Triboelectric nanogenerators as a probe for studying charge transfer at liquid–solid interface

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Contact electrification and charge transfer between liquid and solid interfaces are of the most importance for chemistry, energy, catalysis, and even biology. However, the origin of the charges generated and transferred at liquid–solid interfaces remains to be investigated. Recently, triboelectric nanogenerators have emerged as a promising probe for studying charge transfer at liquid–solid interfaces. When a liquid drop moves on a hydrophobic surface, the electric signal generated on electrodes located under the dielectric solid film provides a chance to probe the charge transfer between liquid and solid surfaces. Here, the most recent advances toward a deeper understanding of the charge generated, accumulated, and transferred at liquid–solid interfaces by TENG, as well as the probes design and physical/chemical effects are discussed. The sensing systems and energy conversions enabled by charge transfer at the liquid–solid interface are also included.

Introduction

When two materials come into contact with each other and then separate, both become statically charged.[1–3] This phenomenon, termed as contact electrification (CE), also known as triboelectricity or static electricity, is an old subject, with the earliest written records dating from 600 BC.[4,5] Most research on CE or triboelectricity focuses on solid–solid interfaces and solid–solid CE has been widely studied by various methods owing to its importance in engineering.[6–9] Although shear forces and shear gradients are a big contributor to the electrification of an insulating object,[8,10–12] CE can occur not only between solids but also at liquid–solid interfaces.[13–17] Research on liquid–solid CE, especially applications related to energy harvesting[18–20] and sensing technology,[21–23] is currently attracting more and more attention from various science and technology disciplines. However, the understanding of liquid–solid CE is rather limited due to the lack of fundamental understanding of interfacial charge transfer.

The triboelectric nanogenerator (TENG), based on the CE between liquid droplet and solid, is widely used in energy harvesting, environmental monitoring, and biomedical sensors. TENG usually contains three layers: the top layer is the dielectric solid film for CE with the liquid droplet; the second layer is a metal electrode for electrostatic induction; and the bottom layer is a solid plate for support.[24] When a water droplet slides on the dielectric surface with the metal electrode located under it, the surface-induced charges on the dielectric surface can be measured by an electrometer connected with the metal electrode at the same time. As liquid–solid CE is highly sensitive to the physical and chemical properties of liquid, it can help study the basic foundations of liquid–solid CE.[25–28] In the year 2020, Wang's group measured the charge transfer at the liquid–solid interface with the TENG and first proposed the concept of the TENG "probe."[24,29] Such a unique probe opens a promising application direction for energy harvesting and sensing systems.

This article provides a summary of recent studies on the TENG probe. First, the fundamental charge-transfer mechanisms at liquid–solid interfaces, including charge carriers, charge transfer, and Wang's hybrid EDL model, are introduced. Second, factors affecting charge transfer at the liquid–solid interface, for example, ambient factors, droplet

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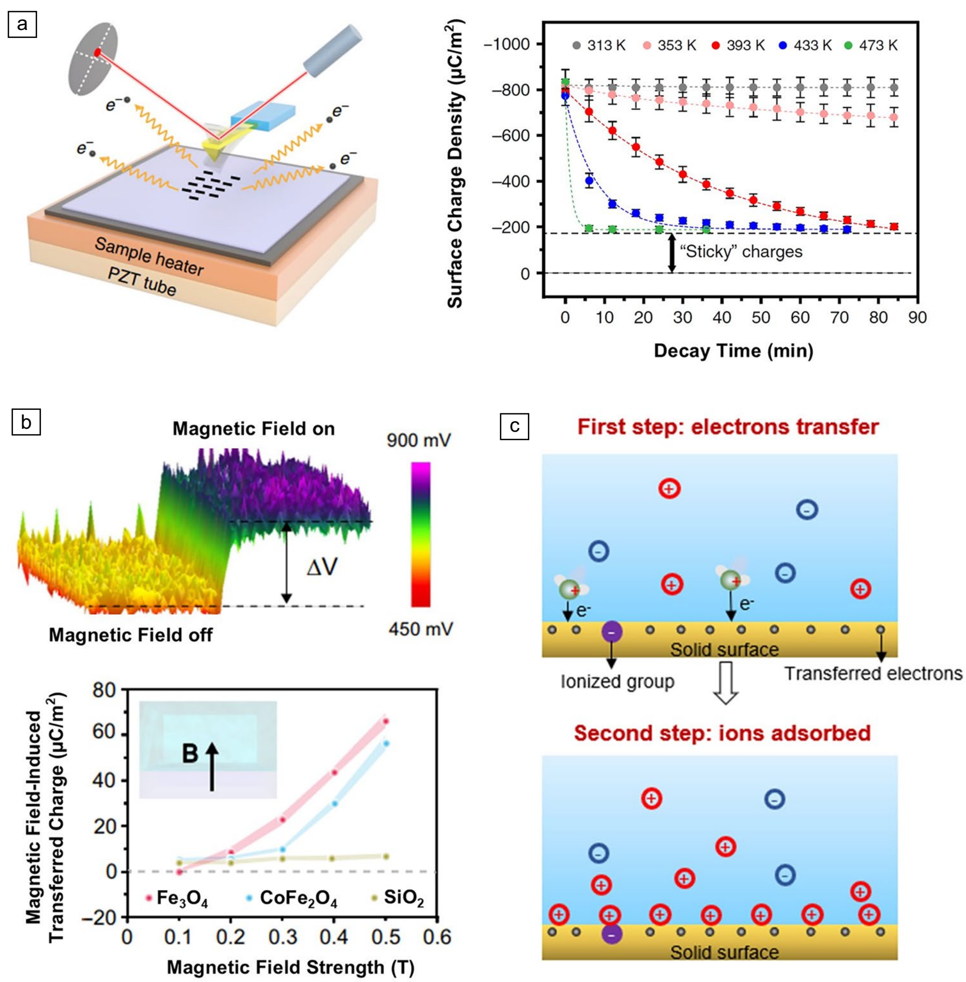

Figure 1. Charge carriers and electric double-layer formation at a liquid–solid interface. (a) Quantitative analysis of electron and ion transfer at liquid–solid interface using thermal electron emission method. After heating a statically charged sample, electrons generated by liquid–solid CE are removed, while ions remain.[30] Reprinted with permission from Reference 30. © 2020 Nature Publishing Group. (b) The surface potential of the Fe_3O_4 in the de-ionized water. ΔV denotes the change of surface potential.[31] Reprinted with permission from Reference 31. © 2022 Nature Publishing Group. (c) "Two-step" process for the formation of electric double layer, which is also called Wang's model.

velocity, liquid chemistry, and magnetic fields are discussed in detail. Third, interface and structure design of the TENG probe are highlighted and further discussed. Finally, the applications of CE at the liquid–solid interface using the TENG as energy harvesters and sensors are systematically summarized.

Charge-transfer mechanisms at liquid–solid interfaces and the hybrid electric double-layer model

Due to the explosion of water droplet energy harvesting,[15,16] research on the CE mechanism at the liquid–solid interface is urgent. In 2020, Wang's group used the thermal electron emission method to study liquid–solid CE,[30] achieving quantitative analysis of electron transfer and ion transfer at the liquid–solid interface. As shown in **Figure 1**a, after heating a statically charged sample at 300–500 K, part of the charges (electrons) generated by liquid–solid CE were removed, while some stubborn charges (ions) remained, proving that both electron transfer and ion adsorption coexist in liquid–solid CE. Moreover, Wang's group explored the response of charge transfer at the liquid–solid interface to magnetic fields in 2022,[31] revealing that the magnetic field at the liquid–solid interface can promote charge transfer at the liquid–solid interface. Based on the difference in spin properties of electrons and ions, the authors prove the existence of electron transfer at the liquid–solid CE (Figure 1b). As previously stated, electron transfer has a strong effect during the CE between the liquid and the solid and thus, electron transfer should also be considered in the formation of the electric double layer (EDL). A hybrid EDL model was first proposed by Wang in 2019 in Figure 1c, which is, the overlap of the electron clouds of the solid atoms and water molecules leads to the electron transfer between them first, and after that free ions in the liquid will transfer to the charged solid surface because of the electrostatic interactions. In this way, an EDL formed.[32] The fundamental principle for liquid–solid TENG to be a power generator or sensor is likely due to the dynamic electric double layer. The dynamic EDL has twofold of meanings: first, the charge carrier composition made of electrons and ions varies at the liquid–solid interface at the initial contact due to the movement of the liquid level and unstable ion diffusion/transport at the vicinity of the surface. This means that the potential profile at the liquid–solid interface could be time-dependent. Second, the covering area of the liquid onto the solid surface varies with the movement of the liquid and experimental condition.

Effects of ambient factors on charge transfer at liquid–solid interface

The environment, for example, temperature, humidity and light, should affect the charge transfer at liquid–solid interfaces. The environmental humidity not only causes surface charge dissipation, but also affects the triboelectrification effect.[33–36] High temperature usually decreases the charge transfer because of the dissipation of triboelectric charges through the thermionic emission.[37] For example, when the temperature rises from 288 K to 323 K, the current generated by water-SiO_2 CE decreases from 17 nA to 9 nA.[38] The strong light intensity will accelerate the charge dissipation and thus will reduce the charge transfer at the liquid–solid interface.[39–41] After CE with poly(methyl methacrylate) (PMMA), the static charge decay behavior on the SiO_2 surface under the irradiation of the xenon light was studied in detail.[39] After 45 min of xenon light irradiation, the static charge density decayed from -160 μC m^{-2} to -10 μC m^{-2} on the solid SiO_2, indicating that the light irradiation should be an effective way to decrease surface triboelectric charges. However, the light effect on the transferred charge dynamic is still unclear. In 2023, Wang's group used a pixeled TENG as a probe to *in situ* study the relationship between light intensity and charge-transfer mapping at a liquid–solid interface. When a water droplet slides on a fluorinated ethylene propylene (FEP) film under different light intensities, it is noted that the charges transfer between the water and the y-axis decreased of light intensity. As the light intensity increased from 0 W/m^2 to 3500 W/m^2, the charge transfer along the *y*-axis decreased from 0.013 nC/mm^{-1} to 0.002 nC/mm^{-1}.[42] The result is consistent with the photoelectron emission theory, suggesting that the electrons should be the charge carriers at liquid–solid CE.

Effects of droplet velocity on charge transfer at liquid–solid interface

In recent years, there have been numerous studies on harvesting water energy by liquid–solid CE. However, the sliding velocity of droplets are rarely considered. In 2023, Wang's group used a TENG as a probe to measure the transferred charges at different sliding velocities of moving droplets. The results show that the transferred charges generated by the liquid–solid CE is related to the droplet sliding velocity: the faster the sliding velocity, the higher the transferred charges detected (**Figure 2**a).[43] The mechanism is likely due to the influence of droplet velocity on the amount of ions adsorbed. When the droplet slides at a slow velocity, more cations are adsorbed onto the FEP surface, leading to a stronger shielding effect and, consequently, a reduced charge transfer. On the other hand, a faster droplet sliding velocity results in the loss of adsorbed cations and an increased charge transfer. This work presents a physical method to maximize contact charging at the liquid–solid interface, offering a new concept for velocity sensing.

Effects of liquid chemistry on charge transfer at liquid–solid interface

Charge transfer at liquid–solid interfaces inevitably involves ion adsorption and the formation of the EDL. Most important, the charge transfer measured by an electrometer is a net charge, which results from both the electron transfer and ion adsorption.[44] Thus, the chemical information of liquid, for example, ion migration kinetics should be considered when involved CE between liquid and solid. In recent years, researchers have started to use a TENG as a probe to study the effect of liquid chemistry, such as ions type, pH and ion concentrations on charge transfer at a liquid–solid interface. Most of the works indicate that when the concentration of ions in the liquid droplet increases, the charge transfer will first increase and then decrease. The reason for charge transfer depend on the ion concentration should be ascribed to the enhancement of the ion transfer process during the CE, while the charge transfer reduced at higher ion concentration may because of the screen effect of free ions. For salt solutions, such as Na^+ and Cu^{2+}, the maximum charge transfer corresponds to an ion concentration of approximately 1×10^{-5} mol L^{-1}.[29] Moreover, Wang's group revealed that the charge accumulation kinetic on a solid surface is closely depending on the liquid chemistry: a droplet with small cationic diameter and stronger ionic bonds between cations and anions, has a relatively fast charge accumulation rate (Figure 2b),[45] highlighting that liquid chemistry should be considered when energy harvesting involved CE between liquid and solid.

Effects of magnetic fields on charge transfer at liquid–solid interface

As stated above, electron transfer is a key step in the CE process at liquid–solid interfaces. Also, the electron transfer between two species is spin conservative and follows the Pauli exclusion principle. Therefore, the electron spin in magnetic field should have an influence on CE at liquid–solid interfaces. In 2022, Wang's group explored the charge transfer between different liquids and ferrimagnetic solids in a magnetic field by dual harmonic Kelvin probe force microscopy (DH-KPFM).[31] Three types of solids (Fe_3O_4, $CoFe_2O_4$ and SiO_2) were used to CE with DI water. As shown in Figure 2c, applying a magnetic field increased the charge transfer between Fe_3O_4-DI water (up to 70 μC m^{-2}) and $CoFe_2O_4$-DI water (up to 40 μC m^{-2}) interfaces in a 0.5 T magnetic field (Figure 2c). However, for SiO_2, such effect was not obvious, which can be ascribed to the small saturated magnetic moment, showing that the magnetic field effect on the liquid–solid charge transfer is depending on the magnetic moment strength of the solids. Moreover, magnetic fields were applied in different directions (upward, downward, rightward, and leftward) to the liquid–solid interfaces, indicating that the magnetic field effect on liquid–solid CE was not related to the direction of the magnetic field. The mechanism proposed by the authors is that due to the Zeeman interaction, the magnetic field can regulate

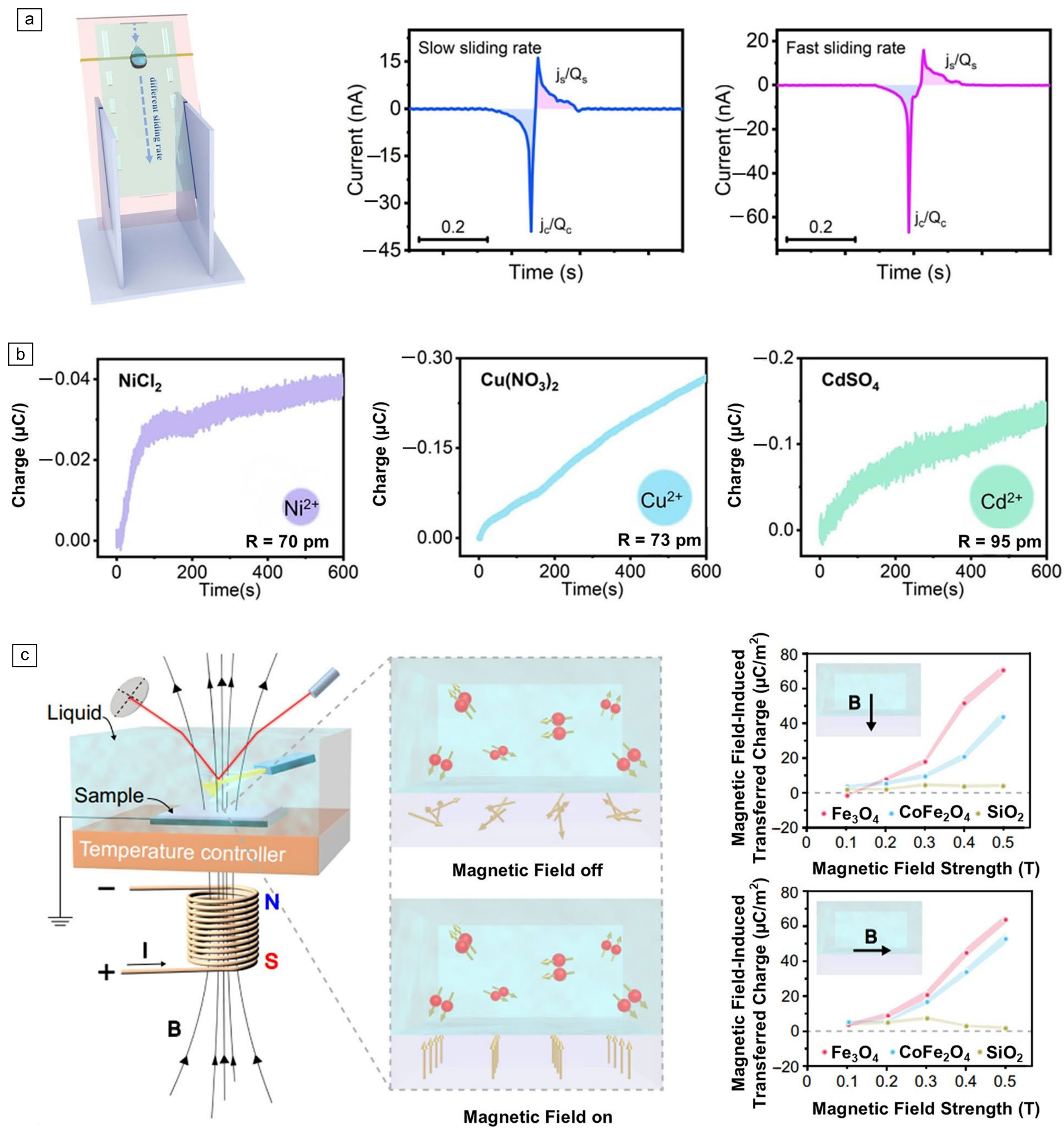

Figure 2. Factors affecting the charge transfer at the liquid–solid interface. (a) Droplet velocity effect: the faster the sliding speed, the higher the transferred charges detected.[43] Reprinted with permission from Reference 43. © 2023 Royal Society of Chemistry. (b) The charge accumulation kinetic on a solid surface is closely dependent on the liquid chemistry: a droplet with a small cationic diameter has a relatively fast charge accumulation rate on the solid surface.[45] Reprinted with permission from Reference 45. © 2023 Royal Society of Chemistry. (c) The magnetic field effects on the contact electrification between water and different solids in different directions. B represents the magnetic field with direction; I is the current.[31] Reprinted with permission from Reference 31. © 2022 Nature Publishing Group.

the spin conversion of radical pairs at the interface between O_2-containing liquid and ferrimagnetic solid and thus promote electron transfer. According to the radical pair mechanism, this work proposed a spin-selected electron transfer model for liquid–solid CE, providing a perspective for understanding magnetic field-controlled chemical reactions.

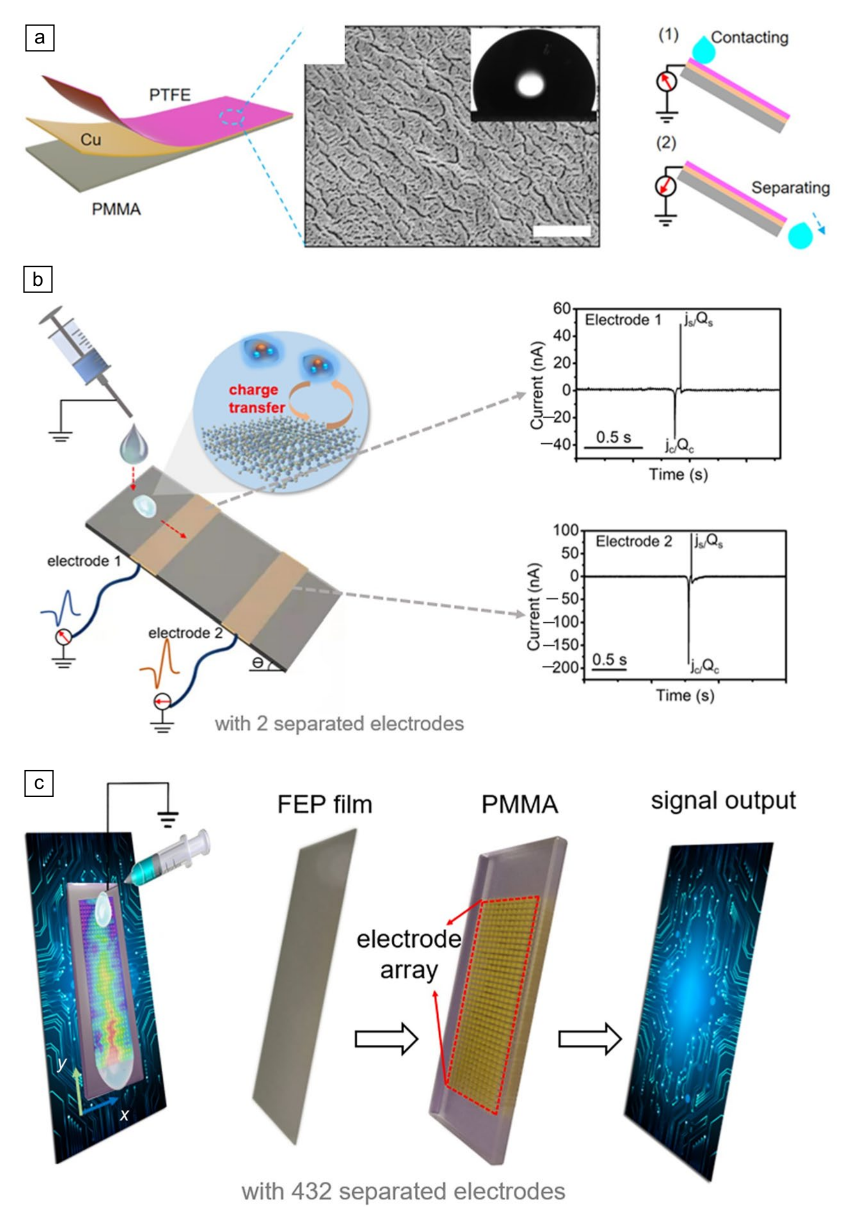

Figure 3. Tribielectric nanogenerator (TENG) probe: from one electrode to electrode array. (a) TENG probe with one electrode.[24] Reprinted with permission from Reference 24. © 2020 American Chemical Society. (b) TENG probe with two separated electrodes.[52] Reprinted with permission from Reference 52. © 2021 American Chemical Society. (c) TENG probe with high-density electrode array.[42] Reprinted with permission from Reference 42. © 2023 American Chemical Society.

Interface design affecting charge transfer at the liquid–solid interface

The static charge density on a dielectric solid surface is closely related to the surface's chemical properties. Modifying the surface chemical environment through appropriate functionalization is a fundamental approach for controlling the output performance of solid–solid TENGs.[46–48] For liquid–solid cases, it has been shown that the charge transfer still highly depends on the functional groups present on material surfaces, and the output performance of liquid–solid TENGs can also be optimized by modifying functional groups on the solid surfaces.[49] Further, modification of surface functional groups can alter the polarity of the triboelectric charges between the polymer and aqueous, and the electrical properties at different pH values will also change. An important question was raised from the identity of the charge carriers in the CE between liquids and the functional group-modified solid surfaces. Ions transfer or electrons transfer needs to be better understood. To address this question, Wang's group systematically studied the CE between SiO_2 with different functional groups and different liquids in 2020.[50] By studying the charge dissipation process when heating the statically charged surface SiO_2 at 413 K, electrons were dissipated, while the transferred ions keep. Therefore, according to the charge decay behavior on the solid, the amount of electron transfer and ion transfer at the liquid–solid interface can be distinguished. In addition, the electron transfer between DI water and SiO_2 is depending on the electron affinity of the functional group grafted onto the SiO_2 surface, while the electron transfer between the organic solution and SiO_2 has not shown related to the functional group, which should be ascribed to the limited ability of organic solutions to donate/gain electrons from SiO_2.

Structure design of TENG probe: From one electrode to array of electrodes

In 2014, a new prototype TENG was established by Wang's group to harvest water drop, which termed as droplet TENG.[51] Thereafter, in 2020, Wang's group first proposed the concept of the TENG probe.[24] When a liquid droplet contacts with and separates from the solid film, the electric signals will be detected by electrometer (**Figure 3**a) and by analyzing the electric signals generated by liquid–solid CE, the TENG could become a promising probe for studying the charge transfer at a liquid–solid interface. Then, Wang's group extended a single electrode to two electrodes in 2021 (Figure 3b).[52] In this work, when comparing the electric signals on two separated electrodes under solid film, the charge transfer at

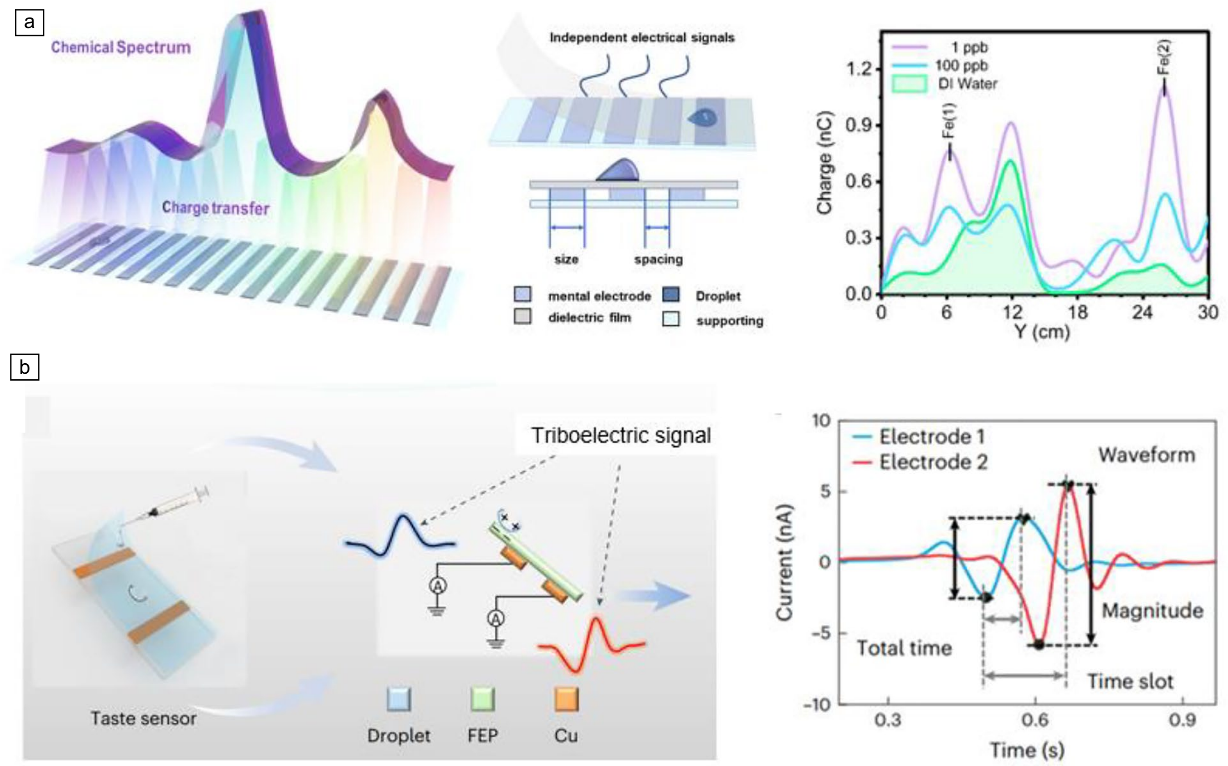

Figure 4. Sensing systems enabled by charge transfer at liquid–solid interface. (a) Triboelectric spectroscopy (TES) for the chemical analysis of liquid samples. When a liquid droplet with dissolved analytes moves across an insulating surface its local triboelectrification will be recorded by electrometer and used to build a TES profile.[53] Reprinted with permission from Reference 53. © 2024 American Chemical Society. (b) The intelligent taste-sensing system based on the two electrodes' TENG probe. The sensor system identifies the types of liquid food by analyzing the shape of the current curves generated on the two electrodes during liquid–solid triboelectric electrification.[54] Reprinted with permission from Reference 54. © 2023 Nature Publishing Group.

various sliding positions on the solid surface was studied. To improve the resolution and sensitivity of the TENG probe, a pixeled TENG with a high-density electrode array (432 separated Cu electrodes) was developed as a probe in 2023 (Figure 3c).[42] The electrical signal at each electrode enables mapping the high-resolution charge-transfer distribution in two dimensions as a water droplet moves over a solid surface at a spatial resolution of 400 µm. Such transferred charge mapping exhibits high sensitivity to the light intensity and pH values of liquid, showing promising applications in the field of sensing. Practically, the ability of such a TENG probe to map the charge transfer in this way is particularly convenient and fast compared with other techniques, such as AFM, and has implications for detection of surface properties of materials, chemical sensing, biomedical sensor, and would also help refine the description of forces acting on the moving droplet.

Sensing systems enabled by charge transfer at liquid–solid interface

Based on the arrayed TENGs, a new form of "electrostatic" spectroscopy was reported, called triboelectric spectroscopy (TES) for the chemical analysis of liquid samples in 2024.[53] Inspired by recent advances both in the fundamental understanding on the origin of static electricity at insulators as well as on the mechanism of charge transfer at liquid–dielectrics interfaces, a series of proof-of-principle experiments were developed to demonstrate the viability of TES for chemical analysis. When a liquid droplet with dissolved analytes moves across an insulating solid its triboelectrification will be recorded by electrometer to retain both spatial and temporal information on the charge exchange (liquid–insulator) (**Figure 4**a). Most important, the charge pattern along the sample trajectory is analyte-specific. By means of building a database of ~30 types of common salts, acids, bases, and organic molecules, and through a simple automated identification program, each TES charge fingerprinting map enables a nondestructive and ultrafast (<1 s) new analytical tool with qualitative and quantitative accuracies close to 93%, and the detection limit of TES is ppb levels. The mechanism of TES curves for different chemicals should be ascribed to the competition of cations with interfacial H_3O^+, leading to the dynamic changes of

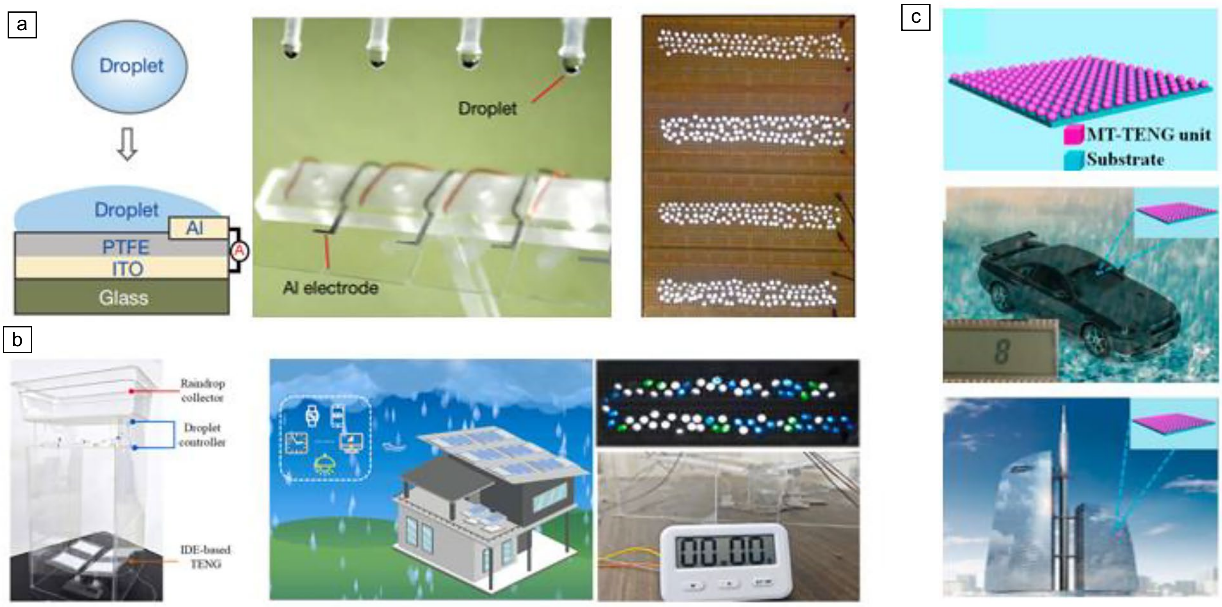

Figure 5. Energy conversion enabled by charge transfer at liquid–solid interface. (a) The device design shows the voltage produced by four parallel droplet energy generators (can light up 100 small LEDs).[57] Reprinted with permission from Reference 57. © 2020 Nature Publishing Group. (b) Triboelectric nanogenerator (TENG) for raindrop energy harvesting can power a light-emitting diode and a clock.[58] Reprinted with permission from Reference 58. © 2024 Elsevier. (c) The device with hundreds of units of TENG, demonstrating the potential applications in integrating with vehicle and building.[59] MT, multiunit transparent. Reprinted with permission from Reference 59. © 2016 Elsevier.

H_3O^+ concentration at the liquid–solid interface when droplet sliding on the solid surface. Recently, based on a TENG probe with two separated electrodes, a droplet-tasting sensor was developed.[54] The sensor system identifies the types of liquid food by analyzing the shape of the current curves generated on the two electrodes during liquid–solid triboelectric electrification, such as the magnitudes of the current signals, the time interval between the peak and the valley of a waveform, and the time interval between the peaks of two wave forms (Figure 4b). The information extracted from electric current signals will be a fingerprint of liquid food, such as liquor, coffee, and white vinegar. Moreover, the TENG probe can also monitor the bubble states. For example, a tube liquid–solid TENG with a single electrode as a probe to explore the bubbles status. The effect of bubble fluid dynamics on liquid–solid CE was detailed studied, including volumes, flow velocity, and release intervals of bubbles. Such sensor with a sensitivity of 13.2 $V \cdot cm^{-3}$, a signal-bubble volume correlation coefficient of 0.9964, and a response time of 0.15 s.[55]

Energy conversion enabled by charge transfer at liquid–solid interface

Water resources are abundant in nature; about 70% of the earth's surface is covered by water resources.[56] However, due to technological limitations, converting the low-frequency kinetic energy, including waves, tides, and rainwater into electrical energy efficiently is still difficult. Liquid–solid CE will generate electric signals, and combination with power management will provide a chance to effectively harvest the water energy. Zuankai Wang's group developed a droplet energy generator (DEG) to highly efficient harvest water energy in 2020. The study shows that a 100-µl water droplet dropped from a height of 15 cm can generate a voltage of more than 140 V and light up 100 LED lights (**Figure 5**a).[57] Another example is an array raindrop TENG, as shown in Figure 5b. By connecting four groups of TENGs in parallel, such a device has the potential to harvest raindrop energy at outdoor locations, for example, rooftops, to power small electronic devices.[58] Arrayed TENGs can also be integrated with glass on vehicles or buildings to harvest raindrop energy and power small electronic devices in cars or buildings (Figure 5c).[59]

Conclusions and perspectives

Based on the charge transfer at the liquid–solid interface, TENG as a probe can be used to study the basic mechanism of charge transfer at the liquid–solid interface, especially for charge-transfer dynamics between liquids and solids. TENG has shown unique application prospects in the fields of sensing and energy harvesting. This unique capability is expected to surpass the limitations of traditional analytical instruments and sensors, which suffer from complex sample preparation and testing procedures, require either bulky or expensive instrumentation, and are unable to provide *in situ* monitoring. For example, TES chemical analysis can be further miniaturized, which will open a path to rapid chemical detection relying on portable low-tech instrumentation.[53,60] Similarly, the TENG for water energy harvesting can be self-powered, inexpensive, and effective. However, the TENG probe is still in its infancy and many challenges and opportunities still exist. These include (1) a fundamental understanding of charge transfer at the liquid–solid interface, especially for charge-transfer dynamics and the basic mechanism of charge distribution on a solid surface. (2) The sensitivity and selectivity of sensors based on a TENG probe need to be improved. (3) The structure design of a TENG probe needs to be diversified to adapt to different application scenarios, such as flexibility. (4) Development of various contact solid materials. (5) The application of the TENG probe in other fields requires further development, for example, biologic detection and environmental pollutant detection. (6) Standardized fabrication and industrialization of TENG probes still have a long way to go. A deep understanding of fundamentals and novel designs of structures could make the TENG probe a powerful tool for sensing systems in the future.

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Author contributions
All authors have conceptualized, designed, and wrote this article.

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Conflict of interest
On behalf of all authors, the corresponding author states that there is no conflict of interest.

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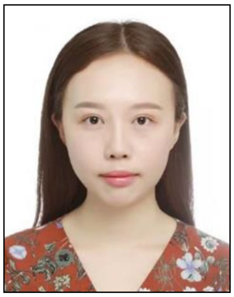

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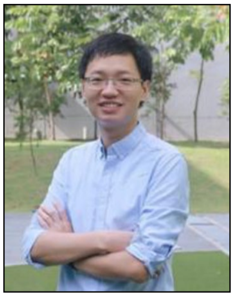

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