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Strongly enhanced sensitivity in elastic capacitive strain sensors

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Strain sensors based on dielectric elastomer capacitors function by the direct coupling of mechanical deformations with the capacitance. The coupling can be improved by enhancing the relative permittivity of the dielectric elastomer. Here, this is carried out through the grafting of conducting polymer (poly-aniline) to the elastomer backbone, leading to molecular composites. An enhancement in capacitance response of 46 times is observed. This could help to extend the possible range of miniaturization towards even smaller device features.

Advances in flexible electronics have created an increasing demand for stretchable mechanoresponsive materials for sensors and actuators.^{1–4} Thin elastomer pressure sensors relying on either resistive² or capacitive⁵ sensing have been successfully developed and suggested for numerous applications such as electronic skin for robotics and intelligent surfaces, wearable communication devices, haptic interfaces, and high-frequency ultrasonic transducers.^{2,5–8} Another application concerns structural health monitoring of civil structures, such as bridges, buildings, dams, *etc.* for damage diagnosis^{9,10} and localization.¹¹ Capacitive sensors based on elastomers provide a particularly attractive option since the device is sensitive in the low stress regime, has fast response times and can sense both small and large strains. Further miniaturization will allow for more localized pressure detection,^{2,5} but is limited to the detection limit of capacitance. Therefore, there is a need to develop strategies for improving the intrinsic capacitive response of materials.

Here, it is demonstrated that the sensing ability of a capacitive sensor based on dielectric elastomer is improved by raising the relative dielectric constant ϵ_r . We focus on the *strain* sensing properties. Nevertheless, because the materials are elastomeric, they can be used in a *pressure* sensor of the type demonstrated recently by Mannsfeld *et al.*⁵

The enhancement in permittivity is achieved by grafting a small amount of conducting polymer on the matrix elastomer backbone to form a molecular composite.¹² The conducting polymer enhances the permittivity *via* the hyper-electronic polarization (Fig. 1), due to the mobile charges within the π -conjugated backbone.¹³ When an external electric field is applied (dotted lines), the charge inside the

polymer is polarized, leading to a net positive amount of charge towards the positive electrode, and *vice versa*. The grafting mitigates agglomeration of the conducting polymer and thereby evens its distribution. It is shown here how the capacitive response of sensors based on such material to mechanical strain is strongly enhanced by augmenting the dielectric constant.

The change in capacitance ΔC of a sensor strip due to an applied strain s is $\Delta C = C_0 s$ (assuming uni-axial deformation). C_0 is the initial capacitance of the sensor strip before any strain has been applied, $C_0 = \epsilon_0 \epsilon_r A_0 / d_0$, where A_0 and d_0 are its area and thickness, respectively. Here, samples with identical dimensions A_0 and d_0 are compared.

The procedures for synthesizing and casting the elastomer materials investigated here has been described in greater detail in ref. 12, the issues relevant to the present investigation are discussed briefly. The base elastomer material was poly-(styrene-*co*-(ethylene-butylene-*g*-maleic-anhydride)-*co*-styrene) (SEBS-*g*-MA, Young's modulus $Y = 4.5$ MPa and $\epsilon_r = 2.0$), which is a thermoplastic SEBS triblock copolymer, grafted with 1–2% maleic anhydride (Kraton FG 1924 G). The maleic anhydride functional groups were grafted to the backbone of poly-aniline (PANI, Panipol T), leading to a covalent amide bond between the backbones of the elastomer and conducting polymer (see Fig. 1, right). The presence of the amide bond is evidenced by analysis of various peaks in the FTIR spectra.¹² The elastomer materials were all fully solvable in toluene.

Films were prepared by casting from toluene solution, directly onto glass slides with electrodes for dielectric relaxation spectroscopy (DRS). Samples with concentrations of 0%, 2.0% and 2.3% by

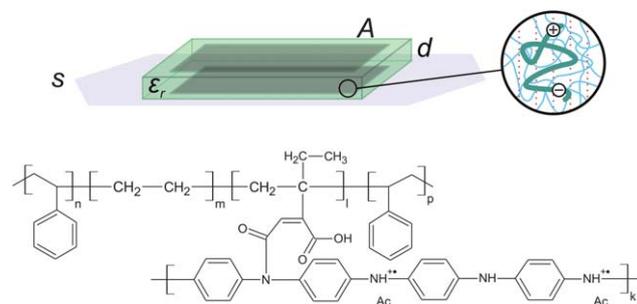


Fig. 1 (above) The deformation of the sensor strip leads to a geometrical change in its capacitance. The dielectric constant was enhanced by the presence of conducting polymer, which allowed for large displacement of mobile charge, essentially forming a macromolecular dipole. (below) The chemical structure of the molecular composite. Not shown is the dopant, which is dodecyl benzene sulphonate (DBS).

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volume of PANI were prepared. Samples with other concentrations were also investigated, however, we focus on these three concentrations here (the data measured on other samples were consistent with the data presented here). DRS was performed at room temperature using a Novocontrol Broadband Dielectric Analyzer, between 0.1 Hz and 1 MHz (see Fig. 3 in ref. 12). The relative dielectric constants at 100 Hz were determined to be 2.1, 24 and 122, respectively.

The cast films ($\approx 40 \mu\text{m}$ in thickness) were removed from the DRS substrates and attached with plastic frame pieces with lead wires, using double sided tape. These were mounted in a tensile tester (Zwick Z005), then carbon grease electrodes were painted on both sides and onto the electrical leads. This defined the geometry of the active sensor with a length of 25 mm, a width of 10 mm and a thickness of $40 \mu\text{m}$. The electrical leads were connected using shielded cables to an LCR meter (HP 4284A) in C_p -G mode. The sample was placed in an oven (without turning it on) for temperature stabilization. The capacitance signal was measured at 100 Hz, without averaging. Before ramping, the samples were prestretched to about 1%. The samples were then stretched at a constant strain rate of $0.5\% \text{ s}^{-1}$. The strain program was either a simple ramp (Fig. 2) or a complicated strain profile with progressively increasing strain targets (Fig. 3).

The change in capacitance with strain is plotted for three different sensor samples in Fig. 2 (below), with 0%, 2.0% and 2.3% of PANI, respectively. The initial capacitances were determined at about 1% prestrain, resulting in the dielectric constant values reported in the graph. The slopes of the capacitance *versus* strain curves were 169 pF, 1629 pF and 7562 pF respectively, which are almost identical to the independently measured C_0 . Comparing the C_0 value of the material with 0% PANI to the material with 2.3%, an enhancement of 45 times is seen.

The upper diagram in Fig. 2 shows noise characteristics measured by first stretching the sample to 10% strain, then recording capacitance for 3600 s. The standard deviations of the full capacitance

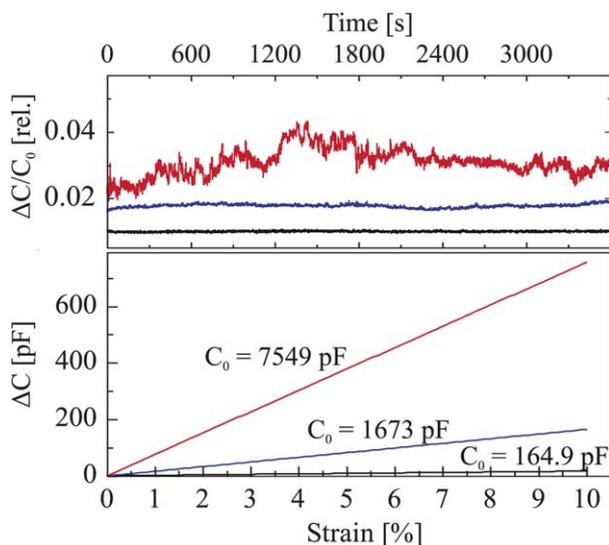


Fig. 2 (below) Change in capacitance with strain, for samples with 0%, 2.0% and 2.3% PANI, respectively. The values for C_0 were measured in the slightly prestretched condition (10% prestretch), from which ϵ_r values were calculated. (above) $\Delta C/C_0$ time traces at constant strain, showing the noise levels.

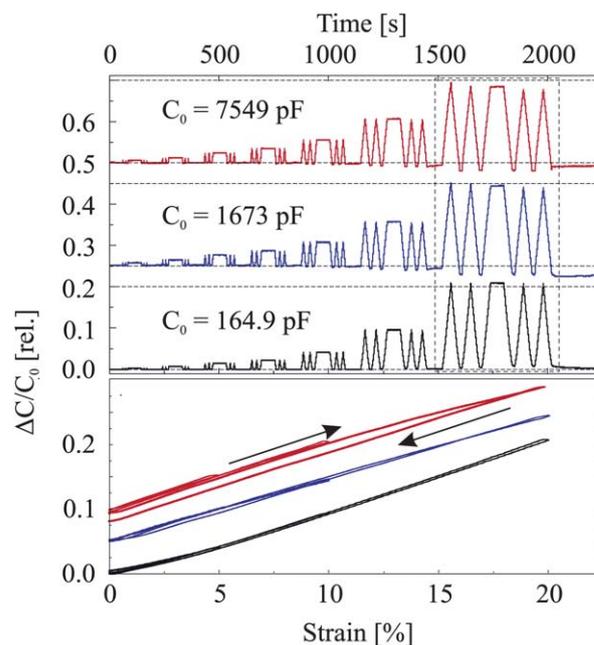


Fig. 3 (above) Time series for strain signals with progressively increasing strain targets. Curve for $C_0 = 164.9 \text{ pF}$ corresponds to a sensor based on pure SEBS-g-MA material, the other two curves to the PANI-doped materials. (below) Cyclic capacitive strain experiment for all three materials (same order as upper graph).

traces, $\sigma(\Delta C/C_0)$, for the three samples were 5.0×10^{-4} , 1.8×10^{-4} and 4.9×10^{-3} , respectively. These values indicate the long-term accuracy and repeatability of the samples when operating as strain sensors. The accuracy is found to proportionally decrease with the increased capacitance of the sample, or with the relative dielectric constant. It is possible that noise level is proportional to the capacitance of the device, which would imply that device miniaturisation and corresponding capacitance reduction would lead to reduced relative noise levels. This would be elucidated by a specific miniaturisation investigation which was not carried out here.

Strain profiles with progressively larger strain targets show the ability of samples to measure intermediate (0.5%) to high (20%) strains (Fig. 3, above). The dashed lines indicate baseline and 20% signal levels. Note that the last five signal 'events' (within the dashed box) are for applied strains between 0 and 20%. As is seen, all samples are able to capture the features of the strain signal, including triangles and plateaus. There is a slight tendency at the highest strain levels for the base line signal to drop for both samples containing PANI. The level of the baseline drop becomes more obvious when the relative capacitance is plotted against actual strain (Fig. 3, below). The effect appears to be more pronounced for the sample with the intermediate C_0 , which indicates that external effects such as temperature drift could be responsible. It is clear that even with the standard deviations determined previously (which were determined over 3600 s), the noise hardly affects sensing accuracy at these strain levels.

The noise levels were estimated at prestretch and on the 20% strain plateaus (Fig. 3). The results so far showed a level of drift, which was ascribed external sources such as temperature variations. An intrinsic noise level can be determined by calculating the standard deviations of the signals over shorter time periods. With a time period of 10 s, the standard deviation of the relative capacitance signals, $\sigma(\Delta C/C_0)$,

are found to be 1.49×10^{-4} , 1.54×10^{-4} and 7.3×10^{-4} respectively. These values are very similar. Interestingly, the noise level is the same for the pure sample and the sample with 2.0% PANI, while it rises 5 times for the sample with 2.3% PANI. For the sample length, which was 25 mm here, these values correspond to detection limits of actual length changes of 3.7, 3.9 and 18.3 μm .

Based on the strain detection limits and the stiffness of the material, the stress sensing capability can also be determined. Since the stress is $T = Ys$, the noise level of a stress measurement can be calculated from the strain detection limit directly. In case of the materials measured here, with the chosen measurement setup, the stress detection limits were $\sigma(T) = 671, 693$ and 3285 Pa respectively (in the local OFET-amplified microstructured system of Mannsfeld *et al.*,⁵ a detection limit of at least below 3 Pa was demonstrated). Using similar local OFET amplification, these detection limits might be improved. The developed materials should also increase the capacitance response of bending or flexing strain sensors, as well as of compressive strain sensors. Since the elastomer is incompressible, it requires of the corresponding device that tensile deformations of the elastomer are possible.

Conclusion

We conclude that the capacitance signal of an elastomeric capacitive strain sensor strip can be strongly enhanced by increasing the dielectric constant of the base material. This was demonstrated using a molecular composite, in which a conducting polymer was grafted to the backbone of a thermoplastic triblock copolymer. Direct ramping with capacitive sensing showed an enhancement in sensitivity proportional to the total capacitance of the strip. An enhancement from 165 pF to more than 7549 pF was observed, an improvement of 46 times. In all materials tested, the change in relative capacitance was found to be equal to the change in strain.

The relative noise levels increased with increased dielectric constant. Long-term (3600 s) fluctuations of the relative capacitance of the high dielectric constant material were found to be much higher than in the short-term (10 s), indicating that drift could cause problems for the determination of absolute strain with this material. The

drift could be due to variations in temperature. However, for applications such as haptic skin and other haptic interfaces, only short-term changes are important. Hence, the use of molecular composites for increasing the capacitance response of capacitive sensors seems highly promising for these applications. In particular, applications where miniaturisation is required, such as robotic sensing skin, these materials can allow for much smaller device features than otherwise possible with regular elastomers.

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