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SPECIAL TOPIC: Flexible Materials

The overlooked key factors for accurate determination of polymer triboelectric series

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Triboelectrification (TE), also known as contact electrification (CE), refers to the phenomenon where two materials acquire opposite charges upon contact and subsequent separation due to charge transfer [1]. The electrostatic accumulation can lead to safety hazards, equipment damage, product defects, and even health risks, making it crucial to mitigate TE in certain industrial and daily scenarios [2]. While TE also enables valuable technological applications [3], such as electrostatic precipitation, spraying, printing, filtration, separation, and contact-electro-catalysis [4,5]. Furthermore, in 2012, Zhong Lin Wang invented the triboelectric nanogenerator (TENG), based on coupling TE effect with electrostatic induction. The triboelectric components of TENG primarily consist of polymers. When two polymers with divergent triboelectric polarities come into contact and then separate under external forces, electrons transfer between them, generating electric current in external circuit and thereby producing electrical energy. This mechanism allows TENGs to efficiently harvest dispersed, high-entropy mechanical energy from the environment or human motion into electrical energy, demonstrating broad potential in energy harvesting, implantable or wearable electronics, the Internet of Things, and self-powered sensing systems (Fig. 1) [5–9]. Establishing reliable triboelectric series for polymers is essential for elucidating the fundamental mechanisms of their triboelectrification, and providing critical guidance for material selection and design to better control over triboelectric effects, such as minimizing electrostatic hazards (e.g., preventing fires) or enhancing electrical output for high-performance TENGs [10]. Using the TENG output analysis enables accurate and direct quantification of transferred charge between different testing polymer films and the reference material (e.g., polytetrafluoroethylene (PTFE) film) during periodic contact-separation. The triboelectric series of these polymer films can be determined by ranking the magnitudes of the transferred charge. In this method, a reliable triboelectric series requires accuracy and consistency of some physical and chemical properties for different testing polymer films, a criterion often overlooked and unmet in current triboelectric polarity studies, leading to questionable accuracy. This perspective analyzes accuracy or consistency challenges in triboelectric polymers (especially biodegradable polymers) films, covering purity, surface roughness, and mass/thickness, offering valuable insights for researchers and engineers engaged in triboelectric polymer research and applications.

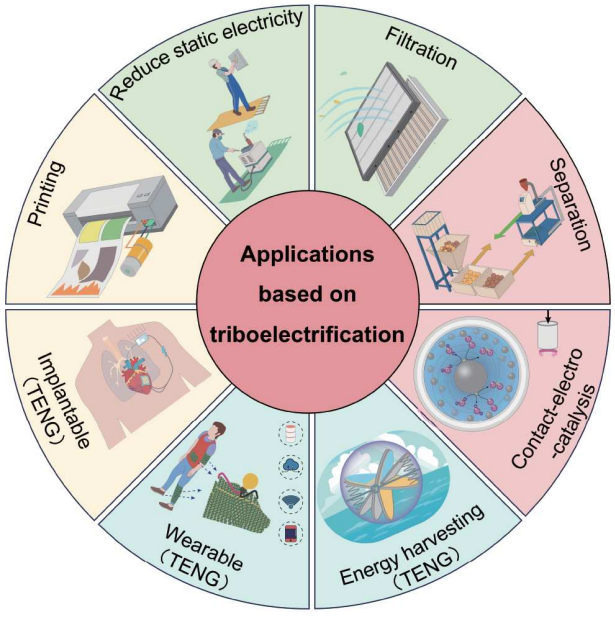

Figure 1 Applications based on triboelectrification effect, including reducing static electricity, electrostatic filtration, separation, printing, contact-electro-catalysis. Reproduced with permission from Ref. [5]. Copyright 2024, Springer Nature. TENGs used in energy harvesting. Reproduced with permission from Ref. [7]. Copyright 2021, Elsevier. Implantable, Reproduced with permission from Ref. [8]. Copyright 2019, Springer Nature. Wearable electronics. Reproduced with permission from Ref. [9]. Copyright 2024, Royal Society of Chemistry.

Charge carriers in CE process can be electrons, ions, or small charged material fragments. Recent experimental evidence strongly suggests that electron transfer driven by the overlap of electron clouds during the CE process plays a dominant role [4]. The triboelectric series of polymers can be established based on their relative tendencies of their triboelectric positive or negative polarity, which corresponds to their strength of electron donating (ED) or electron withdrawing (EW) ability [11,12].

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Since polymers consist of diverse chemical groups with varying ED or EW abilities, the triboelectric polarity and density of these groups fundamentally determine a polymer's triboelectric properties. Despite the first triboelectric series was established over 200 years ago, variations exist among sequences reported by different researchers [10,13,14]. Experimental conditions (e.g., humidity, temperature) can cause some deviations because triboelectric series is determined experimentally [15,16]. However, based on a comprehensive literature investigation and experimental data, we conclude that the accuracy and consistency of properties for tested polymers remains the primary factor influencing the accuracy of triboelectric series when using charge transfer measurement methods (the TENG output analysis), in which different tested polymers films are periodically contacted with and separated from the reference material (e.g., PTFE film) under the drive of linear motor, and using a digital oscilloscope and an electrometer to characterize the short-circuit charge (Fig. 2a, b) [10,17]. We observe that many studies on triboelectric polarity comparison fail to ensure uniformity in these properties [18–23], leading to questionable accuracy. This perspective mainly discusses the relationship between the consistency of properties and the accuracy of triboelectric series for polymers, especially for biodegradable polymers. The properties mentioned include the purity (Fig. 2c), surface roughness (Fig. 2d), mass or thickness (Fig. 2e) of the tested triboelectric polymer films.

Consistency in purity. Sufficient purity is essential for obtaining accurate triboelectric series. During the test, polymers are prepared into thin films. However, some polymers require the addition of organic or inorganic molecules to enhance film formation or improve mechanical properties. The inclusion of these additives results in the triboelectric polarity measurements reflecting the properties of the composite films, rather than those of the pure polymers, a detail often overlooked in some reports. For instance, chitosan tends to be dissolved in acidic aqueous solutions. Consequently, the reported chitosan films inevitably retain residual acid substances (e.g., acetic acid or hydrochloric acid) unless additional acid removal steps are implemented [18,21,24]. Zheng *et al.* [18] reported a triboelectric series (from positive to negative) as follows: glass fiber fabric, starch film, chitosan/starch composite film, chitosan film, aluminum foil. However, the presence of acetic acid in the chitosan film prevents the accurate determination of chitosan's intrinsic triboelectric polarity [24]. This interference also compromises the accurate comparison of the ED abilities between amine and hydroxyl groups, because the main difference for

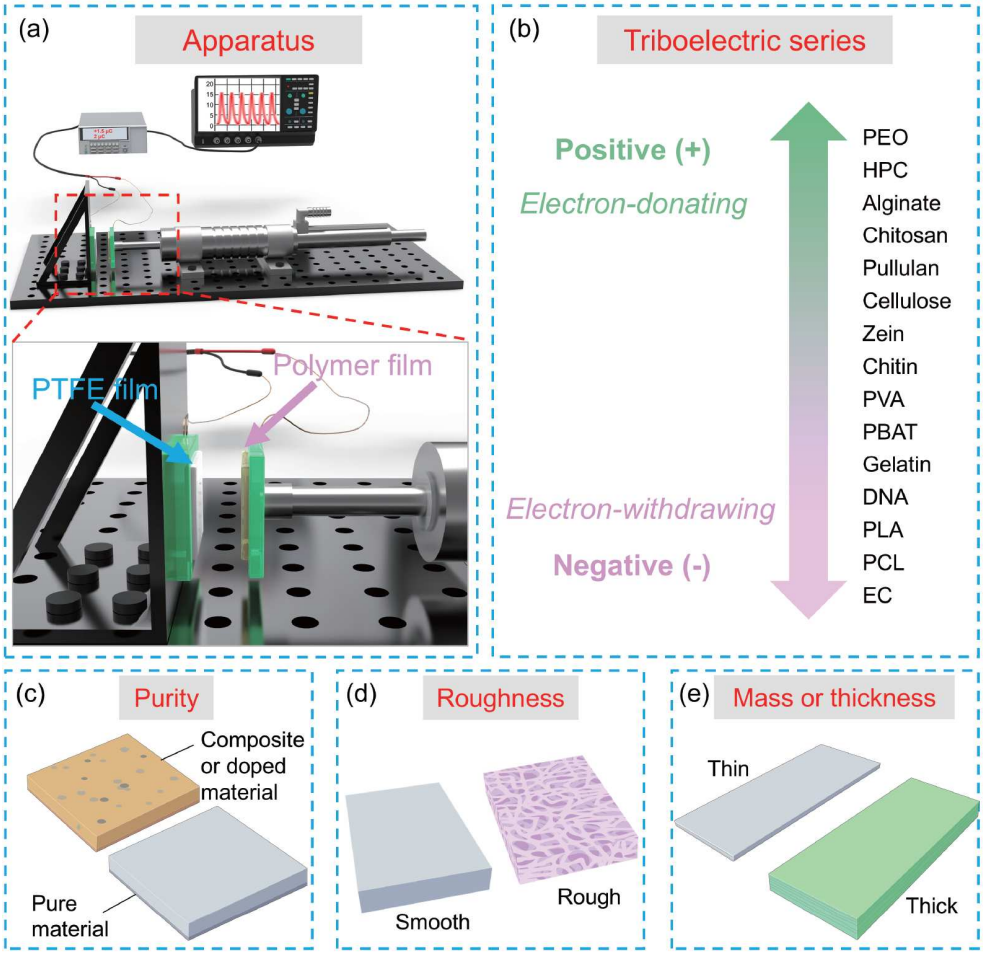

Figure 2 Consistency of polymer properties in triboelectric series experiments. (a) Apparatus for testing the triboelectric series using charge transfer measurement methods. Reproduced with permission from Ref. [17]. Copyright 2024, Elsevier. (b) A triboelectric series of some degradable polymers reported. Reproduced with permission from Ref. [10]. Copyright 2023, Elsevier. Inconsistency of the properties such as (c) purity, (d) surface roughness, and (e) mass or thickness for tested triboelectric polymer films leads to questionable accuracy of their triboelectric polarities.

chitosan is that one of the hydroxyl groups is replaced by an amine group in its molecular repeating unit compared with starch. In addition, glycerol, utilized as a plasticizer, can improve the flexibility of some polymers when incorporated. Jiang *et al.* [19] prepared five distinct protein composite films containing 30 wt% glycerol, and subsequently compared their triboelectric polarities with other polymers, including nylon-11, wool, polydimethylsiloxane (PDMS), and PTFE. However, the introduction of glycerol resulted in a discernible divergence in the measured triboelectric polarities of the protein composite films compared to those of pure protein films. Furthermore, another crucial factor concerns the moisture content requirements for accurate triboelectric evaluation [20]. Since different polymers possess distinct hydrophilicity, rigorous drying protocols are essential prior to testing. This is particularly critical for hygroscopic polymers evaluated under high-humidity conditions. Insufficient drying leads to measurement artifacts, where the recorded triboelectric polarity reflects those of a water-containing polymer composite film rather than the true characteristics of the pure polymer.

Consistency in surface roughness. For TENGs or triboelectric tests, a larger contact area between triboelectric polymers leads to greater charge transfer and higher electrical output [25]. Hence, to obtain accurate triboelectric series assessment, different polymer films should be prepared with identical shape and contact area, and then subjected to periodic contact-separation with the reference PTFE film. Besides, achieving similar surface roughness across samples is crucial for obtaining reliable triboelectric series, because disparities in surface roughness lead to variations in their actual or effective contact areas with the PTFE film, even when their macroscopic areas are identical. Regrettably, some research neglects to account for the impact of surface roughness inconsistencies. For instance, when comparing the triboelectric positive polarity of porous cellulose and chitosan aerogel film, differences in specific surface area (surface roughness) result in unequal effective contact areas. This inconsistency may introduce errors in assessing the true triboelectric series of chitosan and cellulose, as well as in quantifying the contributions of amine and hydroxyl groups to positive ability [21]. Another study compared the triboelectric series of poly(lactic-*co*-glycolic acid) (PLGA), chitin, polylactic acid (PLA), copy paper, rice paper, and silk film. Despite the standardization of film area and thickness across all samples, PLGA and PLA were synthesized via solution-drying, whereas copy paper, rice paper, and silk films were utilized as received, without further processing. These disparate fabrication methods inevitably introduce variations in surface roughness (and thus effective contact area), potentially compromising the accuracy of their triboelectric series [22].

Consistency in mass or thickness. Theoretically, the triboelectric polarity of polymers is determined by their inherent ED or EW ability, independent of their mass or thickness. However, for the testing of triboelectric series, consistency in the mass or thickness is essential, because the tested polymer films and the reference polymer (PTFE) can function as a pair of triboelectric materials of a TENG, and extensive studies have demonstrated that variations in thickness of triboelectric films significantly influence the amount of transferred charge of TENGs [25]. For most biodegradable polymers, composed primarily of C, H, O, or C, H, O, N elements, the density is relatively uniform, typically around 1.2–1.3 g/cm^3 [14,26]. Given this similarity in density, maintaining uniform mass across different polymer films inherently ensures comparable thickness. Therefore, when dealing with these polymers, standardization in film mass or thickness is crucial for ensuring the accuracy of triboelectric series and the proper evaluation of relative ED or EW tendencies of the chemical groups. However, this aspect has not received widespread attention. For instance, the triboelectric series of polyesters [14,27], such as PLA, poly (butylene succinate) (PBS), and polycaprolactone (PCL), varies across different studies [10,23]. The three polyesters share similar molecular structure, consisting of repeating alkane segments and ester groups. The key structure distinction lies in the density of ester groups along the backbone: PLA features one ester group per 2 carbon atoms in the main chain, PBS has one per 4, and PCL has one per 6. Standardization of film mass (or thickness) during triboelectric testing would establish a well-defined quantitative relationship between ester (or alkane) group density and their relative triboelectric polarities [10]. This controlled experimental approach would not only clarify the relative ED or EW abilities of these polyesters, but also provide more reliable insights into how specific chemical group density influences performance, thereby resolving current inconsistencies in reported data. Furthermore, when ranking more kinds of polymers, consistent films mass or thickness can also significantly improve the accuracy in analyzing the relative triboelectric polarity or ED or EW abilities of both the polymers and their groups [10].

In conclusion, the accuracy and reproducibility of the triboelectric series have long been key concerns in scientific research. The relative triboelectric polarities of polymers are fundamentally determined by the types and density of their chemical groups exhibiting varying ED or EW abilities. We conclude that the accuracy and consistency of properties, including material purity, surface roughness, and mass (or thickness) for tested polymer films are crucial for establishing reliable triboelectric series. Establishing reliable triboelectric series is essential for elucidating the fundamental mechanisms of their triboelectrification, and providing critical guidance for material selection and design to better control over triboelectric effects.

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Conflict of interest The authors declare that they have no conflict of interest.

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精确测定高分子摩擦电序中被忽视的关键因素

孟宏宇[1*], 盖延松[1], 白源[1], 于巧[1], 刘卓[1], 李舟[1,2,3*]

摘要 研究和比较高分子的摩擦起电性能，并建立准确的高分子摩擦电序，不仅有助于研究人员深入认识其摩擦起电的潜在机理，而且对于通过合理选择或设计摩擦电高分子来更加可控调节摩擦起电效应具有重要意义．建立准确的摩擦电序的必要条件是高分子理化性能的准确性和一致性，而这在很多相关研究中被忽视，导致报道的摩擦电序的准确性有待进一步探讨．本论文对摩擦电序测试中所研究的摩擦电高分子材料，尤其可降解高分子材料的理化性能一致性对摩擦电序准确性的影响进行了探讨，包括材料纯度、表面粗糙度、质量(或厚度)的一致性等．本论文有望对从事高分子摩擦电研究及相关应用方面的研究人员提供有益参考．