Cyclopean gauge factor of the strain-resistance transduction of indium oxide films

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Abstract. The resistance of indium-oxide covered polyethylene terephthalate foils (IO-PET) shows an extreme sensitivity to tensile strain. In terms of the deformation-resistance transduction, the gauge factor as high as about 60 000 was recorded upon the relative elongation up to 1 %. Except the onset of deformation, the nearly exponential dependence of the resistance on strain suggests that the conductivity of the strained films is governed by tunnelling mechanism; this notion is supported by the formation of scattered cracks in the IO-PET film. The cracks are oriented perpendicularly to the strain vector and are characterized by a rather similar and uniform width. Appropriateness of the standard definition of the gauge factor for strain sensors, which are governed by tunnelling conductance, is critically discussed.

1. Introduction

Applications of strain sensors span from traditional pressure and torsion gauges to tactile sensors [1-3]. Their design flexibility reflects employment of various physical and chemical methods, yet simple resistive strain sensors attract a lot of attention. The principal parameter characterizing the strain sensor is the gauge factor, g

\[ g = \frac{R - R_0}{R_0} \frac{L_0}{L} \]

where \( R_0 \) is the initial resistivity of the pristine sample of the length \( L_0 \), \( R \) is the resistivity of the sample stretched to the length of \( L \). The denominator \( (L-L_0)/L_0 \) is termed the strain, which is commonly expressed in per cents.

Reported more than four decades ago, strain gauges based on metal films formed by discrete islands demonstrated \( g > 100 \) [4], thereby markedly higher compared to the continuous bulk metal films having \( g \sim 2 \). This was shown to be due to the conductance governed by tunneling current mechanism, which occurs in discontinuous films [5-8]. A recent impetus has been provided by employment of nanostructured films (see e.g. [9-16] and references therein). The exponential
dependence of the tunneling current on the separation gap between adjacent nanoobjects results in large conductivity changes responsive to fractional deformation; the gauge factors as high as 200 [10], 300 [15], and even 700 [13] were reported for closely-packed nanoparticle (NP) assemblies. The indium-tin oxide (ITO) films have become standardly used as transparent conductive electrodes in optical planar devices such as solar cells and light emitting diodes. The ITO films supported by either glassy or flexible substrates have been available on commercial basis. Alternatively, indium-oxide (IO) or fluorine-tin-oxide (FTO) films can be considered. The massive usage of ITO in optical devices may overshadow their much less frequent utilization in the role of active sensing films, e.g. for piezo [17] and strain [18,19] sensors. In this study, we report on strain-resistivity transduction of the indium-oxide coated polyethylene terephthalate flexible substrates; the gauge factor up to about two orders higher magnitude as compared to up-to-now reported highest values for the nanoparticle-based strain gauges was determined. The nature of the extremely high response is examined.

2. Experimental
The films under study were stretched using the TS 600 device (Anton Paar, GmbH), which controls the tensile strain by axial stretching of the probed sample. The total length of the examined stripes was 37 mm, while the actual length subjected to the strain was 24 mm. The width of the stripes was chosen to be either of 8 or 14 mm. The samples were indium-oxide coated polyethylene terephthalate (IO-PET) foil.

The IO-PET foil with the surface resistance below 10 Ω/□ and the thickness of 0.2 mm was purchased from Sigma-Aldrich. Except the specification “In2O3/Au/Ag coated PET film”, no further information was ascertainable. Silver was evaporated on a pair of areas separated by 10 mm for resistance characterization and the wired terminals to the resistance meter were glued to these areas by colloidal Ag paste. The film resistance of the stretched film was measured at bias of 1 V. The gradual stretching of the IO-PET foils (typically with a constant ramp rate of ΔL/L0 ≈ 0.23 %/min) was carried out in the direction of the current flow up to a preset maximal strain, e.g. 1 %. The reached maximal strain was maintained for 5 minutes followed by a strain release either with the same ramp or abruptly.

The morphology of the IO-PET films was examined by the field-emission gun Scanning electron microscopy (SEM). The SEM images were recorded by means of the FEG-SEM JEOL JSM 7500F facility using combined signals originating from both secondary and back-scattered electrons with accelerating voltage of 10 kV and working distance of 4.1 mm. For the SEM examination of the strained IO-PET films, the deformation of the pre-stretched film was preserved by gluing a rigid solid substrate to the reverse side of the IO-PET sample.

3. Results
The sequential strain cycles were applied to the IO-PET samples. The cycles consisted of a gradual increase of the strain up to the targeted maximal strains of 0.5 or 1.0 %, the retention of the maximum strain for 5 min, and a gradual decrease of the strain with the same ramp. The first cycle up to the strain of 0.5 % showed relatively low resistance change. The next cycle up to 1 % led to the resistance normalized to the initial resistance R/R0 > 600. The following cycle up to 0.5 % showed markedly higher R/R0 compared to the first 0.5 % cycle. The next cycles either up to 0.5 % or 1.0 % showed R/R0 values resembling to those of the 2nd or 3rd cycle. The reproducibility of the cycling will be addressed below.

Figure 1 presents one cycle of the R/R0 versus strain dependence for the IO-PET film in the semi-logarithmic plot. The dependence shows a hysteresis presumably due to a large time constant of the supporting foil. The gauge factor of the presented curve calculated at the strain of 1 % is about 60 000. The issue of the evaluation of the gauge factor will be discussed in more detail below. The dependence is nearly linear in the log scale in the range between ca 0.5 and 0.8 % suggesting the exponential evolution of the resistance upon the applied strain.
In the given experimental layout, electrical contacts had to be located within the strained area. Yet, our tests showed that the Ag contacts were not affected by strain and accordingly they do not contribute to the resistance-strain transduction of the probed IO films.

The surface mapping of the IO-PET films by the SEM (figure 2) revealed grains arranged into a network of irregular elongated “chaplets” with their elongation equally oriented.

The same technique was employed for the inspection of the surface morphology of the strained IO-PET film. For this purpose, the sample stretched up to 1 % was fixed by gluing its reverse side to a rigid ceramic plate followed by the release of the external force. The strain had partially released in spite of the gluing, yet it sustained the value of about 0.5 %, as deduced from the remaining elevated film resistance still markedly exceeding the initial resistance. The SEM inspection (figure 3) suggests roughly equidistantly parallel-oriented cracks with orientation perpendicular to the deformation vector. Figure 3b details one crack at higher magnification and it reveals fairly uniform width of about 30 nm across the sample. By estimating the crack frequency of about 1 crack per 10 μm (figure 3a), the crack width of ca 30 nm (figure 3b), and considering the stretched length of 24 mm, the sum of crack widths is about 0.072 mm yielding \( \Delta L/L_0 \approx 0.3 \) %. This is in qualitative agreement with the strain of about 0.5 % deduced from the resistance of the pre-stretched sample and supports the reasoning on the presence of cracks over the entire strain sample length.

We presume that the high resistance-strain transduction is due to the current tunneling over the cracks emerging in the IO films subjected to strain. Their uniform widths over the entire width of the sample are presumably owing to both well-defined cleavage of the polycrystalline IO films and the uniform deformation of the PET substrate.
In view of the strain-resistance transduction governed by the formation of cracks, admittedly, a question on the reproducibility of a such response may arise. To illustrate the reproducibility of the transduction for a particular IO-PET sample, five sequential stretching cycles (figure 4a) from unstrained state up to strain $\Delta L/L_0 = 1\%$ were measured after several previous identical cycles. In each cycle, the strain increased with a linear ramp of 0.23 $\%$/min up to the maximum strain of 1 $\%$ that was preserved for five minutes and followed by an abrupt release of the stress. The strain evolution is indicated by red full line in figure 4a. Obviously, the resistance change is reversible and the almost perfect overlapping of the curves of particular cycles suggests the good reproducibility for the probed strain interval.

The same cycling procedure was applied for several IO-PET samples to estimate the sample-to-sample reproducibility. Here, the strain-resistance transduction depended on the stretching orientation with respect to the orientation of “chaplets”, whereas the initial resistance was not sensitive to the orientation of the current flow. Figure 4b presents two triplets of the samples that were cut in mutually perpendicular directions. For each sample, the fifth of the five sequential cycles is shown in figure 4b. The full/open symbols refer to the samples, where the strain was oriented nearly parallel/perpendicular to the longer chaplet axis (the arrows “P” and “V” in figure 2). The latter configuration yields roughly one order higher ratio $R/R_0$ compared to the former one.

4. Discussion

For further discussion, we mention our recent investigations on the strain-resistance transduction of the one monolayer of ordered thiolalkane-linked Au nanoparticles (NPs) with the core diameter of 6-7 nm deposited onto the BoPET foil (Mylar) [16]. The sample structure was the similar to that of the IO-PET samples except the separation of electrodes being of 50 $\mu$m. The gauge factor of ca 45 was assessed at the strain $(L-L_0)/L_0 = 5\%$, where $L_0$ and $L$ are the pristine and strained sample lengths, respectively. As determined by the small-angle x-ray scattering (SAXS) characterization, the nanoparticle distance being of 6.87 nm for the pristine sample changed proportionally with the deformation of the supporting foil; the constant of proportionality was about 0.48 Å per $\%$ of strain.

To provide a qualitative explanation of the NPs- and cracks-derived transductions differing by 2-3 orders of magnitude, simplified models can be considered. We recall that the tunneling resistance depends approximately exponentially on the interparticle gap [10]

$$ R \propto \exp(\beta \Delta l) $$

(2)

where $\Delta l$ is the interparticle gap change induced by strain. The cracks can be considered as the tunneling gaps too. $\beta$ is the tunneling decay constant describing the tunneling of electrons. It ranges from 0.8 to 1.18 Å$^{-1}$ [23,26,27] for thiolalkane molecules and is $\approx 2$ Å$^{-1}$ [15] for the tunneling in air. When comparing the NPs- and IO-derived samples exposed to the same strain, much higher difference of exponent—and accordingly the resistance change—is brought by the distinct width change of the

Figure 3. The SEM maps of the strained IO-PET film with various magnifications.
tunneling gap. This is owing to the high disproportion in their numbers; given the geometries of the respective samples, a rough estimate yields about 2400 cracks and 7000 gaps across the active length of 24 mm and 50 μm, respectively. Consequently, the crack width is about three orders higher than the interparticle distance change to conform the same strain.

The tunneling mechanism of the charge transport seems to be a key factor for getting the high variance of the film conductivity in dependence on the strain. In reference to the nanostructured films, the fabrication approach often leads to the frequency of tunneling gaps commensurate to the density of nanoobjects and with the nano-gap widths being comparable with the size of nanoobjects themselves. Such structures tend to display lower gauge factor due to the deformation distributed to exiguous changes of the interparticle gaps. Conversely, the decreasing number of gaps would increases the width accrualment of gaps upon the unit strain, which in turn increases the gauge factor exponentially due to the exponential character of the tunneling current.

Assuming the crack width of ca 30 nm—as visualized by the SEM (figure 3b) for the strain of ca 0.5 %—one would expect open circuit behavior. Admittedly, $R/R_0 \approx 100$ only. Such a resistance increase would correspond to the much thinner tunneling gap increase by about 2.3 Å. Therefore, we speculate that whereas the number of tunneling gaps in the IO-PET samples corresponds to the number of cracks visualized by SEM, yet the tunneling gaps are effectively much narrower—likely at

Figure 4. The strain-induced resistance normalized by the initial resistance $R/R_0$ for (a) the five sequential stretching cycles from unstrained state up to strain $\Delta L/L_0 = 1\%$ measured for a particular IO-PET film. The strain evolution, which is indicated by the red full line, increased with the ramp of 0.23 %/min and was released abruptly. (b) The fifth of the five sequential cycles measured for two triplets of IO-PET samples cycled under the same conditions as in (a). The strain orientation with respect to the “chaplets” is indicated next to the triplets. The leading edges are shown only.
the IO/PET interface—than the crack width suggested by SEM. Such behavior could be due to the multilayer structure of the IO-PET films: the depth profiling by the x-ray photoemission spectroscopy (XPS) revealed that the indium oxide film is, in fact, composed of two approximately equally thick indium oxide layers sandwiching a thin Au/Ag layer (not shown here).

Assuming the linear transduction, i.e. the linear resistance dependence on strain, the gauge factor evaluated by equation (1) is insensitive to the upper limit of the probed strain range. However, the gauge factors of the nanostructured films are very high owing to their non-linear responses. Typically, the resistance depends on the strain exponentially due to the conduction governed by the tunneling mechanism. The exponential dependence implies that the gauge factor strongly depends on the upper attainable limit of strain. This is illustrated in figure 5 where the empty symbols show the evolution of the gauge factor for the IO-PET sample introduced in figure 1, which was calculated by means of (1).

\[
g = \frac{\log(R/R_0)}{\log(L/L_0)}.
\]

The IO-PET samples get broken at the strain well exceeding about 1.5 %; obviously, a much higher gauge factor is attainable than the one calculated here according to equation (1).

It follows that the gauge factor determined by (1) lacks information on the strain-gauge sensitivity and the probed strain interval. A modified formula, presumably more appropriate for evaluation of the transductions governed by the exponentially-dependent mechanisms, reads

\[
g = \frac{\log(R/R_0)}{\log(L/L_0)}.
\]

The gauge factor defined by (3) reflects also the signal conversion efficiency and it is much less dependent on the measured strain interval provided an ideal mechanism characterized by the exponential strain-to-resistance transduction.

5. Conclusions
The strain-resistivity transduction of the indium-oxide coated polyethylene terephthalate (IO–PET) flexible substrates reaches the gauge factor of tens of thousands, i.e., exceeds by about two orders of magnitude the up-to-now reported highest values for the nanostructured strain gauges. The extremely high response is due to the transduction mechanism governed by the tunneling conductivity across the cracks formed in the strained films. A redefined gauge factor for the transductions based on exponentially dependent mechanism instead of that for the linear transduction was proposed.

Acknowledgment
The work was supported by Scientific Grant Agency (project No. VEGA 2/0010/15) and Slovak Research and Development Agency (project No. APVV-14-0891).
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