Thermo- and electromechanical properties of bucky-gel bending actuators under external loading

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1. Introduction

Bucky-gel actuators are a type of ionic electromechanically active polymers (IEAP) that, when subjected to electric stimulus, exhibit mechanical response such as bending. Conventional bucky-gel actuator consists of ion-containing polymer membrane sandwiched by two electrode layers with carbon nanotubes (CNT) [1].

The exact working principle to explain the mechanical response of bucky-gel actuators is still a matter of on-going dispute [1]. However, in the broadest sense, the electric voltage causes relocation of ions within the multilayer structure and either due to electrostatic forces or ion concentration gradient one electrode layer undergoes expansion while the other is compressed. This imbalance within the bucky-gel laminate is visible as bending.

The end performance of an IEAP actuator is a combination of a diverse range of factors, such as the type of ions, specific surface area and conductance of electrodes, porosity of separator layer, rigidity of the laminate structure, humidity and temperature of the operating environment, input signal frequency and voltage, dimensions and the shape of the actuator, etc (see e.g. refs [1,2]). There are a variety of studies investigating the effect of additives or altered composition ratios on the overall performance of the actuators. The final effects are often contributed to either some mechanical (e.g. elastic modulus) or electric (e.g. capacitance, conductance) property altering due to the particular additive [2].

The performance of a bending actuator is generally characterised by the maximum amplitude of its deflection and blocking force in response to voltage excitation. As the geometry of material and experimental setup also influences these characteristics, deflection and force are often normalised to strain and stress in order to facilitate comparison between different actuators [1,2].

It is often the case that actuators of smaller rigidity exhibit large and rapid deflections but rather small blocking force values [3]. Conversely, thick and more rigid actuators exhibit small and slow bending while blocking force can be relatively high [4]. Optimisation to achieve good deflection values together with high blocking force may be considered one of the main objectives of the researchers developing new IEAP actuators.

On a generalised level, it can be claimed that the mechanical output of an IEAP is the sum of two forces:
1) Electrically induced driving force $F_D$
2) Restoring force of the material $F_R$

It is quite intuitive that in the case of a cantilever actuator the driving force needs to overcome the opposing restoring force for bending to occur. When the difference between these forces is large (i.e. $F_D \gg F_R$), the resultant acceleration is bound to cause relatively rapid deflection. However, as the restoring force has principally elastic nature, it increases with the amplitude of deflection, thus causing the bending to eventually stop.

If IEAP materials were purely elastic, the restoring force could be described by means of the Hooke's law, however, studies indicate that the performance of the actuator is dependent on the input frequency, temperature, etc [5]. Additionally, the polymers in general are known to be viscoelastic, i.e. when subjected to deformation they present both elastic and viscous characteristics [6]. Probably the most frequently registered manifestation of viscoelastic nature of an IEAP is the creep observed during repeated work-cycles of an actuator. Therefore viscous response of polymer actuators must also be considered when modelling actuator performance and/or choosing new materials to create actuators from.

In the current paper the properties of restoring force and driving force are investigated. Mechanical and...
thermal analysis of bucky-gel actuators is carried out. The focus of the paper is to understand how the elasticity and viscosity affect the overall performance of a bucky-gel actuator.

2. Experimental

A dedicated experimental setup was constructed consisting of force sensor B12-115 by Nano Control and laser distance meter Keyence LJ-V7060 mounted on motorised stages. The setup allowed measuring force at a variety of free deflections of a cantilever actuator (fig.1-a). A PC with LabVIEW 2012 was used to control the experimental procedure, including signal generation and acquisition via NI PCI-6251 DAQ card.

The driving voltage for the actuator was a bipolar rectangular signal with the period of 40 s. It was generated at the sampling refresh rate of 1 kHz and amplified using Hokuto Denko HAL3001 potentiostat. Frequency of data acquisition for all the measured signals was 200 Hz.

The full procedure of these measurements is presented in detail in fig.1-b and -c. It was possible to simultaneously measure the force and the displacement for different free deflection paths ($\delta > 0$) of the actuator. Furthermore, the force sensor could also be used to externally bend the actuator ($\delta < 0$), thus imposing constant strain and enabling measurements of viscoelasticity.

In the beginning of the experiment the force sensor is placed just beyond the maximum deflection of the actuator, after measuring both force and displacement under rectangular voltage excitation, the motorised stages shift the force sensor 100 µm towards the actuator and measurements are repeated (fig.1-b). After the force sensor reaches zero-deflection (i.e. conventional measurements of blocking force), it starts to gradually impose constant strain to the actuator. Example of how relevant values are determined from the experimental data is depicted in fig.1-c, where approximately -0.7 mm deflection is used to strain the actuator prior to applying input signal.

Thermo-mechanical analysis of the bucky-gel actuators was carried out using the cyclic tension measurement mode of the mechanical measurement system Hitachi EXSTAR DMS6100. Temperature was varied from 20 ºC to 150 ºC (at the rate of 1 ºC/min), while sinusoidal mechanical excitation with the amplitude of 10 µm and frequencies in the range of 0.01 – 20 Hz was applied to the sample.

Temperatures on the surface of the bucky-gel actuator before, during and after its work-cycle were recorded using a thermal imaging infrared camera CHINO CPA-E60.

While several bucky-gel actuators with different composition were tested, for the sake of brevity, this paper reports results of one type of actuator. It was a tri-layer laminate with electrodes of polyvinylidene fluoride-cohexafluoropropylene (PVdF(HFP)), single-walled CNTs and EMIMBF4 (32/20/48 wt). The separator membrane composed of PVdF(HFP) and EMIMBF4 (50/50 wt). The sample size was $24 \times 5 \times 0.159$ mm.

3. Results

The relation between maximum output force at different distances of deflection (at $L = 5$ mm) is depicted in fig.2. Maximum output force and displacement of the actuator increases with the input voltage, however, series of experiments at different lengths $L$ and input voltages demonstrate that the correlation is not always linear, especially in the case of displacement. That means the slope of the linear fit (depicted in fig.2) is dependent on the input voltage.

Fig.3 depicts the data from the experiments, in which
force sensor imposes gradual strain to the bending actuator. The viscoelastic nature of bucky-gel actuator becomes evident in fig.3-a as the restoring force exhibits non-linear relation to distance (i.e. the Hooke’s law does not apply). The data also indicate that the restoring force is dependent on the input voltage. As the strain histories at different voltages can be considered identical, this phenomenon is probably due to Joule heating occurring within the actuator in response to the current caused by input voltage. Further proof in support of this theory of electrical heating is provided later in the paper.

Intriguingly under external load the maximum force output decreases as the input voltage increases (fig.3-b). By assuming \( F_{OM} = F_R + F_D \), driving force \( F_D \) was calculated and is depicted in fig.3-c. The driving force is rather weakly dependent on input voltage in comparison to, say, \( F_R \).

Results of thermo-mechanical measurements show that the mechanical properties of the bucky-gel laminate are clearly dependent on the temperature (fig.4). It is observed that the storage modulus \( E' \) (related to the elasticity of the material) decreases as the temperature increases, i.e. the material becomes less elastic. Significant changes are observed already in the ranges of few degrees above and below common room temperatures; therefore, even small thermal variations (e.g. due to Joule heating) can noticeably influence the elasticity of a bucky-gel laminate. The loss modulus \( E'' \) (related more to the viscosity of the material) remains constant around room temperature.

Tab.1 lists temperatures measured from the surface of the bucky-gel actuator before, during and after its repeated work-cycles. The temperature increases noticeably on the surface of the actuator during its work-cycle. Using an infrared camera it was observed that the temperature is highest near the fixing clamp and gradually reduces towards the far end of the bending sample. As expected the temperature and current peaks appeared simultaneously. The higher input voltages resulted in higher peak temperatures during the work-cycle of the actuator. Furthermore, even 60 seconds after removing input signal, surface temperature was registered 2 ºC over the ambient temperature.

<table>
<thead>
<tr>
<th>( V_{in} ) (V)</th>
<th>Ambient &amp; actuator ( T ) (ºC)</th>
<th>Peak ( T ) during work-cycle (ºC)</th>
<th>( T ) after 10 periods of ( V_{in} ) (ºC)</th>
<th>( T ) after 60 s (ºC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.4</td>
<td>24</td>
<td>28</td>
<td>27</td>
<td>26</td>
</tr>
<tr>
<td>2.0</td>
<td>24</td>
<td>29</td>
<td>27</td>
<td>26</td>
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Tab.1 Temperatures measured from the surface of a bucky-gel actuator before applying the input signal, during the work cycle, after 10 periods of the input signal, and 60 s later.

4. Discussion

At this point the evidence indicates to a significant thermal dissipation during the work-cycle of the bucky-gel actuator. Temperature at the surface of the actuator can increase nearly 5 ºC under the input of 2 V, however it is likely that within the separator membrane the rise in the temperature may be even higher. One clear effect of such heating is the observed decrease of the material’s restoring force (or elastic modulus) in response to higher voltages. However, as the restoring force values were registered when no input signal was applied to the sample, the observed effect is either due to residual heat from the previous period or the actuator’s shape gets “reprogrammed” by the combination of internal heating and external loading.

The data about restoring force (fig.3-a) and displacement output (fig.2) of the bucky-gel actuator indicate that the increased deflection at higher voltages is mainly due to the reduced storage modulus at higher temperatures (voltages). As the bucky-gel actuator has symmetric structure, the acquired data on restoring force can be reflected over zero-point to describe restoring force during the free actuation of the sample. Even if that maximum driving force is marginally altered by the input voltage, an increase in displacement can already be achieved due to the voltage induced (thermal) change in the elastic modulus. In other words, the voltage-dependent slopes of restoring force (fig.3-a) are the likely reason behind the altered slopes in fig.2.

Presented results point out that thermal response of the materials is critical to the general performance of bucky-gel actuators. For instance, better performances can be achieved by materials that exhibit relatively large decrease in elasticity in response to even small variations of temperature. When actuator is expected to lift loads or close valves, low amplitude input signals can result in better performance as the material remains more rigid.

5. Conclusions

In this paper the results of thermo-mechanical study of bucky-gel actuators are reported. It is demonstrated that that bucky-gel laminates exhibit change in their
viscoelastic properties already at room temperatures. Since the driving signal causes Joule heating of a bucky-gel actuator, during its work-cycle the elastic modulus decreases as the input voltage is increased. Even after removing the driving signal, the temperature of the actuator remains above ambient temperature. Experiments with external loading demonstrate that because of this change in elastic properties, the maximum output force of a bucky-gel actuator may actually decrease at higher voltages.

REFERENCES


