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## PAPER

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#### 1. Introduction

Contact electrification (CE) or triboelectrification (TE) are terms to describe the charge exchanging phenomena across the interface of two materials in contact.<sup>1,2</sup> Despite the prevalence of related natural CE/TE phenomena in our surroundings, identification of the charge transfer mechanism still remains a challenging research subject.<sup>1,3</sup> Meanwhile, the relatively straightforward architectures of triboelectric nanogenerators (TENGs) have made them a promising solution in the energy harvesting field.<sup>2,4</sup> In the TENG device, when a dielectric (usually polymer) touches a metal electrode, mobile charges are electrostatically induced and displaced. As the

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# Tailoring morphological and chemical contributions of nanoscale charge transfer for enhanced triboelectric nanogenerators<sup>†</sup>

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Triboelectric devices, operating through contact electrification (CE) and electrostatic induction, have shown great promise in energy harvesting applications. However, optimizing charge transfer at the interface remains crucial for enhancing device performance. This study introduces a novel approach to harnessing CE by employing morphological and chemical modifications of polymers. Our strategy involves adjusting the elastomer base to curing agent ratio to fine-tune the chemical properties of polydimethylsiloxane (PDMS) and introducing morphological modifications through a peeling and flipping (P/F) process of PDMS off the Si-substrate. Unlike conventional methods, the P/F-method minimally alters the intrinsic properties of PDMS, creating nanoscale surface corrugations adiabatically. We explore the mechanical, tribological, and electrical properties of the surface at the nano-scale and demonstrate that our approach allows for precise control of energy dissipation and electric potential at the surface, thereby optimizing charge transfer. Furthermore, we show that using a plasma-treated Si-substrate can further increase device performance up to 80% without affecting other properties. This study presents a comprehensive strategy for fine-tuning CE to enhance the performance of triboelectric nanogenerators.

capacitor system varies due to the separation between the materials, the gradient causes the current flow. Therefore, facilitating charge transfer on the triboelectric surfaces is the most fundamental strategy for advancing TENGs. The degree of CE depends not only on the pairing selection between triboelectric materials with the different chemical potentials,<sup>1,5,6</sup> but also on morphological<sup>7-18</sup> or other chemical/electrical effects.<sup>19–24</sup> Although various engineering approaches have been applied to TENG devices, they often excessively impact the device in terms of morphological, chemical, and electrical aspects, leading to unexpected CE degradation. Specifically, when CE depends on polymers, unregulated treatment can readily deteriorate the device output.<sup>10,11,21</sup> Therefore, it is essential to carefully identify triboelectric charge and transfer mechanism.

Here, using polydimethylsiloxane (PDMS) as a representative polymer, we evaluate a new engineering strategy to finetune CE and TENG output, which consists of two major steps: (1) manipulating chemical/electrical properties of PDMS by modulating the ratio of base/curing agents and (2) modifying surface corrugation by peeling and flipping (P/F) PDMS off from the Si-substrate. The CE properties were characterized at both nano- and macroscale, respectively. By observing diverse properties of specimens together with morphologies and

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#### Paper

assessing their correlation, we showed that CE is driven by energy-dissipation-assisted thermionic emission. The degree can be systemically and meticulously tuned over a considerable range by using two competing effects: the morphological effect and other chemical/electrical ones. Their coupling can be moderately separated due to the drastic P/F process, unlike other conventional methods. Moreover, our approach can further enhance CE in conjunction with other pretreatments to the Si-substrate, without affecting the intrinsic properties of the polymer.

#### 2. Results and discussion

As shown in Fig. 1(a), to fabricate TENG devices, PDMS elastomer and a curing agent for crosslinking were mixed at various weight ratios of 5/7/10:1, respectively, and followed by degassing under vacuum for 30 min. From the solution, each PDMS film was created on a Si-substrate by spin-coating at 500 rpm for 30 s, and then cured on a hot-plate at 80 °C for 2 h. The resulting thickness of the PDMS was ~310  $\mu$ m (Fig. S1†). For a group (referred to as 'top'), the PDMS membrane was placed

onto the ITO-bottom electrode, keeping the surface as in the as-spun state. Meanwhile, for another group ('bottom'), the bottom interface of PDMS was switched to the surface using P/F-method. Then, the devices' performance was evaluated using a custom-made vibrator in which an ITO top electrode repeated a vertical tapping motion on the underlying PDMS polymer, supported by the ITO bottom electrode, at 1 Hz rate with a preset maximum force of 30 N; the short-circuit current between the two electrodes was continuously monitored during the motion. Each panel of Fig. 1(b) presents the representative device behaviors with three different chemical ratios. In the graphs, the black and red lines indicate the TENG based on top- and bottom-PDMS, respectively. The peak and valley for each pulse were individually averaged and plotted in Fig. 1(c) from which the peak-to-peak values could be calculated, as shown in Fig. 1(d). We use a notation where either "T" or "B" inside the parenthesis, along with the number at the front, represents the surface side, and the chemical ratio of PDMS, respectively. As shown in Fig. 1(d), not only does the average peak-to-peak current output gradually rise with the increase in the chemical ratio, but the bottom side also consistently produces a higher current output than the top side.



**Fig. 1** (a) Schematic for the preparation and characterization process of a TENG device using PDMS (details are given in ESI<sup>†</sup>). (b) The short circuit current obtained from a TENG device based on top (black) or bottom (red) side PDMSs with different chemical ratios. (c) Mean values in peak and valley regimes. (d) Mean peak-to-peak values for six different PDMS conditions. (e) Charge density values for six different PDMS conditions. The auxiliary arrows (magenta) are inserted to clearly indicate the behavior of current and charge density in (d) and (e). We use a notation where either "T" or "B" inside the parenthesis denotes the surface side (top or bottom), with the number at the front signifying the corresponding chemical ratio of PDMS.

#### Nanoscale

Thus, the difference in peak-to-peak current between the top and the bottom with the same chemical ratio gradually decreases with the chemical ratio, being estimated at  $\sim 0.41$ , ~0.33, and ~0.15 µA for 5:1, 7:1, and 10:1, respectively. The amplitude of peak-to-peak current reflects the surface state density, and the difference of electron affinities of the two materials (Fig. S2<sup>†</sup>).<sup>25</sup> Similarly, we estimated the charge density  $(\sigma)$  difference between top and bottom by integrating the current with respect to the elapsed time (Fig. 1(e)). As shown in Fig. S3,† the charge density difference and the peakto-peak current difference between the top side and the bottom side show a very strong positive correlation. As exhibited in the current output of Fig. 1(d), we can reconfirm that the TENG performs better, not only with the increase of chemical ratio, but also on the bottom-based PDMS. However, the obtainable gain by switching from the top-side to the bottomone gradually decreases with the chemical ratio.

To understand such a peculiar CE behavior, scanning probe microscopy (SPM, Multimode 8®) was employed. First, the frictional properties of samples were assessed with lateral force microscopy (LFM) mode. The friction force is obtained by subtraction of the lateral forces measured in the two directions. The friction force ( $F_f$ ) is obtained by halving the difference in lateral forces between the two scan directions, thereby mitigating the morphological effects.

Using a silicon cantilever with an elastic constant of ~0.3 N m<sup>-1</sup>, the half value of lateral deflection difference between the two scanned directions was considered due to friction. Beforehand, the elastic constant and lateral diffraction of the tip had been precisely calibrated based on the thermal tune method<sup>26</sup> and the wedge method,<sup>27</sup> respectively, for quantitative analysis. The loading force, and a scan rate were set at 14 nN and 1 Hz, respectively; the loading conditions were relatively mild so that neither tip wear nor sample surface scratches were observed. Fig. 2(a)



**Fig. 2** (a) A series of friction images obtained on PDMSs with six different conditions. (b) Friction behavior of PDMS depending the conditions; (inset) friction difference between the top and bottom at the same chemical ratio. (c) Representative local load-friction spectroscopy for the six PDMS samples. The fitting lines, based on the linear model, were plotted together using the same color code; (inset) friction coefficients of the six types of PDMS. (d) Lateral force loop obtained in three different regions. (e) The LFM loop area corresponding to energy dissipation.

shows a series of friction images for the six different types of PDMS. From the  $F_{\rm f}$ -images, the area-averaged friction values were plotted in Fig. 2(b), where the inset presents the friction difference between top- and bottom-side. The results exhibit that the friction on all the bottom-side-PDMSs increased substantially compared to the top counterparts, while the friction of the top-side just slightly increased along with the chemical ratio. The tribological difference among the PDMSs can be more evident based on normal load ( $F_{\rm n}$ ) vs. friction ( $F_{\rm f}$ ) spectroscopy (Fig. 2(c)), where a 1-µm single line was continuously scanned, varying the loading force. The resulting friction was observed to be linearly proportional to the loading force for all the cases, following the relation:

$$F_{\rm f} = \mu (F_{\rm n} - F_0) \tag{1}$$

where  $\mu$  and  $F_0$  are friction coefficient and adhesion force, respectively.<sup>28,29</sup> From the linear fitting, the friction coefficients were determined as plotted in the inset. The behavior similar to the previous LFM results validates our analysis well. To interpret such unique friction behavior, we provide the representative LFM loops (Fig. 2(d)), where all the bottom-side-PDMSs reveal the larger friction fluctuation during the sliding regime, as well as the observable drop in slope at the edge, in comparison to those of top-side ones (Fig. S4<sup>†</sup>). Because the slope reflects lateral stiffness in a tip-sample system<sup>30-32</sup> if we consider that the Si-cantilever is usually sufficiently stiff<sup>30,33</sup> and the tip modulus is tremendously higher than that of polymers, the measured slope is attributed to the weakest bonding between the tip and the polymer. Therefore, the declined slope implies the deteriorated modulus in the bottom-side-PDMSs. We also estimated the dissipated energy for the sliding process by calculating the enclosed area of the LFM loop. The results show that the increased energy dissipation occurs together with the enlarged friction fluctuations for all the bottom-side-PDMSs, but the distinction from the top-side gradually decreases with the chemical ratio (Fig. 2(e)). The behavior consistent with friction results conditions validates that the dissipated energy is controlled by the friction altered by the PDMS conditions.

Moreover, to investigate the CE influenced by the dissipated energy, we directly characterize the nanomechanical properties of PDMSs by peak force atomic force microscopy (PF-AFM).<sup>34</sup> In contrast to LFM, PF-AFM relies solely on vertical tapping motion, making it more advantageous in mimicking the contact/separation-based TENG devices. An Au-coated tip with an elastic constant of  $\sim$ 2.8 N m<sup>-1</sup> was used for the study. Due to the quasi-static tip motion in PF-AFM, mechanical properties, such as adhesion ( $F_a$ ), deformation ( $\Delta d$ ), and energy dissipation ( $E_{diss}$ ), can be revealed with the corresponding topography. The modulus of a sample  $(E_s)$  can also be extracted from the slope of the unloading curve (Fig. S5<sup>†</sup>).<sup>34,35</sup> (More detailed estimation procedures were described in the ESI.<sup>†</sup>) For all the samples, the complete PF-AFM data sets are presented in Fig. S6.† A series of topography is selectively exhibited in Fig. 3(a), where the common intensity scale was

adopted for comparison. The figures evidently exhibit that nanoscale protrusions were created on all the bottom-PDMS where their density and fluctuation became less against the increasing chemical ratio. Accordingly, Fig. 3(b) exhibits that the surface roughness significantly increased on all the bottom-side-PDMS, but the distinction from the top counterpart steadily decreased with the chemical ratio. Meanwhile, the top-side-PDMS rarely experiences the roughness variations, and if any, it is only a marginal increase with the chemical ratio. Corresponding to the topographies in Fig. 3(a), a series of  $E_{diss}$  images, and their signal distributions are presented in Fig. 3(c) and (d), respectively. The central values of each distribution were determined by Gaussian fitting and are plotted in the inset. The inset figure depicts that  $E_{diss}$  on the top-side steadily increases with the chemical ratio. All the bottom-side-PDMSs exhibited a significantly increased Ediss compared to the top-side counterpart, but the distinction progressively declined against the chemical ratio. Additionally, we provide exemplary force-distance spectrum curves for several different PDMS conditions (Fig. 3(e)). In the figure, while the dissipation area in 5(B) is significantly larger than in 5(T), the difference in area between 10(T) and 10(B) is very small. Furthermore, the area distinction between 5(T) and 5(B)becomes apparent in the repulsive force regime, due to the noticeable slope variation, which is derived from the degraded modulus and the increased contact time. This induces the elongated/enlarged indentation process, eventually, increasing the friction force and the contact area. This, in turn, is likely to affect the rise of  $E_{\rm diss}$ , even in the attractive force regime,<sup>36</sup> despite similar adhesion force. Additionally, the corresponding signal distributions of  $E_s$ ,  $\Delta d$ , and  $F_a$  are suggested in Fig. S7,† and the central values based on Gaussian fitting were determined as depicted in the insets of each panel. Compared with the behavior of  $E_{diss}$ , while  $E_s$  and  $\Delta d$  show strong negative and positive correlation, respectively,  $F_a$  is likely to be insensitive. Their trends corroborate that  $E_{diss}$  is enhanced on the bottom-side during the indentation, being in accordance with the earlier LFM results.

Moreover, the strong resemblance between the roughness behavior and the  $E_{diss}$  behavior reflects that the indentationinduced  $E_{diss}$  can be further promoted by the surface roughness itself. Eriten et al.37 theoretically predicted that while the frictional energy dissipation is the lowest on the smoothest surface, it increase remarkably with the multiple-asperityinduced roughness. The authors showed that the increasing effect is mainly governed by the height-dependent friction behaviors of asperities, with the contact area contribution to a minor extent. They suggested that taller asperities tend to stick and control the contact stiffness, while shorter asperities slip prefers to dissipate frictionally stored energy. The authors' proposals align well with the findings from PF-AFM results. Indeed, as shown in Fig. S8,† the bottom-side-PDMS not only exhibits greater  $E_{diss}$  over the entire area, but also reveals a more strongly negative correlation between height and energy dissipation compared to the top-side-PDMS. Based on PF-AFM results, we present the contribution diagram to  $E_{diss}$  as a func-



**Fig. 3** (a) A series of topographies obtained on the six different kinds of PDMS. (b) The RMS surface roughness corresponding to topographies of (a). (c) A series of  $E_{diss}$  images corresponding to topographies of (a). (d) The distribution of energy dissipations (dot) and Gaussian fitting (line): (inset) the behaviors of  $E_{diss}$  were extracted from the fitting. (e) Representative force–distance curves measured for the (top panel) 5(T), and 5(B) PDMS, and (bottom panel)10(T), 10(B) PDMS. The arrows indicate the direction of the SPM tip movement. (f) Dependence of  $E_{diss}$  on chemical ratio (x): the lower regime represents the phase where both roughness ( $E_{surf}$ ) and chemical effects ( $E_{chem}$ ) work, while the upper regime represents the  $E_{surf}$ -dominant phase (assuming the linear combination of the two effects,  $E_{diss}$  is presented as the function of x).

tion of chemical ratio (x) (Fig. 3(f)). In the figure, we assumed that the  $E_{\rm diss}$  in the lower phase (corresponding to top-side-PDMSs) was composed of the surface roughness effect  $(E_{surf})$ and other chemical effects  $(E_{chem})$  through a linear combination. In the upper phase corresponding to bottom-side-PDMSs, variations in  $E_{diss}$  ( $\Delta E_{diss}$ ) were assumed to be exclusively attributed to the surface roughness effect  $(E_{surf})$  as described in  $\Delta E_{diss}$  =  $E_{\rm surf} \approx -65.2x + 1444$ , while maintaining  $E_{\rm chem}$  at the same level as in the lower phase. Meanwhile, since  $E_{\rm diss}$  is described as  $E_{\rm diss} = E_{\rm surf} + E_{\rm chem} \approx 45.8x + 735$  in the lower area,  $E_{\rm chem}$  is estimated as 111x + 17.5. The results reveal that  $E_{diss}$  on the topside-PDMS is more influenced by the chemical ratio-driven effect than roughness variation. The chemical ratio-dependent increase of  $E_{diss}$  can be addressed to the increase of friction coefficient and the decrease of modulus, as suggested in Fig. 2(c) and Fig. S7(a),† respectively. Similarly, Lee et al.<sup>38</sup> recently reported that the smaller activation energy at the higher chemical ratio can also enhance  $E_{diss}$  on PDMS through more probable bond-breaking. Therefore, an optimal X can be phenomenologically expected at the point where the two lines meet,

revealing the value of X to be ~12.9. However, if other effects can be involved or the linear trend were varied, the value can be changed, indicating that further studies are required.

The effect of chemical ratio to CE can be characterized by Kelvin probe force microscopy (KPFM) in terms of surface potential. To estimate the dielectric sample's effective work function  $(\Phi_{\text{eff}})$  from the difference between the charge neutral level  $(E_{s,cnl})$  and the vacuum level  $(E_{vac})$ , the surface of every sample was partially coated with Au using a sputter. By measuring the contact potential differences at PDMS and the neighboring Au surface together and obtaining their disparity,  $\Phi_{\rm eff}$  of PDMS can be more effectively derived under the assumption that  $\Phi$  of Au is ~4.8 eV (Fig. S9<sup>†</sup>). Fig. 4(a) reveals the resulting Kelvin probe force microscopy (KPFM) images for all the samples, where the identical tip was utilized to facilitate the comparison between PF-AFM and KPFM.  $\Phi_{\rm eff}$  of PDMS gradually increased along with the chemical ratio, while the difference between top and bottom at the same chemical ratio remained negligible, regardless of the chemical ratio (Fig. 4(b)). Additionally, the  $\Phi$  of ITO was characterized to be



**Fig. 4** (a) A series of  $\Phi_{\text{eff}}$  images for the six types of PDMS, and (b) the corresponding distribution results of  $\Phi_{\text{eff}}$  based on Gaussian fitting. The work function of ITO was characterized as ~4.9 eV, which is used as the top electrode in TENG device. (c) FTIR full-range spectra for top-side PDMS with different chemical ratios: (inset) the reduction of vibration modes of Si–H groups. (d) The negligible differences of Si–H groups between the top and bottom sides for all three chemical ratios. (e) Illustration of the two competing factors harnessing thermionic charge transfer mechanisms: (left) potential barrier manipulation through the chemical ratio-varied  $\Phi_{\text{eff}}$  of PDMS, and (right)  $E_{\text{diss}}$ -enhancement through the surface corrugation.

~4.9 eV based on the CPD difference from Au. (Fig. S10†). Fig. 4(b) suggests that the current output in TENG is propelled by the thermionic emission to overcome the energy barrier created by the work function difference relative to ITO.<sup>39,40</sup> The process is often prominent in metal/dielectric systems.<sup>40–42</sup> Furthermore, the figure indicates that the barrier height consistently decreases against the increase in the chemical ratio. From the KPFM results, we found that surface potential is dependent on the chemical ratio, but not sensitive to the P/F method. Meanwhile, as shown in Fig. 3(b) and (d), both the surface roughness and energy dissipation significantly increase on all the bottom-side PDMS samples, but their distinctions from the top counterparts steadily decrease with the chemical ratio. Therefore, the decrease in output gain obtainable from the P/F method against the increase in chemical ratio is attributable to the reduced distinctions of surface coarsening and energy dissipation with the higher chemical ratio.

To elucidate the variation of  $\Phi_{\rm eff}$  and the correlation with the previous PF-AFM and LFM results in the chemical/electrical aspect, Fourier-transformed infrared spectroscopy (FTIR) was conducted. Fig. 4(c) displays that FTIR spectra for the topside-PDMSs at the different compositions. As shown in the inset, the peak intensity associated with the Si–H vibration mode (~910 cm<sup>-1</sup>) consistently decreased with the increase of chemical ratio for all the top-side-PDMSs.<sup>43,44</sup> Conversely, the asymmetric Si–O–Si modes (~1060 cm<sup>-1</sup>)<sup>45</sup> showed the increasing trends with the chemical ratio (Fig. S11†).<sup>43</sup> While the PDMS elastomer base contains numerous vinyl terminations,

Paper



Fig. 5 (a) (top) Charge density evolution on the back-side of 10:1-PDMS, depending on  $O_2$ -plasma pretreatment time, with the top-side charge density indicated by a dashed line for reference; (bottom) relative charge density difference estimated from the upper panel. (b) Roughness, (c) energy dissipation, (d) friction, and (e) energy dissipation obtained from SPM images for 10-s-plasma-treated back-side-PDMS, in compared with the top and bottom-side-PDMS with no plasma treatment. Full SPM image sets are provided in Fig. S12.<sup>†</sup>

molecules of the curing agent possess several different silicon hydride bonds (Si-H). At the smaller chemical ratio, the molecule chain length of the base is shortened due to the more frequent Si-H-terminated curing agent. As confirmed in PF-AFM and LFM, this leads to an increase in modulus, while, coincidently, decreasing both friction and  $E_{diss}$ . Meanwhile, from the perspective of KPFM, the increased incorporation of Si-H, coupled with the reduction of asymmetric Si-O-Si bonds, induces the n-type effects, rendering the lower  $\Phi_{\rm eff}$  at the smaller chemical ratio, and vice versa. However, in conjunction with the corresponding KPFM results, negligible IR variations between top-side and bottom-side (Fig. 4(d)), suggest that the P/F process rarely impacts the chemical/electrical properties, regardless of chemical ratio. This may be attributed to such an abrupt P/F process, which delivers insufficient energy to alter surface electrical or chemical properties at the room temperature. On the other hand, as shown in Fig. 2 and 3, the dissociation energy of Si-H<sup>46</sup> released during the peel-off process likely increases roughness, friction, and energy dissipation, possibly by generating nano-corrugation on the surface. Conversely, PDMS with a higher chemical ratio can be more smoothly peeled off from the Si-substrate due to the lower energy release, decreasing the P/F-methodinduced output device gain by restricting the expansion of contact area, as shown in Fig. 1. The results validate the earlier  $E_{\text{diss}}$ -evaluation (Fig. 3(f)) based on the separation of the roughness effect from other chemical contributions.

Therefore, our SPM/FTIR results provide evidence that CE is achieved by the  $E_{\rm diss}$ -assisted thermionic emission, and the degree can be controlled by two competing factors, depending on the PDMS conditions; while the variation of  $\Phi_{\rm eff}$  has a dominant impact on CE under mild roughness variation by manipulating the potential barrier height, as shown in the top-side-PDMSs, the enhanced  $E_{\rm diss}$  significantly amplifies CE under large roughness variations, as observed in the back-side-PDMSs (Fig. 4(e)).

Last, to briefly assess the applicability and stretchability of P/F-method to different substrate environments, we pretreated

the Si-substrate with O2-plasma before depositing the PDMS solution. As the dissociation energy of Si-O is known to be larger than that of Si-H,46 we expected that more nano-corrugations would be generated on the surface of bottom side through the P/F-method, by applying O<sub>2</sub>-plama treatment to the bare Si-substrate before dropping the PDMS solution. For 10:1-PDMS, the top and bottom panels of Fig. 5(a) show the plasma effect on the charge densities of bottom- $(\sigma_{bot})$  side, as well as the relative  $\sigma\text{-difference}~((\sigma_{\rm bot}-\sigma_{\rm top})\!/\sigma_{\rm top}),$  respectively, as a function of the plasma time. Surprisingly, after  $\sigma_{\rm bot}$ rapidly grew until ~20 s of treatment, it almost saturated. In reference to the plasma-free case (0 s), the 120 s-treatment dramatically raised the relative  $\sigma$ -difference from ~4% to ~81%. For the 60-s-treated 10:1-PDMS, FFM, PF-AFM, and KPFM were conducted, and compared with the plasma-free 10(T)and 10(B)-PDMSs (Fig. S12<sup>†</sup>). From the SPM results, we confirmed that the 60-s-pretreated 10(B) significantly increased  $E_{\rm diss}$  (Fig. 5(c)) and friction (Fig. 5(d)) with a slight rise of  $R_{\rm q}$ (Fig. 5(b)), while keeping  $\Phi_{\text{eff}}$  almost invariant (Fig. 5(d)).

#### Conclusions

In this study, we present a strategy to enhance TENG performance by tailoring chemical and morphological contributions of CE. Our strategy can systemically tailor the performance of TENG over a wide range by two factors: chemical ratio, and surface corrugation through the P/F-method. Our comprehensive SPM study combined with FTIR not only shows that the CE variation can be derived from the  $E_{diss}$ -leveraged thermion emission, which is differently contributed to from two competing factors, depending on the PDMS conditions. When the roughness-induced energy dissipation marginally varies, due to the smooth surface, the work function-tailored potential barrier height predominantly impacts CE. However, if the surface is sufficiently corrugated, for example, by P/F-method, the surface-enhanced  $E_{diss}$  plays a pivotal role in overcoming the barrier height more easily. Our analysis reveals the extent to which  $E_{\rm diss}$  can be increased simply through the corrugation effect, without considerable coupling with other chemical/electrical properties. Furthermore, due to its indirect surface treatment method, our approach can be easily integrated with other surface treatment to enhance CE, as exemplified in the case of O<sub>2</sub>-plasma.

#### Author contributions

J. H. K.: conceptualization(equal), methodology(equal), formal analysis(equal), investigation(equal), data collection(equal), visualization(equal), and writing. D. W. J.: methodology (equal), formal analysis(equal), investigation(equal), data collection(equal), visualization(equal), and writing. J. H. J.: methodology, investigation, data collection, visualization. D. K.: methodology, investigation, data collection, visualization. H. Y.: methodology, investigation. H. C.: methodology, investigation. H. I.: review and editing. J. Y. P.: project administration (equal), formal analysis, writing – review & editing (equal); resources (equal). J. H. J.: conceptualization (equal); formal analysis, writing – review & editing (equal); project administration (equal); resources (equal).

#### Data availability

The data supporting this article have been included as part of the ESI.<sup>†</sup> Additional data related to this paper may be requested from the authors.

### Conflicts of interest

There are no conflicts to declare.

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