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Scale-like compliant gold electrode: Towards high strain capacitive devices for energy harvesting

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ABSTRACT

Highly compliant electrodes are of primary importance for high strain capacitive energy harvesting. Herein, we present a compliant gold sputtered electrode on a natural rubber substrate. Electrical conductivity remained remarkably good even at strains of 500%. The robustness of the electrodes has been assessed in fatigue tests and resistivity of less than 25 $\Omega\,cm^{-1}$ were observed after 1500 cycles between 200% and 300% strain. These electrodes were then used in harvesting energy for large strains and experimental energy densities up to 3.3 $mJ\,cm^{-3}\,cycle^{-1}$ have been recorded, showing the capabilities of such electrodes for efficiently ensuring electrical contact under high strain for converting mechanical energy into electricity.

1. Introduction

Energy harvesting using electroactive polymeric materials has been a hot topic in the scientific community for the last decade due to their low-cost, processability and light weight [1–8]. Harvesting energy using dielectric elastomers can be done using electrostatic harvesters where the capacitance variation of the material subjected to a mechanical force (vibrations, stretching...) is the main process ([9a–11]). For instance, this type of system could be integrated into fabrics in order to harvest energy from body movement where strain over 100% could be expected. Although it is theoretically possible to harvest energy by stretching elastomers, very few can get a large capacitance difference between stretched and unstretched polymer as the maximum deformation allowed is usually hindered by the compliance of the electrodes deposited onto the dielectric material surface [12,13]. The electrodes should therefore be made in such a way that no loss of conductivity is observed when stretching it.

Techniques such as the use of microstructured metallic electrode [14,15], conducting polymers [16], ion implantation [17,18], dust or carbon grease [19] all lack from conductivity, ease of use or loss of conductivity when stretched, and thus, are not ideal solution for capacitive energy harvesting or, as for carbon loaded grease, are not practical to use in systems. Indeed, the techniques described above for metallic and conducting polymer electrodes can only be employed where strain <100% are recorded. In addition, films covered with dust or carbon grease electrodes are very difficult to handle and can result in messy system when they are implemented.

Recently, Wallace et al. reported the fabrication of compliant gold/polypyrrole electrodes showing loss of conductivity above 30% longitudinal strain [20]. Strain value for the loss of conductivity – <100% strain – has been reported by many in the microelectronic community where stretchable electronics is a source of interest [21–24]. The main technique employed in these papers is to pre-stretch the material before conducting material deposition so that when released a wrinkled metallic electrode is formed making it compliant in the pre-stretching direction.

We believe that the technique described above can be exploited further and that it is possible to form fish scales-like structures where gold scales could slide from one to another, therefore making the electrode compliant in the pre-strain direction even at large strains.

In this paper, a 500% gold sputtered stretchable electrode is presented using natural rubber as substrate. Its conductivity remained below 25 $\Omega\,cm^{-1}$ in the stretching direction. Application of this electrode towards capacitive energy harvesting is demonstrated.

2. Elaboration

The substrate used in this study was a 50 μm thick natural rubber (NR) sheets purchased from Fisher Fabrics. NR was chosen for its highly stretchable property as well as its good mechanical and chemical resistance. An elastic strain of nearly 600% can be reached without any appearance of plastic behaviour. Highly stretchable

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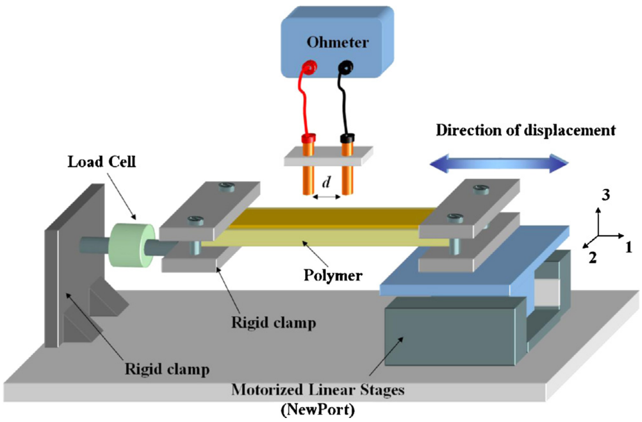

Fig. 1. Fatigue, strain and mechanical test setup. The fatigue test consisted of continuously stretched and unstretched for a sample of 50 mm of length and 20 mm of width until electrode failure (where failure meant that the electrode was "non-conductive at rest").

gold electrodes were deposited in a 2-step process. First, a NR sheet was pre-stretched with a strain ratio (SR) ranging from 0% to 500%. Then, 50 nm thick gold electrodes were sputtered on one or both sides of the polymer using a Cressington® Sputtering Coater (208HR). The samples were then carefully unstretched with a gradual release of the pre-strain.

3. Characterization

3.1. Structure characterization

The gold structure was assessed by SEM using a FEI XL30 FEG ESEM.

3.2. Resistance, fatigue and mechanical characterization

Three types of measurement have been realized: electrode fatigue under mechanical excitation, variation of the resistance with strain and elastic modulus measurements using the test bench described below (Fig. 1).

First the variation of electrical resistance over applied mechanical strain was tested using a dedicated bench test (Fig. 1). The procedure consists of applying strain with a linear motor (XM550 ironless linear motor—Newport Cop., Irvine, CA) and measuring the resistance between the two probes of the Ohmmeter. In order to measure the electrode resistance, the sample was placed between two clamps, as shown in Fig. 1. The resistance of the electrode was measured with the help of a multimeter (ITC-920) where the two probes were separated by 10 mm (d) at all time. The structure of test bench presented in Fig. 1 was composed of two parts: one fixed and a second that could be moved in the 1-direction with the help of the linear motor. As a consequence, the electrodes were driven with a given strain profile and assumed to be strained along their length. The generated stress was measured with a help of force sensor (ELPF-T2M-250N, Measurement Specialities, Paris).

The fatigue of the electrodes over mechanical strain is also required to fully characterize the electrodes. Classical approaches to fatigue design involve the characterization of total fatigue life to failure in terms of the cyclic stress or strain range. In these methods, the number of stress or strain cycles necessary to induce fatigue failure in initially uncracked specimens is estimated under controlled amplitudes of cyclic stresses and strains. The test bench described above was also used for this study. Fatigue resistance of the gold metallized was tested by repeated transverse strain and resistance measurements were made every 5 min during 15,000 cycles for a strain varying from 200% to 300%. The measurements were always realized at the same position on the sample, with a 10 mm distance between the two probes (d). The bench test described above was also use for mechanical characterization. The load cell was used to measure the stress and the linear stage to apply the strain. Experiments were performed on 4 cm by 1 cm samples at 1 Hz.

3.3. Energy harvesting capabilities

Fig. 2 provides a schematic representation of the setup developed for characterizing the energy harvesting by the polymer film. One end was fixed by a rigid clamp fitted with copper foil electrodes that was connected to electrode surface of the sample, and the other end was mounted on the ironless linear motor which produced the mechanical strain. For harvesting energy an electrical preload is necessary, it is produced by a power amplifier (Trek 20/20C) driven by a function generator (Agilent 33220A). The electrical responses due to geometric variation of the sample were measured with the help of voltage probe (Model 820, Trek, USA) and current amplifier (SR570, Stanford Research Systems Inc. Sunnyvale, CA). The data were monitoring with the help of oscilloscope (Agilent—DSO7034A).

4. Results and discussion

4.1. Structure

Fig. 3 shows the observed structures on unstretched (A), after releasing a 500% pre-strain (B), and when applying a 200% strain on electrodes after releasing 500% pre-strain (C and D). The structures obtained after releasing were scale-like and conduction remained good even a large applied strains as conduction paths between gold scales existed.

On unstretched substrates, the gold layer exhibited minor cracks and the overall structure is smooth. However, when sputtering on a pre-stretched substrate, gold scales are formed upon releasing the applied pre-strain. It is believed that during the pre-strain release, wrinkles started to form and that the spatial frequency of the wrinkles increased. At one point during the release the forces on the gold layer were large enough for it to crack, leaving chips of gold on the NR surface attached to it on one side.

When stretching the electrodes (Fig. 3C and D) gold scales are redistributed over the surface of the substrate. It is seen in Fig. 3C and D that although gaps between gold scales begin to appear upon stretching the electrodes at 200%, a conduction paths still exists as some gold scales are still connected one to another.

4.2. Mechanical characterisation

To ensure that the electrodes did not increase the stiffness of the film, mechanical characterization have been performed. The results are depicted in Fig. 4. The Elastic modulus was calculated to be around 2.5 MPa for both samples (with and without electrodes). Therefore, it is demonstrated that the electrode has no effect on the stiffness of the film.

4.3. Electrodes resistivity

Firstly, the resistivity of the fabricated electrodes was assessed using the dedicated bench test described above. Fig. 5-a shows the resitivity *vs.* applied strain for electrodes deposited with pre-strains of 0%, 100%, 250% and 500%. It appeared that loss of conductivity occurred with a strain 50% higher that the pre-strain, *i.e.* at 50%, 150%, 300% for a pre-strain of 0%, 100% and 250%, respectively. The compliance of the electrodes is also clearly demonstrated as resistivity under 25 $\Omega\,cm^{-1}$ in the strain direction was recorded