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Electronic Supplementary Information

Dramatic increase in polymer triboelectrification by transition from glassy to rubbery state

Andris Šutka *[a], Artis Linarts [b], Kaspars Mālnieks [a], Klāvs Stiprais [a], Linards Lapčinskis [b]

Affiliations:

[a.] Research Laboratory of Functional Materials Technologies, Faculty of Materials Science and Applied Chemistry, Riga Technical University, Paula Valdena 3/7, 1048 Riga, Latvia. E-mail: andris.sutka@rtu.lv

[b.] Institute of Technical Physics, Faculty of Materials Science and Applied Chemistry, Riga Technical University, Paula Valdena 3/7, Riga LV-1048, Latvia

*corresponding author e-mail: andris.sutka@rtu.lv

Experimental details

Polymer films were prepared by hot-pressing on perforated copper plate electrodes to ensure adherence of polymer on the conductive electrode without the use of adhesives. Hot-pressing temperature for each of polymers - polystyrene (recycled polymer), polycarbonate (recycled polymer) and poly(methyl methacrylate) (Sigma Aldrich, average MW 120`000) was chosen in accordance with corresponding melting temperatures. To change the surface roughness of samples, the pressed polymer films were immersed in solvent for 30 seconds and immediately placed in antisolvent for 10 seconds which does not dissolve the polymer phase but is miscible with the solvent used for polymer. Solvent for PMMA was chloroform with methanol as an antisolvent. By doing so a swollen porous polymer surface layer is precipitated. In TENG device prepared samples were vertically contacted with copper counter electrode.

The short circuit current and voltage pulses generated by TENG devices were measured by using a custom-made voltage divider in combination with a Keithley 6514 electrometer connected to a Picoscope 5444B PC oscilloscope to provide high time resolution. Contacting area (sample size) was 5 cm^2. Surface charge was calculated from measured current by equation $Q=\int idt$ (where i – instantaneous current). Pressing force (10 N) and frequency (1 Hz), as well as separation speed (100 mm s^{-1}) and separation gap between TENG sides (5 mm) was controlled by INSTRON E1000 All-Electric Dynamic Test Instrument, which also allows measuring the force necessary for separation of the contacted films in TENG device. In non-contact measurements TENG device was contacted for 10 times in the aforementioned manner and further oscillated at 1 Hz by changing gap size from 0.5 mm to 5 mm.

To change the temperature of TENG device during operation both electrode plates were adhered on custom-made heatable sample holders (ESI, Figure S1) using high temperature epoxy. Heatable part with the drilled holes for ceramic tubing was isolated from the rest of the holder by PEEK 1000 polymer layer. Sample holder heating was achieved by DC power supply units connected to nichrome wire heating coils interweaved in bipolar ceramic tubes which were inserted in sample holder holes.

Temperature of polymer sample was determined using Fluke Ti10 Infrared Camera to ensure precise temperature control during testing. The mean square (RMS) surface roughness among five independently prepared samples was measured by AFM (Smena, NT-MDT) in semi-contact mode. RMS surface roughness expresses square root of mean square of height deviations of the surface, measured from the mean height of the data plane. Differential Scanning Calorimetry (Mettler Toledo, model DSC3) was used to determine the glass transition temperature of polymers. It was performed in N_2 at 10 °C min^{-1} heating rate.

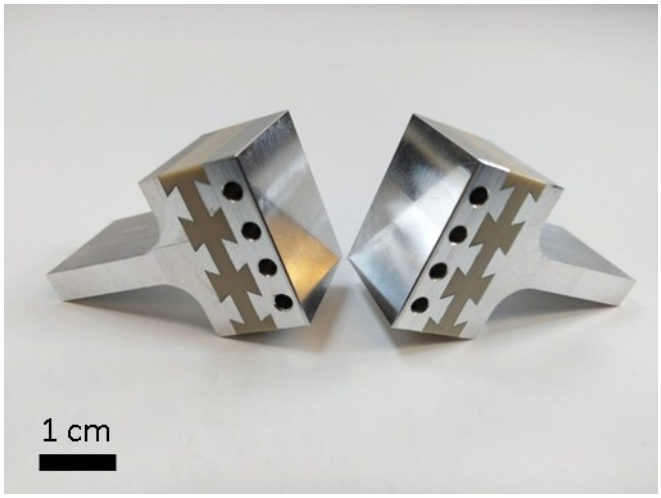

Figure S1. Heatable sample holders. The heating coil was introduced in the holes. The heatable plates were isolated from the rest of the sample holder and device by PEEK 1000 insulator layer (light brown). The sample was attached directly on heatable plate by high temperature epoxy.

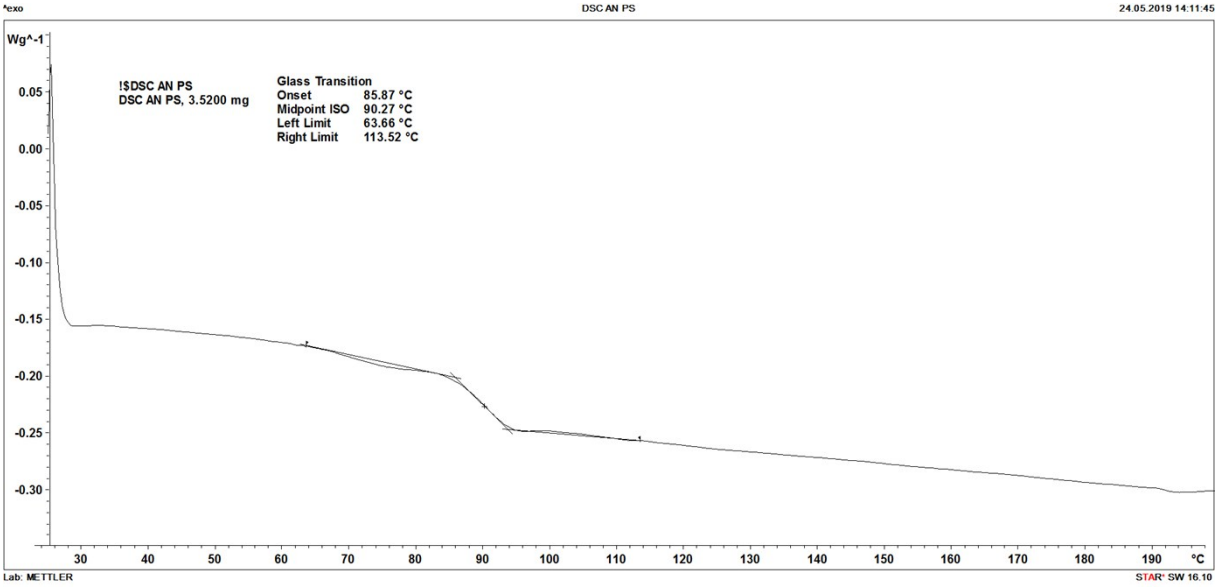

Figure S2. Differential scanning calorimetry (DSC) of PS polymer.

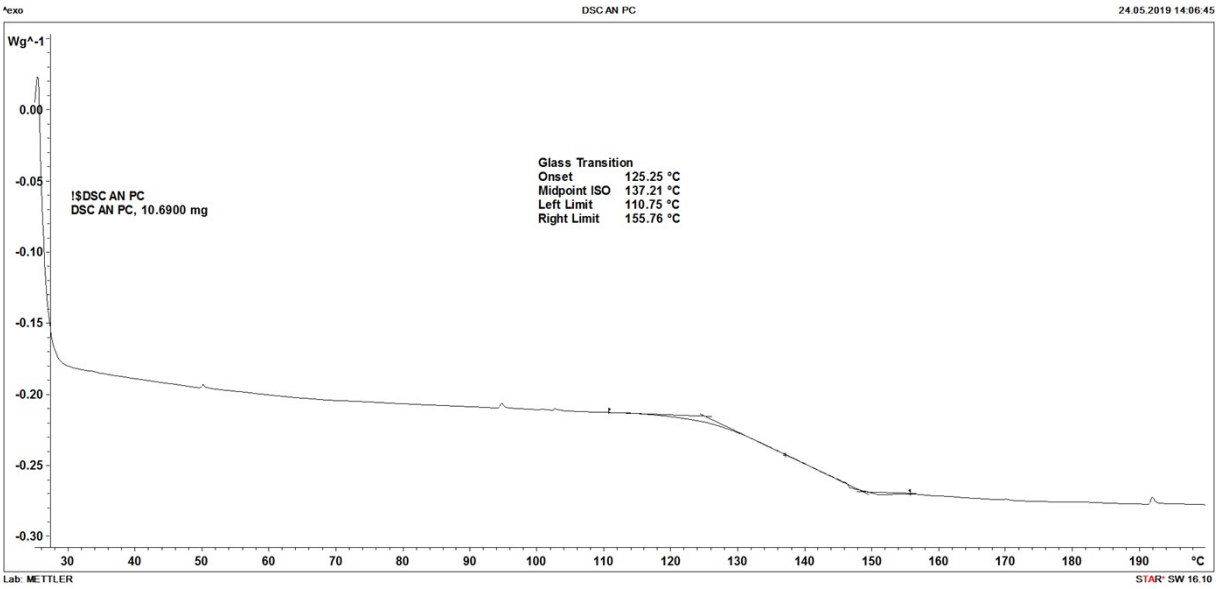

Figure S3. Differential scanning calorimetry (DSC) of PC polymer.

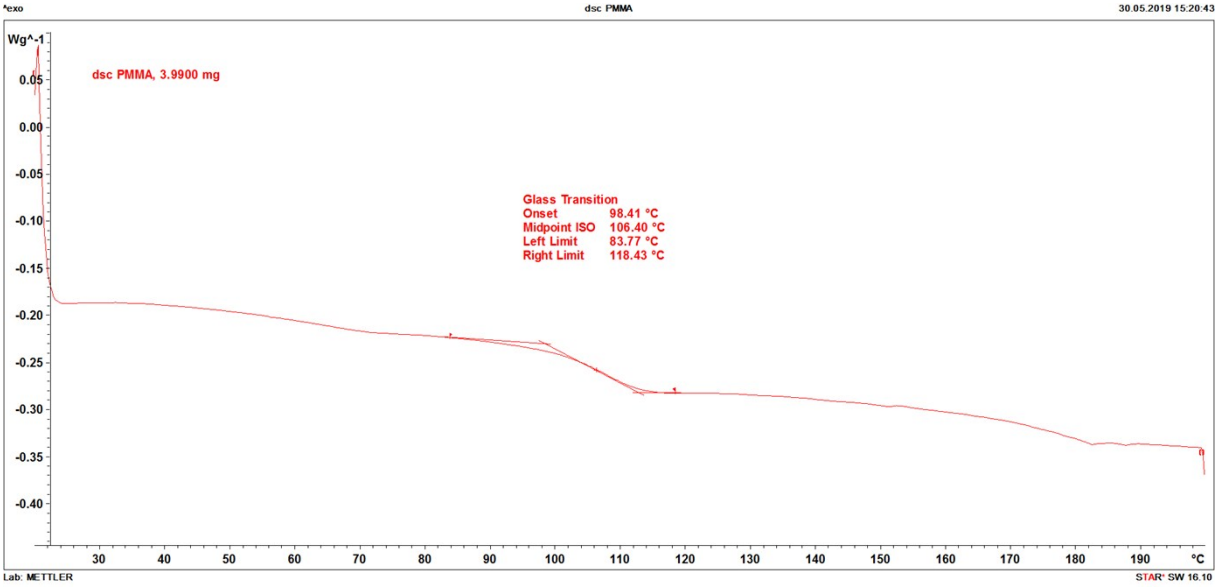

Figure S4. Differential scanning calorimetry (DSC) of PMMA polymer.

Table S1. Glass transition temperature of polymers.

Polymer	T_g, K (Measurement)	T_g, K (Literature)
Polystyrene	363.4	373 [1]
Polycarbonate	410.4	418 [2]
Poly(methyl methacrylate)	379.6	378 [3]

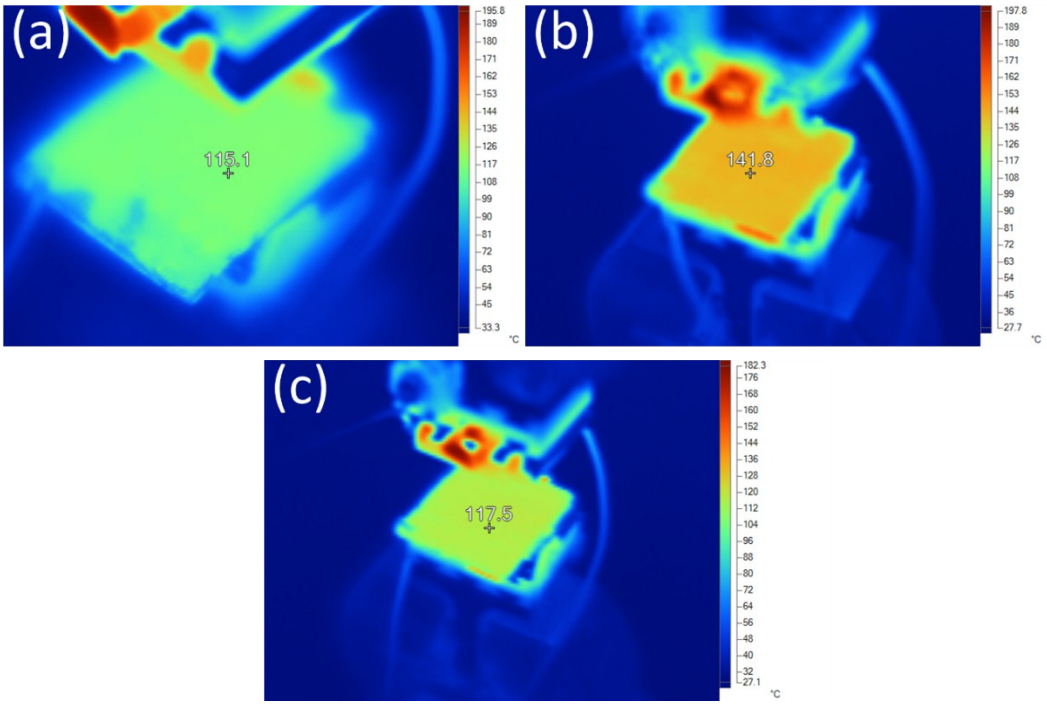

Figure S5. Thermographic images of heated (a) PS sample surface, (b) PC sample surface and (c) PMMA sample surface.

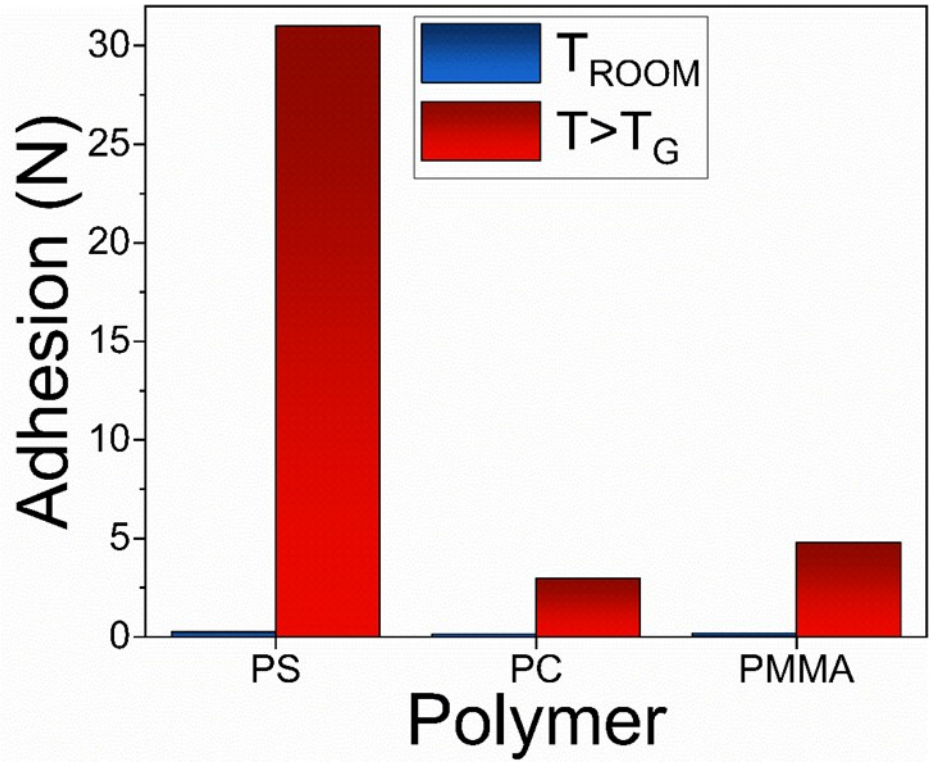

Figure S6. Adhesion force values of polymers PS, PC and PMMA at room temperature and at temperature exceeding glass transition temperature.

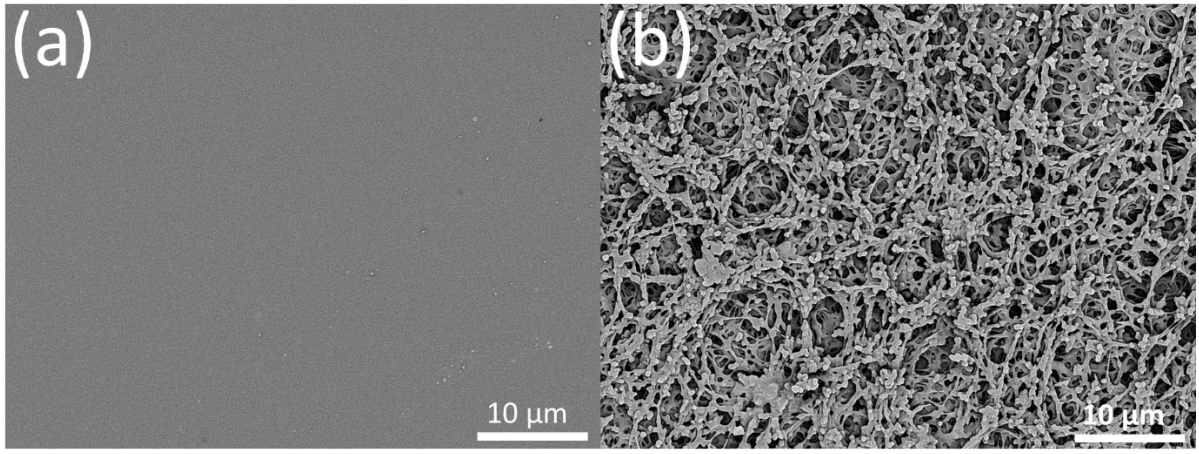

Figure S7. SEM images of (a) smooth (0.08 µm) PMMA sample surface and (b) rough (>1 µm) PMMA sample surface.

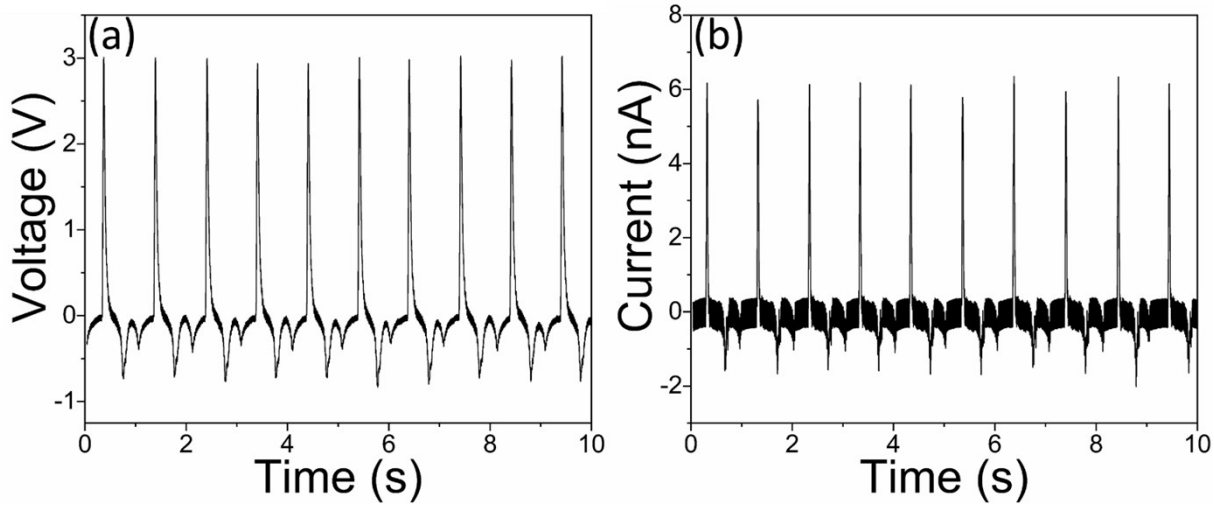

Figure S8. (a) V_{OC} and (b) I_{SC} measurements of rough surface PMMA TENG device.

References

1 J. Brandrup, E. H. Immergut in Polymer Handbook, 4th Edition (Eds.: J. Brandrup, E. H. Immergut, E. A. Grulke) John Wiley & Sons Ltd., Hoboken, 1999, pp. VI/211

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