the SLCE-TENG, they induce mechanical motion, which generates triboelectric charges on the solid surfaces of different materials within the device.^{47–49} These charges are then transferred through an external circuit, converting mechanical motion into electricity.^{50,51} Liang et al. newly proposed liquid wave SLCE-TENG based on the dynamic electric double to harvest energy from the water wave.⁴⁶ Figure 3b demonstrates the working mechanism of the liquid wave SLCE-TENG. Before being placed in the liquid, the two film surfaces of the liquid wave SLCE-TENG, polypropylene (PP) and FEP, are initially and oppositely charged through contact electrification or other methods. The PP film surface is positively charged, while the FEP film surface is negatively charged, as shown in Figure 3b-I. When the liquid wave SLCE-TENG is submerged in liquid, the liquid wave SLCE-TENG is filled with water, as seen in Figure 3b-II. Opposite EDLs are formed to neutralize the surface potentials of the two films, causing free charges to flow in the external circuit through the back electrodes. When the water level drops below the TENG, the EDLs on the film surfaces deconstruct, as depicted in Figure 3b-III. As a result, the surface potentials tend to return to their original state, and the free charges flow back, and if there are continuous waves to rise and fall, dynamic EDLs will be periodically constructed and deconstructed, forming a dynamic boundary at the solid–liquid–gas interfaces. These dynamic EDLs can drive free charges to flow back and forth between the two electrodes, resulting in the ac output of the liquid wave SLCE-TENG. The liquid wave SLCE-TENG is suitable for use in areas with high liquid wave activity, such as coastal regions or near dams. Table 2 compiles recently designed liquid wave SLCE-TENGs and their electrical performances with commonly used materials for solids and liquids.

3.2. Influence of the Solid Conductivity at SLCE-TENGs. This section discusses the contact electrification process in the SLCE-TENG and evaluates the effect of the

Table 2. Performances of Liquid SLCE-TENGs

solid	liquid	electrical output		ref
		voltage	current	
FEP	pure water	93	480 nA	33
FEP	distilled water	20	400 nA	36
PTFE	distilled water		2.68 μ A	45
FEP	DI water	37	100 nA	47
FEP	1 mol L ⁻¹ KCl	2.5	120 nA	47
SiO ₂	ultrapure water	0.4	25 nA	48
PTFE	tap water	22	1.45 μ A	49
PTFE	tap water	27.2	3.84 μ A	50
PTFE/PI	oil	6	12 nA	51
PFTS/PP	DI water	20	400 nA	52

solid conductivity on the performances of SLCE-TENGs. The study distinguishes between semiconductors and insulators based on their electrical properties. For conductor-based SLCE-TENGs, charge accumulation is inherently hindered by the high electrical conductivity characteristic of conductors due to rapid neutralization of the generated charges by the free charge carrier. Therefore, we are focused on the influence of contact electrification at semiconductors and insulators, which have lower conductivity than conductors.

3.2.1. Influence of the Semiconductor Solid at SLCE-TENGs. Lu et al. described the semiconductor-based SLCE-TENGs as a significant advancement in energy-harvesting technology.⁵² Unlike traditional TENGs that generate ac through the triboelectric effect, they found that the semiconductor-based SLCE-TENGs can produce continuous direct current (dc) by utilizing the dynamic characteristics of a PN junction. The design incorporated water droplet as a mobile dielectric layer between p-type and n-type silicon semiconductors shown in Figure 4a. This setup polarized water molecules, propelled by the disparity in the Fermi levels of the semiconductors, resulting in the accumulation of free charge carriers at the solid–liquid interface. This interaction created an electrostatic and polarization equilibrium, resulting in an electrical output of up to 0.3 V and currents as high as 0.64 μ A. This electrical output demonstrated the ability to power portable energy generation devices, representing a significant advancement in energy extraction from water droplets.

Studies are also underway to combine triboelectric energy harvesting with other technologies, such as piezoelectric and electromagnetic, to increase the electrical output. Zheng and his colleagues developed a method that utilizes both the photovoltaic and tribovoltaic effects at the solid–liquid interface to generate dc by sliding a DI water droplet on a semiconductor wafer.⁵³ In Figure 4b, they delve deeper into the tribovoltaic effect by analyzing its occurrence at the interface between a liquid and a semiconductor, especially with light irradiation. Notably, the intensity of light has a significant effect on the triboelectric current, as opposed to the triboelectric voltage. They analyzed that the light can highlight how photoexcited electron–hole pairs contribute to the triboelectric current, which enhances the electrical output by increasing the charge-carrier concentration at the interface. Similar to p-type silicon, the triboelectric current for n-type silicon also increased with rising light intensity. The study supported the role of light-induced electron–hole pairs in enhancing triboelectric charge generation and provides a coupling effect that can enhance the electrical property.

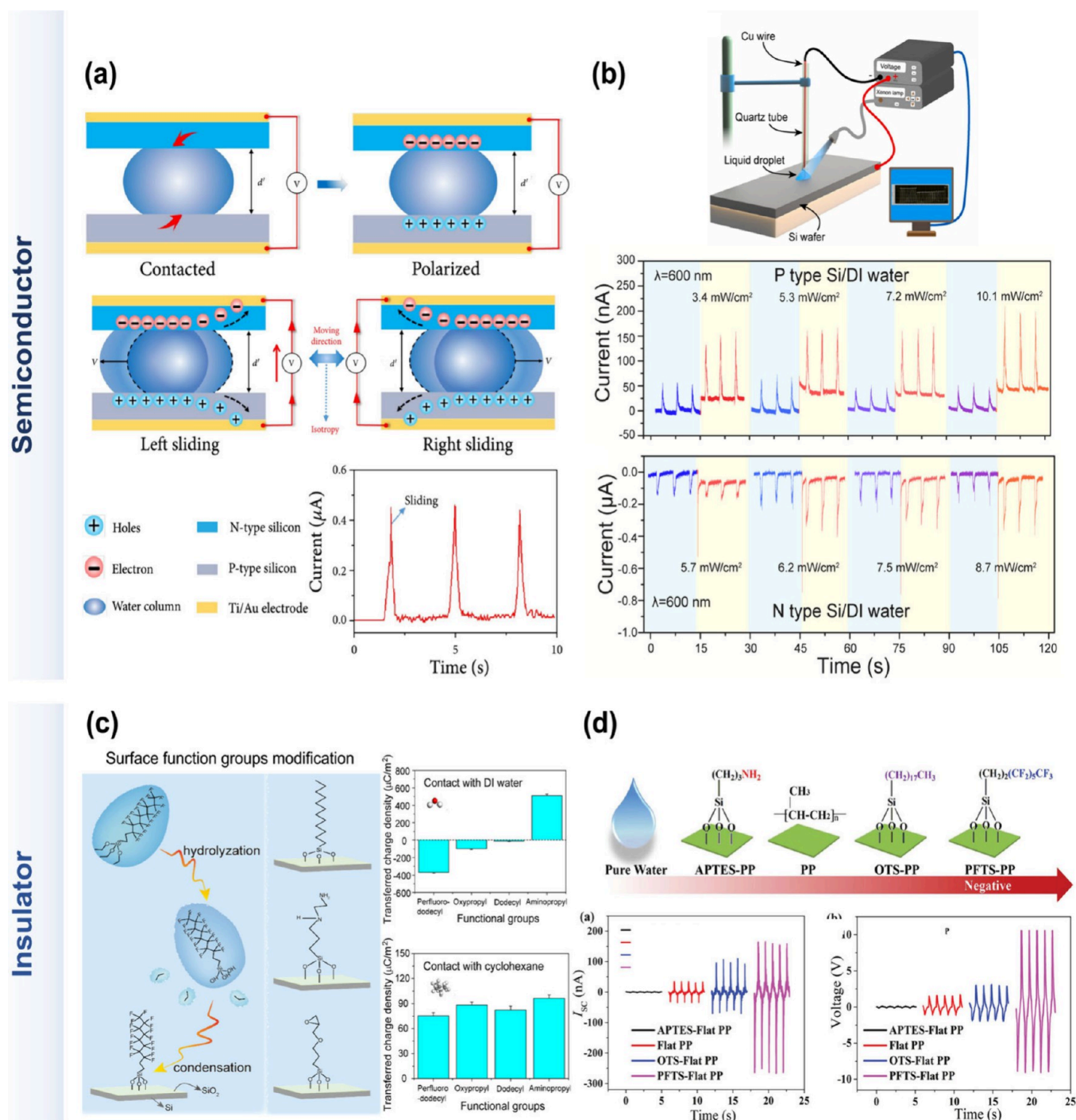


Figure 4. Influence of the semiconductor/insulator solid at SLCE-TENGs. (a) Schematic image of charge transfers due to water movement between two semiconductors, output of the current. Reproduced from ref 52. Available under a CC-BY 4.0 license. Copyright 2021 Lu, Y. et al. (b) Setup of the measurement of the triboelectric current between water and p-type silicon (n-type silicon). Reproduced with permission from ref 53. Copyright 2021 Elsevier. (c) Illustration of the chemical-functional-group-modified surface by hydrolyzation of the silane coupling agents. Transferred charge density according to different functional-group-modified SiO₂ surfaces in contact with DI water and cyclohexane. Reproduced from ref 54. Copyright 2020 American Chemical Society. (d) Triboelectric series, TENG based on the diverse modified PP performance of the current and voltage. Reproduced with permission from ref 55. Copyright 2019 John Wiley and Sons.

3.2.2. Influence of the Insulator Solid at SLCE-TENGs. SLCE focused on insulator solid surfaces and liquids has also been studied. Lin et al. developed an energy band model to explain electron transfer between solid–liquid interfaces and determine its contribution.⁵⁴ They modified SiO₂ surfaces with various chemical functional groups and measured the surface charge density using KPFM. They modified the SiO₂ surface

using silane coupling agents with different functional groups (perfluorododecyl, oxypropyl, dodecyl, and aminopropyl) containing F, O, C, and N elements in the experiments. The results indicated that more electrons transferred from the impurity states of the DI water to the SiO₂ surface. Modifying low-electron-affinity functional groups, such as aminopropyl, on the SiO₂ surface introduced occupied high energy levels and

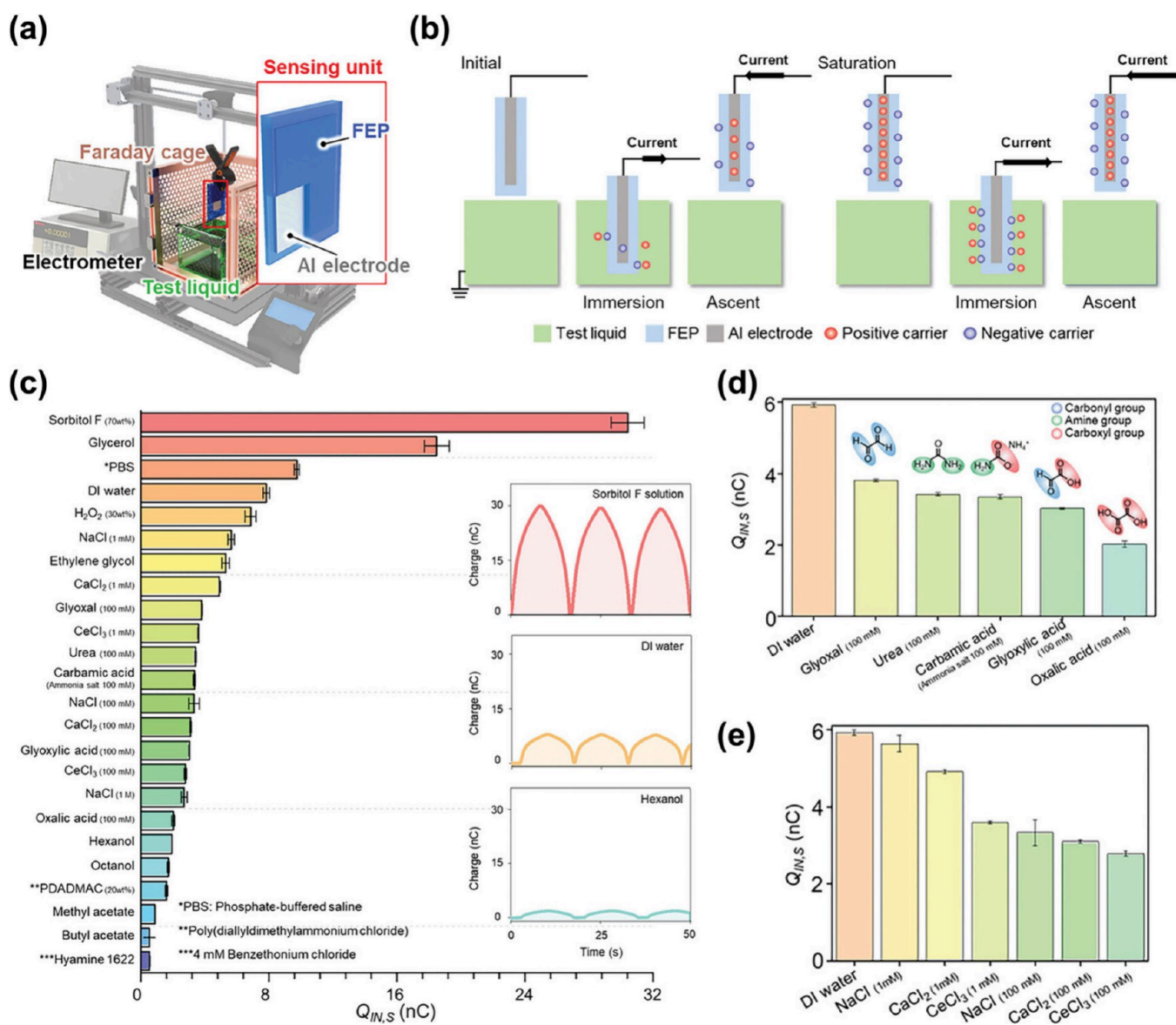


Figure 5. Liquid triboelectric series at SLCE. (a) Setup to measure liquid triboelectric series. (b) Sensing mechanism at initial and saturation state. Induced charge (c) at saturation state of various liquids. (d) at saturation state according to carboxyl, amino, and aldehyde groups. (e) at saturation state according to effect of cations. Reproduced with permission from ref 56. Copyright 2023 John Wiley and Sons.

raises the highest occupied energy level in the surface states of SiO₂. Therefore, electron transfer occurred from the SiO₂ surface to the DI water side, as depicted in Figure 6c. Li and colleagues enhanced insulator surfaces by chemically modifying flat PP films using 3-aminopropyltriethoxysilane (APTES), octadecyltrichlorosilane (OTS), and 1H,1H,2H,2H-perfluorooctyltrichlorosilane (PFTS), as shown in Figure 6d.⁵⁵ This modification optimized the film's triboelectric performance by minimizing the retention of DI water on the PP film's surface during solid–liquid interaction. During examination of the insulators in SLCE-TENGs, the electrical output of these insulators was observed to increase as their triboelectric charge became more negative.

3.3. Liquid Triboelectric Series at SLCE-TENG.

Quantifying the triboelectric series has mainly focused on solids, which provides a fundamental understanding of the way materials can readily donate or accept electrons. In a pioneering study of the liquid triboelectric series, Yoo et al. extended these studies to SLCE to introduce the liquid

triboelectric series.⁵⁶ To measure the triboelectricity of liquids, they developed a platform for measuring the amount of charge, as shown in Figure 5a. The sensing unit consisted of an aluminum electrode connected to an electrometer and an FEP film that encapsulated the electrode. Figure 5b illustrates the operational mechanism of the platform, which begins by immersing the sensing unit into the test liquid after the initially induced solid surface charge and test liquid charge. When the solid with FEP was immersed in each test liquid, and a carrier is detected and increases with the increase in the contact area between the sensing unit and the liquid. After the motion stopped, the amount of charge reached its maximum. An induced charge was calculated from the difference between the maximum and minimum values of the measured electrical signal.

Yoo and their colleagues used this validated measurement of liquid triboelectric charges to study different types of liquids and summarized it in Figure 5c, which allowed them to establish a “liquid triboelectric series”. They also compared the

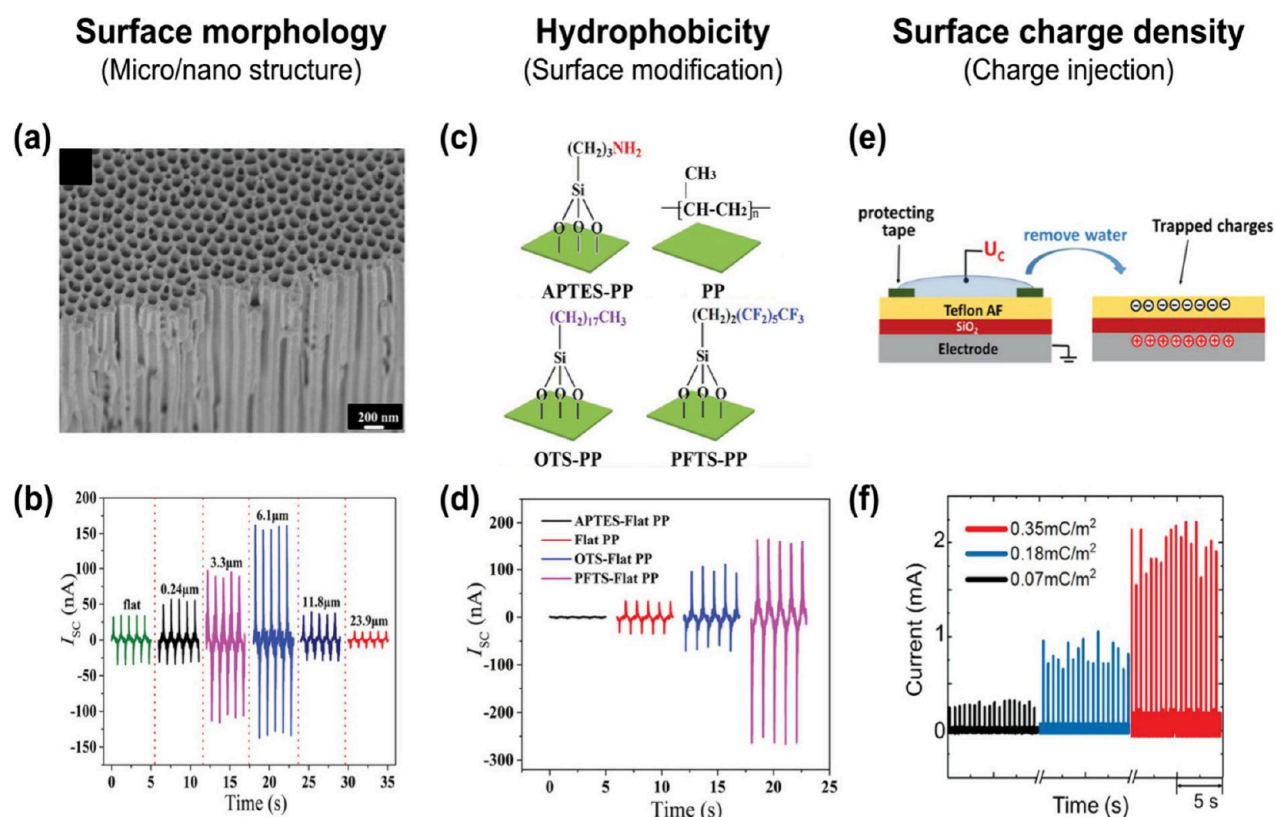


Figure 6. Surface modification factors to enhance the SLCE-TENG performance. (a) PP NWs view on AAO. (b) Output of SLCE-TENG based on PP NWs with different lengths. Effect of the hydrophobicity on the performance. (c) Schematics of the surface functional group with APTES-PP, PP, OTS-PP, and PFTS-PP. (d) Output performance varying to the functional group. Reproduced with permission from ref 55. Copyright 2019 John Wiley and Sons. Effect of the surface charge density on the performance. (e) Schematic representation of the homogeneous electrowetting-assisted charge-injection method. (f) Output change according to the charged surface. Reproduced with permission from ref 58. Copyright 2020 John Wiley and Sons.

effects of different chemical groups, including amino, carboxyl, and aldehyde groups, as shown in Figure 5d. The carboxyl group was found to be ionized when dissolved in DI water, which hindered SLCE. Unlike the carboxyl group, amino and aldehyde groups maintain their original molecular form when dissolved in DI water. However, when these chemical groups dissolved in water, they also degraded contact electrification, causing the triboelectric charges to gradually be decreased in the following order: carbonyl groups to carboxyl group. The study also investigated the effect of ion valence on the liquid triboelectric effect, comparing NaCl, CaCl_2 , and CeCl_3 solutions with concentrations of 10^{-3} to 1 M. The results indicate that each solution contains Na^+ , Ca^{2+} , and Ce^{3+} with the charges gradually decreasing in the following order: DI water > NaCl > CaCl_2 > CeCl_3 , with low-to-high concentration shown in Figure 5e. These results can be analyzed in terms of the ion valence and total ion concentration. A higher triboelectric output was observed in DI water as compared with other ionic solutions. This effect is primarily attributed to impurities, specifically ions, which inhibit SLCE. This reduction in triboelectric charges is also confirmed to intensify with an increasing ion concentration. Moreover, the valence of the ions is of great importance. Yoo et al. found CeCl_3 , which has a higher ion, exhibits significantly lower triboelectricity compared to NaCl. Further studies have corroborated the substantial influence of ion valence on the triboelectric properties of solutions.⁵⁶

3.4. Optimizing the SLCE-TENG Performance. To enhance the performance of SLCE-TENGs, it is crucial to achieve advanced material engineering at each interface to maximize the triboelectric output. Here, we first introduce strategies of solid surface modification such as adjust surface roughness or coatings and increase surface charge density.⁵⁷ We then focus on the liquid part to optimize the formation of the EDL through contact electrification. This requires selecting liquids with specific ionic characteristics that, when interfaced with suitably modified solid surfaces, enhance the EDL's ability to separate charges, resulting in a significant increase in the electrical output of the SLCE-TENGs.

3.4.1. Solid Modification to Enhance the SLCE-TENG Performance. In Figure 6, we summarize several methods to modify solid which can produce high performance SLCE-TENG. In Figure 6a,b, Li and co-workers explored the impact of surface morphology on the efficacy of SLCE-TENGs, using films with varying lengths of nanowires (NWs).⁵⁵ They modified the surface roughness of PP films adeptly by employing anodic aluminum oxide (AAO) as a template to synthesize PP NWs. The investigation covered TENGs that used flat films and those that featured NWs up to 23.9 μm long. The hypothesis was that an increase in the length of NWs would amplify the output due to the expanded surface area. Empirical findings supported this hypothesis, showing that elongated NWs significantly increased the solid-liquid contact area, thereby enhancing the triboelectric output. However, they found when the NW length exceeds a certain threshold,

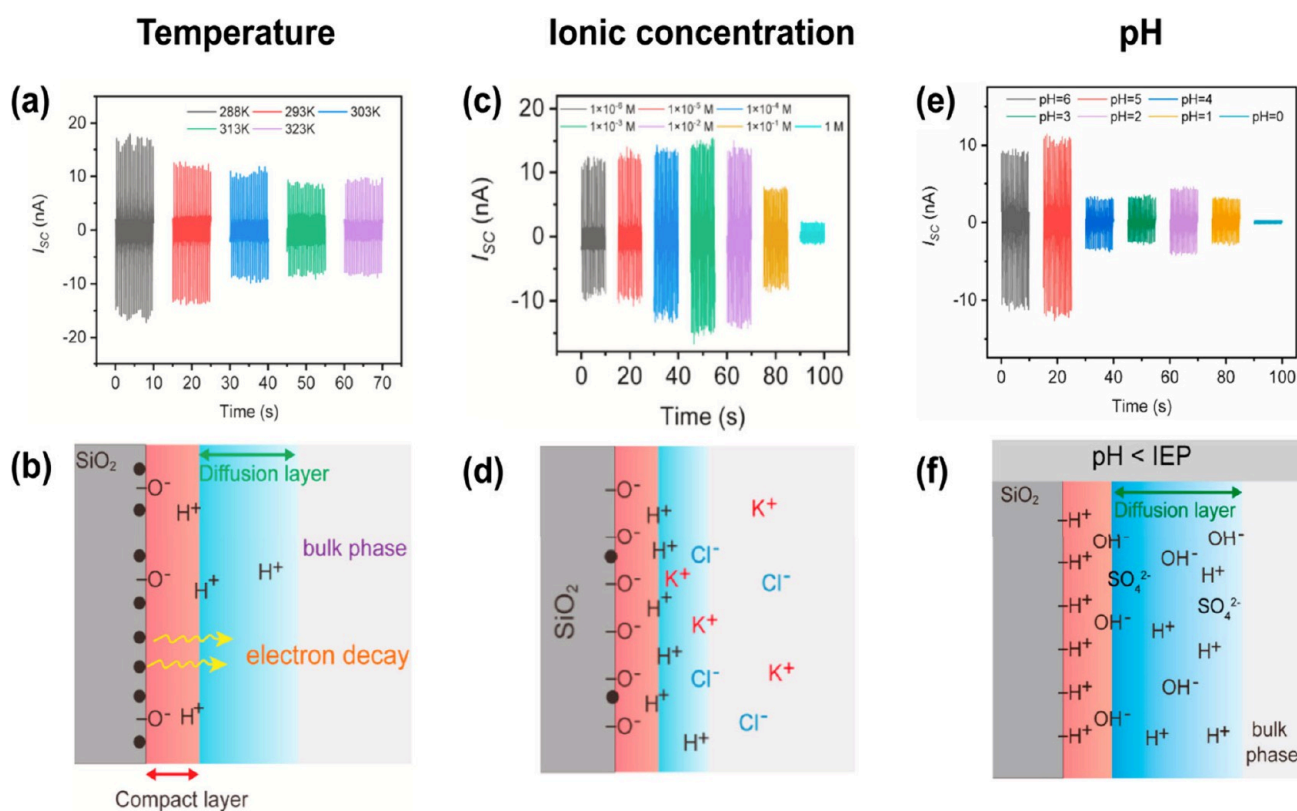


Figure 7. Liquid modification factors to enhance the SLCE-TENG performance. (a) Output change according to the solid–liquid interface temperature. (b) Schematic image of charge transfer at the solid–liquid interface when the temperature is 323 K. (c) Output change according to the ionic concentration. (d) Schematic image of charge transfer at the solid–liquid interface when the ionic concentration is high. (e) Output change according to the pH. (f) Schematic image of charge transfer at the solid–liquid interface when the pH value is less than 4. Reproduced with permission from ref 48. Copyright 2020 Elsevier.

water permeates into the nanoclusters, hindering effective separation and reducing the electrical output. This highlighted the importance of an optimal NW length for maximizing the triboelectric performance of the SLCE-TENGs. The efficiency of SLCE-TENGs was also evaluated based on the effect of the surface composition. PP films were treated with APTES, OTS, and PFTS, as shown in Figure 6c. Both untreated and surface-functionalized PP films were tested by interacting with DI water to facilitate contact electrification. The results, depicted in Figure 6d, showed a clear performance ranking: APTES–PP < bare PP < OTS–PP < PFTS–PP. The PFTS-coated PP film exhibited the highest short-circuit current and open-circuit voltage compared to the others. The superior performance of the PP substrate was mainly attributed to the incorporation of fluorine atoms, and this incorporation significantly enhanced the triboelectric polarity contrast between the contacting surfaces, leveraging fluorine's exceptional electronegativity. Additionally, the fluorination process reduced the surface free energy, making the PP material more hydrophobic and less prone to water adhesion. Surface modifications could be used to improve the triboelectric properties of insulators, including changes in the surface morphology to increase the solid–liquid contact area and chemical treatments to enhance the triboelectric properties. These modifications resulted in a significant improvement in the performance of SLCE-TENGs. These advancements not only enhanced the triboelectric output but also demonstrated the potential for optimizing energy-harvesting capabilities through meticulous material and surface engineering.

For increasing the surface charge density of the solid, Wu and colleagues utilized a charge-injection method to propose the charge-trapping-based SLCE-TENG depicted in Figure 6e.⁵⁸ For a droplet SLCE-TENG with a homogeneous electrowetting-assisted charge-injection (h-EWCI) process, the solid surface trapped more stable charges and the charge density does not degrade even in the water vapor environment. By utilizing the h-EWCI process, the SLCE-TENG was able to significantly increase its surface charge density, reaching up to 1.8 mC m^{-2} . This method significantly enhances the electrical output, with the device demonstrating currents beyond 2 mA, a power density above 160 W m^{-2} , and an energy-harvesting efficiency of over 11% from continuously falling water droplets. Additionally, the device's longevity and reliability were demonstrated by the enduring stability of the internally generated charge.

3.4.2. Liquid Modification to Enhance the SLCE-TENG Performance. Zhang et al. investigated the influence key factors of liquids based on the hybrid EDL to enhance the triboelectric performance.⁴⁸ They defined three different properties that can influence liquid, which are temperature, ion concentration, and pH, and conducted how those factors influence at the solid–liquid interface to form the hybrid EDL and electrical output of SLCE-TENG. Parts a and b of Figure 7 demonstrate that the electrical output performance of SLCE-TENG decreases significantly as the temperature increases. This decrease in the triboelectric output was due to increased electron dissipation at higher temperatures. At higher temperatures, electrons transferred due to contact electrification are

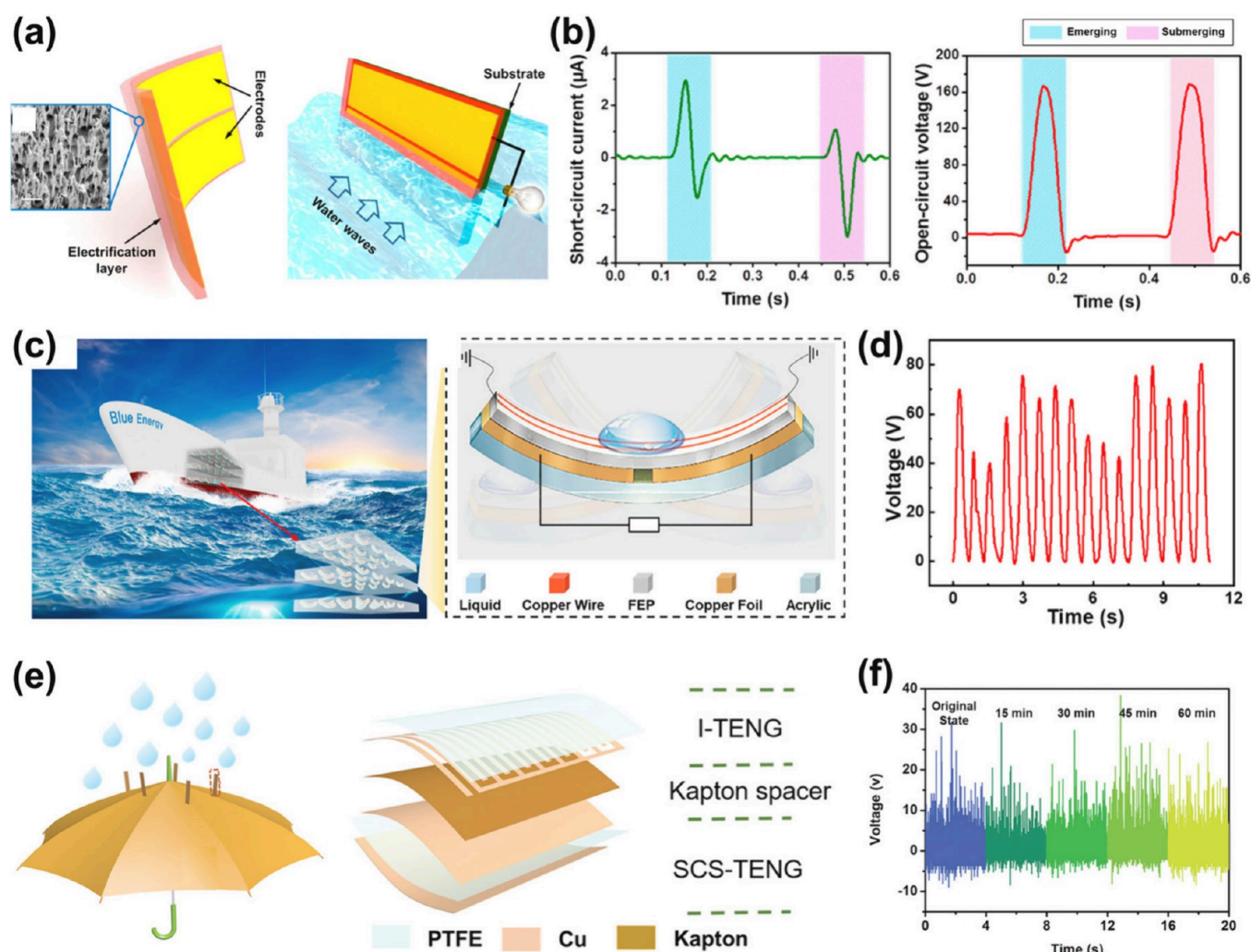


Figure 8. SLCE-TENG for applicable energy harvesting. (a) Schematic image of the electrode layer and electrification layer, SEM image of the polymer NWs on the triboelectric surface (tilted 30 degrees), and a schematic image that harvests energy as the wave contacts the electrode surface of SLCE-TENG. (b) Electric output at a velocity of 0.5 m s^{-1} . Reproduced from ref 60. Copyright 2014 American Chemical Society. (c) Structural design of TENG on the ship. (d) Performance of TENG in simulated ocean waves (open-circuit voltage). Reproduced from ref 61. Copyright 2021 American Chemical Society. (e) Schematic of a TENG-integrated umbrella. (f) Output and stability by water drop energy. Reproduced with permission from ref 62. Copyright 2019 John Wiley and Sons.

more likely to dissipate into the environment, while ions remain on the surface. Thus, they concluded that the reduction in the stern layer of the hybrid EDL and the output of the SLCE-TENG were due to an overall attenuation in induced charge. They also suggested that the ionic concentration also affects the electrical output of SLCE-TENG. They first used different concentrations of KCl as a liquid ionic material to verify contact electrification at the solid–liquid interface and showed an optimal peak within the increasing ionic concentration range, specifically from 10^{-3} to 1 M, as depicted in Figure 7c. The ion concentration indicated a balance between enhancing and diminishing the triboelectric output due to ion concentration changes. At low concentrations, ions behaved more like a conductor, promoting electron transfer across the interface, which is beneficial for the triboelectric performance. However, as the concentration increased, the solution's conductivity increased, leading to greater electron dissipation of triboelectricity and ultimately reducing the triboelectric output performance, as shown in Figure 7d. They suggested a delicate balance in the ion concentration to maximize the triboelectric output performance, highlighting

the critical role of ions in modulating the efficiency of SLCE-TENG. Confirming that the KCl ion concentration affected the SLCE-TENG performance, Zhang and colleagues also compared different ion salts, including NaCl, CuCl_2 , and AlCl_3 , and found similar trends in the electrical output across these solutions, indicating a generalizable effect of the ion concentration on the SLCE-TENG performance.

Verifying that the temperature and ion concentration affected SLCE-TENG, Zhang et al. also investigated the relationship between the liquid pH value and the triboelectric output performance at the solid–liquid interface. In Figure 7e, they proved that the pH value affects the triboelectric output performance, which alters the composition and polarity of adsorbed ions on the surface (Figure 7f). They found that the triboelectric output exhibits two peaks, one before and one after the isoelectric point (IEP), due to pH-induced changes. The presence of hydrogen ions (H^+) in acidic solutions and hydroxide ions (OH^-) in alkaline solutions altered the surface state, which affected the charge elimination behavior. The IEP played a crucial role in the charge elimination process. The charge polarity could be reversed depending on whether the

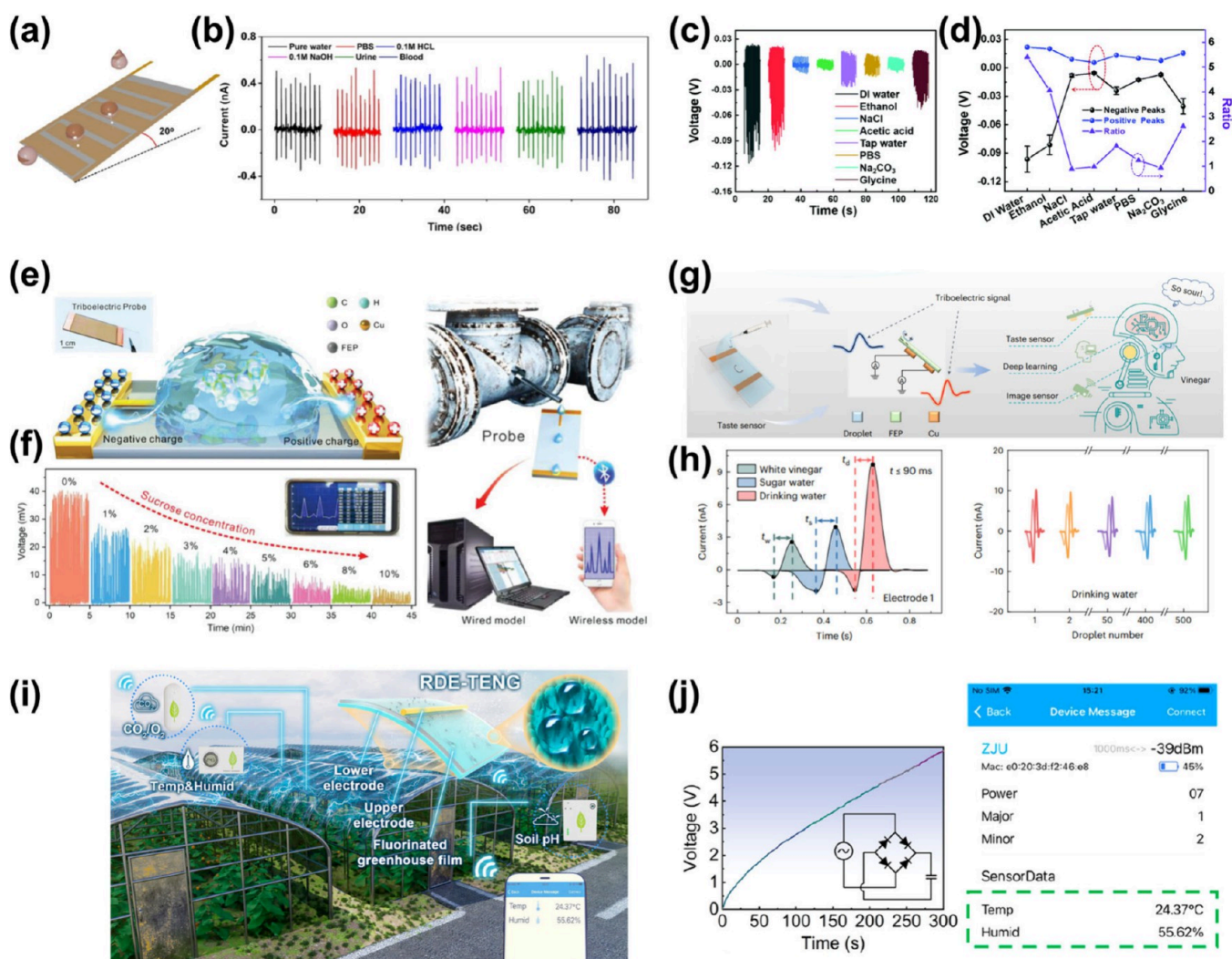


Figure 9. Solid–liquid interfaces applications for self-powered sensors. (a) Schematic image of droplet superhydrophobic SLCE-TENG as a biomedical sensor. (b) Current generated by six types of droplets over the SLCE-TENG (20° tilted angle). Reproduced from ref 64. Copyright 2020 American Chemical Society. (c) Voltage responses of the liquid sensor with droplets of different analytes (except water, other analytes were prepared with a concentration of 0.1 mol L⁻¹ in DI water). (d) Positive and negative voltage peaks and the ratio between the positive and negative peak values for different analytes. Reproduced with permission from ref 65. Copyright 2021 Royal Society of Chemistry. (e) Illustration of a SLCE-TENG-based sucrose monitoring sensor and schematic applied to an intelligent factory. (f) Output change according to the sucrose concentration and mobile phone screenshot in wireless mode. Reproduced with permission from ref 66. Copyright 2023 John Wiley and Sons. (g) Schematic of liquid droplet SLCE-TENG-based self-powered sensors. (h) Changes in the output and response time according to three types of liquid and output changes when contacted with hundreds of water droplets as if drinking water. Reproduced with permission from ref 67. Copyright 2023 Springer Nature. Corrosion protection through wave energy conversion. (i) Illustration of a TENG-based self-powered intelligent greenhouse. (j) Charging a 10 μF capacitor with TENG and screenshots confirming the operation of SLCE-TENG-based sensors. Reproduced from ref 68. Copyright 2021 American Chemical Society.

pH value was above or below the IEP. The reversal of triboelectric generation was caused by the adsorption of different ions. OH⁻ were responsible for the reversal at pH levels above the IEP, while H⁺ caused the reversal at pH levels below the IEP. The significance of considering the chemical properties of materials used in SLCE-TENGs to enhance their energy-harvesting performance is highlighted by this change in the surface charge density.

4. ADVANCED SLCE-TENG APPLICATIONS

Currently, the SLCE-TENG produces a high voltage but low current output, which limits its practical application. However, recent advancements in output enhancement techniques have increased their output efficiency, making them suitable for

practical applications such as energy-harvesting applications, self-powered sensors, and contact electrocatalysis. These advancements have broadened the range of potential applications in various fields. This section examines the specific applications where solid–liquid triboelectric energy-harvesting technologies have had a significant impact.

4.1. Energy Harvesting. Blue energy can harness the ocean's mechanical energy, providing a sustainable alternative to conventional sources and potentially reducing pollution. With oceans covering over 70% of Earth, there are vast opportunities for renewable energy harvesting. SLCE-TENG is a highly applicable energy-harvesting application that can produce electric energy simply by encountering solids and liquids.⁵⁹ Additionally, the output can be optimized by controlling the solid roughness and functionalizing the surface.

In Figure 8a, Zhu et al. presented an SLCE-TENG that operates through liquid wave motion and the impact of falling liquid droplets. They fabricated SLCE-TENG with FEP film parallel strip electrodes separated by a slender gap.⁶⁰ The nanoscale-patterned opposite FEP layer aimed to increase the surface roughness, enhancing its interaction with water for optimal triboelectric charge generation. The interaction between the solid and liquid phases was significantly improved by the NWs. The NWs, which have dimensions of approximately 100 nm in diameter and 2 μm in length, were designed to increase the contact area with water. The efficiency rate of this generator was 7.7%, achieved by increasing the contact surface area between the liquid and solid phases through the strategic modification of the surface with polymer NWs. The electrical output of the SLCE-TENG was observed when tap water was used as the medium and moved repetitively against the TENG at a speed of 0.5 m s^{-1} . This generated a short-circuit current of 3 μA and an open-circuit voltage of 160 V (Figure 8b). They demonstrated that these SLCE-TENGs can effectively harvest energy in rain-impacted and coastal environments, making them broadly applicable.

SLCE-TENGs are considered to be a stable energy-harvesting method of ocean energy due to their excellent energy harnessing capabilities at low frequencies, making them ideal for ocean energy. Wei et al. integrated a SLCE-TENG within a ship's structure to harvest ocean energy derived from the motion of ships, as shown in Figure 8c.⁶¹ The SLCE-TENG was constructed using FEP as the triboelectric layer and copper as the electrode material. To improve the energy-transfer output, a thin copper wire was placed on the triboelectric surface to ground the droplet continuously. The performance of the SLCE-TENG was simulated by analyzing the results of wave simulations depicted in Figure 8d. An open-circuit voltage of 70 V was achieved, indicating significant potential for large-scale ocean energy harvesting. The potential for energy harvesting extends beyond the ocean to include raindrops. Liu et al. demonstrated the use of an umbrella paired with the SLCE-TENG to harvest energy from both the kinetic energy of the raindrops and the energy generated through contact electrification between the water and PTFE.⁶² Due to PTFE's hydrophobicity, water droplets naturally flowed over its surface, preventing pooling and allowing continuous power. To maintain the stability of the electric output, water seepage into the SLCE-TENG inner layer was prevented. As shown in Figure 8f, this resulted in no decrease in the voltage output and high stability.

4.2. Self-Powered Sensors. The SLCE-TENG has the potential for biomedical applications through interaction with biofluids. It has recently been established that contact electrification at the solid–liquid interfaces can be used to detect a wide range of ions in water, enabling the detection of organic and inorganic chemicals.⁶³ With a superhydrophobic solid interface, triboelectric charges can repel blood, urine, and water, increasing their utility in healthcare with applicable self-powered sensors. In Figure 9a, Hu et al. presented a SLCE-TENG-based sensor that combines flexibility, self-cleaning ability, and adhesion, with a demonstrated superhydrophobicity.⁶⁴ The sensor was composed of an interdigitated copper foil electrode and a hydrophobic fumed silica nanoparticle layer for its superhydrophobic surface. In Figure 9b, Hu and colleagues fabricated SLCE-TENG-based self-powered sensors to detect six different liquid types: pure water, phosphate-buffered saline (PBS), 0.1 M HCl, 0.1 M NaCl, urine, and

blood. The electrical pulse generation highlighted the sensor's efficacy and potential for optimization. They also showed that biofluids can interact with TENGs. The rapid transit of droplets across the superhydrophobic surface of the TENG facilitated quick sensor response times. Through these results, Hu and colleagues suggested that its rapid response time holds promise for accurately monitoring clinical procedures including drainage and fluid transfusion.

The detection of liquids is necessary in various fields including chemistry, medicine, and the environment. Figure 9c depicted another SLCE-TENG-based self-powered triboelectric sensor that can detect liquid chemicals.⁶⁵ When a droplet passed the electrode on the surface of the SLCE-TENG, the EDL charge generated and the SLCE-TENG surface-induced charge interact. Ying and his researchers demonstrated the sensor's capability to detect different liquids by analyzing the positive and negative voltage spikes and ratios resulting from SLCE interactions. To show the sensor's ability to detect various liquids, output checks were shown with DI water, ethanol, NaCl, acetic acid, tap water, PBS, Na_2CO_3 , and glycine. The negative and positive peaks as well as the ratio between them reflected the chemical and dielectric properties of the droplets being analyzed. Figure 9d illustrates that the corresponding pattern can be used to identify the type of liquid.

SLCE-TENG-based self-powered sensors can also be useful for real-time monitoring of sucrose solutions via SLCE. Sucrose, a significant component in both the food and chemical industries, interacted with a SLCE-TENG comprising a FEP film and a copper electrode, as described in Figure 9e.⁶⁶ Upon contact between the solution and FEP, water molecule electrons tended to migrate toward the FEP surface. This interaction was characterized by a competitive dynamic between ion adsorption and electron transfer, influencing the SLCE process. Self-powered sensors could be integrated within fluid transport, as shown in Figure 9f. The concentration of sucrose can be wirelessly detected in real time through the generated triboelectricity. Liu et al. reported that the sensor could demonstrate remarkable linearity (0.966) and sensitivity in detecting the sucrose concentration, along with a swift response time of 90 ms, making it a high-performance sucrose sensor suitable for industrial use in the future.

Wei et al. proposed a self-powered droplet-tasting sensor-system-based SLCE.⁶⁷ This sensor system achieved an accuracy of over 90% in liquid recognition by employing triboelectric signals combined with deep-learning techniques. The accuracy was further improved to 96% with the integration of an image sensor. The taste system, as shown in Figure 9g, was created to imitate the multisensory interactions that are present in the human taste system. It consisted of a SLCE-TENG that functioned through triboelectric signals, eliminating the need for an external power source. This configuration enabled the extraction of liquid characteristics based on triboelectric signals. The SLCE-TENG exhibited an immediate response to various liquids, such as water, sugar water, and white vinegar, regardless of their viscosity. Furthermore, based on SLCE, Figure 9g indicates that the system maintained high detection stability and achieved a consistent performance even after the application of 500 droplets. This technology could offer a promising method for distinguishing and monitoring liquids, making it suitable for a wide range of industrial applications.

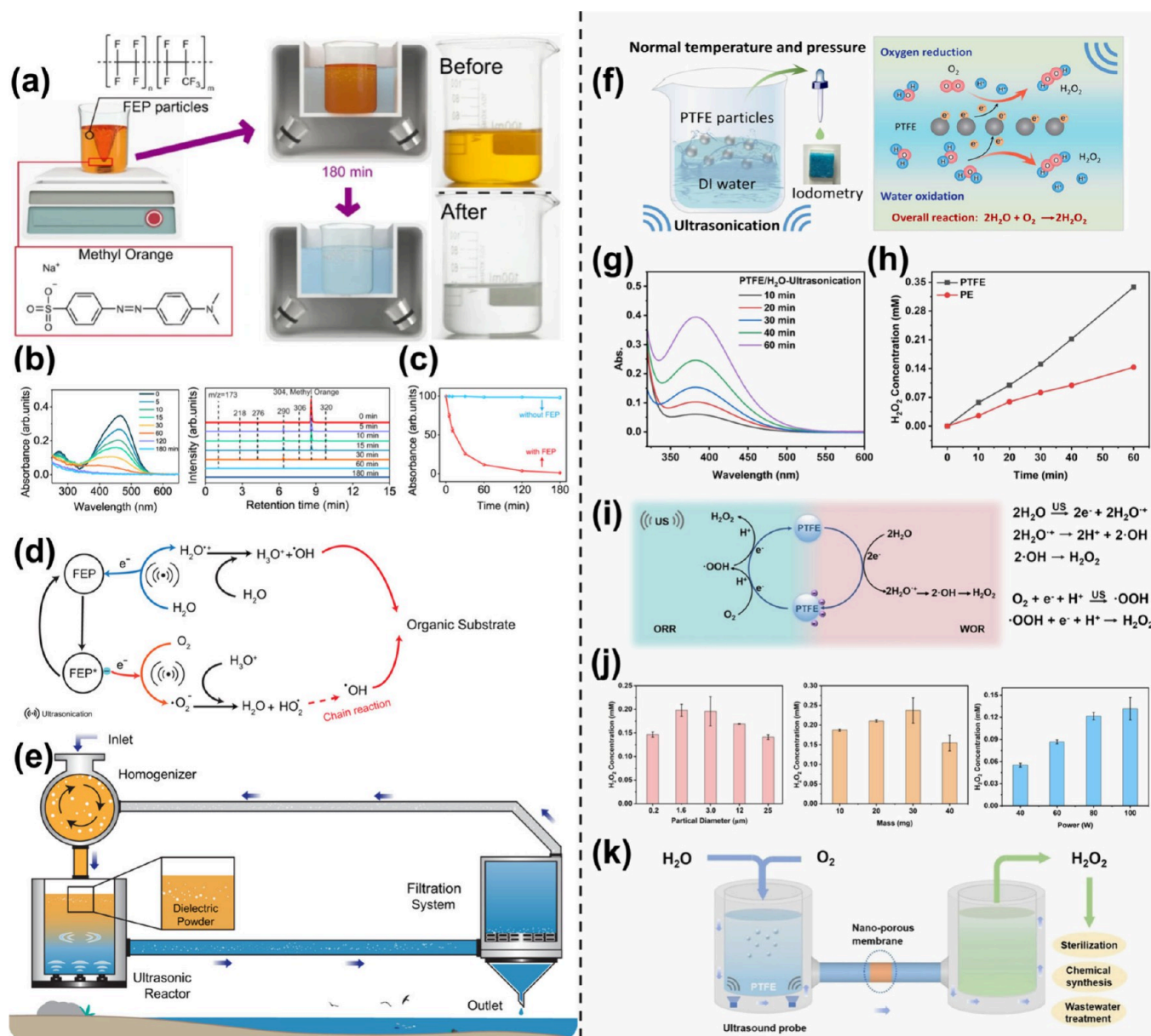


Figure 10. SLCE applications for contact electrocatalysis. (a) Experimental setup and overall process. (b) Changes in the UV–vis and liquid chromatography–mass spectroscopy spectra of the MO solution over time. (c) Comparison of the MO solution absorbance with and without FEP powder. (d) MO decomposition mechanism by radicals. (e) Expected application to large-scale organic wastewater treatment. Reproduced from ref 24. Copyright 2022 American Chemical Society. Reproduced from ref 25. Available under a CC-BY 4.0 license. Copyright 2022 Wang, Z. et al. (f) Schematic of the setup and overall reaction. (g) Absorbance difference according to the ultrasonic time. (h) Comparison of the H₂O₂ concentration between PTFE and PE particles. (i) Mechanism of H₂O₂ generation by contact electrocatalysis. (j) Comparison of H₂O₂ generation according to particle and ultrasonic cleaner conditions. (k) Expected application to the efficient production of H₂O₂ technology. Reproduced with permission from ref 69. Copyright 2023 John Wiley and Sons.

Raindrops can operate as self-powered sensors to monitor and modulate temperature and humidity levels in agricultural greenhouses. Zhang et al. presented a novel greenhouse design that combines a greenhouse with a hydrophobic film-based SLCE-TENG, as shown in Figure 9i.⁶⁸ This system harvested energy from raindrops to operate the temperature and humidity sensors. Recognizing the crucial importance of light permeability in greenhouses, they selected a polyethylene (PE) membrane, which underwent inductively coupled plasma etching to fabricate the SLCE-TENG. The triboelectric charges generated by SLCE between the top and bottom electrodes and the film thus eliminate the interfacial screening

phenomenon and increase the efficiency of the triboelectric charge transfer. Figure 9j shows that water droplets applied at a frequency of 2 Hz from a height of 15 cm for 3 min electrified a 10 μF capacitor to approximately 6 V. This result confirmed the reliability and durability of the energy generated by the SLCE-TENG, which is sufficient to power small electronic sensors. Zhang et al. adjusted self-powered sensors to propose a foundation for an intelligent greenhouse system that can autonomously regulate and monitor the temperature and humidity.

4.3. Contact Electrocatalysis. Recent advancements at the solid–liquid interface led to contact electrocatalysis as a

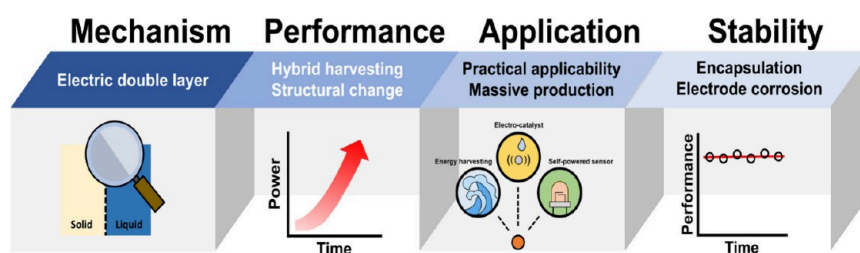


Figure 11. Perspective of the development of SLCE.

promising application. Contact electrocatalysis was based on SLCE and acted as a catalyst by moving charges through forces acting on powdered solids. Electrons exchanged at the solid–liquid interface can be utilized through the contact electrocatalysis process. As demonstrated in Figure 10a, ultrasonic treatment facilitated electron exchange at the interface between FEP powder and water, leading to degradation of an aqueous methyl orange (MO) solution.²⁵ The effects of the sonication time on the degradation process are presented in Figure 10b, showcasing the results from UV–vis and liquid chromatography–mass spectroscopy analyses. Wang et al. observed that both the absorbance peak and the mass-to-charge ratios indicative of MO diminish with increasing sonication time. As shown in Figure 10c, further analysis was conducted to compare the MO absorbance in the presence and absence of FEP powder. The results indicated that the pivotal role of FEP powder in MO degradation is underscored by the noticeable reduction in absorbance observed in its absence. Figure 10d elucidates the mechanism through which FEP facilitates the degradation of organic substances. Contact between water and FEP led to the formation of water radical cations via electron transfer. This process also prompted the migration of protons, resulting in the generation of hydronium cations and hydroxyl radicals. Concurrently, FEP, enriched with electrons, interacted with oxygen to produce O_2^- radicals, which, in turn, generated hydroxyl radicals. These radicals engaged with organic compounds in the solution, facilitating their breakdown. Wang and his research team reported that FEP powder is expected to be used commercially because its MO decomposition performance does not deteriorate even after recycling. With these results, they expected that FEP powder can be a promising prospect for its application in organic wastewater treatment, as depicted in Figure 10e. This innovation not only highlighted a novel approach to addressing organic pollutants but also underscored the potential for reducing reliance on traditional catalysts in environmental remediation efforts.

Hydrogen peroxide (H_2O_2) served as a crucial component across various industries, notably within the environmental and chemical sectors. Traditional production methods for H_2O_2 necessitated the use of noble metals or high-entropy alloys, presenting significant cost challenges. To address these issues, Zhao et al. introduced a novel approach employing contact electrocatalysis between PTFE and water.⁶⁹ Figure 10f illustrates the electron exchange facilitated by inducing contact separation between PTFE and water via ultrasonic treatment. The increase in the H_2O_2 concentration as the sonication time increased can be seen in Figure 10g by using UV–vis spectroscopy. Furthermore, Figure 10h highlights the differential H_2O_2 concentrations observed in the presence versus absence of PTFE, with comparisons to PE. Notably, H_2O_2 concentrations were found to be higher in the presence of PTFE, attributing the enhanced H_2O_2 yield to PTFE's superior

electron attraction capabilities. The underlying mechanism of H_2O_2 production is detailed in Figure 10i. It was postulated that H_2O_2 formation occurs via two primary reactions: water oxidation and oxygen reduction. During the water oxidation reaction, the interaction of PTFE with water prompted the transfer of electrons from water molecules to the PTFE surface, resulting in the formation of water radical cations, hydronium cations, and hydroxyl radicals, ultimately producing H_2O_2 synthesis. In the oxygen reduction reaction, ultrasonically induced bubbles in the water facilitated the transfer of electrons from PTFE surface particles to oxygen, forming $\cdot\text{O}_2^-$ radicals. These radicals subsequently interacted with water ions to produce H_2O_2 . The optimization of H_2O_2 production relative to PTFE powder characteristics and sonication conditions is shown in Figure 10j. PTFE particles with diameters of 1.6 and 3.0 μm demonstrated optimal H_2O_2 production efficiencies of 0.221 and 0.215 $\text{mmol L}^{-1} \text{h}^{-1}$, respectively. A total of 30 mg of powder provided the most effectiveness for maximizing SLCE, thereby enhancing the H_2O_2 concentration. Also, it was observed that utilizing a sonication power of 100 W significantly improved the interface contact separation, yielding the highest H_2O_2 concentration. For economical H_2O_2 production based on contact electrocatalysis, it was possible to design an H_2O_2 production system, as shown in Figure 10k.

Contact electrocatalysis is a promising application for solid–liquid interfaces, especially in a large scale of liquids. Using mechanical vibration to initially produce a solid powder in catalysis, it presents a new candidate for preventing marine pollution by degrading various organic solvents. This method has the potential to eliminate pathogens in water through reactive oxygen species reactions, making it useful in water purification and desalination plants.

5. CONCLUSIONS AND PERSPECTIVES

In this Spotlight on Applications, we aim to introduce the SLCE with fundamental studies of its applications. We began with the traditional EDL to hybrid EDL to understand the SLCE. In solid interfaces, electron transfer first exists to form the stern layer with ions, and it is identified using diversified experiments. Then we introduced the SLCE-TENG with two different modes to utilize the SLCE. We further explored the influence of the SLCE with solid types and organized the liquid triboelectric series. Then we delved into the key factors to improve the triboelectric properties in each phase. Finally, we probed the advances in technology of the SLCE from energy harvesting to contact electrocatalysis. Despite significant advancements in the SLCE, there are still several challenges that need to be addressed before widespread adoption can occur. The four challenges are summarized in Figure 11.

5.1. Deeper Understanding of the Charge Dynamics of SLCE. The traditional EDL model is limited by its focus on

ions. The emergent hybrid EDL model, which integrates electron-driven charge transfer, remains underexplored, particularly in the later stages of EDL formation. Rigorous experimental and computational studies are warranted to elucidate the role of electrons in charge dynamics at interfaces given their sensitivity to environmental factors.

5.2. Optimizing the Performance of TENGs. SLCE-TENGs, while promising for energy harvesting, currently operate at microwatt power levels, which hinders their wider application. Additionally, the output performance of the SLCE-TENG is markedly impacted by ambient humidity conditions. An investigation of the output performance of SLCE-TENG under conditions of high humidity could provide valuable insights into its potential applications.⁷⁰ Intensive research into materials engineering and integration with other energy conversion methods, such as piezoelectric, electromagnetic, or other energy-harvesting technologies, is essential to increase their energy output, particularly to improve the current output.

5.3. Diversifying Applications. SLCE-TENGs have found applications in blue energy harvesting and raindrop energy conversion with growing interest in contact electrocatalysis. Groundbreaking research is essential to extend their utility to biologically relevant systems, such as harnessing energy from blood flow, thereby broadening the scope of SLCE applications.

5.4. Long-Term Operation Stability. The longevity of SLCE-TENGs is paramount, especially in scenarios such as contact electrocatalysis, where the solid substrate must resist liquid-mediated degradation. Investigation of protection strategies, including advanced encapsulation technologies, is essential to maintaining the integrity and functionality of these applications over time.

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Notes

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