

Effect of Surface Pre-Charging and Electric Field on the Contact Electrification between Liquid and Solid

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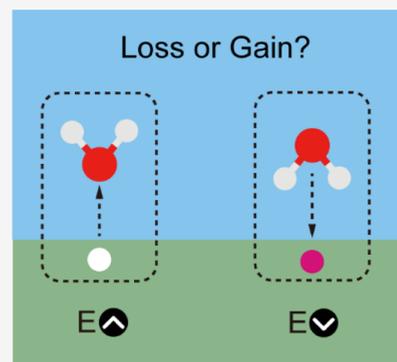
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ABSTRACT: Contact electrification at the solid–liquid interface is a ubiquitous phenomenon. In spite of the extensive studies about its origin, it remains difficult to reveal the mechanism of charge transfer under special conditions (e.g., in the case of pre-charging or applied electric field). Here, we measured the charge transfer between solids and liquids with different pre-charged pairs using an acoustic levitation–electric field measurement device. It is demonstrated that the interaction between the electronegativity and the electrostatic attraction by the net charge of the solid can facilitate or suppress the amount of transferred charges after contacting with the liquid. Considering that the pre-charges may affect the charge transfer by establishing an interfacial electric field, the effect of the applied electric field on the charge transfer at the solid–liquid interface was further explored. Particularly, charge transfer along the opposite direction of the driving electric-field force is observed, which is more likely due to electron transfer. The results are well explained using Wang’s hybrid electric double layer model, which consists of transferred electrons, specifically oriented water molecules, and adsorbed ions.



INTRODUCTION

Contact electrification (CE) is a common phenomenon in nature. When two materials come into contact, the charge will transfer from one surface to the other. Based on the phenomenon of CE, diverse technologies such as triboelectric nanogenerators (TENGs),^{1–3} electrophotography,⁴ air filtration,^{5,6} and interface spectroscopy^{7,8} have been considerably developed. CE not only occurs between the solid–solid interface but also exists on the solid–liquid interface, which is also the most common contact interfaces on earth. The flow of liquid over a solid surface normally results in the solid surface being electrically charged.^{9–12} Moreover, when the liquid flows over the charged solid surface again, it shows fascinating physical–chemical changes. This charged interface is widely employed in droplet manipulation,^{13,14} energy conversion,^{15–17} catalysis^{18,19} and electrochemistry,^{20,21} and so forth. However, almost all of these works have focused on the charged solid, while the charged liquid or both charged cases remain unexplored, which leads to an incomplete understanding of the solid–liquid interface.

The internal electric field established due to the charge difference at the solid–liquid interface may trigger many novel effects and phenomena, such as the tribovoltaic effect,²² hydrovoltaic effect,²³ dewetting,²⁴ and so forth. Behind their curtain is a vigorous discussion to determine the origin of charge transfer caused by material transfer,²⁵ ion transfer,²⁶ or electron transfer.^{27–29} Through introducing thermal excitation³⁰ and photoexcitation^{31,32} at the solid–liquid interface, the relationship between electrons and ions is further elaborated. However, experiments involving solid–liquid

interfaces under an applied electric field are rare. In particular, the effect and mechanism of charge transfer under the action of an electric field are still elusive.³³ Thoroughly elucidating the above processes is of critical importance for basic science.

Very recently, we developed an acoustic levitation–electric field charge measurement device that provides a contact-free test environment.³⁴ Based on this device, further by improving and optimizing, we can apply an electric field perpendicular to the solid–liquid interface during CE in this work. The charge transfer between different charged pairs at the solid–liquid interface was investigated. Most importantly, the unusual charge transfer phenomenon at the solid–liquid interface is found when an electric field is applied, and the effect of aqueous solution and electric field direction are also taken into account. Wang’s hybrid electric double layer (EDL) model³⁵ applicable to the case of an external electric field is proposed to explain this intriguing phenomenon.

RESULTS AND DISCUSSION

Solid–Liquid Interface Scenarios and Principles of Charge Measurement. As shown in Figure 1a, the charge measurement device consists of three parts. The top acoustic

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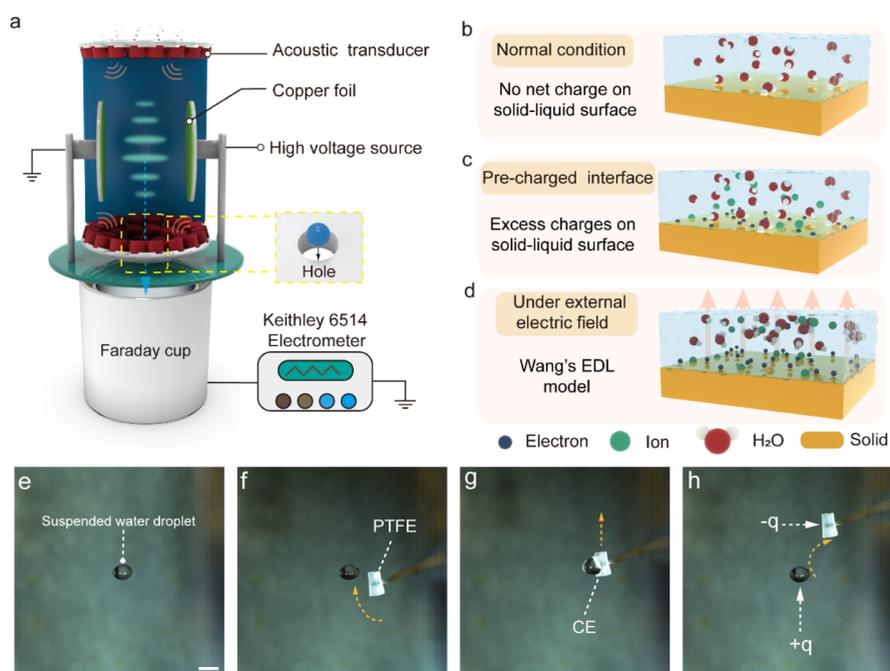


Figure 1. CE at the solid–liquid interface under different initial conditions. (a) The schematic diagram of the charge measurement device based on an acoustic levitation–electric field. (b) Solid–liquid interface is initially electrically neutral. (c) CE between the charged solid surface and the charged liquid. (d) CE at the solid–liquid interface under an applied electric field. (e–h) Schematic diagram of the solid–liquid CE process. The orange line represents the direction of movement of polytetrafluoroethylene (PTFE). Scale bar: 2 mm.

transducer unit is responsible for generating standing waves to provide the acoustic radiation force needed to levitate the object. Two parallel metal plates are connected to a high-voltage power supply to provide an electric field in the horizontal direction. The Faraday cup at the bottom is connected to an electrostatic meter to record the charge. In experiments with solid–liquid CE, solid and liquid are initially both considered electrically neutral (Figure 1b), CE is only regulated by the nature of the material itself, and numerous experiments were performed under this ideal setting. However, in practical situations, it is impossible to completely exclude the interference of the container, the solid or liquid still retains subtle residual charges (net positive or negative charges) at the pC level or even more, and the transferred charges are then influenced by the initial charges on the surface. CE in this situation is called a pre-charged interface (Figure 1c). More importantly, the classical solid–liquid EDL theory is based on the concept of gradient distribution of ions at the solid–liquid interface and ignores the contribution of electrons, and Wang et al. introduced the contribution of electrons into the model of EDL, which furthered the understanding of the solid–liquid interface. However, the charge transfer at the solid–liquid interface under an applied electric field has been rarely reported before owing to the limitations of the experimental setup. Moreover, Wang’s EDL model is based on experimental conditions without external field effects, and it becomes urgent to investigate whether the model continues to be applicable in special conditions (e.g., CE under applied electric field, Figure 1d), which will be discussed in the subsequent sections. For the current research dilemma, a device based on acoustic levitation and electric field was developed to open a new path for studying charge transfer at the solid–liquid interface in a special environment, and the work principle of the device is described in methods. As shown in Figure 1e, a 5 μL deionized (DI) water droplet is stably suspended in the air medium due

to the capture center formed by the acoustic radiation force to counteract gravity, and this contactless measurement environment avoids the interference of the containers with the experiment. Subsequently, a PTFE film is used to contact with the DI water droplet, and the dynamic process of CE is shown in Figure 1e–h and Movie S1. In this case, the water droplet is usually positively charged, while the PTFE is negatively charged. Finally, the charge variation on the droplet before and after contact is measured by a Faraday cup to represent the charge transfer during the whole CE process.

Effect of a Single Pre-Charged Object on Charge Transfer. As shown in Figure 2a, the initially negatively and positively charged PTFE is used to contact with uncharged DI water droplets. Fluorinated ethylene propylene (FEP) and Cu films were used as triboelectric material, and PTFE serves as another triboelectric material and was pre-charged by TENG in the vertical contact-separation mode³⁰ (Figure 2b). Detail of the process of pre-charging the material is given in methods and illustrated in Figure S1. The amount of charges on the PTFE surface is regulated by controlling the time of contact cycles, the amount of transferred charges will gradually increase as the number of contacts increases, and the charge density σ is derived from the following equation: $\sigma = Q/S$, where Q is the total PTFE charges measured by Faraday cup, and S is the surface area.

Seven groups of PTFE samples with different initial charges were purposely selected for CE with almost electrically neutral DI water droplets. The amount of transferred charges to the water droplet after being contact with PTFE is shown in Figure 2c. Interestingly, when PTFE is already initially negatively charged in a small amount ($\sigma = -0.37 \pm 0.11 \text{ pC/mm}^2$). Due to the attraction of negative charges to positive charges, upon contacting with water, the PTFE should have received positive charges or the water droplets should have received negative charges. However, the fact is that the water droplet continually

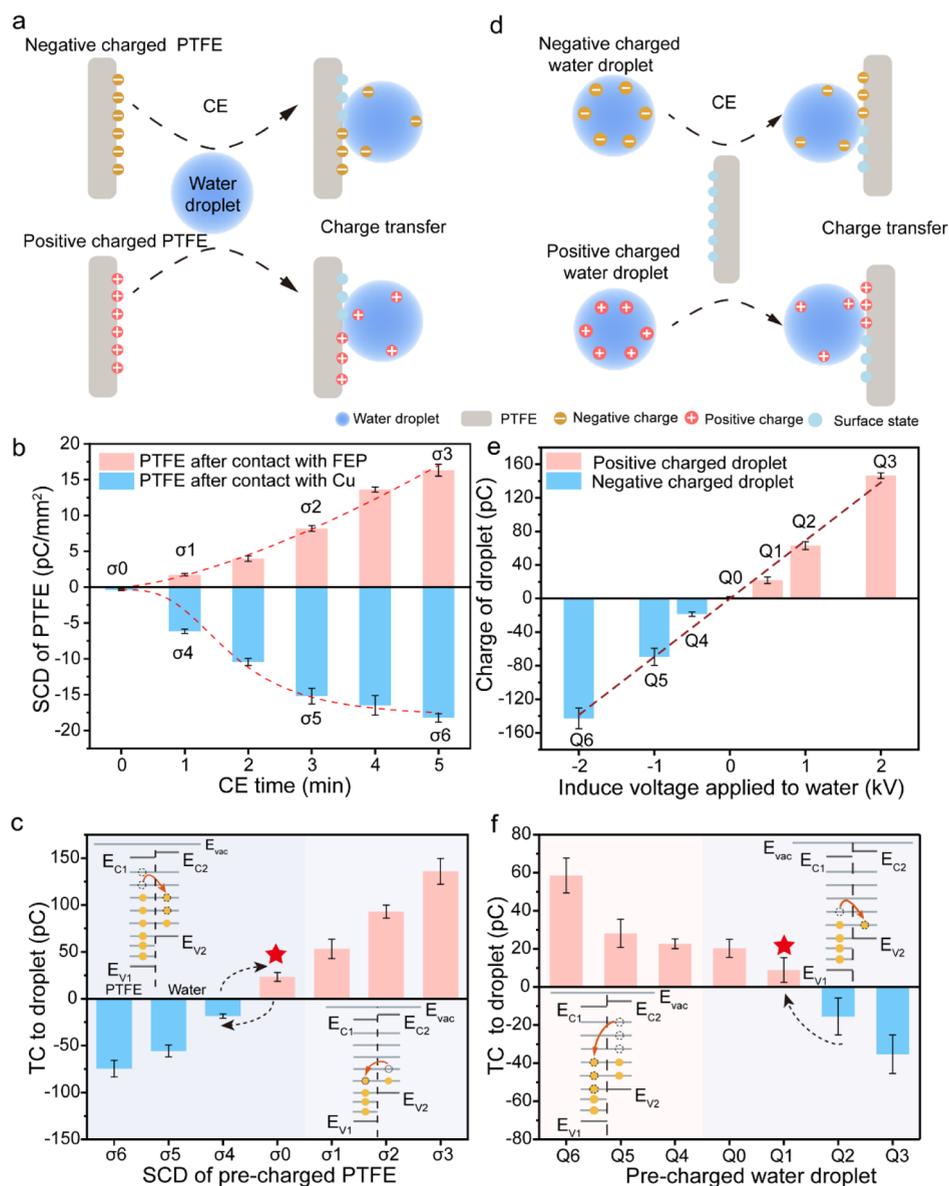


Figure 2. Charge transfer between charged/uncharged pairs at the solid–liquid interface. (a) Schematic diagram of CE between an initially positively or negatively charged PTFE and a neutral water droplet. (b) The evolution of the surface charge density of PTFE with contact time. (c) The evolution of the transferred charges to the droplet, after contact with selected PTFE with different initial charges. (d) Schematic diagram of charge transfer between an initially positively or negatively charged droplet and an almost uncharged PTFE. (e) Plot of the initial charges of the droplet and induced voltage. (f) The variation of the transferred charges on the droplets, after the selected droplets contact with uncharged PTFE.

loses its negative charges and becomes positively charged, while the PTFE can still get more negative charges (red star in Figure 2c). The negative charges obtained by PTFE is unlikely to be the anion in water because the initial negative charges on the PTFE surface forms an electric field at a distance ranging from a few angstroms to tens of nanometers^{37–39} (Debye length) at the solid–liquid interface that has a tremendous electrostatic repulsive effect on the anion transfer in water, and the free anion ions in water cannot cross the EDL to reach and adsorb on the PTFE surface. This result implies ions are not the cause of charge transfer in this case. Instead, electronegativity is only responsible for electrons,^{40,41} and for the electrons in water molecules, the electrostatic repulsion of the EDL is overcome by the electronegativity of the PTFE, so the electrons are still transferred from the water to the surface of PTFE. The phenomenon is also observed when the initial

charge density on the solid surface changes from σ_0 to σ_1 . It is not just the electrons in the water that are transferred to PTFE due to electronegativity, and the initial excess net positive charge on the PTFE surface will also cause attraction to the anions in the water, and the amount of charge transferred increases with the initial positive charge density of PTFE (σ_1 to σ_3). More anions in the water are adsorbed to the PTFE surface, thus enhancing the amount of charge transfer between the solid and liquid, the transfer of ions in the water dominates the charge transfer. Accordingly, the amount of charges transferred to the water droplet increases with the initial negative charges of PTFE (σ_4 to σ_6), and the charges on PTFE is generated by the electrons obtained after contact with copper, which means that electrons dominate the charge transfer process. We also examined the amount of charges transferred to the PTFE and found that the PTFE gained the

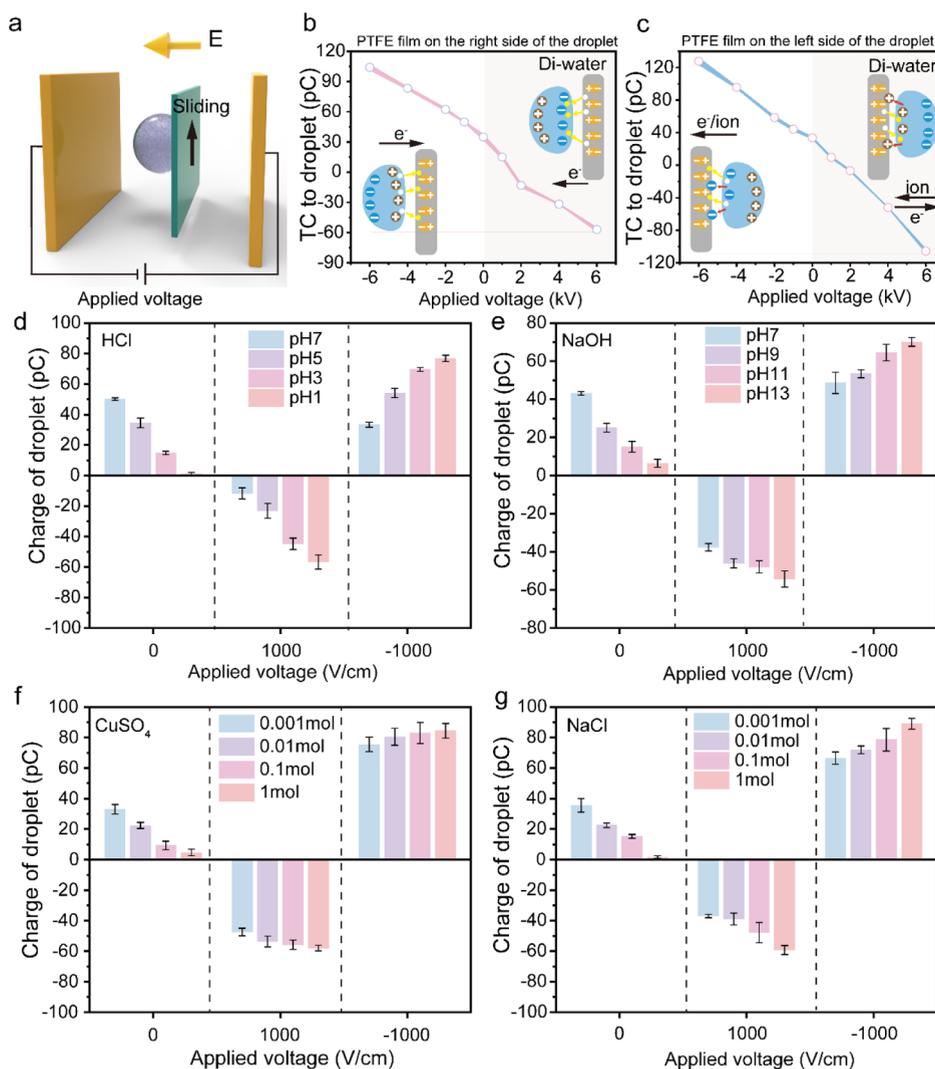


Figure 3. Charge transfer at the solid–liquid interface under an applied electric field. (a) Experimental schematic diagram of charge transfers with the applied electric field. When the PTFE film contact with the droplet on the right side (b) and the left side (c), the transfer charge versus the applied electric field. (d) HCl, (e) NaOH, (f) CuSO_4 , and (g) NaCl, four solutions after being contacted with PTFE, the amount of charge transfer versus the applied electric field. Charge transfer at the solid–liquid interface under external electric field.

same amount of charge as the droplet, but with opposite charge sign, proving the experimental reliability (Figure S2).

On the other side, pre-charged DI-water water droplets were used to contact with uncharged PTFE (Figure 2d). The water droplet is negatively (Figure S3a) or positively (Figure S3b) charged. The relationship between the initial charge on the droplet and the induced voltage is shown in Figure 2e. Similarly, seven groups of water droplets with different initial charges were used to contact with the almost electrically neutral PTFE film (Figure 2f). According to the classic perspective for ion transfer,²⁶ a water droplet with net positive charges means that there is an excess of cations in the droplet, and after being contacted with PTFE, the water droplet should lose its positive ions and become negative charged due to the massive ion adsorption. However, we were surprised to find that the initially positively charged droplets (Q0) and (Q1), after CE with PTFE, still got more positive charges (red star in Figure 2f), a phenomenon that cannot be explained by ion transfer. Moreover, the PTFE used in this experiment initially carries extremely little negative charges and attracts cations more preferentially, but this is not consistent with our

observations (Figure 2b σ_0). The physical nature of this process is due to the transfer of electrons from the water to the PTFE. From the surface state theory perspective,⁴² electrons occupying higher energy levels will transfer from the higher energy levels to the lower energy level surface states (inset in Figure 2c,f, more discussion shown in the Supporting Information). In addition, for the liquid–gas interface, hydroxide aggregation at the interface may cause the droplet to lose electrons more easily.^{43–45} The CE cycles for charged droplets after being contacted with same locations on the PTFE surface, as shown in Figure S4. However, it is undeniable that when the initial positive or negative charge on the droplet exceeds a certain value (e.g., Q2 and Q3), the contribution to transferred charges of electrons is hidden by ions, in which case, the number of ions electrostatically adsorbed on the solid surface is greater than the number of transferred electrons, which follows the principle that the transferred charges to the PTFE increase with the initial charges on the water droplet. Moreover, the discussion about the effect of double charged pairs on the charge transfer between the solid–liquid interface is shown in Figure S5 and the Supporting Information

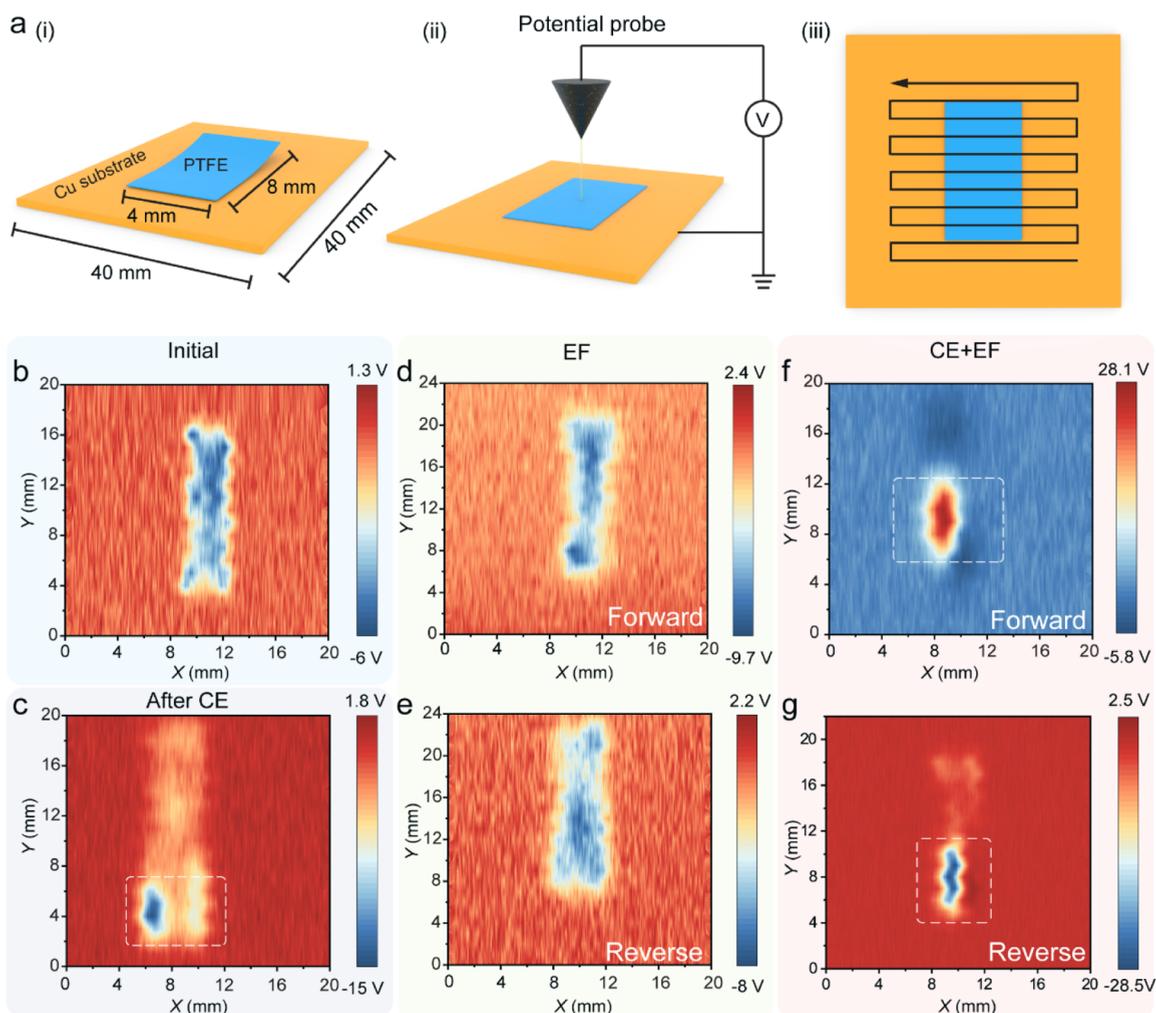


Figure 4. PTFE surface potential distribution. (a) Schematic diagram of the surface potential measurement process. (b) The initial state. (c) After contact with the water droplet. Under positive (d) and reverse (e) electric field treatment. After contact with water, when positive (f) or reverse (g) electric field is applied, respectively. The white wire frame represents the area in contact with the water droplet.

The ability of the device to apply an electric field perpendicular to the direction of the solid–liquid contact interface was developed to explore the effect of an applied electric field on the solid–liquid charge transfer, and a schematic diagram of the device is shown in Figure 3a. We investigate the charge transfer behavior between solid and liquid by varying the relative positions of the film and droplet contact positions as well as the strength and direction of the electric field. First, DI water droplets are used to come into contact with the PTFE film. The relationship of the transferred charge and the applied voltage is shown in Figure 3b. When a positive electric field is applied, after contacting with PTFE, the water droplet becomes negatively charged; while when a negative electric field is applied, the water droplet becomes positively charged after CE. Then, we repeated the above experiment by placing the solid sheet to the left of the droplet, and the CE results are shown in Figure 3c. Coincidentally, the overall trend of the transferred charge remained consistent. However, we found that when both applied a positive electric field (right insert in the Figure 3c,b), PTFE film on the right and left side of the droplet after contact with the droplet, respectively, the charge transfer of the droplets is all negative. However, what is obvious is that the dipole symbol on the contact surface has changed. Similarly, when keeping the

polarization direction of the droplet fixed (left insert in the Figure 3c,b), after the PTFE film is rubbing on the left side of the droplet and the right side of the droplet, the droplets are positively charged. However, the dipole sign at the solid–liquid interface is also different, which suggests that different charge transfer mechanisms are responsible for such a result. Therefore, we speculate that it is not ions but electrons that cause the charge transfer. Because whether the applied positive or negative electric field, the water will be polarized under the action of the external field. The anions and cations in the water are pulled to the two sides of the droplet under the electric field force, respectively, and cannot get close to each other. Therefore, when a negative electric field is applied, only the transfer of electrons from the droplet to the PTFE can make the droplet positively charged (left insert in Figure 3b). Moreover, when a negative electric field is applied, only the electrons on the PTFE are transferred to the water to make the droplets negatively charged (right insert in Figure 3b). For the case where the film is rubbed on the left side of the droplet (Figure 3c), the dipole is generated inside the polarized PTFE by the applied electric field, but the dipole (bound charge) cannot be transferred. When a positive electric field is applied, the negative charge on the droplet may be due to the combined transfer of ions and electrons (left insert in Figure

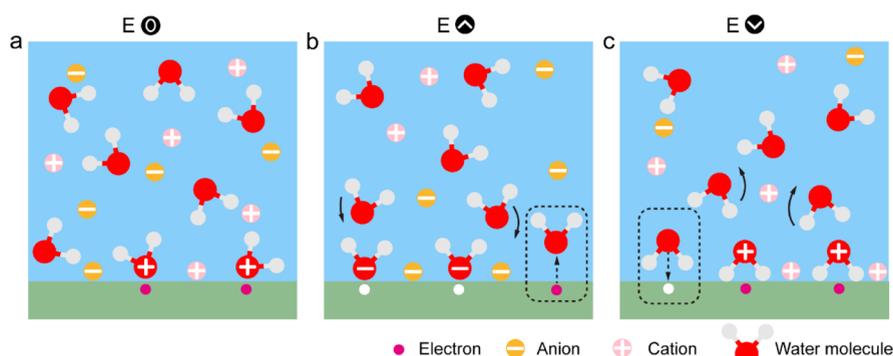


Figure 5. Schematic diagram of charge transfers at the solid–liquid interface. Illustration of electron transfer in the case of (a) without an electric field applied, (b) electric field direction from solid to body liquid, and (c) electric field direction from the liquid droplet to solid.

3c), when the negative electric field is applied, the electrons and negative ions on the droplet are transferred to the PTFE, making the droplet positively charged (right inset in Figure 3c). For ruling out the possible influence due to the material properties, we repeated the above experiments immediately using both FEP and polydimethylsiloxane under the same conditions, as shown in Figure S6. The overall trend of charge transfer for different solid materials remains similar to PTFE, but the difference in electronegativity for electrons of the three materials results in different amounts of charge transfers.⁴⁶

In order to comprehensively investigate the effect of ions in solution on the transferred charge, we set the intensity of the electric field to 1 kV/cm. Droplets from four solutions of HCl, NaOH, CuSO₄, and NaCl were selected to bring into contact with PTFE under electric field, and the transferred charges to the water droplet are shown in Figure 3d–g. As shown in the dates, the amount of transferred charges decreases with the increase in ion concentration in solution for all four solutions without an external electric field. This is consistent with the reported literature and indicates that the high concentration of ions in the solution inhibits electron transfer.^{30,47,48} Subsequently, when an electric field is applied during CE, we find that the transferred charges increase with the increase in ion concentration of the droplets from four aqueous solutions. Although the specific adsorption of ions in the EDL might affect the experiment, assuming that the PTFE is specific adsorption for one certain ion from water, the charge transfer is not affected by the direction of the electric field.⁴⁹ In other words, the sign of the charge transferred to the droplet does not change by changing the direction of the external electric field. However, after altering the electric field direction, four solutions in contact with PTFE, the charge transfer direction changes. Therefore, above results cannot support the explanation of ion specific adsorption. Besides, after contacting with PTFE, it is easy for all of the droplets from four solutions to get negative charges when a positive electric field is applied and get more counter (positive) charges when a negative electric field is applied. The charge transfer regulated by the external electric field in this case defies the traditional physicochemical expectation but in good agreement with the results in Figure 3b,c. We speculate that it may be due to the addition of ions to the solution that new impurity energy levels are introduced in the energy band of water.⁵⁰ The number of energy levels increases with the number of added ion concentration, and these new defective states play a crucial role in electron capture. The results of the molecular dynamics simulations also support the above assumptions (Figure S7),

and the specific experimental setup and discussion can be found in the Supporting Information.

Surface Potential Measurement. As shown in Figure 4, the PTFE surface potential distribution was further analyzed under different solid–liquid contact conditions. Figure 4a(i–iii) shows the schematic diagram of the experiment. A PTFE film is taped to a back copper electrode and a potential probe is used to detect the surface potential, and the scanning path is shown in Figure 4a(iii). We investigated the distribution of PTFE surface potential in the original state (b), after contact with liquid droplets (c), after electric field treatment (d,e), and CE under an electric field (f,g). It can be seen from Figure 4b that the initial PTFE surface has a slightly negative potential, and when rubbed with water droplets, the potential in the CE region decreases significantly (Figure 4c), indicating that the PTFE gets more negative charges, and all of the above experiments are consistent with the experimental data in Figure 2. In order to exclude the possibility that the polymer may be polarized in the electric field and possess an inherent surface potential, the PTFE film was placed in the forward and reverse electric fields, and Figure 4d,e shows that the surface potential changes negligibly compared to the initial state. Subsequently, when PTFE was rubbed with water droplets under an electric field, we unexpectedly found that the surface potential of the CE area became positive (Figure 4f), indicating that PTFE lost electrons, when the direction of the electric field is changed, the potential of PTFE surface decreases, which proves that PTFE gets electrons (Figure 4g). We also measured the potential of the FEP surface, as shown in Figure S8. It can be found that the initial surface of FEP has more negative charges, so the potential is even lower (Figure S8a). After contact with water droplets, the surface potential of FEP further decreases after it gets negatively charged (Figure S8b). However, the electric field polarization also shows little effect on the FEP (Figure S8c,d). Surprisingly, in Figure S8e,f, we unexpectedly observe that in the solid–liquid contact region, it is apparently centrosymmetric or surrounded by a distribution of both positive and negative potentials. This indicates that throughout the charge transfer process, ion adsorption due to external electric field and electron transfer due to electronegativity are coexisting. However, the amount of charges of transferred electrons is more than the amount of changes of ions. This can be seen from the magnitude of the surface potential, and electron transfer dominates the charge transfer process is further demonstrated. However, when measuring the charge of water droplets, only the dominant factor (i.e., the result of electron transfers) is shown, so the contribution of

ions to the transferred charges is hidden by electrons, which exactly explains the phenomenon in Figure 3. Here, we reveal that the charge transfer at the solid–liquid interface is influenced by the co-competition of electrons and ions under the electric field and that the transfer of electrons dominates the whole process by observing the potential distribution on the solid surface.

Wang's Hybrid EDL under the Applied Electric Field.

The above experimental results can be explained by the hybridized EDL model at the solid–liquid interface first proposed by Wang,³⁵ they further demonstrated the existence of electrons in the EDL.³⁰ As shown in Figure 5a, in general, when the liquid is in contact with PTFE, the water molecules first lose electrons (step one) and become positively charged, then the PTFE gets electrons and becomes negatively charged, thus attracting the positive ions in the water (step two), and thus, a “two-step” EDL is established.³⁰ Although the “two-step” model generalizes the charge transfers at the solid–liquid interface, some modifications are needed when an external electric field is applied. Combined with the experimental and simulation results in this paper, Wang's hybrid EDL model applicable under the applied electric field is proposed, as shown in Figure 5b,c. When the direction of the applied electric field is from the solid to the liquid (Figure 5b), the anions and cations in the water are gathered on the two sides of the droplet, respectively. At this time, under the action of the external electric field, water molecules to the electric field force direction are in preferential arrangement, so that the oxygen atoms in the water molecules are closer to the solid surface. After CE, the electrons on the PTFE surface are captured by the water molecules and leave holes with positive charges in PTFE. Solid surface holding a net electric charge acts to align the static dipole of water molecules at the surface.⁵¹ Moreover, the specifically oriented water molecules are attracted to the positively charged solid surface together with other anions in the aqueous solution, forming EDL. When the direction of the electric field is pointed from the liquid to the solid (Figure 5c), the orientation of the water molecules changes so that the hydrogen atoms in the water molecules are closer to the interface under the action of the electric field. Meanwhile, the electrons on the water molecules transferred to the solid surface, and specifically oriented water molecules and other cations in aqueous solution are adsorbed to the negatively charged solid surface to form EDL. Transferred electrons (or holes) could absorb opposite ions at the solid–liquid interface, but electron transfer is highly sensitive to changes in the ion concentration in solution as well as the external electric field. Ultimately, adsorbed specifically oriented water molecules on the solid surface are in obvious competition with other ions for the solid surface adsorption sites.

CONCLUSIONS

In summary, with the recent prevalence of solid–liquid TENG, the phenomenon of CE at the solid–liquid interface has been revisited, and the two main problems (i.e., charge carriers and the EDL) plaguing CE at the solid–liquid interface have been further discussed. The measurement of charge transfer between pre-charged pairs at the solid–liquid interface is achieved by an acoustic levitation–electric field charge measurement device. It was found that for CE between solids and liquid droplets, the initial charges of both have a significant effect on the amount as well as the transfer direction of charges at the solid–liquid

interface. It is further concluded that electrons are the main contributors to charge transfer when dominated by material electronegativity, while ions are the main contributors to charge transfer when dominated by the electrostatic attraction of excess net charges of materials. The interaction between the electronegativity and the net charges can facilitate or suppress the amount of transferred charges. The dividing point between electronegativity and electrostatic attraction depends on the magnitude and polarity of the initial charge of the two contacting materials. In addition, we observed the transfer behavior of electrons against the electric field direction when an electric field is applied. Charge transfer is the result of competition between electrons and ions by solid surface potential analysis, and electrons are more likely the dominant factor. Besides the consideration of the role of Wang's hybrid layer,⁴⁸ our experiments imply that the orientation of the water molecules near the surface of the solid plays a key role in charge transfer under the action of the external electric field. The hybrid EDL model first proposed by Wang (based on the electron transfer, and the adsorption of ions in aqueous solution to the solid surface), renewed by specifically oriented water molecules, is used to explain the phenomenon. This innovative model may renew our understanding of EDL and further advance research including TENG, electrocatalysis, organic chemistry, and so on.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.jpcc.2c01713>.

Materials and methods; acoustic levitation–electric field droplet charge measurement device; pre-charging process; droplet charge measurement; computational methods; surface state theory for the mechanism of charges charge transfer; effect of double charged pairs on charge transfer between the solid–liquid interface; molecular dynamics simulation under applied electric field at the solid–liquid interface; PTFE pre-charging process and charge transfer mechanism in energy band theory; transferred charges to the PTFE and droplet, after CE; pre-charging process of droplet and charge transfer mechanism; charge transfer versus number of contact cycles; charge transfer network diagram between solid–liquid pre-charged pairs; variation of charge transfer with the applied electric field; molecular dynamics images; and FEP surface potential distribution (PDF)

Process of CE between liquid and solid (MP4)

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Author Contributions

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Notes

The authors declare no competing financial interest.

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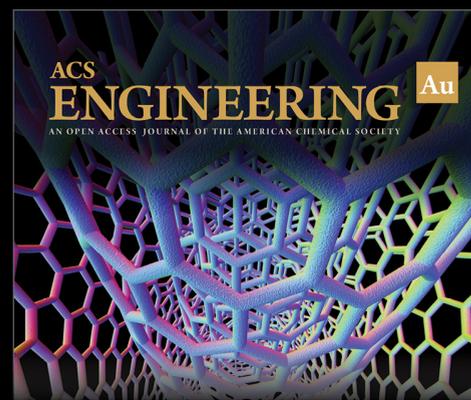
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