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Scalable fabrication of ionic and capacitive laminate actuators for soft robotics

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ABSTRACT

Soft electrochemically-driven actuators based on high specific surface area electrodes and having a laminate structure similar to the electrical double-layer capacitors are attractive for future generations of biomimetic robotics. Successful incorporation of such ionic and capacitive actuators into soft robots demands highly repeatable and scalable fabrication techniques. As a novel approach in fabrication of ionic and capacitive laminates, the electroactive layers are deposited layer-by-layer around a woven fabric substrate, which is pertained as the centermost layer of the laminate. The electromechanical performance of the actuators having the textile reinforcement layer was on a par with similar actuators without the reinforcement layer; however, the reinforcement layer significantly simplifies the manufacturing process and increases repeatability of the actuators characteristics. More importantly, the reinforcement layer pertains its tensile strength, which gives new opportunities for incorporation of the soft ionic actuators directly onto fabric without compromising their excellent electromechanical performance. Fortunately, the same electroactive laminates, directly exposed to ambient air, have outstanding performance as electric double-layer capacitors for energy storage. Electromechanical characterization with a galvanostatic input was carried out on a large pool of actuators to identify the factors influencing the repeatability of their actuation. The new fabrication procedure yields electroactive laminates with outstanding uniformity in thickness and areal capacitance (standard deviations at 95% confidence interval are 9.2% and 9.4%, respectively) and with a lifetime of more than ten thousand cycles.

1. Introduction

The emerging fields of soft biomimetic robotics are in demand of compatible soft actuators, sensors, and energy-storage materials. Flexible electroactive laminates having a sandwich structure of an ionically conductive layer interleaved in-between electronically conductive layers are ideal candidates for the applications, as they can either simultaneously or intermittently operate as actuators [1], sensors [2], and energy-storage units [3]. These laminates have a similar structure as the electrical double-layer capacitors (EDLCs), which can store energy by electrostatic attraction of mobile ions onto and into high-surface-area electrodes. The electro-osmotic relocation of mobile ions during charging of the energy-storage laminate also results in expansion or contraction of the electrodes due to the unequal size [4] and mobility of the cations and anions in the ionic domain.

Commonly, the soft electrochemically activated artificial muscles are classified based on their particular electrode materials (as the bucky-gel actuators (BGAs) [5,6]), the particular membrane materials (e.g. the ionic polymer transducers (IPTs) [7]), or a combination of constituent materials (e.g. the carbon-polymer composites (CPCs) [8] and ionic polymer-metal composites (IPMCs) [9]). However, the underlying working principles of these actuators are not determined by the particular constituent materials only. The recent years have yielded new classes of structural [10], flexible [11], and even stretchable [12] laminates, which are structurally similar to the ionic actuators, but which are optimized for energy storage as EDLCs only. As a remark, the electrodes and the membrane of flexible EDLCs and ionic actuators are fused into a single entity, whereas the functional layers of a ‘traditional’, commercial EDLC are simply in touch contact. On the whole, it is evident that the

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difference between EDLCs and electrochemically activated artificial muscles is gradually diminishing.

In this paper we introduce a new term – ionic and capacitive laminate (ICL), which identifies a class of soft ionically conductive laminates that have simultaneously the best qualities of an electromechanical actuator and a flexible energy-storage unit.

Soft and flexible actuators are needed for a new generation of soft and shape-morphing robots [13,14]. These robots find application, for example, in civil or military surveillance and in rescue missions, as they are able to pass through small gaps and to grasp gently delicate objects of varying shape and surface texture. ICLs have a number of exclusive properties that make them particularly suitable as actuators in biomimetic soft robotics. ICLs also offer the desired level of compliance and benefit from their high level of integration due to their flexibility and multifunctionality. ICLs operate at voltage levels not exceeding five volts, which can be considered safe for wearable applications. Moreover, the operating voltage of an ICL matches perfectly with the typical voltage levels found in contemporary digital complementary metal–oxide–semiconductor (CMOS) microelectronics (usually not exceeding 3.3 V) and in single-cell lithium-ion batteries (up to 4.2 V), which makes interfacing ICLs to their control units very straightforward. ICL actuators with ionic liquid electrolytes exhibit long-term stable operation even in vacuum [15], and they even endure low-Earth orbit conditions [16], which make it possible to design robots and manipulators with ICL actuators for space applications. ICLs are particularly beneficial for robots in a size scale of a few centimeters and below: at a larger size, the ICLs could not compete with conventional electromagnetic actuators, whereas the ICLs can be fabricated in a few-micrometer scale [17], which is not easily reachable by using conventional actuator technologies.

To date, the use and even early-stage prototyping of ICLs in soft robots has been constrained by the availability of ICLs, because the manufacturing process of ICLs (a) requires diligent manual labor, as the currently available manufacturing methods could not be easily automated; (b) is relatively complex, which reflects in its poor repeatability; and (c) often relies on prohibitively expensive materials (e.g. both platinum and ionic polymers in case of IPMCs). Concerning the potential applications, a practical ICL manufacturing process must be reproducible and scalable, and preferably should not rely on prohibitively costly materials. To date, a number of available manufacturing techniques for ICLs are advantageous for small (around 1-cm^2) batches in laboratory conditions, but the scalability is still challenging. For example, it was shown by Addinall et al. [18] that the layer-by-layer assembly process developed by Fukushima et al. [5], which involves fusing the layers by hot-pressing, has a tendency of forming short-circuit ‘hotspots’ when the manufacturing process was scaled up. Another successful method for ICL fabrication, first employed by Akle et al. [7,19], is the direct assembly process (DAP), that is, spray-coating of the electrodes on a free-standing cast membrane. A disadvantage of this method is the fact that the extensive and anisotropic swelling of the free-standing membrane during the manufacturing process can cause bucking of the laminate and can result in inconsistent electrode coverage. Most importantly, the loss in the mechanical strength of the membrane during the fabrication process limits the possibilities for scaling up the fabrication. Limited scalability is a common issue also for producing flexible EDLCs [20]. Consequently, to facilitate further applied research on ICLs, there is a huge demand for scalable methods for preparing ICL actuators in a repeatable fashion.

In this work, we report on an improved process that is not limited to laboratory-scale batches, but can yield ICLs in quantity and size appropriate for industrial manufacturing. Recently, methods for fabrication of ICLs using structural, fabric *electrodes*, have been reported [21,22]. Although fabric electrodes can be optimal for EDLCs, the high axial tensile strength of the fabric electrodes can limit the performance of ICL actuators, as the axial strain governs their actuation. Thus, instead of using fabric electrodes, we have fabricated an ICL with a fabric-incorporated *separator*: the newly-developed ICL is formed layer-by-layer on the sides of a fibrous woven substrate. The fibrous substrate is under tension in a fixed position that prevents swelling of the material in the axial direction during fabrication, thus providing a pathway to fabricate ICL actuators using a continuous process. The presence of a non-stretchable reinforcement layer is a significant benefit in comparison to the previous DAP fabrication technique, as extensive swelling of the membrane during application of the subsequent electrode layers can eventuate in a mechanical failure of the membrane. Without the reinforcement layer, the non-homogenous swelling causes the membrane to curl, which can eventuate in a non-uniform coating of the subsequent layers. In the worst cases, the softened membrane can get mechanically torn apart or the electrodes could get short-circuited. A non-swelling membrane and a uniform electrode coating are especially critical in making materials with patterned electrodes [23]. The improved DAP is demonstrated in a semi-laboratory-scale, however, this can be easily scaled up – automated industrial roll-to-roll conveyors can be used for preparing ICLs in an unprecedented repeatability, cost, and quantity. We have characterized a large pool (17 pcs) of actuators fabricated using the improved method to identify the standard deviation of the fabrication process in terms of mechanical, electrical, and electromechanical properties. Further, we have analyzed the measured data to identify the possible correlation between the electrically measurable parameters and the actuation performance.

As a step of symbolic significance, the resulting ICL is completely ionic-polymer-free. Actuators with non-ionic polymer membranes have been demonstrated in previous work [1]; however, ionic polymer Nafion® has been still employed as glue. The omission of ionic polymers from the ICL constitution increases their price and consequently also their availability.

The incorporation of a woven fibrous interlayer has a greater impact to the ICL technology than merely the improved scalability. To date, the incorporation of ICL actuators into or onto textiles has been impeded due to the incompatibility between fabrication processes of the textiles and the ICLs. The newly developed process creates a new technological framework for incorporation of ionic actuators with a large variety of textile and mesh supports. An ICL having extra reinforcement layer has an excellent load-bearing capacity that provides an opportunity to create smart textiles with inherently integrated ICLs, in similar to the fiber-reinforced soft pneumatic actuators [24]. As a perspective, this process can be further extended to create patterned functional textiles, where the membrane and electrode layers have been applied only to certain areas of the textile, rendering only these specific areas electroactive. The high areal capacitance of ICLs suggests their perspective use also as flexible supercapacitors, either as the sole function or in combination with actuation. Therefore, the textile-incorporated ICLs are desired not only for soft robotics, but also for smart textile applications and for energy storage.

2. Experimental

2.1. Materials

1-Ethyl-3-methylimidazolium trifluoromethanesulfonate ionic liquid (EMIM-Otf, >99.0%) and N,N-dimethylacetamide (DMAc) were purchased from Fluka. Poly(vinylidene fluoride-co-hexafluoropropene) (PVdF-HFP) and 4-methyl-2-pentanone (MP) were purchased from Sigma-Aldrich. Potassium acetate (CH_3CO_2K) was obtained from Fischer Scientific. Titanium carbide-derived