Experimental Investigations on Carbon Nanotube Actuators Defining the Operation Point and Its Standard Deviation

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ABSTRACT

Carbon nanotube (CNT) actuators have been extensively investigated from the perspective of materials, their composition, and system construction as well as from three main performance features, which are displacement, force and velocity. However, up till now none of the CNT actuators have reached the stage of implementation into products. It is due to the fact that even though from the point of view of performance each property can reach satisfactory values, their combination is much more difficult, as they are not proportional. This relation of properties motivated the work to test and investigate currently available CNT-polymer actuators to define their operation point. Under this term one should understand a performance of actuator where displacement, force and velocity do not affect each other. In other words, any change in one of the properties will adversely affect at least one of the remaining ones.

The measurements are performed in out-of-plane mode on 2 cm diameter samples in low frequency range (0.01 - 1 Hz) under application of low voltage (2 V).

Measurement curves of three main actuator properties are plotted together against the frequency resulting in operation point as the intersection point of those curves. Additionally the deviations in actuator performance are assessed to reflect the actuators' reproducibility and their production process stability by means of standard deviation.

Knowledge about the relation between actuator properties and the value of operation point will facilitate evaluation of the existing CNT actuator against its potential applications.

Keywords: Carbon nanotube, actuator, displacement, force, velocity, performance, experimental investigations

1 INTRODUCTION

Actuator is a component of prime importance in all smart structures. Its purpose is to induce strain into the system in order to change its shape or to compensate disturbing vibrations. Currently, besides many materials of minor importance, there are two major groups that are commercially available – piezoceramics and shape memory alloys [1]. Even though, these materials are widely used in industrial applications, they exhibit several disadvantages. Among those one can list necessity for high operating voltages and currents, together with high material density [1]. These characteristics lead to search for more effective substitutes. Actuators that are based on carbon nanotubes have the potential to overcome drawbacks of currently used materials [2]. They can possibly offer higher work per cycle than previous actuator technologies and generate much higher mechanical strength. Additionally, CNTs require very low driving voltages for their operation which is the major advantage over ferroelectric, magnetostrictive and electrostrictive materials. Another advantage is the direct conversion of electrical energy to mechanical energy followed by high actuation strain, high strength, high elastic modulus and low density. However, up till now, none of the developed carbon nanotube actuators has made it to the commercialization stage. The choice of suitable application was mostly hindered by the compromises that have to be accounted when characterizing the performance of such actuators. From this point of view they can be mainly divided into those generating high strains at relatively high speed and the others which bring or sustain high stresses. This compromise is hardly ever accepted from the industry, especially after great characteristics promises shown by CNTs at the early stage of development but which were not fully realized thus far. For this reason an attempt is made to characterize currently available CNT-polymer actuators focusing on the three main features and find the performance at which all the features can be utilized to their optimum. Such an analysis should make it easier to define the applications at which the carbon nanotube actuators could be integrated in the near future.

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2 CARBON NANOTUBE ACTUATORS

2.1 Actuator Types

The first macroscopic carbon nanotube actuator was reported in 1999 by Baughman et al. [3]. That actuator was composed of three layers. The inner layer was a commercially available double-sided scotch tape. Two outer layers that were fixed on the tape were carbon nanotube sheets, so called Bucky Papers. The actuator was immersed in salt-water electrolyte. Application of the voltage to the nanotube electrodes caused the actuator structure to bend. Reversing the polarity caused the bimorph to bend in the opposite direction.

Even though the operation of the CNT actuator was successful, from the point of industrial applications this actuator had several disadvantages. First of all, it was of low mechanical stability which made it difficult to handle. However, a bigger drawback was the necessity to operate such an actuator in an electrolyte bath. This would have made the actuator system much too big, complicated and expensive to gain real attention from the industry. That is why there was a need to develop actuators that could be operated in air.

The first dry actuator, fabricated through layer-by-layer casting with "bucky gel", a gelatinous room-temperature ionic liquid that contains single walled carbon nanotubes (SWNTs) [4] was reported by Fukushima et al. [5]. That actuator was adopted in a bimorph configuration with a polymer-supported internal ionic liquid electrolyte layer, sandwiched by bucky gel electrode layers, which allow operation in air at low applied voltages.

The bucky gel actuator presented several advantages. First, the fabrication does not require any special apparatus and can readily be extended to printing-based processing essential for miniaturization of machinery. The observed performance and durability were one of the highest among those reported for low-voltage driven, dry electromechanical actuators at the time. That development provided an important step toward the realization of miniaturized mechanical devices.

Dry actuators, under investigation here, were constructed on the basis of actuators reported by Fukushima et al. [5] and Ionic Polymer Metal Composites (IPMCs). They are composed of three layers where two outer ones are composite materials comprising of CNTs, electrolyte and polymer matrix and the inner layer is commercially available ion exchange membrane. This membrane due to its high mechanical stability and stiffness promotes bending of the actuator structure and adds to the values of generated force.

Dry operation is possible due to the fact that electrolyte is embedded in the polymer matrix. During solution processing of composites, a polymer of choice is obtaining porous structure which promotes the retention of electrolyte within the composite structure. Ionic liquids, which are used here, are organic, water free, electrolytes that allow application of higher voltages without a risk of chemical decomposition and decrease of electrolyte performance. It was already shown that the amplitude of displacement of carbon nanotube structures increases proportionally with an increase in input voltage [6].

The difference between actuators implemented in this study and other CNT actuators is the shape of the sample and the arrangement of the measurement setup [6]. These dry actuators are stamped into circular form and for the measurement the sample is positioned horizontally to the surface of the electrode and contacted at its whole surface. The measurement is called out of plane actuation measurement, where the displacement is measured in the direction perpendicular to thickness of sample, thus the results are often presented as percent values of displacement in respect to the thickness of sample. Actuation principle for dry, CNT-polymer composite actuators is similar to that described by Baughman [3]. Application of square wave voltage results in bending of the actuator. Reverse in the polarity of voltage results in bending motion of the actuator in the opposite direction.

2.2 Actuation Mechanism

The physical phenomenon responsible for exhibiting strains in this kind of actuator is not yet completely understood. Up till now, several different mechanisms for actuation of carbon nanotubes were reported. Physical phenomena, such as electrochemical double layer charging [3], electrostatic actuation [7], and photothermal actuation [8] etc. have been considered as factors responsible for the actuation. Most plausible mechanism for carbon nanotube actuation is double-layer charge injection, wherein a nanotube acts as an electrochemical capacitor with charge injected into the nanotube which is balanced by the electrical double-layer formed by the movement of electrolyte ions to the nanotube surface. The charge injection causes quantum chemically based dimensional changes in the carbon-carbon bond length of the surface atoms [9]. As a result expansion/contraction of single carbon nanotube can be observed. Nevertheless, since the first observation of this effect, less is known about the macroscopic actuation properties of bucky papers or composite

actuators. Currently it can be only taken into consideration that the actuation of individual nanotubes is transferred to macroscopic dimensions and so it is possible to see the actuation of an entire mat of nanotubes [9].

There are several parameters, previously reported in [10], [11], that influence the electromechanical performance of carbon nanotube actuators. These parameters can be separated into two main categories. The first category involves the actuator itself including the type of carbon nanotubes used, their different preparation techniques, the purification grade, the dimensions, and the homogeneity of the nanotubes distribution in the actuator as well as its size and thickness. The second category includes the parameters of the test equipment and arrangement, like the type of electrolyte, electrode materials, surface electrode resistance, arrangement of electrodes, applied voltage, frequency of the applied voltage. In order to characterize the influence of all the above mentioned parameters, a great amount of tests would have to be carried out. Thus, here described are investigations primarily focusing on the influence of operational frequency on the performance of the CNT-polymer actuator.

3 EXPERIMENTAL DETAILS

3.1 Materials

MWNT material under investigation was purchased from Ahwahnee Technology Inc. As given by the manufacturer this material had a purity of 95% carbon content and was used as received, without further purification. The diameter and length range for the material is 7-15 nm and 0.5-200 μ m, respectively. For the experimental investigations, CNT-polymer actuators (three layer structure) with total thickness of 1250 μ m were produced. All of them were stamped into a circular form with a diameter of 20 mm.

3.2 Instrumentation

The experimental setup and the out of plane test configuration have been described by Haque et al. [11] and Kosidlo et al [6]. Figure 1 schematically shows the position of the electrodes and the measurement direction of the mechanical deformation. Three layer carbon nanotube actuators have been placed between the working and counter graphite electrodes. From the actuation principle, dry actuators work in a similar way as the Bucky Paper actuators mentioned earlier. Application of voltage of different polarity at each side of the actuator causes one of the carbon nanotube layers to expand and the second one to contract, and the membrane in between those layers causes the whole structure to bend (see Fig. 1). The top electrode, positioned on the surface of the actuator (see Fig. 1), is free to move and as a consequence of the actuator movement, it is lifted up and down. The change of distance, between the surface of top electrode and the sensor positioned above it, is read out as the actual displacement of actuator.



Figure 1. Schematic representation of the measurement setup configuration (left) and the principle of the actuator displacement (right).

The measurement setup allows also investigations of the force generated by the actuator. For these measurements additional load is applied on the top electrode. The weight which is lifted by the actuator can be correlated with the force it generates.

3.3 Data Analysis

Measured values of displacement are plotted in the form of time dependent curves with the use of LabView software. The recording and analysis of the data is conducted with a program using LabView software. Simultaneously with the running measurement this program is displaying data, in forms of time depended diagrams, considering applied voltage, current, displacement and temperature. Additionally, from the data collected by the LabView program it is possible to calculate further values useful for characterization of actuators like charge injected into nanotubes, efficiency of the actuator and energy density. These calculations are supported by the use of Excel or Origin software.

A description of different influencing parameters responsible for carbon nanotubes actuation was reported by Haque et al. [10], [11]. In this paper, number voltage patterns of different frequency have been applied on the same CNT-polymer actuator sample to analyze the corresponding actuation behavior.



Figure 2: Applied potential-time profile for five different frequencies under investigation and the corresponding displacement as actuator response.

For the purpose of these investigations, the voltage pattern as shown in Figure 2, with varying frequencies, has been applied. The rectangular light gray line represents the voltage profile and the black line the corresponding actuation (or active displacement) profile. Please note that for all the tests in this paper, when the actuation curve moves upwards, it represents contraction of the material and expansion is represented by the curve moving downwards.

4 RESULTS AND DISCUSSION

4.1 Displacement and Velocity

Initially, displacement tests under the input signal of square wave voltage of ± 2 V were conducted with the variation of the potential frequency. Five frequencies, in the range from 0.01 Hz to 1 Hz were chosen. The displacement results of

those investigations, for samples measured with varying pre-loads, as well as the velocities calculated for those samples are presented in Figure 3.



Figure 3: Displacement (left) and velocity (right) dependent on frequency for CNT-polymer actuators measured with five different pre-loads.

It can be seen clearly from Figure 3 that the level of actuator's displacement is inversely proportional with the frequency. Increase in frequency leads to a decrease in actuation performance. An especially sudden drop in performance can be observed at the first change in the frequency input, from 0.01 Hz to 0.05 Hz.

A similar trend can be observed for the velocity calculated for those samples. In general the velocity is decreasing with increasing frequency, however in this case the top performance is being recorded at the frequency of 0.05 Hz.

These results are expected of CNT-polymer actuators, whose actuation is based on the principle of ion movement and intercalation, a process which is never of a high speed, especially in a solid matrix, but which can be optimized by the variation of the electrolyte based on its ion mobility and conductivity properties.

4.2 Force

In the second stage, the sample which was investigated for its performance at varying frequencies was evaluated for its performance under application of increasing stress, in order to state the trend in the force generation. The results of this investigation are presented in the Figure 4.

From the two diagrams it can be seen that both the displacement and velocity are inversely proportional to the applied stress, behavior similar to that in relation to increasing frequency. The best displacement results were obtained from the measurement where the applied pre-load was 60 g, what equates to approximately 0.6 N.

The improvement which can be observed in the displacement and velocity observed with the first increase of the applied stress can be explained by the fact that due to this increase, the contact between the electrodes in the measurement setup and the surface of actuator is improved. The improved electrical contact is responsible for faster charging of carbon nanotubes in the actuator layers and thus for faster and higher displacement of the actuator. Any further increase in the applied stress is working against the actuator requiring the higher forces to be generated.

It is important to state, however that at all applied stresses, up to approximately 3.7 N, it was possible to record the actuation performance.



Figure 4: Displacement (left) and velocity (right) dependent on force for CNT-polymer actuators measured at five different frequencies.

4.3 **Optimum point**

Based on the analysis of the above presented results it can be stated that the actuation performance of CNT-polymer actuators is inversely proportional to the frequency of operation as well as to the application of stress. The same trend is observed for the calculated velocity as a function of frequency and applied stress. Due to these dependencies it is not possible to define one operation point at which all of the performance properties would be at their optimum. As has been seen earlier the CNT actuators can be divided into two classes where the first includes actuators characterized by high displacement and quick velocity and the second one consisting of actuators generating high forces.

However, the calculation of the mechanical energy of the actuator gives a possibility to define the optimum operation region of the actuator. As can be seen from the diagram in Figure 5, which is presented for the actuator operating at frequency of 0.01 Hz, the optimum operation can be defined in the range from 0.6 N to 1,2 N.



Figure 5. Displacement and mechanical energy as a function of force for CNT-polymer actuators measured at 0.01 Hz.

The assumption can be made that with increase of the applied stress the mechanical energy is as well increasing due to the improvement of the contact properties between actuator and electrodes, what can further influence the resistance of the system as well as the charge transfer.

As in the first class of actuators defined here two of performance characteristics (displacement and velocity) are in their optimum and additionally for the given point the mechanical energy is also at its maximum, this actuator and its performance are suggested as the optimum operation point of the actuator.

4.4 Standard deviation of operation point

In the case of our investigations a single measurement (using the same operating parameters) was made on the sample loaded with a force of 0.6 N and using a frequency of 0.01 Hz, with a corresponding probability of 68 % that the result is within the standard deviation σ_x of the correct value. Thus, we can adopt σ_x to mean "uncertainty". It is reasoned as follows: We measure d (displacement) for one actuator 20 times, and then the mean of these measurements should give a good estimate of d for the one actuator. The standard deviation σ_d of these 20 measurement cycles provides us with an estimate of the uncertainity in our method of measuring d. Provided all other actuators are reasonably similar and we use the same method to measure each one; we can reasonably expect the same uncertainity in each measurement [12]. 20 measurement cycles were conducted on the actuator and the obtained displacement d results are shown in Table 1.

Measurement number	1	2	3	4	5	6	7	8	9	10
Displacement	27,5	36,7	25,6	32,8	25,9	30,7	29,9	30,3	29,2	29,0
[µm]										
Measurement	11	12	13	14	15	16	17	18	19	20
number										
Displacement	29,3	28,6	29,1	28,4	29,0	27,9	28,6	27,2	27,6	25,6
[µm]										

Table 1. Displacement results from 20 measurement cycles performed on one sample.

From these values we can calculate $d_{mean} = 28,992 \mu m$ and, using the equation:

$$\sigma_{\mathbf{x}} = \sqrt{\frac{1}{N-1}\sum d_i^2} = \sqrt{\frac{1}{N-1}\sum (x_i - \overline{x})^2}.$$

Where σ_x is the standard deviation, N is number of measurements, d_i^2 is deviation squared, x_i is measured value and \overline{x} is the mean value of the measured ones.

The uncertainity in any one measurement of d is therefore approximately $\sigma_d = 2.52 \ \mu m$.

The same calculations are conducted for the velocity s based on the results (Table 2) of measurement on one sample within 20 measurement cycles.

Table 2. Velocity results from 20 measurement cycles performed on one sample.

Measurement	1	2	3	4	5	6	7	8	9	10
number										
Velocity	0,56	0,69	0,54	0,66	0,53	0,60	0,57	0,63	0,54	0,62
[µm/s]										
Measurement	11	12	13	14	15	16	17	18	19	20
number										
Velocity	0,56	0,56	0,59	0,55	0,59	0,54	0,5	0,53	0,57	0,54
[µm/s]										

From these values we obtain $s_{mean} = 0.5824 \ \mu m/s$ and the standard deviation as uncertainity in one measurement is $\sigma_s = 0.043 \ \mu m/s$.

When evaluating those results in correlation with the mean values of performance we obtain that the standard deviations as the uncertainity in a single measurement correspond to 8.7 % and 7.4 % for displacement and velocity, respectively.

5 CONCLUSIONS

In this paper we have presented experimental investigations on the performance of CNT-polymer actuators. A number of experiments have been performed varying only the frequency pattern while keeping the rest of the input parameters constant in the actuation and velocity test cases. In the further step, measurements with application of pre-load were performed for each frequency in order to evaluate the performance of actuators under stress.

Summarizing, two classes of actuators were defined where the first one is characterized by high displacement and quick response time and the second one by high forces.

It was found that currently available actuators show the best performance in terms of displacement and velocity at low frequency patterns. Similar behavior is observed for the performance under applied stress. With the increase of the preload the displacement and velocity are decreasing. At high frequencies and high forces the displacement of the actuator can still be measured, however at the very low scale. The calculation of the mechanical energy, however, enabled the definition of the operation range of the actuator. The best performance in terms of mechanical energy was defined in the range of 0.6 N-1.2 N and was assumed to be the operation range of the actuator. More detailed investigations in this range may allow in the future defining the operation point only.

Based on the single sample measured it was calculated that the standard deviation for displacement and velocity is 8.7 % and 7.4 %, respectively. In the future, the investigations should be extended to a multitude of samples in order to evaluate the reproducibility.

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