Designing Non-charging Surfaces from Non-conductive Polymers

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Contact electrification is a phenomenon in which solid surfaces become charged when they are brought into contact and are then separated. This phenomenon is useful in a number of applications, such as electrophotography, particle separation, and the recently developed triboelectric nanogenerators for harvesting mechanical energy.[1–4] On the other hand, the consequences of this phenomenon can also be undesirable, and, at times, hazardous. For example, charge accumulated on solid surfaces by contact electrification can cause slight annoyances in many everyday activities, such as the sticking of clothes onto each other while drying, the experience of a shock when one touches a doorknob in dry weather, and the adhesion of dust particles on surfaces (e.g., on screens of computers due to attractive electrostatic force). More importantly, electrical discharges (e.g., sparks) due to the accumulated charge may potentially result in dangerous situations such as the explosion of flammable gases, dusts, and organic liquids (e.g., during the fueling of vehicles). Electrical discharges are also responsible for the damage of equipment; these damages are reported to cost the electronic industry billions of dollars per year.[5] In many other types of industries, the accumulation of charge on solid surfaces can also make processes less efficient. For example, charged particles that adhere onto the walls of reactor vessels can hinder effective heat transfer; charged powder in the pharmaceutical industry can lead to non-uniform blending, thus resulting in non-uniform dosages of the products.[6] Therefore, it is important to eliminate charge on solid surfaces due to contact electrification.

In particular, it is especially important to develop ways to eliminate charge due to contact electrification of polymeric materials. Although polymers have a wide range of applications, they are especially problematic: they have the natural tendency to charge highly on contact, and are typically insulating. Conventionally, the approach to eliminate static charge involves making the polymers conductive in order to dissipate charge away from their surfaces. Methods include fabricating polymeric composite materials (with a conductive material),[7,8] doping (e.g., doped conjugated polymers),[9,10] or adding antistatic agents. A wide variety of antistatic agents have been developed,[11,12] such as powdered metals, carbon nanotubes, and graphene.[7,8,13] In general, these methods have their disadvantages and limitations. For example, modifying the materials (e.g., through doping or adding antistatic agents) may change their properties in unfavorable ways (e.g., by reducing their mechanical strength, or changing their color). They may also become incompatible with certain applications; for example, the materials may become corrosive, toxic, or instable to heat.[12,14] In order to preserve the properties of the materials, one approach is to coat their surfaces with a layer of conductive film.[11] Other methods include coating surfaces with charged molecules (e.g., self-assembled monolayer of ionic molecules,[15] and multilayers of polyelectrolytes[16]). Chemical modification and coatings of the surface, however, are susceptible to degradation through wear and tear, and may require additional considerations (e.g., the bonding of the coat and the influence of the surface modification on the bulk material).[11,17–21] Another recent study proposed to dope materials with radical-scavenging molecules.[5]

In this study, we introduce a general strategy of fabricating polymers that resist charging due to contact electrification. First, we propose to refer to the triboelectric series (see **Figure 1** for an example)[1] for selecting the molecules for polymerization. This series is an empirically determined list of materials ranked according to their tendency to charge upon contact with other materials. In general, a material has the tendency to charge positively against another material ranked lower than it, and vice versa. Hence, a material ranked close to the top of the series has a tendency to charge positively against many other materials (**Figure 2**a, scheme on the left), and a material ranked close to the bottom has a tendency to charge negatively against many other materials (Figure 2a, scheme on the right). Here, we show that by copolymerizing an appropriate proportion of molecules that tend to charge positively with molecules that tend to charge negatively, we can fabricate a surface that resists charging against another material (Figure 2a, scheme in the middle). This strategy is fundamentally different from methods proposed in previous studies—importantly, it does not rely on increasing the conductivity of the material. In addition, the polymer is structurally homogeneous and does not require further modification (e.g., addition of antistatic agents or dopants).

However, one point to consider is that the triboelectric series includes only a limited set of materials, and is known to be unpredictable.[6,22] One reason is because the mechanisms that cause the separation of charge in contact electrification of insulating materials are still incompletely understood.[1,6] For contacts that involve a metallic surface, previous studies suggest that the separation of charge may be due to the transfer of electrons; this rationalization has led to ways of chemically modifying the surfaces of materials for altering the amount of

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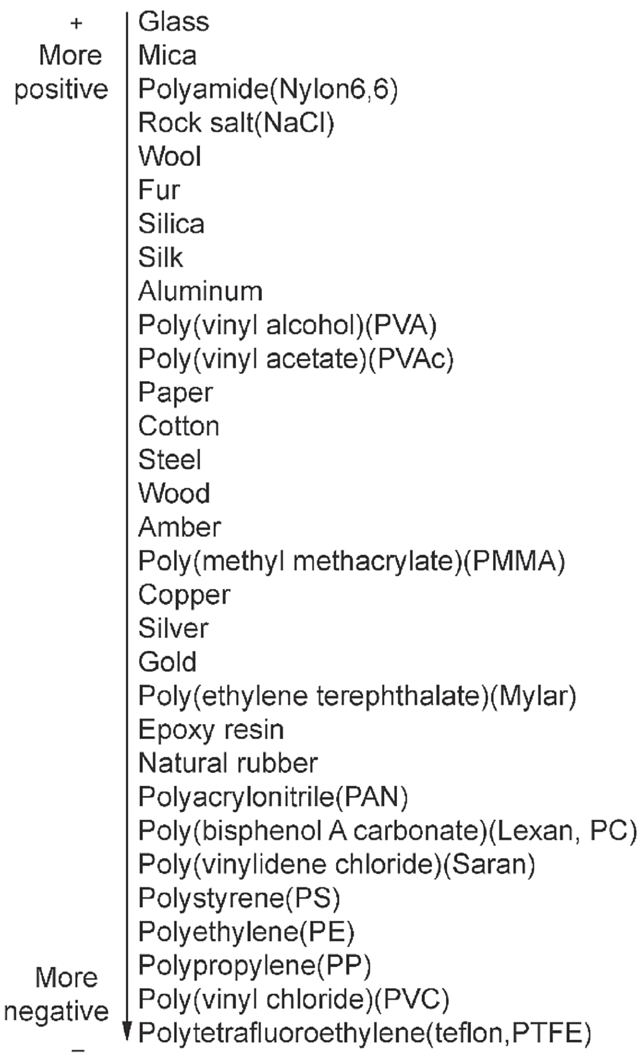

Figure 1. The triboelectric series. Reproduced with permission.[1] Copyright 2008, Wiley-VCH.

charge separated by contact electrification.[23,24] For contacts between an insulating surface and an insulating surface, however, the significances of a number of mechanisms are still under investigation; examples include electron transfer,[25,26] ion transfer,[1] materials transfer,[27,28] and others (e.g., contact de-electrification).[29,30] Therefore, we also propose to rely on a previously reported observation that can complement the triboelectric series: an acidic surface has the tendency to charge negatively, while a basic surface has the tendency to charge positively.[31]

As a demonstration, we chose poly(ethylene glycol) diacrylate (PEGDA) (average molecular weight 575) as the molecule that has the tendency to charge positively (Figure 2b). The reason for choosing PEGDA is that the oxygen atoms in the polyethylene glycol linker might serve as the Lewis basic sites for binding cations present on any contacting surfaces. Transfer of these cations to the surface of PEGDA would charge PEGDA positively. After fabricating the PEGDA polymer (i.e., the 100% PEGDA) by free radical polymerization (see Section 1 in the Supporting Information), we contacted it with other materials.

In a typical experiment, we first discharged the polymerized PEGDA and a reference material using a Zerostat gun. After discharging the materials, we contacted the polymerized PEGDA with the reference material 20 times. The charges of both materials were measured by a Faraday Cup connected to an electrometer (Keithley, model 6514). We repeated the experiment for different types of reference materials as shown in the *x*-axes of **Figure 3**a,b. We found that the polymerized PEGDA charged positively against all these materials (Figure 3a), while the reference materials charged negatively (Figure 3b). Importantly, the polymerized PEGDA was found to charge positively against materials (i.e., nylon, mica, and glass) that are ranked around the top of the triboelectric series shown in Figure 1. Therefore, PEGDA is expected to charge positively against a wide variety of materials.

For the molecule that has the tendency to charge negatively, we chose 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyl methacrylate (HDFDM) (Figure 2c). This molecule was chosen because it contains many fluorine atoms. Therefore, the polymerized HDFDM (PHDFDM) may share similar contact-charging characteristics with poly(tetrafluoroethylene) (PTFE), a polymer that contains a high number of fluorine atoms. Importantly, PTFE ranks at the bottom of the triboelectric series (Figure 1). After contacting PHDFDM (i.e., the 0% PEGDA) against other reference materials, we found that it indeed charged negatively against all other reference materials (Figure 3a), while the other reference materials were found to charge positively (Figure 3b). (Note that because PHDFDM charged positively against PTFE, we did not use PTFE as the reference material.)

After verifying that PEGDA has the tendency to charge positively and PHDFDM has the tendency to charge negatively against many materials, we copolymerized a mixture of PEGDA (50 wt%) and HDFDM (50 wt%) by free radical polymerization (Section 1, Supporting Information). We then contacted the copolymer with other reference materials. Figure 3a compares the charge of the 50:50 wt% copolymer, the 100% PEGDA, and the 100% PHDFDM after the contact with the respective reference materials. For all the reference materials, the charges of the 50:50 wt% copolymer were between that of the 100% PEGDA and 100% PHDFDM. Similarly, the charges of the reference materials in contact with the 50:50 wt% copolymer were also in between those in contact with 100% PEGDA and 100% PHDFDM (Figure 3b). These results suggest that it is possible to tune the charge on the polymeric surface generated by contact electrification by copolymerizing varying amounts of PEGDA versus PHDFDM.

Consequently, we demonstrated that a non-charging copolymer can be fabricated with an appropriate proportion of PEGDA versus HDFDM. Specifically, we contacted copolymers that are composed of different proportions of PEGDA and HDFDM against poly(vinyl chloride) (PVC) as the reference material. Results from the measurements of their charges show that the charge of the polymers varies approximately linearly with increasing proportion of PEGDA. We found that at around 40% PEGDA, both the copolymer and PVC did not charge by contact electrification (Figure 3c). We repeated the experiment for glass as the reference material, and found that the trends are qualitatively similar to the case when PVC is the reference

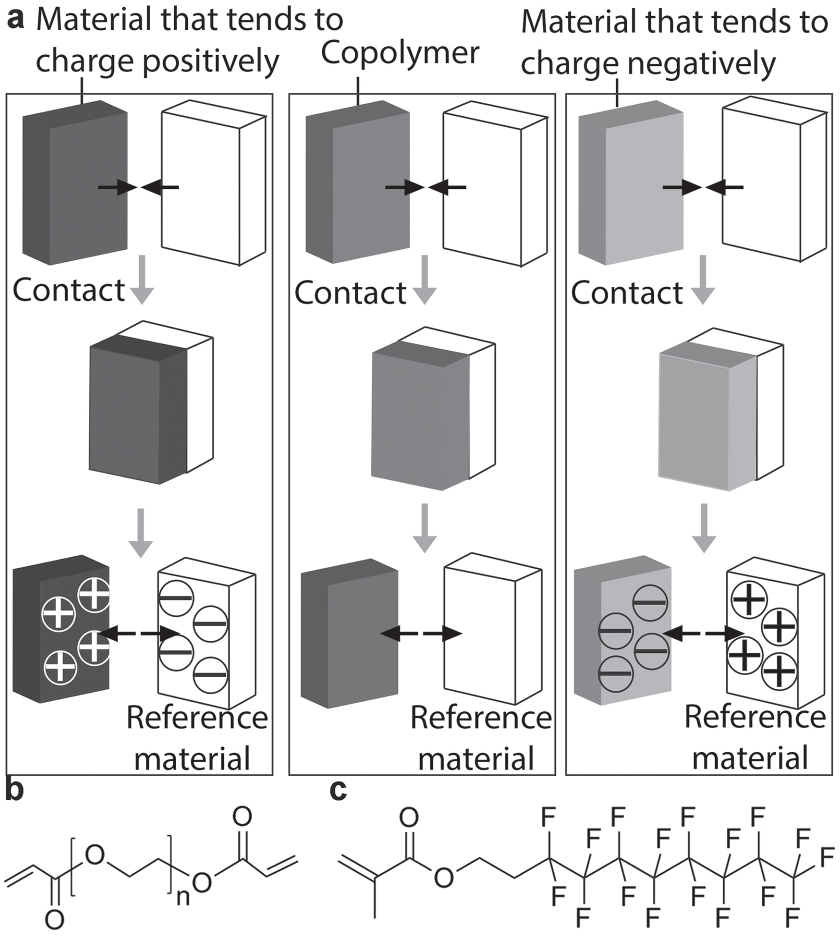

Figure 2. Designing a copolymer that resists charging against a reference material. a) Scheme on the left shows a polymer that has a tendency to charge positively after contact electrification. The scheme on the right shows a polymer that tends to charge negatively. By copolymerizing an appropriate proportion of a molecule that has the tendency to charge positively with a molecule that has the tendency to charge negatively, it is possible to fabricate a copolymer that resists charging against the reference material (scheme in the middle). b) Poly(ethylene glycol) diacrylate (PEGDA), and c) 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-HDFDM. Polymerized PEGDA has the tendency to charge positively, and polymerized HDFDM has the tendency to charge negatively.

material. In this case, both the copolymer and glass did not charge at around 55% PEGDA (Figure 3d). (Section 2 in the Supporting Information compares this non-charging polymer with a polymer added with radical scavenging molecules.)

The percentages of PEGDA mentioned in the previous paragraphs represent the amount of PEGDA that we added in the pre-polymer solution mixture for the fabrication of the solid copolymer. Results from elemental analysis of the solid copolymers showed that the compositions of the solid copolymers are similar to the compositions of the pre-polymer mixtures (Section 3, Supporting Information).

Mechanically, we measured the compressive moduli of the copolymers of different proportions by a material testing system (5500, Instron). The compressive moduli are similar to other types of polymers, such as chlorinated polyethylene and low-density polyethylene (Section 4, Supporting Information).[32] We also measured the surface roughness of the PEGDA–HDFDM copolymers; it seems to be approximately constant for all the compositions of the copolymers studied (Section 5, Supporting Information).

In addition, we determined the bulk conductivities of the polymers by measuring their resistances using an electrometer (model 6514, Keithley). Resistance can be converted to bulk conductivity by the formula, $\sigma = L/(R_B \times A)$, where σ is the conductivity, L is the length, A is the cross-sectional area, and R_B is the resistance of the polymer as measured by the electrometer. In general, the bulk conductivities are on the order of 10^{-9} S cm^{-1} or less for polymers of all proportions of PEGDA (**Figure 4**); hence, the polymers are non-conductive.[9,33] Note that the measurement of the conductivity of the 0% PEGDA is not available because its conductivity is too low for the electrometer to measure.

In order to show that this strategy for fabricating non-charging surfaces is general, we prepared another type of copolymer based on the monomers vinyl acetate and acrylonitrile (Section 6, Supporting Information). The reason for choosing these two monomers is that poly(vinyl acetate) (PVAc) and polyacrylonitrile (PAN) are reasonably far apart in the triboelectric series (Figure 1). After contacting the copolymer with acrylonitrile butadiene styrene (ABS), we found that the results are qualitatively similar to those obtained from the copolymers fabricated from PEGDA and HDFDM. First, the 100% PVAc charged positively, while the 100% PAN charged negatively against ABS (**Figure 5**). This result is in accordance with their rankings in the triboelectric series since PVAc is ranked above ABS and PAN is ranked below ABS (see Figure 1 and ref.[34]). The charge of the 50:50 wt% PVAc–PAN copolymer is in between that of the 100% PVAc and 100% PAN (Figure 5, plot on the left); the charge of ABS in contact with the 50:50 wt% PVAc–PAN copolymer is also in between the charge of ABS in contact with 100% PVAc and 100% PAN (Figure 5, plot on the right). This result suggests that it is possible to fabricate copolymers that resist charging against other materials with appropriate proportions of PVAc versus PAN (except when the reference material is not in between PVAc and PAN in the triboelectric series; see Section 7 in the Supporting Information).

In addition, we measured the conductivity of the polymers. For 100% PVAc, the bulk conductivity is 3.5×10^{-12} S cm^{-1}; for 100% PAN, it is 6.6×10^{-11} S cm^{-1}; and for 50–50 wt% PVAc–PAN, it is 4.7×10^{-11} S cm^{-1}. These results show that the polymers are insulators. The surface resistivities corresponding to these bulk conductivities are 2.4×10^{11} Ω sq^{-1} for the 100% PVAc, 2.5×10^{10} Ω sq^{-1} for the 50:50 wt% PVAc–PAN, and 1.9×10^{10} Ω sq^{-1} for the 100% PAN. These low surface resistivities mean that they do not dissipate charge readily away from their surfaces.[10] The compressive modulus of the 50:50 wt% PVAc–PAN copolymer is measured to be around 2 GPa.

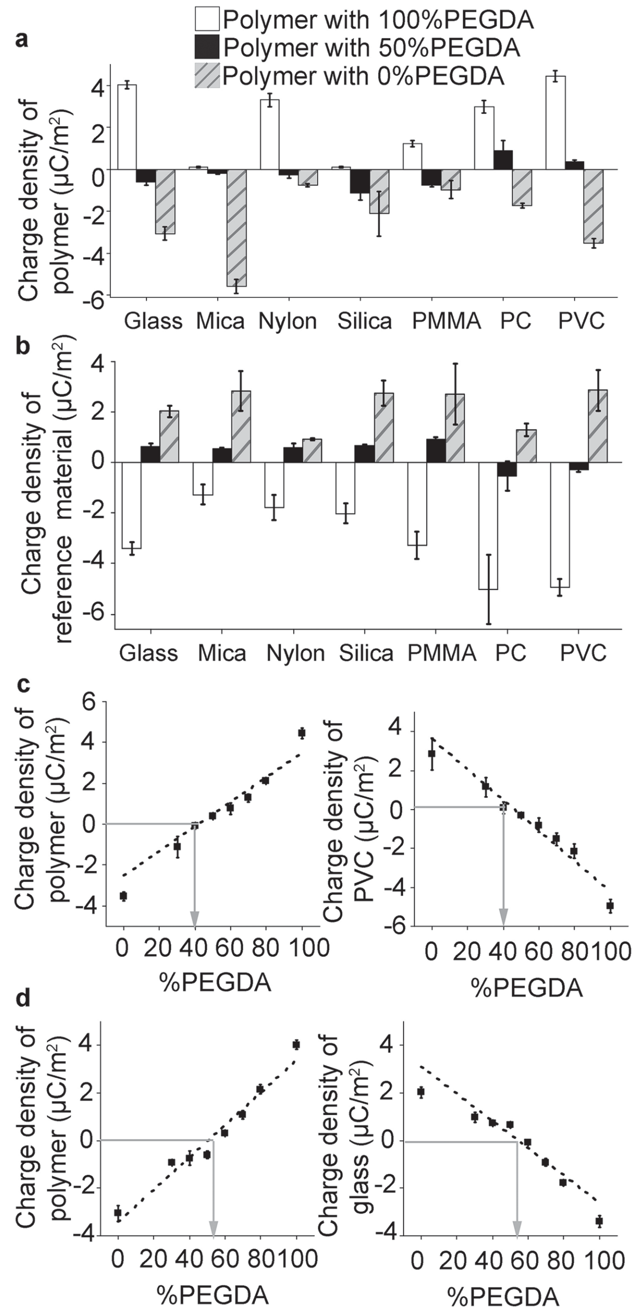

Figure 3. The tendency of the polymers to charge with different proportions of PEGDA and HDFDM. a) Plot showing the charge density (i.e., charge per unit surface area of contact) of the 100% PEGDA polymer, 50%–50 wt% PEGDA–HDFDM copolymer (or 50% PEGDA), and 100% PHDFDM (or 0% PEGDA) polymer after contacting them with the reference materials. The various reference materials used are indicated in the *x*-axis (PMMA: poly(methyl methacrylate), PC: polycarbonate, and PVC: poly(vinyl chloride)). b) Plot showing the charge density of the reference materials as indicated in the *x*-axis after contact with the respective polymers. Contacting the polymers of different proportions of PEGDA versus HDFDM with c) PVC and d) glass. Plot on the left shows the charge density of the polymer and plot on the right shows the charge density of the reference material.

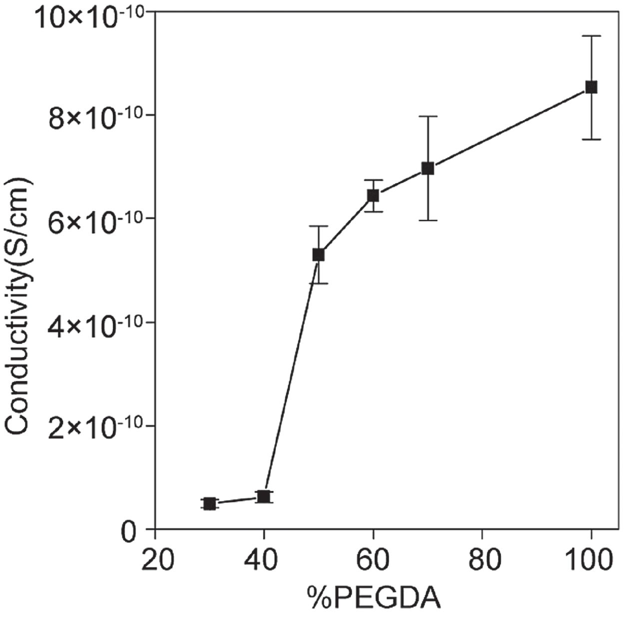

Figure 4. Bulk conductivities of the PEGDA–HDFDM copolymers.

One important property of an antistatic surface is that small (e.g., microscopic) particles do not have the tendency to adhere onto it due to attractive electrostatic force. As mentioned, the adhesion of charged particles on surfaces is highly undesirable in many circumstances in industry, and in our daily lives. In order to demonstrate that the copolymer we fabricated can resist charging against, and prevent the adhesion of, microscopic particles, we brought dextrose powder (diameter ≈100 μm) into contact with polymers composed of different proportions of PEGDA and HDFDM. The reason for choosing to work with dextrose is because it is important for many purposes, including using it as a pharmaceutical excipient.[35] Specifically, we transferred 0.03 g of dextrose powder onto the surface, and agitated the polymer using a vortex mixer (**Figure 6**a). As the powder particles rolled across the surface, both the powder and the surface gained charge due to contact electrification. An indication of the attractive electrostatic force is the number of particles that remained adhered onto the surface after it was turned upside down. If the attractive electrostatic force was negligible, the particles would fall off the surface due to gravity. For the case when the surface was composed of either 100% PEGDA or 100% PHDFDM, a large portion of the surface was covered with the powder (see Figure 6b,c). At 38% PEGDA, only a few particles adhered to the surface. Thus, this surface prevents the adhesion of the microscopic dextrose particles.

In order to demonstrate that the adhesion of the powder is mainly due to the attractive electrostatic force and not due to van der Waals force (or other types of forces), we conducted two types of control experiments as described in Section 8 in the Supporting Information.

When two pieces of polymeric surfaces are rubbed against each other, they typically gain significantly more charge than when they merely come into contact. We demonstrated that the non-charging surface that we fabricated did not charge

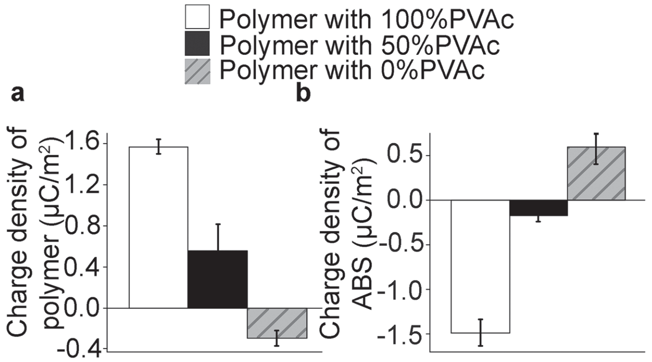

Figure 5. Charge density of the 100% PVAc, 50:50 wt% PVAc–PAN (or 50% PVAc), and 100% PAN (or 0% PVAc) after contacting with the reference material ABS (plot on the left). The plot on the right shows the charge of ABS after the contact.

significantly even after rubbing with another polymeric surface. First, we prepared three types of samples, the 100% PEGDA, 40% PEGDA, and 0% PEGDA, as previously described. After preparation, we discharged these samples and a piece of PVC using a Zerostat gun. Subsequently, we rubbed the samples against the piece of PVC for 40 s using a force of around 3 N (or an equivalent pressure of around 8 kPa, as measured by using the same amount of force on a mass balance). After rubbing, the charges of the 100% PEGDA, 40% PEGDA, and 0% PEGDA were measured to be 7.4 μC m^{-2}, –0.6 μC m^{-2}, and –11.2 μC m^{-2}, respectively. In a separate step, we cut 10 pieces of paper from a fresh piece of A4-sized copy paper. After cutting, each piece of paper had a weight of 1.2 mg, and an average residual charge of around 0.2 μC m^{-2}. The samples were then brought to a distance of around 1.5 mm away from the pieces of paper. **Figure 7**a shows that the positively charged surface of the 100% PEGDA repelled the positively charged pieces of paper away from the surface. For the negatively charged 0% PEGDA, the positively charged pieces of paper were attracted to, and stuck onto, the surface (Figure 7b). However, for the 40% PEGDA, it had no observable effect on the paper (Figure 7c); thus, this experiment demonstrated that the 40% PEGDA did not charge significantly after rubbing against PVC.

In conclusion, we are introducing a general strategy for fabricating polymers that resist charging against another material based on copolymerizing an appropriate proportion of a molecule that has the tendency to charge positively and a molecule that has the tendency to charge negatively. Importantly, these copolymers were determined to be non-conductive—this characteristic is in contrast with conventional methods in which surfaces are made conductive in order to eliminate static charge. These surfaces can prevent the generation of charge entirely for both the contacting surfaces; on the other hand, conductive materials usually first generate charge after the contact, then dissipate it through conduction. The copolymers thus fabricated are structurally homogeneous, and do not require further modification or addition of other substances (e.g., antistatic agents or dopants).

Because PEGDA ranks around the top and PHDFDM ranks close to the bottom of the triboelectric series, it is possible to fabricate PEGDA–HDFDM copolymers that resist charging

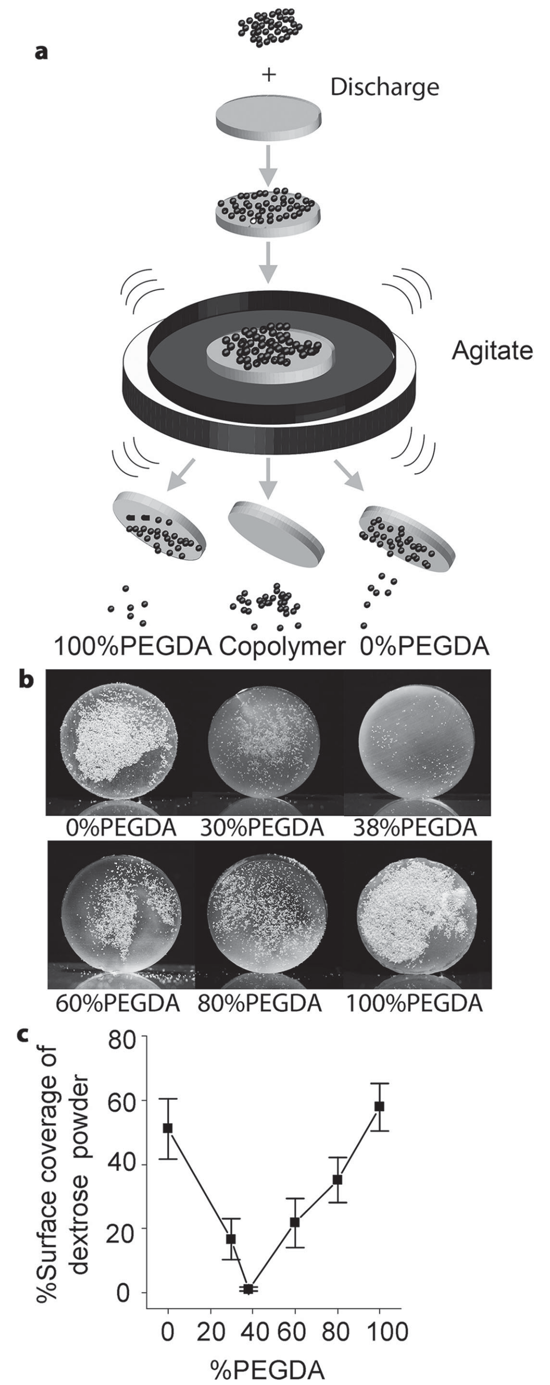

Figure 6. Resisting the adhesion of microscopic particles on the non-charging surface. a) Scheme illustrating the procedure of the experiment in which dextrose powder was agitated on the surface of a polymer composed of a predetermined proportion of PEGDA and HDFDM. The polymer was first discharged by a Zerostat gun and placed on a Petri dish. Dextrose powder was added onto the surface, and the whole Petri dish was agitated by a vortex mixer. After turning the polymer upside down, the powder might remain on the polymeric surface due to attractive electrostatic forces. The experiment was repeated once by adding dextrose powder, agitating the dish, and turning the polymer upside down again. b) Side views of the surfaces of different polymeric compositions adhering to different amounts of the powder after agitation. c) Plot of the surface coverage of the powder that adhered onto the surfaces of different compositions.

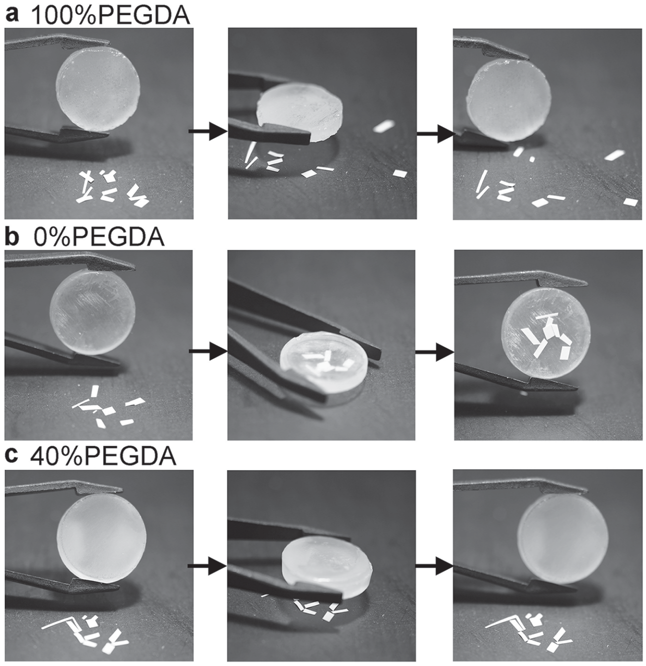

Figure 7. Copolymer resists charging even after rubbing. The samples (100% PEGDA, 0% PEGDA, and 40% PEGDA) were first discharged and were then rubbed against a piece of PVC. After rubbing, the 100% PEGDA charged positively, while the 0% PEGDA charged negatively. The charge of the 40% PEGDA was negligible. The samples were brought close (1.5 mm) to pieces of paper, which had slight residual positive charges. a) For the 100% PEGDA, the pieces of paper repelled away from the surface. b) For the 0% PEGDA, the pieces of paper were attracted to the surface, and stuck onto it. c) For the 40% PEGDA, the charge on it was too small to have any observable effect on the pieces of paper.

against a wide variety of materials. In addition, the strategy is general: it is possible to select different sets of molecules (i.e., besides the combination of PEGDA and HDFDM) for fabricating the non-charging surface, as long as one of the monomers has the tendency to charge positively and the other has the tendency to charge negatively, against the reference material. We demonstrated that the method is general by fabricating a copolymer based on the vinyl acetate and acrylonitrile monomers. It may also potentially be possible to select two types of monomers which are close to each other in the triboelectric series (after polymerization), as long as the reference material is in between them. Therefore, this strategy opens up a new avenue for designing a wide range of copolymers that can resist charging against other materials. A wide range of choices is necessary because of the wide diversity of applications related to non-charging surfaces; in particular, different applications typically require different material properties of the polymer (e.g., chemical compatibility, mechanical property, and degree of transparency).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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