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Transient aspects of plasma luminescence induced by triboelectrification of polymers

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ABSTRACT

Transient electric gas discharges that occur around sliding interfaces during contact electrification of polymers were studied at millisecond timescales and with micrometre resolution. Deduced vibrational temperatures indicate cold plasma resulting from positive corona discharge. At millisecond timescales, previously unseen rapid discharge events are observed, and modelling suggests that these result from streamer development, triggered by electron emission from the polymer surface. Those which occur over a period of several seconds are shown to be caused by competition between charge generation and the formation of polymer films. The findings explain the interplay between charging and plasma generation and their dependence on wear processes.

1. Introduction

Tribocharging, or triboelectrification, is defined as the electrical charging of contacting insulators that occurs when they are rubbed together or brought in contact and then separated. The underlying physics of tribocharging is only now becoming understood [1], despite documents of its occurrence dating back thousands of years. Emergence of new technologies such as distributed energy harvesting by triboelectric nanogenerators [2] [3], nano-coulomb molecular mass spectrometry [4] and triboelectric X-ray source [5] as well as rapid development of the existing technologies, *e.g.* xerography [6], laser printing [7], electrostatic separations [8], and so on, have led to renewed interest in deepening our knowledge about the relevant phenomena of charge generation and dissipation [9]. Although the details of charging mechanisms are still incompletely understood, many efforts have been made on surface engineering of polytetrafluoroethylene (PTFE) and other polymer membranes to enhance the triboelectric output and to increase the surface charge density, which is closely approaching the maximum value that is constrained by air breakdown [9–11]. What has made this field challenging is that charging is not only an electrostatics problem, i.e. charge generation and dissipation, but in reality involves tribological processes, which are system dependent.

Early research led to the construction of a triboelectric series which ranks materials based on charge accumulation ([*e.g.* Refs. [12–14]]). However, the exact order of this series is inconsistent [15,16], and even rubbing together identical materials can produce charge [17,18] or de-electrification [19]. Furthermore, whereas for metals electron transfer between valence levels is understood to be the dominant mechanism [20], there has been uncertainty regarding charge transfer for insulating materials such as polymers, which by definition have no free charge carriers. Charge carriers in these contacts were initially assumed to be electrons [21–23] and then evidence suggested molecular or atomic ions [15,24–26], with the latter now being the more accepted view.

During the last decade, the picture of how both micro- and nanoscale events are responsible for tribocharging and inconsistencies in triboelectric series has become clearer, thanks in part to the introduction of Kelvin Probe Force Microscopy measurements [27]. Baytekin et al. showed that a mass transfer mechanism for tribocharging of polymers gives rise to nanoscale regions of positive and negative charge [28]. Following this, Galembeck and co-workers showed that polyethylene (PE) and PTFE, tribocharged due to the mechanochemical process of chain scission followed by electron transfer [29], with further support of mass transfer as a mechanism coming from Williams [30], Burgo and Erdemir [31].

An additional complication, which affects charging behaviour, is the role of the ambient environment [31] and humidity [32]. Furthermore, as shown by Sow et al., material strain can reverse the direction of

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charge transfer [33] and may play a key role in triboelectrification in general. Despite these system-specific issues, several studies [31,34,35] suggest that tribocharging and friction force have a common origin and that measurements of the former provide fundamental information on the origins of the latter.

Discharging mechanisms have received considerably less attention and are not well understood [25]. Some evidence suggests that corona discharge limits the formation of charge during contact electrification [36–38]. When an electric field exceeds a threshold (30 kV/cm for air [39]), breakdown occurs that leads to the generation of plasma. The presence of this plasma was first predicted in 1965 [40], but it was not until the beginning of the XXI century that Nakayama and co-workers proved its existence by obtaining images of UV photon distributions in the vicinity of sliding contacts between a diamond tip and a dielectric surface [41–43]. It was found that the gas discharge at a sliding contact is not exactly the same as the gas discharges, which appear during the breaking of adhesive bonds, such as peeling a tape [44]. Besides controlling charge recombination, the generation of plasma is responsible for tribochemical behaviour that cannot be explained by classical means such as flash temperature rise [45,46]. For instance, plasma may induce reactions that alter contact conditions by forming breakdown products that deposit on component surfaces [47]. Plasma generated in this way can cause the degradation of polymeric components and organic lubricants, particularly in the case of computer hard drive applications [48]. It can also influence electric and mechanical properties of the materials of the sliding parts [49], that leads to complex nonlinear behaviour of energy and charge dissipation. At a larger scale, plasma generation at fault asperities, has been linked to seismo-electromagnetic radiation preceding earthquakes [50].

Despite the advances described above, there is still much uncertainty regarding triboplasma phenomena and its links with triboelectrification. Most critically, the vast majority of triboplasma measurements to date have been either time-averaged (*e.g.* Refs. [41,45,50–53]) or spatially averaged [25], and as a result, there is little understanding of the transient nature of this phenomenon. Furthermore, the focus of triboplasma and triboelectrification studies has been on crystalline inorganic materials, with only very few studies [25,54–57] addressing polymer materials, known to be important in tribocharging applications [9].

The aim of the current study is to elucidate the transient behaviour of tribocharging and its relationship to triboplasma for two polymers: ultra-high molecular weight polyethylene (UHMWPE) and polytetrafluoroethylene (PTFE). These two polymers are widely used as tribological materials in engineering and biomedical applications for their low wear (UHMWPE) and low friction (PTFE) properties. Furthermore, PTFE and PE have been studied extensively with respect contact electrification behaviour [29,58,59]. It is understood that, when rubbed, its dominant charging mechanism is the homolytic scission of the C—C bond which produces free radicals with markedly different electronegativities followed by electron transfer to leave regions of cations and anions on surfaces [60]. In this study, plasma triboluminescence distributions around sliding polymer contacts in UV-vis range were measured simultaneously with surface electrostatic potential generated on the polymer. The obtained data were fed into a simplified model of electric field configuration in order to figure out the basic mechanisms of discharge ignition.

2. Test methods

A sliding contact was produced by loading a 5 mm diameter, single crystal Al_2O_3 hemisphere against a rotating polymer disc (see Fig. 1), while a UV sensitive camera (Hamamatsu ImagEM X2) with UV-13X lens (Newport) was focussed through the Al_2O_3 specimen (Swiss jewel, USA) onto the contact to obtain images of the generated plasma luminescence. A charge sensor (of the type developed in Ref. [61]) was positioned diametrically opposite the contact with its tip 2 mm above

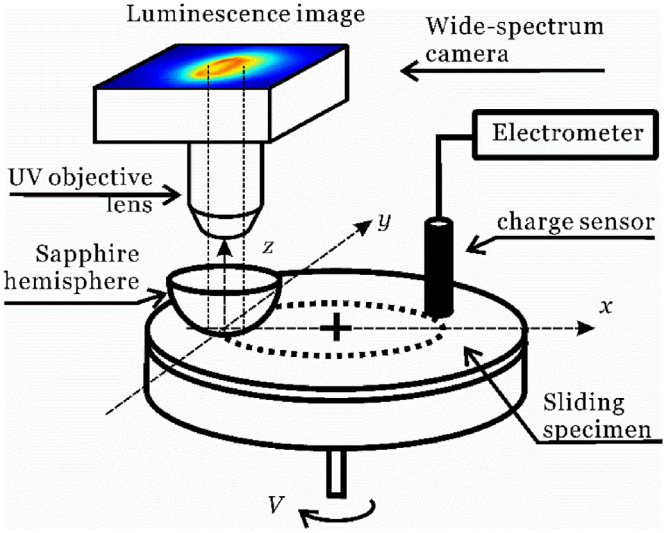

Fig. 1. Schematic diagram of test set up showing the sliding contact between Al_2O_3 hemisphere and polymer disc.

the disc surface and was connected to an electrometer (Keithley 6517B) to record the accumulated charge signal. For certain tests, the camera was replaced by a spectrometer (Ocean Optics QE Pro) to measure the optical spectra of the plasma. All tests were carried out at 2 N load (corresponding to the initial average Hertzian contact pressure of 40 MPa for PTFE and 16–52 MPa for UHMWPE) and 2.4 m/s sliding speed. After each test, scans of the specimen surfaces were performed using a variable pressure SEM (pressure of 60 Pa, 15 kV beam voltage and ×100 magnification) and a Veeco optical profilometer.

Test specimens were discs of UHMWPE and PTFE (Goodfellows, UK). The crystallinity of polymer samples was measured using X-ray diffraction and found to be 54% and 40% for UHMWPE and the PTFE respectively. Single crystal alumina (Al_2O_3) was used for the stationary specimen, since it is transparent to UV and visible wavelengths and enables plasma discharge within the contact to be viewed. All tests were conducted in an ambient atmosphere and room temperature and relative humidity varied between 20 and 50%.

3. Results and discussion

3.1. Experimental transient plasma measurements

Fig. 2 shows time-averaged plasma luminescence intensity images, recorded during rubbing tests on the PTFE and UHMWPE specimens with a frame rate of 1.2 fps for 120 s. The shapes of the triboplasma regions generally agree with those obtained by Nakayama for single crystal oxides (*e.g.* Refs. [41,51]): for PTFE, the luminescence zone has an almost circular geometry with a reasonably uniform luminosity distribution, whereas for UHMWPE it is elongated. Weak protrusions along the wear track at both the inlet and exit sides can be distinguished. For PTFE, a bow-shaped glow is observed around the contact zone that extends to a point approximately 200 μm behind it. This corresponds to Miura's β-peak (the location past the exit of the contact where the geometry of the gap and the configuration of electric field provide a maximum propensity for discharge) [42]. For UHMWPE, a peak is observed in front of the inlet of the contact. This resembles Miura's γ-peak (the location before the inlet of the contact where re-entering charge and gap geometry provide a maximum propensity for discharge) [42], however, it is located closer to the contact zone. In addition to this, both polymers show weak luminescence at the contact areas (Miura's α-peak). Overall, these findings are broadly in line with the work of Nakayama [51] who found that UV luminescence image became symmetrical with respect to the radial axis when a relatively

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