

Supporting Information

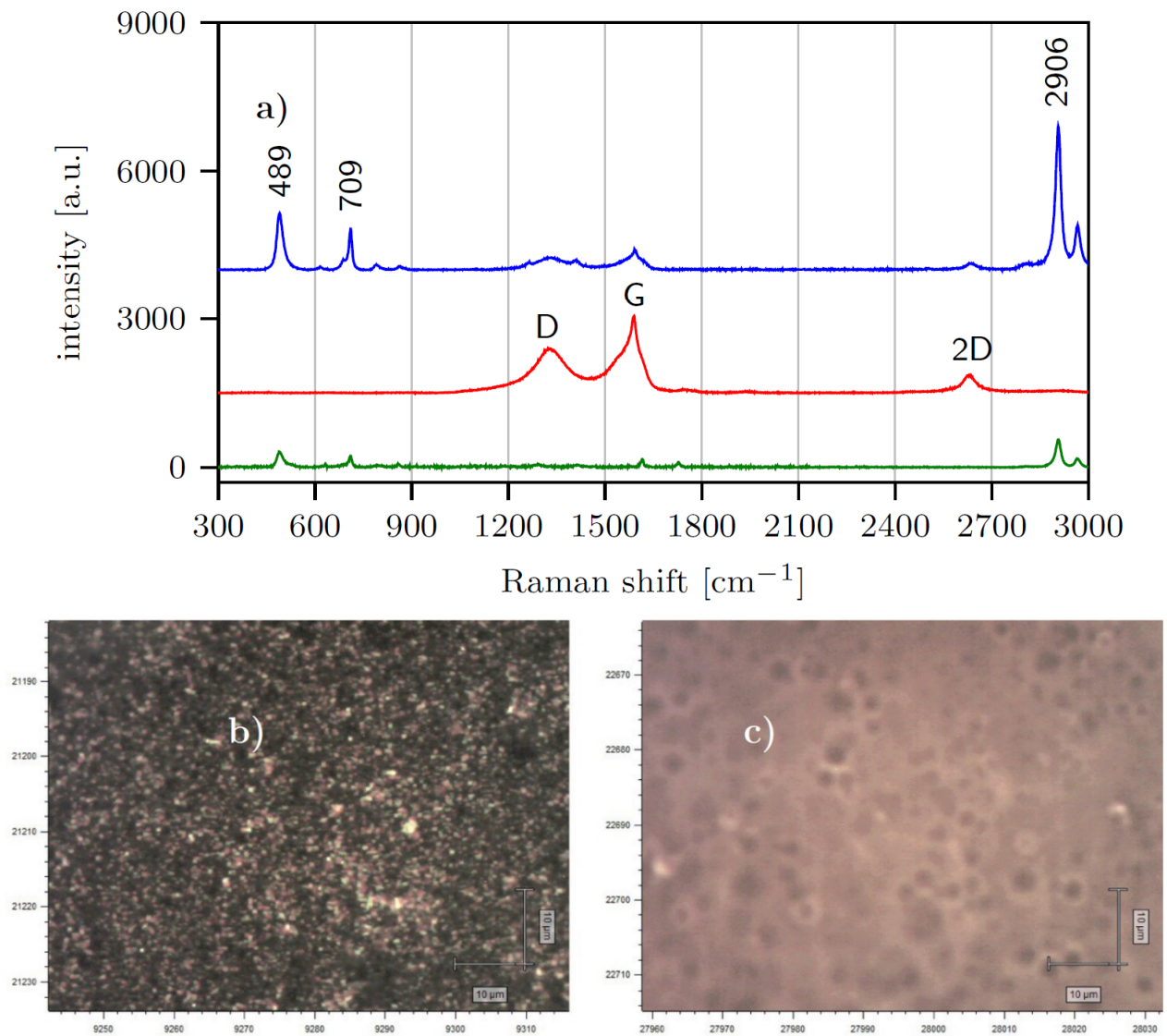


Figure S1. (a) Raman spectra of pristine DE film (green), DE film with the CNT/Gr-NH₂ deposited on it (red) and DE film with the CNT/Gr-NH₂ coated with the functional organosilicone polymer composite (blue). The images of the latter two are shown in (b) and (c), respectively. Description is presented in the text in detail.

Table S1. Comparison among various rolled DEAs reported so far [37,39–42].

| | Zhang et al [37] | Trujillo et al [39] | Kunze et al [42] | Benslimane et al [40] | Lau et al [41] | This work |
|------------------------------|------------------------------------|-------------------------------------|-----------------------------------|------------------------------------|---|-----------------------------------|
| Dielectric elastomer (DE) | Acrylic VHB 4910 | Dow Corning silicone Sylgard 184 | Wacker silicone ELASTOSIL 2030 | Wacker silicone ELASTOSIL RT625 | Silicone BJB TC-5005 | Wacker silicone ELASTOSIL 2030 |
| Electrode material | Graphite powder (TIMREX LB1300) | Sputtered gold | Carbon grease | Corrugated silver | Graphite powder (TIMREX KS6) | CNT/graphene |
| DE roll diameter | 12 mm | 10 mm | 4 mm | — | 11 mm | 12 mm |
| DE roll length | 45 mm | 25 mm | 60 mm | 100 mm | 61 mm | 40 mm |
| Pre-stretch ratio | 3 × 6.5 | 1 × 1 | 1.05 × 1.05 | 1 × 1 | 1.15 × 1.15 | 2 × 1 |
| Pre-tensioner | Spring | Core free | Core free | Core free | Shell design I | Spring |
| Pre-tensioner stiffness | 200 N m ⁻¹ | 0 N m ⁻¹ | 0 N m ⁻¹ | 0 N m ⁻¹ | 103.0 N m ⁻¹ | — |
| Rolled DEA stiffness | 1440 N m ^{-1a} | — | — | 3052 N m ^{-1b} | 305.0 N m ^{-1c} | 1500 N m ⁻¹ |
| Maximum active strain | 31.25 % ^{c,d} | 6.0 % ^c | 2.5 % ^c | 7.0 % ^c | 6.1 % ^c 11.0 % ^e | 6.0 % ^c |
| DE breakdown field (E_b) | 68.2 MV m ^{-1f} | 50 MV m ⁻¹ | — | 35 – 40 MV m ⁻¹ | 33.5 MV m ⁻¹ | 80 MV m ⁻¹ |
| Pre-tensioner weight | 7.2 g | — | — | — | 1.7 g | 4.5 g |
| Total weight | 8.0 g | — | — | — | 7.0 g | 5.0 g |
| Static work density | 2.25 J kg ⁻¹ | — | — | — | 0.133 J kg ⁻¹ | 1.44 J kg ⁻¹ |
| % of max. work density | 20.7 % ^g | — | — | — | 31.0 % | 24.4 % |

^a Calculated using max. blocking force/max. stroke = (7.2 N/5 mm).

^b Calculated using blocking force/max. stroke = (3 N cm⁻² × 500 cm × 0.0082 cm)/(6.5 % × 62 mm).

^c Longitudinal.

^d Calculated using max. displacement of actuator/active length = (5 mm/16 mm).

^e Transverse.

^f Calculated using activation voltage/(initial thickness/(pre-stretch factor)) = (3500 V/(1 mm/3 × 6.5)).

^g VHB properties: Young’s modulus 1.8 MPa, dielectric constant 4.7, density 960 kg m⁻³