Supplementary Materials for

Real Time High Voltage Capacitance for Rapid Evaluation of Dielectric Elastomer Actuators

Ang (Leo) Li, Siyoung Lee, Haleh Shahsa, Mihai Duduta

*Corresponding author. Email: shffleo.li@mail.utoronto.ca

This PDF file includes:

Figs. S1 to S15 Legends for Movies S1 References (1 to 3)

Other Supplementary Materials for this manuscript include the following:

Movies S1



Fig. S1. Fabrication process and key dimensions of the DEAs used in this research. **(A)**, the VHB 4910 film, cut in a circle, is pre-stretched onto the rigid pre-stretching frame. Electrode masks are then applied on both sides of the film. CNT left on the PTFE filter after filtration is then stamped on top of the masks, and bottom support (isolate the VHB with the surface by non-adhesive material) can ease the stamping process. Take off the filter and masks, and the CNT electrode of the desired shape is left on the elastomer. The final step is to attach electrical connection wires and seal the connection point with carbon tapes. **B**, Key dimensions of the DEA device. Notice the VHB4910 film is pre-stretched to 3x of its original diameter (from 1 inch to 3 inches) onto the rigid frame. The electrode area is a circle with a diameter of 3cm, with an electrical connection point as a square with a side length of 0.5 cm.



Fig. S2. CNT electrode fabrication process. **(A)**, First transfer CNT ink through a pipette. **(B)**, mix the CNT ink with excessive solvent: a mixture of IPA alcohol and distilled water, with a volume ratio of 3:1. The ink mixture, is then sonicated for 5 minutes. **(C)**, The mixture is then transferred to the standard setup for filtration. **(D)**, comparison of the finished electrodes for ink undergoes sonication or not. As can be seen, the electrode surface is smooth after sonication, while lots of CNT aggregates (black points) are presented for the one that did not go through sonication.





(A), capacitance measurement on devices made with the same geometry but different CNT concentrations. As can be seen, as increasing CNT areal concentration, capacitance increases until reaches a maximum value, indicating that CNTs reach a full conductive network coverage on elastomer surfaces. Also, ink A forms full network coverage with a lower concentration than ink B, indicating longer CNTs tend to form network coverages at lower concentrations than shorter CNTs. (B and C), equivalent series resistance (Resr) and equivalent parallel resistance (Rp) of devices with identical geometry and differences in CNT electrode composition (average CNT length and areal concentrations). As the increase of CNT areal concentration, Resr is decreasing while Rp is increasing, indicating the formation of more ideal capacitors. (D), sheet resistance measurement of CNT with different areal concentrations and compositions on PTFE filter. (E), sheet resistance measurement after transferring the CNT from filters to VHB4910 film. At the same CNT areal concentration and composition, the sheet resistance is increased on VHB film than on the PTFE filter, indicating the change of substrate surface structure on the electrode conductive network.



Fig. S4. SEM images of the microstructure of the electrodes at different concentrations for the three inks. **(A-C)** show the high magnification images of 150uL of ink A, B, and 1:1 mix, respectively. The long nanotubes in ink A lead it to form a noodle-like microstructure by tangling the nanotubes to each other. For ink B, however, the nanotubes' ends can be seen thanks to their short length. For a similar reason, these nanotubes are less likely to tangle. The 1:1 mixed microstructure of ink A and B looks very similar to the ink A microstructure which confirms the formed CNT network is dominated by ink A. **(D-F)** shows the lower magnification images of 40uL of the ink and there are pores between the nanotube clusters. **(G-I)** show 150uL of ink A, B, and the 1:1 mix at lower magnification to compare the ink coverage. Unlike the 40uL concentration, a full ink coverage can be seen for all three inks. No large pores are found, and almost the whole surface is covered by the compact network of the CNTs.



Fig. S5. SEM images of CNT local network on VHB elastomer at the stationary condition in comparison with the network under 100% uni-axial strain. (A), CNT local conductive network formed by 90uL ink A. The network is in the form of a net with CNTs intersecting each other. (B), CNT conductive network formed by 90uL ink A, but the elastomer undergoes a uniaxial stretch of 100% to mimic the condition under high voltage actuation. (C), in a zoomed-in view of a stationary local network, CNTs intersect with each other and form a mesh-like network. (D), a zoomed-in view of stretched CNT local network. Under stretching, the CNTs transform to more parallel alignments. The change of the network alignments may impact the CNT network surface conductivity and the coverage of the conductive region on the elastomer, and provide insights into the network degradation under high-voltage actuation of DEAs.



Fig. S6. Real-time high voltage measurement setup and the circuit diagram of the customdesigned isolation box. Our measurement setup is built by using a custom-designed isolation box circuit to connect the high voltage power source, LCR meter, and the DEA being measured. The data acquisition is done by a LabVIEW program on a computer connected to the LCR meter via USB. The isolation box circuit is shown at the bottom of the figure. The circuit isolates the current input from the power supply through a large high voltage-rated capacitance (C_1), meanwhile delivers the required current and voltage by the DEA. The calibration of the isolation box is required for measurement when the capacitor C_1 is not sufficiently larger than the DEA capacitance (1).



Fig. S7. Circuit calibration procedure.

The decoupling capacitor used in this work is a high voltage ceramic capacitor, with a known capacitance value of 26.5nF (denoted as C_1 in Fig. S6). This ceramic capacitor has stable capacitance measurements after high voltage charging. The capacitance value of a typical ceramic capacitor decreases under high voltage actuation, and that variation in its capacitance will impact the DEA capacitance measurement. The capacitance reading of the LCR meter is influenced by both C1 and the DEA. Thus, to get accurate DEA capacitance measurement, the value of C1 at the high voltage steps needs to be measured prior to experiments. This figure shows the capacitance measurement of the decoupling ceramic capacitor C1 at different voltage steps. In this work, all the DEA devices are tested under 4.5kV, 5kV, 5.5kV, and 6kV. Thus the capacitance values C₁ under these four bias voltage steps are of particular interest. The measured capacitance is shown on the LCR meter screen C_{tot} is actually the series capacitance of C₁ and the DEA (C_{DEA}). Knowing C₁, C_{tot}, we can recover the true DEA capacitance C_{DEA}:

$$C_{tot} = \frac{C_1 C_{DEA}}{C_1 + C_{DEA}}$$

$$C_{tot} (C_1 + C_{DEA}) = C_1 C_{DEA}$$

$$C_{tot} C_1 = (C_1 - C_{tot}) C_{DEA}$$
Thus, we have $C_{DEA} = \frac{C_{tot} C_1}{C_1 - C_{tot}}$



Fig. S8. Area expansion measurement sequence. Notice a video camera is capturing the electrode change during actuation.



Fig. S9. Finite element method simulation. **(A)**, Configuration of the circular capacitive structure designed in geometry and material section. The view was 30 times scaled up in Z direction. **(B)**, Results of the radial deformation of the structure at the applied voltage of 5 kV between the top and bottom electrodes. Colors on the structure indicate the radial displacement of each point from the original position. **(C and D)**, Relative changes of geometry such as volume, capacitive circular area and thickness (C), and capacitance (D) depending on the applied voltage varying from 1 to 5 kV.



Fig. S10. Visualizing the CNT electrode degradation process.

All the devices are actuated under continuous DC voltage of 6kV. The electrodes made with lower CNT areal concentration degrade faster than the ones made with higher CNT areal concentration. However, devices made with higher CNT areal concentration may result in earlier dielectric breakdown. The devices with electrodes made of 150uL ink B do not show obvious electrode defects but experience a much earlier breakdown. We speculate a large number of non-observable ultra-small defect areas appeared on the electrode with higher CNT areal concentrations, the more exposed CNT ending points of ink B may cause an earlier breakdown during continuous actuation.



Fig. S11. Time domain capacitance measurement upon DC actuation at 5kV for 30 minutes of different electrode CNT areal concentration of ink A. Top, the horizontal axis in the log domain. Bottom, the horizontal axis in the time domain.



Fig. S12. Time domain capacitance measurement upon DC actuation at 5kV for 30 minutes for devices with electrodes of the different mixing ratios of ink A and B, controlling identical total CNT areal concentration. Top, the horizontal axis in the log domain. Bottom, the horizontal axis in the time domain.



Fig. S13. Time domain capacitance measurement upon DC actuation at 5kV for 30 minutes of different electrode CNT areal concentration of ink B. Top, the horizontal axis in the log domain. Bottom, the horizontal axis in the time domain. As can be seen, the capacitance decreases much earlier for electrodes made of ink B than ink A.



Fig. S14. Capacitance voltage response. **(A)**, capacitance measurement for devices with different electrode CNT areal concentrations of ink A at different high voltage steps. B, capacitance measurement for devices with a different mixing ratio of ink A and B, with the identical total CNT areal concentration at different high voltage steps.



Fig. S15. Energy transfer model of DEAs during continuous DC high voltage actuation.

During actuation, the device receives input electrostatic energy from the power source. The instantaneous electrostatic energy stored in the capacitance is continuously transferred to strain potential energy, which accumulates to build up and maintain areal strain. The strain potential energy accumulates, maintains, and does not transfer to other forms as long as the voltage supply is constant. Thus, in terms of calculating energy consumption at a specific actuation time, the total capacitive energy consumed is the integration of the instantaneous capacitive energy over actuation time, whereas the total strain potential energy is the energy value at the specific actuation time instant.



Fig. S16. Calculation procedures for strain specific energy at time $t e_{strain}$, electrical specific energy consumption at time $t e_{electrical}$, and energy conversion efficiency η . (A), calculation procedure for e_{strain} , (2). Notice the volume of DEA is kept constant during actuation, and Young's modulus of VHB is also kept constant for ease of calculation. (B), calculation procedure for $e_{electrical}$ and η (3).



Fig. S17. Comparison of High voltage capacitance and strain data on DEAs made with carbon grease and CNT electrodes (150uL ink A). (A), Capacitance evolution (presented in C/C_0 as the ratio to the initial capacitance C_0 of DEAs made with carbon grease electrodes. These devices are actuated with DC voltage of different magnitude. Testing ends when the dielectric breakdown occurs (at 5.5~kV and 6~kV) or after 30 minutes. (B), Electrode area expansion ratio, A/A_0 as a function of time. The measurement was conducted simultaneously as the same devices being tested in Fig. S17 A. (C), comparison of the capacitance evolution of DEAs made with carbon grease electrodes and the ones made with CNT electrodes. Notice except for electrode materials, all the other testing conditions and fabrication parameters are made the same for CNT and carbon grease-made devices. As can be seen, carbon grease electrodes have gone through a more unstable capacitance evolution, and the measured capacitance dropped much earlier compared to the CNT electrodes. At the same actuation voltages, carbon grease electrodes also experience breakdown much earlier. (D), Comparison of the electrode area expansions of DEAs made with carbon grease electrodes and the ones made with CNT electrodes. As can be seen, under the same testing conditions, devices made with CNT electrodes can withstand higher breakdown voltages, and ultimately reach much higher peak areal strain compare to the ones made with carbon grease electrodes.

Movie S1 (separate file).

Real-time electrode degradation VS capacitance evolution.

SI References

- A. M. Goodman, "Safe operation of Capacitance Meters Using High Applied-Bias Voltage." National Bureau of Standards, Princeton, New Jersey 08540, available at <u>https://nvlpubs.nist.gov/nistpubs/Legacy/SP/nbsspecialpublication400-34.pdf</u> Last visited on March 20, 2022
- 2. E. Hajiesmaili, D. R. Clarke, Dielectric elastomer actuators. *Journal of Applied Physics*, *129*(15), 151102 (2021).
- J. P. L. Bigué, J. S. Plante, Experimental study of dielectric elastomer actuator energy conversion efficiency. *IEEE/ASME Transactions on Mechatronics*, 18(1), 169-177 (2011).