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Ionic liquid-based actuators working in air: The effect of ambient humidity

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ABSTRACT

The ionic electromechanically active polymer (iEAP) actuators with ionic liquid electrolytes are distinguished by their ability for operation in ambient air, but also respond to the change in ambient relative humidity. Contrarily to the iEAP actuators with water-solvated electrolytes, water content of an ionic liquid electrolyte has been considered either of little importance in iEAP actuators or to cause unfavorable effects such as hydrolysis of some anions such as BF_4^-.

The effect of ambient relative humidity on the electromechanical properties of an iEAP actuator with 1-ethyl-3-methylimidazolium trifluoromethanesulphonate (EMITFS) electrolyte, porous carbon electrodes and Nafion membrane is investigated. The ambient humidity has an extensive impact on the frequency response of the actuator. In addition, electrochemical impedance spectroscopy is employed to specify the equivalent schematics for the iEAP actuator.

The physicochemical parameters, especially viscosity and conductivity, of ionic liquids are strongly dependent on their water content and have therefore huge impact on actuation characteristics. The other constituents of the laminate are also hygroscopic, which amplifies and accelerates the water sorption even further.

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1. Introduction

Recently, the development of soft, flexible, stretchable, foldable and printable electronics and robotics has attracted a lot of interest, including the goal of development of wearable and biointegrated devices [1–8]. The transition from rigid silicon-based technology toward the soft polymer-based technology evokes the need for compatible power sources, sensors and actuators. The electric double-layer capacitors (also called supercapacitors) are often considered as power conditioning or even energy storage media in soft and flexible electronics [9–13]. In addition, supercapacitor-like ionic electromechanically active polymer (iEAP) actuators with ionic liquid (IL) electrolytes [14–16] are considered as a promising technology for use as actuators embedded with soft electronics and robotics. A common requirement for the iEAPs is the ability of operation in non-liquid environments, which can contain different gaseous substances at variable partial pressures. The ambient conditions set a number of prerequisites for the materials iEAPs are composed of. The well-developed technology of conventional, sealed supercapacitors or iEAPs could not be directly taken over to soft electronics (or ionics) and robotics.

The iEAP actuators considered for use in robotic and micromanipulation appliances have been developed since the early 1990s [17]. One particular representative type of iEAPs – ionic polymer–metal composites (IPMC) with anionic polymer – is normally operated while immersed in deionized water. Their electromechanical actuation mechanism is based on the formation of cation concentration gradient in the thickness direction of a thin laminate. The electro-osmosis of cations also induces water concentration gradient and results in macroscopic actuation due to anisotropic swelling [18]. On the contrary, the iEAP actuators with IL as electrolytes can be operated in air for over 250 000 cycles [19].

The electromechanical properties of an iEAP actuator with carbonaceous electrodes and IL electrolyte working in air have been thoroughly investigated [15,16,20]. The three-layer laminate constructed in this work consists of an Nafion membrane, sandwiched between two electronically conductive electrode layers. The electrode is a composite of high surface area porous carbon particles bonded by ionic polymer chains. In particular, carbide-derived carbon (CDC) is used as an electrode material. The whole laminate is

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impregnated with an IL. The cluster network of the Nafion membrane swollen in an IL serves two purposes: (a) is responsible for the ionic conductivity between the electrodes; (b) acts as a reservoir for IL. Within one electrode the carbon particles are in direct physical contact with each other, ensuring electronic conductivity. Due to relatively high electron transfer resistance between the separate carbon particles within one electrode, the extra current collector layers made of gold foil are added on top of the electrodes. When electric potential is applied between the electrodes, electric double-layer is formed in the electrode–electrolyte boundary. The magnitude of actuation is related to the size and shape difference between the cations and anions [21].

The actuation properties of the ionic polymer–metal composite (IPMC) actuators with water-solvated electrolyte have a huge dependence on the ambient RH level [22]. This is expected, because the working mechanism of IPMC includes water convection fluxes. The RH level also has an influence on back-relaxation of IPMCs. On the contrary, the working mechanism of a solvent-free IL-based iEAP actuator is considerably different from the water-based IPMC actuators, which suggests different behavior in relation to the RH level. However, to date the impact of water content upon the electrical and electromechanical characteristics of iEAP actuators with IL electrolytes has been considered as insignificant. There are three major considerations for doing so: (a) the effects of ambient water absorption are typically not considered significant in the performance of actuators working in air. Interestingly, Kikuchi et al. [23] have demonstrated that the relation between bending curvature and transported charge is not considerably affected by the ambient relative humidity level. (b) A number of potential application fields for iEAPs presume operation in dry environments or in vacuum such as in spacecrafts. (c) It is well-known that an important presumption for the long-term in-air operation of iEAPs is the fact that the ILs have negligible vapor pressure even in vacuum at temperatures below their boiling temperature [24]. The low interest in the humidity-related effects is particularly expressed in the fact that the state-of-the-art prototypes of iEAP actuators (and also flexible supercapacitors) with IL electrolytes are often investigated in ambient air conditions, without specifying the instantaneous RH value of the surrounding environment.

This paper is focused on characterizing the effect of ambient relative humidity (RH) on the electrical as well as electromechanical properties of the iEAP laminate. The results lead to an useful feature of the material – the ability to measure the relative humidity value of the environment, in which the laminate is located.

The measured impedances are fitted with a simple equivalent circuit that consists of a charge transfer resistance, a Warburg element and a parallel connection of resistance and a constant phase element. The dependence of frequency response on ambient RH level and therefore also humidity content of the laminate is explained by the impedance results. Besides, this work also indicates the important role of absorbed water on the overall working principle of iEAP actuators with IL electrolytes.

2. Experimental

2.1. Material fabrication

The three-layer iEAP actuator was manufactured using the 'direct assembly process' introduced by Akle et al. [25]. A Nafion 117 ionomer membrane purchased from FuelCellStore.com was first cleaned by boiling in 1 M hydrochloric acid, washed by boiling in deionized water, and then ion-exchanged by immersing in $LiClO_4$ solution. After subsequent removal of solvents in vacuum, the membrane was immersed in 1-ethyl-3-methylimidazolium trifluoromethanesulphonate (EMITFS) IL (Fluka, ≥99.0%). By doing so,

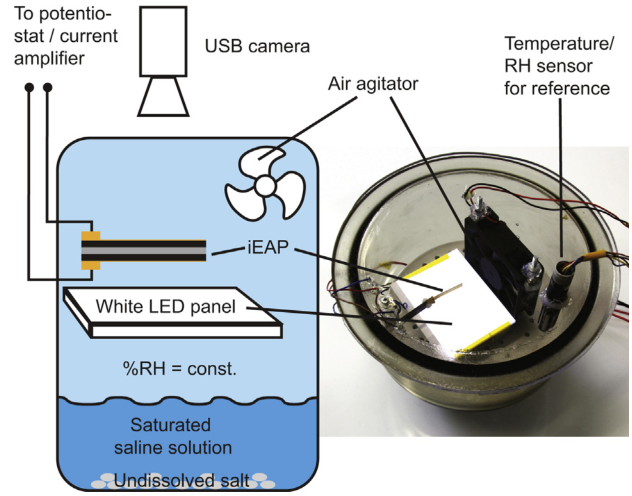

Fig. 1. The measurement set-up.

virtually all of the Li+ cations were exchanged by the EMI^+ cations. The electrode material was prepared by dispersing titanium carbide-derived carbon (TiC-CDC) powder (Skeleton Technologies) with 15 wt% Nafion solution (LIQUION® LQ-1115 1100EW; Ion Power, Inc.). Isopropanol was used as solvent. The weight ratio between TiC-CDC powder and Nafion in the electrode was 41:59. An ultrasonic probe was used to promote homogenization. The dispersion was painted layer-by-layer directly on both sides of the previously IL-impregnated Nafion membrane using an airbrush. After application of electrode layers on both sides of the membrane, volatile solvents were evaporated under an infrared lamp. Finally, the membrane was sandwiched between ∼100 nm gold foils (Gold-Hammer) and fused together by hot-pressing under 3.5 MPa at 70 °C for 5 s. Some extra Nafion solution was used to promote adhesion of the gold foil. The gold foils act as current collectors, minimizing the voltage drop along the electrode. The thickness of the finished iEAP laminate was 250–280 μm. The manufacturing method and the electromechanical properties of the laminate have been described in detail by Palmre et al. [15].

2.2. Humidity generation

The impact of humidity was measured by placing the iEAP laminate in a chamber with controlled RH, as depicted in Fig. 1A. The iEAP laminate was held in an environment of constant RH for at least 3 h before the measurements, while the RH value in the chamber was continuously equilibrated using an air agitator. During the measurement, the air agitator was turned off. A Rotronic HC2 temperature and humidity sensor was used for RH value reference. Five fixed humidity points were generated. The lowest humidity (0 ± 4%RH was generated by using calcium chloride ($CaCl_2$) as absorbent, and the highest humidity value (100 ± 4%RH) settled over a bath of distilled water. The fixed RH values of 23 ± 2, 53 ± 2, and 75 ± 2%RH were generated by using saturated solutions of potassium acetate (CH_3CO_2K), magnesium nitrate ($Mg(NO_3)_2$), and sodium chloride (NaCl), respectively [26].

The measurements were conducted using two different approaches: (a) direct measurement of the actuation properties; and (b) measurement of electrical impedance in a variable RH level. In the course of the measurements the iEAP sample was not removed from the humidity chamber. All experiments were conducted at room temperature (24 ± 1 °C).

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