Sensors and Actuators B 261 (2018) 135–143

Contents lists available at ScienceDirect

Sensors and Actuators B: Chemical

journal homepage: www.elsevier.com/locate/snb

Stretchable composite monolayer electrodes for low voltage dielectric elastomer actuators

Xiaobin Ji[a], Alae El Haitami[b], Francesca Sorba[a], Samuel Rosset[a], Giao T.M. Nguyen[b], Cédric Plesse[b], Frédéric Vidal[b], Herbert R. Shea[a], Sophie Cantin[b,*]

[a] Soft Transducers Laboratory (LMTS), Eícole Polytechnique Feídeírale de Lausanne (EPFL), Rue de la Maladieŕe 71B, Neuchâtel 2000, Switzerland
[b] Laboratoire de Physicochimie des Polymères et des Interfaces (LPPI, EA2528), Institut des Matériaux, Université de Cergy-Pontoise, 5 mail Gay Lussac 95031 Cergy-Pontoise cedex, France

ARTICLE INFO

Article history:
Received 15 September 2017
Received in revised form 15 January 2018
Accepted 17 January 2018

Keywords:
Langmuir-Schaefer
Low-voltage dielectric elastomer actuator
Patterned monolayer electrodes
MWCNT polythiophene composite
Large strain actuator
Electroactive polymer

ABSTRACT

In this work, a Multiwalled Carbon Nanotube/poly(alkylthiophene) (MWCNT/PT) composite is developed as the electrodes for dielectric elastomer actuators (DEAs) using the Langmuir-Schaefer (LS) method. These composites form stable monolayers at the air-water interface that can then be LS transferred onto a poly(dimethylsiloxane) (PDMS) elastomer membrane. The monolayer electrode remains conductive up to 100% uniaxial strain. We present a method to fabricate DEAs using the LS transferred electrodes. By using a mask during the transfer step, the electrodes can be patterned with better than 200 μm resolution on both sides of a 1.4 μm-thick pre-stretched PDMS membrane to produce an ultra-low voltage DEA. The DEA generates 4.0% linear strain at an actuation voltage of 100 V, an order of magnitude lower than the typical DEA operating voltage.

1. Introduction

Soft actuators are required when one wishes to integrate active motion or deformation control in compliant or stretchable objects. Soft actuators are thus used in a broad range of fields where the system must be both soft yet capable of actively changing shape, such as soft robotics [1,2], tunable optics [3,4], and compliant grippers [5,6]. Dielectric elastomer actuators (DEAs) are a promising soft actuator technology due to their high energy density [7], large deformation strain [8], and fast response [3]. A DEA consists of a dielectric elastomer (DE) (usually silicone [9] or acrylic [7,10]), sandwiched between two compliant electrodes. When a potential difference is applied between such electrodes, the DE is squeezed in thickness and expands in plane [7]. For deformations of less than approximately 10% (depending on materials and pre-stretch), the in-plane strain S_x is given by [11]:

$$S_x = \varepsilon \frac{E^2}{2Y_m} = \frac{1}{2}\frac{\varepsilon}{Y_m t_m^2} V^2 \quad (1)$$

where ε is the dielectric permittivity of the DE membrane, Y_m is the Young's modulus of the DE membrane, E is the electric field between the two electrodes, V is the applied voltage, and t_m is the DE membrane thickness.

The electrodes used for DEAs are generally assumed not to contribute to the stiffness of the devices. This has been the case for many DEAs fabricated using carbon grease electrodes on acrylic films of thickness several tens of μm. The electrode stiffness can only be ignored when the following inequality is respected [12]:

$$Y_e * t_e << Y_m * t_m, \quad (2)$$

where Y_e is the Young's modulus of the electrode, t_e is the electrode thickness, Y_m is the Young's modulus of the DE, and t_m is the DE membrane thickness.

If Eq. (2) is not satisfied (eg, for very thin elastomer membranes, or stiff metallic electrodes), Eq. (1) must be corrected to account for the stiffening impact of the electrodes. This highlights the role of the electrode mechanical properties on the performance of DEAs.

The maximum strain of a DEA is limited by the breakdown field of the DE (more correctly, the failure mode of DEAs is an electromechanical instability [13], but the breakdown field is the key limiting factor for small strain actuator configurations). For typical elastomer thicknesses of 10 μm to 100 μm, and working near typical elastomer breakdown fields of 100–150 V/μm, DEAs require from 1 kV to 15 kV to reach maximum strain. These high operating voltages limit some possible applications of DEAs, due to the cost, size, and efficiency of high voltage electronics. Therefore, significant

* Corresponding author.
E-mail address: sophie.cantin-riviere@u-cergy.fr (S. Cantin).

https://doi.org/10.1016/j.snb.2018.01.145
0925-4005/

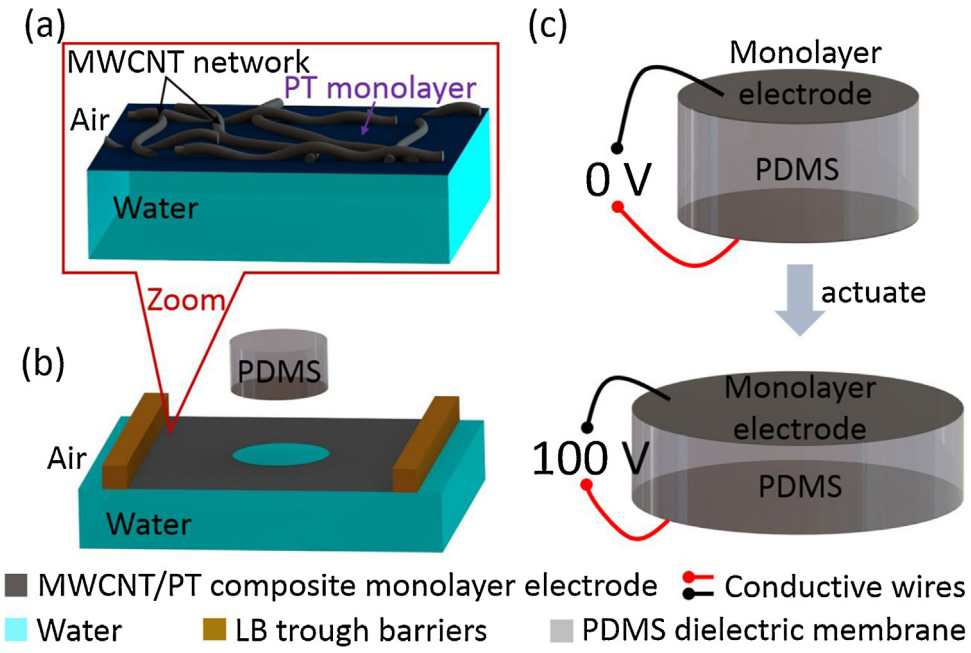

Fig. 1. Low-voltage DEAs made using Langmuir-Schaefer transferred monolayer electrodes (a) Monolayer composite electrode formed at the air-water interface: interconnected MWCNT network embedded inside an polyt(alkylhiophene) (PT) monolayer. (b) Langmuir-Schaefer (LS) transfer of the composite monolayer from the air-water interface onto the PDMS dielectric membrane to make one electrode of the DEA. (c) The DEA consists of a 1.4 μm-thick silicone membrane sandwiched between two sub–100 nm thick composite monolayer electrodes.

research has been carried out to decrease the DEA operating voltage, while keeping the same actuation performance. As can be seen from Eq. (1), to maintain a given actuation strain while decreasing the voltage, either ε must be increased [9,14], or Y_m and t_m must be decreased [15,16]. The strain-to-voltage-squared ratio (S_x/V^2) metric has been used to compare the performance of DEAs with different operating voltages [16]. Decreasing the membrane thickness t_m has resulted in the highest previously reported S_x/V^2 value of 125%/kV^2, for a 3 μm-thick DEA generating 7.5% linear strain at 245 V [16]. Given that the electrodes are generally stiffer than the elastomer ($Y_e > Y_m$ for nearly all materials used in DEAs), when the membrane is made thinner, then the electrode thickness t_e or the electrodes stiffness Y_e must also be further decreased to maintain actuation strain, as summarized in Eq. (2).

Technologies for stretchable electrodes for DEAs, reviewed in references [12,17], include metal ion-implantation [18], transfer of SWCNT layers [19]., ionogels and hydrogels [20,21], or silicones or silicone oils doped with carbon black applied by pad printing [3,16], spray-coating [10], blade casting [22] or screen-printing [23]. Those electrode fabrication methods are not suitable for DEAs with a membrane thickness of 1 μm, which would allow for DEAs working at 100 V, as the electrode thickness is either not negligible with respect to that of the DE membrane, or, for the ion-implanted method, with 50 nm thick electrodes, the electrode stiffness is much too high. A major breakthrough in low-voltage DEAs could be achieved by developing nanometer-thick and stretchable electrodes presenting a low $Y_e{}^*t_e$ value. This would allow the fabrication of DEAs with 1 μm-thick membrane with strain of over 10% at 100 V. To date, electrodes that enable DEAs with more than 1% linear actuation strain under 100 V have not been reported [16,19,22].

Langmuir technology is an appealing alternative to commonly used electrode fabrication methods as it allows the formation of nm-thick films. It is a powerful tool to transfer molecular monolayers from an air-water interface to a solid substrate. By first spreading and then compressing amphiphilic molecules at the air-water interface, one can fabricate monomolecular films, called Langmuir monolayers, that are highly ordered over areas of several hundreds of cm^2 [24]. In addition to producing films one single molecule thick, this technique has the advantage of controlling the density of molecules in the monolayer. These Langmuir monolayers can then be transferred to a chosen substrate using the vertical Langmuir-Blodgett (LB) or horizontal Langmuir-Schaefer (LS) method [25]. Several devices including molecular sensors [26], photo-electrochemical devices [27], organic semi-conductor devices [28], and field effect transistors [29], have been fabricated using Langmuir technology. DEA electrodes based on stretchable monolayer conductors fabricated by the Langmuir technology have not yet been reported.

In this article, Langmuir technology is used to fabricate stretchable monolayer electrodes for DEAs (Fig. 1). Hydrophobic poly(alkylthiophene) (PT) and hydrophilic Multiwalled Carbon Nanotubes (MWCNT) are mixed in a solvent to form a stable amphiphilic composite dispersion. This dispersion can be spread on the water surface to form a composite electrode, in which the MWCNT network is embedded in a PT monolayer (Fig. 1a). PT, as conducting polymer, should contribute to the electronic conductivity and stabilize non-functionalized hydrophilic MWCNT at the air-water interface. Two types of PT with different linear alkyl side chains poly(3-hexylthiophene) (P3HT) and poly(3-decylthiophene) (P3DT)) are studied, since it has been shown that the side chain length has a significant effect on the PT material properties. The longer this chain is (up to 12C), the smaller the Young's modulus and the electrical conductivity [30]. The MWCNT/PT composite monolayer is then transferred from the air-water interface to a poly(dimethylsiloxane) (PDMS) elastomer membrane using the LS method (Fig. 1b). The adhesion is provided by the hydrophobic affinity between the PDMS substrate and PT from the composite monolayer. The 1.4 μm-thick pre-stretched PDMS membrane is sandwiched between two LS transferred composite electrodes to make a 100 V operating DEA (Fig. 1c). The electrodes are evaluated based on their morphological, electrical and mechanical properties with regards to DEA application.