



Heterogeneity of surface potential in contact electrification under ambient conditions: A comparison of pre- and post-contact states

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ABSTRACT

We measured the pattern of charging by contact electrification, following contact between a polydimethylsiloxane (PDMS) stamp and a glass substrate with gold electrodes. We used scanning Kelvin probe microscopy to map the surface potential at the same regions before and after contact, allowing a point-by-point comparison. After contact, the mean surface potential of the glass shifted by 360 mV and micron-scale heterogeneity appeared with a magnitude of ~100 mV. The gold electrodes showed charge transfer but no discernible heterogeneity. These results show that contact electrification causes heterogeneity of surface potential even on non-polymer surfaces such as glass under ambient conditions.

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1. Introduction

The transfer of charges between two surfaces that are brought into contact and then separated is a familiar everyday occurrence. An example of this process, which is known as contact electrification, is rubbing a balloon on one's head and observing that hair clings to the balloon. Contact electrification is prevalent in many technological applications, including laser printing and photocopying [1–5] and triboelectric generators [6,7]. It is also perceived to be the cause for adhesion of granular media [8,9], which can lead to explosions in grain silos [10] as well as equipment malfunction in space probes traversing the surfaces of planets and moons [11,12]. Despite centuries of investigation and the frequent appearance of contact electrification in technology, the process is still poorly understood [4,13].

The most basic characterization of contact electrification is the triboelectric series, which ranks dielectric materials according to their tendency to charge positively or negatively [14,15]. The ranking in the triboelectric series is not universal but instead depends on atmospheric conditions such as humidity and surface cleanliness [9,13,16,17]. As an example that will be relevant for this article, rubber elastomer (polydimethylsiloxane, PDMS) tends to

adopt a negative charge of approximately $15 \times 10^{-5} \text{ C/m}^2$ after contact with Au, Al, Cr, steel, or brass [18]. PDMS is a useful material for studying contact electrification owing to its ability to deform and hence make conformal contact with the other substrate so that the true contact area is known.

Since the 1980s, scanning-probe methods have been used to map the spatial dependence of the charge developed by contact electrification. Scanning probe studies have used either electrostatic force microscopy (EFM) or scanning Kelvin-probe force microscopy (KPFM). These studies have shown that contact electrification results in spatially heterogeneous charge, and even bipolar charge patterns. For instance, bringing a metallized atomic force microscope (AFM) tip into contact with a polymer film results in a charge dipole, consisting of micron-scale domains of positive and negative net charge [19,20]. Rubbing a polymer film with another polymer film also leads to bipolar charge distributions with characteristic sizes that can extend from μm scale [21] to cm scale [22]. More recently, Baytekin et al. showed that contact between two macroscopic polymer surfaces under ambient conditions (without rubbing) leads to a random pattern of net positive and negative regions, which they termed charge mosaic [23]. Most surprisingly, these authors reported that the charge mosaic had two characteristic length scales of $0.45 \mu\text{m}$ and 45 nm and a characteristic magnitude of 250 mV. The emergence of charge mosaic indicates that average charge (as described by the triboelectric series) misses much of the important phenomenology. To understand the mechanisms of contact electrification, we clearly need further measurements of the spatial array of charge on different

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substrates with controlled initial conditions [24,25].

Remaining questions about contact electrification that stem from the prior scanning-probe results include: does non-uniform surface charging occur on non-polymer surfaces (*i.e.*, other than PDMS, PC, PTFE, acrylic or other polymer surface that have been studied so far)? Specifically, does the heterogeneity arise on metallic surfaces or on inorganic insulating surfaces such as glass? How do specific locations on the surface change as a result of contact electrification and does the initial surface electric potential alter the result?

In this article, we report on the use of scanning Kelvin probe force microscopy (KPFM) to measure the change in surface electric potential following contact between a polydimethylsiloxane (PDMS) stamp and a glass substrate. We demonstrate an experimental technique that brings two advantages to the use of scanning-probe methods to study contact electrification. First, our sample consists of a region of glass bounded by two 5- μm -wide grounded gold electrodes; this allows us to monitor separately the response of the gold, which acts as a control surface. This geometry was used previously to probe surface potential induced by an applied bias [26] but here we keep the electrodes grounded. Second, this approach allows us to scan the same region of the sample immediately before and after contact with the PDMS, thereby providing a point-by-point comparison. By comparing the same region before and after contact, we can isolate the changes coming from the contact itself. Our results show that the gold electrodes develop a surface potential of approximately +260 mV following contact with the PDMS, and we discern no heterogeneous (mosaic) pattern. The glass region develops a positive surface potential of, on average, 360 mV. Moreover, the glass exhibits a much broader range of potential values both before and after contact with the PDMS. Following the charging process, we identify heterogeneity at the μm scale on glass. The results show that heterogeneity of the surface potential (and hence heterogeneity of the transferred charge) appears on glass surfaces but is not detected on gold. We anticipate that future studies that make a point-by-point comparison of pre- and post-contact surface potentials will be very helpful in revealing the microscopic mechanisms of contact electrification. Aside from the importance in understanding contact electrification in general, the results also show that microcontact printing on glass surfaces using PDMS stamps may lead to microscopic charge heterogeneity.

2. Materials and methods

The glass substrate (ABTECH Scientific, Inc., IAME 0504.3 Series) has four gold parallel electrodes that are 5 μm wide and spaced 5 μm apart on top of Schott D263 borosilicate glass. The sample is illustrated schematically in Fig. 1A. The electrodes were approximately 400 nm thick. The electrodes were held at ground by the AFM controller, providing a built-in check of the voltage magnitudes and ruling out the possibility that the AFM tip might accumulate contaminant or charge, which would appear as a sudden change in the potential of the gold electrodes. In this way, variations of the measured surface potential were attributed to the surface itself and not to the tip.

The stamp used to induce contact electrification was made of polydimethylsiloxane (PDMS). PDMS is useful for stamping owing to its flexibility and ability to maximize its contact area with the substrate [18,23]. We prepared the PDMS using Sylgard 184 elastomer base and Sylgard 184 elastomer curing agent in a 10:1 w/w ratio. The mixture was degassed in a vacuum and cured in a conical mold at 40 $^{\circ}\text{C}$ for 24 h. The stamp was made large enough to grip by hand. PDMS was kept in the mold, never making contact with air until immediately before use.

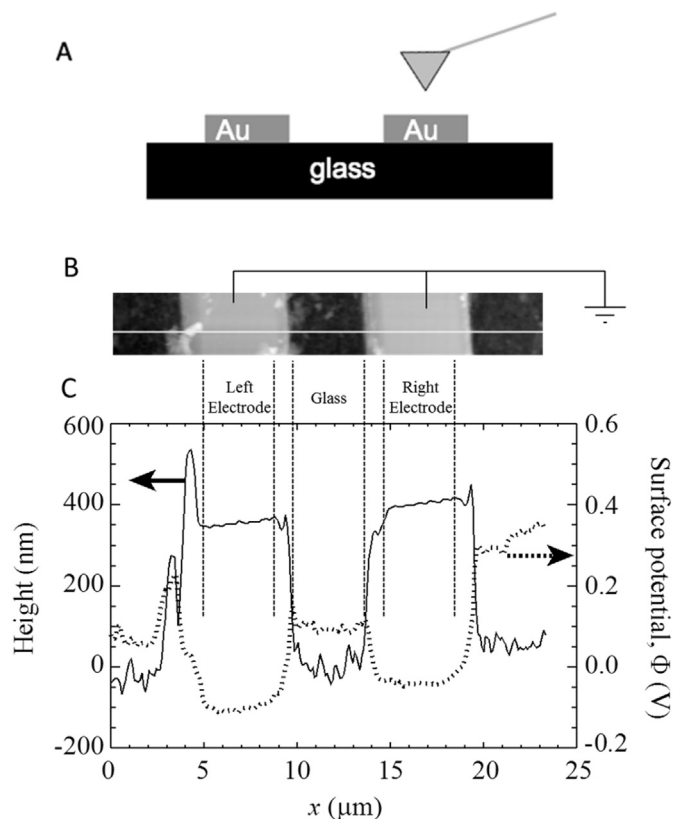


Fig. 1. (A) Schematic of the AFM tip interacting with the glass-electrode sample. (B) Image of the height of a region 240×35 pixels ($23.4 \times 3.4 \mu\text{m}$). (C) Line plot of height (left axis, solid line) and measured surface potential (right axis, dashed line) prior to contact, along the line shown in white in (B). The difference in heights of the electrodes and glass is clearly visible, as are the differences in surface potentials. The regions defined as left and right electrodes and glass are labeled by the vertical dashed lines.

Our procedure was to clean the sample first by gently rinsing with acetone for 2 min followed by gently rinsing with methanol for 1 min. The sample was left to air dry for 24 h. Following this process, we scanned the sample using KPFM (as described below). We then made contact with the sample using the PDMS stamp, and then scanned the sample again. To minimize delay between the contact and the second scanning, we left the sample on the AFM stage and applied the PDMS stamp by hand. (Our first stamping process consisted of using a micrometer to press the PDMS onto the glass substrate, but due to limited space in the enclosure we found this to be difficult.) The delay between contact and the completion of the second scan was approximately 10 min. During the contact/stamping process, the PDMS was pressed gently against the glass once and held in contact for 3 s. We found this method was sufficient to observe charge transfer. Previously, Cottrell found that varying the contact time for PDMS on gold from 3s–24 h had small effect on the charge density [18]; Baytekin et al. also found consistent results for contact times 2 s–1.5 h for PDMS/PDMS contact [23]. After contact we remounted the AFM scan head, making sure to image the same location on the glass surface. Small topographical features on the glass made it possible to identify the same location as the before-contact scans.

All topography and surface potential images were acquired with an atomic force microscope (Asylum Research MFP-3D). The sample was mounted on a grounded stage and the sample electrodes were connected to 0 V. Surface potential (Φ) images were taken using a platinum-coated tip (App Nano ANSCM-PT) with a nominal resonant frequency of 70 kHz and spring constant 3 N/m. Scanning

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